Rabi oscillations in the scattering-state representation

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This paper presents an analysis of the Rabi oscillations of two-level atoms in the scattering-state representation. A collision complex obtained from a pair of identical two-level atoms is coupled to a field mode and it is found that there can be three distinct Rabi frequencies. These reduce to a single Rabi frequency for a photon distribution that is sharply peaked around an average number $\bar{n} \gg 1$. One then finds that, in addition to the three frequency components in the off-diagonal element of the atomic density matrix, there are two components that are symmetrically displaced around the mode frequency at intervals of twice the Rabi frequency. They represent the collisionally induced cooperative motion of the atoms. When various observable quantities are averaged over the orientation of the complex, one obtains results that apply to many-atom systems in the binary collision approximation. Collisional effects do not enter the above results as lifetimes due to the fact that scattering states have precisely defined energies and correspond to entire particle histories. The collision potential opens up new radiative coupling channels, which collectively represent spectral line broadening. The collision potential and the radiative coupling combine to produce static and quasistatic dipole moments for the complex. At the threshold for atomic population inversion, the density matrix for the complex, as well as for an atom, becomes a constant multiple of the identity matrix, demonstrating that the threshold of stimulated emission is also a critical point for collisional effects, within the approximations of this paper.

I. INTRODUCTION

Popular theories of light propagation in many-body systems may be roughly divided into two groups: semiclassical theories and projection algebra of density-matrix operators. In both types of approaches, one usually starts a theoretical analysis with severely restrictive assumptions about relative strengths of radiative and nonradiative couplings. The widely used semiclassical theory of light propagation[1,2] is valid, provided that there are no significant nonradiative interactions among atoms, or in the opposite limit, if nonradiative interactions, such as collisions, are intense and rapid, as well as localized, so that the incident field has no effect on individual events. In this latter limit, collisions are taken into account by means of finite linewidths. The most precise approach to radiation fields plus an N-body system is to treat the entire collection as one quantum system, with one wave function for each distinct many-body state. The quantum properties of its components, such as those of atoms and molecules, or of radiation fields, may then be deduced by means of appropriate projection operators. Various density-matrix operator methods are based on this systematic reduction technique.[2,3] These methods become quite complex as soon as one tries to impose some sort of self-consistency,[4] or to treat mutual influences between the material system and the radiation field on equal terms. Usually the physics becomes completely obscured behind a complicated formalism.

With strongly interacting atoms and molecules which spontaneously organize themselves into a crystalline order, there is no alternative to some form of projection algebra, either on density-matrix operators or directly on wave functions. However, when such spontaneous organized behavior does not take place, for example, in gases, it is possible to develop an intermediate approach in which one identifies sufficiently small units which may be considered as independent in the absence of radiation fields. An independent unit may be a pair of colliding atoms ("binary-collision approximation"), a triplet of mutually scattering atoms, etc. These units may be described by scattering states and referred to as collision complexes. Collision complexes may then be directly coupled to radiation fields in a dressed atom formalism. Once the density matrix of a complex is determined as a function of time, it can be partially traced over to yield the density matrix for individual atoms. Thus, the proposed method permits one to take into account significant correlations arising from collisions while treating the coupling between the radiation field and the material system rigorously. Earlier we applied the method to a collisionally triggered coherence phenomenon in atomic vapors[5] and to an alternative description of the collisionally opened radiative channels.[6] In the present paper, we apply the method to the problem of Rabi oscillations in a simple model system.

One uses a dressed atom representation[7] in the description of the interaction of a nearly resonant intense field with an atom (well-known examples are the resonance fluorescence and Rabi oscillations[8] of atoms). The num-

ber of atomic states involved in radiative transitions induced by an incident optical field is usually small. When one associates these states with the number states of the incident field quanta, the Hamiltonian of the system of one atom plus many quanta becomes truncated in the product Hilbert space, at least in some form of the rotating-wave approximation. The determination of the eigenvalues and eigenstates of the Hamiltonian is then reduced to the problem of the removal of degeneracies. This extremely simple and direct method has provided an efficient tool with which quantum properties of the system of one atom plus many quanta have been investigated rigorously.

Scattering states are well known from the early work on the quantum theory of scattering.[9] Let us consider a pair of colliding atoms. One can solve the Schrödinger equation for the collision process by describing the quantum state of the pair by the scattering state corresponding to a specified asymptotic state of the pair in the limit of vanishing collision potential. If V is the localized collision potential and H_A represents the asymptotic Hamiltonian of the two atoms, then the scattering state corresponding to an asymptotic state $|\psi\rangle_A$ is given by

$$|\psi\rangle=\Omega|\psi\rangle_A\ , \tag{1.1a}$$

$$\Omega=1+(E-H_A-V+i\varepsilon)^{-1}V\ , \tag{1.1b}$$

where E is the energy of the asymptotic state $|\psi\rangle_A$ and is treated as a c-number parameter. The transformation (1.1a) can be viewed as a canonical transformation when there are no bound states of the two atoms induced by V, or even if there are such bound states for an attractive V, if they can be neglected with little effect on the dynamics of the pair. This follows from a fundamental theorem[10] which states that, to every solution ψ_t of the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi_t=(H_A+V)\psi_t \tag{1.2a}$$

which is orthogonal to all bound states, there corresponds a unique solution of the particle Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\phi_t=H_A\phi_t\ , \tag{1.2b}$$

such that the norm obeys

$$\lim_{t\to\infty}\|\psi_t-\phi_t\|=0\ . \tag{1.2c}$$

Conversely, to every solution of (1.2b), there corresponds exactly one solution of (1.2a) such that (1.2c) holds. The asymptotic pair states can therefore be put into one-to-one correspondence with the scattering states in the absence of bound states. In fact, the operator Ω satisfies the relations[11]

$$\Omega^\dagger\Omega=1\ , \tag{1.3a}$$

$$\Omega\Omega^\dagger=1-\sum_\beta|\psi_\beta\rangle\langle\psi_\beta|\ , \tag{1.3b}$$

where $|\psi_\beta\rangle$'s are the bound states of H_A+V. Thus, Ω is actually a unitary operator and (1.1a) is a unitary transformation if there are no bound states. Once a collision potential is specified, (1.1a) gives the state of the pair parametrized by its energy.

The dressed atom formalism can be directly applied to the scattering states generated by the transformation (1.1a). This has several advantages. First, by coupling the scattering states directly with the appropriate number states of the radiation field and solving for the dressed states of the complex, one takes into account the effect of the field on the collision complex, hence, on the collisional process, in a direct and physically transparent way. A collision event is no longer an irreducible event as in the semiclassical theories. The new formalism reflects the fact that a collision event is influenced by the radiation field. Second, the effect of the collisional process on the radiative coupling is also taken into account in a direct way. Specifically, collisionally triggered cooperative effects, as well as collisional broadening, are exhibited in a unified formalism. Of course, the degree of accuracy to which the mutual influences between the collisional and radiative processes are taken into account in a many-body system depends on the number of atoms in the complex and on the approximations made in order to solve the effective Hamiltonian when particular photon-number states are associated with the atomic states. If the collision complex has two atoms, that is, if $2N$ atoms of a gas are paired into N collision complexes, the results are accurate in the binary-collision approximation. One can systematically increase the number of atoms in the complex. For example, a two-atom complex may be paired with another atom to obtain a three-atom complex. In the second pairing, the scattering states of the two-atom complex are treated as if they were the asymptotic states with respect to the scattering states of the three-atom complex. Calculations with three-atom complexes take into account three-body correlations as well as two-body correlations. This procedure may be repeated to obtain larger complexes for more accuracy in calculations of many-body effects. It is seen that our method of approach to radiative interactions in many-body systems is inductive, in contrast to projection algebra methods, which are didactic.

In this paper our main purpose is to illustrate the new method. Consistent with this purpose, we chose the well-known Rabi oscillations of two-level atoms for analysis. However, we also obtain new results. In summary these are the following: (a) For the complex of a pair of two-level atoms, there is more than one Rabi frequency. These frequencies are determined by a cubic equation. (b) At high field intensities, one recovers a single Rabi frequency, but there are still collisionally induced cooperative effects. For example, the off-diagonal element of the atomic density matrix has four frequency satellites which are symmetrically displaced from the mode frequency, two by an amount equal to the Rabi frequency and two by twice the Rabi frequency. The latter are induced by the collisional process. (c) Under the influence of the fundamental mode, static or quasistatic dipole moments of the collision complex can be produced. These moments are associated with degenerate states of the complex. (d) Within the approximations of

Sec. III, the threshold for atomic population inversion is a critical point not only for radiative transitions, but also for collisional effects. At this threshold, the collisional effects vanish and the density matrix for the complex, as well as the reduced density matrix for the atoms, becomes a multiple of the identity matrix, independent of time.

In Sec. II, we demonstrate the formation of a collision complex from a pair of indistinguishable two-level atoms. We use the straightline trajectory approximation. The dipole moment operator of the complex in the scattering-state representation exhibits new radiative coupling channels which are induced by V. The coupling coefficients between the complex and the radiation field become dependent on energy as well as on V. We couple the complex to a single field mode (occasionally referred to as the fundamental mode in this paper), and assume that the mode frequency is sufficiently close to the atomic resonance frequency so that the rotating-wave approximation can be made. The diagonalization of this simple Hamiltonian with the aid of dressed states yields a biquadratic equation for the energy eigenvalues. One solution corresponds to an unshifted energy level of the complex. The remaining cubic equation yields three distinct Rabi frequencies. These solutions are quite sensitive to the detuning between the mode frequency and the atomic resonance frequency and to the number of the quanta in the mode. The exact solutions of the eigenstates and eigenenergies permit the determination of the time-dependent density matrix for the complex plus the field once the initial conditions are specified. We discuss these initial conditions, assuming that they are specified in the asymptotic limit.

In Sec. III, we consider the case where the number of the field quanta is sharply peaked around an average number which is much larger than one. The collisionally split Rabi frequencies merge together and yield one Rabi frequency, which is still modified by V through the coupling coefficient. Some collisionally induced cooperative effects appear at twice this Rabi frequency. With a further simplification of the initial conditions in the scattering-state representation, the elements of the time-dependent density matrix for the complex are explicitly displayed. In the following, we refer to the frequency components of the diagonal elements of density matrices as the Rabi spectrum, to those of the off-diagonal elements as the Mollow spectrum. The Rabi spectrum of the complex for a sharply peaked distribution of photon numbers has five components at 0, $\pm\xi'$, and $\pm 2\xi'$, where ξ' is the Rabi frequency. The Mollow spectrum of the complex has frequency components at 0, $\pm\xi'$, $\pm 2\xi'$, ω, $\omega\pm\xi'$, $\omega\pm 2\xi'$, 2ω, $2\omega\pm\xi'$, and $2\omega\pm 2\xi'$. The same spectra are found for the atomic density matrix, which is obtained by partially tracing over the density matrix of the complex. The vanishing of the detuning from the atomic resonance and/or the limit of infinite average number of quanta in the mode define the strong-coupling limit. In this limit, the amplitudes of many of the above frequency components vanish. For example, the Mollow spectrum of an atom reduces to just five components at ω, $\omega\pm\xi'$, and $\omega\pm 2\xi'$. In comparison, the standard near-resonant Mollow spectrum has just the components at ω and $\omega\pm\xi'$. Thus, the components at $\omega\pm 2\xi'$ represent the collisionally induced cooperation between the two atoms in the complex. They vanish when $V=0$. The components of the Mollow spectrum of the complex at 0, $\pm\xi'$, and $\pm 2\xi'$ represent the static and quasistatic polarizations of the complex. Under the combined action of the field and the collision potential, a complex acts as if it were an independent permanent dipole. As discussed further in Sec. V, small static electric fields can align such dipoles in large domains. The density matrix of the complex, as well as the atomic density matrix, becomes a constant multiple of the identity matrix at the threshold for the inversion of the atomic population. Collisional effects vanish. This indicates that the threshold for atomic population inversion is a critical point for both radiative and collisional processes.

It should be emphasized that the collisional effects do not appear as energy uncertainties or lifetimes in the above results. This is due to a different perspective of collisions in the scattering-state representation, as discussed in Sec. IV. Normally, collisions are viewed in the time domain, where an atom rushes from one collision event to another, which leads to uncertainties in atomic energy levels and hence to spectral line broadening. By contrast, scattering states have precisely defined energies and therefore each scattering state defines an entire particle history. In this picture, radiative transitions correspond to hopping from one particular history to another. The collision potential opens up many new radiative coupling channels. These collectively describe spectral line broadening. The collision potential V also modifies the magnitude of the Rabi frequency (or frequencies). In Sec. IV we also discuss the binary-collision approximation. When the results of Sec. III are properly averaged over the orientation of the complex, one obtains expressions which are valid for many-atom systems in the binary-collision approximation. Section V gives a few concluding remarks.

A final remark of this introduction concerns the neglect of the bound states that permits the treatment of the transformations induced by Ω as unitary transformations. This means that our method can be applied to gases of atoms (or molecules) if there are no chemical phase transitions. Temperatures, pressures, as well as the type of atoms, must be such that no chemical transformations must take place, and that chemical species must preserve their identity.

II. A SIMPLIFIED HAMILTONIAN AND ITS SOLUTION

In this section we consider a simple collision complex consisting of a pair of identical two-level atoms in a single-mode radiation field and obtain the density matrices for the pair and the individual atoms. We further simplify the general Hamiltonian in the scattering-state representation by assuming that momenta exchanged during collisions, as well as recoil momenta associated with radiative transitions, are negligible. Thus, the atoms of the complex preserve their center-of-mass (c.m.) momenta throughout various transitions. This is equivalent

to the straightline trajectory approximation used in many semiclassical theories. It restricts the validity of the results to long-range collisions.

Since the atoms of the complex are identical, the asymptotic states are either symmetric or antisymmetric with respect to the interchange of the atoms. Symmetric operators like the collision potential and the dipole operator of the complex do not couple symmetric states to antisymmetric ones. All possible states of the complex are therefore either symmetric or antisymmetric, with no mixing between the states of different parity. In the following we assume that the states of the complex are symmetric.

For two identical two-level atoms, there are four asymptotic states. They can be written as

$$|0\rangle_A=\frac{1}{\sqrt{2}}(|0;\mathbf{p}_1\rangle_a\otimes|0;\mathbf{p}_2\rangle_b+|0;\mathbf{p}_1\rangle_b\otimes|0;\mathbf{p}_2\rangle_a)\ , \tag{2.1a}$$

$$|1\rangle_A=\frac{1}{\sqrt{2}}(|0;\mathbf{p}_1\rangle_a\otimes|1;\mathbf{p}_2\rangle_b+|0;\mathbf{p}_1\rangle_b\otimes|1;\mathbf{p}_2\rangle_a)\ , \tag{2.1b}$$

$$|2\rangle_A=\frac{1}{\sqrt{2}}(|1;\mathbf{p}_1\rangle_a\otimes|0;\mathbf{p}_2\rangle_b+|1;\mathbf{p}_1\rangle_b\otimes|0;\mathbf{p}_2\rangle_a)\ , \tag{2.1c}$$

$$|3\rangle_A=\frac{1}{\sqrt{2}}(|1;\mathbf{p}_1\rangle_a\otimes|1;\mathbf{p}_2\rangle_b+|1;\mathbf{p}_1\rangle_b\otimes|1;\mathbf{p}_2\rangle_a)\ . \tag{2.1d}$$

Here, the state vectors $|\alpha;\mathbf{p}_i\rangle_{a,b}$ ($\alpha=0,1;i=1,2$) refer to the individual atomic states. α designates the internal atomic state, $\mathbf{p}_i$ the c.m. momentum. a,b label the atoms. $\alpha=0$ is the ground state, $\alpha=1$ is the excited state. One should interpret the meaning of, for example, $|0;\mathbf{p}_1\rangle_a$ as

$$|0,\mathbf{p}_1\rangle_a\rightarrow\langle A_a;\mathbf{X}_a|0;\mathbf{p}_1\rangle=\frac{1}{\sqrt{V_{\rm OL}}}e^{i\mathbf{p}_i\cdot\mathbf{X}_a}\psi_0(A_\alpha)\ , \tag{2.2}$$

where $A_{a,b}$ refers to the sets of the internal variables of the atoms a and b, and $\mathbf{X}_{a,b}$ to the c.m. coordinates. ψ_0 is the wave function for the internal ground state. Under the straightline trajectory approximation, $\mathbf{p}_1$ and $\mathbf{p}_2$ are constant parameters without dynamical significance. It is therefore entirely sufficient to label these asymptotic states by $|\mu\rangle_A$ ($\mu=0,1,2,3$). They are the eigenstates of the asymptotic Hamiltonian H_A and are orthonormal:

$$H_A|\mu\rangle_A=E_\mu^c|\mu\rangle_A\ , \tag{2.3a}$$

$${}_A\langle\mu|\mu'\rangle_A=\delta_{\mu\mu'}\ , \tag{2.3b}$$

where

$$E_0^c=2E_0+\frac{\hbar^2\mathbf{p}_1^2}{2m}+\frac{\hbar^2\mathbf{p}_2^2}{2m}\ , \tag{2.3c}$$

$$E_1^c=E_2^c=E_0+E_1+\frac{\hbar^2\mathbf{p}_1^2}{2m}+\frac{\hbar^2\mathbf{p}_2^2}{2m}\ , \tag{2.3d}$$

$$E_3^c=2E_1+\frac{\hbar^2\mathbf{p}_1^2}{2m}+\frac{\hbar^2\mathbf{p}_2^2}{2m}\ . \tag{2.3e}$$

Here E_0 and E_1 are the energies and the atomic ground and excited states, respectively. The scattering states corresponding to (2.1) are given by

$$|\mu\rangle=[1+(E_\mu^c-H_A-V+i\varepsilon)^{-1}V]|\mu\rangle_A\ , \tag{2.4a}$$

which are the eigenstates of H_A+V with the same eigenenergies as in (2.3c)–(2.3e):

$$(H_A+V)|\mu\rangle=E_\mu^c|\mu\rangle\ . \tag{2.4b}$$

Next, let us consider the dipole moment operator for the complex and the coupling coefficients to the field. For any operator $\mathcal{O}$ which is specified in the space of the asymptotic states, the matrix elements in the scattering-state representation are obtained from the transformation induced by Ω:

$$\langle\mu|\mathcal{O}|\mu'\rangle=\sum_{\mu_1\mu_2}\Omega^\dagger_{\mu\mu_1}(E_\mu)\,{}_A\langle\mu_1|\mathcal{O}|\mu_2\rangle_A\,\Omega_{\mu_2\mu'}(E_{\mu'})\ , \tag{2.5a}$$

where

$${}_A\langle\mu'|\mu\rangle={}_A\langle\mu'|\Omega(E_\mu)|\mu\rangle_A=\Omega_{\mu'\mu}(E_\mu)\ . \tag{2.5b}$$

Some mathematical care needs to be exercised in the evaluation of the diagonal matrix elements and the matrix elements between degenerate states. Gell-Mann and Goldberger[11] have shown that in the limit $\varepsilon\rightarrow0$,

$$\lim_{\varepsilon\rightarrow0}{}_A\langle\mu|\mu\rangle=\lim_{\varepsilon\rightarrow0}\Omega_{\mu\mu}(E_\mu)=1\ . \tag{2.6}$$

Furthermore, the limit $\varepsilon\rightarrow0$ must be accompanied by the limit $L\rightarrow\infty$, where $V_{\rm OL}=L^3$=the quantization volume, for consistent physical interpretation. If two distinct states $|\mu\rangle$ and $|\mu'\rangle$ have the same energy $E_\mu=E_{\mu'}$, then the Gell-Mann-Goldberger limiting procedure yields

$$\lim_{\varepsilon\rightarrow0}\Omega_{\mu'\mu}(E_\mu)=0\ . \tag{2.7}$$

Thus, for the asymptotic states in (2.1), the diagonal matrix elements of the operator Ω are 1, and some of its other matrix elements are given by

$$\Omega_{12}(E_2^c)=\Omega_{21}(E_1^c)=0\ , \tag{2.8a}$$

$$\Omega_{01}(E_1^c)=\Omega_{02}(E_2^c)=\tfrac{1}{2}[({}_a\langle0|\otimes{}_b\langle0|)\Omega(E_1+E_0)(|0\rangle_a\otimes|1\rangle_b)+({}_a\langle0|\otimes{}_b\langle0|)\Omega(E_1+E_0)(|1\rangle_a\otimes|0\rangle_b)]\ , \tag{2.8b}$$

$$\Omega_{31}(E_1^c)=\Omega_{32}(E_2^c)=\tfrac{1}{2}[({}_a\langle1|\otimes{}_b\langle1|)\Omega(E_1+E_0)(|1\rangle_a\otimes|0\rangle_b)+({}_a\langle1|\otimes{}_b\langle1|)\Omega(E_1+E_0)(|0\rangle_a\otimes|1\rangle_b)]\ , \tag{2.8c}$$

$$\Omega_{30}(E_0^c)=\tfrac{1}{2}{}_a\langle1|\otimes{}_b\langle1|\Omega(2E_0)|0\rangle_a\otimes|0\rangle_b\ , \tag{2.8d}$$

where product state vectors at the right-hand side refer only to the internal states of the atoms. Other matrix elements such as $\Omega_{10}(E_0^c)$, $\Omega_{13}(E_3^0)$, etc., may be obtained from (2.8b)–(2.8d) by appropriate interchanges of the ket and bra vectors and by inserting the appropriate energy arguments for the operator Ω. Note that the reason for the appearance of just the internal states and internal energies on the right-hand side of (2.8b)–(2.8d) is the straightline trajectory approximation. In this approximation, V has no effect on the c.m. states. When one factorizes the atomic internal states and the c.m. states, e.g.,

$$|\alpha;\mathbf{p}_i\rangle_a=|\alpha\rangle_a\otimes|\mathbf{p}_i\rangle_a ,$$

and uses $\langle\mathbf{p}_i|\mathbf{p}_j\rangle=\delta_{ij}$, the translational energy parts are canceled in the energy arguments of Ω and one is left with only the internal states and internal energies.

For a pair of atoms, the dipole moment density and its Fourier transform are given by

$$\mathbf{D}(\mathbf{r})=\sum_{j=a,b}\delta(\mathbf{r}-\mathbf{X}_j)\mathbf{d}_j , \tag{2.9a}$$

$$\int d\mathbf{r}e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{D}(\mathbf{r})=\sum_{j=a,b}e^{i\mathbf{k}\cdot\mathbf{X}_j}\mathbf{d}_j=\mathcal{D}(\mathbf{k}) , \tag{2.9b}$$

where $\mathbf{d}_j$ is the electric dipole moment operator of the jth atom. The indistinguishability of the atoms and the neglect of radiative recoil permit one to simplify (2.9b). Let $\mathbf{x}=(\mathbf{X}_a+\mathbf{X}_b)/2$ and $\mathbf{x}'=\mathbf{X}_a-\mathbf{X}_b$. Then

$$\mathcal{D}(\mathbf{k})=2\mathbf{d}e^{i\mathbf{k}\cdot\mathbf{x}}\cos(\mathbf{k}\cdot\mathbf{x}'/2)\simeq 2\mathbf{d} , \tag{2.10}$$

where, for pair separations which are much less than the wavelength of the mode, we set $\cos(\mathbf{k}\cdot\mathbf{x}'/2)$ equal to 1. Furthermore, in the calculations the exponential factor $\exp(i\mathbf{k}\cdot\mathbf{x})$ always appears with the exponential factors $\exp(i\mathbf{p}_i\cdot\mathbf{x})$. As long as the atomic c.m. momenta are much larger than the photon momentum, one can replace $\exp(i\mathbf{k}\cdot\mathbf{x})$ by 1. With these approximations, $\mathcal{D}$ is reduced to $(2\mathbf{d})$. In the space of the asymptotic states, the matrix elements of $\mathcal{D}$ are given by

$${}_A\langle\mu|\mathbf{D}|\mu'\rangle_A=\tfrac{1}{2}{}_A\langle\alpha_1\alpha_2|\mathcal{D}|\alpha_1'\alpha_2'\rangle_A , \tag{2.11}$$

where $(\alpha_1\alpha_2)$ and $(\alpha_2'\alpha_2')$ are the internal states corresponding to μ and μ', taken in the same order as in the first terms of (2.1a)–(2.1d). Using (2.10), the nonvanishing matrix elements are

$$\begin{aligned}{}_A\langle 0|\mathbf{D}|1\rangle_A={}_A\langle 0|\mathbf{D}|2\rangle_A={}_A\langle 1|\mathbf{D}|3\rangle_A\\ ={}_A\langle 2|\mathbf{D}|3\rangle_A=\mathbf{d}_{01}\end{aligned} \tag{2.12}$$

and their Hermitian conjugates. The subscripts on $\mathbf{d}$ refer to the atomic internal states. For a two-level atom, only $\mathbf{d}_{01}$ and $\mathbf{d}_{10}=\mathbf{d}_{01}^*$ are nonzero. The matrix elements of $\mathbf{D}$ in the scattering-state representation are obtained from (2.5a) and (2.12):

$$\begin{aligned}\langle\mu|\mathbf{D}|\mu'\rangle=\mathbf{d}_{01}[\Omega_{0\mu}^*(E_\mu^c)\Omega_{1\mu'}(E_{\mu'}^c)+\Omega_{0\mu}^*(E_\mu^c)\Omega_{2\mu'}(E_{\mu'}^c)+\Omega_{1\mu}^*(E_\mu^c)\Omega_{3\mu'}(E_{\mu'}^c)+\Omega_{2\mu}^*(E_\mu^c)\Omega_{3\mu'}(E_{\mu'}^c)]\\ +\mathbf{d}_{01}^*[\Omega_{1\mu}^*(E_\mu^c)\Omega_{0\mu'}(E_{\mu'}^c)+\Omega_{2\mu}^*(E_\mu^c)\Omega_{0\mu'}(E_{\mu'}^c)+\Omega_{3\mu}^*(E_\mu^c)\Omega_{1\mu'}(E_{\mu'}^c)+\Omega_{3\mu}^*(E_\mu^c)\Omega_{2\mu'}(E_{\mu'}^c)] .\end{aligned} \tag{2.13}$$

Finally, the coupling coefficients to a radiation mode can be written as

$$g_{\mu\mu'}=\left[\frac{2\pi\omega}{\hbar V_{\mathrm{OL}}}\right]^{1/2}\langle\mu|\hat{\epsilon}\cdot\mathbf{D}|\mu'\rangle , \tag{2.14a}$$

where ω is the frequency of the mode, $\hat{\epsilon}$ is its polarization vector. One can show from (2.8) and (2.13) that $g_{\mu\mu'}$ obeys the symmetry relations

$$g_{01}=g_{02},\quad g_{13}=g_{23} . \tag{2.14b}$$

The fact that the collisional process opens up new radiative coupling channels can be seen from (2.13). For example, the dipole matrix element $\langle 0|\mathbf{D}|3\rangle$ is nonzero and arises entirely from the collision potential. It couples the complex to radiation modes which are at twice the atomic transition frequency. This coupling describes the cooperation of the atoms through the collisional process in that individual photons absorbed or emitted in this channel are shared by the two atoms. Such cooperative effects can cause second harmonic generation in atomic vapors.[5]

In the scattering-state representation the general form of the Hamiltonian is[6]

$$\begin{aligned}H=&\sum_{\alpha_1,\mathbf{p}_1,\alpha_2,\mathbf{p}_2,\sigma}E_{\alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2}|\alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2\sigma\rangle\langle\alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2\sigma|-\sum_{\lambda,\mathbf{k}}\hbar\omega_{\lambda\mathbf{k}}a_{\lambda\mathbf{k}}^\dagger a_{\lambda\mathbf{k}}\\ &+\left[i\sum_{\sigma;\alpha_1,\mathbf{p}_1,\alpha_2,\mathbf{p}_2;\alpha_1',\mathbf{p}_1',\alpha_2',\mathbf{p}_2';\lambda,\mathbf{k}}g^{\sigma\lambda}_{\alpha_1\alpha_2;\alpha_1'\alpha_2'}(\mathbf{p}_1\mathbf{p}_2;\mathbf{p}_1'\mathbf{p}_2';\mathbf{k})|\alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2\sigma\rangle\langle\alpha_1'\mathbf{p}_1'\alpha_2'\mathbf{p}_2'\alpha|a_{\lambda\mathbf{k}}+\mathrm{H.c.}\right] .\end{aligned} \tag{2.15a}$$

Here $E_{\alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2}$ is the energy of the complex. σ designates the symmetry ($\sigma=+1$) or the antisymmetry ($\sigma=-1$) of the pair of the atoms with respect to the interchange of the atoms. $a_{\lambda\mathbf{k}}$ and $a^{\dagger}_{\lambda\mathbf{k}}$ are the field operators for the mode $(\lambda\mathbf{k})$. $g^{\sigma\lambda}$ is the coupling coefficient in the scattering-state representation given by

$$g^{\sigma\lambda}_{\alpha_1\alpha_2;\alpha_2'\alpha_2'}(\mathbf{p}_1\mathbf{p}_2;\mathbf{p}_1'\mathbf{p}_2') = \left[\frac{2\pi\hbar\omega_{\lambda\mathbf{k}}}{V_{\mathrm{OL}}}\right]^{1/2} \langle \alpha_1\mathbf{p}_1\alpha_2\mathbf{p}_2\sigma|\hat{\epsilon}_{\lambda\mathbf{k}}\cdot\mathcal{D}(\mathbf{k})|\alpha_1'\mathbf{p}_1'\alpha_2'\mathbf{p}_2'\sigma\rangle \ . \tag{2.15b}$$

In the present paper we will work with a simplified version of this Hamiltonian by omitting all field modes except one, with frequency ω, which we will refer to as the fundamental mode, and by making the rotating-wave approximation with respect to this fundamental mode. Under the assumption that momenta exchanged in collisions, as well as radiative recoil momenta, are negligible, we may use the compact notation of (2.1)–(2.4) and our working Hamiltonian becomes

$$\begin{aligned} H=&\sum_{\mu} E^c_{\mu}|\mu\rangle\langle\mu|+\hbar\omega a^{\dagger}a \\ &+[i\hbar(g_{10}|1\rangle\langle 0|+g_{10}|2\rangle\langle 0|+g_{31}|3\rangle\langle 1| \\ &\quad +g_{31}|3\rangle\langle 2|)a+\mathrm{H.c.}] \ , \end{aligned} \tag{2.16}$$

where a and $a^{\dagger}$ are destruction and creation operators for the fundamental mode. Even though (2.16) omits the cooperative effects associated with second harmonic generation, the density matrix of the complex may have frequency components which oscillate at 2ω due to two-photon transitions.

The eigenvalues and eigenstates of the Hamiltonian in (2.16) can be obtained by writing an arbitrary state $|\psi\rangle$ as a linear superposition of the states of the complex and the radiation field in the form

$$\begin{aligned} |\psi_n\rangle =& l_0|0\rangle\otimes|n+2\rangle+l_1|1\rangle\otimes|n+1\rangle \\ &+l_2|2\rangle\otimes|n+1\rangle+l_3|3\rangle\otimes|n\rangle \\ \equiv& \sum_{\mu} l_{\mu}|\mu\rangle\otimes|n+m_{\mu}\rangle \ , \end{aligned} \tag{2.17a}$$

where $|n\rangle$ designates the radiation state with n quanta ($n\geq 0$) and

$$m_{\mu}=\begin{cases} 2 & \text{for } \mu=0 \ , \\ 1 & \text{for } \mu=1,2 \ , \\ 0 & \text{for } \mu=3 \ . \end{cases} \tag{2.17b}$$

When H operates on $|\psi_n\rangle$, the resulting state vector remains within the subspace defined by the basis vectors in (2.17a). From

$$H|\psi_n\rangle=E|\psi_n\rangle \tag{2.18a}$$

one finds that

$$\begin{bmatrix} E^c_0+(n+2)\hbar\omega & -i\hbar g^*_{10}\sqrt{n+2} & -i\hbar g^*_{10}\sqrt{n+2} & 0 \\ i\hbar g_{10}\sqrt{n+2} & E^c_1+(n+1)\hbar\omega & 0 & -i\hbar g^*_{31}\sqrt{n+1} \\ i\hbar g_{10}\sqrt{n+2} & 0 & E^c_2+(n+1)\hbar\omega & -i\hbar g^*_{31}\sqrt{n+1} \\ 0 & i\hbar g_{31}\sqrt{n+1} & i\hbar g_{31}\sqrt{n+1} & E^c_3+n\hbar\omega \end{bmatrix} \begin{bmatrix} l_0 \\ l_1 \\ l_2 \\ l_3 \end{bmatrix} = E \begin{bmatrix} l_0 \\ l_1 \\ l_2 \\ l_3 \end{bmatrix} \ . \tag{2.18b}$$

The eigenvalue problem for H is thus reduced to the diagonalization of a 4×4 Hermitian matrix. With the definitions

$$E=\hbar\xi+E^c_1+(n+1)\hbar\omega \ , \tag{2.19a}$$

$$\hbar\Delta=E^c_1-E^c_0-\hbar\omega \ , \tag{2.19b}$$

one finds the eigenvalue equation

$$\xi^4-[2(n+2)]|g_{10}|^2+2(n+1)|g_{31}|^2+\Delta^2]\xi^2+2\Delta[(n+2)|g_{10}|^2-(n+1)|g_{31}|^2]\xi=0 \ . \tag{2.20}$$

Let us designate the roots of (2.20) by $\xi_{n\lambda}$, where $\lambda=0,1,2,3$. The overall ξ factor gives a root that is zero. Let this root be ξ_{n3}. The remaining three roots are the solutions of a cubic equation. Define the quantity

$$w_n=u_n^3-v_n^2 \ , \tag{2.21a}$$

where

$$u_n=\tfrac{1}{3}[\Delta^2+2(n+2)|g_{10}|^2+2(n+1)|g_{31}|^2] \ , \tag{2.21b}$$

$$v_n=\Delta[(n+2)|g_{10}|^2-(n+1)|g_{31}|^2] \ . \tag{2.21c}$$

Since the matrix in (2.18b) is Hermitian, $w_n>0$, the cubic equation is irreducible and there are three real roots:[12]

$$\xi_{n0}=2\sqrt{u_n}\cos\left[\frac{\phi_n}{3}\right] \ , \tag{2.22a}$$

$$\xi_{n1}=-2\sqrt{u_n}\cos\left[\frac{\pi-\phi_n}{3}\right] \ , \tag{2.22b}$$

$$\xi_{n2}=-2\sqrt{u_n}\cos\left[\frac{\pi+\phi_n}{3}\right] \ , \tag{2.22c}$$

where

$$\phi_n=\tan^{-1}\left[\frac{-\sqrt{w_n}}{v_n}\right] . \tag{2.22d}$$

Let $l_{n\lambda\mu}$ represent the components of the eigenvector corresponding to $\xi_{n\lambda}$. Define

$$\begin{aligned}N_{n\lambda}=\{&[\xi_{n\lambda}(\xi_{n\lambda}-\Delta)-2(n+1)|g_{31}|^2]^2\\&+2(n+2)(\xi_{n\lambda}-\Delta)^2|g_{10}|^2\\&+4(n+1)(n+2)|g_{10}|^2|g_{31}|^2\}^{1/2} .\end{aligned} \tag{2.23a}$$

Then

$$l_{n\lambda 0}=\frac{1}{N_{n\lambda}}[\xi_{n\lambda}(\xi_{n\lambda}-\Delta)-2(n+1)|g_{31}|^2] , \tag{2.23b}$$

$$l_{n\lambda 1}=l_{n\lambda 2}=\frac{i}{N_{n\lambda}}\sqrt{n+2}(\xi_{n\lambda}-\Delta)g_{10} , \tag{2.23c}$$

$$l_{n\lambda 3}=-\frac{2}{N_{n\lambda}}\sqrt{(n+1)(n+2)}g_{10}g_{31} . \tag{2.23d}$$

These eigenvectors are complete as well as orthonormal:

$$\sum_{\mu} l^*_{n\lambda\mu}l_{n\lambda'\mu}=\delta_{\lambda\lambda'} , \tag{2.24a}$$

$$\sum_{\lambda} l^*_{n\lambda\mu}l_{n\lambda\mu'}=\delta_{\mu\mu'} . \tag{2.24b}$$

In summary, the eigenstates of H are given by

$$|\psi_{n\lambda}\rangle=\sum_{\mu} l_{n\lambda\mu}|\mu\rangle\otimes|n+m_\mu\rangle , \tag{2.25a}$$

$$\begin{aligned}H|\psi_{n\lambda}\rangle&=E_{n\lambda}|\psi_{n\lambda}\rangle\\&=[\hbar\xi_{n\lambda}+E_1^c+(n+1)\hbar\omega]|\psi_{n\lambda}\rangle .\end{aligned} \tag{2.25b}$$

An arbitrary state of the collision complex plus the radiation field can be written as

$$|\psi\rangle=\sum_{n=0}^{\infty}\sum_{\lambda=0}^{3} c_{n\lambda}|\psi_{n\lambda}\rangle . \tag{2.26a}$$

If the state of the total system at time $t=0$ is given by a state like (2.26a), then the state at time t is given by

$$|\psi(t)\rangle=e^{-iHt/\hbar}|\psi\rangle=\sum_{n,\lambda} c_{n\lambda}e^{-iE_{n\lambda}t/\hbar}|\psi_{n\lambda}\rangle . \tag{2.26b}$$

The corresponding density-matrix operator is given by

$$\begin{aligned}\rho(t)&=|\psi(t)\rangle\langle\psi(t)|\\&=\sum_{n,\lambda,n',\lambda'} c_{n\lambda}c^*_{n'\lambda'}e^{-i(E_{n\lambda}-E_{n'\lambda'})t/\hbar}|\psi_{n\lambda}\rangle\langle\psi_{n'\lambda'}| .\end{aligned} \tag{2.26c}$$

If at time $t=0$ the system can be described only by a mixture of states, for example, by the density-matrix operator $\rho(0)$,

$$\rho(0)=\sum_{n,\lambda,n',\lambda'}|\psi_{n\lambda}\rangle\rho_{n\lambda;n'\lambda'}(0)\langle\psi_{n'\lambda'}| , \tag{2.27a}$$

then

$$\begin{aligned}\rho(t)&=\sum_{n,\lambda,n',\lambda'}|\psi_{n\lambda}\rangle[e^{-i(E_{n\lambda}-E_{n'\lambda'})t/\hbar}\rho_{n\lambda;n'\lambda'}(0)]\langle\psi_{n'\lambda'}|\\&=\sum_{n,\lambda,n',\lambda'}|\psi_{n\lambda}\rangle\rho_{n\lambda;n'\lambda'}(t)\langle\psi_{n'\lambda'}| .\end{aligned} \tag{2.27b}$$

Clearly, the specification of either the initial-state amplitudes $c_{n\lambda}$ in (2.26a) or the initial density-matrix elements $\rho_{n\lambda;n'\lambda'}(0)$ in (2.27a) is equivalent to a complete solution for the dynamics of the system. Since (2.27) includes (2.26) as a special case, let us concentrate on the density matrices.

We postulate that the initial (boundary) conditions are specified in the asymptotic limit and for individual atoms. We also assume that, at $t=0$, the density matrix of the total system can be factored into an outer product of the density matrix for the complex and the density matrix for the field. These assumptions correspond to a physical picture in which the radiation field and the collision potential are turned on simultaneously on $t=0$. We therefore need to relate $\rho_{n\lambda;n'\lambda'}(0)$ to the asymptotic states of the complex at $t=0$ and determine the general connection formulas between the density matrix of the complex and the density matrix of the individual atoms. From (2.25a),

$$\rho_{n\lambda;n'\lambda'}(0)=\langle\psi_{n\lambda}|\rho(0)|\psi_{n'\lambda'}\rangle=\sum_{\mu\mu'} l^*_{n\lambda\mu}l_{n'\lambda'\mu'}\langle n+m_\mu|\otimes\langle\mu|\rho(0)|\mu'\rangle\otimes|n'+m_{\mu'}\rangle . \tag{2.28}$$

Factoring the density matrix as

$$\rho(0)=\rho^c(0)\otimes\rho^\gamma(0) , \tag{2.29a}$$

where $\rho^c(0)$ is the density matrix operator for the complex and $\rho^\gamma(0)$ is for the field, one finds

$$\begin{aligned}&\langle n+m_\mu|\otimes\langle\mu|\rho(0)|\mu'\rangle\otimes|n'+m_{\mu'}\rangle\\&\quad=\langle\mu|\rho^c(0)|\mu'\rangle\rho^\gamma_{n+m_\mu,n'+m_{\mu'}}(0) ,\end{aligned} \tag{2.29b}$$

$$\begin{aligned}\rho_{n\lambda;n'\lambda'}(0)=\sum_{\mu,\mu',\mu'',\mu'''} &l^*_{n\lambda\mu}l_{n'\lambda'\mu'}\rho^\gamma_{n+m_\mu,n'+m_{\mu'}}(0)\\&\times\Omega^\dagger_{\mu\mu''}(E^c_\mu)\rho^{Ac}_{\mu''\mu'''}(0)\Omega_{\mu'''\mu'}(E^c_{\mu'}) ,\end{aligned} \tag{2.29c}$$

where $\rho^{Ac}_{\mu\mu'}(0)$ denotes ${}_A\langle\mu|\rho^c(0)|\mu'\rangle_A$. To compute $\rho^{Ac}_{\mu\mu'}(0)$, we note that Eqs. (2.1) can be summarized as

$$|\mu\rangle_A=\sum_{\alpha_1,i_1,\alpha_2,i_2} C^{\mu}_{\alpha_1 i_1;\alpha_2 i_2}|\alpha_1;\mathbf{p}_{i_1}\rangle_a\otimes|\alpha_2;\mathbf{p}_{i_2}\rangle_b . \tag{2.30}$$

Since both the collision potential and the radiation field are turned on at $t=0$, it is reasonable to assume that $\rho^{Ac}(0)$ may be further factored into the atomic density matrices for the two atoms of the complex:

$$\rho^{Ac}_{\mu\mu'}(0)={}_A\langle\mu|\rho^{\rm at}_a(0)\otimes\rho^{\rm at}_b(0)|\mu'\rangle_A \ . \tag{2.31}$$

Substituting (2.30) into (2.31), we obtain $\rho^{Ac}_{\mu\mu'}(0)$ in terms of the elements of the atomic density matrix $\rho^{\rm at}(0)$:

$$\rho^{Ac}_{\mu\mu'}(0)=\sum_{\substack{\alpha_1,i_1,\alpha_2,i_2\\ \alpha_1',i_1',\alpha_2',i_2'}}(C^{\mu}_{\alpha_1 i_1;\alpha_2 i_2})^*\rho^{\rm at}_{\alpha_1 i_1;\alpha_1' i_1'}(0)\times\rho^{\rm at}_{\alpha_2 i_2;\alpha_2' i_2'}(0)C^{\mu'}_{\alpha_1' i_1';\alpha_2' i_2'} \ . \tag{2.32}$$

Note that all of the nonvanishing $C^{\mu}_{\alpha i;\alpha' i'}$'s are equal to $1/\sqrt{2}$:

$$C^0_{01;02}=C^0_{02;01}=C^1_{01;12}=C^1_{12;01}=C^2_{11;02}=C^2_{02;11}=C^3_{11;12}=C^3_{12;11}=\frac{1}{\sqrt{2}} \ . \tag{2.33}$$

Suppose that $\rho^{\rm at}(0)$ is diagonal and that the occupation probabilities of the atomic states are given by

$$\rho^{\rm at}_{\alpha i;\alpha' i'}(0)=\delta_{\alpha\alpha'}\delta_{ii'}f_\alpha(\mathbf{p}_i) \ , \tag{2.34}$$

where $f_\alpha(\mathbf{p}_i)$ is the conditional probability that the internal state of the atom is α if the atom is in the c.m. momentum state $\mathbf{p}_i$. Because each atom is either in the ground state or the excited state,

$$f_0(\mathbf{p}_i)+f_1(\mathbf{p}_i)=1 \ . \tag{2.35}$$

Substituting (2.34) into (2.32), one finds that the off-diagonal elements of ρ^{Ac} vanish and the diagonal elements are given by

$$\rho^{Ac}_{00}(0)=f_0(\mathbf{p}_1)f_0(\mathbf{p}_2) \ , \tag{2.36a}$$

$$\rho^{Ac}_{11}(0)=f_0(\mathbf{p}_1)f_1(\mathbf{p}_2) \ , \tag{2.36b}$$

$$\rho^{Ac}_{22}(0)=f_0(\mathbf{p}_2)f_1(\mathbf{p}_1) \ , \tag{2.36c}$$

$$\rho^{Ac}_{33}(0)=f_1(\mathbf{p}_1)f_1(\mathbf{p}_2) \ . \tag{2.36d}$$

When these initial conditions are transformed into the scattering-state representation, one obtains the initial density matrix in the scattering-state representation, which we designate by F:

$$\begin{aligned}F_{\mu\mu'}&\equiv\sum_{\mu'',\mu'''}\Omega^{\dagger}_{\mu\mu''}(E^c_\mu)\rho^{Ac}_{\mu''\mu'''}(0)\Omega_{\mu'''\mu'}(E^c_{\mu'})\\&=f_0(\mathbf{p}_1)f_0(\mathbf{p}_2)\Omega^{\dagger}_{\mu 0}(E^c_\mu)\Omega_{0\mu'}(E^c_{\mu'})+f_0(\mathbf{p}_1)f_1(\mathbf{p}_2)\Omega^{\dagger}_{\mu 1}(E^c_\mu)\Omega_{1\mu'}(E^c_{\mu'})\\&\quad+f_0(\mathbf{p}_2)f_1(\mathbf{p}_1)\Omega^{\dagger}_{\mu 2}(E^c_\mu)\Omega_{2\mu'}(E^c_{\mu'})+f_1(\mathbf{p}_1)f_1(\mathbf{p}_2)\Omega^{\dagger}_{\mu 3}(E^c_\mu)\Omega_{3\mu'}(E^c_{\mu'}) \ .\end{aligned} \tag{2.37}$$

The time-dependent density matrices for the complex and the individual atoms are obtained by tracing over the states of the field mode. From (2.27b) and (2.37), one finds for the density matrix of the complex in the scattering-state representation

$$\rho^c_{\mu\mu'}(t)=\sum_{n,n',\lambda,\lambda',\mu'',\mu'''}\delta(n+m_\mu,n'+m_{\mu'})e^{-it[\xi_{n\lambda}-\xi_{n'\lambda'}-(m_\mu-m_{\mu'})\omega]}\rho^{\gamma}_{n+m_{\mu''},n'+m_{\mu'''}}(0)l_{n\lambda\mu}l^*_{n\lambda\mu''}F_{\mu''\mu'''}l_{n'\lambda'\mu'''}l^*_{n'\lambda'\mu'} \ . \tag{2.38}$$

Tracing over the states of one of the atoms in (2.38) and transforming back to the space of asymptotic states, one finds the time-dependent atomic density matrix:

$$\rho^{\rm at}_{\alpha i;\alpha' i'}(t)=\sum_{\mu,\mu',\alpha'',i''}C^{\mu}_{\alpha i;\alpha'' i''}\rho^{Ac}_{\mu\mu'}(t)(C^{\mu'}_{\alpha' i';\alpha'' i''})^* \ , \tag{2.39a}$$

where

$$\rho^{Ac}_{\mu\mu'}(t)=\sum_{\mu'',\mu'''}\Omega_{\mu\mu''}(E^c_{\mu''})\rho^c_{\mu''\mu'''}(t)\Omega^*_{\mu'\mu'''}(E^c_{\mu'''}) \ . \tag{2.39b}$$

It is seen from (2.38), (2.25b), and (2.22) that the original energy levels are split into four components. One component is unshifted and corresponds to $\xi_{n3}=0$. The other three components have distinct shifts in the most general case, represented by ξ_{n0}, ξ_{n1}, and ξ_{n2}. This means that, in principle, there are three distinct Rabi frequencies. This is surprising in view of the conventional three-peak Mollow spectrum,[8,13] where each of the energy levels is split into two components and the shifts which are determined by a single Rabi frequency are symmetric with respect to the original level. Clearly, the additional levels and the corresponding Rabi frequencies in the present analysis arise from the collision-induced cooperation between the two atoms. The signs and magnitudes of ξ_{n0}, ξ_{n1}, and ξ_{n2} are quite sensitive to the detuning from the atomic resonance and to the intensity of the field. The phase angle ϕ_n in (2.22d) approaches $-\pi/2$ for $\Delta\to 0$ and/or $n\to\infty$. This causes $\xi_{n1}\to\xi_{n3}=0$ and $\xi_{n2}\to-\xi_{n0}$, producing two symmetric levels around the

unshifted level, and recovering something similar to the standard Mollow spectrum. The intense field case will be discussed in more detail in the next section. Both v_n and u_n are sensitive to Δ. However, v_n should be relatively insensitive to the field intensity since, on the basis of symmetry, one would expect g_{10} and g_{31} either to have the same magnitudes or to differ very little. If we let

$$d_{10}=d_{10}^{*}, \quad \mathcal{V}_{11;00}(0)=-\mathcal{V}_{11;00}^{*}(0) , \tag{2.40}$$

then to first order in V and in the limit $\epsilon\rightarrow 0$:

$$g_{10}=\left[\frac{2\pi\omega}{\hbar V_{\mathrm{OL}}}\right]^{1/2}\hat{\epsilon}\cdot\mathbf{d}_{10}\left[1-\frac{\mathcal{V}_{11;00}(0)}{2E_1-2E_0}\right]+O(V^2) , \tag{2.41a}$$

$$g_{31}=\left[\frac{2\pi\omega}{\hbar V_{\mathrm{OL}}}\right]^{1/2}\hat{\epsilon}\cdot\mathbf{d}_{10}\left[1+\frac{\mathcal{V}_{11;00}(0)}{2E_1-2E_0}\right]+O(V^2) , \tag{2.41b}$$

$$g_{31}^{*}\simeq g_{10}\equiv g . \tag{2.41c}$$

Here $\mathcal{V}$ is the Fourier transform of V with respect to atomic separations[6]

$$\mathcal{V}_{\alpha_1\alpha_2;\alpha_1'\alpha_2'}(\mathbf{k})=\sum_{A_a A_b}\frac{1}{V_{\mathrm{OL}}}\int d\mathbf{R}e^{i\mathbf{k}\cdot\mathbf{R}}\psi_{\alpha_1}^{*}(A_a)\psi_{\alpha_2}^{*}(A_b)\times V(A_a A_b;\mathbf{R})\times\psi_{\alpha_1'}(A_a)\psi_{\alpha_2'}(A_b) . \tag{2.42}$$

In (2.42), $A_{a,b}$ refers to the internal variables of the atoms, $\mathbf{R}$ is the atomic separation $\mathbf{X}_a-\mathbf{X}_b$, ψ_α''s are the asymptotic atomic states. When (2.41c) is used in (2.21b) and (2.21c), v_n and u_n become

$$u_n=v=\Delta|g|^2 , \tag{2.43a}$$

$$u_n=\tfrac{1}{3}[\Delta^2+(4n+6)|g|^2] . \tag{2.43b}$$

v is completely independent of the intensity. Whatever the intensity is, ϕ_n approaches $-\pi/2$ as one approaches the atomic resonance, and one obtains a single Rabi frequency. Clearly, any possibility for the observation of the multiple Rabi frequencies lies with large detunings from the atomic frequency and with extremely low intensities. These requirements can make experimental observation extremely difficult. Nevertheless, it may be possible to inject a few Rydberg atoms into high-Q cavities[14] or into optical traps[15] and to look for the collisionally split Rabi frequencies, for example, in the free-induction decays of atoms or in the spectrum of scattered light.

The appearance of just one angle variable in (2.41a) and (2.41b) may be surprising at first glance, since there are two atoms in the complex. However, we assumed that the two atoms of the complex are distinguishable. If the internal $\mathbf{d}_{10}$ were to point in different directions for each atom, then the atoms would be distinguishable. Of course, one can still use the symmetric and antisymmetric combinations of asymptotic atomic states for distinguishable atoms. For the symmetric combinations, one then has the replacement

$$\hat{\epsilon}\cdot\mathbf{d}_{10}\rightarrow\tfrac{1}{2}\hat{\epsilon}\cdot(\mathbf{d}_{10}^{(a)}+\mathbf{d}_{10}^{(b)})$$

in (2.41a) and (2.41b) when the atoms a and b are distinguishable. Formulas for distinguishable atoms are more cumbersome than those for indistinguishable atoms and will not be pursued here.

III. INTENSE FIELD

In this section we discuss the case in which the average number of quanta in the mode $\bar{n}\gg 1$ and the distribution of n's is sharply peaked in the vicinity of $\bar{n}$ (for example, when the mode is in a coherent state). By the word "intense" in the heading of this section, we simply mean that $\bar{n}\gg 1$, even though this may, in practical terms, mean a relatively weak optical field. When the field is intense in this sense, one recovers the familiar features of the Rabi oscillations in the two-level atom model. The collisionally split distinct Rabi frequencies discussed in Sec. II collapse into a single Rabi frequency, which has the familiar form. Nevertheless, there are still collisionally induced cooperative effects which appear as oscillations at the second harmonic of the Rabi frequency. The simplifications induced by the intense field assumption, as well as some further reasonable approximations, permit one to display explicit expressions for the density matrices. These expressions show an interesting effect arising from a particular set of initial conditions. If the initial conditions correspond to the threshold for the inversion of the atomic population, then the collisional effects vanish and the density matrices remain constant and proportional to the unit matrix. It follows that the threshold for stimulated emission is a critical point for collisional, as well as for radiative, cooperative effects.

Using (2.41c), replacing n's by $\bar{n}$ and ignoring 1's and 2's in (2.20), one obtains the simplified eigenvalue equation

$$\xi^4-(\Delta^2+4\bar{n}|g|^2)\xi^2=0 . \tag{3.1a}$$

The eigenvalues are

$$\xi_{0,1}=\pm(\Delta^2+4\bar{n}|g|^2)^{1/2}\equiv\pm\xi' , \tag{3.1b}$$

$$\xi_{2,3}=0 . \tag{3.1c}$$

It is clear from (2.41) and (3.1b) that the quantity ξ' is the generalized Rabi frequency for an atom in a coherent field mode.[8] If θ is the angle between the polarization vector and the bare atomic dipole moment

$$\hat{\epsilon}\cdot\mathbf{d}_{10}=|\mathbf{d}_{10}|\cos\theta , \tag{3.2a}$$

then ξ' can be written as

$$\xi'=|\Delta|(1+\zeta^2\cos^2\theta)^{1/2} , \tag{3.2b}$$

where

$$\zeta^2=\frac{4\bar{n}}{\Delta^2}|g(\theta=0)|^2\equiv\frac{\Omega_R^2}{\Delta^2} \tag{3.2c}$$

is the square of the ratio of the standard Rabi frequency to detuning. It is also seen from (2.41) that the collision

potential V modifies the magnitude and the phase of the atomic dipole moment.

The eigenvectors corresponding to $\pm\xi'$ are

$$l_0=\begin{bmatrix} \frac{\xi'-\Delta}{2\xi'} \\ \frac{i}{\xi'}g\sqrt{2\bar{n}} \\ \frac{i}{\xi'}g\sqrt{2\bar{n}} \\ -\frac{\xi'+\Delta}{2\xi'} \end{bmatrix}, \quad l_1=\begin{bmatrix} -\frac{\xi'-\Delta}{2\xi'} \\ \frac{i}{\xi'}g\sqrt{2\bar{n}} \\ \frac{i}{\xi'}g\sqrt{2\bar{n}} \\ \frac{\xi'+\Delta}{2\xi'} \end{bmatrix} . \tag{3.3a}$$

The other eigenvectors corresponding to degenerate $\xi_{2,3}=0$ may be chosen to be orthogonal to the vectors in (3.3a) as well as to each other:

$$l_2=\begin{bmatrix} -\frac{i}{\xi'}g^*\sqrt{2\bar{n}} \\ \frac{\Delta}{\xi'\sqrt{2}} \\ \frac{\Delta}{\xi'\sqrt{2}} \\ -\frac{i}{\xi'}g^*\sqrt{2\bar{n}} \end{bmatrix}, \quad l_3=\begin{bmatrix} 0 \\ \frac{1}{\sqrt{2}} \\ -\frac{1}{\sqrt{2}} \\ 0 \end{bmatrix} . \tag{3.3b}$$

These vectors form an orthonormal set.

Before we use the preceding eigenvectors in (2.38) to obtain the time-dependent density matrix of the complex, we simplify (2.38) by taking only the diagonal elements of the matrix F of the initial conditions. To order V^2, $F_{\mu\mu}$ is given by

$$F_{\mu\mu}=\delta_{\mu 0}f_0(\mathbf{p}_1)f_0(\mathbf{p}_2)+\delta_{\mu 1}f_0(\mathbf{p}_1)f_1(\mathbf{p}_2) +\delta_{\mu 2}f_0(\mathbf{p}_2)f_1(\mathbf{p}_1)+\delta_{\mu 3}f_1(\mathbf{p}_1)f_1(\mathbf{p}_2)+O(V^2) . \tag{3.4}$$

If the conditional probabilities are independent of $\mathbf{p}_i$, then

$$F_{00}=f_0^2, \quad F_{11}=F_{22}=f_0f_1, \quad F_{33}=f_1^2 . \tag{3.5}$$

The neglected off-diagonal elements of F are of the order of V; they play the primary role in the collisionally triggered second-harmonic generation.[5] However, in the present paper our focus is on Rabi oscillations, and this approximation considerably simplifies the final results. With respect to a single complex, it preserves the essential features of the cooperative effects in that the density-matrix elements have the same number of frequency components as when the off-diagonal matrix elements of F are not omitted.

From (2.38), (3.3), and (3.4), one finds that the elements of $\rho^c(t)$ in the intense field approximation are given by

$$\rho_{00}^c(t)\simeq\frac{1}{8\xi'^4}[(3\xi'^4+2\xi'^2\Delta^2+3\Delta^4)F_{00}+(\xi'^2-\Delta^2)(\xi'^2+3\Delta^2)(F_{11}+F_{22})+3(\xi'^2-\Delta^2)^2F_{33}] +\frac{(\xi'^2-\Delta^2)}{2\xi'^4}[(\xi'^2+\Delta^2)F_{00}-\Delta^2(F_{11}+F_{22})-(\xi'^2-\Delta^2)F_{33}]\cos\xi' t+\frac{(\xi'^2-\Delta^2)^2}{8\xi'^4}(F_{00}-F_{11}-F_{22}+F_{33})\cos 2\xi' t , \tag{3.6a}$$

$$\rho_{11}^c(t)\simeq\frac{1}{8\xi'^4}\{(\xi'^2-\Delta^2)(\xi'^2+3\Delta^2)F_{00}+[(\xi'^2-\Delta^2)^2+2\xi'^4+2\Delta^4](F_{11}+F_{22})+(\xi'^2-\Delta^2)(\xi'^2+3\Delta^2)F_{33}\} +\frac{\Delta^2}{2\xi'^2}(F_{11}-F_{22})+\left[\frac{\Delta^2(\xi'^2-\Delta^2)}{2\xi'^4}(-F_{00}+F_{11}+F_{22}-F_{33})+\frac{(\xi'^2-\Delta^2)}{2\xi'^2}(F_{11}-F_{22})\right]\cos\xi' t +\frac{(\xi'^2-\Delta^2)^2}{8\xi'^4}(-F_{00}+F_{11}+F_{22}-F_{33})\cos 2\xi' t , \tag{3.6b}$$

$$\rho_{22}^c(t)=\rho_{11}^c(t;F_{11}\leftrightarrow F_{22}) , \tag{3.6c}$$

$$\rho_{33}^c(t)\simeq\frac{1}{8\xi'^4}[3(\xi'^2-\Delta^2)^2F_{00}+(\xi'^2-\Delta^2)(\xi'^2+3\Delta^2)(F_{11}+F_{22})+(3\xi'^4+2\xi'^2\Delta^2+3\Delta^4)F_{33}] +\frac{(\xi'^2-\Delta^2)}{2\xi'^4}[-(\xi'^2-\Delta^2)F_{00}-\Delta^2(F_{11}+F_{22})+(\xi'^2+\Delta^2)F_{33}]\cos\xi' t +\frac{(\xi'^2-\Delta^2)^2}{8\xi'^4}(F_{00}-F_{11}-F_{22}+F_{33})\cos 2\xi' t , \tag{3.6d}$$

$$\rho_{01}^{c}(t)\simeq\left[\frac{-ig^{*}\sqrt{\bar{n}}\,e^{i\omega t}}{4\xi'^{4}}\right]\{-\Delta(\xi'^{2}+3\Delta^{2})F_{00}-\Delta(\xi'^{2}-3\Delta^{2})(F_{11}+F_{22})+3\Delta(\xi'^{2}-\Delta^{2})F_{33}$$
$$+e^{-i\xi' t}[-(\xi'-\Delta)(\xi'^{2}+\xi'^{2}\Delta+2\Delta^{2})F_{00}$$
$$+\Delta(\xi'-\Delta)(\xi'+2\Delta)(F_{11}+F_{22})+(\xi'^{2}-\Delta^{2})(\xi'-2\Delta)F_{33}]$$
$$+e^{i\xi' t}[(\xi'+\Delta)(\xi'^{2}-\xi'\Delta+2\Delta^{2})F_{00}+\Delta(\xi'-2\Delta)(\xi'+\Delta)(F_{11}+F_{22})-(\xi'^{2}-\Delta^{2})(\xi'+2\Delta)F_{33}]$$
$$+\tfrac{1}{2}(\xi'^{2}-\Delta^{2})[(\xi'+\Delta)e^{2i\xi' t}-(\xi'-\Delta)e^{-2i\xi' t}](F_{00}-F_{11}-F_{22}+F_{33})$$
$$+\xi'^{2}[2\Delta+(\xi'-\Delta)e^{-i\xi' t}-(\xi'+\Delta)e^{i\xi' t}](F_{11}-F_{22})\}\ , \tag{3.6e}$$

$$\rho_{02}^{c}(t)=\rho_{01}^{c}(t;F_{11}\leftrightarrow F_{22})\ , \tag{3.6f}$$

$$\rho_{03}^{c}(t)\simeq\frac{e^{2i\omega t}(\xi'^{2}-\Delta^{2})}{16\xi'^{4}}(F_{00}-F_{11}-F_{22}+F_{33})$$
$$\times[2(\xi'^{2}-3\Delta^{2})+4\Delta(\xi'+\Delta)e^{i\xi' t}-4\Delta(\xi'-\Delta)e^{-i\xi' t}-(\xi'+\Delta)^{2}e^{2i\xi' t}-(\xi'-\Delta)^{2}e^{-2i\xi' t}]\ , \tag{3.6g}$$

$$\rho_{12}^{c}(t)\simeq\frac{1}{8\xi'^{4}}(F_{00}-F_{11}-F_{22}+F_{33})[\xi'^{4}+2\xi'^{2}\Delta^{2}-3\Delta^{4}-4\Delta^{2}(\xi'^{2}-\Delta^{2})\cos\xi' t-(\xi'^{2}-\Delta^{2})^{2}\cos 2\xi' t]\ , \tag{3.6h}$$

$$\rho_{13}^{c}(t)\simeq\frac{ig\sqrt{\bar{n}}\,e^{i\omega t}}{8\xi'^{4}}\{2\Delta[3(\xi'^{2}-\Delta^{2})F_{00}-(\xi'^{2}-3\Delta^{2})(F_{11}+F_{22})-(\xi'^{2}+3\Delta^{2})F_{33}+2\xi'^{2}(F_{11}-F_{22})]$$
$$+e^{+i\xi' t}2(\xi'+\Delta)[-(\xi'-\Delta)(\xi'+2\Delta)F_{00}+\Delta(\xi'-2\Delta)(F_{11}+F_{22})$$
$$+(\xi'^{2}-\xi'\Delta+2\Delta^{2})F_{33}-\xi'^{2}(F_{11}-F_{22})]$$
$$+e^{-i\xi' t}2(\xi'-\Delta)[(\xi'+\Delta)(\xi'-2\Delta)F_{00}+\Delta(\xi'+2\Delta)(F_{11}+F_{22})$$
$$-(\xi'^{2}+\xi'\Delta+2\Delta^{2})F_{33}+\xi'^{2}(F_{11}-F_{22})]$$
$$+(\xi'^{2}-\Delta^{2})[(\xi'+\Delta)e^{2i\xi' t}-(\xi'-\Delta)e^{-2i\xi' t}](F_{00}-F_{11}-F_{22}+F_{33})\}\ , \tag{3.6i}$$

$$\rho_{23}^{c}(t)=\rho_{13}^{c}(t;F_{11}\leftrightarrow F_{22})\ . \tag{3.6j}$$

The notation $F_{11}\leftrightarrow F_{22}$ means that F_{11} and F_{22} should be interchanged, which reverses the sign of the terms that are proportional to $(F_{11}-F_{22})$.

It is seen from (3.6a)–(3.6d) that the spectrum of the diagonal matrix elements of ρ^{c} consists of five frequency components of 0, $\pm\xi'$, and $\pm 2\xi'$. In the standard Rabi oscillations of two-level atoms, one has only three components corresponding to 0 and $\pm\xi'$. Thus, the spectral lines of $\rho_{\mu\mu}^{c}$ at $\pm 2\xi'$ arise from the pairing of the two atoms. Note that the components at $\pm\xi'$ and $\pm 2\xi'$ disappear when $\zeta\ll 1$, which may be called the weak-coupling regime:

$$\rho_{00}^{c}(t)\xrightarrow[\zeta\ll 1]{}f_{0}^{2}\ ,$$
$$\rho_{11}^{c}(t),\rho_{22}^{c}(t)\xrightarrow[\zeta\ll 1]{}f_{0}(1-f_{0})\ , \tag{3.7}$$
$$\rho_{33}^{c}(t)\xrightarrow[\zeta\ll 1]{}(1-f_{0})^{2}\ .$$

As expected, the diagonal elements of ρ^{c} reduce in this limit to the probabilities of the formation of the pair states calculated from the asymptotic atomic probabilities. In the other limit $\zeta\gg 1$, which may be called the strong-coupling regime, ρ_{00}^{c} and ρ_{33}^{c} have five frequency components, but ρ_{11}^{c} and ρ_{22}^{c} do not have the components at $\pm\xi'$:

$$\rho_{00}^{c}(t)\xrightarrow[\zeta\gg 1]{}\tfrac{1}{4}+\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}+(f_{0}-\tfrac{1}{2})\cos(\Omega_{R}|\cos\theta|t)$$
$$+\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}\cos(2\Omega_{R}|\cos\theta|t)\ ,$$
$$\rho_{11}^{c}(t),\rho_{22}^{c}(t)\xrightarrow[\zeta\gg 1]{}\tfrac{1}{4}-\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}$$
$$-\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}\cos(2\Omega_{R}|\cos\theta|t)\ , \tag{3.8}$$
$$\rho_{33}^{c}(t)\xrightarrow[\zeta\gg 1]{}\tfrac{1}{4}+\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}-(f_{0}-\tfrac{1}{2})\cos(2\Omega_{R}|\cos\theta|t)$$
$$+\tfrac{1}{2}(f_{0}-\tfrac{1}{2})^{2}\cos(2\Omega_{R}|\cos\theta|t)\ ,$$

where we used the fact that ξ' becomes $\Omega_{R}|\cos\theta|$ for $\zeta\gg 1$. In order to compare (3.8) with the standard atomic Rabi spectrum, let us assume that asymptotically both of the atoms are in the excited state, that is $f_{0}=0$:

$$\rho_{00}^{c}(t)\rightarrow[\sin^{2}(\tfrac{1}{2}\Omega_{R}|\cos\theta|t)]^{2}\ ,$$
$$\rho_{11}^{c}(t),\rho_{22}^{c}(t)\rightarrow\sin^{2}(\tfrac{1}{2}\Omega_{R}|\cos\theta|t)\cos^{2}(\tfrac{1}{2}\Omega_{R}|\cos\theta|t)\ , \tag{3.9}$$
$$\rho_{33}^{c}(t)\rightarrow[\cos^{2}(\tfrac{1}{2}\Omega_{R}|\cos\theta|t)]^{2}\ .$$

Here $\sin^{2}(\Omega_{R}|\cos\theta|t/2)$ and $\cos^{2}(\Omega_{R}|\cos\theta|t/2)$ are the time-dependent probabilities of the lower and upper states of a two-level atom which are obtained in the standard Rabi oscillation problem. The limits in (3.9)

represent the occupation probabilities obtained simply from the pairing of two statistically independent atoms. The cooperative nature of the motions of the two atoms is more clearly manifested in the off-diagonal elements of $\rho^c(t)$.

It is seen from (3.6e)–(3.6j) that the spectral lines of ρ^c_{01}, ρ^c_{02}, ρ^c_{13}, and ρ^c_{23} are at ω, $\omega\pm\xi'$, and $\omega\pm2\xi'$. The lines of ρ^c_{12} are at 0, $\pm\xi'$, and $\pm2\xi'$. ρ^c_{03} has spectral lines at 2ω, $2\omega\pm\xi'$, and $2\omega\pm2\xi'$. In the weak-coupling limit all of these off-diagonal matrix elements vanish, ρ^c_{03} and ρ^c_{12} as ζ^2, the others as ζ. On the other hand, in the strong-coupling limit,

$$\begin{aligned}
&\rho^c_{01},\rho^c_{02}\xrightarrow[\zeta\gg1]{}\tfrac{1}{2}e^{i\omega t-i\phi_g}(f_0-\tfrac{1}{2})\\
&\qquad\times[\sin(\Omega_R|\cos\theta|t)\\
&\qquad\quad+(f_0-\tfrac{1}{2})\sin(2\Omega_R|\cos\theta|t)]\ ,\\
&\rho^c_{13},\rho^c_{23}\xrightarrow[\zeta\gg1]{}\tfrac{1}{2}e^{i\omega t+i\phi_g}(f_0-\tfrac{1}{2})\\
&\qquad\times[\sin(\Omega_R|\cos\theta|t)\\
&\qquad\quad-(f_0-\tfrac{1}{2})\sin(2\Omega_R|\cos\theta|t)]\ ,\\
&\rho^c_{12}\xrightarrow[\zeta\gg1]{}\tfrac{1}{2}(f_0-\tfrac{1}{2})^2\ ,\\
&\rho^c_{03}\xrightarrow[\zeta\gg1]{}\tfrac{1}{2}e^{2i\omega t}(f_0-\tfrac{1}{2})^2[1-\cos(2\Omega_R|\cos\theta|t)]\ ,
\end{aligned}\tag{3.10}$$

where we set $g=|g|\exp(i\phi_g)$. From (2.41), one finds

$$\sin\phi_g=\frac{i\mathcal{V}_{11;00}\cos\theta}{[4(E_1-E_0)^2+\mathcal{V}_{11;00}|^2]^{1/2}}\ .\tag{3.11}$$

The first four elements in (3.10) oscillate at $\omega\pm\Omega_R|\cos\theta|$ and $\omega\pm2\Omega_R|\cos\theta|$, which can be expected from single photon transitions and from the fact that both atoms are executing Rabi oscillations with frequency $\Omega_R|\cos\theta|$. It is interesting that ρ^c_{12} is independent of time as well as of the field. In other words, there is a static polarization between the two degenerate states of the complex in the strong-coupling limit. The implications of ρ^c_{12} are discussed further in Sec. V. ρ^c_{03} oscillates at 2ω and $2\omega\pm2\Omega_R|\cos\theta|$ in the strong-coupling limit. If we put $f_0=0$ as before, the limits in (3.10) become

$$\begin{aligned}
&\rho^c_{01},\rho^c_{02}\xrightarrow[\zeta\gg1]{}-e^{i(\omega t-\phi_g)}\sin^3(\tfrac{1}{2}\Omega_R|\cos\theta|t)\\
&\qquad\times\cos(\tfrac{1}{2}\Omega_R|\cos\theta|t)\ ,\\
&\rho^c_{13},\rho^c_{23}\xrightarrow[\zeta\gg1]{}-e^{i(\omega t+\phi_g)}\sin(\tfrac{1}{2}\Omega_R|\cos\theta|t)\\
&\qquad\times\cos^3(\tfrac{1}{2}\Omega_R|\cos\theta|t)\ ,\\
&\rho^c_{12}\xrightarrow[\zeta\gg1]{}\tfrac{1}{8}\ ,\\
&\rho^c_{03}\xrightarrow[\zeta\gg1]{}e^{2i\omega t}\sin^2(\tfrac{1}{2}\Omega_R|\cos\theta|t)\cos^2(\tfrac{1}{2}\Omega_R|\cos\theta|t)\ .
\end{aligned}\tag{3.12}$$

The off-diagonal matrix elements in (3.12) should be compared with the off-diagonal density-matrix element of a two-level atom executing Rabi oscillations, starting from its excited state, which is given by

$$-\exp(-i\omega t)\sin(\Omega^A_R|\cos\theta|t/2)\cos(\Omega^A_R|\cos\theta|t/2)\ ,$$

where $\Omega^A_R=\Omega_R(V=0)$. The off-diagonal elements of $\rho^c(t)$ generate a polarization field through the matrix elements of the dipole moment operator of the complex in the scattering-state representation, given by (2.13). In principle, the polarization field can have frequency components at 0, ξ', $2\xi'$, ω, $\omega\pm\xi'$, $\omega\pm2\xi'$, 2ω, $2\omega+\xi'$, and $2\omega\pm2\xi'$. Relative strengths of various components depend on the detuning from the atomic resonance, on the intensity of the fundamental mode used in (2.16), as well as on its statistics, and on the collision potential V. Some of the spectral components can be suppressed in certain coupling regimes. The time-dependent polarization field of the complex couples to the other modes of the electromagnetic field ($\neq\omega$) which have the same frequencies as those listed above. Thus, several new components appear in the usual Mollow spectrum. These new components carry information about the individual matrix elements of the collision potential.

The matrix elements of $\rho^c(t)$ in (3.6) exhibit an interesting effect if the initial conditions correspond to the threshold for the inversion of the atomic population. At this threshold

$$f_0=f_1=\tfrac{1}{2}\ ,\tag{3.13a}$$

$$\rho^c_{\mu\mu'}(t;f_0=\tfrac{1}{2})=\tfrac{1}{4}\delta_{\mu\mu'}\ .\tag{3.13b}$$

All of the off-diagonal elements vanish and the density matrix of the complex reduces to a constant times the unit matrix [Eq. (3.13b) also implies that ρ^c is proportional to the unit matrix in the representation of the asymptotic states since the transformation induced by Ω is unitary; see Eq. (2.39b)]. Thus, the collisionally induced effects vanish, and the threshold for the stimulated emission from the atoms is also a critical point for the collisional effects.

We should emphasize that this conclusion holds to the degree of the validity of the approximation made for $F_{\mu\mu'}$ in going from (2.37) to (3.5). At the threshold, the factors arising from f_0 and f_1 in (2.37) are equal to $\frac{1}{4}$, and $F_{\mu\mu'}$ can be written as

$$F_{\mu\mu'}=\tfrac{1}{4}\sum_{\mu''}\Omega^\dagger_{\mu\mu''}(E^c_\mu)\Omega_{\mu''\mu'}(E^c_{\mu'})\ .\tag{3.14}$$

Although this expression yields $\frac{1}{4}$ for the diagonal elements due to the unitarity of Ω, it does not vanish for the off-diagonal elements of F because of the different energy parameters in Ω's. $F_{\mu\mu'}$ for $\mu\neq\mu'$ is at least of the order of V. Whether the threshold is still a critical point when such terms are included is an open question.

As the preceding discussion makes it clear, it is the whole complex that couples to the electromagnetic field. $\rho^c(t)$ is therefore a physically more significant quantity than the atomic density matrix $\rho^{\rm at}(t)$ whose elements are given by (2.39a). Nevertheless, we will discuss at least certain parts of $\rho^{\rm at}(t)$ in order to exhibit certain features that arise from collisions, as well as those which are familiar from the standard two-level atom problem. Because of the form of $\Omega\Omega^\dagger$, $\rho^{\rm at}(t)$ can be decomposed into three parts. From (1.1b) and (2.5b) one sees that

$$\Omega_{\mu\mu'}(E_{\mu'})\Omega^{*}_{\mu''\mu'''}(E_{\mu'''})=\delta_{\mu\mu'}\delta_{\mu''\mu'''}+\left[\delta_{\mu\mu'}\langle\mu'''|V\frac{1}{E_{\mu'''}-E_{\mu''}-V-i\epsilon}|\mu''\rangle+\delta_{\mu''\mu'''}\langle\mu|\frac{1}{E_{\mu'}-E_{\mu}-V+i\epsilon}V|\mu'\rangle\right]$$
$$+\langle\mu|\frac{1}{E_{\mu'}-E_{\mu}-V+i\epsilon}V|\mu'\rangle\langle\mu'''|V\frac{1}{E_{\mu'''}-E_{\mu''}-V-i\epsilon}|\mu''\rangle\ . \tag{3.15a}$$

We have grouped the terms according to the number of Kronecker δ's. The term with two Kronecker δ's is of the zeroth order in V. The terms with one Kronecker δ are at least of the order of V. The last term is at least of the order of V^2. One can decompose $\rho^{\rm at}(t)$ accordingly:

$$\rho^{at}(t)=\rho^{\rm I}(t)+\rho^{\rm II}(t)+\rho^{\rm III}(t)\ . \tag{3.15b}$$

$\rho^{\rm I}(t)$ corresponds to the first term of (3.15a). $\rho^{\rm II}(t)$ corresponds to the terms with one Kronecker δ. $\rho^{\rm III}(t)$ corresponds to the last term of (3.15a). We will discuss only $\rho^{\rm I}(t)$. Let us write it as

$$\rho^{\rm I}_{\alpha i;\alpha' i'}=e^{-i(\alpha-\alpha')\omega t}\sum_{s=0,\pm1,\pm2}r^{\rm Is}_{\alpha i;\alpha' i'}e^{is\xi' t}\ . \tag{3.16}$$

Then the diagonal elements are given by

$$r^{\rm I\pm2}_{\alpha i;\alpha i}=0\ , \tag{3.17a}$$
$$r^{\rm I0}_{0i;0i}=\frac{1}{4}+\frac{(2f_0-1)}{4(1+\xi^2\cos^2\theta)}\ , \tag{3.17b}$$
$$r^{\rm I\pm1}_{0i;0i}=\frac{\xi^2\cos^2\theta}{8(1+\xi^2\cos^2\theta)}(2f_0-1)\ , \tag{3.17c}$$
$$r^{\rm Is}_{1i;1i}=r^{\rm Is}_{0i;0i}(f_0\rightarrow1-f_0)\ . \tag{3.17d}$$

Equation (3.17) means that, if we focus on atomic behavior, we find that the atomic population probabilities execute the standard Rabi oscillations. In $\rho^{\rm I}(t)$, the effect of the scattering on atomic population probabilities is to modify the standard Rabi frequency by modifying the coupling coefficient to the radiation field. The off-diagonal elements of $\rho^{\rm I}(t)$, however, have spectral components at $\omega\pm2\xi'$ as well as at ω and $\omega\pm\xi'$:

$$r^{\rm I0}_{0i;1i}=\frac{i\xi|\cos\theta|}{16(1+\xi^2\cos^2\theta)^2}\{(4+\xi^2\cos^2\theta)[e^{-i\phi_g}f_0^2-e^{i\phi_g}(1-f_0)^2]$$
$$+3\xi^2\cos^2\theta[e^{i\phi_g}f_0^2-e^{-i\phi_g}(1-f_0)^2]+4i\sin\phi_g(2-\xi^2\cos^2\theta)f_0(1-f_0)\}\ , \tag{3.18a}$$
$$r^{\rm I1}_{0i;1i}=\frac{i\xi|\cos\theta|[1+(1+\xi^2\cos^2\theta)^{1/2}]}{16(1+\xi^2\cos^2\theta)^2}\{[-1+\xi^2\cos^2\theta+(1+\xi^2\cos^2\theta)^{1/2}][-f_0^2e^{i\phi_g}+(1-f_0)^2e^{-i\phi_g}]$$
$$+[3+\xi^2\cos^2\theta-(1+\xi^2\cos^2\theta)^{1/2}][-f_0^2e^{i\phi_g}+(1-f_0)^2e^{-i\phi_g}]$$
$$+4i\sin\phi_g[-2+(1+\xi^2\cos^2\theta)^{1/2}]f_0(1-f_0)\}\ , \tag{3.18b}$$
$$r^{\rm I-1}_{0i;1i}=r^{\rm I1}_{0i;1i}[(1+\xi^2\cos^2\theta)^{1/2}\rightarrow-(1+\xi^2\cos^2\theta)^{1/2}]\ , \tag{3.18c}$$
$$r^{\rm I2}_{0i;1i}=\frac{-\xi^3|\cos\theta|^3[1+(1+\xi^2\cos^2\theta)^{1/2}]}{4(1+\xi^2\cos^2\theta)^2}(\sin\phi_g)(f_0-\tfrac{1}{2})^2\ , \tag{3.18d}$$
$$r^{\rm I-2}_{0i;1i}=r^{\rm I2}_{0i;1i}[(1+\xi^2\cos^2\theta)^{1/2}\rightarrow-(1+\xi^2\cos^2\theta)^{1/2}]\ . \tag{3.18e}$$

The components at $\omega\pm2\xi'$ are generated entirely by the collision process. In the limit of strong coupling,

$$\rho^{\rm I}_{\alpha i;\alpha i}(t)\xrightarrow[\xi\gg1]{}\tfrac{1}{4}+(-1)^{\alpha}\tfrac{1}{2}(f_0-\tfrac{1}{2})\cos(\Omega_R|\cos\theta|t)\ ,$$
$$\rho^{\rm I}_{0i;1i}(t)\xrightarrow[\xi\gg1]{}\frac{e^{i\omega t}}{2}(f_0-\tfrac{1}{2})[\cos\phi_g\sin(\Omega_R|\cos\theta|t)-i(f_0-\tfrac{1}{2})\sin\phi_g\sin(2\Omega_R|\cos\theta|t)]\ . \tag{3.19}$$

These have the same form as one might have expected on the basis of the Rabi oscillations of the two-level atoms except for the second term in the off-diagonal element. This second term represents the collisionally induced components of the atomic Mollow spectrum, which are separated from the fundamental frequency by twice as much as are the standard components. Note that there is a tradeoff between the standard side components of the Mollow spectrum and the collisionally induced components in (3.19). The amplitudes of the former are proportional to $\cos\phi_g$, the amplitudes of the latter to $\sin\phi_g$. It is seen from (3.11) that $\sin\phi_g$ compares the potential energy to the energy separation of the atoms. $\sin\phi_g$ goes to zero linearly as $V\rightarrow0$. On the other hand, if the potential energy is much larger than the atomic-energy separation, the standard Mollow side peaks tend to disappear and one is left with the collisionally induced components alone. The direct experimental observation of

the atomic density matrix as given in (3.19) by means of optical method appears to be quite difficult but feasible. One needs to design experiments in which the atoms are transferred (one at a time on average and within a finite time period) from the region where collisions and the coupling to the fundamental mode take place to a region where they are isolated and decay.

The threshold for the atomic population inversion is also a critical point for the reduced density matrix $\rho^{\rm at}(t)$. Using (2.39), one can write the matrix elements of $\rho^{\rm at}(t)$ as

$$\rho^{\rm at}_{01;01}(t)=\tfrac{1}{2}\sum_{\eta=1,2}\sum_{\mu,\mu'}\Omega_{\eta\mu}(E_\mu)\Omega^*_{\eta\mu'}(E_{\mu'})\rho^c_{\mu\mu'}(t)\ , \tag{3.20a}$$

$$\rho^{\rm at}_{02;02}(t)=\rho^{\rm at}_{01;01}(t;F_{11}\leftrightarrow F_{22})\ , \tag{3.20b}$$

$$\rho^{\rm at}_{11;11}(t)=\tfrac{1}{2}\sum_{\eta=2,3}\sum_{\mu,\mu'}\Omega_{\eta\mu}(E_\mu)\Omega^*_{\eta\mu'}(E_{\mu'})\rho^c_{\mu\mu'}(t)\ , \tag{3.20c}$$

$$\rho^{\rm at}_{12;12}(t)=\rho^{\rm at}_{11;11}(t;F_{11}\leftrightarrow F_{22})\ , \tag{3.20d}$$

$$\rho^{\rm at}_{01;11}(t)=\tfrac{1}{2}\sum_{\mu,\mu'}[\,\Omega_{0\mu}(E_\mu)\Omega^*_{2\mu'}(E_{\mu'})+\Omega_{1\mu}(E_\mu)\Omega^*_{3\mu'}(E_{\mu'})]\rho^c_{\mu\mu'}(t)\ , \tag{3.20e}$$

$$\rho^{\rm at}_{02;12}(t)=\rho^{\rm at}_{01;11}(t;F_{11}\leftrightarrow F_{22})\ . \tag{3.20f}$$

It follows from the unitarity of Ω and from (3.13b) that, at the threshold,

$$f_0=\tfrac{1}{2}\Longrightarrow\rho^{\rm at}_{\alpha i;\alpha' i'}(t)=\tfrac{1}{4}\delta_{\alpha\alpha'}\delta_{ii'}\ , \tag{3.21a}$$

$$\sum_i\rho^{\rm at}_{\alpha i;\alpha' i}(t)=\tfrac{1}{2}\delta_{\alpha\alpha'}\ . \tag{3.21b}$$

Thus, all off-diagonal atomic correlations, radiative as well as collisional, vanish at the point where stimulated emission at the atomic frequency can begin.

IV. COMMENT ON THE BINARY-COLLISION APPROXIMATION

The discussions of Secs. II and III concern just one collision complex. When there are N independent complexes, the orientation angle θ becomes a stochastic variable. If $\mathcal{O}$ is an additive operator for the N-complex system,

$$\mathcal{O}=\sum_{p=1}^{N}\mathcal{O}_p\ , \tag{4.1}$$

then its average value is given by a stochastic average over the traces of $\mathcal{O}_p$'s with $\rho^c(t)$:

$$\langle\langle\mathcal{O}(t)\rangle\rangle_\theta=\sum_{p=1}^{N}\tfrac{1}{2}\int_{-1}^{1}d(\cos\theta){\rm Tr}[\rho^c(\cos\theta,t)\times\mathcal{O}_p(\cos\theta)]=\tfrac{1}{2}N\int_{-1}^{1}d(\cos\theta){\rm Tr}[\rho^c(\cos\theta,t)\mathcal{O}_1(\cos\theta)]\ . \tag{4.2}$$

$\langle\langle\mathcal{O}(t)\rangle\rangle_\theta$ describes a property of the $2N$-atom system in the binary-collision approximation. Thus, quantities like the time-dependent distribution of atoms, the dipole moment density, etc., become simple averages over θ.

The simplicity of the above stochastic average hides the significant shift in the perspective with which one views collisions in radiative interactions in the scattering-state representation as compared to the standard picture of many-particle systems interacting with radiation fields. In the standard picture, one visualizes an atom wandering around and undergoing collisions at different places and times. After each collision, the atom loses the memory of any previous collision event. This means that after each collision, a new particle history starts. One also assumes that each collision event takes only a short while and that most of radiative interactions of the atom occur when the atom is in between two collisional events. In contrast, the scattering states have precisely defined energies. As far as collisional processes are concerned, they take into account entire particle histories, that is to say, entire particle trajectories. For the description of a $2N$-particle system, one superposes N histories of collision complexes one upon another. In this new picture, a radiative transition means substituting one pair history for another under the influence of the field. This is analogous to a Feynman path integration in which one uses expansions in terms of entire trajectories rather than individual space-time points.[16]

The contrast between the standard and scattering-state pictures indicates that the scattering-state representation can be particularly useful in analyses of coherent phenomena in many-particle systems. When entire particle histories are used in their descriptions, atoms may have partial memories about collision events in the presence of coherent radiation fields. Memories are partial because of superpositions of many distinct histories. The various quantum amplitudes of the independent complexes interfere. What survives these interferences represents a collective mode of the entire system. Surviving memory effects correspond to correlations among atoms and are embodied in collective modes. Because radiative interactions and collisions are treated on equal terms, the order that a coherent field may impose on the material system is taken into account in a rigorous quantum formalism, but in a physically transparent way. We have applied the present method to a collisionally triggered second-harmonic generation in atomic vapors[5] and expect it to be useful for analyses of other collisionally triggered coherence phenomena, such as the collision-induced four-wave mixing.[17]

As examples of the stochastic average in (4.2), we briefly consider the state distribution of complexes and the dipole moment density in the strong-coupling limit. From (3.8) and (4.2), one finds that the average number of complexes in the state μ is given by

$$\langle\langle N^c_\mu(t)\rangle\rangle_\theta=N\left[\eta^{(0)}_\mu+\eta^{(1)}_\mu\left[\frac{\sin\Omega_R t}{\Omega_R t}\right]+\eta^{(2)}_\mu\left[\frac{\sin 2\Omega_R t}{2\Omega_R t}\right]\right]\ , \tag{4.3a}$$

where

$$\eta_0^{(0)}=\eta_3^{(0)}=\tfrac{1}{4}+\tfrac{1}{2}(f_0-\tfrac{1}{2})^2 ,$$
$$\eta_1^{(0)}=\eta_2^{(0)}=\tfrac{1}{4}-\tfrac{1}{2}(f_0-\tfrac{1}{2})^2 ,$$
$$\eta_0^{(1)}=-\eta_3^{(1)}=f_0-\tfrac{1}{2} , \quad (4.3b)$$
$$\eta_1^{(1)}=\eta_2^{(1)}=0 ,$$
$$\eta_0^{(2)}=-\eta_1^{(2)}=-\eta_2^{(2)}=\eta_3^{(2)}=\tfrac{1}{2}(f_0-\tfrac{1}{2})^2 .$$

It is seen that the time-dependent distribution of the complexes exhibits decaying oscillations about certain constants. The same type of ringing behavior is also observed in the polarization density. Writing (2.13) in the form

$$\langle\mu|\mathbf{D}|\mu'\rangle=\mathbf{d}_{01}S_{\mu\mu'} , \quad (4.4)$$

one finds that the polarization density of the N-complex system is given by

$$\mathbf{P}(t)=N\langle \mathrm{Tr}\mathbf{D}\rho^c(t)\rangle_\theta \xrightarrow[\xi\gg 1]{} \frac{i}{2}\hat{\epsilon}|\mathbf{d}_{01}|(f_0-\tfrac{1}{2})e^{-i\omega t}\sin\tilde{\phi}_g$$
$$\times[(S_{01}+S_{02}-S_{13}-S_{23})R(\Omega_R t)+(f_0-\tfrac{1}{2})(S_{01}+S_{02}+S_{13}+S_{23})R(2\Omega_R t)]+\text{c.c.}, \quad (4.5a)$$

where

$$\sin\tilde{\phi}_g=\frac{i\mathcal{V}_{11;00}}{[4(E_1-E_0)^2+|\mathcal{V}_{11;00}|^2]^{1/2}} , \quad (4.5b)$$

$$R(x)=\left[\frac{2}{x^3}-\frac{1}{x}\right]\cos x+\frac{2\sin x}{x^2}-\frac{2}{x^3} . \quad (4.5c)$$

If the two-level atoms were like independent spins, one would expect their dipole moments to align along the polarization vector $\hat{\epsilon}$ of the intense field mode without any reference to the collision potential. But, in (4.5a), $\mathbf{P}$ is proportional to $\sin\tilde{\phi}_g$ which depends on the collision potential. Clearly, (4.5a) shows that the collisional process does not permit the atoms to act independently. The quantity $\sin\tilde{\phi}_g$ measures the degree to which the two field-excited atomic dipoles within one complex can coordinate with each other. The polarization of the entire system is then proportional to $\sin\tilde{\phi}_g$.

$P(t)$ given by (4.5a) has frequency components at $\omega\pm\Omega_R$ and $\omega\pm 2\Omega_R$. The frequency components of $\rho_{02}^c(t)$ near 2ω, as given by (3.10), do not survive the average in (4.2). If we had kept the off-diagonal elements of $F_{\mu\mu'}$ in (2.38), some of these would have introduced an extra factor of g into the amplitudes of $\rho_{02}^c(t)$. Such terms survive the average in (4.2) and contribute to the second-harmonic generation in atomic vapors.[5]

V. CONCLUDING REMARKS

In the preceding sections we used the scattering-state representation in conjunction with a dressed atom formalism in the analysis of the Rabi oscillations of a pair of two-level atoms. This method provided for the simultaneous analysis of collisional and radiative processes in a mathematically simple fashion. It also provided a new perspective of the effects of collisions on radiative transitions. The preceding discussion makes it clear that the method is particularly suitable for analyses of collisionally induced cooperative effects. Such cooperative effects can clearly be seen in the Mollow spectrum.

For a pair of colliding two-level atoms we found that there is, in principle, more than one Rabi frequency. The magnitudes of these different Rabi frequencies depend on the detuning from the atomic resonance and on the number of quanta in the field mode. For an intense coherent mode, these distinct frequencies merge, and one obtains a single Rabi frequency for the problem. One also recovers the usual Rabi and Mollow spectra. However, there are additional components in these spectra at twice the Rabi frequency which represent the pairing and the collisionally induced cooperative motions of the atoms.

It is important to emphasize that in the above results, the effect of the collisional process on radiative transitions appears as the modification of the magnitude and phase of the coupling coefficient between the complex and the field, hence, the modification of the magnitude of the Rabi frequency or frequencies. Collisional effects do not enter the results as lifetime parameters or energy uncertainties. This is a direct consequence of the radically different physical picture of collisions in the scattering-state representation compared to the standard time-dependent picture, as discussed in Sec. IV. In the scattering-state representation, spectral line broadening is interpreted as the opening of new coupling channels by V, due to partial breaking of the translational symmetry. This is discussed more extensively in Ref. 6. Because of the straightline trajectory approximation used in this paper, this symmetry-breaking aspect is somewhat hidden in the results. One deals only with the internal transitions of the atoms. Nevertheless, V opens up new radiative coupling channels with respect to internal transitions as seen from (2.13) and these represent a "line broadening" in the straightline trajectory approximation.

An interesting result of the analysis of the Rabi oscillations in the scattering-state representation is the fact that $\rho^c(t)$, as well as $\rho^{\mathrm{at}}(t)$ obtained by partially tracing over $\rho^c(t)$, becomes a multiple of the indentity matrix at the threshold for atomic population inversion, independent of time and orientation of atomic dipoles. The collisional effects disappear. As a result, the density-matrix elements in (4.2) are unaffected by the stochastic average at the threshold. One can visualize performing Kadanoff transformations[18] on the state distributions of the $2N$-atom system by partitioning it into smaller units of paired atoms. At the threshold, such transformations would simply reproduce the original distribution. This means that the threshold for the atomic population inversion is a "fixed point" not only with respect to radiative processes but also with respect to collisional processes. Strictly speaking, we demonstrated this only in the binary-

collision approximation and to the validity of using (3.5) for (2.37). It would be interesting to pursue the question of whether the threshold remains a critical point when these approximations are relaxed.

Another quite interesting result of the analysis is the appearance of a quasistatic polarization ρ_{12}^c. We will discuss this phenomenon in more detail elsewhere. Here we simply note that the dipole moment of the complex corresponding to ρ_{12}^c is given by

$$\mathbf{p}_s = \langle 1|\mathbf{D}|2\rangle \rho_{21}^c + \text{c.c.} , \tag{5.1}$$

which becomes completely static in the strong-coupling regime. Both collisions and radiative transitions are necessary to produce a finite $\mathbf{p}_s$. ρ_{21}^c vanishes without the coupling to the fundamental mode, $\langle 1|\mathbf{D}|2\rangle$ vanishes if V vanishes. The existence of two degenerate states for the complex is also critical. According to (5.1), each complex behaves as if it were a static dipole. Although the fundamental mode contributes to the existence of $\mathbf{p}_s$, it does not align these moments. This follows from the fact that a stochastic average over $\mathbf{p}_s$'s yields zero. However, such dipole moments can be aligned by static or quasistatic electric fields. Minute amounts of static electric fields may create large domains in which $\mathbf{p}_s$'s are aligned. Such responses yield information about matrix elements of collision potentials which may not be readily observable by other methods. For example, according to (2.8), (2.13), and (2.42), $\langle 1|\mathbf{D}|2\rangle$ is approximately given by

$$\langle 1|\mathbf{D}|2\rangle \cong 2\mathbf{d}_{01}\text{Re}\left[\frac{\mathcal{V}_{00;01}(0)}{E_1 - E_0 + i\epsilon}\right] . \tag{5.2}$$

Here we expanded (1.1b) in powers of V and kept only the linear term. The matrix element $\mathcal{V}_{00;01}(0)$ describes a transition in which one atom is deexcited while its partner remains in the ground state, with no momentum exchange between them. Finally, the formation of $\mathbf{p}_s$'s in intense light may be called the "dynamic paraelectricity," since dynamically induced $\mathbf{p}_s$'s in an electric field act very much like independent spins in a magnetic field.

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