Fixed bound atom interacting with a coherent pulse of quantized radiation: Excitation and short-time behavior

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A model of a coupled atom—quantum-field system is presented, consisting of a time-independent Hamiltonian and a realistic dynamical initial condition. The Hamiltonian describes a fixed bound atom coupled to the complete electromagnetic field. The initial condition is a consistent quantum-mechanical description of an atom in the exact ground state of the coupled atom-field system, on which is superimposed an approaching quasiclassical excitation of the field which will cause the interesting dynamical processes later on. Tools are presented for the manipulation of this model, with emphasis towards series expansions of all relevant quantities suitable for small strength λ of the atom-field coupling and (potentially) large magnitude α of the coherent field excitation, such that the "influence of the coherent excitation on the atom" (i.e., the product $\lambda\alpha$) remains constant as λ tends to zero. The following results are obtained, at the lowest nontrivial order in λ, in an approximation suitable for times much shorter than any radiative lifetime of the atom: (a) The change in free-field energy due to the interaction with the atom is *exactly* compensated at all times by the change in bare-atom energy plus the atom-field coupling energy. This checks the consistency of the proposed scheme. (b) The frequency distribution of this change in free-field energy has a (somewhat unexpected) component, with a smooth spectrum, spreading over the whole width of the spectrum of the exciting pulse, in addition to δ and dispersive singularities centered on the atomic transition frequencies. This frequency distribution is discussed in detail and its features are illustrated by a number of figures, in a simple case, numerically tractable. (c) The heterodyne detection of the radiation emitted by the atom is investigated, with emphasis on the issue of causality and propagation of light. *Strict causality* is obtained, without further approximation, in a model with an atom-field coupling involving the field at a single point and a detector similarly sensitive to properties of the field at a single point. This result breaks down if one uses nonlocal approximations such as the rotating-wave approximation or a detection observable $\mathbf{E}^-(\mathbf{r})\cdot\mathbf{E}^+(\mathbf{r})$. Finally, an appendix is devoted to the use, in quantum mechanics, of bases moving with respect to each other.

I. INTRODUCTION

Time-dependent quantum theory is still far from being as well understood and developed as the quantum theory of stationary situations. This reflects the formidable difficulty of dynamics in general (particularly for systems involving a field and/or a large number of particles), and also the scarcity of time-dependent experimental results requiring precise quantum theory for their interpretation. Even such a basic and apparently simple model as the interaction of a fixed bound (i.e., not ionized) atom with the quantized electromagnetic field presents considerable difficulties when one undertakes to predict its *dynamical* behavior entirely from the exact quantum equations of motion.

A first source of problems is the initial condition for which no satisfactory general solution is available. For instance, (a) assuming the irradiation of the atom stationary for a long time (i.e., removing the problem at $t\rightarrow-\infty$) is not directly compatible with the study of the decay of an excited state by spontaneous emission, (b) turning on the atom-field interaction adiabatically does not fare better, and (c) approximations like "the atom in a bare excited state and the field in its ground state at $t=t_0$" are *very rough* descriptions of any realistic situation and do not correspond to any physically plausible past for $t<t_0$.

A second source of problems is particularly acute when one deals with irradiation pulses of high intensity; either one describes the field classically with the advantage that the corresponding Bloch equations for the atom easily predict the strongly nonlinear short-time response, and the major drawback that radiative effects (such as spontaneous emission) are absent from such a model in which they can only be simulated by the *ad hoc* incorporation of phenomenological damping terms, or one uses a properly quantized description of the field and faces serious difficulties in discussing the strongly nonlinear behavior of the atom by time-dependent perturbation techniques (even for short times).

A third source of problems, of a more fundamental type, arises in the discussion of the long-time behavior. For instance, little is known, entirely from first principles, about the asymptotic situation of a coupled atom-field system with persistent irradiation or the transient approach to it. Even in the absence of irradiation, the standard solution is still the approximation proposed by

Weisskopf and Wigner[1] in 1930 (and its variants), which does not lead to a systematic perturbation scheme.

In this and a subsequent paper, we present a model of a coupled atom-field system which alleviates the above-mentioned problems of quantum dynamics. The model consists of a time-independent Hamiltonian *and* a realistic dynamical initial condition, with well-defined expansion parameters. The Hamiltonian describes a bound N-level atom, at a fixed position in space, interacting with the complete electromagnetic field. The initial situation is a consistent quantum-mechanical description of the "naive" idea of an atom in the exact dressed ground state of the coupled atom-field system, on which is superimposed an approaching quasiclassical (also called coherent) excitation of the field which will cause the interesting dynamical processes later on. This initial situation is easily generalized to the thermal equilibrium state of the coupled atom-field system, again with superposition of an approaching quasiclassical excitation. The main parameters are the strength λ of the atom-field coupling and the magnitude α of the coherent-field excitation. We shall consistently treat λ as a small expansion parameter, and let α increase when λ tends to zero in such a way that the "influence of the coherent excitation on the atom" (i.e., the product $\lambda\alpha$) remains constant.

In this paper we present the model together with some tools for its manipulation and a detailed discussion of its short-time behavior, using fairly standard quantum mechanics. The examples show that this, indeed, provides a particular, usable solution for the "first source of problems" (initial condition) mentioned above and also, for times much shorter than all radiative lifetimes, for the "second source of problems" (classical versus quantized field). Of course, the short-time expansion techniques could be pursued to higher order, but this would rapidly lead to forbidding complications.

In a forthcoming paper we shall show how this model can be treated with nonunitary transformations (in "Liouville" or operator space) of the type introduced by Prigogine and his co-workers.[2] The second law of thermodynamics, which remains the guiding principle for the long-time behavior of complex systems, is incorporated in the construction of these basic "star unitary" transformations. The method leads to a transformed evolution equation for the transformed density operator in which dressing, decay, and excitation are disentangled. Using this equation one obtains satisfactory results for times comparable to the lifetimes of atomic excited states, hence providing a solution to all three sources of problems mentioned above. We hope to present specific predictions for situations of actual experimental interest in further publications.

In Sec. II of this paper we first define the notation for the Hamiltonian and basis states, and recall some properties of the quasiclassical states of the field. Next we use a time-dependent unitary transformation [denoted $M(t)$], in the way indicated by Mollow,[3] to disentangle the quasiclassical field pulse from the rest of the problem. The result is a transformed state of the system from which the (potentially large) field pulse has been removed, and a transformed equation of motion with a generator of the motion containing a time-dependent term describing the coupling between the atom and the classical-field pulse *together* with all the terms in the original Hamiltonian. The time-dependent term is proportional to the product $\lambda\alpha$ of the strength λ of the atom-field coupling by the magnitude α of the field pulse. Hence this term can be kept constant, possibly describing a large effect of the field pulse on the atom, even in the limit of $\lambda\to 0$, by suitably increasing α when λ decreases.

The first significant use of this transformation $M(t)$ is to provide an explicit quantum-mechanical expression for a realistic and stable initial condition valid for all times before the beginning of the overlap between the atom and the classical field associated with the quasiclassical excitation of the quantized field. Let us emphasize that the unitary transformation actually describes the quasiclassical-field excitation, hence this transformation depends upon the initial situation for the field and not upon the bare-atom Hamiltonian or the atom-field coupling. Let us also note that, in its present form, the whole procedure used here hinges on a transformation which is defined only for quasiclassical excitations of the field.

From this point on the discussion in our forthcoming paper will involve nonunitary transformations and strongly deviate from the more conventional techniques used in the rest of this paper.

In Sec. III we set up a perturbative scheme for solving the transformed equation of motion obtained in Sec. II, in which the "unperturbed" starting point is the exact solution of the corresponding Bloch equations (classical-field approximation) and the "perturbation" is a transformed version of the atom-field coupling Hamiltonian. For this we use two additional time-dependent unitary transformations to successively "eliminate" the motion due to the bare-atom and free-field Hamiltonians, and the motion which would take place in the classical-field approximation. The result is a transformed equation of motion with a generator of the motion proportional to the strength λ of the atom-field coupling, and a transformed state which is close to the ground state of the free field and bare atom. This combination is very suitable for using the standard expansion of the transformed state (described by a density operator for convenience) as a power series in λ; such an expansion is very useful for times much shorter than any radiative lifetime of the atom as, in such conditions, one can restrict oneself to the first few terms as in standard perturbation theory.

As an illustration of the use of the proposed procedure, we have examined a few examples in the perspective of a strength λ of the atom-field coupling tending to zero while the magnitude α of the (quasiresonant) field pulse increases in order to keep $\lambda\alpha$ constant. The first example, in fact a check on the consistency of the procedure, rests on the fact that the basic Hamiltonian H_S of the model is time independent, hence the quantum average energy $\langle H_S\rangle$ is an exact invariant of the motion and this must also hold true for each term in a series expansion in powers of λ. In Sec. IV we first show that the leading term (in α^2, hence in λ^{-2}) of such a series expansion is the time-independent energy of the unperturbed field pulse. At the next nontrivial order (in λ^0), some care is required

in the discussion to show that the changes in bare-atom energy *and* in atom-field coupling energy are *exactly* compensated at all times by corresponding opposite changes in free-field energy. This result holds in situations in which the properties of the atom may have very fast time dependences, hence indicating that the heuristic notion of "wide energy level" which is often associated with fast evolution, is in no way required for a satisfactory interpretation of *exact* calculations.

In Sec. V we examine the frequency distribution of the change in free-field energy caused by the atom-field interaction, again in a short-time approximation which ignores spontaneous emission. For an experiment of the type sketched in Fig. 1 this would be the difference in the response of the "high-resolution spectrometer" to a pulse with and without atom, for pulses much shorter than the radiative lifetimes of the atom (and neglecting the fraction of spontaneous emission which hits the spectrometer). In the approximation of weak pulses, which leave the atom almost unaffected, the spectrum of the absorbed radiation has the anticipated structure of one δ function at each unperturbed atomic-transition frequency, with the coefficients which would be given by Fermi's golden rule. However, for pulses which strongly affect the state of the atom, two qualitatively new components of the "absorption spectrum" appear: one with a *continuous* frequency distribution spreading over the whole width of the pulse spectrum, and one with dispersionlike singularities centered on each δ function. In the particular cases where the pulse leaves the atom "exactly" in an eigenstate of the bare-atom Hamiltonian, the singular spectral features are exactly zero, leaving only the continuous component. For large pulses, the various spectral features can be positive or negative, and the energy moved by the atom from one frequency range to another in a single short pulse can be larger than the maximum energy stored by the atom itself. This somewhat unexpected spectral behavior is illustrated by a number of figures, which have been worked out numerically for a simplified model with a two-level atom.

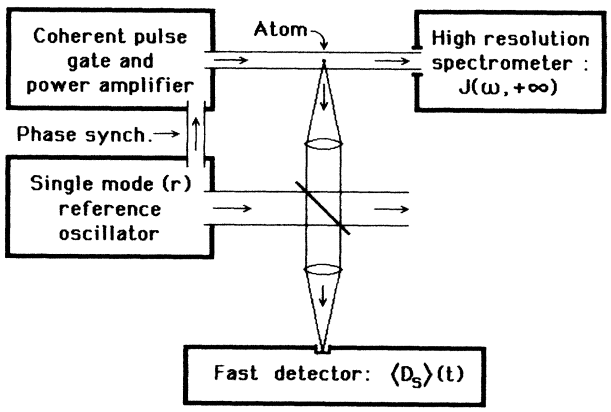

FIG. 1. Sketch of the fast-pulse experiments for which the theory is discussed in this paper. The high-resolution spectrometer measures the pulse spectrum and the spectrum of the energy exchanged between the atom and the field modes which contribute to the coherent pulse. The fast detector measures the beats between the radiation emitted by the atom and the coherent reference radiation (r).

In Sec. VI we briefly discuss the heterodyne (also called phase-sensitive) detection of radiation emitted by the atom in the context of the experimental setup roughly sketched in Fig. 1, where the relevant output is now the average counting rate of the "fast detector." The phase-sensitive detection technique provides detailed information about the nondiagonal part of the atom reduced density operator (in a representation which diagonalizes the bare-atom Hamiltonian), and is, in this respect, complementary to the direct detection of spontaneous emission. We have also examined the issue of causality and propagation of light for phase-sensitive detection with a point atom and a point detector. The result of our calculations is that the detector output at time t depends only upon the state of the atom at the single time $t-\Delta t$, where Δt is the propagation delay from atom to detector, provided that the atom-field coupling involves the field *at a single point* and that the detector is similarly sensitive to properties of the field *at a single point.* This gratifying exact result breaks down if one uses a nonlocal approximation of the electric dipolar atom-field coupling such as the rotating-wave approximation, or if one replaces the (local) electric field operator $\mathbf{E}(\mathbf{r})$ by its (nonlocal) "components" $\mathbf{E}^{+}(\mathbf{r})$ or $\mathbf{E}^{-}(\mathbf{r})$ in the construction of a model for the photoelectric observable.

In Appendix A we give some details about the coherent states of a single harmonic oscillator and propose a basis of *orthogonal* states which is very suitable for the discussion of states close to a coherent state. The particular case of a two-level atom is discussed in Appendix B, and miscellaneous calculations are collected in Appendix C.

Finally, in Appendix D, we discuss the use of bases moving with respect to each other in quantum dynamics, and the relations between mobile bases and interaction representations.

II. DESCRIPTION OF THE MODEL

A. Hamiltonian and sketch of the initial condition

The model presented here consists of a quantum state space, a time-independent Hamiltonian, and an initial condition. The state space and Hamiltonian describe a bare and bound atom with a nucleus at a fixed position in space, the complete electromagnetic field, and an atom-field coupling. The simplest initial condition corresponds to the naive picture in which the atom and the field in its vicinity are in the ground state of the coupled atom-field problem, whereas the distant field contains a (perhaps large) quasiclassical excitation which will interact with the atom later on. In Sec. II D this naive picture will be formulated in a consistent quantum way.

The quantum state space for the coupled atom-field system (labeled by the index S) is the tensorial product of the state space for the isolated atom (labeled by the index A) with that for the unperturbed field (labeled by the index F) which, itself, can be represented as a tensorial product of state spaces for each individual field oscillator (labeled by the oscillator index k). For clarity, all quantum objects such as bras, kets, operators, and traces, will

usually be labeled to indicate the relevant quantum space. To avoid too clumsy notations, the operators a_k and $a_k^\dagger$ will be tacitly extended to the whole field whenever necessary (by tensorial multiplication with the unit operators $1_{k'}$ for all the other modes k' of the field), and the indices F, A, and S will be dropped when this does not cause ambiguity.

The Hamiltonian H_S of the coupled atom-field system can be written in the form

$$H_S=(1_A\otimes H_{0F})+(H_{0A}\otimes 1_F)+\lambda V_S\ , \tag{2.1}$$

where $\otimes$ denotes tensorial multiplication and 1_A and 1_F are unit operators (respectively for the bare-atom and unperturbed field state spaces). The unperturbed field Hamiltonian H_{0F} is given by

$$H_{0F}=\sum_k \hbar\omega_k a_k^\dagger a_k\ , \tag{2.2}$$

where $a_k^\dagger$ and a_k are the usual boson creation and annihilation operators for the field oscillator with label k, and the zero-point energy has been omitted. The bare-atom Hamiltonian is given by

$$H_{0A}=\sum_i \hbar\omega_i\,|\,i\rangle_{A\,A}\langle i\,|\ , \tag{2.3}$$

where $i=1,2,\ldots,$ labels the eigenstates of the bare atom. In this paper, we shall implicitly ignore the ionized states of the atom, hence use the fiction of a bare-atom state space of finite dimension spanned by bound states. For further simplicity we shall often assume that the ground state of H_{0A} is nondegenerate (and label it $i=1$). We shall usually approximate the atom-field coupling by an expression linear in the a_k and $a_k^\dagger$,

$$\begin{aligned}\lambda V_S(\mathbf{r}_A)&=\lambda\sum_k\sum_{i,j}\{V_{kij}^*(\mathbf{r}_A)\,|\,i\rangle_{A\,A}\langle j\,|\otimes a_k+V_{kij}(\mathbf{r}_A)\,|\,j\rangle_{A\,A}\langle i\,|\otimes a_k^\dagger\}\\&=\lambda\sum_k\sum_{i,j}\{\,|\,i\rangle_{A\,A}\langle j\,|\otimes[V_{kij}^*(\mathbf{r}_A)a_k+V_{kji}(\mathbf{r}_A)a_k^\dagger]\}\ ,\end{aligned} \tag{2.4}$$

where the complex quantities $V_{kij}(\mathbf{r}_A)$ are functions of the (fixed) position $\mathbf{r}_A$ of the atom and the real number λ is introduced for further use as an expansion parameter in perturbation calculations. For the time being, the "strength" λ of the atom-field coupling may be replaced by unity. For instance, the linearized electric dipolar approximation for λV_S is given by

$$\lambda V_{\mathrm{ed},S}(\mathbf{r}_A)=-\lambda\boldsymbol{\mu}_{eA}\cdot\mathbf{E}_F(\mathbf{r}_A)\ , \tag{2.5}$$

where $\boldsymbol{\mu}_{eA}$ is the atomic electric dipole operator and $\mathbf{E}_F$ the electric-field operator [see (2.7)], and the magnetic dipolar interaction of a spin with the radiation field is described by

$$\lambda V_{\mathrm{md},S}(\mathbf{r}_A)=-\lambda\boldsymbol{\mu}_{mA}\cdot\mathbf{B}_F(\mathbf{r}_A)=-\lambda\gamma\hbar\mathbf{I}_A\cdot\mathbf{B}_F(\mathbf{r}_A)\ , \tag{2.6}$$

where $\boldsymbol{\mu}_{mA}$ is the spin magnetic moment operator, $\hbar\mathbf{I}_A$ the actual spin angular momentum operator, $\mathbf{B}_F$ the magnetic induction operator [see (2.7)], and γ the magnetogyric ratio. In (2.5) and (2.6) the dot indicates scalar product in configuration space *and* tensorial product of quantum operator spaces. The particular case of the "two-level atom" is discussed in Appendix B.

The quasiclassical excitation, which is the time-dependent feature of the initial condition *at the initial time* t_i is completely specified by the corresponding classical field $\alpha f_{\mathrm{cl}}(t_i)$, which is chosen to be exactly zero in the vicinity of the atom. The notations $f_{\mathrm{cl}}(\mathbf{r},t)$ and $f_{\mathrm{cl}}(t)$ symbolically stand for the $\mathbf{E}_{\mathrm{cl}}$ *and* $\mathbf{B}_{\mathrm{cl}}$ fields at time t and, respectively, at point $\mathbf{r}$ and at all points in space. The real number α is introduced for further use as an expansion parameter in perturbation calculations. For the time being, the strength α of the irradiation field may be replaced by unity.

B. Quasiclassical ("coherent") states of the unperturbed field

We consider an unperturbed classical electromagnetic field with a linear and conservative set of equations of motion with constant coefficients and no source (this includes the specifications about boundary conditions). The state of this classical field can be completely specified (for all positions $\mathbf{r}$ accessible to the field and for all times t) by the electric field $\mathbf{E}_{\mathrm{cl}}(\mathbf{r},t_0)$ *and* the magnetic induction $\mathbf{B}_{\mathrm{cl}}(\mathbf{r},t_0)$, both at the single time t_0. In Coulomb gauge, an alternative complete specification is provided by the (real) function $\mathbf{A}_{\mathrm{cl}}(\mathbf{r},t_0)$ *and* its time derivative $\partial\mathbf{A}_{\mathrm{cl}}(\mathbf{r},t_0)/\partial t_0$. As a preparation for quantization, we follow standard practice and introduce a discrete basis of (complex-valued) normal modes of the classical field, labeled by an index k and such that the unperturbed classical field at any time t is given by

$$\begin{aligned}\mathbf{E}_{\mathrm{cl}}(\mathbf{r},t)&=\mathrm{Re}\sum_k\{\mathbf{e}_k(\mathbf{r})\alpha_k(t)\}\\&=\tfrac{1}{2}\sum_k\{\mathbf{e}_k(\mathbf{r})\alpha_k(t)+\mathbf{e}_k^*(\mathbf{r})\alpha_k^*(t)\}\ ,\\\mathbf{B}_{\mathrm{cl}}(\mathbf{r},t)&=\mathrm{Re}\sum_k\{\mathbf{b}_k(\mathbf{r})\alpha_k(t)\}\\&=\tfrac{1}{2}\sum_k\{\mathbf{b}_k(\mathbf{r})\alpha_k(t)+\mathbf{b}_k^*(\mathbf{r})\alpha_k^*(t)\}\ .\end{aligned} \tag{2.7}$$

We shall not further specify this basis here in order to retain the possibility of describing the (linear) optical instruments used in the experiments (mirrors, lenses, polarization analyzers, etc.) by the corresponding basis of normal modes of the field. The complex numbers $\alpha_k(t)$ evolve in time according to[4]

$$\alpha_k(t)=\alpha_k(t_0)\exp[-i\omega_k(t-t_0)]\ , \tag{2.8}$$

with real positive frequencies $\omega_k/2\pi$, and the total field energy $E_{\rm cl}$ is given by

$$E_{\rm cl}=\sum_k \hbar\omega_k |\alpha_k|^2 . \tag{2.9}$$

The set of complex numbers $\{\alpha_k(t_0)\}$ provides an alternative complete specification of the classical field.

A very important tool in the discussion of such a classical field is the classical-superposition principle. Namely, if the fields $f_{{\rm cl},1}(\mathbf{r},t)$ and $f_{{\rm cl},2}(\mathbf{r},t)$ both satisfy the equations of motion and boundary conditions, then the field $f_{{\rm cl},1}(\mathbf{r},t)+f_{{\rm cl},2}(\mathbf{r},t)$ also does.

In the standard quantum description the unperturbed field Hamiltonian (omitting the zero-point energy) is given by (2.2) and the quantum observables for the electric field and the magnetic induction are

$$\begin{aligned}\mathbf{E}_F(\mathbf{r})&=\tfrac{1}{2}\sum_k\{\mathbf{e}_k(\mathbf{r})a_k+\mathbf{e}_k^*(\mathbf{r})a_k^\dagger\} ,\\ \mathbf{B}_F(\mathbf{r})&=\tfrac{1}{2}\sum_k\{\mathbf{b}_k(\mathbf{r})a_k+\mathbf{b}_k^*(\mathbf{r})a_k^\dagger\} .\end{aligned} \tag{2.10}$$

This quantization procedure ensures that, for any quantum state of the unperturbed field (this includes fields known only in a statistical way), the quantum averages $\langle f_F(\mathbf{r})\rangle(t)$ of the field operators exactly satisfy the classical equations of motion and boundary conditions. The notations $f_F(\mathbf{r})$ and f_F symbolically stand for the pair of observables (2.10), respectively at point $\mathbf{r}$ and at all points in space.

To any given unperturbed classical field $f_{\rm cl}(t)$, we can associate a single, well-defined "quasiclassical" (or coherent) quantum state of the field, which we shall denote $|f_{\rm cl}(t)\rangle_F$, by the requirements that (a) the quantum average field is equal to $f_{\rm cl}(t)$, i.e., $\langle f_F(\mathbf{r})\rangle(t)={}_F\langle f_{\rm cl}(t)|f_F(\mathbf{r})|f_{\rm cl}(t)\rangle_F=f_{\rm cl}(\mathbf{r},t)$ at all positions and times, and (b) the quantum average field energy $\langle H_{0F}\rangle$ has the minimum value compatible with the given average field. These requirements are satisfied by the state

$$|f_{\rm cl}(t)\rangle_F=|\{\alpha_k(t)\}\rangle_F=\prod_k|\alpha_k(t)\rangle_k , \tag{2.11}$$

where $\prod_k$ indicates an infinite tensorial product, $|\alpha_k(t)\rangle_k$ is such that

$$a_k|\alpha_k(t)\rangle_k=\alpha_k(t)|\alpha_k(t)\rangle_k , \tag{2.12}$$

and the set of complex numbers $\{\alpha_k(t)\}$ specifies the classical field $f_{\rm cl}(t)$ in the sense of (2.7). If the above requirements are used to specify a density operator, the result is the *pure* state $|f_{\rm cl}(t)\rangle_{F\,F}\langle f_{\rm cl}(t)|$.

The extension to quantum theory of ideas borrowed from classical theory of fields (such as the classical superposition principle), and the manipulation of quantum states which are exactly or approximately quasiclassical, are greatly simplified by the use of the "displacement" operator introduced by Glauber,[5] which we shall denote M. To any given unperturbed classical field $f_{\rm cl}(t)$, one associates a unitary displacement operator $M_F(f_{\rm cl}(t))$ defined by

$$M_F(f_{\rm cl}(t))=M_F(\{\alpha_k(t)\})=\prod_k M_k(\alpha_k(t)) , \tag{2.13}$$

where

$$M_k(\alpha_k(t))=\exp[\alpha_k(t)a_k^\dagger-\alpha_k^*(t)a_k] . \tag{2.14}$$

As discussed in more details in Appendix A for a single oscillator, the displacement operator provides a simple tool for constructing coherent states from the ground state $|0\rangle_F$ of the field,

$$\begin{aligned}&|f_{\rm cl}(t)\rangle_F=M_F(f_{\rm cl}(t))|0\rangle_F ,\\ &|f_{\rm cl}(t)\rangle_{F\,F}\langle f_{\rm cl}(t)|=M_F(f_{\rm cl}(t))|0\rangle_{F\,F}\langle 0|M_F^\dagger(f_{\rm cl}(t)) ,\end{aligned} \tag{2.15}$$

and has the simple symmetries

$$M_F^{-1}(f_{\rm cl}(t))=M_F^\dagger(f_{\rm cl}(t))=M_F(-f_{\rm cl}(t)) \tag{2.16}$$

and $M_F(0)=1_F$, and the group property

$$M_F(f_{{\rm cl},2})M_F(f_{{\rm cl},1})=M_F(f_{{\rm cl},2}+f_{{\rm cl},1})\exp(i\varphi_{21}) , \tag{2.17}$$

where the real phase

$$\varphi_{21}=-\tfrac{1}{2}i\sum_k(\alpha_{k,2}\alpha_{k,1}^*-\alpha_{k,2}^*\alpha_{k,1}) \tag{2.18}$$

only causes minor inconveniences which completely disappear in a density-operator formalism. We have dropped the time variable in expressions valid at all times, in which all quantities are taken at the same time and no time derivative is involved.

Useful properties of the unitary transformation M are

$$\begin{aligned}M_k(\alpha_k(t))a_kM_k^\dagger(\alpha_k(t))&=a_k-\alpha_k(t) ,\\ M_k(\alpha_k(t))a_k^\dagger M_k^\dagger(\alpha_k(t))&=a_k^\dagger-\alpha_k^*(t) ,\end{aligned} \tag{2.19}$$

and similar relations with the roles of M and $M^\dagger$ interchanged using (2.16) or (A7). These relations can be extended to any function $g(a_k,a_k^\dagger)$ which can be expanded in a power series [see (A12)], and to the complete quantum field space [see (2.13)]. As an example, combining (2.10) and (2.19), we obtain

$$M_F(f_{\rm cl}(t))f_FM_F^\dagger(f_{\rm cl}(t))=f_F-f_{\rm cl}(t)1_F . \tag{2.20}$$

A straightforward quantum version of the classical-superposition principle for the classical fields $f_{{\rm cl},1}$ and $f_{{\rm cl},2}$ is obtained by combining (2.15) and (2.17),

$$\begin{aligned}&|f_{{\rm cl},2}+f_{{\rm cl},1}\rangle\langle f_{{\rm cl},2}+f_{{\rm cl},1}|\\ &\quad=M_F(f_{{\rm cl},2}+f_{{\rm cl},1})|0\rangle_{F\,F}\langle 0|M_F^\dagger(f_{{\rm cl},2}+f_{{\rm cl},1})\\ &\quad=M_F(f_{{\rm cl},2})|f_{{\rm cl},1}\rangle_{F\,F}\langle f_{{\rm cl},1}|M_F^\dagger(f_{{\rm cl},2}) .\end{aligned} \tag{2.21}$$

The coherent state associated with the classical superposition $f_{{\rm cl},2}+f_{{\rm cl},1}$ is generated by the M transformation associated with one of the classical fields ($f_{{\rm cl},2}$), acting on the coherent state associated with the other classical field ($f_{{\rm cl},1}$). A useful extension of this idea is obtained by starting with any quantum statistical state ρ_{1F} of the field, in which the (quantum statistical) average field will be denoted by

$$\langle f_F\rangle_1={\rm Tr}_F\{f_F\rho_{1F}\} , \tag{2.22}$$

and acting on this state with the unitary transformation $M_F(f_{{\rm cl},2})$. In the resulting transformed state, the average field is given by

$$\mathrm{Tr}_F\{f_F M_F(f_{\mathrm{cl},2})\rho_{1F}M_F^\dagger(f_{\mathrm{cl},2})\}=\langle f_F\rangle_1+f_{\mathrm{cl},2}\ . \tag{2.23}$$

Clearly, the transformation $M_F(f_{\mathrm{cl},2})$ superimposes the classical field $f_{\mathrm{cl},2}$ on the previously existing quantum average field $\langle f_F\rangle_1$. Note that if ρ_{1F} is *not* a coherent state, then $M_F(f_{\mathrm{cl},2})\rho_{1F}M_F^\dagger(f_{\mathrm{cl},2})$ is also *not* a coherent state.

Next, we briefly examine the time dependence of $M_F(t)$, where the explicit reference to the classical field will be omitted for simplicity. Combining (A13) and (2.13) we see that $M_F(t)$ satisfies the evolution equation

$$i\hbar\frac{\partial}{\partial t}M_F(t)=[H_{0F}(t),M_F(t)]\ . \tag{2.24}$$

Further discussions of time evolution will be simplified by the introduction of the unitary evolution operator $Q_F(t,t_0)$ for the unperturbed field, defined by[4,6]

$$i\hbar\frac{\partial}{\partial t}Q_F(t,t_0)=H_{0F}Q_F(t,t_0)\ \text{ with }\ Q_F(t_0,t_0)=1\ , \tag{2.25}$$

or equivalently by

$$Q_F(t,t_0)=\exp[-i(t-t_0)H_{0F}/\hbar]\ . \tag{2.26}$$

The solution of (2.24) can be written in various forms,

$$M_F(t)=Q_F(t,t_0)M_F(t_0)Q_F(t_0,t)\ , \tag{2.27}$$

$$M_F(t)Q_F(t,t_0)=Q_F(t,t_0)M_F(t_0)\ , \tag{2.28}$$

$$[H_{0F},M_F(t)]Q_F(t,t_0)=Q_F(t,t_0)[H_{0F},M_F(t_0)]\ . \tag{2.29}$$

Similar relations hold with $M_F^\dagger(t)$ and $M_F(t_0)$ replaced, respectively, by $M_F^\dagger(t)$ and $M_F^\dagger(t_0)$; more relations of the same type are obtained by multiplication by $Q_F(t_0,t)$ from the left and from the right and use of $Q_F(t,t_0)Q_F(t_0,t)=1_F$.

C. Disentangling the quasiclassical-field pulse from the rest of the problem

Using the naive picture for the initial condition presented at the beginning and at the end of Section II A we anticipate that the field will remain for a long time "close" to the quasiclassical state $|\alpha f_{\mathrm{cl}}(t)\rangle_F$. This suggests using the unitary transformation

$$M_S^\dagger(t)=1_A\otimes M_F^\dagger(\alpha f_{\mathrm{cl}}(t))=1_A\otimes M_F(-\alpha f_{\mathrm{cl}}(t)) \tag{2.30}$$

in the way shown by Mollow,[3] to "remove" the undisturbed coherent excitation $\alpha f_{\mathrm{cl}}(t)$ from the field [see (2.23)], hopefully leaving the field close to its ground state and, hence, easier to discuss.

The transformed quantities will be indicated by a bar above the symbols, and defined as follows: for kets and bras,

$$|\bar{t}\rangle_S=M_S^\dagger(t)\,|t\rangle_S,\quad {}_S\langle\bar{t}|={}_S\langle t|M_S(t)\ , \tag{2.31}$$

for operators involving a single time t or no explicit time dependence,

$$\bar{R}_S(t)=M_S^\dagger(t)R_S(t)M_S(t)\ , \tag{2.32}$$

and for operators connecting two times (e.g., evolution operators),

$$\bar{U}_S(t,t_0)=M_S^\dagger(t)U_S(t,t_0)M_S(t_0)\ , \tag{2.33}$$

with $M_S(t)$ given by (2.30). Whenever a relation between the original quantities involves *neither time derivative nor time integration,* it holds also between the corresponding transformed quantities. For instance, if $\rho_S(t)=U_S(t,t_0)\rho_S(t_0)U_S(t_0,t)$, then $\bar{\rho}_S(t)=\bar{U}_S(t,t_0)\bar{\rho}_S(t_0)\bar{U}_S(t_0,t)$.

The original equation of motion

$$i\hbar\frac{\partial}{\partial t}\rho_S(t)=[H_S,\rho_S(t)] \tag{2.34}$$

for $\rho_S(t)$ leads to the transformed equation of motion

$$i\hbar\frac{\partial}{\partial t}\bar{\rho}_S(t)=[H_S+\lambda(\bar{V}_S(t)-V_S),\bar{\rho}_S(t)] \tag{2.35}$$

for $\bar{\rho}_S(t)$ [see (2.24), (2.1), (2.32)].

In many respects, the "bar" transformation introduced here is analogous to the standard transformation to an interaction representation used to remove parts of the Hamiltonian from the transformed equation of motion. However, there are also significant differences: the bar transformation removes the coherent excitation of the field from the transformed state of the system, but the corresponding unitary operator $M_S(t)$ is *not* associated with a large part of the Hamiltonian evolution operator, hence no large part of the Hamiltonian is absent from (2.35).

When the atom-field coupling is given by (2.4), the supplementary term $\lambda[\bar{V}_S(t)-V_S]$ which appears in the generator of the motion of $\bar{\rho}_S(t)$ has the very simple form [see (2.19) and (2.4)] of a *time-dependent* operator acting in a nontrivial way on the bare atom only,

$$\begin{aligned}\lambda(\bar{V}_S(t)-V_S)&=\lambda\alpha\sum_{i,j}\{\,|\,i\rangle_{A\,A}\langle j\,|\,V_{ij}(\mathbf{r}_A,t)\}\otimes 1_F\\&=\lambda[\bar{V}(t)-V]_A\otimes 1_F\ ,\end{aligned} \tag{2.36}$$

where the complex functions of time $V_{ij}(\mathbf{r}_A,t)$ are given by

$$V_{ij}(\mathbf{r}_A,t)=\sum_k[V_{kij}^*(\mathbf{r}_A)\alpha_k(t)+V_{kji}(\mathbf{r}_A)\alpha_k^*(t)]\ , \tag{2.37}$$

and the notation $[\bar{V}(t)-V]_A$ indicates an operator proportional to α and acting in the state space of the atom only. The operator $\lambda[\bar{V}(t)-V]_A$ is exactly the time-dependent term which would be introduced to describe the atom-field interaction in the conventional semiclassical description in terms of a quantized atom and a classical field. As an example, in the linearized electric dipolar approximation (2.5), one has

$$\lambda[\bar{V}_{\mathrm{ed}}(\mathbf{r}_A,t)-V_{\mathrm{ed}}(\mathbf{r}_A)]_A=-\lambda\boldsymbol{\mu}_{eA}\cdot\mathbf{E}_{\mathrm{cl}}(\mathbf{r}_A,t)\ . \tag{2.38}$$

If the atom-field coupling is nonlinear in the field, the operator $\lambda[\bar{V}_S(t)-V_S]$ is slightly more complicated than (2.36), and is no longer the semiclassical atom-field coupling. For instance, in atomic spectroscopy, the term quadratic in the field has a coefficient of the order of the fine-structure constant whereas the term linear in the field is of the order of the square root of this small constant. Using λ to denote this square root, we can write the atom-field coupling under the form $\lambda V_S=\lambda V_{S,1}+\lambda^2 V_{S,2}$,

where $\lambda V_{S,1}$ is the term linear in the field discussed above and $\lambda^2 V_{S,2}$ is quadratic in the field. The contribution of the term $\lambda^2 V_{S,2}$ to $\lambda[\bar{V}_S(t)-V_S]$ involves terms linear in the field operators a_k and $a_k^\dagger$, with a coefficient $\lambda(\lambda\alpha)$, in addition to terms of the semiclassical type (i.e., acting nontrivially on the bare atom only), with a coefficient proportional to $(\lambda\alpha)^2$.

As no approximation is involved in going over from (2.34) to (2.35), the generator of the motion of $\bar{\rho}_S(t)$ still contains the *complete* quantum-mechanical atom-field interaction problem, although the semiclassical coupling of the atom with a time-dependent classical field appears explicitly in (2.35).

During all time periods in which the unperturbed classical field $f_{\rm cl}(\mathbf{r},t)$ is exactly zero in the region of space occupied by the atom, the "supplementary term" $\lambda[\bar{V}_S(t)-V_S]$ given by (2.36) is also exactly zero. This property still holds for couplings which are nonlinear in the field. Hence, during these time periods, $\bar{\rho}_S(t)$ evolves exactly as if the (perhaps large) quasiclassical excitation $\alpha f_{\rm cl}(\mathbf{r},t)$ of the field was absent, and $f_{\rm cl}(\mathbf{r},t)$ itself appears only in the unitary transformation $M_S(t)$ which leads back from $\bar{\rho}_S(t)$ to $\rho_S(t)$.

The standard basis for the present problem is obtained by tensorial multiplication of the basis of eigenstates of the bare-atom Hamiltonian by a basis of eigenstates of the energy for each normal mode of the field,

$$|i,\{n_k\}\rangle_S = |i\rangle_A \otimes \prod_k |n_k\rangle_k \ . \tag{2.39}$$

When matrix elements of "bar-transformed" operators [see (2.32)] are needed in this $\{i,\{n_k\}\}$ basis, the following identity may be useful:

$$\begin{aligned} {}_S\langle i',\{n_k'\}|\bar{R}_S(t)|i,\{n_k\}\rangle_S &= {}_S\langle i',\{n_k'\}|[M_S^\dagger(t)R_S(t)M_S(t)]|i,\{n_k\}\rangle_S \\ &= [{}_S\langle i',\{n_k'\}|M_S^\dagger(t)]R_S(t)[M_S(t)|i,\{n_k\}\rangle_S] \ , \end{aligned} \tag{2.40}$$

where the new basis formed by the kets $\{M_S(t)|i,\{n_k\}\rangle_S\}$ is the extension to the case of atom *and* field of the basis discussed from (A17)–(A19).

D. Realistic, stable initial conditions

For all times t_i during the initial idle period, the absence of overlap between the atom and the classical field pulse $f_{\rm cl}(\mathbf{r},t)$ implies that the term $\lambda[\bar{V}_S(t)-V_S]$ in (2.35) is exactly zero, hence the equation of motion for $\bar{\rho}_S(t_i)$ is

$$i\hbar\frac{\partial}{\partial t_i}\bar{\rho}_S(t_i)=[H_S,\bar{\rho}_S(t_i)] \ . \tag{2.41}$$

When the initial idle period is also one of rest, during which the *only* thing that "happens" is the propagation of the quasiclassical pulse excitation of the field, then $\bar{\rho}_S(t_i)$ is time independent during this period.

The simplest stationary solution of (2.41) is

$$\bar{\rho}_S(t_i)=|G\rangle_S\,{}_S\langle G| \ , \tag{2.42}$$

where $|G\rangle_S$ is the exact ground state of the complete Hamiltonian H_S for the coupled atom-field system. This ground state is close to the tensorial product of the ground state $|1\rangle_A$ of the bare atom with the ground state $|0\rangle_F$ of the unperturbed field. More accurate expressions for $|G\rangle_S$ can be obtained by perturbation techniques. Going back from $\bar{\rho}_S(t_i)$ to $\rho_S(t_i)$, we can write the initial condition corresponding to (2.42) under the form

$$\rho_S(t_i)=M_S(f_{\rm cl}(t_i))|G\rangle_S\,{}_S\langle G|M_S^\dagger(f_{\rm cl}(t_i)) \ , \tag{2.43}$$

which describes a situation in which the field is close to the quasiclassical excitation $|f_{\rm cl}(t_i)\rangle_F\,{}_F\langle f_{\rm cl}(t_i)|$ and the atom is almost in its bare ground state; the only deviations are the direct radiative effects of the atom-field coupling which already exist in the absence of the approaching quasiclassical excitation. Expression (2.43) for $\rho_S(t_i)$ is an *exact* solution of the equation of motion (2.34) during the initial idle period, and the strong time dependence of $\rho_S(t_i)$ is quite natural because it describes the propagation of the quasiclassical excitation of the field.

An atom can be prepared in the stable initial condition described by (2.42) and (2.43) by first being left alone without any irradiation for a period much longer than any of its radiative lifetimes, before being subjected to the coherent radiation which will cause the interesting dynamical processes.

If the whole experiment, including the long preparation period, takes place in the presence of blackbody radiation at temperature T, then the relevant stationary solution of (2.41) is the equilibrium density operator

$$\bar{\rho}_S(t_i)=[1/Z(T)]\exp(-H_S/k_BT) \ , \tag{2.44}$$

where k_B is Boltzmann's constant and the partition function $Z(T)$ is given by $Z(T)=\mathrm{Tr}_S\{\exp(-H_S/k_BT)\}$, hence the state of the system during the initial idle period is given by

$$\rho_S(t_i)= M_S(f_{\rm cl}(t_i))(1/Z(T)) \times\exp(-H_S/k_BT)M_S^\dagger(f_{\rm cl}(t_i)) \ . \tag{2.45}$$

E. Removing H_{0A} and H_{0F} from the equation of motion

For the rest of this paper, it will be convenient to remove the unperturbed Hamiltonian $(H_{0A}\otimes 1_F)+(1_A\otimes H_{0F})$ from the equation of motion by going to the usual "interaction representation" based on the unitary operator

$$Q_S(t)=Q_A(t)\otimes Q_F(t) \ , \tag{2.46}$$

where[4]

$$Q_A(t)=\exp[-i(t-t_0)H_{0A}/\hbar]$$

and[6,7]

$$Q_F(t)=\exp[-i(t-t_0)H_{0F}/\hbar] \ . \tag{2.47}$$

The new transformation will lead from the bar quantities to the "tilde" quantities, much in the same way as (2.31)–(2.33) led from the original quantities to bar quantities. For instance, the transformed density operator is

$$\tilde{\rho}_S(t)=Q_S^\dagger(t)\bar{\rho}_S(t)Q_S(t)$$
$$=Q_S^\dagger(t)M_S^\dagger(t)\rho(t)M_S(t)Q_S(t) \quad (2.48)$$

and obeys the transformed equation of motion

$$i\hbar\frac{\partial}{\partial t}\tilde{\rho}_S(t)=[\lambda\tilde{V}_S(t),\tilde{\rho}_S(t)]$$
$$=[\{\lambda Q_S^\dagger(t)V_SQ_S(t)$$
$$+\lambda Q_S^\dagger(t)(\bar{V}_S(t)-V_S)Q_S(t)\},\tilde{\rho}_S(t)] , \quad (2.49)$$

from which H_{0A} and H_{0F} have been removed. The transformed initial condition (2.42) during the whole idle period is

$$\tilde{\rho}_S(t_i)=Q_S^\dagger(t_i)[\,|\,G\rangle_{S\,S}\langle G\,|\,]Q_S(t_i) . \quad (2.50)$$

Both terms in the curly brackets of (2.49) are time dependent. As it does not involve M_S, the first term is independent of the state of the field. The second one is different from zero only during the overlap of the classical-field pulse $f_{\rm cl}(\mathbf{r},t)$ with the atom.

III. SHORT TIMES, WEAK-COUPLING λ, ARBITRARY EXCITATION STRENGTH $\lambda\alpha$: A CONVENTIONAL SCHEME OF SUCCESSIVE APPROXIMATIONS

In the remaining part of this paper we will discuss a successive approximation scheme, suitable for short times (i.e., much shorter than the radiative lifetime of the atom), weak coupling λ, but arbitrary excitation strength $\lambda\alpha$. In order to avoid unessential complications we shall consider a coupling linear in the field and the simple initial condition (2.42) involving the ground state of the atom-field system. The results for a two-level atom will be given, and further discussed in Appendix B.

A. The traditional classical field approximation (Bloch equations without relaxation)

In the traditional classical-field approximation the effect of the classical field on the atom is taken into account as a time-dependent contribution to the Hamiltonian of the atom, whereas all effects of the atom on the field are ignored: Lamb shift, spontaneous emission, any reaction of the atom on the exciting field. In the present model, with linear atom-field coupling, this corresponds to keeping the term $\lambda[\bar{V}_S(t)-V_S]$, which is proportional to $\lambda\alpha$, in the generator of the motion of $\bar{\rho}_S(t)$ or $\tilde{\rho}_S(t)$ [see (2.35) and (2.49)] and dropping the isolated term involving λV_S, which is independent of the magnitude α of the classical field. In this approximation, taking into account (2.36), where $[\bar{V}(t)-V]_A$ is an operator acting only in the atom space, (2.49) becomes

$$i\hbar\frac{\partial}{\partial t}\tilde{\rho}_S^B(t)=[Q_A^\dagger(t)\lambda(\bar{V}(t)-V)_AQ_A(t)\otimes 1_F,\tilde{\rho}_S^B(t)] , \quad (3.1)$$

where the superscript B stands for "Bloch equations without relaxation" (a similar notation will be used for average values of observables) and the stable initial condition before the atom–classical-field overlap is given by

$$\tilde{\rho}_S^B(t_i)=\bar{\rho}_S^B(t_i)=(\,|\,1\rangle_{A\,A}\langle 1\,|\,)\otimes(\,|\,0\rangle_{F\,F}\langle 0\,|\,) . \quad (3.2)$$

The above two equations are derived from the exact equations (2.34) and (2.43) by an *ad hoc* procedure, one consequence of which is to make the generator of the motion of $\rho_S^B(t)$ time dependent so that the average "energy" evaluated with $\rho_S^B(t)$ is no longer an invariant and conservation of total energy loses its role as a simple, general property. As we shall show in Sec. III B a more acceptable approach is to use a perturbation scheme in which λ tends to zero and α to infinity in a correlated way such that the excitation strength $\lambda\alpha$ remains finite and constant. Such a systematic procedure indeed leads to very satisfactory results in its limited range of validity (i.e., short times). Nevertheless, we shall briefly discuss the solution of (3.1) and (3.2) because this provides a satisfactory solution for the evolution of the atom reduced density operator and useful tools for further calculations.

Like the generator of the motion in (3.1), the evolution operator $B_S(t,t_0)$, such that $\tilde{\rho}_S^B(t)=B_S(t,t_0)\tilde{\rho}_S^B(t_0)B_S(t_0,t)$, is the tensorial product of an operator acting only on the atom by the unit operator 1_F for the field

$$B_S(t,t_0)=B_A(t,t_0)\otimes 1_F , \quad (3.3)$$

where[4]

$$i\hbar\frac{\partial}{\partial t}B_A(t,t_0)=\{Q_A^\dagger(t)\lambda[\bar{V}(t)-V]_AQ_A(t)\}B_A(t,t_0) \quad (3.4)$$

and $B_A(t_0,t_0)=1_A$. Furthermore, starting with the factorized initial situation (3.2), the density operator will remain factorized at all times,

$$\tilde{\rho}_S^B(t)=\tilde{\rho}_A^B(t)\otimes(\,|\,0\rangle_{F\,F}\langle 0\,|\,)$$
$$=[Q_A^\dagger(t)\rho_A^B(t)Q_A(t)]\otimes(\,|\,0\rangle_{F\,F}\langle 0\,|\,)$$

and

$$i\hbar\frac{\partial}{\partial t}\tilde{\rho}_A^B(t)=[Q_A^\dagger(t)\lambda(\bar{V}(t)-V)_AQ_A(t),\tilde{\rho}_A^B(t)]$$

and

$$\tilde{\rho}_A^B(t_i)=|\,1\rangle_{A\,A}\langle 1\,| . \quad (3.5)$$

The tilde transformation used here for operators acting in the bare-atom state space is defined by $\tilde{A}_A(t)=Q_A^\dagger(t)A_A(t)Q_A(t)$.

In most cases of practical interest in pulse spectroscopy, the generator of the motion in (3.4) does not commute with itself at different times, so that solving this equation is not an easy task. This generator of the motion is Hermitian at all times, hence $B_A(t,t_0)$ is unitary and has all the usual properties of evolution operators. Useful tools for approximate evaluations of $B_A(t,t_0)$ are provided by numerical methods (in the case of finite time intervals and atom state space of finite dimension) and by the Magnus transformation.[8] The simple case of a two-level atom is discussed in Appendix B.

B. Iterative procedure for weak coupling λ, short times, and arbitrary excitation strength $\lambda\alpha$

The solution of an equation of the type (2.34), (2.35), or (2.49) by the conventional short-time expansion limited to

a few terms is a useful approximation only for time intervals during which the corresponding density operator never deviates much from its initial condition. For (2.49) and (2.50), in which the generator of the motion still contains a term proportional to the excitation strength $\lambda\alpha$, this restricts the usefulness of such an expansion to irradiations which have little effect on the state of the atom (hence to small values of $\lambda\alpha$) even for pulses much shorter than the radiative lifetime of the atom. This limitation to very weak irradiation pulses can be overcome by a further time-dependent unitary transformation which removes the atom—classical-field coupling from the generator of the motion. A suitable unitary operator $B_S(t)$ is constructed from the "single-time" version[6,7] $B_A(t)$ of the evolution operator defined by (3.4),

$$B_S(t)=B_A(t)\otimes 1_F \ , \tag{3.6}$$

where[4]

$$i\hbar\frac{\partial}{\partial t}B_A(t)=\{Q_A^\dagger(t)\lambda[\bar{V}(t)-V]_A Q_A(t)\}B_A(t) \tag{3.7}$$

with $B_A(t_0)=1_A$.

The transformed quantities will be denoted by a caret above the symbol. For instance,

$$\begin{aligned}\hat{\rho}_S(t)&=B_S^\dagger(t)\bar{\rho}_S(t)B_S(t)\\&=B_S^\dagger(t)Q_S^\dagger(t)\bar{\rho}_S(t)Q_S(t)B_S(t)\\&=B_S^\dagger(t)Q_S^\dagger(t)M_S^\dagger(t)\rho_S(t)M_S(t)Q_S(t)B_S(t) \ ,\\ \hat{V}_S(t)&=B_S^\dagger(t)Q_S^\dagger(t)\bar{V}_S(t)Q_S(t)B_S(t) \ .\end{aligned} \tag{3.8}$$

With the linear atom-field coupling (2.4), $\hat{\rho}_S(t)$ obeys the equation of motion [see (2.49)]

$$i\hbar\frac{\partial}{\partial t}\hat{\rho}_S(t)=[\lambda B_S^\dagger(t)Q_S^\dagger(t)V_S Q_S(t)B_S(t),\hat{\rho}_S(t)] \ . \tag{3.9}$$

Note that the generator of the motion in (3.9) is *not* $\lambda\hat{V}_S(t)$. For a reference time t_0 for $B_S(t)$ in the initial idle period, $B_A(t_i)=1_A$ and the stable initial condition (2.42), (2.50) has the simple form

$$\hat{\rho}_S(t_i)=Q_S^\dagger(t_i)\bar{\rho}_{GS}Q_S(t_i)=Q_S^\dagger(t_i)\,|\,G\rangle_{S\,S}\langle G\,|\,Q_S(t_i) \ . \tag{3.10}$$

A strong irradiation of the atom (i.e., $\lambda\alpha$ large) causes a fast time dependence of the unitary operator $B_S(t)$ during the overlap between the irradiation pulse and the atom, but the large parameter α does *not* appear in a multiplicative way in the generator of the motion of $\hat{\rho}_S(t)$. Hence, this generator of the motion is of the order of magnitude of the strength λ of the atom-field coupling at all times, and a short-time expansion of the solution of (3.9), limited to a few terms, will be satisfactory at least for time intervals Δt such that $\lambda\Delta t$ is small. If no strong irradiation takes place during the time interval, it is well known that this range of validity extends to Δt much shorter than the radiative lifetime, i.e., $\lambda^2\Delta t$ small.

In the conventional technique for short-time approximation, the solution of (3.9) and (3.10) is a formal series expansion in the (small) strength λ of the atom-field coupling,

$$\hat{\rho}_S(t)=\hat{\rho}_S^{(0)}+\lambda\hat{\rho}_S^{(1)}(t)+\lambda^2\hat{\rho}_S^{(2)}(t)+\cdots \ , \tag{3.11}$$

with [see (2.42)]

$$\bar{\rho}_{GS}=|\,G\rangle_{S\,S}\langle G\,|=\bar{\rho}_{GS}^{(0)}+\lambda\bar{\rho}_{GS}^{(1)}+\lambda^2\bar{\rho}_{GS}^{(2)}+\cdots \ . \tag{3.12}$$

The usual perturbative expansion of the ground state $|\,G\rangle_S$ of the complete Hamiltonian H_S in powers of λ, assuming a nondegenerate ground state $|\,1\rangle_A$ for the bare-atom Hamiltonian, leads to

$$\bar{\rho}_{GS}^{(0)}=(\,|\,1\rangle_{A\,A}\langle 1\,|\,)\otimes(\,|\,0\rangle_{F\,F}\langle 0\,|\,) \ , \tag{3.13}$$

and to a first-order term

$$\bar{\rho}_{GS}^{(1)}=\bar{\rho}_{GS}^{(0)}(W_S^{(1)})^\dagger+W_S^{(1)}\bar{\rho}_{GS}^{(0)} \ , \tag{3.14}$$

where

$$W_S^{(1)}=\sum_k\sum_j\frac{-V_{k1j}}{\hbar(\omega_j-\omega_1+\omega_k)}\,|\,j\rangle_{A\,A}\langle 1\,|\otimes a_k^\dagger \ . \tag{3.15}$$

Combining (3.9) to (3.13) we obtain

$$\hat{\rho}_S^{(0)}=(\,|\,1\rangle_{A\,A}\langle 1\,|\,)\otimes(\,|\,0\rangle_{F\,F}\langle 0\,|\,) \tag{3.16}$$

and, for $n\geq 1$, the recursion relations

$$i\hbar\frac{\partial}{\partial t}\hat{\rho}_S^{(n)}(t)=[B_S^\dagger(t)Q_S^\dagger(t)V_S Q_S(t)B_S(t),\hat{\rho}_S^{(n-1)}(t)] \ , \tag{3.17}$$

which can also be written in integral form as

$$\hat{\rho}_S^{(n)}(t)=Q_S^\dagger(t_i)\bar{\rho}_{GS}^{(n)}Q_S(t_i)+(1/i\hbar)\int_{t_i}^{t}[B_S^\dagger(t')Q_S^\dagger(t')V_S Q_S(t')B_S(t'),\hat{\rho}_S^{(n-1)}(t')]dt' \ , \tag{3.18}$$

where t_i is chosen in the initial idle period. Using (2.41) and (3.10), one can easily verify, through derivation of (3.18) with respect to t_i, that $\hat{\rho}_S^{(n)}$ is a function of t, but *not* of t_i.

Going back to the tilde version, we have $\tilde{\rho}_S^{(0)}(t)=B_S(t)\hat{\rho}_S^{(0)}B_S^\dagger(t)=\tilde{\rho}_S^B(t)$ [see (3.1)–(3.5)]. Hence, the starting point $\rho_S^{(0)}$ of the proposed iterative procedure is the traditional classical-field approximation, and higher-order terms in (3.11) can be seen as correcting for the error made by not properly treating the field quantum mechanically in the first place.

When $\bar{\rho}_S(t)$ is known, average values of observables can be evaluated from the relation

$$\langle C_S\rangle(t)=\mathrm{Tr}_S\{C_S\rho_S(t)\}=\mathrm{Tr}_S\{\hat{C}_S(t)\hat{\rho}_S(t)\} \ , \tag{3.19}$$

and may also be expanded in powers of λ,

$$\langle C\rangle(t)=\sum_m\lambda^m\langle C\rangle^{(m)}(t) \tag{3.20}$$

with m taking integer values. However, great care must be exercised in going over from the formal series expansion (3.11) of $\hat{\rho}_S(t)$ to an expansion of the type (3.20) because the large parameter α, which appears in many caret-transformed observables combines with the small parameter λ to give the constant excitation strength $\lambda\alpha$.

IV. ENERGY BALANCE UP TO ORDER λ

The Hamiltonian of our model, as given by (2.1), is time independent, hence the probability distribution in total energy is time independent and, in particular, the average total energy

$$\langle H_S\rangle(t)=\langle 1_A\otimes H_{0F}\rangle(t)+\langle H_{0A}\otimes 1_F\rangle(t)+\langle V_S\rangle(t) \tag{4.1}$$

is a constant. As an illustration of the use of the iterative scheme, we now evaluate in some detail all the traces and integrals needed to check that

$$\langle H_S\rangle^{(m)}(t)=\langle 1_A\otimes H_{0F}\rangle^{(m)}(t)+\langle H_{0A}\otimes 1_F\rangle^{(m)}(t)+\langle V_S\rangle^{(m)}(t) \tag{4.2}$$

is indeed exactly constant for all values of m up to $m=1$, for a linear atom-field coupling.

In these calculations we make extensive use of the cyclic invariance of the trace and of the factorization properties (2.30) for $M_S(t)$, (2.46) for $Q_S(t)$, (3.3) for $B_S(t)$, and (2.36) for $\bar{V}_S(t)-V_S$. One should also keep in mind that the single-time operators $Q_S(t)$ and $B_S(t)$ still involve t_0 parametrically and that t_0 and t_i are both assumed to be in the initial idle period. Moreover, in the calculations involving H_{0F}, we have repeatedly used the commutation properties of M_F, Q_F, and H_{0F} which are given in Appendix C 1.

We begin with $\langle H_{0A}\otimes 1_F\rangle(t)$. The contribution K_1 arising from $\hat{\rho}_S^{(0)}$,

$$\begin{aligned}K_1&=\mathrm{Tr}_S\{B_S^\dagger(t)Q_S^\dagger(t)M_S^\dagger(t)(H_{0A}\otimes 1_F)M_S(t)Q_S(t)B_S(t)(\,|\,1\rangle_{A\,A}\langle 1\,|\otimes|\,0\rangle_{F\,F}\langle 0\,|\,)\}\\&=\mathrm{Tr}_S\{H_{0A}Q_A(t)B_A(t)\,|\,1\rangle_{A\,A}\langle 1\,|B_A^\dagger(t)Q_A^\dagger(t)\otimes|\,0\rangle_{F\,F}\langle 0\,|\,\}\\&=\mathrm{Tr}_A\{H_{0A}\rho_A^B(t)\}=\langle H_{0A}\rangle^B(t)\,,\end{aligned} \tag{4.3}$$

is the average atomic Hamiltonian in the traditional classical-field approximation. The contribution arising from $\hat{\rho}_S^{(1)}(t)$ is identically zero because $\hat{\rho}_S^{(1)}(t)$ is linear in a_k or $a_k^\dagger$, and the caret version of $\langle H_{0A}\otimes 1_F\rangle$ acts on the field as 1_F. Contributions from higher-order terms $\hat{\rho}_S^{(n)}(t)$ will be of order λ^n with $n>1$. As a conclusion,

$$\begin{aligned}&\lambda^m\langle H_{0A}\otimes 1_F\rangle^{(m)}(t)=0,\quad\text{for } m<0\\&\langle H_{0A}\otimes 1_F\rangle^{(0)}(t)=\langle H_{0A}\rangle^B(t)\,,\\&\lambda\langle H_{0A}\otimes 1_F\rangle^{(1)}(t)=0\,.\end{aligned} \tag{4.4}$$

Next we evaluate $\langle\lambda V_S\rangle(t)$. The contribution K_2 arising from $\hat{\rho}_S^{(0)}$ (taking into account that V_S is linear in a_k or $a_k^\dagger$),

$$\begin{aligned}K_2&=\mathrm{Tr}_S\{\lambda\hat{V}_S\hat{\rho}_S^{(0)}\}\\&=\mathrm{Tr}_S\{B_S^\dagger(t)Q_S^\dagger(t)\{\lambda[\bar{V}_S(t)-V_S]+\lambda V_S\}Q_S(t)B_S(t)(\,|\,1\rangle_{A\,A}\langle 1\,|\otimes|\,0\rangle_{F\,F}\langle 0\,|\,)\}\\&=\mathrm{Tr}_S\{\lambda[\bar{V}(t)-V]_A\rho_A^B(t)\otimes|\,0\rangle_{F\,F}\langle 0\,|\,\}=\langle\lambda[\bar{V}(t)-V]_A\rangle^B(t)\,,\end{aligned} \tag{4.5}$$

is the average atom-field coupling in the traditional classical-field approximation. This is a quantity of order $\lambda\alpha$, hence a contribution to $\langle V_S\rangle^{(0)}(t)$. The evaluation of contributions to $\langle\lambda V_S\rangle(t)$ arising from further terms $\rho_S^{(n)}(t)$ is simplified by writing $\lambda\bar{V}_S(t)$ again as $\lambda[\bar{V}_S(t)-V_S]+\lambda V_S$. For n odd, the contribution is only from λV_S and is of order λ^{n+1}, whereas for n even the contribution is from $\lambda[\bar{V}_S(t)-V_S]$ and is of order $(\lambda\alpha)\lambda^n$. As a conclusion,

$$\begin{aligned}&\lambda^m\langle\lambda V_S\rangle^{(m)}(t)=0,\quad\text{for } m<0\\&\langle\lambda V_S\rangle^{(0)}(t)=\langle\lambda[\bar{V}(t)-V]_A\rangle^B(t)\,,\\&\lambda\langle\lambda V_S\rangle^{(1)}(t)=0\,.\end{aligned} \tag{4.6}$$

Finally, we evaluate $\langle 1_A\otimes H_{0F}\rangle(t)$. The contribution K_3 from $\hat{\rho}_S^{(0)}$,

$$\begin{aligned}K_3&=\mathrm{Tr}_S\{B_S^\dagger(t)Q_S^\dagger(t)M_S^\dagger(t)(1_A\otimes H_{0F})M_S(t)Q_S(t)B_S(t)(\,|\,1\rangle_{A\,A}\langle 1\,|\otimes|\,0\rangle_{F\,F}\langle 0\,|\,)\}\\&=\mathrm{Tr}_S\{\,|\,1\rangle_{A\,A}\langle 1\,|\otimes H_{0F}M_F(t)\,|\,0\rangle_{F\,F}\langle 0\,|M_F^\dagger(t)\}=\alpha^2\sum_k\hbar\omega_k\,|\,\alpha_k\,|^2\,,\end{aligned} \tag{4.7}$$

is exactly the average energy which the field would have if the atom was absent. In the present notation this term in α^2 appears as "of order λ^{-2}," hence a contribution to $\langle 1_A \otimes H_{0F} \rangle^{(-2)}(t)$.

The contribution K_4 to $\langle 1_A \otimes H_{0F} \rangle(t)$ from the initial value $\hat{\rho}_S^{(1)}(t_i)$ in (3.18) for $\hat{\rho}_S^{(1)}(t)$ is

$$K_4 = \mathrm{Tr}_S \{ B_S^\dagger(T) Q_S^\dagger(t) M_S^\dagger(t) (1_A \otimes H_{0F}) M_S(t) Q_S(t) B_S(t) \times Q_S^\dagger(t_i) \{ |1\rangle_{A\,A}\langle 1| \otimes |0\rangle_{F\,F}\langle 0| (W_S^{(1)})^\dagger + \mathrm{H.c.} \} Q_S(t_i) \} , \tag{4.8}$$

where $W_S^{(1)}$ is given by (3.15). As shown in Appendix C 2 this can be written as

$$K_4 = \mathrm{Tr}_S \left\{ \sum_k \hbar\omega_k [a_k^\dagger + \alpha_k^*(t_i)][a_k + \alpha_k(t_i)] \times [|1\rangle_{A\,A}\langle 1| \otimes |0\rangle_{F\,F}\langle 0| (W_S^{(1)})^\dagger + \mathrm{H.c.}] \right\}$$

$$= \lambda\alpha \sum_k [-V_{k11}\alpha_k^*(t_i) + \mathrm{c.c.}]$$

$$= -\langle \lambda [\bar{V}(t_i) - V]_A \rangle^B(t_i) . \tag{4.9}$$

This is minus the average atom-field interaction energy at time t_i in the traditional classical-field approximation, hence

$$K_4 = 0 . \tag{4.10}$$

The contribution K_5 to $\langle 1_A \otimes H_{0F} \rangle(t)$ from the time integral in (3.18) for $\hat{\rho}_S^{(1)}(t)$ is given by

$$K_5 = (1/i\hbar) \int_{t_i}^t dt' \mathrm{Tr}_S \{ B_S^\dagger(t) Q_S^\dagger(t) M_S^\dagger(t) (1_A \otimes H_{0F}) M_S(t) Q_S(t) B_S(t) \times [B_S^\dagger(t') Q_S^\dagger(t') \lambda V_S Q_S(t') B_S(t'), |1\rangle_{A\,A}\langle 1| \otimes |0\rangle_{F\,F}\langle 0|] \} . \tag{4.11}$$

The integrand in the integral over t' can be recast as a sum of derivatives with respect to t', the equations of motion of $M_F(t)$ and $\tilde{\rho}_A^B(t)$ [see (2.24) and (3.5)] being used to this aim. As (2.24) involves $[H_{0F}, M_F(t)]$, a first very useful step is to replace $H_{0F}M_F(t)$ by $M_F(t)H_{0F} + [H_{0F}, M_F(t)]$ in (4.11). The contribution of $M_F(t)H_{0F}$ to the trace is zero because, for this term, the M_F operators cancel and V_S remains the only nondiagonal field operator in a representation which diagonalizes H_{0F}. After some algebra (see Appendix C 3), one obtains

$$K_5 = \int_{t_i}^t dt' \mathrm{Tr}_S \{ B_A(t') |1\rangle_{A\,A}\langle 1| B_A^\dagger(t') \otimes M_F(t_0) |0\rangle_{F\,F}\langle 0| M_F^\dagger(t_0) Q_S^\dagger(t') \frac{\partial}{\partial t'} [M_S(t') \lambda V_S M_S^\dagger(t') + \lambda V_S] Q_S(t') \} . \tag{4.12}$$

The next step is to introduce the operator $\bar{V}_S(t) - V_S$ with the help of relation (2.30) under the form $M_F^\dagger(\{\alpha\alpha_k(t)\}) = M_F(\{-\alpha\alpha_k(t)\})$, which implies that interchanging M_F and $M_F^\dagger$ is equivalent to a change of sign of the parameter α. After some algebra (see Appendix C 4), one then obtains

$$K_5 = -\int_{t_i}^t dt' \frac{\partial}{\partial t'} \langle \bar{V}(t') - V \rangle^B(t') - \int_{t_i}^t dt' \frac{\partial}{\partial t'} \langle H_{0A} \rangle^B(t') . \tag{4.13}$$

The order in λ of the contributions to $\langle 1_A \otimes H_{0F} \rangle(t)$ arising from a general $\hat{\rho}_S^{(n)}(t)$ can be evaluated easily by noting that $M_F(t)H_{0F}M_F^\dagger(t) = \sum_k \hbar\omega_k [a_k^\dagger a_k + \alpha(\alpha_k^* a_k + \alpha_k a_k^\dagger) + \alpha^2 |\alpha_k|^2]$. Contributions in α^2 arise only from $n=0$ because $\hat{\rho}_S^{(n)}(t)$ is traceless for $n>0$. For n odd, the contribution is only from the term in α and is of order λ^{n-1}. For n even (larger than zero), the contribution is only from the term $a_k^\dagger a_k$ and is of order λ^n. As a conclusion,

$$\begin{aligned} &\lambda^{-2}\langle 1_A \otimes H_{0F} \rangle^{(-2)}(t) = \alpha^2 \sum_k \hbar\omega_k |\alpha_k|^2 , \\ &\lambda^{-1}\langle 1_A \otimes H_{0F} \rangle^{(-1)}(t) = 0 , \\ &\langle 1_A \otimes H_{0F} \rangle^{(0)}(t) = +\langle H_{0A} \rangle^B(t_i) - \langle H_{0A} \rangle^B(t) - \langle [\bar{V}(t) - V]_A \rangle^B(t) , \\ &\lambda\langle 1_A \otimes H_{0F} \rangle^{(1)}(t) = 0 . \end{aligned} \tag{4.14}$$

Collecting relations (4.4), (4.6), and (4.14), we can easily check that the average value of the energy of the atom-field system is indeed *exactly* time independent at each order in λ up to the highest order (i.e., $m=1$) for which detailed calculations are shown. Of course, this detailed verification of (4.2) provides only a check of the soundness of the model, iterative scheme and series expansions, and *not* a check about the general features of quantum mechanics. However, the way in which various contributions to total energy conspire to keep it constant is not trivial and will now be discussed briefly.

The leading term in the series expansion of the average energy in powers of the small strength λ of the atom-field coupling is the (time-independent) unperturbed field energy $\alpha^2 \sum_k \hbar\omega_k |\alpha_k|^2$. The fact that this term is of order λ^{-2} is quite natural in the present approximation scheme in which the small λ and the large α are related by the requirement that the strength $\lambda\alpha$ of the atom-field coupling is a constant.

The next nontrivial contributions to the average energy are the terms of order λ^0, i.e., independent of λ. In a typical experiment, the coupling of the atom with the irradiation field is much smaller than the bare-atom Hamiltonian; more precisely, $(1/\hbar)\langle [\bar{V}(t) - V]_A \rangle^B(t)$ is much smaller than the Bohr atomic frequencies $\omega_1 - \omega_j$. As a consequence, for irradiations which strongly affect the state of the atom (i.e., the atom reduced density operator), the contributions of order λ^0 to the average energy involve terms of two different orders of magnitudes. The larger terms $\langle H_{0A} \rangle^B$, involving the Bohr frequencies, corre-

spond to the idea that any energy gained by the bare atom is exactly taken from the field energy. The smaller terms $\langle[\bar{V}(t)-V]_A\rangle^B$, in $\lambda\alpha$, show that the same idea also holds exactly for the atom-field coupling energy, also at order λ^0, of course.

Finally, we emphasize that the relevant contributions of order λ^0 to the average free-field energy $\langle 1_A\otimes H_{0F}\rangle$ arise from the term $\hat{\rho}_S^{(1)}$, of *first order in* λ in the power expansion of the transformed density operator. This is clearly due to the presence of multiplicative factors α in the transformed version (C 2) of the free-field Hamiltonian.

V. SPECTRAL DISTRIBUTION OF THE ENERGY EXCHANGED BETWEEN THE ATOM AND THE COHERENT FIELD PULSE, UP TO ORDER λ

A. Frequency distributions

In order to display frequency as the essential parameter, and ignore the detailed mode structure of the field as much as possible, we introduce a time-dependent frequency distribution $J(\omega,t)$ for the field energy, defined for $\omega\geq 0$, by lumping together the average energies $\langle 1_A\otimes\hbar\omega_k a_k^\dagger a_k\rangle(t)$ of all the field modes in each small frequency interval $(\omega,\omega+\Delta\omega)$,

$$J(\omega,t)=\sum_k \langle 1_A\otimes\hbar\omega_k a_k^\dagger a_k\rangle(t)\delta(\omega-\omega_k)$$
$$=\hbar\omega \lim_{\Delta\omega\to 0}\left\{(1/\Delta\omega)\sum_{k\ (\omega<\omega_k<\omega+\Delta\omega)}\langle 1_A\otimes a_k^\dagger a_k\rangle(t)\right\} \tag{5.1}$$

where the last summation is over all modes k for which $\omega<\omega_k<\omega+\Delta\omega$. Clearly, $J(\omega,t)$ is such that

$$\int_0^\omega J(\omega',t)d\omega'=\sum_{k\ (\omega_k<\omega)}\langle 1_A\otimes\hbar\omega_k a_k^\dagger a_k\rangle(t)\ , \tag{5.2}$$

where the summation is over all modes k for which $\omega_k<\omega$. Following the same procedure as for all other observables, we shall express $J(\omega,t)$ as a series expansion in powers of λ,

$$J(\omega,t)=\sum_m \lambda^m J^{(m)}(\omega,t)\ , \tag{5.3}$$

valid for short times [see the discussion between (3.10) and (3.11)].

The complex functions $G_{ij}(\omega,t)$ defined as [see (2.37) where the explicit indication of the location $\mathbf{r}_A$ of the atom has been dropped]

$$G_{ij}(\omega,t)=\sum_k V_{kij}\alpha_k^*(t)\delta(\omega-\omega_k)$$
$$=\lim_{\Delta\omega\to 0}\left\{(1/\Delta\omega)\sum_{k\ (\omega<\omega_k<\omega+\Delta\omega)}V_{kij}\alpha_k^*(t)\right\}, \tag{5.4}$$

are useful to discuss the first contributions (up to $m=1$) to (5.3). The ω and ωt dependences of these functions are factorized [see (2.8)]

$$e^{-i\omega t}G_{ij}(\omega,t)=e^{-i\omega t_0}G_{ij}(\omega,t_0)\ . \tag{5.5}$$

Notice that the only nonvanishing contributions to (5.4) are from those modes k which contribute to the pulse (i.e., $|\alpha_k|\neq 0$) *and* are coupled to the atom (i.e., $|V_{kij}|\neq 0$ for one pair ij at least).

The functions $G_{ij}(\omega,t)$ are directly related to the Fourier transform of the time-dependent Hamiltonian used in the linearized classical-field approximation. Indeed, using (2.8) and (2.37), the Fourier transform of the function $V_{ij}(t)$ is

$$\int_{-\infty}^{+\infty}dt\,V_{ij}(t)e^{i\omega t}$$
$$=e^{i\omega t_0}\int_{-\infty}^{+\infty}dt\sum_k\left[V_{kij}^*\alpha_k(t_0)e^{i(\omega-\omega_k)(t-t_0)}+V_{kji}\alpha_k^*(t_0)\,e^{i(\omega+\omega_k)(t-t_0)}\right]. \tag{5.6}$$

Interchanging the summation and the integral, noting that, for real values of ω, the integrals of the exponentials give $2\pi\delta(\omega-\omega_k)$ or $2\pi\delta(\omega+\omega_k)$ and that $\omega_k\geq 0$ by definition, and using (5.4), one obtains the relations

$$\int_{-\infty}^{+\infty}dt\,V_{ij}(t)e^{i\omega t}$$
$$=\begin{cases}2\pi e^{-i|\omega|t_0}G_{ji}(|\omega|,t_0), & \text{if } \omega<0\\ 2\pi e^{i\omega t_0}G_{ij}^*(\omega,t_0), & \text{if } \omega>0\ .\end{cases} \tag{5.7}$$

Measuring $J(\omega,t)$ would involve the use of a dispersive spectrometer which (in a somewhat idealized picture) would sort out the incoming field in "frequency bins" of width $\Delta\omega$ and measure the amount of energy received in each bin. In any conventional dispersive system, a frequency resolution $\Delta\omega$ implies a distribution of propagation times between input and output of width Δt, with a lower bound given by $\Delta\omega\Delta t\geq 1$ (this is a rough statement concerning orders of magnitude for ill-defined quantities $\Delta\omega$ and Δt). As a consequence of this, the relevant frequency distribution of field energy is $J(\omega,t)$ for times t as late as possible after the beginning of the overlap between the classical-field pulse and the atom. The validity of the approximations of the present paper, which retain only the terms (5.11) and (5.13) of $J(\omega,t)$, is limited to durations much shorter than any radiative lifetime of the atom. However, these radiative lifetimes go to infinity when λ tends to zero so that useful indications will be obtained by taking the limit of $J(\omega,t)$ for $t\to+\infty$. Of course, the δ and principal part singularities generated by this limit process may only be taken as an indication about the presence of fast-frequency-dependent features, presumably extending over frequency ranges of the order of the inverse of the radiative lifetimes.

B. Evaluation of $J^{(m)}(\omega,+\infty)$ for $m\leq 1$

Using the same technique as in Sec. IV, one can easily see that the contributions to the average energy

$\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle(t)$ of the field mode k, arising from the first terms of (3.11) for $\hat{\rho}_S(t)$, can be expressed as $\alpha^2\hbar\omega_k \mid \alpha_k \mid^2$ for $n=0$ [see (3.16)], and as

$$K_6 = -\lambda\alpha[V_{k11}\alpha_k^*(t_i) + \text{c.c.}] \tag{5.8}$$

and

$$K_7 = \lambda\alpha\omega_k 2\mathrm{Im}\left\{\sum_{i,j} V_{kij}\int_{t_i}^{t} dt'\alpha_k^*(t') \times \langle \mid j \rangle_{A\,A}\langle i \mid \rangle^B(t')\right\}, \tag{5.9}$$

respectively, for the initial value and the integral terms for $n=1$ in (3.18). Contributions for $n \geq 2$ are of order 2 or higher in λ in the present short-time approximation, hence the contributions to $\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle(t)$ up to order 1 in λ are given by

$$\begin{aligned}
&\lambda^{-2}\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle^{(-2)}(t) = \alpha^2\hbar\omega_k \mid \alpha_k \mid^2 , \\
&\lambda^{-1}\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle^{(-1)}(t) = 0 , \\
&\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle^{(0)}(t) = K_6 + K_7 , \\
&\lambda\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle^{(1)}(t) = 0 .
\end{aligned} \tag{5.10}$$

The time-independent term of order α^2 (i.e., of order λ^{-2}) is the unperturbed average energy of mode K. The time-dependent term K_6+K_7, of order λ^0, is the energy "given by the atom to mode k of the field" in a first, short-time approximation which ignores radiative effects such as spontaneous emission and Lamb shifts. Equations (5.8) and (5.9) show that $\langle 1_A \otimes \hbar\omega_k a_k^\dagger a_k \rangle^{(0)}(t)$ is different from zero only for those modes k which contribute to the pulse (i.e., $\mid \alpha_k \mid \neq 0$) *and* are coupled to the atom (i.e., $\mid V_{kij} \mid \neq 0$ for one pair ij at least).

Combining (5.1)–(5.10), we can express $\lambda^m J^{(m)}(\omega,t)$ for $m \leq 1$ as

$$\lambda^{-1}J^{(-1)}(\omega,t) = 0, \quad \lambda J^{(1)}(\omega,t) = 0 ,$$
$$\lambda^{-2}J^{(-2)}(\omega,t) = \alpha^2\hbar\omega \lim_{\Delta\omega\to 0}\left\{(1/\Delta\omega)\sum_{k\ (\omega<\omega_k<\omega+\Delta\omega)} \mid \alpha_k \mid^2\right\}, \tag{5.11}$$

$$J^{(0)}(\omega,t) = -\lambda\alpha[G_{11}(\omega,t_i) + \text{c.c.}] + 2\lambda\alpha\omega\mathrm{Im}\left\{\sum_{i,j} G_{ij}(\omega,t_0)\int_{t_i}^{t} dt' e^{i\omega(t'-t_0)}\langle \mid j \rangle_{A\,A}\langle i \mid \rangle^B(t')\right\} \tag{5.12}$$

$$= -\lambda\alpha[G_{11}(\omega,t_0)e^{i\omega(t-t_0)} + \text{c.c.}] + 2\lambda\alpha\omega\mathrm{Im}\left\{\sum_{i,j} G_{ij}(\omega,t_0)\int_{t_i}^{t} dt' e^{i\omega(t'-t_0)}[\langle \mid j \rangle_{A\,A}\langle i \mid \rangle^B(t') - \delta_{ij,11}]\right\} . \tag{5.13}$$

The leading term (5.11) in this series expansion is the time-independent frequency distribution of the unperturbed field pulse. The next nontrivial term (5.13), of order λ^0, is the frequency distribution of the change in free-field energy due to the interaction with the atom, in a first approximation valid for times $t < t_b + \Delta t$, where t_b is the beginning of the overlap between the classical-field pulse and the atom, and Δt is a time interval much shorter than any radiative lifetime of the atom [see the discussion between (3.10) and (3.11)].

We now discuss some of the properties of the limit $J^{(0)}(\omega,+\infty)$. When $(t-t_0)$ is large, the first term in (5.13) has a fast oscillatory dependence upon ω, arising from the factor $\exp\{i\omega(t-t_0)\}$. As a result, *the average of this term over any finite frequency interval (which is the quantity of actual interest)* approaches zero when $t \to +\infty$ for any smooth function $G_{11}(\omega,t_0)$. Hence, we can ignore this first contribution to $J^{(0)}(\omega,+\infty)$.

To evaluate the time integral in the second term in (5.13), we choose the initial time t_i close before the beginning of the overlap between the classical-field pulse and the atom, and introduce an intermediate time t_f close after the end of this overlap. The first part of the integral, over the finite time interval from t_i to t_f, is a continuous function of ω, appreciably differing from zero over a frequency range at least of the order of magnitude of the inverse of the pulse duration $(t_f - t_i)$. For a pulse of finite duration, the $G_{ij}(\omega,t_0)$ are also functions of ω, with similar limitations on frequency range. Hence, the first part (from t_i to t_f) of the integral in (5.13) will, in general, contribute a nontrivial continuous function of ω to $J^{(0)}(\omega,+\infty)$.

For $t' \geq t_f$, i.e., after the end of the overlap between the classical pulse and the atom, one has, in the classical-field approximation,

$$\langle \mid j \rangle_{A\,A}\langle i \mid \rangle^B(t') = \langle \mid j \rangle_{A\,A}\langle i \mid \rangle^B(t_f)e^{-i(\omega_j-\omega_i)(t'-t_f)} . \tag{5.14}$$

In the particular case of a pulse which leaves the atom exactly in the eigenstate i' of H_{0A}, the only average value given by (5.14) which differs from zero is $\langle \mid i' \rangle_{A\,A}\langle i' \mid \rangle^B(t') = 1$, hence the second part (from t_f to $t \to \infty$) of the integral in (5.13) is easily evaluated as

$$(1/i\omega)(e^{i\omega(t-t_0)} - e^{i\omega(t_f-t_0)})(\delta_{ij,i'i'} - \delta_{ij,11}) . \tag{5.15}$$

The terms in $\exp\{i\omega(t_f-t_0)\}$ in (5.15) give a contribution to $J^{(0)}(\omega,+\infty)$, which is a continuous function of ω, with a frequency dependence essentially governed by that of $G_{i'i'}(\omega,t_0)$ and $G_{11}(\omega,t_0)$, and the terms in $\exp\{i\omega(t-t_0)\}$ can be ignored in the limit $t \to \infty$ because of their fast oscillatory dependence upon ω.

Hence, a first conclusion is that, if a short pulse transfers the atom *completely* from the ground state to any eigenstate of H_{0A} (in the classical-field approximation), then the change of the field energy due to the excitation of the atom is continuously distributed over the field modes in a whole range of frequencies with no singular

spectral feature at the atomic-transition frequency. Figures 2–5 (for the angles θ which are integral multiples of π) illustrate this situation for the particular model of a two-level atom which is discussed in detail in Sec. V D and in Appendix B. Almost all graphs display positive and negative excursions of $J^{(0)}(\omega,+\infty)$ and the curves for $\theta=7\pi$ and 9π in Fig. 5 clearly show that a single atom excited by a pulse not only takes its excitation energy $\hbar\omega_0$ from the field pulse as anticipated, but can also move even larger amounts of energy from one region of the pulse spectrum to another.

Whenever the pulse does not leave the atom exactly in an eigenstate of H_{0A} in the classical-field approximation, the present "pure-state" model implies that at least one pair of complex-conjugate average values $\langle\,|j\rangle_{A\,A}\langle i\,|\,\rangle^{B}(t_f)$ and $\langle\,|i\rangle_{A\,A}\langle j\,|\,\rangle^{B}(t_f)$ with $i\neq j$, is different from zero at the end of the atom-pulse overlap. Using (5.14), the second part (from t_f to $+\infty$) of the integral in (5.13) for one of these average values can be evaluated as

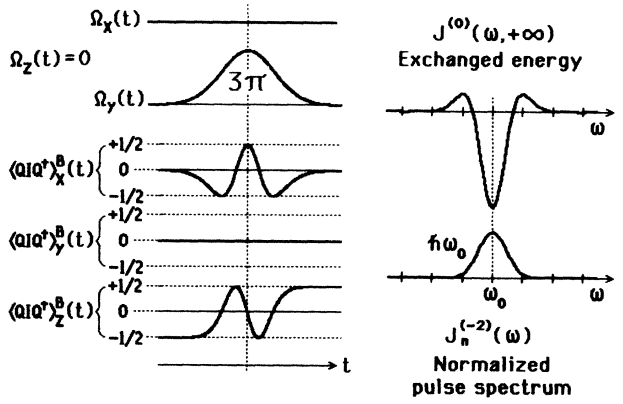

FIG. 2. Frequency distribution of the change in free-field energy $J^{(0)}(\omega,+\infty)$, due to the interaction of a two-level atom with a short pulse of radiation, ignoring spontaneous emission (negative excursions correspond to energy absorbed by the atom). The vertical scales for the exchanged energy and for the normalized pulse spectrum are such that equal areas under the curves correspond to equal amounts of energy, and the pulse spectrum has been normalized to correspond to the energy $\hbar\omega_0$ of a single resonant photon. The vectors $\lambda\alpha\boldsymbol{\Omega}(t)$ and $\langle QIQ^{\dagger}\rangle^{B}(t)$ are defined by (B9)–(B12) and (B16), and related by (B18). The "flip angle" θ (which is equal to 3π here) is defined by (B25). In a standard NMR experiment, $\lambda\alpha\boldsymbol{\Omega}(t)$ is the spin angular velocity as seen from the "rotating frame," $\hbar\langle QIQ^{\dagger}\rangle^{B}(t)$ is the average spin angular momentum and XYZ is the rotating frame with Z parallel to the constant magnetic field. Before the pulse, the atom is in its ground state. The techniques used to generate the data are discussed in Sec. V D. *In this figure,* a resonant pulse with a Gaussian envelope and $\theta=3\pi$ eventually leaves the atom exactly in its excited state. Note the smooth, nontrivial frequency distribution of the exchanged energy, for a total exchange of energy of $\hbar\omega_0$ from field to atom.

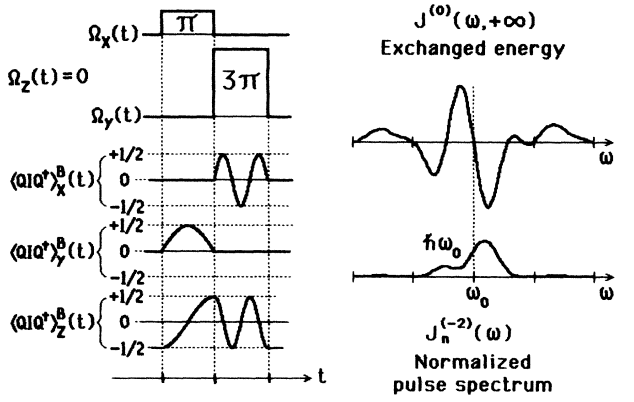

FIG. 3. Similar to Fig. 2 except that, here, a sequence of two resonant pulses, phase shifted by $\pi/2$ with respect to each other, eventually bring the atom back exactly in its ground state. Note that amounts of energy of order $\hbar\omega_0$ are moved from one spectral region to another, for a total exchange of energy of exactly zero.

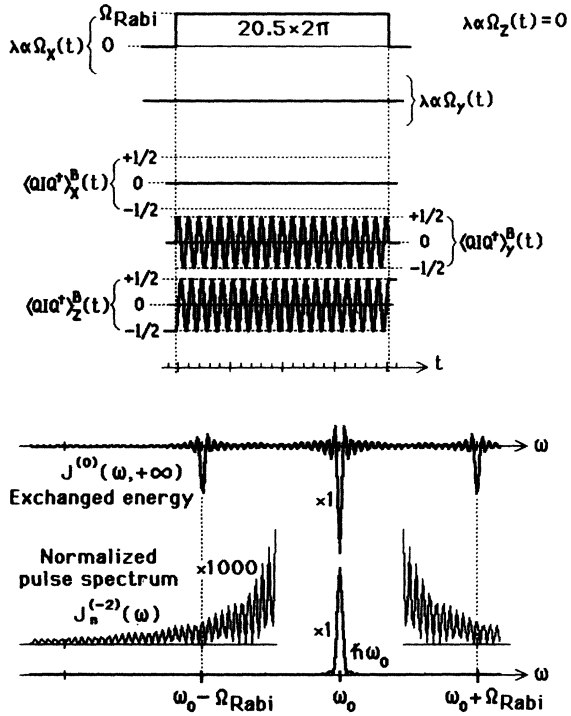

FIG. 4. Similar to Fig. 2 except that, here, a resonant pulse of constant amplitude and large flip angle $\theta=20.5\times2\pi$ eventually leaves the atom exactly in its excited state. Note the smooth frequency distribution of the exchanged energy, with the expected peaks at the resonant frequency plus or minus the Rabi frequency. For convenience, the wings of the normalized pulse spectrum are also shown enlarged vertically by a factor of 1000.

$$\lim_{t\to+\infty}\left\{\int_{t_f}^{t}dt' e^{i\omega(t'-t_0)}e^{-i(\omega_j-\omega_i)(t'-t_f)}\langle\,|j\rangle_{A\,A}\langle i|\,\rangle^{B}(t_f)\right\}$$
$$=e^{i\omega(t_f-t_0)}\langle\,|j\rangle_{A\,A}\langle i|\,\rangle^{B}(t_f)\{\pi\delta(\omega-(\omega_j-\omega_i))+i\mathscr{P}[1/\{\omega-(\omega_j-\omega_i)\}]\}\ . \quad (5.16)$$

Combining (5.13) and (5.16) we conclude that, for a general pulse, the extension to $+\infty$ of the integral in (5.13) can add a δ and a principal part singularity at each classical atomic-transition frequency $|\omega_j-\omega_i|$,

$$J^{(0)}(\omega,+\infty)=2(\lambda\alpha)\mathrm{Im}\left[\omega\sum_i\sum_j G_{ij}(\omega,t_0)\int_{t_i}^{t_f}dt' e^{i\omega(t'-t_0)}[\langle\,|j\rangle_{A\,A}\langle i|\,\rangle^{B}(t')-\delta_{ij,11}]\right.$$
$$+ie^{i\omega(t_f-t_0)}\sum_j G_{jj}(\omega,t_0)[\langle\,|j\rangle_{A\,A}\langle j|\,\rangle^{B}(t_f)-\delta_{jj,11}]$$
$$\left.+\omega e^{i\omega(t_f-t_0)}\sum_i\sum_{j\,(\neq i)}\langle\,|j\rangle_{A\,A}\langle i|\,\rangle^{B}(t_f)\{\pi\delta(\omega-(\omega_j-\omega_i))+i\mathscr{P}[1/\{\omega-(\omega_j-\omega_i)\}]\}\right]\ . \quad (5.17)$$

Of course, the "tails" of the principal parts add to the continuous parts of $J^{(0)}(\omega,+\infty)$. Figures 5–7 provide examples of functions $J^{(0)}(\omega,+\infty)$ showing either type of singularity, or both together, superimposed on the continuous part. Figure 5 also shows that the amount of energy exchanged in a δ singularity can be positive or negative, and may be larger in absolute value than the excitation energy of the atom.

C. Comparison with standard elementary time-dependent perturbation theory

One of the main assumptions used in the traditional evaluation of atomic-transition probabilities is that the interaction only causes a very small change in the state of the atom-field system during the irradiation. In the model discussed in this paper, this condition will be satisfied by taking the limit of weak pulses, i.e., by treating $\lambda\alpha$ as a small parameter and retaining only the first nontrivial term in a series expansion of the relevant observables in powers of $\lambda\alpha$.

For the frequency distribution of the energy given by the atom to the field, (5.13) indicates that the relevant term is in $(\lambda\alpha)^2$ and arises from contributions in $(\lambda\alpha)$ to $\langle\,|j\rangle_{A\,A}\langle i|\,\rangle^{B}(t')=\mathrm{Tr}_A\{\,|j\rangle_{A\,A}\langle i|\rho_A^B(t')\}$. These contributions can be evaluated by integration of an approximate form of (3.5) in which $\tilde{\rho}_A^B(t)$ is replaced in the commutator by its initial value $|1\rangle_{A\,A}\langle 1|$,

$$\tilde{\rho}_A^B(t')=|1\rangle_{A\,A}\langle 1|+(1/i\hbar)\int_{t_i}^{t'}dt''[Q_A^{\dagger}(t'')\lambda(\bar{V}(t'')-V)_A Q_A(t''),|1\rangle_{A\,A}\langle 1|\,]+O((\lambda\alpha)^2)\ . \quad (5.18)$$

As shown in Appendix C 5, this leads to

$$J^{(0)}(\omega,+\infty)=-\hbar\omega\sum_{j\,(\neq 1)}\{(4\pi/\hbar^2)(\lambda\alpha)^2\times|G_{j1}((\omega_j-\omega_1),t_0)|^2\times\delta(\omega-(\omega_j-\omega_1))\}+O((\lambda\alpha)^3)\ , \quad (5.19)$$

where t_0 is only a dummy variable as shown by (5.5). Equation (5.19) agrees with the standard prediction that, *for weak irradiation,* energy is exchanged between the atom and the field *only* at the atomic-transition frequencies. As a further check of agreement, one can easily verify that the coefficient $(4\pi/\hbar^2)(\lambda\alpha)^2|G_{j1}((\omega_j-\omega_1),t_0)|^2$ of the δ function in (5.19) is exactly the probability of transition $\langle\,|j\rangle_{A\,A}\langle j|\,\rangle^{B}(t_f)$ from state $|1\rangle_A$ to state $|j\rangle_A$, as evaluated from (3.5), to second order in $(\lambda\alpha)$.

D. Simple examples for a two-level atom

In this section we discuss the techniques and approximations used to generate the data shown in Figs. 2–7. The calculations were performed for a two-level atom, using the techniques and results discussed in Appendix B.

The bare-atom Hamiltonian can be written as $H_{0A}=\hbar\omega_0 I_{ZA}$, and the generator of the motion for $\tilde{\rho}_A^B(t)$ as $\hbar\mathbf{I}_A\cdot\lambda\alpha\boldsymbol{\Omega}(t)$, where [see (B15)–(B19)] $\mathbf{I}_A$ is the pseudospin associated with the atom, $\lambda\alpha\boldsymbol{\Omega}(t)$ is the pseudo-angular-velocity describing the action of the classical field on the atom, and ω_0 is the unperturbed atomic-transition frequency. We assumed a situation analogous to NMR of a single spin $\frac{1}{2}$, with a near-resonant irradiation, circularly polarized in such a way that $\boldsymbol{\Omega}(t)$ lies in the XY plane [hence $\Omega_Z(t)=0$] and differs from zero in a simple and slow way over a time interval (duration of the pulse) much longer than $1/\omega_0$. The time dependence of $\boldsymbol{\Omega}(t)$ is used as a starting point in the calculations, and is shown in the figures by its real X and Y components.

When $\Omega_Z(t)$ and $\Omega_{\pm}=\Omega_X(t)\pm i\Omega_Y(t)$ are known, $V_{ij}(t)$ can be evaluated easily by combining (2.36), (B7), and (B9), with the result that $V_{11}(t)=V_{22}(t)=0$ and

$$V_{12}^{*}(t)=V_{21}(t)=(\hbar/2)e^{-i\omega_0(t-t_0)}\Omega_{-}(t)\ , \quad (5.20)$$

hence $G_{ij}(\omega,t_0)$ is given by (5.7) as $G_{11}=G_{22}=0$ and

$$(\hbar/4\pi)\int_{-\infty}^{+\infty}dt\, e^{i(\omega-\omega_0)(t-t_0)}\Omega_{-}(t)=\begin{cases}G_{12}(|\omega|,t_0), & \text{if } \omega<0\\ G_{21}^{*}(\omega,t_0), & \text{if } \omega>0\ .\end{cases} \quad (5.21)$$

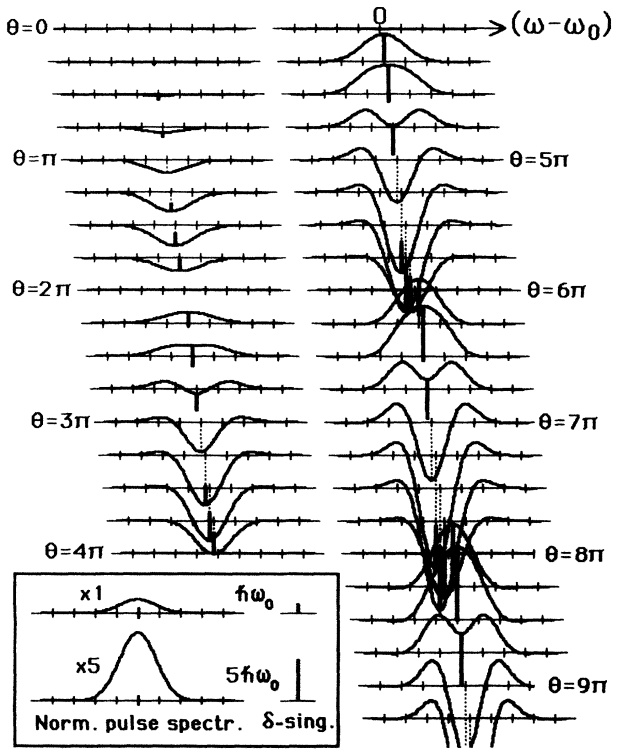

FIG. 5. Frequency distribution of the change in free-field energy due to the interaction of a two-level atom with resonant pulses with Gaussian envelopes of increasing magnitude (but constant duration between half-power points). The δ singularities in the exchanged energy spectrum are graphically shown as thick vertical bars, with heights proportional to the integral over the singularity. The inset shows the normalized pulse spectrum and the δ singularity, for amounts of energy $\hbar\omega_0$ and $5\hbar\omega_0$. Other details are as in Fig. 2.

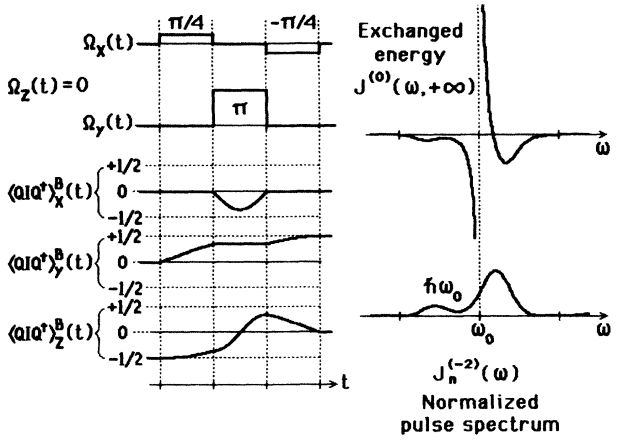

FIG. 6. Similar to Fig. 2 except that, here, the atom is eventually left half way between ground state and excited state, and the exchanged energy spectrum has a dispersive principal part singularity in addition to a continuous (mainly negative) part.

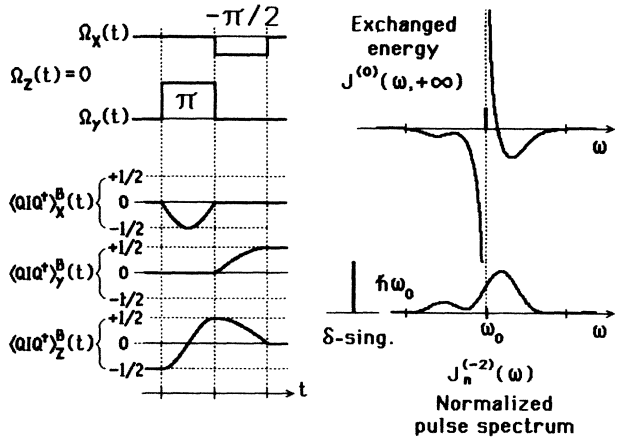

FIG. 7. Similar to Fig. 6, except that, here, the exchanged energy spectrum has all three types of components: δ singularity, dispersive singularity and continuous part. The graphical representation of the δ singularity is the same as in Fig. 5.

In the present case $G_{21}(\omega,t_0)$ is appreciably different from zero in a narrow frequency range around $\omega=\omega_0$, and much smaller outside that range, and $G_{12}(\omega,t_0)$ is very small for all ω.

When $\lambda\alpha\boldsymbol{\Omega}(t)$ is known, the equation of motion (B18) and (B19) for $\langle QIQ^\dagger\rangle^B(t)$ can be solved (numerically in the present case), hence the functions $\langle\,|\,j\rangle_{A\,A}\langle i\,|\,\rangle^B(t)$ are known and (5.17) and (5.21) can be combined to evaluate $J^{(0)}(\omega,+\infty)$. Of course, in our numerical calculations, the smooth part of $J^{(0)}(\omega,+\infty)$ is treated separately from the singularities.

The "pulse spectra" $J^{(-2)}(\omega,t)$ have been evaluated from the given function $\Omega_-(t)$ by assuming that the relevant matrix elements V_{kij} have negligible frequency dependence over the narrow frequency range of interest around ω_0. With this approximation (5.4) and (5.11) show that $J^{(-2)}(\omega,t)$ is proportional to $\omega\,|\,G_{12}(\omega,t_0)\,|^2$. The normalized pulse spectra

$$J_n^{(-2)}(\omega)=\frac{\hbar\omega_0}{\langle 1_A\otimes H_{0F}\rangle^{(-2)}}J^{(-2)}(\omega,t) \tag{5.22}$$

shown for reference in the figures are such that the area under the $J_n^{(-2)}(\omega)$ curve corresponds to a total energy $\hbar\omega_0$ (note also that the irrelevant time variable has been dropped). The same vertical scale is used in the figures for $J^{(0)}(\omega,+\infty)$ and $J_n^{(-2)}(\omega)$, so that equal areas correspond to equal amounts of energy.

Let us emphasize that, in the present context, the results shown in the figures do not involve any truncation of the atom-field coupling Hamiltonian (except for the assumed linear dependence in the field). In the case of more general (linear) coupling or irradiation, the rotating-wave approximation would lead to rather similar results for $J^{(-2)}(\omega,t)$ and $J^{(0)}(\omega,+\infty)$ for ω close to ω_0, with the slowly time-dependent truncated quantity $\boldsymbol{\Omega}_R(t)$, defined by (B12) in Appendix B, playing the role of $\boldsymbol{\Omega}(t)$.

VI. PHASE-SENSITIVE DETECTION OF THE RADIATION "EMITTED" BY THE ATOM

A. A simple model for phase-sensitive detection

The scheme which we shall discuss here is indicated in the lower part of Fig. 1: radiation emitted by the atom is superposed with a much stronger coherent "reference" radiation in a detector and, for a photoelectric detector, the relevant measured quantity is the *change* in the counting rate of photoelectrons due to the atom radiation (often called "beats" between reference radiation and atom radiation).

In spite of much effort and progress, the general theory of photoelectric detection is still, in many respects, an open problem.[9] Following the tradition, we shall replace this missing theoretical link by a conventional "educated guess" which, hopefully, is suitable for the problem at hand. Hence, we shall assume that the average measured photoelectric counting rate at time t can be predicted by evaluating the quantum average value

$$\langle D_S \rangle(t)=\mathrm{Tr}_S\{D_S\rho_S(t)\} \tag{6.1}$$

of a Hermitian "observable" D_S describing the detection process, for a state of the atom-field system described by $\rho_S(t)$. As usual, the detector will not be included in the Hamiltonian H_S of the system.

In the simple type of phase-sensitive detector discussed here, the relevant beats are in narrow frequency bands around the differences (and sums) between the reference oscillator frequency $(\omega_r/2\pi)$ and the atomic-transition frequencies, hence $(\omega_r/2\pi)$ will usually be chosen very close to one of the atomic frequencies in order to make the corresponding component of the beat a slow-enough function of time to be directly observable. As suggested by Fig. 1, it will be convenient to assume that the reference mode r does not interact with the atom [hence $V_{rij}(\mathbf{r}_A)=0$ for all i and j in (2.4) and (2.37)] and does not participate to the pulse, and that the pulse does not interact directly with the detector.

The detector is *directly coupled to the field but not to the atom,* hence the observable D_S acts trivially in the atomic state space,

$$D_S=1_A\otimes D_F \ . \tag{6.2}$$

For simplicity we shall assume that the detector is operated at such a low light level that the counting rate observable D_F is well approximated by a bilinear combination of field operators, which depends on the type, shape, and position $\mathbf{r}_D$ of the detector,

$$D_F(\mathbf{r}_D)=\sum_{k,k'}[D_{kk'}^{--}(\mathbf{r}_D)a_k^\dagger a_{k'}^\dagger+D_{kk'}^{-+}(\mathbf{r}_D)a_k^\dagger a_{k'} +D_{kk'}^{+-}(\mathbf{r}_D)a_k a_{k'}^\dagger+D_{kk'}^{++}(\mathbf{r}_D)a_k a_{k'}] \ . \tag{6.3}$$

Using the boson commutation relations, we may write D_F under the more convenient form

$$D_F(\mathbf{r}_D)=\sum_{k,k'}[D_{kk'}^{--}(\mathbf{r}_D)a_k^\dagger a_{k'}^\dagger+2D_{kk'}^{0}(\mathbf{r}_D)a_k^\dagger a_{k'} +D_{kk'}^{++}(\mathbf{r}_D)a_k a_{k'}]+\left[\sum_k D_{kk}^{+-}(\mathbf{r}_D)\right]1_F \ , \tag{6.4}$$

where $D_{kk'}^{--}=\frac{1}{2}(D_{kk'}^{--}+D_{k'k}^{--})$ and $D_{kk'}^{++}=\frac{1}{2}(D_{kk'}^{++}+D_{k'k}^{++})$ are invariant for permutation of the mode labels k and k', and $D_{kk'}^{0}=\frac{1}{2}(D_{k'k}^{+-}+D_{kk'}^{-+})$; the Hermiticity of D_F implies that $(D_{kk'}^{++})^*=D_{kk'}^{--}$ and $(D_{kk'}^{0})^*=D_{k'k}^{0}$, hence D_{kk}^0 is real. The idealized detector which we consider here will have a counting rate of exactly zero whenever the field is exactly in its ground state, hence

$$\left[\sum_k D_{kk}^{+-}\right]=0 \ . \tag{6.5}$$

The reference radiation used in the phase-sensitive detection scheme will be described as a quasiclassical excitation of a single field mode, with label r (standing for reference), parameter $\alpha_r(t)$, and frequency ω_r. In order to avoid ambiguities in the discussion of weak atom-field coupling (i.e., $\lambda\to 0$ with $\lambda\alpha$ remaining constant), we have *not* included the real number α [see (2.30), (2.36), and the end of Sec. II A] in the definition of the $\alpha(t)$ parameter of the reference mode r. With this notation, the properties of the detection scheme are not affected by the limit process and the disentangling operator $M_S(t)$, originally given by (2.30), can be written in the form

$$M_S(t)=1_A\otimes M_F(\{\alpha\alpha_k(t)\})M_F(\alpha_r(t)) \ . \tag{6.6}$$

The assumption that the pulse does not interact with the detector implies that $D_{kk'}^{+-}=D_{kk'}^{0}=0$ for all modes k for which $|\alpha_k(t)|\neq 0$ (or, more compactly, that $[D_F,M_F(\{\alpha\alpha_k(t)\})]=0$), and hence the bar version of D_S is

$$\bar{D}_S(t)=M_S^\dagger(t)D_S M_S(t)=1_A\otimes M_F^\dagger(\alpha_r(t))D_F M_F(\alpha_r(t)) =1_A\otimes\left[\sum_{k,k'}(D_{kk'}^{--}a_k^\dagger a_{k'}^\dagger+D_{kk'}^{0}a_k^\dagger a_{k'})+2\alpha_r^*(t)\sum_k(D_{rk}^{--}a_k^\dagger+D_{rk}^{0}a_k)+\{D_{rr}^{--}[\alpha_r^*(t)]^2+D_{rr}^{0}|\alpha_r(t)|^2\}1_F\right]+\text{H.c.} \tag{6.7}$$

We can now use the same techniques as in Sec. V to evaluate the first terms of a series expansion of the quantum-average value $\langle \bar{D}_S\rangle(t)$ in powers of λ [see (3.20)]. Equation (6.7) for $\bar{D}_S(t)$ does not involve the parameters λ or α, hence

$$\lambda^n\langle D_S\rangle^{(n)}(t)=\lambda^n\mathrm{Tr}_S\{D_S\rho_S^{(n)}(t)\}=\lambda^n\mathrm{Tr}_S\{\hat{D}_S\hat{\rho}_S^{(n)}(t)\} \ .$$

The leading term is easily evaluated because all nontrivial field operators in (6.7) have an average value zero in the ground state of the field,

$$\langle D_S\rangle^{(0)}(t)=\mathrm{Tr}_S\{\hat{D}_S\hat{\rho}_S^{(0)}\}$$
$$=\{D_{rr}^{-}[\alpha_r^*(t)]^2+D_{rr}^{0}\,|\,\alpha_r(t)\,|^2\}+\text{c.c.}$$
$$=2D_{rr}^{0}\,|\,\alpha_r(t_0)\,|^2$$
$$+2\,\mathrm{Re}\{D_{rr}^{-}[\alpha_r^*(t_0)]^2e^{2i\omega_r(t-t_0)}\}\ . \tag{6.8}$$

The first term in the right-hand side (rhs) of (6.8) is the expected time-independent rate of photoelectric detection of the reference radiation in the absence of the atom (i.e., $\lambda=0$). The second term oscillates around zero at twice the reference-mode frequency, a frequency much too high for direct observation with present technology for $(\omega_r/2\pi)$ in the optical range.

As $\hat{\rho}_S^{(1)}(t)$ is linear in a_k and $a_k^\dagger$, only the terms linear in a_k and $a_k^\dagger$ in (6.7) contribute to $\lambda\langle D_S\rangle^{(1)}(t)$ and this contribution is proportional to $|\,\alpha_r\,|$. The contribution K_9 to $\lambda\langle D_S\rangle^{(1)}(t)$ arising from the initial condition in (3.18) for $\hat{\rho}_S^{(1)}(t)$ is easily evaluated, and is exactly zero in the linearized dipolar approximation (see Appendix C), hence we shall ignore it here. The contribution to $\lambda\langle D_S\rangle^{(1)}(t)$ arising from the integral in (3.18) for $\hat{\rho}_S^{(1)}(t)$, computed in much the same way as $\langle H_{0F}\rangle(t)$ in Sec. IV, can be written as

$$\lambda\langle D_S\rangle^{(1)}(t)-K_9$$
$$=\lambda 2\,\mathrm{Re}\left[(1/i\hbar)2\alpha_r(t_0)e^{i\omega_r(t-t_0)}\right.$$
$$\times\int_{t_i}^{t}dt'\sum_{i,j}\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t')$$
$$\left.\times T_{ij}(\mathbf{r}_A,\mathbf{r}_D,t-t')\right]\ , \tag{6.9}$$

where

$$T_{ij}(\mathbf{r}_A,\mathbf{r}_D,t-t')=\sum_k\{-V_{kij}^*(\mathbf{r}_A)D_{rk}^{-}(\mathbf{r}_D)e^{i\omega_k(t-t')}$$
$$+V_{kji}(\mathbf{r}_A)D_{rk}^{0}(\mathbf{r}_D)e^{-i\omega_k(t-t')}\}\ . \tag{6.10}$$

Equation (6.9) expresses the behavior of the phase-sensitive detector in terms of the quantities $T_{ij}(\mathbf{r}_A,\mathbf{r}_D,t-t')$ which are joined properties of (i) the atom-field coupling $V_S(\mathbf{r}_A)$, (ii) the detection observable $D_S(\mathbf{r}_D)$, (iii) the reference mode r, and (iv) the mode structure of the electromagnetic field which may be strongly affected by the optical instruments inserted between the atom and the detector. For practical instruments with high collection efficiency, good approximations for (6.10) should be provided by techniques of the type given, for instance, by Kline and Kay.[10]

In Appendix C we make an *exact* evaluation of (6.10) in the simple case of (a) the linearized electric-dipolar coupling (2.5), (b) a point detector sensitive to the electric-energy density (with sensitivity K), (c) a free electromagnetic field (i.e., no optical instrument), and (d) a reference mode with propagation vector $\mathbf{k}_r$, frequency $\omega_r/2\pi$, and linear polarization vector $\hat{\mathbf{s}}_r$. In this computation, the electric-energy density operator is taken proportional to the normally ordered version $:\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r}):$ of the squared modulus $\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r})$ of the electric-field operator (i.e., the version of $\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r})$ obtained by replacing each term $a_ka_k^\dagger$ by $a_k^\dagger a_k$; we shall indicate this transformation by writing the operator between colons). The corresponding detection observable,

$$D_F(\mathbf{r}_D)=K:\mathbf{E}_F(\mathbf{r}_D)\cdot\mathbf{E}_F(\mathbf{r}_D):\ , \tag{6.11}$$

where K is a constant, is Hermitian, bilinear in the field operators, and satisfies condition (6.5) for zero detection in the ground state of the field. Arguments for this choice will be given in Sec. VI B. Using the result (C18) of this computation, expression (6.9) for $\lambda\langle D_S\rangle^{(1)}(t)$ is easily evaluated as ($\mathbf{R}=\mathbf{r}_D-\mathbf{r}_A$ is the relative position of atom and detector and $\hat{\mathbf{R}}=\mathbf{R}/|\,R\,|$)

$$\lambda\langle D_S\rangle^{(1)}(t)=\lambda K(1/8\pi)\mathrm{Re}\left[\alpha_r(t_0)(\hbar\omega_r/2L^3)^{1/2}e^{-i\mathbf{k}_r\cdot\mathbf{r}_D}e^{i\omega_r(t-t_0)}(1/c^2\,|\,R\,|)\right.$$
$$\times\sum_{i,j}-\left[\frac{d^2}{dt^2}\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t-|\,R\,|/c)\right]$$
$$\left.\times\{(\langle i\,|\,\boldsymbol{\mu}_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_r)-2(\langle i\,|\,\boldsymbol{\mu}_{eA}\,|\,j\rangle\cdot\hat{\mathbf{R}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{R}})\}\right]+(\cdots) \tag{6.12}$$

where the ellipsis represents similar terms in $|\,R\,|^{-2}$ and $|\,R\,|^{-3}$, also involving $\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t-|\,R\,|/c)$. At distances $|\,R\,|$ much larger than the wavelengths at the atomic-transition frequencies, the terms in $|\,R\,|^{-2}$ and $|\,R\,|^{-3}$ in the rhs of (6.12) can be neglected.

In typical experimental situations, the time evolution of $\rho_A^B(t)$ is caused essentially by H_{0A}, hence the time evolution of $\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B$ over a few periods is well approximated by

$$\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t)$$
$$=e^{i(\omega_i-\omega_j)(t-t_1)}\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t_1)\ , \tag{6.13}$$

and the second time derivative at time $(t-|\,R\,|/c)$ is well approximated by

$$-(\omega_i-\omega_j)^2\langle\,|\,i\,\rangle_A\,{}_A\langle\,j\,|\,\rangle^B(t-|\,R\,|/c)\ .$$

The standard photoelectric detectors are unable to follow time dependences of the electric-energy density at optical frequencies, hence the scheme discussed here requires adjusting ω_r close to *one* of the atomic-transition frequencies, for instance $(\omega_j-\omega_i)$ with $\omega_j>\omega_i$, in such a way that the combined time dependence $\exp(i\omega_r t)\exp[-i(\omega_j-\omega_i)t]$ which appears in (6.12) is slow enough for actual direct observation. With this tech-

nique, the time evolution of each term $\langle\,|\,i\,\rangle_{A\,A}\langle\,j\,|\,\rangle^{B}$ can be measured in great detail by suitably choosing ω_r.

The detection of beats between the atom radiation and a reference radiation is qualitatively different from the direct detection of "spontaneous emission": spontaneous emission manifests itself from $n=2$ on in (3.11) and causes observable effects of order λ^2 in the present short-time-approximation scheme, whereas $\lambda\langle D_S\rangle^{(1)}(t)$ is an observable quantity of order λ, arising from the term $n=1$ in (3.11). Also $\langle\lambda D_S\rangle^{(1)}(t)$ is proportional to the amplitude $|\alpha_r|$ of the reference radiation, hence can be made large (without affecting the atom in any way) by suitably increasing $|\alpha_r|$. Note, however, that increasing $|\alpha_r|$ does not improve the detectability of the radiative effects of the atom because $\lambda\langle D_S\rangle^{(1)}(t)$ is superimposed on the direct detection $\langle D_S\rangle^{(0)}(t)$ of the reference radiation itself, which gives a photoelectric counting rate proportional to $|\alpha_r|^2$, hence an rms fluctuation in counting rate proportional to $|\alpha_r|$. Furthermore, for a point detector and free field, spontaneous emission is detected proportional to $|R|^{-2}$ whereas (6.12) clearly shows that the beats are detected proportional to $|R|^{-1}$.

Performing an experiment in the optical-frequency range with the type of setup suggested by Fig. 1 would be very difficult because it would require an exceedingly accurate and permanent alignment of the optical system, detector and atom to avoid averaging the beats to zero. However, the beats discussed here have already been observed in the optical range many years ago, using a more suitable experimental setup.[11]

B. Digression about locality, causality, and propagation of light

In contradistinction with the rest of this paper, the discussion presented in this section rests on heuristic arguments.

As far as causality and propagation of light are concerned, Fig. 1 suggests the bold idea that the response of the fast detector at location $\mathbf{r}_D$ and time t depends upon the radiation emitted at location $\mathbf{r}_A$ and time $t-\Delta t$ (where Δt is the delay for propagation of light from $\mathbf{r}_A$ to $\mathbf{r}_D$), hence depends upon the state of the atom at time $t-\Delta t$ and upon the irradiation field at location $\mathbf{r}_A$ and time $t-\Delta t$. However, in a description in which atom and field are treated as a single system, using the notion of "state of the atom" may be misleading. The model used in this paper allows a more cautious formulation of the idea, which avoids this weakness: if the classical field associated with the coherent excitation pulse has a sharp front which crosses the location $\mathbf{r}_A$ of the atom at time t_b, the effect of the pulse on the atom should not be felt at the location $\mathbf{r}_D$ of the detector at any time earlier than $(t_b+\Delta t)$. These simple ideas can be expected to hold in the above form for a point detector for which the detection observable D_S is "local at $\mathbf{r}_D$" (i.e., involves properties of the field at the single location $\mathbf{r}_D$ and *not* at other locations), and for an atom-field coupling V_S which is similarly local at $\mathbf{r}_A$. Furthermore, the "bold" idea above clearly assumes that there is a *single* propagation time Δt from $\mathbf{r}_A$ to $\mathbf{r}_D$, as would be the case for a free field in vacuum (or for an ideal optical system imaging $\mathbf{r}_A$ onto $\mathbf{r}_D$).

Let us briefly examine the question of locality in general. For the sake of uniformity of presentation, we shall use the Schrödinger picture in which the field operators at fixed locations are time independent. In a Heisenberg picture all these operators would be at the same time. We assume that the operators $A_F(\mathbf{r}_1)$ and $B_F(\mathbf{r}_2)$ are both local, respectively at $\mathbf{r}_1$ and at $\mathbf{r}_2$, meaning, for instance, that $A_F(\mathbf{r}_1)$ describes some physical condition at the particular location $\mathbf{r}_1$ (and *not* at other locations). In analogy with the well-known situation for the components of the $\mathbf{E}$ and $\mathbf{B}$ fields,[12] we expect that the local character of A and B implies that

$$[A_F(\mathbf{r}_1),B_F(\mathbf{r}_2)]=0,\quad \text{whenever } \mathbf{r}_1\neq\mathbf{r}_2 \tag{6.14}$$

including the particular case $[A_F(\mathbf{r}_1),A_F(\mathbf{r}_2)]=0$, of course.

Let us now consider a state $\rho_{\mathscr{R}F}$ of the field such that, in a three-dimensional region $\mathscr{R}$ of space (and on its border), the average value of all functions of field components have the same value as in the ground state ρ_{GF} of the field. Another necessary condition for locality of the observable $A_F(\mathbf{r})$ is that

$$\mathrm{Tr}_F\{A_F(\mathbf{r})\rho_{\mathscr{R}F}\}=\mathrm{Tr}_F\{A_F(\mathbf{r})\rho_{GF}\}\ , \tag{6.15}$$

whenever $\mathbf{r}$ is in $\mathscr{R}$.

It is quite obvious that operators such as $\mathbf{E}_F(\mathbf{r})$, $\{\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r})\}$, and

$$:\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r}):=\mathbf{E}_F(\mathbf{R})\cdot\mathbf{E}_F(\mathbf{R})-1_F\sum_k|\,\mathbf{e}_k(\mathbf{r})\,|^2\ , \tag{6.16}$$

where $\sum_k|\,\mathbf{e}_k(\mathbf{r})\,|^2$ is a c-number, are local in the above sense and satisfy (6.14) and (6.15). In contrast, the operators

$$\mathbf{E}_F^-(\mathbf{r})=\sum_k\mathbf{e}_k^*(\mathbf{r})a_k^\dagger,\quad \mathbf{E}_F^+(\mathbf{r})=\sum_k\mathbf{e}_k(\mathbf{r})a_k\ , \tag{6.17}$$

such that $\mathbf{E}_F(\mathbf{r})=\mathbf{E}_F^-(\mathbf{r})+\mathbf{E}_F^+(\mathbf{r})$ and $\mathbf{E}_F^-(\mathbf{r})=(\mathbf{E}_F^+(\mathbf{r}))^\dagger$, appear as nonlocal (in spite of the typography used). Indeed, one can show after some calculations that, for a free field and for two *different* locations $\mathbf{r}_1$ and $\mathbf{r}_2$,

$$[\mathbf{E}_F^-(\mathbf{r}_1),\mathbf{E}_F^+(\mathbf{r}_2)]=1_F(\hbar c/\pi^2)\,|\,\mathbf{r}_1-\mathbf{r}_2\,|^{-4}\ , \tag{6.18}$$

in clear contradiction to (6.14). One can also show easily that the operator $\mathbf{E}_F^+(\mathbf{r})$ does not satisfy (6.15) by considering the particular quasiclassical state of the free field in which $\alpha_{\mathbf{k}s}=(8L/\hbar\omega_{\mathbf{k}})^{1/2}$ for all $\mathbf{k}$ pointing *exactly* in the $+z$ direction (in a discrete $\mathbf{k}$ space) and $\hat{\mathbf{s}}_{\mathbf{k}}=\hat{\mathbf{x}}$, and all other α parameters are equal to zero. The average field in this state is given by

$$\langle\,\mathbf{E}_F(\mathbf{r})\,\rangle=\hat{\mathbf{x}}\delta(z)\ , \tag{6.19}$$

and one can easily verify that other combinations of field components, such as $:\mathbf{E}_F(\mathbf{r})\cdot\mathbf{E}_F(\mathbf{r}):$ for instance, also deviate from their ground-state value only if $\mathbf{r}$ is in the $z=0$ plane. However, the average value of $\mathbf{E}_F^+(\mathbf{r})$ in this state is given by

$$\langle\,\mathbf{E}_F^+(\mathbf{r})\,\rangle=\hat{\mathbf{x}}\{\tfrac{1}{2}\delta(z)+(i/2\pi)\mathscr{P}(1/z)\}\ , \tag{6.20}$$

and is clearly different from zero for locations $\mathbf{r}$ outside

the $z=0$ plane, in contradiction to (6.15).

Coming back to the specific problem at hand, the linearized electric dipolar coupling

$$V_{\mathrm{ed},S}(\mathbf{r}_A)=-\boldsymbol{\mu}_{eA}\cdot\mathbf{E}_F(\mathbf{r}_A)$$
$$=-\sum_{i,j}|j\rangle_A\,{}_A\langle i|\otimes\langle j|\boldsymbol{\mu}_{eA}|i\rangle\cdot\mathbf{E}_F(\mathbf{r}_A) \tag{6.21}$$

appears as a local operator because it involves the field only through the local operator $\mathbf{E}_F(\mathbf{r}_A)$. In contrast, the rotating-wave approximation (RWA) to $V_{\mathrm{ed},S}(\mathbf{r}_A)$, which amounts to replacing $\mathbf{E}_F(\mathbf{r}_A)$ in (6.21) by $\mathbf{E}_F^+(\mathbf{r}_A)$ when $\omega_j>\omega_i$, and by $\mathbf{E}_F^-(\mathbf{r}_A)$ when $\omega_j<\omega_i$, appears as a nonlocal operator because $\mathbf{E}_F^+$ and $\mathbf{E}_F^-$ are nonlocal.

The usual rough caricatures for the photoelectric detection observable are proportional to the electric-energy density. Of course, $\mathbf{E}_F(\mathbf{r}_D)\cdot\mathbf{E}_F(\mathbf{r}_D)$ is not a suitable observable because of its (location-independent) divergent average value in the ground state of the field. One way out of this difficulty is to subtract the offending ground-state average value from the operator, as shown by equation (6.16), hence leading to the proposal $D_F(\mathbf{r}_D)=K{:}\mathbf{E}_F(\mathbf{r}_D)\cdot\mathbf{E}_F(\mathbf{r}_D){:}$, much in the same way which leads to the Hamiltonian $H_F=\sum_k\hbar\omega_k a_k^\dagger a_k$ for the free field itself. This proposal is local at the single point $\mathbf{r}_D$. Another way out of the difficulties with $\mathbf{E}_F(\mathbf{r}_D)\cdot\mathbf{E}_F(\mathbf{r}_D)$ is to replace it by $\mathbf{E}_F^-(\mathbf{r}_D)\cdot\mathbf{E}_F^+(\mathbf{r}_D)$. However, as indicated above, this replacement leads to a proposal for $D_F(\mathbf{r}_D)$ which is *not* local at $\mathbf{r}_D$.

It is very gratifying that, in the framework of the present model and to first order in λ, exact causality [see (6.12)] is obtained without any approximation when the atom-field coupling $V_S(\mathbf{r}_A)$ and the detection observable $D_F(\mathbf{r}_D)$ are both local, and the field propagates freely in vacuum. Equation (6.12) is even in agreement with the bold version of causality described at the beginning of Sec. VI B. This result, however, should be taken with caution because it was obtained in a first approximation in λ which is still very close to classical physics. It is also gratifying to verify (after some calculations) that replacing $V_{\mathrm{ed},S}(\mathbf{r}_A)$ by its RWA, or using a $D_F(\mathbf{r}_D)$ proportional to $\mathbf{E}_F^-(\mathbf{r}_D)\cdot\mathbf{E}_F^+(\mathbf{r}_D)$, or both, destroys this exact causality, as expected for nonlocal interaction or detection. The same relation between exact causality and the choice of interaction and detection observable has been obtained by de Haan[13] in a different context.

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APPENDIX A: QUASICLASSICAL STATES OF A SINGLE HARMONIC OSCILLATOR

1. Generalities and "displacement operator"

The state of a classical harmonic oscillator of frequency $\omega/2\pi$ can be completely specified by the complex number $\alpha(t)$ which evolves in time according to

$$\alpha(t)=\alpha(t_0)\exp[-i\omega(t-t_0)]\,, \tag{A1}$$

and is defined such that the oscillator energy is given by $E_{\mathrm{cl}}=\hbar\omega\,|\alpha(t)|^2$.

To each classical state, specified by $\alpha(t)$, we can associate a quasiclassical or coherent quantum state $|\alpha(t)\rangle$ by requiring that (a) the quantum average annihilation operator $\langle\alpha(t)|a|\alpha(t)\rangle$ is equal to $\alpha(t)$ and (b) the quantum average energy has the lowest value compatible with the first requirement. These requirements imply that

$$a|\alpha(t)\rangle=\alpha(t)|\alpha(t)\rangle\,, \tag{A2}$$

and, if we omit the "zero-point energy" from the oscillator Hamiltonian

$$H=\hbar\omega a^\dagger a\,, \tag{A3}$$

the quantum average energy $\langle\alpha(t)|H|\alpha(t)\rangle$ is equal to the classical energy.

Quasiclassical states can be constructed from the ground state $|0\rangle$ by the action of the Glauber[5] displacement operator M,

$$|\alpha(t)\rangle=M(\alpha(t))|0\rangle\,, \tag{A4}$$

where

$$M(\alpha(t))=\exp[\alpha(t)a^\dagger-\alpha^*(t)a]\,. \tag{A5}$$

We shall now collect some properties of $M(\alpha(t))$ which can be derived easily with the help of the rich set of theorems and methods given by Louisell.[14] The displacement operator is unitary,

$$M^\dagger(\alpha(t))=M^{-1}(\alpha(t))\,, \tag{A6}$$

has the simple symmetries

$$M^\dagger(\alpha(t))=M(-\alpha(t)),\quad M(0)=1\,, \tag{A7}$$

and the group property

$$M(\alpha_2(t))M(\alpha_1(t))=M(\alpha_2(t)+\alpha_1(t))\exp(i\varphi_{21})\,, \tag{A8}$$

where φ_{21} is a real, time-independent phase

$$\varphi_{21}=-\tfrac{1}{2}i[\alpha_2(t)\alpha_1^*(t)-\alpha_2^*(t)\alpha_1(t)]\,. \tag{A9}$$

Useful commutation properties of M are

$$\begin{aligned}&[a,M(\alpha(t))]=\alpha(t)M(\alpha(t))\,,\\ &[a^\dagger,M(\alpha(t))]=\alpha^*(t)M(\alpha(t))\,,\\ &[a^\dagger a,M(\alpha(t))]=[\alpha(t)a^\dagger+\alpha^*(t)a\\ &\qquad\qquad-|\alpha(t)|^2]M(\alpha(t))\,.\end{aligned} \tag{A10}$$

The first two relations of (A10) can also be written as

$$M(\alpha(t))aM^{\dagger}(\alpha(t))=a-\alpha(t)\ ,$$
$$M(\alpha(t))a^{\dagger}M^{\dagger}(\alpha(t))=a^{\dagger}-\alpha^{*}(t)\ , \tag{A11}$$

and similar relations with the roles of M and $M^{\dagger}$ interchanged using (A7). Following the lines indicated by Louisell,[15] the above results can be extended to any function $g(a,a^{\dagger})$ which can be expanded in a power series,

$$M(\alpha(t))g(a,a^{\dagger})M^{\dagger}(\alpha(t))=g(a-\alpha(t),a^{\dagger}-\alpha^{*}(t))\ . \tag{A12}$$

A little algebra shows that

$$i\hbar\frac{\partial}{\partial t}M(\alpha(t))=[H,M(\alpha(t))]\ . \tag{A13}$$

2. "Large" and "small" operators, states close to a coherent state $|\alpha(t)\rangle$

With the notation $|||\psi\rangle||^2=\langle\psi|\psi\rangle$ for the square of the norm of a ket, one can easily show that

$$||[a-\alpha(t)]|\alpha(t)\rangle||^2=0,\quad ||a|\alpha(t)\rangle||^2=|\alpha(t)|^2 \tag{A14}$$

for a normalized ket $|\alpha(t)\rangle$ [i.e., $\langle\alpha(t)|\alpha(t)\rangle=1$]. In this way, when acting on the coherent state $|\alpha(t)\rangle$, the operator $[a-\alpha(t)]$ appears as the zero operator and the operator a as of order $|\alpha(t)|$. If these operators act on a state $|\varphi(t)\rangle$ (not necessarily coherent) which is close to $|\alpha(t)\rangle$ in the sense that $|||\varphi(t)\rangle-|\alpha(t)\rangle||\ll 1$, $a-\alpha(t)$ will appear as a very small operator and a will still appear as of order $|\alpha(t)|$. Conversely, when acting on a state which is close to the ground state $|0\rangle$, $a-\alpha(t)$ appears as of order $|\alpha|$, and a as very small. We have found these remarks helpful in sorting out contributions involving various powers of $|\alpha(t)|$.

The obvious relation

$$||\,|\varphi(t)\rangle-|\alpha(t)\rangle||^2=||M^{\dagger}(\alpha(t))[\,|\varphi(t)\rangle-|\alpha(t)\rangle]||^2$$
$$=||M^{\dagger}(\alpha(t))\,|\varphi(t)\rangle-|0\rangle||^2 \tag{A15}$$

shows that, if $|\varphi(t)\rangle$ is close to the coherent state $|\alpha(t)\rangle$, then $M^{\dagger}(\alpha(t))\,|\varphi(t)\rangle$ is close to the ground state $|0\rangle$.

3. A convenient basis for states close to $|\alpha(t)\rangle$

The representation and manipulation of states which are all close to some known quasiclassical state $|\alpha(t)\rangle$ are simplified by the use of a basis which is constructed by the action of the unitary transformation $M(\alpha(t))$ on the usual basis $\{|n\rangle\}$, which diagonalizes the occupation number operator $N=a^{\dagger}a$. Starting from

$$a^{\dagger}a\,|n\rangle=n\,|n\rangle,\quad n=0,1,2,\ldots\ , \tag{A16}$$

we multiply both terms of this equality by $M(\alpha(t))$ at the left, insert the unit operator $1=M^{\dagger}(\alpha(t))M(\alpha(t))$ at the left and at the right of a, use (A11), and obtain the relation

$$\{[a^{\dagger}-\alpha^{*}(t)][a-\alpha(t)]\}[M(\alpha(t))\,|n\rangle]$$
$$=n[M(\alpha(t))\,|n\rangle]\ . \tag{A17}$$

Hence, the kets $M(\alpha(t))\,|n\rangle$ are the eigenkets of the Hermitian operator

$$[a^{\dagger}-\alpha^{*}(t)][a-\alpha(t)]=M(\alpha(t))NM^{\dagger}(\alpha(t))\ ,$$

and the corresponding eigenvalues are the non-negative integers. The eigenstate of $[a^{\dagger}-\alpha^{*}(t)][a-\alpha(t)]$ with the eigenvalue 0 is the coherent state $|\alpha(t)\rangle$.

Further insight in the relations between the kets $|n\rangle$ of the usual basis and the kets $M(\alpha(t))\,|n\rangle$ of the new basis can be gained by starting from the well-known relation

$$|n\rangle=(n!)^{-1/2}(a^{\dagger})^{n}\,|0\rangle\ , \tag{A18}$$

multiplying both sides at the left by $M(\alpha(t))$ and using (A10) to obtain

$$M(\alpha(t))\,|n\rangle=(n!)^{-1/2}[a^{\dagger}-\alpha^{*}(t)]^{n}M(\alpha(t))\,|0\rangle$$
$$=(n!)^{-1/2}[a^{\dagger}-\alpha^{*}(t)]^{n}\,|\alpha(t)\rangle\ . \tag{A19}$$

Clearly, going over from $\{|n\rangle\}$ to $\{M(\alpha(t))\,|n\rangle\}$ only requires replacing a by $a-\alpha(t)$, $a^{\dagger}$ by $a^{\dagger}-\alpha^{*}(t)$, and the vacuum state $|0\rangle$ by the coherent state $|\alpha\rangle$. In this replacement, the commutation properties of the basic operators are not changed: $[a,a^{\dagger}]=1=[a-\alpha(t),a^{\dagger}-\alpha^{*}(t)]$.

Going over from the usual time-independent basis to a time-dependent basis such as $\{M(\alpha(t))\,|n\rangle\}$ calls for some precautions and gives some new possibilities which are discussed in Appendix D. For the convenience of the reader we mention that, using the notation of Appendix D, with $\{|n\rangle\}$ as basis c and $\{M(\alpha(t))\,|n\rangle\}$ as basis b, one has the relations $W_{bc}(t)=M(\alpha(t))$ and $D_{bc}(t)=H-M(\alpha(t))HM^{\dagger}(\alpha(t))$, with H given by (A3).

If one prefers to avoid the use of a time-dependent basis, the *time-independent* basis $\{M(\alpha(t_0))\,|n\rangle\}$ still provides a very useful tool.

APPENDIX B: TWO-LEVEL ATOM

In this appendix we shall use the fiction of a two-level atom to illustrate some features of our model in a simple case.

1. The bare two-level atom

Let the two orthogonal and normalized kets $|1\rangle_A$ and $|2\rangle_A$ form a basis in the two-level atom ket space. From these two kets we can construct a convenient set of four orthogonal Hermitian operators, which spans the corresponding four-dimensional operator space, as

$$1_A=|1\rangle_{AA}\langle 1|+|2\rangle_{AA}\langle 2|\ ,$$
$$I_{XA}=\tfrac{1}{2}(|1\rangle_{AA}\langle 2|+|2\rangle_{AA}\langle 1|)\ ,$$
$$I_{YA}=i\tfrac{1}{2}(|1\rangle_{AA}\langle 2|-|2\rangle_{AA}\langle 1|)\ , \tag{B1}$$
$$I_{ZA}=\tfrac{1}{2}(|2\rangle_{AA}\langle 2|-|1\rangle_{AA}\langle 1|)\ .$$

This set contains the unit operator, and the other three operators have the same simple commutation relations as the Cartesian components of angular momentum (divided by $\hbar$), hence can be considered as the components of a pseudo-angular-momentum $\hbar\mathbf{I}_A$ for a pseudo-spin-$\frac{1}{2}$ associated with the atom in some abstract XYZ space. In gen-

eral, this abstract space has nothing to do with the usual configuration space in which one describes such things as positions of atoms, polarization and propagation of fields etc.

Any operator is completely specified by the coefficients of its expansion as a linear combination of the operators of a set such as (B1), and vice versa. For instance, the density operator $\rho_A(t)$ describing any quantum-statistical state of the two-level atom can be expressed as (using the relation $\mathrm{Tr}_A\{\rho_A\}=1$)

$$\rho_A(t)=\tfrac{1}{2}[1_A+4\langle \mathbf{I}_A\rangle(t)\cdot\mathbf{I}_A]\ , \tag{B2}$$

in terms of the real vector

$$\langle \mathbf{I}_A\rangle(t)=\mathrm{Tr}_A\{\rho_A(t)\mathbf{I}_A\}\ , \tag{B3}$$

and any Hamiltonian $H_A(t)$ for the atom can be expressed as

$$H_A(t)=[\tfrac{1}{2}\,\mathrm{Tr}_A\{H_A(t)\}]1_A+[2\,\mathrm{Tr}_A\{H_A(t)\mathbf{I}_A\}]\cdot\mathbf{I}_A\ . \tag{B4}$$

With this notation, and using the relation[16] $[\mathbf{I}_A\cdot\mathbf{a},\mathbf{I}_A\cdot\mathbf{b}]=i(\mathbf{a}\times\mathbf{b})\cdot\mathbf{I}_A$ valid for any c-number vectors $\mathbf{a}$ and $\mathbf{b}$, where $\times$ denotes a vectorial product, the quantum equation of motion $i\hbar\partial\rho_A(t)/\partial t=[H_A(t),\rho_A(t)]$ can be written as the equivalent relation

$$\frac{d}{dt}\langle \mathbf{I}_A\rangle(t)=(2/\hbar)\mathrm{Tr}_A\{H_A(t)\mathbf{I}_A\}\times\langle \mathbf{I}_A\rangle(t)\ , \tag{B5}$$

which only involves real c-number vectors of XYZ space, and describes a rotation of vector $\langle \mathbf{I}_A\rangle(t)$ at the instantaneous angular velocity $(2/\hbar)\mathrm{Tr}_A\{H_A(t)\mathbf{I}_A\}$. It is a remarkable feature of the two-level model that exact quantum-statistical predictions can be obtained from such a simple and easily visualized classical calculation.

In the case of the time-independent bare-atom Hamiltonian H_{0A}, it is convenient to take the ground state of H_{0A} as $|1\rangle_A$ and its excited state as $|2\rangle_A$, and to choose a zero of energy halfway between the two eigenvalues of H_{0A}, so that

$$H_{0A}=\hbar\omega_0 I_{ZA}\ , \tag{B6}$$

where $\omega_0=\omega_2-\omega_1$ is a positive angular velocity. We also introduce the convenient operators

$$I_{+A}=I_{XA}+iI_{YA}=|2\rangle_A\,{}_A\langle 1| \quad\text{and}\quad I_{-A}=(I_{+A})^\dagger\ . \tag{B7}$$

2. The coupled two-level atom and field system

For the two-level atom expression (2.4) for the linear atom-field coupling becomes

$$\begin{aligned}\lambda V_S(\mathbf{r}_A)=&\tfrac{1}{2}1_A\otimes\lambda\sum_k[V_{k0}^*(\mathbf{r}_A)a_k+V_{k0}(\mathbf{r}_A)a_k^\dagger]\\&+I_{ZA}\otimes\lambda\sum_k[V_{kU}^*(\mathbf{r}_A)a_k+V_{kU}(\mathbf{r}_A)a_k^\dagger]\\&+I_{+A}\otimes\lambda\sum_k[V_{kR}^*(\mathbf{r}_A)a_k+V_{kV}(\mathbf{r}_A)a_k^\dagger]\\&+I_{-A}\otimes\lambda\sum_k[V_{kV}^*(\mathbf{r}_A)a_k+V_{kR}(\mathbf{r}_A)a_k^\dagger]\ ,\end{aligned} \tag{B8}$$

where the first two terms are the contributions of $i=j$ while the last two are the contributions of $i\neq j$ in the summations over atomic states.

In the linearized electric-dipole approximation all V_{k0} and V_{kU} are identically zero and V_{kR} and V_{kV} are related by $V_{kR}=V_{kV}$. In the case of NMR, V_{k0} is identically zero and the other three coefficients are typically of the same order of magnitude for each mode k, with a notable exception for modes which are circularly polarized in the XY plane (see below). The subscripts R and V stand for the traditional denominations "real" and "virtual."

Using (B8) we have

$$\begin{aligned}\lambda Q_S^\dagger(t)[\bar V_S(t)-V_S]Q_S(t)&\\=\hbar\mathbf{I}_a\cdot\lambda\alpha\boldsymbol{\Omega}(t)\otimes 1_F+\hbar\lambda\alpha\Omega_0(t)1_A\otimes 1_F\ ,&\end{aligned} \tag{B9}$$

where

$$\Omega_0(t)=(1/2\hbar)\sum_k V_{k0}\alpha_k^*(t_0)e^{i\omega_k(t-t_0)}+\text{c.c.}\ , \tag{B10}$$

and the pseudo-angular-velocity $\lambda\alpha\boldsymbol{\Omega}(t)$ is given by

$$\lambda\alpha\boldsymbol{\Omega}(t)=\lambda\alpha\boldsymbol{\Omega}_R(t)+\lambda\alpha\boldsymbol{\Omega}_V(t)+\lambda\alpha\boldsymbol{\Omega}_U(t)\ , \tag{B11}$$

where $\boldsymbol{\Omega}_R(t)$, $\boldsymbol{\Omega}_V(t)$, and $\lambda\boldsymbol{\Omega}_U(t)$ are real vectors of XYZ space, with components given by

$$\begin{aligned}\Omega_{R+}(t)&=\Omega_{RX}(t)+i\Omega_{RY}(t)\\&=(2/\hbar)\sum_k V_{kR}\alpha_k^*(t_0)e^{i(\omega_k-\omega_0)(t-t_0)}\ ,\\\Omega_{V+}(t)&=\Omega_{VX}(t)+i\Omega_{VY}(t)\\&=(2/\hbar)\sum_k V_{kV}^*\alpha_k(t_0)e^{-i(\omega_k+\omega_0)(t-t_0)}\ ,\\\Omega_{UZ}(t)&=(1/\hbar)\sum_k V_{kU}\alpha_k^*(t_0)e^{i\omega_k(t-t_0)}+\text{c.c.}\ ,\\\Omega_{RZ}(t)&=\Omega_{VZ}(t)=\Omega_{UX}(t)=\Omega_{UY}(t)=0\ ,\end{aligned} \tag{B12}$$

(c.c. means complex conjugate). In these relations, all quantities of the type V_{kR}, etc., are tacitly evaluated at the location $\mathbf{r}_A$ of the atom.

Using (B9), the equation of motion for $\tilde\rho_S(t)$ becomes

$$\begin{aligned}i\hbar\frac{\partial}{\partial t}\tilde\rho_S(t)=[\{&\lambda Q_S^\dagger(t)V_S Q_S(t)\\&+\hbar\mathbf{I}_A\cdot\lambda\alpha\boldsymbol{\Omega}(t)\otimes 1_F\},\tilde\rho_S(t)]\ .\end{aligned} \tag{B13}$$

[The last term in (B9) leads to a vanishing contribution in the commutator in (2.49)].

The initial condition, given by (2.50), is easily obtained explicitly, to first order in λ, using (3.12), (3.14), and the fact that (3.15) becomes here

$$W_S^{(1)}=\sum_k\left\{\frac{-V_{kV}}{\hbar(\omega_0+\omega_k)}I_{+A}\otimes a_k^\dagger+\frac{(V_{kU}-V_{k0})/2}{\hbar\omega_k}(|1\rangle_A\,{}_A\langle 1|)\otimes a_k^\dagger\right\}\ . \tag{B14}$$

3. The classical-field approximation

Using (B9)—(B13), the equations of motion (3.1) and (3.4) can be written in the compact forms

$$i\hbar\frac{\partial}{\partial t}\tilde{\rho}_A^B(t)=[\hbar\mathbf{I}_A\cdot\lambda\alpha\boldsymbol{\Omega}(t),\tilde{\rho}_A^B(t)]\ ,$$

$$i\hbar\frac{\partial}{\partial t}B_A(t,t_0)=[\hbar\mathbf{I}_A\cdot\lambda\alpha\boldsymbol{\Omega}(t)+\hbar\lambda\alpha\Omega_0(t)1_A]B_A(t,t_0)\ . \tag{B15}$$

The behavior of the solutions can be visualized easily by introducing the real vector of XYZ space

$$\begin{aligned}\langle QIQ^\dagger\rangle^B(t)&=\langle Q_A(t)\mathbf{I}_AQ_A^\dagger(t)\rangle^B(t)\\&=\mathrm{Tr}_A\{\rho_A^B(t)Q_A(t)\mathbf{I}_AQ_A^\dagger(t)\}\\&=\mathrm{Tr}_A\{\tilde{\rho}_A^B(t)\mathbf{I}_A\}\ ,\end{aligned} \tag{B16}$$

where $\langle QIQ^\dagger\rangle^B(t)$ is a shorthand notation. When ρ_A^B is a pure state (i.e., a projector), the length of the vector is $\frac{1}{2}$. Smaller lengths correspond to mixed states and lengths above $\frac{1}{2}$ are impossible. The density operator is given by [see (B2)]

$$\tilde{\rho}_A^B(t)=\tfrac{1}{2}[1_A+4\langle QIQ^\dagger\rangle^B(t)\cdot\mathbf{I}_A]\ , \tag{B17}$$

and the equation of motion for $\langle QIQ^\dagger\rangle^B(t)$ is the Bloch equation without relaxation,

$$\frac{d}{dt}\langle QIQ^\dagger\rangle^B(t)=\lambda\alpha\boldsymbol{\Omega}(t)\times\langle QIQ^\dagger\rangle^B(t)\ , \tag{B18}$$

where the time derivative is for an observer which moves with the XYZ frame of reference. The initial condition (3.5) takes the form

$$\langle QIQ^\dagger\rangle^B(t_i)=-\tfrac{1}{2}\hat{\mathbf{Z}}\ , \tag{B19}$$

where the caret denotes a unit vector.

Equation (B18) describes the motion of the classical vector $\langle QIQ^\dagger\rangle^B(t)$ as a rotation at the known angular velocity $\lambda\alpha\boldsymbol{\Omega}(t)$ with respect to the XYZ frame. Solutions of (B18) and (B19) have been discussed extensively in the literature of NMR and quantum optics, so that only a few remarks will be made here.

In a typical experiment in pulse spectroscopy, the pulse is approximately at the resonance frequency ω_0 of the atom, its duration is much longer than the atomic period $2\pi/\omega_0$, and the strength of the atom-pulse coupling is much smaller than the bare-atom Hamiltonian (i.e., $\lambda\alpha\,|\,\boldsymbol{\Omega}(t)\,|\ll\omega_0$ at all times). Under these circumstances $\boldsymbol{\Omega}_R(t)$ varies with time at frequencies much lower than ω_0, $\boldsymbol{\Omega}_U(t)$ oscillates at frequencies close to ω_0 and $-\omega_0$, and $\boldsymbol{\Omega}_V(t)$ oscillates at frequencies close to $2\omega_0$ and $-2\omega_0$. As a consequence, a useful (although sometimes misleading) approximation for the solution of (B18) for not-too-long times can be obtained by first dropping the "fast oscillating" parts $\boldsymbol{\Omega}_V$ and $\boldsymbol{\Omega}_U$ of $\boldsymbol{\Omega}$, and then solving (B18) with $\boldsymbol{\Omega}(t)$ replaced by the remnant $\boldsymbol{\Omega}_R(t)$ of this truncation. Usually this makes the equation soluble analytically, or, at least, amenable to numerical methods. If the above approximation [i.e., keeping only $\boldsymbol{\Omega}_R(t)$] is not accurate enough, it can be improved in a particularly efficient way by using the Magnus transformation.[8] Of course, the rotating-wave approximation would also replace $\boldsymbol{\Omega}(t)$ by $\boldsymbol{\Omega}_R(t)$, but this procedure would involve a major truncation of the atom-field coupling Hamiltonian itself, with far-reaching consequences (see Sec. VI). These remarks also apply in the case of a multilevel atom.

4. A simple example from Zeeman spectroscopy

Let us briefly discuss the application of these ideas to the case of NMR of a single spin $\frac{1}{2}$, in which XYZ space and ordinary configuration space are directly related. The spin Hamiltonian can be written as $-\gamma\hbar\mathbf{I}_A\cdot(\mathbf{B}_0+\mathbf{B}_{\mathrm{cl}}(t))$, where γ is the magnetogyric ratio of the spin, $\mathbf{B}_0$ is the constant magnetic induction which is the source of H_{0A}, and $\mathbf{B}_{\mathrm{cl}}(t)$ is the irradiation magnetic induction at the location of the nucleus. In order to satisfy (B6), the unit vector $\hat{\mathbf{Z}}$ must point in the direction of the vector $-\gamma\mathbf{B}_0$, so that $\omega_0=-\gamma\mathbf{B}_0\cdot\hat{\mathbf{Z}}$ is positive. The supplementary term $\lambda[\bar{V}_S(t)-V_S]$, which appears in the equation of motion (2.35) of $\bar{\rho}_S(t)$, is given here by $-\gamma\hbar\mathbf{I}_A\cdot\mathbf{B}_{\mathrm{cl}}(t)\otimes 1_F$. The following relations are helpful in going over to the tilde version (3.1) of the equation of motion:

$$\begin{aligned}&Q_A^\dagger(t)I_{\pm A}Q_A(t)=I_{\pm A}\exp[\pm i\omega_0(t-t_0)]\ ,\\&Q_A^\dagger(t)I_{ZA}Q_A(t)=I_{ZA}\ ,\\&\mathbf{I}_a\cdot\mathbf{B}_{\mathrm{cl}}(t)=\tfrac{1}{2}[I_{+A}B_{\mathrm{cl}-}(t)+I_{-A}B_{\mathrm{cl}+}(t)]\\&\qquad\qquad+I_{ZA}B_{\mathrm{cl}Z}\end{aligned}$$

with

$$B_{\mathrm{cl}\pm}(t)=B_{\mathrm{cl}X}(t)\pm iB_{\mathrm{cl}Y}(t)\ . \tag{B20}$$

The angular velocity $\lambda\alpha\boldsymbol{\Omega}(t)$ can be evaluated from the relation

$$Q_A^\dagger(t)\{-\gamma\hbar\mathbf{I}_A\cdot\mathbf{B}_{\mathrm{cl}}(t)\}Q_A(t)=\hbar\mathbf{I}_A\cdot\lambda\alpha\boldsymbol{\Omega}(t)\ .$$

A particularly simple situation arises when $\mathbf{B}_{\mathrm{cl}}(t)$ results from the amplitude modulation of a wave of constant frequency, circularly polarized in the XY plane,

$$\begin{aligned}B_{\mathrm{cl}+}(t)&=B_{\mathrm{cl}X}(t)+iB_{\mathrm{cl}Y}(t)\\&=h(t)e^{i\varphi}e^{i\omega(t-t_0)}\ ,\end{aligned} \tag{B21}$$

where $h(t)$ is the modulation amplitude, φ is the phase at time t_0, and $\omega\hat{\mathbf{Z}}$ is the angular velocity of rotation of $\mathbf{B}_{\mathrm{cl}}(t)$ around $\hat{\mathbf{Z}}$ (with the sign of ω giving the sense of rotation). Then, $\boldsymbol{\Omega}(t)$ is given by

$$\begin{aligned}&\lambda\alpha\Omega_+(t)=-\gamma h(t)e^{i\varphi}e^{i(\omega-\omega_0)(t-t_0)}\ ,\\&\Omega_Z(t)=0\ .\end{aligned} \tag{B22}$$

In the simple case where $\omega=\omega_0$ and $h(t)$ is any real function,

$$\lambda\alpha\boldsymbol{\Omega}(t)=-\gamma h(t)\hat{\boldsymbol{\varphi}}\ , \tag{B23}$$

where the time-independent unit vector $\hat{\boldsymbol{\varphi}}=\hat{\mathbf{X}}\cos\varphi+\hat{\mathbf{Y}}\sin\varphi$, lying in the XY plane, describes the phase of the irradiation field, the exact solution of (B18) satisfying (B19) is

$$\langle QIQ^\dagger\rangle_X^B(t)+i\langle QIQ^\dagger\rangle_Y^B(t)=(i/2)e^{i\varphi}\sin\theta(t)\ ,$$
$$\langle QIQ^\dagger\rangle_Z^B(t)=-\tfrac{1}{2}\cos\theta(t)\ , \tag{B24}$$

where the angle $\theta(t)$ of rotation around $\hat{\varphi}$ is given by

$$\theta(t)=\int_{t_0}^{t}-\gamma h(t')dt'\ . \tag{B25}$$

When $\theta=(\pi/2+n\pi)$ for any integer n, the vector $\langle QIQ^\dagger\rangle^B$ lies in the XY plane, hence the two atomic states are equally populated and the quantum average of $|2\rangle_{AA}\langle 1|$ is maximum in absolute value. When $\theta=(\pi+2n\pi)$, the atomic populations have been interchanged and, when $\theta=2n\pi$, the atom goes back to its initial situation.

When $\omega\neq\omega_0$, an exact analytical solution of (B18) is available in a few other particular situations.[17] In all other cases, approximations have to be used (including numerical methods).

In this example, one sees clearly that the simple form (B23) of $\lambda\alpha\boldsymbol{\Omega}(t)$ results from a realistic (although particular) choice of the bare atom (NMR) *and* of the exciting classical field $f_{\rm cl}(\mathbf{r}_A,t)$ and is *not* the consequence of some truncation of the atom-field coupling Hamiltonian such as, for instance, the rotating-wave approximation (RWA) in which all terms for the atom-field coupling Hamiltonian V_S [see (B8) for instance] involving V_{kV} or V_{kU} are replaced by zero. This remark is well illustrated in the case where the field [see (2.7)] described by (B21) involves a single mode k', circularly polarized and propagating parallel to the Z direction for a plane wave. Then $h(t)$ is a constant for all times. If we compare (B12) and (B22), keeping in mind that ω_0 and $\omega_{k'}=|\omega|$ are both positive whereas the sign of ω indicates the sense of rotation of $\mathrm{Re}\{\mathbf{b}_{k'}(\mathbf{r}_A)\alpha_{k'}(t)\}$, we conclude that $V_{k'V}=0$ if the field rotates in the same sense as the free precession of the spin ($\omega_{k'}$ positive) and that $V_{k'R}=0$ for the opposite sense of rotation, in agreement with the well-known selection rules.

In the case of NMR there is a direct relation between XYZ space and ordinary inertial configuration space, which can be visualized in two useful ways. In the first visualization, the XYZ frame rotates with respect to the inertial frame at the angular velocity $\omega_0\hat{\mathbf{Z}}$ of the free precession of the spin, $\hbar\langle QIQ^\dagger\rangle^B(t)$ is the averaged spin angular momentum *as seen from from the rotating XYZ frame,* and $-(1/\gamma)\lambda\alpha\boldsymbol{\Omega}(t)$ is the magnetic induction $\mathbf{B}_{\rm cl}(t)$. In the alternative visualization, the XYZ frame is at rest with respect to the inertial frame, $\hbar\langle\mathbf{I}\rangle^B(t)$ is the (averaged) spin angular momentum so that $\hbar\langle QIQ^\dagger\rangle^B(t)$ is $\hbar\langle\mathbf{I}\rangle^B(t)$ *rotated back* at the angular velocity $-\omega_0\hat{\mathbf{Z}}$ to undo the effect of H_{0A}, and $-(1/\gamma)\lambda\alpha\boldsymbol{\Omega}(t)$ is the magnetic induction $\mathbf{B}_{\rm cl}(t)$ similarly rotated back at the angular velocity $-\omega_0\hat{\mathbf{Z}}$.

APPENDIX C: MISCELLANEOUS CALCULATIONS

1. Commutation properties of H_{0F}, $M_F(t)$, and $Q_F(t)$

The commutation properties (2.27)—(2.29) involving the time-displacement operator $Q_F(t,t_0)$ can be rewritten[6,7] as similar relations involving the single-time operator $Q_F(t)$, keeping in mind that $Q_F(t)$ still depends on t_0 parametrically and that (2.27)—(2.29) remain valid if t and t_0 are interchanged. The result is the following (the operators U_{cF} introduced in Appendix D have not been written explicitly because they are irrelevant when all bases used are immobile with respect to each other):

$$\begin{aligned}&M_F(t)Q_F(t)=Q_F(t)M_F(t_0)\ ,\\ &Q_F^\dagger(t)M_F(t)=M_F(t_0)Q_F^\dagger(t)\ ,\\ &[H_{0F},M_F(t)]Q_F(t)=Q_F(t)[H_{0F},M_F(t_0)]\ ,\\ &Q_F^\dagger(t)[H_{0F},M_F(t)]=[H_{0F},M_F(t_0)]Q_F^\dagger(t)\ ,\end{aligned} \tag{C1}$$

and similar relations with $M_F(t)$ and $M_F(t_0)$ replaced, respectively, by $M_F^\dagger(t)$ and $M_F^\dagger(t_0)$.

2. Derivation of (4.9)

We start from (4.8) and use the commutation relations (C1) for the operators Q_F, M_F, and H_{0F} to move successively the operators $Q_S(t)$, $B_S(t)$, and $Q_S^\dagger(t_i)$ to the left until they cancel with their counterparts $Q_S^\dagger(t)$, $B_S^\dagger(t)$, and $Q_S(t_i)$. As a result, the operator outside the inner curly bracket can be written as

$$\begin{aligned}&1_A\otimes M_F^\dagger(t_i)H_{0F}M_F(t_i)\\ &\quad=1_A\otimes\sum_k\hbar\omega_k[a_k^\dagger+\alpha_k^*(t_i)][a_k+\alpha_k(t_i)]\ .\end{aligned} \tag{C2}$$

The only remaining operators which do not act trivially in the atomic state space are $W_S^{(1)}$ and $(W_S^{(1)})^\dagger$, hence only the terms $j=1$ in expression (3.15) contribute to the trace. The only nonvanishing contributions to the trace over the field state space arise from the product of each a_k from $(W_S^{(1)})^\dagger$ with the corresponding $a_k^\dagger$ from (C2), and similar combinations for $W_S^{(1)}$, with the result that (4.8) can be written under the form (4.9).

3. Derivation of (4.12)

We start from (4.11), replace $H_{0F}M_F(t)$ by $M_F(t)H_{0F}+[H_{0F},M_F(t)]$ and use

$$\mathrm{Tr}\{E[F,G]\}=\mathrm{Tr}\{G[E,F]\}\ . \tag{C3}$$

As explained in the main text, only $[H_{0F},M_F(t)]$ gives a nonvanishing contribution. Moving $B_A^\dagger(t')$ and $Q_A^\dagger(t')$ through the long string of field operators, one obtains

$$\begin{aligned}K_5=(1/i\hbar)\int_{t_i}^{t}dt'\mathrm{Tr}_S\{&B_A(t')|1\rangle_{AA}\langle 1|B_A^\dagger(t')\otimes|0\rangle_{FF}\langle 0|\\ &\times[1_A\otimes Q_F^\dagger(t)M_F^\dagger(t)[H_{0F},M_F(t)]Q_F(t),Q_S^\dagger(t')\lambda V_SQ_S(t')]\}\ .\end{aligned} \tag{C4}$$

To go from this to (4.12), write the outer commutator explicitly, use the commutation relations (C1) repeatedly to

move M_F and $[H_{0F},M_F]$ close to λV_S, insert the unit operator $1=M_F^\dagger M_F=M_F M_F^\dagger$ when necessary, use (2.24) and note that V_S is time independent (hence $\partial\lambda V_S/\partial t'=0$).

4. Derivation of (4.13)

Using (3.12), (2.32), and (2.36), the right-hand part of the operator in the trace in (4.12) can be written as $Q_S^\dagger(t')\{\partial[-\bar{V}_S(t')+V_S]/\partial t'\}Q_S(t')$, which can be further written as $-\partial\{Q_S^\dagger(t')[\bar{V}_S(t')-V_S]Q_S(t')\}/\partial t'$ plus terms involving time derivatives of Q_S and $Q_S^\dagger$. Using (2.36) and (2.46), the result is

$$K_5=\int_{t_i}^{t}dt'\mathrm{Tr}_S\left\{B_A(t')\,|\,1\rangle_{A\,A}\langle 1\,|\,B_A^\dagger(t')\otimes\,|\,0\rangle_{F\,F}\langle 0\,|\right.$$
$$\left.\times\left\{-\frac{\partial}{\partial t'}\{Q_S^\dagger(t')\lambda(\bar{V}_S(t')-V_S)Q_S(t')\}+(1/i\hbar)[Q_S^\dagger(t')\lambda(\bar{V}_S(t')-V_S)Q_S(t'),H_{0A}\otimes 1_F+1_A\otimes H_{0F}]\right\}\right\}. \tag{C5}$$

The contribution of the first term in the inner large braces is evaluated easily by taking the operators $B_A(t')$ and $B_A^\dagger(t')$ inside the derivative with respect to t' and showing that the correcting terms cancel. Then one is left with the first term in (4.13). The contribution from the second term in the inner large braces is shown to be equal to the second term in (4.13) by noting that the terms involving $1_A\otimes H_{0F}$ cancel in the commutator, using (3.5) and (2.36).

5. Derivation of (5.19)

Equation (2.36) for $\lambda[\bar{V}(t)-V]_A$ shows that the only contributions of order $\lambda\alpha$ to $\tilde{\rho}_A^B(t')$ are of the types $|\,j\rangle_{A\,A}\langle 1\,|$ and $|\,1\rangle_{A\,A}\langle j\,|$, and, using (3.5), one obtains for $j\neq 1$,

$$\langle\,|\,1\rangle_{A\,A}\langle j\,|\,\rangle^B(t')=\mathrm{Tr}_A\{Q_A^\dagger(t')\,|\,1\rangle_{A\,A}\langle j\,|\,Q_A(t')\tilde{\rho}_A^B(t')\}$$
$$=(\lambda\alpha)e^{-i(\omega_j-\omega_1)(t'-t_0)}(1/i\hbar)\int_{t_i}^{t'}dt''e^{i(\omega_j-\omega_1)(t''-t_0)}V_{j1}(t'')+O((\lambda\alpha)^2)\,. \tag{C6}$$

Noting that $\langle\,|\,j\rangle_{A\,A}\langle 1\,|\,\rangle^B(t')=\{\langle\,|\,1\rangle_{A\,A}\langle j\,|\,\rangle^B(t')\}^*$, and using (5.13), we can now write the contribution in $(\lambda\alpha)^2$ to $J^{(0)}(\omega,t)$ in the form K_8,

$$K_8=2(\lambda\alpha)^2\omega(1/\hbar)\mathrm{Im}\left[\sum_{j(\neq 1)}\left\{-iG_{j1}(\omega,t_0)\int_{t_i}^{t}dt'e^{i\omega(t'-t_0)}e^{-i(\omega_j-\omega_1)(t'-t_0)}\int_{t_i}^{t'}dt''e^{i(\omega_j-\omega_1)(t''-t_0)}V_{j1}(t'')\right.\right.$$
$$\left.\left.+iG_{1j}(\omega,t_0)\int_{t_i}^{t}dt'e^{i\omega(t'-t_0)}e^{i(\omega_j-\omega_1)(t'-t_0)}\int_{t_i}^{t'}dt''e^{-i(\omega_j-\omega_1)(t''-t_0)}V_{1j}(t'')\right\}\right]. \tag{C7}$$

Expression K_8 can be evaluated in the limit $t\to\infty$ by the following procedure: (i) write the double integrals as an outer integral over t'' from t_i to t and an inner integral over t' from t'' to t, (ii) evaluate the inner integral which is easy because the t' dependence is through exponentials only, (iii) take the limit of the inner integral for $t\to\infty$ with the usual result

$$\lim_{t\to\infty}\left\{\int_{t''}^{t}dt'e^{i\omega_0 t''}e^{i(\omega-\omega_0)t'}\right\}$$
$$=e^{i\omega t''}\{\pi\delta(\omega-\omega_0)+i\mathscr{P}[1/(\omega-\omega_0)]\}\,, \tag{C8}$$

where $\mathscr{P}$ denotes a principal part and ω_0 may be $(\omega_j-\omega_1)$ or $(\omega_1-\omega_j)$, (iv) use (5.7) to evaluate the outer integral in which t_i may be replaced by $-\infty$ because the $V_{ij}(t)$ differ from zero only during the overlap between the atom and the classical-field pulse, (v) note that ω and $(\omega_j-\omega_1)$ are non-negative quantities. This leads to (5.19).

6. Exact evaluation of (6.10) for a particular choice of interaction, detector, and optics

We evaluate here the function (6.10) in the particular case described in Sec. VI, immediately before (6.11).

When the field evolves in empty space with periodic boundary conditions in a cube of side L, a convenient basis of normal modes is given by linearly polarized plane traveling waves with real polarization unit vectors $\hat{\mathbf{s}}_\mathbf{k}$,

$$\mathbf{e}_k(\mathbf{r})=\mathbf{e}_{\mathbf{k}s}(\mathbf{r})=(\hbar\omega_\mathbf{k}/2L^3)^{1/2}\hat{\mathbf{s}}_\mathbf{k}e^{i\mathbf{k}\cdot\mathbf{r}}\,, \tag{C9}$$

where $\omega_\mathbf{k}=c\,|\,\mathbf{k}\,|$, the x, y, and z components of $\mathbf{k}$ are integral multiples of $2\pi/L$, and $\mathbf{k}$ and the two corresponding $\hat{\mathbf{s}}_\mathbf{k}$ are mutually orthogonal.

Combining (C9), (2.10), (6.4), and (6.11), we obtain

$$D^-_{\mathbf{k}s\mathbf{k}'s'}(\mathbf{r}_D)=K(\hbar/8L^3)(\omega_\mathbf{k}\omega_{\mathbf{k}'})^{1/2}\hat{\mathbf{s}}_\mathbf{k}\cdot\hat{\mathbf{s}}_{\mathbf{k}'}e^{-i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}_D}\,,$$
$$D^0_{\mathbf{k}s\mathbf{k}'s'}(\mathbf{r}_D)=K(\hbar/8L^3)(\omega_\mathbf{k}\omega_{\mathbf{k}'})^{1/2}\hat{\mathbf{s}}_\mathbf{k}\cdot\hat{\mathbf{s}}_{\mathbf{k}'}e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_D}\,. \tag{C10}$$

Combining (C9), (2.10), and (2.5), we also obtain, for the linearized electric-dipolar coupling,

$$V_{\mathbf{k}sij}(\mathbf{r}_A)=-(\hbar/8L^3)^{1/2}(\omega_\mathbf{k})^{1/2}$$
$$\times\,{}_A\langle j\,|\,\mu_{eA}\,|\,i\rangle_A\cdot\hat{\mathbf{s}}_\mathbf{k}e^{-i\mathbf{k}\cdot\mathbf{r}_A}\,, \tag{C11}$$

with $V_{ksii}=0$ because all diagonal matrix elements of μ_{eA} are equal to zero in a representation which diagonalizes H_{0A}. A first consequence of this last property is that the contribution K_9 to $\lambda\langle D_S\rangle^{(1)}(t)$ arising from the initial contribution in (3.18) for $\hat{\rho}_S^{(1)}(t)$ is exactly zero for the electric-dipolar coupling because $D_S=1_A\otimes D_F$ acts trivially in the atomic state space. A second consequence is that all contributions with $i=j$ in (6.9) and (6.10) are equal to zero.

To evaluate the sum over $\mathbf{k}$, we introduce a frame of reference based on the orthogonal vectors $\mathbf{U}$, $\mathbf{V}$, and $\mathbf{R}$ (and the corresponding unit vectors $\hat{\mathbf{U}}$, $\hat{\mathbf{V}}$, and $\hat{\mathbf{R}}$) defined by

$$\begin{aligned}&\mathbf{R}=\mathbf{r}_D-\mathbf{r}_A\ ,\\ &\mathbf{U}=\langle i\,|\,\mu_{eA}\,|\,j\rangle-\hat{\mathbf{R}}(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{R}})\ ,\\ &\mathbf{V}=\hat{\mathbf{s}}_r-\hat{\mathbf{R}}(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{R}})-\hat{\mathbf{U}}(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{U}})\ ,\end{aligned}\tag{C12}$$

in which the direction of vector $\mathbf{k}$ is specified by the usual polar angles θ and φ, such that $\hat{\mathbf{k}}\cdot\hat{\mathbf{R}}=\cos\theta$, $\hat{\mathbf{k}}\cdot\hat{\mathbf{U}}=\sin\theta\cos\varphi$ and $\hat{\mathbf{k}}\cdot\hat{\mathbf{V}}=\sin\theta\sin\varphi$. Using (C10) and (C11), we can write (6.10) in the form

$$\begin{aligned}&T_{ij}(\mathbf{r}_A,\mathbf{r}_D,t-t')\\ &\quad=K(\hbar/8L^3)^{3/2}(\omega_r)^{1/2}e^{-i\mathbf{k}_r\cdot\mathbf{r}_D}\\ &\qquad\times\sum_{\mathbf{k}}\omega_{\mathbf{k}}\left\{\sum_s(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_{\mathbf{k}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{s}}_{\mathbf{k}})\right\}\\ &\qquad\times(e^{i\mathbf{k}\cdot(\mathbf{r}_A-\mathbf{r}_D)}e^{i\omega_{\mathbf{k}}(t-t')}-\text{c.c.})\ ,\end{aligned}\tag{C13}$$

where the reference mode has propagation vector $\mathbf{k}_r$ and polarization unit vector $\hat{\mathbf{s}}_r$. Using the mutual orthogonality of $\mathbf{k}$ and the two corresponding polarization unit vectors, one can show easily (using these three directions as a system of coordinates for instance) that the sum over polarizations in (C13) is given by

$$\begin{aligned}K_{10}&=\sum_{s=1,2}(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_{\mathbf{k}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{s}}_{\mathbf{k}})\\ &=\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_r-(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{k}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{k}})\ ,\end{aligned}\tag{C14}$$

where $\hat{\mathbf{k}}$ is a unit vector pointing in the direction of $\mathbf{k}$. Using the UVR frame of coordinates, (C14) can be written as

$$\begin{aligned}K_{10}&=\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_r-(\cos\theta)^2(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{R}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{R}})\\ &\quad-[1-(\cos\theta)^2]\{(\cos\varphi)^2(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{U}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{U}})\\ &\qquad+(\sin\varphi)^2(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{V}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{V}})\}\\ &\quad+(\cdots)\ ,\end{aligned}\tag{C15}$$

where the ellipsis contains terms in $\sin\varphi$, $\cos\varphi$, and $\sin\varphi\cos\varphi$, and the specification of $\mathbf{k}$ appears only in the angles θ and φ.

In the limit of $L\to\infty$ we can now replace the sum over $\mathbf{k}$ by an integral over the continuous variable $\mathbf{k}$ and proceed with the evaluation of T_{ij} as given by (C13) and (C15). The integral over φ is trivial and the integral over $\cos\theta$ is easily evaluated. In the evaluation of the integral over $|\,k\,|$, use is made of the relation $\omega_k=c\,|\,k\,|$, and the dummy variable ω can be equal to ω_k or to $-\omega_k$. The integral over $|\,k\,|$ will be separated in three parts, according to the dependence in $|\,R\,|=|\,\mathbf{r}_D-\mathbf{r}_A\,|$. The contribution in $|\,R\,|^{-3}$ involves an integral of the type

$$\begin{aligned}&\int_0^{+\infty}d\,|\,k\,|\;|\,k\,|^2\omega_k(e^{i\omega_k(t-t')}-e^{-i\omega_k(t-t')})\frac{2}{(i\,|\,k\,|\,|\,R\,|)^3}(e^{i\,|\,k\,|\,|\,R\,|}-e^{-i\,|\,k\,|\,|\,R\,|})\\ &\qquad=-(2i/|\,R\,|^3)\int_{-\infty}^{+\infty}d\omega(e^{i\omega(t-t'-|\,R\,|/c)}-e^{i\omega(t-t'+|\,R\,|/c)})\\ &\qquad=-(4\pi i/|\,R\,|^3)[\delta(t-t'-|\,R\,|/c)-\delta(t-t'+|\,R\,|/c)]\ .\end{aligned}\tag{C16}$$

The contributions in $|\,R\,|^{-2}$ and $|\,R\,|^{-1}$ involve similar integrals which are easily evaluated with the help of the relation

$$\frac{\partial^n}{\partial y^n}[2\pi\delta(y)]=\frac{\partial^n}{\partial y^n}\int_{-\infty}^{+\infty}d\omega\,e^{i\omega y}=i^n\int_{-\infty}^{+\infty}d\omega\,\omega^n e^{i\omega y}\tag{C17}$$

for $n=1$ and 2.

Combining the above relations from (6.11) on, we obtain, for a free electromagnetic field,

$$\begin{aligned}&T_{ij}(\mathbf{r}_A,\mathbf{r}_D,t-t')=K(\hbar\omega_r/2L^3)^{1/2}(i\hbar/32\pi)e^{-i\mathbf{k}_r\cdot\mathbf{r}_D}\\ &\qquad\times\left[-(1/c^2\,|\,R\,|)\left\{\frac{\partial^2}{\partial t^2}\delta(t-t'-|\,R\,|/c)-\frac{\partial^2}{\partial t^2}\delta(t-t'+|\,R\,|/c)\right\}\right.\\ &\qquad\times\{(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{s}}_r)-2(\langle i\,|\,\mu_{eA}\,|\,j\rangle\cdot\hat{\mathbf{R}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{R}})\}\end{aligned}$$

$$+\left\{(1/c\,|R|^2)\left\{\frac{\partial}{\partial t}\delta(t-t'-|R|/c)-\frac{\partial}{\partial t}\delta(t-t'+|R|/c)\right\}\right.$$

$$-(1/|R|^3)\{\delta(t-t'-|R|/c)-\delta(t-t'+|R|/c)\}$$

$$\left.\times\{(\langle i|\mu_{eA}|j\rangle\cdot\hat{\mathbf{s}}_r)-3(\langle i|\mu_{eA}|j\rangle\cdot\hat{\mathbf{R}})(\hat{\mathbf{s}}_r\cdot\hat{\mathbf{R}})\}\right]. \tag{C18}$$

Let us stress that the calculations that lead to this result involve no approximation, and that the δ functions (and their derivatives) in (C18) are obtained without any *ad hoc* extension of integration range or neglect of fast oscillating terms.

APPENDIX D: THE USE OF TIME-DEPENDENT BASES IN QUANTUM MECHANICS

It is traditional in classical mechanics to use a frame of reference specially adapted to the problem at hand, or even to discuss various aspects of the same problem using different frames of reference moving with respect to each other. Of course, this procedure introduces the minor complication that the time derivatives of nonscalar quantities depend upon the frame of reference used, but this is often compensated by considerable simplifications of the equations and improvements in the qualitative understanding of phenomena. Surprisingly, a similar procedure is almost never used, in the strict sense, as far as the state space of quantum mechanics is concerned, in spite of the fact that "absolute rest" is not a valid concept in quantum state space any more than it is in ordinary configuration space (if we use two inertial frames moving with respect to each other for the description of a position observable, the two corresponding bases in quantum state space obtained by the usual quantization procedure also move with respect to each other). However, we have found "moving" bases in state space quite useful and this appendix is devoted to an elementary discussion of some of their properties, in the simple case of discrete bases.

1. Representations

As a starting point, we choose a basis b in ket space, which is a collection of normalized kets $|b_i(t)\rangle$ which, at any time t, satisfies the orthonormality condition

$$\langle b_i(t)|b_j(t)\rangle=\delta_{i,j}\ , \tag{D1}$$

and the closure relation

$$\sum_i |b_i(t)\rangle\langle b_i(t)|=1\ , \tag{D2}$$

where 1 denotes the unit operator. If we take a ket $|\psi(t)\rangle$ and multiply it from the left by the closure relation (D2), we obtain the relation

$$|\psi(t)\rangle=1\,|\psi(t)\rangle=\sum_i |b_i(t)\rangle\langle b_i(t)|\psi(t)\rangle\ , \tag{D3}$$

which expresses ("represents") $|\psi(t)\rangle$ as a linear combination of the b basis kets $|b_i(t)\rangle$. This procedure is easily extended to linear operators $A(t)$ involving a single time. Such operators are defined by the linear relation between $|\psi(t)\rangle$ and $|\varphi(t)\rangle=A(t)|\psi(t)\rangle$ for any $|\psi(t)\rangle$. When a basis $\{|b_i(t)\rangle\}$ has been chosen in ket space, it is convenient to use the corresponding single-time level-shift basis $\{|b_i(t)\rangle\langle b_j(t)|\}$ in operator space. A representation of operator $A(t)$ as a linear combination of the basis operators $|b_i(t)\rangle\langle b_j(t)|$ is easily obtained by multiplying $A(t)$ with the closure relation (D2) from both left *and* right,

$$A(t)=\sum_{i,j}|b_i(t)\rangle\langle b_j(t)|\langle b_i(t)|A(t)|b_j(t)\rangle\ . \tag{D4}$$

The extension of this procedure to Liouville operators (or superoperators) involving a single time, and further on, is straightforward.

Time displacement operators, like evolution operators for instance, deserve a somewhat more elaborate treatment. Such operators $K(t_1,t_0)$ are defined by the linear relation between the ket $|\psi(t_0)\rangle$ at time t_0 and the ket

$$|\varphi(t_1)\rangle=K(t_1,t_0)|\psi(t_0)\rangle \tag{D5}$$

at the different time t_1, for any ket $|\psi(t_0)\rangle$. In this case it is convenient to construct from the basis $\{|b_i(t)\rangle\}$ of ket space a "time displacement level-shift" basis $\{|b_i(t_1)\rangle\langle b_i(t_0)|\}$ in operator space, and a representation of $K(t_1,t_0)$ is easily obtained by multiplication from the left with (D2) at time t_1 and from the right with (D2) at time t_0,

$$K(t_1,t_0)=\sum_{i,j}|b_i(t_1)\rangle\langle b_j(t_0)|\langle b_i(t_1)|K(t_1,t_0)|b_j(t_0)\rangle\ . \tag{D6}$$

The extension of this procedure to Liouville operators ("superoperators"), and further on, is also straightforward (note that the maximum number of different times involved increases by a factor of 2 at each step of such an extension).

2. Immobility as seen from a basis

Quite naturally, a ket $|\psi(t)\rangle$ is called immobile (or constant) *as seen from basis b* if all the projections of $|\psi(t)\rangle$ on this basis are time independent. Hence, any ket $|\psi(t)\rangle$ which is *immobile as seen from basis b* can be expressed in the form

$$|\psi(t)\rangle=\sum_i |b_i(t)\rangle\langle b_i(t)|\psi(t)\rangle$$
$$=\sum_i |b_i(t)\rangle\langle b_i(t_0)|\psi(t_0)\rangle$$
$$=U_b(t,t_0)|\psi(t_0)\rangle\ , \tag{D7}$$

where t_0 is some fixed time, and the unitary time displacement operator

$$U_b(t,t_0)=\sum_i |b_i(t)\rangle\langle b_i(t_0)| \tag{D8}$$

has all the usual properties of evolution operators, including the group property for connected time intervals

$$U_b(t,t_0)=U_b(t,t_1)U_b(t_1,t_0) \tag{D9}$$

and the relations

$$\{U_b(t,t_0)\}^\dagger=\{U_b(t,t_0)\}^{-1}=U_b(t_0,t)\ . \tag{D10}$$

A trivial example of immobile kets as seen from basis b is provided by the basis kets $|b_i(t)\rangle$ themselves. Obviously, if one uses two bases which are mobile with respect to each other, a ket which is immobile as seen from one basis will, in general, be mobile as seen from the other basis. Similarly, a linear operator is called immobile or constant as seen from basis b if all its matrix elements in the basis are time independent, and examples of such operators are given by

$$A'(t)=U_b(t,t_0)A'(t_0)U_b(t_0,t)$$

and (D11)

$$K'(t,t')=U_b(t,t_0)K'(t_0,t_1)U_b(t_1,t')\ ,$$

where t and t' are variable times and t_0 and t_1 are fixed times. Again, the extension of this idea to Liouville operators, and further on, is straightforward. Note also that for scalar quantities (numbers) the notion of immobility is much simpler and does *not* depend upon the choice of basis.

3. Time derivatives as seen from a basis

In the perspective of the use of bases which are mobile with respect to each other, the naive definition of the time derivative of a ket as a limit of $(|\varphi(t+\Delta t)\rangle-|\varphi(t)\rangle)/\Delta t$ for $\Delta t\to 0$ has to be supplemented with a procedure for comparing (subtracting) kets at two *different* times. A natural way out of this problem is to subtract kets which are both defined at the same time $(t+\Delta t)$ and to interpret $|\varphi(t)\rangle$ in the above formula as the ket which would be obtained at time $(t+\Delta t)$ if the ket $|\varphi(t)\rangle$ remained immobile as seen from basis b during the time interval t to $(t+\Delta t)$. With this procedure the time derivative of a ket as seen from basis b is given by

$$\left[\frac{\partial}{\partial t}\right]_b|\varphi(t)\rangle=\lim_{\Delta t\to 0}(1/\Delta t)[\,|\varphi(t+\Delta t)\rangle - U_b(t+\Delta t,t)|\varphi(t)\rangle]$$
$$=\sum_i |b_i(t)\rangle\frac{\partial}{\partial t}\langle b_i(t)|\varphi(t)\rangle\ , \tag{D12}$$

where the time derivative of the ket has to be indexed by the relevant basis whereas the time derivatives of scalar quantities, such as scalar products or matrix elements, are not indexed because they do not depend upon the choice of basis.

The same problems arise with the definition of time derivatives of bras, operators, Liouville operators etc., and the procedure described above leads, for example, to the following definitions of time derivatives as seen from basis b:

$$\left[\frac{\partial}{\partial t}\right]_b\langle\varphi(t)|=\sum_i\langle b_i(t)|\frac{\partial}{\partial t}\langle\varphi(t)|b_i(t)\rangle\ ,$$
$$\left[\frac{\partial}{\partial t}\right]_b A(t)=\sum_{i,j}|b_i(t)\rangle\langle b_j(t)|\frac{\partial}{\partial t}\langle b_i(t)|A(t)|b_j(t)\rangle\ , \tag{D13}$$
$$\left[\frac{\partial}{\partial t}\right]_b K(t,t_0)=\sum_{i,j}|b_i(t)\rangle\langle b_j(t_0)| \times\frac{\partial}{\partial t}\langle b_i(t)|K(t,t_0)|b_j(t_0)\rangle\ ,$$

$$\left[\frac{\partial}{\partial t_0}\right]_b K(t,t_0)$$
$$=\sum_{i,j}|b_i(t)\rangle\langle b_j(t_0)|\frac{\partial}{\partial t_0}\langle b_i(t)|K(t,t_0)|b_j(t_0)\rangle\ .$$

It is worth noting that, if the time derivative of a scalar quantity is evaluated from the separate time derivatives of the components (bra, operators, ket) of the scalar quantity, care must be taken to evaluate all the time derivatives of nonscalar quantities as seen from the same basis. For instance,

$$\frac{\partial}{\partial t}\langle\varphi(t)|\psi(t)\rangle=\left\{\left[\frac{\partial}{\partial t}\right]_b\langle\varphi(t)|\right\}|\psi(t)\rangle + \langle\varphi(t)|\left\{\left[\frac{\partial}{\partial t}\right]_b|\psi(t)\rangle\right\}\ . \tag{D14}$$

4. Quantum dynamics as seen from different bases

We shall consider two bases. The first is basis b, with orthogonality and closure relations given by (D1) and (D2) and the characteristic evolution operator $U_b(t,t_0)$ given by (D8). When seen from basis b, the operator $U_b(t,t_0)$ is independent of time and behaves like an identity operator. However, when seen from any basis c which is not immobile with respect to basis b, the operator $U_b(t,t_0)$ is a time-dependent evolution operator. The second basis is denoted c, with orthogonality and closure relations analogous to those of basis b, and a characteristic evolution (or time displacement) operator

$$U_c(t,t_0)=\sum_i |c_i(t)\rangle\langle c_i(t_0)|\ . \tag{D15}$$

The relation between the two bases can be described completely by the single-time unitary operator

$$W_{bc}(t)=\sum_i |b_i(t)\rangle\langle c_i(t)| = U_b(t,t_0)W_{bc}(t_0)U_c(t_0,t) , \quad \text{(D16)}$$

such that, for any j, $|b_j(t)\rangle = W_{bc}(t)|c_j(t)\rangle$. If we interchange the roles of the two bases, we can define the unitary operator $W_{cb}(t)=\sum_i |c_i(t)\rangle\langle b_i(t)| = W_{bc}^{\dagger}(t)$. Another convenient tool for describing the relative motion of the two bases is provided by the operator $D_{bc}(t)$ defined by the set of differential equations

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c |b_j(t)\rangle = D_{bc}(t)|b_j(t)\rangle \quad \text{(D17)}$$

for all values of j. This operator has the dimension of energy and is Hermitian because the kets $|b_j(t)\rangle$ are normalized at all times. If we interchange the roles of the two bases, the corresponding operator $D_{cb}(t)$ can be evaluated by the following procedure: (i) write (D12) with any $|c_j(t)\rangle$ playing the role of $|\varphi(t)\rangle$, (ii) use (D14), with time derivatives as seen from basis c, to evaluate the time derivative of $\langle b_i(t)|c_j(t)\rangle$, (iii) note that $[\partial/\partial t]_c |c_j(t)\rangle = 0$ and use the adjoint of (D17). The result is

$$i\hbar\left[\frac{\partial}{\partial t}\right]_b |c_j(t)\rangle = -D_{bc}(t)|c_j(t)\rangle \quad \text{(D18)}$$

for all values of j, hence, $D_{cb}(t)=-D_{bc}(t)$. Using similar techniques one obtains the following differential relations:

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c U_b(t,t_0)=D_{bc}(t)U_b(t,t_0) , \quad \text{(D19)}$$

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c W_{bc}(t)=D_{bc}(t)W_{bc}(t) , \quad \text{(D20)}$$

which have to be supplemented with the initial conditions $U_b(t_0,t_0)=1$, and $W_{bc}(t_0)$ known at some fixed time t_0.

Finally, still using the same techniques, the following relations are easily obtained between time derivatives as seen from the two bases b and c:

$$\begin{aligned}
&i\hbar\left[\frac{\partial}{\partial t}\right]_b |\psi(t)\rangle = i\hbar\left[\frac{\partial}{\partial t}\right]_c |\psi(t)\rangle - D_{bc}(t)|\psi(t)\rangle ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_b \langle\psi(t)| = i\hbar\left[\frac{\partial}{\partial t}\right]_c \langle\psi(t)| + \langle\psi(t)|D_{bc}(t) ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_b A(t) = i\hbar\left[\frac{\partial}{\partial t}\right]_c A(t) - [D_{bc}(t),A(t)] , \quad \text{(D21)}\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_b K(t,t_0) = i\hbar\left[\frac{\partial}{\partial t}\right]_c K(t,t_0) - D_{bc}(t)K(t,t_0) ,\\
&i\hbar\left[\frac{\partial}{\partial t_0}\right]_b K(t,t_0) = i\hbar\left[\frac{\partial}{\partial t_0}\right]_c K(t,t_0) + K(t,t_0)D_{bc}(t_0) .
\end{aligned}$$

In order to display some simple and obvious uses of Eqs. (D21), we choose basis c as a conventional basis in which the state of a physical system, described by $|\varphi(t)\rangle$ or $\rho(t)$, evolves according to the usual Schrödinger or von Neumann equation of motion,

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c |\varphi(t)\rangle = H(t)|\varphi(t)\rangle$$

or (D22)

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c \rho(t)=[H(t),\rho(t)] ,$$

where the Hermitian operator $H(t)$ is the Hamiltonian of the system. The complete specification of a physical situation requires some additional information which may be supplied, for instance, by the state of the system, $|\varphi(t_0)\rangle$ or $\rho(t_0)$, at some fixed time t_0. With this initial condition a formal solution of (D22) can be written in the form

$$|\varphi(t)\rangle = U(t,t_0)|\varphi(t_0)\rangle$$

or (D23)

$$\rho(t)=U(t,t_0)\rho(t_0)U(t_0,t) ,$$

where the unitary evolution operator $U(t,t_0)$ describing the motion of the system is the solution of the differential equation

$$i\hbar\left[\frac{\partial}{\partial t}\right]_c U(t,t_0)=H(t)U(t,t_0) \quad \text{(D24)}$$

with the initial condition $U(t_0,t_0)=1$. Note that the operator $U(t,t_0)$ is a property of the physical system which is not related to any specific basis; it is only the *particular* form (D24) of the equation of motion of $U(t,t_0)$ which is expressed in a particular basis.

In order to prepare motivations for the coming discussion we shall decompose the Hamiltonian as

$$H(t)=H_0(t)+[H(t)-H_0(t)] . \quad \text{(D25)}$$

In a first type of problem, the interesting properties of the system *as seen from basis c* are obscured mainly by fast time dependences generated by a large and trivial part $H_0(t)$ of the Hamiltonian. By analogy with the situation in classical mechanics, one suspects that these fast time dependences will not be "visible" any more if the system is seen from a basis b which moves with respect to basis c according to the dynamics generated by $H_0(t)$. Such a basis b is easily constructed from basis c by choosing basis vectors $|b_i(t)\rangle$ which move with respect to basis c in the same way as the kets describing the state of a virtual physical system with Hamiltonian $H_0(t)$. Comparing (D17) and (D22) we see that such a basis b obeys (D17) with

$$D_{bc}(t)=H_0(t) . \quad \text{(D26)}$$

Hence the equations of motion (D22) and (D24) can be rewritten with time derivatives as seen from basis b [see (D21)] in the forms

$$
\begin{aligned}
&i\hbar\left[\frac{\partial}{\partial t}\right]_b |\psi(t)\rangle=\{H(t)-H_0(t)\}\,|\psi(t)\rangle\ ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_b \rho(t)=[H(t)-H_0(t),\rho(t)]\ ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_b U(t,t_0)=\{H(t)-H_0(t)\}U(t,t_0)\ ,
\end{aligned}
\tag{D27}
$$

which clearly show that the fast time dependences generated by the large term $H_0(t)$ are not visible any more when the system is seen from basis b. Furthermore, if the initial situation is known at time t_0, it will often be convenient that the two bases b and c coincide at time t_0, hence $W_{bc}(t)$ will be the solution of the differential equation

$$
i\hbar\left[\frac{\partial}{\partial t}\right]_c W_{bc}(t)=H_0(t)W_{bc}(t) \tag{D28}
$$

with the initial condition $W_{bc}(t_0)=1$.

In a second type of problem, the interesting properties of the system are also obscured by the complexity of the initial situation (at time t_0), *as seen from basis c.* This type of problem can be solved or mitigated by choosing a basis b in which the initial situation is simpler. If the initial situation is specified by $|\psi(t_0)\rangle$, a very suitable basis b would have $|\psi(t_0\rangle$ as one of its basis kets at time t_0. Of course, such a choice for basis b does, in general, prevent the unitary "basis changing" operator $W_{bc}(t)$ from being equal to 1 at any time, but this is a minor detail. After having solved the problems with the initial condition by a proper choice of $W_{bc}(t_0)$, one is still free to solve further problems with fast time dependences by a suitable choice of $D_{bc}(t)$ as discussed above.

5. Interaction pictures (or representations)

The appealing simplicity of the equations of motion (D27) and of the corresponding initial situation may be utterly deceptive because these describe the state of the system *as seen from basis b,* whereas almost everything else in the problem (initial conditions, Hamiltonian, observables, etc.) is usually known in its form *as seen from basis c.* Hence, actually working in basis b very often implies many changes of basis to express kets in terms of $\{|b_i(t)\rangle\}$, operators in terms of $\{|b_i(t)\rangle\langle b_j(t)|\}$ or $\{|b_i(t)\rangle\langle b_j(t_0)|\}$, and great care to keep track of the basis in which each time derivative, ket or operator is expressed.

These deviations from tradition, and inconveniences in notation, can be avoided, without losing the advantages provided by the use of a suitably chosen basis b, by the standard procedure called "going over to an interaction picture." This procedure can be visualized in the following geometrical way: at each time t, we lock all the relevant objects (kets, bras, operators, state of the system, etc.) to basis b, then we (instantaneously) move the basis and locked objects until the basis coincides with basis c (transformation $W_{bc}^{\dagger}$ does exactly this), and we call each moved object the interaction picture of its unmoved counterpart. More explicitly, if we denote interaction pictures by a tilde above the symbol, the correspondence is the following for kets, bras and operators:

$$
\begin{aligned}
&|\tilde{\varphi}(t)\rangle=W_{bc}^{\dagger}(t)\,|\varphi(t)\rangle\ ,\\
&\langle\tilde{\varphi}(t)|=\langle\varphi(t)|\,W_{bc}(t)\ ,\\
&\tilde{A}(t)=W_{bc}^{\dagger}(t)A(t)W_{bc}(t)\ ,\\
&\tilde{K}(t,t_0)=W_{bc}^{\dagger}(t)K(t,t_0)W_{bc}(t_0)\ .
\end{aligned}
\tag{D29}
$$

By this procedure, the interaction picture objects as seen from basis c have the same appealing simple behavior as the original objects seen from basis b,

$$
\begin{aligned}
&i\hbar\left[\frac{\partial}{\partial t}\right]_c |\tilde{\psi}(t)\rangle=\{\tilde{H}(t)-\tilde{H}_0(t)\}\,|\tilde{\psi}(t)\rangle\ ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_c \tilde{\rho}(t)=[\tilde{H}(t)-\tilde{H}_0(t),\tilde{\rho}(t)]\ ,\\
&i\hbar\left[\frac{\partial}{\partial t}\right]_c \tilde{U}(t,t_0)=\{\tilde{H}(t)-\tilde{H}_0(t)\}\tilde{U}(t,t_0)\ .
\end{aligned}
\tag{D30}
$$

The unitary transformation $W_{bc}(t)$ used in (D29) satisfies (D28) with an initial condition $W_{bc}(t_0)$ chosen to suit the initial condition of the physical system. In this way, a single basis (c) is used and there is no need to index time derivatives according to the relevant basis. This is the procedure used in this paper (except in this appendix, of course).

Summing up, the use of moving bases and that of interaction pictures appear as two very closely related ways for disentangling the discussion of problems in quantum dynamics in a succession of steps. The interaction-picture technique avoids the multiplicity of bases and enables one to perform the quantum calculations in the conventional way, at the price of introducing transformed versions of the state of the system and of all dynamical operators. Conversely, using bases moving with respect to each other avoids the multiplicity of transformed versions of each ket, operator, etc.

6. Further remarks about situations involving more than a single time

In the perspective of using a multiplicity of bases moving with respect to each other, one must be extremely careful in dealing with situations involving more than a single time. As an illustration of this idea, we shall now come back to the two related objects of this type explicitly discussed so far: time displacement operators and time derivatives as seen from a basis.

Time displacement operators $K(t_1,t_0)$ were defined by the linear relation (D5), namely, $|\varphi(t_1\rangle=K(t_1,t_0)\,|\psi(t_0)\rangle$, between any $|\psi(t_0)\rangle$ and the corresponding $|\varphi(t_1)\rangle$. In this linear relation, the two times t_0 and t_1 do *not* play equivalent roles: $K(t_1,t_0)$ is defined *only* in the perspective where the object on its right is at time t_0 and the object on its left is at time t_1. We have systematically reflected this dissymmetry in the typographical order of the time arguments of the operators.

A minor source of ambiguity arises with the inverse or the adjoint of a time displacement operator. The adjoint is defined by taking the adjoint of both sides of (D5), namely, $\langle\varphi(t_1)| = \langle\psi(t_0)|\{K(t_1,t_0)\}^\dagger$. Clearly, the adjoint of $K(t_1,t_0)$ is defined with t_1 on the right and t_0 on the left so that, if we give this adjoint the name K' and use the standard typography for the arguments of K', we shall write $\{K(t_1,t_0)\}^\dagger = K'(t_0,t_1)$. Of course, it is tempting to replace the prime by a dagger in this last formula and to define the new notation $K^\dagger(t_0,t_1) = \{K(t_1,t_0)\}^\dagger$. However, we felt that this natural notation would conflict too much with traditions, hence we have not used it in this paper. In practice, we avoided ambiguities by almost never using adjoints of such operators.

An equivalent situation arises with the inverse of $K(t_1,t_0)$, defined by the relation $1 = \{K(t_1,t_0)\}^{-1} K(t_1,t_0)$, in which the identity operator behaves as an operator involving the single time t_0.

Let us now examine, as an example, the differential equation

$$i\hbar\left[\frac{\partial}{\partial t}\right]_b |\xi(t)\rangle = G_b(t)|\xi(t)\rangle \ , \tag{D31}$$

where no particular assumption is made about $G_b(t)$, so that $|\xi(t)\rangle$ does not, in general, describe the state of any physical system. $G_b(t)$ is given an index b to recall its particular role in basis b. As a preparation for solving this equation by iterative techniques, we shall first rewrite (D31) and its initial condition $|\xi(t_0)\rangle$ as an integral equation, using the definition of the time derivative given by (D12). The result is

$$|\xi(t)\rangle = U_b(t,t_0)|\xi(t_0)\rangle + (1/i\hbar)\int_{t_0}^{t} dt' U_b(t,t') G_b(t')|\xi(t')\rangle \ . \tag{D32}$$

We note that the left-hand side (lhs) and each term in the sum (and integral) in the rhs of (D32) are kets which are all defined at the same time t, with the required changes in time provided by the characteristic evolution operator U_b for basis b. The presence of the operator U_b in (D32) may seem superfluous in the perspective of a calculation performed in basis b. However, (D32) as it stands is a general, base-independent relation in which U_b is the remnant of the fact that $G_b(t)$ is the generator of the motion of $|\xi(t)\rangle$ *as seen from basis b.*

Pursuing the calculation in the standard way, we can express the evolution operator $K(t,t_0)$ for $|\xi(t)\rangle$, defined by the requirement that $|\xi(t)\rangle = K(t,t_0)|\xi(t_0)\rangle$ for any $|\xi(t_0)\rangle$, in the form

$$K(t,t_0) = 1 + \sum_{n=1}^{\infty} (1/i\hbar)^n \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-1}} dt_n U_b(t,t_1) G_b(t_1) U_b(t_1,t_2) G_b(t_2) \cdots U_b(t_{n-1},t_n) G_b(t_n) U_b(t_n,t_0) \ . \tag{D33}$$

Of course, if $G_b(t)$ is not Hermitian, then, in general, $K(t,t_0)$ is not unitary in the sense that its adjoint and its inverse are not equal.

If the operator $G_b(t)$ satisfies the commutation relation

$$[U_b(t_2,t_1)G_b(t_1)U_b(t_1,t_2), G_b(t_2)] = 0 \tag{D34}$$

for all values of t_1 and t_2 in the range between t_0 and t, then (D33) is "easily" evaluated as

$$K(t,t_0) = U_b(t,t_0)\exp\left\{(1/i\hbar)\int_{t_0}^{t} dt_1 U_b(t_0,t_1) G_b(t_1) \times U_b(t_1,t_0)\right\} , \tag{D35}$$

where the quadrature is to be evaluated in a first step, and the exponential of the result taken in a subsequent independent step. If condition (D34) is not satisfied, the usual problems of time ordering arise. The lhs of (D34) is a satisfactory expression, in the viewpoint of this appendix, of the usual idea of the commutator of a time-dependent operator with itself taken at two different times; we note that it is an operator acting at a *single time* (t_2 in this particular case), and that it depends upon the basis used to relate the situations at the two different times. Of course, the same situation exists for the commutator of two different operators taken at two different times.

In the main part of this paper, care has been taken to systematically denote single-time operators with a single time argument and time-displacement operators with two time arguments, but the precautions shown in (D32)—(D35) have not been taken, so that all relations involving more than a single time (or time derivatives) are valid only as seen from the standard basis in which the Hamiltonian is the operator of the motion.

[1]V. Weisskopf and E. Wigner, Z. Phys. 63, 54 (1930).
[2]C. George, F. Mayné, and I. Prigogine, Adv. Chem. Phys. 61, 223 (1985), and references therein.
[3]B. R. Mollow, Phys. Rev. A 12, 1919 (1975).
[4]The dummy initial times denoted t_0 and defined by Eqs. (A1), (2.25), (2.8), (2.26), (2.47), (3.4), and (3.7) are not required to be all the same. However, for simplicity, we have given them a common notation implying a common value. It is convenient to choose t_0 in the initial idle period of the model [see (3.10)].
[5]R. J. Glauber, Phys. Rev. 131, 2766 (1963).
[6]In this paper, a clear typographical distinction is introduced between single-time operators $A(t)$ which, acting on a ket defined at some time t, generate a ket defined at the same time t (these operators are written with a single time variable or no

time variable, even if they involve various times parametrically), $A(t)|\psi(t)\rangle = |\varphi(t)\rangle$, and time-displacement operators $K(t_2,t_1)$ which, acting on a ket defined at some time t_1, generate a ket defined at some (usually different) time t_2 (these operators are written with two time variables in the order specified here), $K(t_2,t_1)|\psi(t_1)\rangle = |\varphi(t_2)\rangle$. Motivations for such a distinction can be found, e.g., in (2.33) and in Appendix D (see particularly Sec. D 6). Also, the notations t, t_0, $t_1, \ldots$ always denote unique instants ("date and time of the day"), whereas durations are denoted $(t-t_0)$, Δt, or dt.

[7]Using the ideas discussed in Appendix D the relations between the single-time and time-displacement versions of the unitary operators Q_F and B_A can be expressed as $Q_F(t,t_0)=Q_F(t)U_{cF}(t,t_0)$ and $B_A(t,t_0)=B_A(t)U_{cA}(t,t_0)$ where $U_{cF}(t,t_0)$ and $U_{cA}(t,t_0)$ are the characteristic evolution operators of the standard bases used here for the free-field and bare-atom state spaces.

[8]See, for instance, P. Pechukas and F. C. Light. J. Chem. Phys. **44**, 2897 (1966); U. Haeberlen, Adv. Magn. Reson. Suppl. **1**, (1976), Appendix B; M. Mehring, *High Resolution NMR Spectroscopy in Solids* (Springer, Berlin, 1976).

[9]See, for instance, R. S. Bondurant, Phys. Rev. A **32**, 2797 (1985); H. J. Kimble and L. Mandel, Phys. Rev. A **30**, 844 (1984), and references therein.

[10]M. Kline and I. W. Kay, *Electromagnetic Theory and Geometrical Optics* (Wiley, New York, 1965).

[11]R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. **27**, 631 (1971); R. G. Brewer and A. Z. Genack, Phys. Rev. Lett. **36**, 959 (1976).

[12]See, for instance, Louisell, Ref. 14 (p. 254) and J. M. Jauch and F. Rohrlich, *The Theory of Photons and Electrons* (Addison-Wesley, Reading, Mass., 1955), pp. 7 and 22.

[13]M. de Haan, Physica **132A**, 375 (1985).

[14]W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).

[15]Louisell, Ref. 14, p. 151.

[16]A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1962), p. 510.

[17]See, for instance, A. Bambini and M. Lindberg, Phys. Rev. A **30**, 794 (1984), and references therein.