Reduced-density-matrix description for pump-probe optical phenomena in moving atomic systems

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Linear and nonlinear (especially coherent) electromagnetic interactions of moving many-electron atoms are investigated using a reduced-density-matrix description, which is applied to electromagnetically induced transparency and related resonant pump-probe optical phenomena. External magnetic fields are included on an equal footing with the electromagnetic fields and spin-Zeeman interactions are taken into account. Complimentary time-domain (equation-of-motion) and frequency-domain (resolvent-operator) formulations of the reduced-density-matrix description are self-consistently developed. The general nonperturbative and non-Markovian formulations provide a fundamental framework for systematic evaluations of corrections to the standard Born (lowest-order-perturbation) and Markov (short-memory-time) approximations. The macroscopic electromagnetic response is described semiclassically, employing a perturbation expansion of the reduced-density operator in powers of the classical electromagnetic field. Our primary results are compact Liouville-space operator expressions for the linear and general (nth-order) nonlinear macroscopic electromagnetic-response tensors, which can be evaluated for nonlocal and nonstationary optical media described by multilevel atomic-system representations. Interactions among atoms and with environmental photons are treated as line-broadening effects by means of a general Liouville-space self-energy operator, for which the tetradic-matrix elements are explicitly evaluated in the diagonal, lowest-order, and Markov approximations. The compact Liouville-space operator expressions that are derived for the macroscopic electromagnetic-response tensors are introduced into the dynamical description of the electromagnetic-field propagation. It is pointed out that a quantized-electromagnetic-field approach will be required for a fully self-consistent quantum-mechanical treatment of local-field effects and radiative corrections.

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I. INTRODUCTION

Nonlinear electromagnetic interactions can significantly influence the propagation of (probe or signal) light pulses in ensembles of many-electron atomic systems that are simultaneously undergoing resonant and coherent excitations by intense (pump or control) electromagnetic fields [1,2]. The fundamental, microscopic description of coherent optical processes in quantized electronic systems and the precise evaluation of the linear and the nonlinear macroscopic electromagnetic response are the primary objectives of our investigation. A brief and preliminary account of our reduce-density-matrix description of pump-probe optical phenomena in moving many-electron atomic systems has been reported in Ref. [3]. We now report more details and further developments of our investigation.

A. Quantized electronic systems

We have been developing a general reduced-density-matrix approach for electromagnetic interactions of quantized electronic systems [4]. Our primary goal is a comprehensive fundamental framework for detailed and systematic theoretical and computational investigations of a wide variety of electromagnetic phenomena. Moving many-electron atomic systems in warm vapors are treated in this paper. Electromagnetic processes in solid-state systems, including metamaterials and quantum-confinement structures, are beyond the scope of the present investigation.

B. Electromagnetically induced transparency and related pump-probe optical phenomena

Harris *et al.* [5] presented the original description of electromagnetically induced transparency (EIT), in which a rapid frequency variation of the index of refraction within a narrow spectral range leads to a reduction in the group velocity of a propagating electromagnetic pulse within a coherently excited optical medium. Slow light produced by EIT, as well as by related resonant pump-probe optical phenomena, offers promising applications for optical communication [6,7]. Electromagnetically induced transparency in moving many-electron atomic systems and slow light are the primary applications of interest in this investigation. However, our general reduced-density-matrix description is applicable to a wide variety of resonant pump-probe optical phenomena and a diverse class of modified light-propagation characteristics.

C. Applications of the reduced-density-operator approach to atomic systems

Figure 1 schematically illustrates a pump-probe optical process, such as EIT, for a three-state Λ atomic-system model. Our reduced-density-operator description is applicable to electromagnetic processes involving an intense (pump or control) electromagnetic field and a weak (probe or signal) electromagnetic field, as well as to processes involving three or more different electromagnetic fields. We present the derivation of a compact Liouville-space operator expression for the general (nth-order) macroscopic electromagnetic response, which can be readily evaluated for a larger, more extensive manifold of many-electron atomic states, including bound excited, autoionizing, and nonresonant continuum states from the complete basis set.

An explicitly time-dependent approach based on the equation of motion for the reduced density operator is expected to be necessary for ultrafast optical excitation processes.

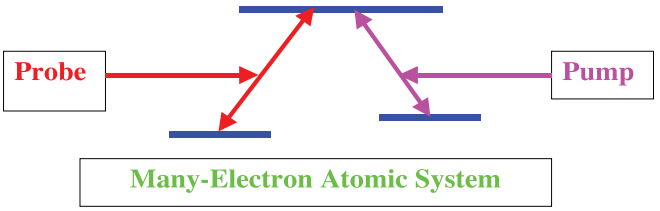

FIG. 1. (Color online) A pump-probe process, such as electromagnetically induced transparency, is illustrated for a three-state Λ atomic-system model and two distinct electromagnetic fields, which are referred to as the pump and probe fields.

A multiple-mode Floquet-Fourier expansion of the reduced density operator can provide the basis for a nonperturbative dressed-state approach that would be applicable for intense but stationary (continuous-wave) electromagnetic fields [8]. In our approach, the reduced density operator can be expanded in powers of either the weaker probe field alone or the entire combined electromagnetic field. In our semiclassical description, the ensemble of many-electron atomic systems corresponding to the optical medium is treated as a quantum system, while the electromagnetic field is assumed to be governed by the Maxwell equations of classical electrodynamics. However, we will point out that the development of a quantized-field formulation will be necessary for a fully self-consistent quantum-mechanical description.

D. External magnetic fields

We will treat pump-probe optical phenomena in the presence of external magnetic fields. Slow light within an enhanced spectral range has been produced in warm atomic vapors by applying an external magnetic field in the light propagation direction, with a transverse spatial dependence [9,10]. When different frequency components enter the EIT medium at suitably separated transverse locations, the required two-photon resonance condition, involving the pump and the probe electromagnetic fields, can be maintained at each transverse spatial location by the Zeeman shifts that are generated in the atomic systems. The individual Zeeman-split EIT spectral patterns have been observed for spatially homogeneous external magnetic fields [11,12].

E. Atomic collisions and other environmental interactions

The general expressions for the Liouville-space self-energy operators, which are derived in the frequency-domain and time-domain formulations of our reduced-density-operator description, provide a fundamental framework for the systematic treatment of the effects of interactions among atoms, on an equal footing with radiative processes. We point out that an unambiguous treatment of binary atom-atom collision processes can be most readily achieved for an optical medium consisting of two different atomic species. In many observations, a gas of different atomic systems has been intentionally injected into the optical medium in order to minimize the inhomogeneous (diffusion and transit-time) broadening produced in the many-electron atomic systems that can participate in the resonant electromagnetic interactions [9,13]. The tetradic matrix elements of the Liouville-space self-energy operators can be most simply evaluated in the Born (lowest-order-perturbation), Markov (short-memory-time), and diagonal-resolvent approximations. We explicitly evaluate the lowest-order spectral linewidths and line shifts and demonstrate that they can be expressed as sums of the conventional frequency-independent binary-collision cross sections and the radiative transition rates [14]. We emphasize that our general nonperturbative and non-Markovian tetradic-matrix expressions for the Liouville-space self-energy operators can be systematically evaluated for complex spectra consisting of a multitude of overlapping spectral lines, which may be produced by radiative transitions between enormous sets of closely spaced many-electron (fine-structure or hyperfine-structure) atomic levels, and our approach is not restricted to binary atomic interactions.

F. Organization of this paper

The remainder of this paper is organized in the following manner. In Sec. II, we present the frequency-domain (resolvent-operator) and time-domain (equation-of-motion) formulations of our reduced-density-matrix approach. The Liouville-space projection-operator formalism is exploited to obtain the Markovian and non-Markovian descriptions in compact Liouville-space operator forms. In Sec. III, a preliminary semiclassical treatment of the electromagnetic interaction is adopted. The reduced density operator, describing the ensemble of moving many-electron atomic systems corresponding to the optical medium, is expanded in powers of either the combined classical (pump and probe) electromagnetic field or the probe field alone. Time-domain and frequency-domain hierarchies of coupled relationships for the field components of the reduced density operator are obtained, incorporating the classical center-of-mass motions of the many-electron atomic systems in the presence of an external magnetic field as well as the pump and probe electromagnetic fields. Section IV is devoted to the precise evaluation of the macroscopic electromagnetic response, with particular emphasis on the frequency and wave-vector representation. Compact Liouville-space operator expressions are derived for the linear ($n = 1$) and the general ($n > 1$) nonlinear macroscopic electromagnetic-response tensors. The classical center-of-mass motions of the atomic systems are analytically incorporated by means of a Doppler-shifted Liouville-space resolvent operator. The macroscopic electromagnetic-response tensors can be evaluated for coherent initial atomic excitations and for the full tetradic-matrix form of the Liouville-space self-energy operator representing the environmental interactions either in the Markov approximation or adopting a model for non-Markovian behavior. In Sec. V, the tetradic-matrix elements of the Liouville-space self-energy operator are explicitly evaluated in the Born (lowest-order-perturbation), Markov (short-memory-time), and diagonal-resolvent approximations, taking into account binary atomic collisions and environmental radiative interactions. In Sec. VI, the linear and nonlinear propagation characteristics of the electromagnetic field are considered, based on the introduction of our general analytical expressions for the macroscopic electromagnetic response tensors into the macroscopic Maxwell field equations. It is pointed out that, in a self-consistent treatment of the electromagnetic

propagation, one would encounter the coupling with the longitudinal fields arising from any electrostatic interactions that are not included in the zeroth-order Hamiltonian operator. The need for the development of a fully quantum-mechanical description of the electromagnetic interaction is also emphasized. Our conclusions and future plans for various extensions of this investigation are presented in Sec. VII.

II. GENERAL DENSITY-OPERATOR APPROACH

We adopt a density-operator approach to develop a general nonperturbative and nonequilibrium quantum-statistical description of (possible coherent) electromagnetic interactions involving quantized many-electron systems. In order to provide a fundamental treatment for decoherence and relaxation processes, together with spectral line broadening mechanisms, we develop a quantum-open-systems (reduced-density-operator) description [15–18] to incorporate the interactions arising from a much larger system, which is designated as the environment.

The environment is represented by a time-independent density operator in the conventional reservoir approximation, which is illustrated in Fig. 2. Accordingly, the environment is assumed to remain essentially unaltered during its interactions with the relevant quantum system. The influences of the environment on the relevant quantum system are illustrated in Fig. 3, in terms of decoherence and relaxation processes together with spectral line broadening mechanisms. These stochastic kinetics and spectral phenomena are systematically and self-consistently incorporated by means of the Liouville-space self-energy corrections that are derived in the complimentary time-domain and frequency-domain formulations of our reduced-density-operator description.

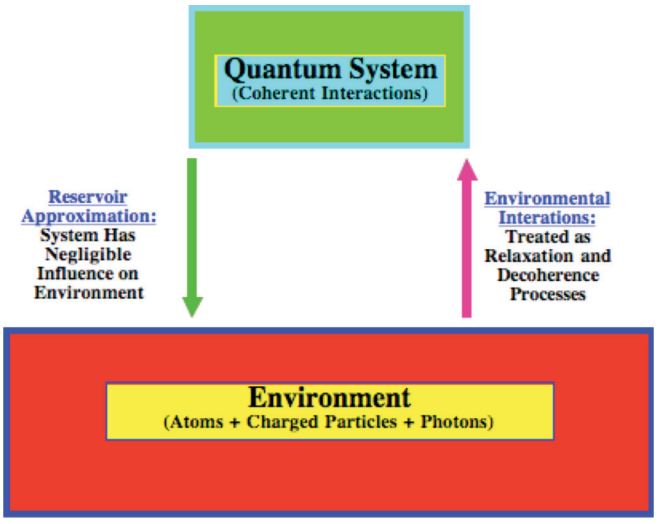

FIG. 2. (Color online) The reduced-density-operator description is illustrated in terms of the partition of the entire interacting system into a relevant quantum system (which may consist of an ensemble of many-electron atomic systems and a set of emitted or observable photons) and an environment (which may consist of many-electron atoms, charged particles, and photons), which is treated using the traditional reservoir approximation.

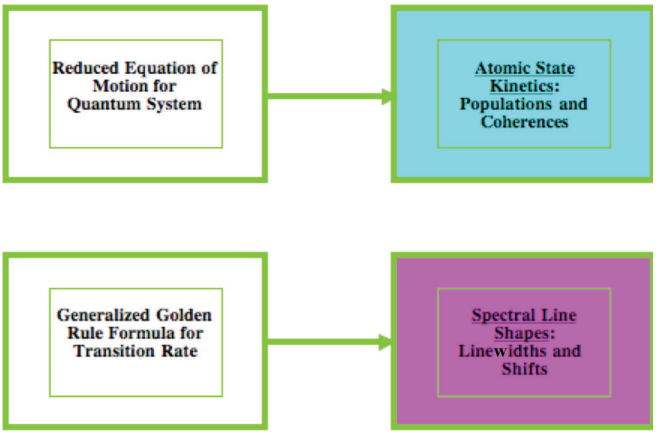

FIG. 3. (Color online) Time-domain (equation-of-motion) and frequency-domain (resolvent-operator) formulations of the reduced-density-operator approach can provide the fundamental framework for a unified treatment of atomic-state kinetics and spectral line shapes.

The partition of the entire interacting quantum system into a relevant quantum system and an environment is inherently arbitrary and by no means apparent. In the ordinary Hilbert-space description, different divisions of the total Hamiltonian operator into a zeroth-order (unperturbed) Hamiltonian operator and an interaction (or perturbation) operator would be equivalent if the interaction could be incorporated to all orders. In contrast, different partitions in the reduced-density-operator description are fundamentally inequivalent and will inevitably lead to dissimilar predictions. In this investigation, we point out the consequences of different partitions for ensembles of interacting many-electron atomic systems in an optical medium.

The statistical state of the combined, interacting (closed) quantum system is conventionally assumed to be initially expressible in the uncorrelated tensor-product form $\rho(t_0) = \rho^S \otimes \rho^E$. The statistical state of the relevant quantum system at the initial time t_0 is represented by the density operator ρ^S, while the quantum-statistical state of the environment is represented by the time-independent (reservoir-approximation) density operator ρ^E. In order to take into account the correlations between the relevant quantum system and the environment that will be generated by the environmental interactions, the complete density operator $\rho(t)$ representing the statistical state of the combined interacting quantum system, at an arbitrary time $t > t_0$, must not be constrained to the simple uncorrelated tensor-product form.

A. Reduced-density-operator description

The reduced relevant density operator defined by $\rho^r(t) = \mathrm{Tr}_E\{\rho(t)\}$ provides a proper representation of the statistical state of the relevant quantum system, at an arbitrary time t. The partial-trace operation indicated by Tr_E introduces a quantum-statistical average involving the large set of quantum numbers specifying the environmental degrees of freedom.

The reduced-density-operator description can be presented in compact forms by adopting the Liouville-space operator representation [8,19–24], in which the density operators play

the role of state vectors. We will make extensive use of the complex inner product defined as the trace $\langle\langle\rho_1|\ \rho_2\rangle\rangle = \mathrm{Tr}(\rho_1^+\rho_2)$ of the two Liouville-space state vectors $|\rho_1\rangle$ and $|\rho_2\rangle$, where the plus superscript indicates the adjoint.

B. Frequency-domain (resolvent-operator) formulation

In the frequency-domain (resolvent-operator) formulation of the reduced-density-operator description, the transition rate is given by the Liouville-space Fermi-golden-rule formula [3,8]

$$A_R(i \to f) = -\,i\ \lim_{\varepsilon\to 0}\left\langle\left\langle P_f^r\middle|\bar{T}^r(+i\varepsilon)\middle|\rho_i^r\right\rangle\right\rangle. \tag{1}$$

The asymptotic boundary condition is introduced by the $\varepsilon \to 0$ limit. We will employ the Lippmann-Schwinger relationship $\bar{T}^r(+i\varepsilon) = \bar{V}^r + \bar{V}^r\,\bar{G}^r(+i\varepsilon)\,\bar{V}^r$ to evaluate the reduced relevant Liouville-space transition operator $\bar{T}^r(+i\varepsilon)$. The reduced relevant Liouville-space resolvent (or Green) operator is defined by $\bar{G}^r(+i\varepsilon) = [+i\varepsilon - \bar{L}^r - \bar{\Sigma}(+i\varepsilon)]^{-1}$. The Liouville-space operators, which are denoted by overbars, must be specified by four indices in their matrix representations.

The relevant Liouvillian operator $\bar{L}^r$ is defined by means of the commutator relationship

$$\bar{L}^r\rho^r = (1/\hbar)\,[H^r,\rho^r]. \tag{2}$$

The relevant Hamiltonian operator H^r describes the many-electron quantum system together with the restricted set of relevant (observable) modes of the quantized electromagnetic field. The relevant Liouvillian operator is given by $\bar{L}^r = \bar{L}_0^r + \bar{V}^r$, where $\bar{L}_0^r$ is the zeroth-order (unperturbed) relevant Liouvillian operator corresponding to the zeroth-order relevant Hamiltonian operator H_0^r and the Liouville-space operator $\bar{V}^r$ includes the relevant electromagnetic interactions.

The initial statistical state of the relevant quantum system is represented by the reduced density operator ρ_i^r. The final state in the transition is specified by the relevant projection operator P_f^r. These operators act on the combined tensor-product states of the many-electron quantum system and the relevant (observable) modes of the quantized electromagnetic field.

1. Frequency-domain self-energy operator

The frequency-domain Liouville-space self-energy operator $\bar{\Sigma}(z)$ can be expressed, as a function of the complex variable z, in terms of the Zwanzig Liouville-space projection operators $\bar{P} = |\rho^E\rangle\rangle\langle\langle I^E|$ and $\bar{Q} = 1 - \bar{P}$, where I^E denotes the environmental identity operator [3,8]:

$$\begin{aligned}\bar{\Sigma}(z) &= \bar{P}\bar{V}^{\mathrm{ir}}\bar{P} + \bar{P}\bar{V}\bar{Q}\frac{1}{z-\bar{Q}\bar{L}\bar{Q}}\bar{Q}\bar{V}\bar{P}\\ &= \mathrm{Tr}_E\left[\left(\bar{V}^{\mathrm{ir}} + \bar{V}\bar{Q}\frac{1}{z-\bar{Q}\bar{L}\bar{Q}}\bar{Q}\bar{V}\right)\rho^E\right].\end{aligned} \tag{3}$$

The complete Liouvillian operator $\bar{L}$ is obtained from the total Hamiltonian operator $\bar{H}$ for the entire interacting quantum system. The total Liouville-space interaction operator $\bar{V}$ is decomposed as $\bar{V} = \bar{V}^r + \bar{V}^{\mathrm{ir}}$, where the irrelevant Liouville-space interaction operator $\bar{V}^{\mathrm{ir}}$ includes the environmental interactions. As a result of the quantum-statistical average (partial-trace operation Tr_E) involving the quantum numbers associated with the environmental degrees of freedom, the projection operator $\bar{P}$ projects onto the subspace of states corresponding to the relevant-system degrees of freedom (uncorrelated with the environmental degrees of freedom). The complementary projection operator $\bar{Q}$ projects onto the orthogonal subspace of states corresponding to the irrelevant (environmental) degrees of freedom (taking into account the system-environment correlations). We emphasize that the general reduced-density-operator description is applicable to nonequilibrium quantum-statistical distributions and that our fundamental quantum-mechanical treatment for the Liouville-space self-energy corrections can provides a systematic microscopic description of the environmental interactions, which are almost always treated in the literature by the introduction of phenomenological parameters.

When the system-environment interactions are sufficiently weak, the Liouville-space self-energy operator $\bar{\Sigma}$ may be expanded in a perturbation series involving increasing powers of the total Liouville-space interaction operator $\bar{V}$. Retaining only the lowest-order nonvanishing contribution, which corresponds to the Born approximation, we show in Sec. V that the total spectral line shift and linewidth in the diagonal-resolvent approximation can be reduced to the sums of the partial contributions from elementary collisional and radiative processes acting alone. Interference between transition amplitudes can occur in the high-order contributions to the linewidth and shift, as well as in our general tetradic-matrix expression valid for overlapping spectral lines.

2. Initial-state coherences

Coherent excitations of the many-electron quantum system are manifest by nondiagonal elements of the initial-state reduced density operator ρ_i^r in the tensor-product representation $|\alpha\rangle = |a\rangle \otimes |\{n_{\vec{k}\lambda}\}\rangle$ based on the unperturbed many-electron eigenstates $|a\rangle$ and the relevant photon states $|\{n_{\vec{k}\lambda}\}\rangle$. For a single-photon spontaneous-emission process, the relevant final-state projection operator P_f^r projects onto the manifold of tensor-product states corresponding to the unperturbed many-electron eigenstates $|b\rangle$ that can be created by the spontaneous emission of a single photon with momentum $\hbar\vec{k}$ (and energy $\hbar\omega$) and polarization (or helicity) λ. Using the Liouville-space Dirac notation $|\alpha\rangle\langle\beta| = |\alpha,\beta\rangle\rangle$, the operator eigenstate decompositions appropriate for a single-photon spontaneous-emission process can be expressed as follows:

$$\left|\rho_i^r\right\rangle\rangle = \sum_a\sum_{a'}|aa',00\rangle\rangle\left\langle\left\langle aa',00\middle|\rho_i^r\right\rangle\right\rangle, \tag{4}$$

$$\left|P_f^r\right\rangle\rangle = \sum_b |bb,11\rangle\rangle. \tag{5}$$

Our general formalism can be employed for single-photon absorption processes and for multiphoton (nonlinear optical) transitions, which involve photon-occupation numbers $n_{\vec{k},\lambda} > 1$.

C. Time-domain (equation-of-motion) formulation

The time-domain (equation-of-motion) formulation of the reduced-density-operator description is based on the

generalized master equation [3,8,25]

$$\frac{\partial}{\partial t}\,\rho^r(t) = -\,i\,\bar{L}^r(t)\rho^r(t) - i\int_{t_0}^{t} dt'\,\bar{\Sigma}(t,t')\rho^r(t')\,. \quad (6)$$

This closed-form equation of motion for the reduced, relevant density operator $\rho^r(t) = \bar{P}\rho(t)$ has been derived by neglecting the initial-state correlations. Initial-state correlations are automatically excluded by the conventional assumption that the entire initial-state density operator for the combined interacting system can be represented as an uncorrelated tensor product of individual density operators for the separate isolated subsystems.

1. Time-domain self-energy operator

The time-domain Liouville-space self-energy operator kernel $\bar{\Sigma}(t,t')$ can be expressed in terms of the Q-subspace projection of the Liouville-space propagator and related to the Liouville-space self-energy operator $\bar{\Sigma}(z)$ in Eq. (1) by a Fourier transformation [3,8,25]. The self-energy operator $\bar{\Sigma}(t,t')$ is independent of time in the traditional Markov (short-memory-time) approximation [8]

$$\bar{\Sigma}(t,t') = \lim_{z\to i0}\bar{\Sigma}(z)\delta(t-t'). \quad (7)$$

The frequency-domain Liouville-space self-energy operator $\bar{\Sigma}(i0)$, which subsequently will be denoted simply by $\bar{\Sigma}$, is independent of the frequency. For a completely consistent treatment of the non-Markovian dynamics, which may be important in ultrafast electromagnetic interactions, a suitable model should be introduced for the initial-state correlation term that was excluded in the derivation of the closed-form equation of motion for the reduced relevant density operator.

2. Many-electron-system and electromagnetic-field equations

An equation of motion for the density operator describing the ensemble of many-electron atomic systems can be derived from Eq. (6) by performing the additional quantum-statistical average (partial-trace operation) involving the relevant (observable) photon states. The dynamical equation for the density operator representing the quantum-statistical state of the electromagnetic field can be obtained from Eq. (6) by carrying out the complimentary additional quantum-statistical average (partial-trace operation) pertaining to the states representing the ensemble of many-electron atomic systems.

In the Born (lowest-order) and Markov (short-memory-time) approximations, the set of dynamical equations involving only the electronic-state population densities (diagonal reduced-density-matrix elements) can be expressed in terms of the familiar (lowest-order) radiative and nonradiative transition rates that are obtained from an evaluation of the standard Fermi-golden-rule formula of ordinary Hilbert-space perturbation theory. We will treat electromagnetic interactions in terms of the complete set of reduced-density-matrix equations, taking into account the electronic-state coherences (nondiagonal reduced-density-matrix elements).

III. SEMICLASSICAL REDUCED-DENSITY-OPERATOR DESCRIPTION

The reduced relevant density operator describing the ensemble of moving many-electron atomic systems will be assumed to satisfy a semiclassical equation of motion of the form [26]

$$\begin{aligned}&\partial\rho^r(\vec{r},\vec{v},t)/\partial t + \vec{v}\cdot\vec{\nabla}\rho^r(\vec{r},\vec{v},t)\\ &\quad= -i\big[\bar{L}_0^r + \bar{V}^r(t)\big]\rho^r(\vec{r},\vec{v},t)\\ &\qquad - i\int_{t_0}^{t} dt'\,\bar{\Sigma}(t,t')\rho^r(\vec{r},\vec{v},t')\\ &\quad\to -i\big[\bar{L}_0^r + \bar{V}^r(t)\big]\rho^r(\vec{r},\vec{v},t) - i\,\bar{\Sigma}\rho^r(\vec{r},\vec{v},t). \quad (8)\end{aligned}$$

Our general non-Markovian form is indicated by the equality, while the Markovian result is indicated by the arrow. The zeroth-order relevant Liouvillian operator $\bar{L}_0^r$ now describes the entire unperturbed many-electron quantum system alone. The explicit dependences of the reduced density operator $\rho^r(\vec{r},\vec{v},t)$ on the laboratory-frame position vector $\vec{r}$ and velocity vector $\vec{v}$ have been introduced to incorporate the classical translational (center-of-mass) motions of the many-electron atomic systems. The Liouville-space operators in Eq. (8) may also depend on these vectors. In the remainder of our analysis, the superscript r will be omitted.

For a completely consistent treatment of the quantum-statistical and quantum-coherence properties, the electromagnetic field must be included as an integral part of the relevant quantum system. This will also be necessary for a fully consistent treatment of local-field effects and radiative corrections. The quantization and relativistic treatment of the translational degrees of freedom will also be necessary for a full quantum-electrodynamics (QED) description.

A. Nonrelativistic electromagnetic interaction in the Coulomb gauge

We treat the atom-field interaction using a nonrelativistic approximation in the Coulomb gauge, for which the electromagnetic scalar potential may be set equal to zero in the absence of free charges. However, we will incorporate the magnetization due to the electron spins by including a lowest-order relativistic correction corresponding to the spin-Zeeman interaction. The interaction of the many-electron atomic system with an external magnetic field $\vec{B}_{\rm ext}(\vec{r},t)$ will be included by expressing the combined vector potential as the sum of the electromagnetic vector potential $\vec{A}(\vec{r},t)$ and the external vector potential $A_{\rm ext}(\vec{r},t)$. The total atomic-field interaction operator can be expressed as a sum of linear and quadratic contributions as follows:

$$V_{\rm AF}(t) = V_{\rm AF}^1(t) + V_{\rm AF}^2(t). \quad (9)$$

1. Linear and quadratic atom-field interactions

The manifestly Hermitian form of the total linear contribution $V_{\rm AF}^1(t)$ to the atom-field interaction can be written as

follows:

$$V^1_{\rm AF}(t) = \left(\frac{e}{2mc}\right)\sum_j [\vec{p}_j\cdot\vec{A}(\vec{r}_j,t) + \vec{A}(\vec{r}_j,t)\cdot\vec{p}_j] + \left(\frac{eg_s}{4mc}\right)\sum_j [\vec{s}_j\cdot\vec{\nabla}_j\times\vec{A}(\vec{r}_j,t) + \vec{\nabla}_j\times\vec{A}(\vec{r}_j,t)\cdot\vec{s}_j] + \left(\frac{e}{4mc}\right)\sum_j [\vec{p}_j\cdot\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j + \vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j\cdot\vec{p}_j] + \left(\frac{eg_s}{4mc}\right)\times\sum_j [\vec{s}_j\cdot\vec{B}_{\rm ext}(\vec{r}_j,t) + \vec{B}_{\rm ext}(\vec{r}_j,t)\cdot\vec{s}_j]. \quad (10)$$

Our detailed expression for the single-electron canonical-momentum operators $\vec{p}_j$ is given by

$$\vec{p}_j = -i\hbar\vec{\nabla}_j = m\left(\frac{d\vec{r}_j}{dt}\right) - \left(\frac{e}{c}\right)\vec{A}(\vec{r}_j,t) - \left(\frac{e}{2c}\right)\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j. \quad (11)$$

The single-electron spin operators may be expressed in the form $\vec{s}_j = (\hbar/2)\vec{\sigma}_j$. The external magnetic field $\vec{B}_{\rm ext}(\vec{r},t)$ will be assumed to be spatially uniform on the scale of the atomic dimensions and stationary relative to the relevant interaction times. The external vector potential is then given by $A_{\rm ext}(\vec{r},t) = \frac{1}{2}B_{\rm ext}(\vec{r},t)\times\vec{r}$. The magnetic field associated with the electromagnetic vector potential $\vec{A}(\vec{r},t)$ is given by $\vec{B}(\vec{r},t) = \vec{\nabla}\times\vec{A}(\vec{r},t)$. The sums over j include all electrons (with charge $-e$) in the many-electron system. For long-wavelength (e.g., infrared) radiation, the contributions involving the nuclear canonical momentum and spin operators can play an important role. The nuclear contributions are beyond the scope of the present paper. The total quadratic (diamagnetic) contribution is given in the manifestly Hermitian form

$$V^2_{\rm AF}(t) = \left(\frac{e^2}{2mc^2}\right)\sum_j\left[\vec{A}(\vec{r}_j,t) + \frac{1}{2}\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j\right]\cdot\left[\vec{A}(\vec{r}_j,t) + \frac{1}{2}\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j\right]. \quad (12)$$

2. Perturbation operator for the electromagnetic interaction

The zeroth-order Hamiltonian operator H_0 will now be redefined to include the external-magnetic-field-dependent but electromagnetic-field-independent interactions, which are given by

$$\Delta H_0 = \left(\frac{e}{4mc}\right)[(\vec{L} + g_s\vec{S})\cdot\vec{B}_{\rm ext}(\vec{r},t) + \vec{B}_{\rm ext}(\vec{r},t)\cdot(\vec{L} + g_s\vec{S})] + \left(\frac{e^2}{8mc^2}\right)\sum_j [\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j]\cdot[\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j]. \quad (13)$$

The total orbital angular momentum operator is defined by $\vec{L} = \sum_j \vec{r}_j\times\vec{p}_j$ and the total spin angular momentum operator is defined by $\vec{S} = \sum_j \vec{s}_j$. The redefined zeroth-order atomic eigenstates must be determined by taking into account both the linear and the quadratic external-field interactions.

We can now introduce a more convenient perturbation expansion of the (reduced) density operator, in powers of the electromagnetic field, by redefining the linear and quadratic components of the relevant interaction (or perturbation) operator as follows:

$$V^1(t) = \left(\frac{e}{2mc}\right)\sum_j [\vec{p}_j\cdot\vec{A}(\vec{r}_j,t) + \vec{A}(\vec{r}_j,t)\cdot\vec{p}_j] + \left(\frac{eg_s}{4mc}\right)\sum_j [\vec{s}_j\cdot\vec{\nabla}_j\times\vec{A}(\vec{r}_j,t) + \vec{\nabla}_j\times\vec{A}(\vec{r}_j,t)\cdot\vec{s}_j] + \left(\frac{e^2}{4mc^2}\right)\sum_j [\vec{A}(\vec{r}_j,t)\cdot\vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j + \vec{B}_{\rm ext}(\vec{r}_j,t)\times\vec{r}_j\cdot\vec{A}(\vec{r}_j,t)] \quad (14)$$

and

$$V^2(t) = \left(\frac{e^2}{2mc^2}\right)\sum_j \vec{A}(\vec{r}_j,t)\cdot\vec{A}(\vec{r}_j,t). \quad (15)$$

Alternatively, H_0 can be redefined to include the intense-field electromagnetic interaction, in addition to the external-magnetic-field interaction. These alternatively redefined zeroth-order atomic eigenstates would then correspond to dressed-atom Zeeman states and the perturbation expansion would then involve powers of the weak electromagnetic field alone.

3. Hyperfine interaction

The hyperfine interaction, which can play an important role in high-resolution spectroscopic investigations, will be included in the zeroth-order Hamiltonian operator H_0, together with the external-magnetic-field interaction. For a self-consistent treatment, the nuclear-spin contributions should be included in the electromagnetic interaction. If the external magnetic field is neglected, the electromagnetic interaction (or perturbation) operator can be treated in a zeroth-order representation based on atomic eigenstates corresponding to the total (combined electronic and nuclear) angular momentum operator $\vec{F} = \vec{J} + \vec{I}$ and the total z component F_z. Only the total z component F_z remains conserved in the presence of an external magnetic field.

4. Electron-electron interactions

In accurate atomic-structure calculations, a correlated basis set is usually acquired, in which the zeroth-order many-electron atomic eigenstates are represented as superpositions of antisymmetrized products (Slater determinants) of single-electron atomic basis states. The component of the electrostatic electron-electron interaction operator that incorporates the longitudinal field pertaining to an isolated atomic system is then explicitly included in the zeroth-order Hamiltonian operator H_0. However, electrostatic interactions involving

different many-electron atomic systems of the ensemble are neglected. In Sec. VI, an equation for the longitudinal field associated with these electrostatic interactions will be included in the set of equations describing the dynamics of the electromagnetic-field propagation. The systematic and self-consistent incorporation of relativistic contributions to both the electron-electron and the electromagnetic interactions within the framework of a fully relativistic many-electron QED formulation is reserved for future investigation.

B. Perturbation expansion of the reduced density operator in the time domain

The traditional perturbation expansion of the (reduced) density operator has the form

$$\rho(\vec{r},\vec{v},t)=\sum_{n=0}^{\infty}\rho^{n}(\vec{r},\vec{v},t)=\rho^{0}(\vec{r},\vec{v},t)+\rho^{1}(\vec{r},\vec{v},t)+\cdots. \tag{16}$$

The dependence on the nth power of the classical electromagnetic field is contained in the component $\rho^{n}(\vec{r},\vec{v},t)$. This expansion is advantageous when only sufficiently weak electromagnetic fields are included in the perturbation operator. The initial-state (reduced) density operator $\rho^{0}(\vec{r},\vec{v},t)$ specifies the quantum-statistical state of the unperturbed many-electron quantum system, in the absence of the classical electromagnetic field that is included in the perturbation operator. As a first approximation, the initial-state (reduced) many-electron-system density operator may include a Maxwellian (thermodynamic-equilibrium) center-of-mass velocity distribution $\rho^{0}(\vec{v})$, which is independent of the position vector $\vec{r}$ as well as of the time t.

1. Time-domain hierarchy of coupled reduced-density-operator equations

An infinite set (hierarchy) of coupled integral-differential equations for the field components $\rho^{n}(\vec{r},\vec{v},t)$ can be obtained by substituting the perturbation expansion given by Eq. (16) for the (reduced) density operator $\rho(\vec{r},\vec{v},t)$ into our semiclassical equation of motion expressed by means of Eq. (8). The following detailed matrix-element form is thereby obtained for a general member of this set of coupled integral-differential equations:

$$\begin{aligned}&[\partial/\partial t+\vec{v}\cdot\vec{\nabla}+i(\omega_a-\omega_b)]\rho^{n}_{ab}(\vec{r},\vec{v},t)\\&\quad+i\sum_{c}\sum_{d}\int_{t_0}^{t}dt'\,\bar{\Sigma}_{ab,cd}(t,t')\rho^{n}_{cd}(\vec{r},\vec{v},t')\\&=\left(\frac{-i}{\hbar}\right)\sum_{c}\left[V^{1}_{ac}(t)\rho^{n-1}_{cb}(\vec{r},\vec{v},t)-\rho^{n-1}_{ac}(\vec{r},\vec{v},t)V^{1}_{cb}(t)\right.\\&\quad\left.+V^{2}_{ac}(t)\rho^{n-2}_{cb}(\vec{r},\vec{v},t)-\rho^{n-2}_{ac}(\vec{r},\vec{v},t)V^{2}_{cb}(t)\right].\end{aligned} \tag{17}$$

The lowercase latin letters a, b, c, and d are used to designate members of the complete set of zeroth-order many-electron-system eigenstates. The frequency variables corresponding to the eigenvalues of our redefined zeroth-order Hamiltonian operator H_0, which are given by $E_a=\hbar\omega_a$ and $E_b=\hbar\omega_b$, are understood to be functions of the external magnetic field and will also be functions of the intense (pump or control) electromagnetic field if this field is included in our alternatively redefined zeroth-order Hamiltonian operator H_0. A formal solution for the various field components $\rho^{n}(\vec{r},\vec{v},t)$ of the (reduced) many-electron-system density operator $\rho(\vec{r},\vec{v},t)$ can be obtained by means of a natural generalization of the procedure described by Mukamel [27] and by Tanaka *et al.* [28].

2. Alternative forms of the electromagnetic interaction

Our semiclassical perturbation-theory approach is related to that introduced by Bloembergen and Shen [29], who presented a pioneering investigation of linear and nonlinear optical phenomena based on the equivalent of a reduced-density-matrix formulation. However, they employed a phenomenological treatment of the rates occurring in the Markovian description of environmental decoherence and relaxation processes. In addition, they adopted an alternative form of the semiclassical electromagnetic interaction, which involves the electric and magnetic fields together with the various dipole and multipole moments. We provide a systematic microscopic description of the Liouville-space self-energy corrections. Furthermore, the multipole expansion is not required in our treatment of the electromagnetic interaction.

Horsley and Babiker [30] utilized a gauge transformation of the electromagnetic vector and scalar potentials to obtain relativistic corrections to the electromagnetic interaction that depend on the center-of-mass velocity. This alternative form of the electromagnetic interaction is expressed in terms of closed integral forms of the polarization and magnetization, representing the entire electric and magnetic multipole expansions to all orders.

C. Perturbation expansion of the reduced density operator in the continuous frequency and wave-vector representation

We obtain the perturbation expansion of the (reduced) density operator in the continuous frequency and wave-vector representation by introducing the Fourier transformation

$$\begin{aligned}&\rho^{n}(\vec{r},\vec{v},t)\\&\quad=\left(\frac{1}{2\pi}\right)^{4}\int d^{3}q\int d\omega\rho^{n}(\vec{q},\vec{v},\omega)\exp[i(\vec{q}\cdot\vec{r}-\omega t)].\end{aligned} \tag{18}$$

It should be noted that the frequency and the wave vector are treated as independent variables.

1. Hierarchy of coupled equations in the frequency and wave-vector representation

Our complimentary set (hierarchy) of coupled equations for the field components $\rho^{n}(\vec{q},\vec{v},\omega)$ of the (reduced) many-electron-system density operator, in the continuous frequency and wave-vector representation, is derived in the

detailed-matrix form

$$[-i\omega + i\vec{v}\cdot\vec{q} + i(\omega_a - \omega_b)]\rho^n_{ab}(\vec{q},\vec{v},\omega) + i\sum_c\sum_d \int d^3q' \int d\omega' \bar{\Sigma}_{ab,cd}(\vec{q},\omega;\vec{q}',\omega')\rho^n_{cd}(\vec{q}',\vec{v},\omega')$$
$$= \left[\frac{-i}{(2\pi)^4\hbar}\right]\sum_c \int d^3q' \int d\omega' \left[V^1_{ac}(\vec{q}',\omega')\rho^{n-1}_{cb}(\vec{q}-\vec{q}',\vec{v},\omega-\omega') - \rho^{n-1}_{ac}(\vec{q}-\vec{q}',\vec{v},\omega-\omega')V^1_{cb}(\vec{q}',\omega')\right]$$
$$+ \left[\frac{-i}{(2\pi)^4\hbar}\right]\sum_c \int d^3q' \int d\omega' \left[V^2_{ac}(\vec{q}',\omega')\rho^{n-2}_{cb}(\vec{q}-\vec{q}',\vec{v},\omega-\omega') - \rho^{n-2}_{ac}(\vec{q}-\vec{q}',\vec{v},\omega-\omega')V^2_{cb}(\vec{q}',\omega')\right]. \quad (19)$$

The Fourier transforms of the linear and quadratic field components of our relevant interaction operator are denoted by $V^1(\vec{q},\omega)$ and $V^2(\vec{q},\omega)$, respectively. The required tetradic-matrix elements of our Liouville-space self-energy operator kernel are expressed as follows:

$$\bar{\Sigma}_{ab,cd}(\vec{q},\omega;\vec{q}',\omega') = \left(\frac{1}{2\pi}\right)^4 \int d^3r \int_{-\infty}^{\infty} dt \int d^3r' \int_{t_0}^{t} dt' [e^{-i(\vec{q}'\cdot\vec{r}-\omega' t)}\bar{\Sigma}_{ab,cd}(\vec{r},t;\vec{r}',t')e^{+i(\vec{q}'\cdot\vec{r}'-\omega' t')}]e^{-i(\vec{q}-\vec{q}')\cdot\vec{r}}e^{i(\omega-\omega')t}$$
$$\rightarrow \bar{\Sigma}_{ab,cd}\delta(\vec{q}-\vec{q}')\delta(\omega-\omega'). \quad (20)$$

The arrow indicates the simplified result that we obtain for spatial uniformity and in the Markov approximation, based on Eq. (7). In the next section, we will employ the formal solution for the various field components of the (reduced) density operator, in the continuous frequency and wave-vector representation, which follows from the compact operator form of Eq. (19).

2. *Three-state (Λ-system) atomic-structure model*

Numerous experimental and theoretical investigations of EIT and related resonant pump-probe optical phenomena in a warm vapor of ^{87}Rb atoms have been based on the three-state (Λ-system) atomic-structure model, which is schematically illustrated in Fig. 1. The three atomic states that have been included are the $F = 1$ and 2 hyperfine-structure substates of the $5S_{1/2}$ ground state together with the $F' = 2$ hyperfine-structure substate of the $5P_{1/2}$ excited state. The intense pump electromagnetic field is chosen to be nearly resonant with the $F' = 2 \leftrightarrow F = 2$ hyperfine-structure component of the D_1 radiative transition, while the weak probe electromagnetic field is selected to be closely resonant with the $F' = 2 \leftrightarrow F = 1$ hyperfine-structure component of this radiative transition.

In the presence of an external magnetic field, the more detailed Zeeman-substate structure must be taken into account. In investigations of the influence of a homogeneous external magnetic field on EIT, the individual Zeeman-split components of the basic hyperfine-structure spectral feature have been observed [11,12]. If the external magnetic field is collinear with both the pump and probe electromagnetic fields and these electromagnetic fields have identical circular polarization, the selection rules pertaining to the magnetic quantum numbers indicate that three individual Zeeman-split spectral features can be observed [12,31]. For more general field orientations and polarizations, four Zeeman-split spectral features can be produced.

The observed Zeeman-split spectral features have been analyzed by adopting a description in terms of the relevant set of independent magnetic Λ subsystems. In this analysis, the initial magnetic substates, which may be designated by $\langle 5S_{1/2}, F = 1, M_F = 1,0,-1\rangle$, have been assumed to have equal populations and the (Zeeman) coherences pertaining to pairs of magnetic Λ subsystems have be neglected. In an analysis and optimization of a proposed channelization architecture involving an inhomogeneous external magnetic field [9], the ^{87}Rb atoms were assumed to have been initially prepared in one of the three magnetic substates, i.e., the state $\langle 5S_{1/2}, F = 1, M_F = 1\rangle$. In our general reduced-density-operator description, not only the entire manifold of the 13 relevant atomic magnetic substates, but also additional excited states from the complete basis set can be taken into account on an equal footing and the influence of the magnetic-substate (Zeeman) coherences can be systematically investigated.

IV. MACROSCOPIC ELECTROMAGNETIC RESPONSE

In the absence of free electrical charges, the macroscopic electromagnetic response can be evaluated in terms of the microscopic electronic current density. We initially employ the expansion of the (reduced) many-electron-system density operator in powers of the entire classical (combined pump and probe) electromagnetic field. The propagation dynamics of the probe field might be more advantageously investigated using an alternative expansion in powers of the weak probe field alone, together with an alternative zeroth-order representation corresponding to dressed states, which are to be determined by including in the zeroth-order relevant Hamiltonian operator the interactions with the intense pump electromagnetic field together with the external magnetic field. A fully quantum-mechanical evaluation of the macroscopic electromagnetic response, within the framework of an open-systems QED formulation, is left to future investigation.

A. Microscopic electronic current density

For a many-electron quantum system, the microscopic electronic current-density operator $\vec{j}(\vec{r},t)$ can be expressed

in the form [32,33]

$$\vec{j}(\vec{r},t) = -\left(\frac{e}{2}\right)\sum_j \left[\left(\frac{d\vec{r}_j}{dt} - \frac{ig_s}{m\hbar}\vec{p}_j \times \vec{s}_j\right)\delta^3(\vec{r}-\vec{r}_j) + \delta^3(\vec{r}-\vec{r}_j)\left(\frac{d\vec{r}_j}{dt} + \frac{ig_s}{m\hbar}\vec{p}_j \times \vec{s}_j\right)\right]. \quad (21)$$

The laboratory-frame position vectors $\vec{r}_j = \vec{R} + \vec{r}'_j$ depend on the relative position vectors $\vec{r}'_j$ and the center-of-mass position vector $\vec{R}$. The electromagnetic interaction can be expressed as

$$V(t) = -\left(\frac{1}{c}\right)\int d^3r\, \vec{j}(\vec{r},t)\cdot\vec{A}(\vec{r},t). \quad (22)$$

Using Eq. (11), the single-electron velocity operators $\vec{v}_j = d\vec{r}_j/dt$ can be expressed in terms of the single-electron canonical momentum operators $\vec{p}_j$, the single-electron spin operators $\vec{s}_j$, the electromagnetic vector potential $\vec{A}(\vec{r},t)$, and the external magnetic field $\vec{B}_{\rm ext}(\vec{r},t)$. The microscopic electronic current-density operator $\vec{j}(\vec{r},t)$ can then be written as follows:

$$\vec{j}(\vec{r},t) = \vec{j}^{\,0}(\vec{r},t) + \vec{j}^{\,1}(\vec{r},t), \quad (23)$$

where the electromagnetic-field-independent component is given by

$$\vec{j}^{\,0}(\vec{r},t) = \left(\frac{-e}{2m}\right)\sum_j \left[\vec{p}_j - \frac{ig_s}{\hbar}\vec{p}_j \times \vec{s}_j + \frac{e}{2c}\vec{B}_{\rm ext}(\vec{r},t)\times\vec{r}_j\right] \times \delta^3(\vec{r}-\vec{r}_j) + \left(\frac{-e}{2m}\right)\sum_j \delta^3(\vec{r}-\vec{r}_j) \times \left[\vec{p}_j + \frac{ig_s}{\hbar}\vec{p}_j \times \vec{s}_j + \frac{e}{2c}\vec{B}_{\rm ext}(\vec{r},t)\times\vec{r}_j\right] \quad (24)$$

and the electromagnetic-field-dependent component is given by

$$\vec{j}^{\,1}(\vec{r},t) = \left(\frac{-e^2}{2mc}\right)\sum_j [\vec{A}(\vec{r},t)\delta^3(\vec{r}-\vec{r}_j) + \delta^3(\vec{r}-\vec{r}_j)\vec{A}(\vec{r},t)] = \left(\frac{-e^2}{mc}\right)\vec{A}(\vec{r},t)n(\vec{r}). \quad (25)$$

The electronic number-density operator is denoted by $n(\vec{r})$. The electromagnetic interaction given by Eq. (22) is fully consistent with the predominately nonrelativistic Coulomb-gauge form for the electromagnetic interaction that we have introduced in the preceding section.

B. Macroscopic electronic current density

1. Space and time representation

In the space and time representation, the macroscopic electronic current density $\vec{J}(\vec{r},t)$ can be defined as a Liouville-space inner product (quantum-statistical average) involving the microscopic electronic current-density operator $\vec{j}(\vec{r},t)$ and the (reduced) many-electron-system density operator $\rho(\vec{r},\vec{v},t)$. The macroscopic electronic current density $\vec{J}(\vec{r},t)$ can then be obtained, as an expansion in powers of the entire classical electromagnetic field, as follows:

$$\vec{J}(\vec{r},t) = \langle\langle \vec{j}(\vec{r},t)|\rho(\vec{r},\vec{v},t)\rangle\rangle = \sum_{n=0}^{\infty}\langle\langle \vec{j}^{\,0}(\vec{r},t)|\rho^n(\vec{r},\vec{v},t)\rangle\rangle + \sum_{n=1}^{\infty}\langle\langle \vec{j}^{\,1}(\vec{r},t)|\rho^{n-1}(\vec{r},\vec{v},t)\rangle\rangle. \quad (26)$$

The average over the center-of-mass velocity distribution is taken into account in the quantum-statistical average (trace operation) represented by the Liouville-space inner product.

2. Frequency and wave-vector representation

The macroscopic electronic current density can be expressed, in the continuous frequency and wave-vector representation, as follows:

$$\vec{J}(\vec{q},\omega) = \sum_{n=0}^{\infty}\left(\frac{1}{2\pi}\right)^4\int d^3q' \times \int d\omega'\langle\langle \vec{j}^{\,0}(\vec{q}',\omega')|\rho^n(\vec{q}-\vec{q}',\vec{v},\omega-\omega')\rangle\rangle + \sum_{n=1}^{\infty}\left(\frac{1}{2\pi}\right)^4\int d^3q' \times \int d\omega'\langle\langle \vec{j}^{\,1}(\vec{q}',\omega')|\rho^{n-1}(\vec{q}-\vec{q}',\vec{v},\omega-\omega')\rangle\rangle. \quad (27)$$

C. Macroscopic electromagnetic-response tensors

The expansion for the macroscopic electronic current density can be obtained either in the space and time representation or in the frequency and wave-vector representation:

$$\vec{J}(\vec{r},t) = \sum_{n=0}^{\infty}\vec{J}^n(\vec{r},t) \quad \text{or} \quad \vec{J}(\vec{q},\omega) = \sum_{n=0}^{\infty}\vec{J}^n(\vec{q},\omega). \quad (28)$$

1. General nth-order macroscopic electromagnetic-response tensors

The general (arbitrary n) field component $\vec{J}^n(\vec{r},t)$ of the macroscopic electronic current density $\vec{J}(\vec{r},t)$ can be expressed in the form of a $4n$-dimensional integral relationship as follows:

$$\vec{J}^n(\vec{r},t) = \int d^3r_1\cdots\int d^3r_n\int dt_1\cdots \int dt_n\boldsymbol{\sigma}^n(\vec{r},\vec{r}_1,\ldots,\vec{r}_n;t,t_1,\ldots,t_n) \cdot\vec{E}(\vec{r}_1,t_1)\times\cdots\times\vec{E}(\vec{r}_n,t_n). \quad (29)$$

The general nth-order macroscopic electromagnetic response of the many-electron system (comprising the optical medium) to the classical electric field $\vec{E}(\vec{r},t) = -(1/c)(\partial/\partial t)\vec{A}(\vec{r},t)$, incorporating spatial nonlocality together with retardation, is thereby represented in terms of the $(n+1)$th-rank conductivity tensor $\boldsymbol{\sigma}^n(\vec{r},\vec{r}_1,\ldots,\vec{r}_n;t,t_1,\ldots,t_n)$. It is well known that the nonlinear conductivity tensor $\boldsymbol{\sigma}^n(\vec{r},\vec{r}_1,\ldots,\vec{r}_n;t,t_1,\ldots,t_n)$ is not uniquely defined, because the product of the n electric

fields can be rearranged according to $n!$ permutations. The arbitrariness can be removed by defining the $(n+1)$th-rank nonlinear conductivity tensor to include the symmetrization operation $(1/n!)\sum_P P$, where P indicates a permutation involving the n sets consisting of the space and time coordinates and the tensor indices [34], thereby incorporating the intrinsic permutation symmetry. In the quantized-field formulation, this symmetrization operation cannot be introduced because the n electric fields would then be represented by noncommuting operators.

The $4(n+1)$-dimensional Fourier transformation of the relationship given by Eq. (29), incorporating both spatial and temporal dispersion, can be expressed in the general form

$$\vec{J}^n(\vec{q},\omega) = \int d^3q_1 \cdots \int d^3q_n \int d\omega_1 \cdots \int d\omega_n \boldsymbol{\sigma}^n(\vec{q},\vec{q}_1,\ldots,\vec{q}_n;\omega,\omega_1,\ldots,\omega_n)\cdot\vec{E}(\vec{q}_1,\omega_1)\times\cdots\times\vec{E}(\vec{q}_n,\omega_n). \quad (30)$$

The $(n+1)$th-rank conductivity tensor $\boldsymbol{\sigma}^n(\vec{q},\vec{q}_1,\ldots,\vec{q}_n;\omega,\omega_1,\ldots,\omega_n)$, characterizing the nth-order macroscopic electromagnetic response in the continuous frequency and wave-vector representation, is most generally defined by means of the relationship

$$\boldsymbol{\sigma}^n(\vec{q},\vec{q}_1,\ldots,\vec{q}_n;\omega,\omega_1,\ldots,\omega_n) = \left(\frac{1}{2\pi}\right)^{4n}\int d^3r\int d^3r_1\cdots\int d^3r_n\int dt\int dt_1\cdots\int dt_n$$
$$\times\exp[-i(\vec{q}\cdot\vec{r}-\omega t)]\boldsymbol{\sigma}^n(\vec{r},\vec{r}_1,\ldots,\vec{r}_n;t,t_1,\ldots,t_n)\exp\left[i\sum_{j=1}^{n}(\vec{q}_j\cdot\vec{r}_j-\omega_j t_j)\right]. \quad (31)$$

For single field modes, the general definition for the nth-order contribution to the macroscopic electronic current density given by Eq. (30) can be reduced to a simple algebraic form. If in addition the magnetization, which arises from the individual electron orbital and spin angular momenta, is neglected, the analogous relationship expressing the nth-order macroscopic polarization in terms of the nth-order electrical susceptibility tensor and the product of the n electric fields becomes equivalent to the nth-order macroscopic current-density relationship. For a spatially uniform and stationary optical medium, the nth-order electrical susceptibility tensor can be simply related to the nth-order conductivity tensor in the following manner:

$$\boldsymbol{\chi}^n(\vec{q}_1,\ldots,\vec{q}_n;\omega_1,\ldots,\omega_n) = \left(\frac{i}{\Omega_n}\right)\boldsymbol{\sigma}^n(\vec{q}_1,\ldots,\vec{q}_n;\omega_1,\ldots,\omega_n). \quad (32)$$

We emphasize that the orbital-angular-momentum-dependent and lowest-order spin-dependent contributions, giving rise to the magnetization, are automatically taken into account in our definition of the microscopic current-density operator. Consequently, our formulation of the macroscopic electromagnetic response should be equivalent to the more general form of the alternative description, which would include the nth-order macroscopic magnetization, in terms of the nth-order magnetic susceptibility tensor and the product of the n magnetic fields, in addition to the macroscopic polarization relationship.

2. *Self-consistent description of the macroscopic electromagnetic response*

As discussed in Sec. VI, the macroscopic electromagnetic response can be self-consistently described [33] by introducing our perturbation expansion for the macroscopic electronic current density as a source term into the Maxwell field equations, as illustrated in Fig. 4. The local-field corrections, which are almost always treated using empirical models [34], can be systematically incorporated by including the equations governing any longitudinal electric fields that have not been taken into account by the inclusion of the corresponding electrostatic interactions in the zeroth-order Hamiltonian operator representing the many-electron quantum system, e.g., the electrostatic interactions involving different many-electron atoms.

The set of Maxwell field equations thus obtained governs the dynamics of the mean or macroscopic electromagnetic fields. These fields can be defined by invoking the ergodic hypothesis, i.e., that the time average of the rapidly fluctuating microscopic electromagnetic fields is equivalent to the average involving the degrees of freedom of the quantum-statistical ensemble corresponding to the optical medium. In some approaches, an average of the electromagnetic fields within a small volume is introduced. Since a premature spatial average could interfere with the precise description of nonlocal

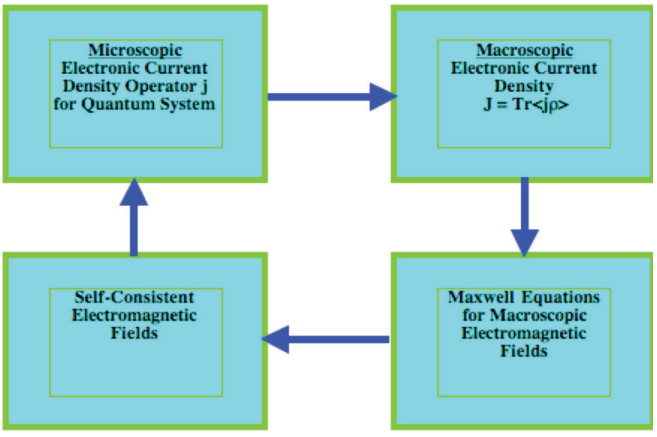

FIG. 4. (Color online) In the self-consistent semiclassical description of the electromagnetic interaction, the macroscopic electronic current density $\vec{J}(\vec{r},t)$, defined in terms of the microscopic electronic current-density operator $\vec{j}(\vec{r},t)$ and the (reduced) many-electron-system density operator $\rho(\vec{r},\vec{v},t)$, is introduced as a current-source term into the Maxwell field equations.

correlations and short-wavelength transitions, the macroscopic electromagnetic fields introduced in our investigation should be understood to be defined only with respect to the statistical-ensemble average.

For ultrafast electromagnetic interactions, the assumption of ergodic behavior may not be valid and it may be necessary to describe the electromagnetic interactions entirely in terms of the microscopic electromagnetic fields. In our formulation, this can be simply accomplished by redefining the trace operation to include only the quantum-mechanical expectation value.

D. Compact expressions for the macroscopic electromagnetic-response tensors

The nth-order field components of the macroscopic electronic current density can be evaluated in the continuous frequency and wave-vector representation by introducing the field components of the (reduced) many-electron-system density operator that are obtained from the formal solution of the set (hierarchy) of coupled relationships given by Eq. (19). The conductivity tensors can then be expressed in terms of the Doppler-shifted Liouville-space resolvent operator $\bar{G}(\omega-\vec{v}\cdot\vec{q})=(\omega-\vec{v}\cdot\vec{q}-\bar{L}_0-\bar{\bar{\Sigma}})^{-1}$. The Liouvillian operator $\bar{L}_0$ corresponds to the field-free many-electron-system Hamiltonian operator augmented with the external-field-dependent contributions and $\bar{\bar{\Sigma}}$ denotes the Liouville-space self-energy operator obtained in the Markov approximation or by introducing a model of non-Markovian behavior for which the required Fourier transformation can be analytically performed. Compact expressions for the macroscopic electromagnetic-response tensors can also be obtained in the space and time representation from the formal solution of the set of coupled relationships given by Eq. (17).

1. Linear ($n=1$) conductivity tensor

After evaluating the linear contribution $\vec{J}^1(\vec{q},\omega)$ to the macroscopic electronic current density using $\rho^1(\vec{q},\vec{v},\omega)=\bar{G}(\omega-\vec{v}\cdot\vec{q})\bar{V}^1(\vec{q},\omega)\rho^0(\vec{v})$ and $\rho^0(\vec{q},\vec{v},\omega)=(2\pi)^4\delta^3(\vec{q})\delta(\omega)\rho^0(\vec{v})$, the linear ($n=1$) conductivity tensor is obtained in the generalized Kubo [35] form

$$\begin{aligned}\sigma^1(\vec{q},q_1;\omega,\omega_1)=&\left(\frac{i}{\omega_1}\right)\left(\frac{1}{2\pi}\right)^5\int d^3q_1'\int d\omega_1'\delta^3(\vec{q}-\vec{q}_1'-\vec{q}_1)\\&\times\langle\langle\vec{j}^0(\vec{q}_1',\omega_1')|\bar{G}(\omega-\omega_1'-\vec{v}\cdot(\vec{q}-\vec{q}_1'))\bar{\bar{j}}^0(-\vec{q}_1,\omega-\omega_1'-\omega_1)|\rho^0(\vec{v})\rangle\rangle\\&+i\left(\frac{1}{2\pi}\right)^3\frac{\delta(\omega-\omega_1)}{\omega_1}\mathbf{1}\left(\frac{e^2}{m}\right)\langle\langle n(\vec{q}-\vec{q}_1)|\rho^0(\vec{v})\rangle\rangle.\end{aligned}\tag{33}$$

The symbol $\bar{\bar{j}}$ denotes the Liouville-space microscopic many-electron current-density operator defined by the commutator relationship $\bar{\bar{j}}\rho=(1/\hbar)[\vec{j},\rho]$ and $\mathbf{1}$ denotes the rank-2 unity tensor. The field-independent microscopic electronic current-density operator $\vec{j}^0(\vec{q},\omega)$ is obtained as follows:

$$\begin{aligned}\vec{j}^0(\vec{q},\omega)=2\pi\delta(\omega)\vec{j}^0(\vec{q})=\frac{-e}{2m}(2\pi)\delta(\omega)\sum_j\Bigg\{&\left(\vec{p}_j-\frac{ig_s}{\hbar}\vec{p}_j\times\vec{s}_j+\frac{e}{2c}\vec{B}_{\rm ext}\times\vec{r}_j\right)\exp[-i(\vec{q}\cdot\vec{r}_j)]\\&+\exp[-i(\vec{q}\cdot\vec{r}_j)]\left(\vec{p}_j+\frac{ig_s}{\hbar}\vec{p}_j\times\vec{s}_j+\frac{e}{2c}\vec{B}_{\rm ext}\times\vec{r}_j\right)\Bigg\}.\end{aligned}\tag{34}$$

2. General nonlinear ($n>1$) conductivity tensors

In our evaluation of the general nth-order contribution to the macroscopic many-electron-system current density, we directly obtain a compact Liouville-space operator expression for the component that does not involve the quadratic-field term in the semiclassical electromagnetic interaction. The remaining components, which involve the quadratic-field interaction, are expressed in terms of contractions among the electric-field vectors and therefore correspond to lower-rank-tensor contributions to the nth-order macroscopic electromagnetic response. After introducing compact Liouville-space operator expressions for the field components of the (reduced) many-electron-system density operator, we derive for tensor providing the highest-rank contribution the following symmetrized analytical relationship:

$$\begin{aligned}&\boldsymbol{\sigma}^{n,n+1}(\vec{q},\vec{q}_1,\ldots,\vec{q}_n;\omega,\omega_1,\ldots,\omega_n)\\&=(i)^n\left(\frac{1}{2\pi}\right)^{5n}\prod_{m=1}^n\left\{\int d^3q_m'\int d\omega_m'\right\}\left(\frac{1}{n!}\right)\\&\times\sum_P P\left\langle\langle\vec{j}^0(\vec{q}_1',\omega_1')|\prod_{m=1}^n\frac{\bar{G}((\omega-\Omega_m')-\vec{v}\cdot(\vec{q}-\vec{Q}_m'))}{\omega_m}\delta^3(\vec{q}-\vec{Q}_m'-\vec{q}_m)\bar{\bar{j}}^0(-\vec{q}_m,\omega-\Omega_m'-\omega_m)|\rho^0(\vec{v})\rangle\right\rangle.\end{aligned}\tag{35}$$

The symbols $\vec{Q}'_m$ and Ω'_m denote the sums, from $j = 1$ to $j = m$, of $\vec{q}'_j$ and ω'_j, respectively.

For the $n = 2$ and 3 nonlinear conductivity tensors, our compact Liouville-space operator expressions have forms analogous to those obtained in the space and time representation by Tanaka *et al.* [28]. In addition to the generalization of their results for arbitrary n, we have also included a lowest-order spin-dependent component of the microscopic electronic current-density operator and have taken into account the full tetradic-matrix form of the Liouville-space self-energy operator in our definition of the Doppler-shifted Liouville-space resolvent operator. Our results for the linear and the general (arbitrary n) nonlinear conductivity tensors can be applied for a spatially nonuniform and nonstationary optical medium under arbitrary (possibly coherent) initial excitation conditions.

3. The n = 1 and 2 conductivity tensors including initial atomic-state coherences

The coherent excitation of a specific manifold of many-electron quantum states can be described by including appropriate nondiagonal matrix elements in the eigenstate decomposition of the initial-state many-electron-system density operator given by Eq. (4). The linear conductivity tensor expressed by Eq. (33) gives the most general form of the lowest-order electromagnetic response for a spatially and temporally dispersive medium. For the analysis of EIT and related pump-probe processes, it is necessary to consider the nonlinear electromagnetic response. The contributions to the linear and the $n = 2$ nonlinear conductivity tensors that do not involve the quadratic-field interaction can be expressed, in terms of the tetradic matrix elements of the Doppler-shifted Liouville-space resolvent operator $\bar{G}(\omega - \vec{v}\cdot\vec{q})$, in the forms

$$\boldsymbol{\sigma}^{1,2}(\vec{q},\vec{q}_1;\omega,\omega_1) = \delta(\omega-\omega_1)\left(\frac{i}{\hbar\omega}\right)\left(\frac{1}{2\pi}\right)^3 \sum_a\sum_{a'}\sum_b\sum_{b'}\sum_c\sum_{c'} \langle c'|\,\vec{j}^{\,0}(\vec{q}-\vec{q}_1)|c\rangle$$
$$\times \langle\langle cc'|\bar{G}(\omega_1-\vec{v}\cdot\vec{q}_1)|bb'\rangle\rangle[\langle b|\vec{j}^{\,0}(-\vec{q}_1)|a\rangle\delta_{b'a'} - \delta_{ba}\langle a'|\vec{j}^{\,0}(-\vec{q}_1)|b'\rangle]\langle a|\rho^0(\vec{v})|a'\rangle \tag{36}$$

and

$$\boldsymbol{\sigma}^{2,3}(\vec{q},\vec{q}_1,\vec{q}_2;\omega,\omega_1,\omega_2) = \frac{\delta(\omega-\Omega_2)}{\omega_1\omega_2}(-i)^2\left(\frac{1}{2\pi}\right)^7\left(\frac{1}{2!}\right)\sum_P P\sum_a\sum_{a'}\sum_b\sum_{b'}\sum_c\sum_{c'}\sum_d\sum_{d'}\sum_e\sum_{e'}$$
$$\times \langle e'|\vec{j}^{\,0}(\vec{q}-\vec{q}_1)|e\rangle\langle\langle ee'|\bar{G}(\omega_1-\vec{v}\cdot\vec{q}_1)|dd'\rangle\rangle\langle\langle dd'|\vec{\bar{j}}^{\,0}(-\vec{q}_1)|cc'\rangle\rangle$$
$$\times \langle\langle cc'|\bar{G}(\omega_2-\vec{v}\cdot\vec{q}_2)|bb'\rangle\rangle\langle\langle bb'|\vec{\bar{j}}^{\,0}(-\vec{q}_2)|aa'\rangle\rangle\langle a|\rho^0(\vec{v})|a'\rangle. \tag{37}$$

The averages over the center-of-mass velocity distribution must be performed to obtain the final results. If the initial quantum-state coherences are neglected, the initial-state many-electron-system density operator can be represented entirely in terms of its diagonal matrix elements, which can be obtained from a simpler set of rate equations governing the population densities.

If the external-magnetic-field interaction is neglected, the electromagnetic-field-independent microscopic electronic current-density operator given by Eq. (34) can be expressed in the more familiar electric dipole form, which has been frequently employed in the nonlinear optics literature [36–38]. After making the electric dipole approximation, which corresponds to $\vec{q} = 0$, neglecting the spin-dependent contributions, and employing the commutator relationship $\vec{p}_j = (im/\hbar)[H_0,\vec{r}_j]$, the current-density operator $\vec{j}^{\,0}(\vec{q})$ can be expressed in terms of the single-electron position operators $\vec{r}_j$. We emphasize that the commutator relationship is rigorously valid only when the various matrix elements are evaluated using exact eigenstates of the zeroth-order many-electron-system Hamiltonian operator H_0.

Electromagnetically induced transparency and related pump-probe optical phenomena have often been investigated by evaluating the linear polarization within the framework of a dressed-atom representation [7], which has been determined by including the electromagnetic interaction involving the intense pump field in the zeroth-order Hamiltonian operator, together with the external-magnetic-field interaction. This alternative dressed-atom approach can be adopted by taking our states a, b, c, d, etc., to be these field-dependent states. The coherent excitation of the Zeeman states produced by the intense pump field is automatically incorporated by this nonperturbative solution of the equation of motion for the many-electron-system density operator. Consequently, the linear macroscopic electromagnetic response can be expressed entirely in terms of the diagonal matrix elements of the many-electron-system density operator in a dressed-atom representation.

4. Ensemble of noninteracting many-electron atomic systems

The states a, b, c, d, etc., and the microscopic electronic current-density operators $\vec{j}^{\,0}(\vec{q},\omega)$ and $\vec{j}^{\,1}(\vec{r},t)$ pertain to the entire many-electron quantum system comprising the optical medium. The optical medium will now be assumed to be composed of an ensemble of N identical many-electron atoms. If interactions among different many-electron atoms are neglected, the density operator representing the initial quantum-statistical state of the entire optical medium can be expressed in the tensor-product form $\rho^0(\vec{v}) = \otimes_{j=1}^N \rho_j^0(\vec{v})$, where $\rho_j^0(\vec{v})$ denotes to the initial-state density operator describing a single many-electron atom. The electronic eigenstates of the entire quantum system corresponding to the

optical medium can then be expressed as tensor products of the N eigenstates, each of which describes a single many-electron atomic system. Our general expression for the nth-order macroscopic electromagnetic response can then be reduced to a simpler result involving the initial-state density operator describing a single many-electron atomic system and the matrix elements of the microscopic electronic current-density operators can be reduced to the corresponding matrix elements for a single many-electron atomic system. The final result for an ensemble of N identical many-electron atoms is obtained by simply multiplying the contribution of each of the identical many-electron atomic systems by their number density in the optical medium. In order to systematically incorporate the interactions among various clusters of coherently excited many-electron atomic systems, a hierarchical reduced-density-operator formulation should be introduced in a future extension of this investigation. This extension will be necessary for the description of cooperative electromagnetic phenomena such as superradiance.

V. EVALUATION OF THE SELF-ENERGY CONTRIBUTIONS FOR ATOMIC COLLISIONAL AND RADIATIVE INTERACTIONS

The entire set of tetradic matrix elements of the Liouville-space self-energy operator $\bar{\Sigma}(z)$, which occurs in our expression for the Doppler-shifted Liouville-space resolvent operator, can be evaluated using the general definition given by Eq. (3), which involves the total Liouville-space interaction operator $\bar{V}$, the Liouville-space environmental (irrelevant) interaction operator $\bar{V}^{\rm ir}$, and the Zwanzig Liouville-space projection operators $\bar{P} = |\rho^E\rangle\rangle\langle\langle I^E|$ and $\bar{Q} = 1 - \bar{P}$.

In our semiclassical description of the electromagnetic interaction, the radiative components of the total Liouville-space interaction operator $\bar{V}$ that are associated with the classical electromagnetic fields should not be included. The self-energy corrections that are induced by the classical electromagnetic fields, together with the local-field corrections arising from any additional electrostatic interactions that are not included in the zeroth-order Hamiltonian operator, are systematically incorporated when our expansion for the macroscopic electronic current density (and perhaps also the electronic charge density) in powers of the classical electromagnetic fields is introduced as a source term into the Maxwell field equations, as discussed in the following section. However, this semiclassical treatment of the radiative self-energy corrections, which would be obtained at the level of a mean field, is not expected to be rigorously correct. In a future investigation, the semiclassical description should be replaced by a quantized-field formulation.

A. Explicit expressions for the Zwanzig Liouville-space projection operators

The zeroth-order states of the combined quantum system, consisting of the relevant many-electron system and the environment, can be represented by the tensor-product states $|\alpha\rangle = |a,\{n_i\},\vec{p}\rangle = |a\rangle \otimes |\{n_i\}\rangle \otimes |\vec{p}\rangle$. The quantum states of the many-electron system, taking into account the external-magnetic-field interaction and perhaps also the intense-pump electromagnetic-field interaction, will be denoted by lowercase latin letters, e.g., a, b, c, and d. The set of occupation numbers corresponding to the environmental photon modes will be denoted by $\{n_i\}$. Finally, the quantum state pertaining to the projectile atomic system in an atom-atom collision process, for which the internal atomic degrees of freedom will be neglected, will be characterized by the relative momentum variable $\vec{p}$. The Zwanzig Liouville-space projection operator $\bar{P} = |\rho^E\rangle\rangle\langle\langle I^E|$ can be explicitly expressed using the following expansions:

$$|\rho^E\rangle\rangle = \sum_{\{n_i\}}\sum_{\vec{p}} |\{n_i\}\{n_i\},\vec{p}\,\vec{p}\rangle\rangle\langle\langle\{n_i\}\{n_i\}|\rho^R\rangle\rangle\langle\langle\vec{p}\,\vec{p}||\rho^C\rangle\rangle, \tag{38}$$

$$|I^E\rangle\rangle = \sum_{\{n_i\}}\sum_{\vec{p}} |\{n_i\}\{n_i\},\vec{p}\,\vec{p}\rangle\rangle. \tag{39}$$

The environmental density operator has been expressed in the tensor-product form $\rho^E = \rho^R \otimes \rho^C$, in terms of the density operators for the radiation field R and the projectile atomic system C. The environmental photons can be represented by the diagonal density-matrix elements corresponding to a Planck (thermal-equilibrium) distribution. The projectile atomic systems can be represented by a Maxwellian (thermal-equilibrium) distribution corresponding to the relative momentum variable and only binary collisions with the target many-electron atomic system will be taken into account. The nondiagonal density-matrix elements will be assumed to vanish.

The most unambiguous treatment of the atomic collision processes involves those pertaining to different (buffer-gas) projectile atomic systems, which are assumed to not resonantly interact with the electromagnetic fields. These (buffer-gas) projectile atomic systems can be most consistently considered as a component of the environment. Collision processes involving the atomic systems comprising the EIT medium can also be treated in terms of environmental interactions, provided the electrostatic interactions among different atomic systems are not included in the zeroth-order Hamiltonian operator. In this case, the internal degrees of freedom for both atomic systems should be taken into account.

B. Liouville-space linewidth and line-shift operators

It is traditional to introduce the Liouville-space relaxation operator $\bar{R}(z)$ as follows:

$$\begin{aligned}\bar{R}(z) &= \bar{V} + \bar{V}\bar{Q}\frac{1}{z - \bar{Q}\bar{L}\bar{Q}}\bar{Q}\bar{V} \\ &= \bar{V} + \bar{V}\bar{Q}\frac{1}{z - \bar{Q}\bar{L}_0\bar{Q}}\bar{Q}\bar{R}(z).\end{aligned} \tag{40}$$

The second of the two equivalent forms is conveniently expressed in terms of the zeroth-order Liouvillian operator $\bar{L}_0$ for the combined system, without the total Liouville-space interaction operator $\bar{V}$. Since we will include all quantized electromagnetic-field modes in the Liouville-space environmental (irrelevant) interaction operator $\bar{V}^{\rm ir}$ and omit radiative self-energy corrections that are induced by the classical electromagnetic fields, the distinction between Liouville-space environmental (irrelevant) interaction operator $\bar{V}^{\rm ir}$ and the

total Liouville-space interaction operator $\bar{V}$ can be ignored. Consequently, the Liouville-space self-energy operator $\bar{\Sigma}(z)$ can be expressed in terms of the Liouville-space relaxation operator $\bar{R}(z)$ as follows:

$$\bar{\Sigma}(z) = \bar{P}\bar{R}(z)\bar{P} = \mathrm{Tr}_E[\bar{R}(z)\rho^E]. \tag{41}$$

Since the number of the relevant degrees of freedom is very small in comparison with the total number of degrees of freedom, we can make the approximation $\bar{Q} = 1 - \bar{P} = 1$.

In the spectral description of the emission or absorption of electromagnetic radiation that is composed of a set of spectral lines, which are associated with transitions between two groups of many-electron quantum states $\{a\}$ and $\{b\}$, it will be convenient to express the Liouville-space self-energy operator $\bar{\Sigma}(z)$, in terms of the Liouville-space shift and width operators $\bar{\Delta}(x)$ and $\bar{\Gamma}(x)$, by employing the following relationship [39,40]:

$$\lim_{\eta\to 0}\bar{\Sigma}(x \pm i\eta) = \bar{\Delta}(x) \mp i\frac{\bar{\Gamma}(x)}{2}. \tag{42}$$

In the diagonal-resolvent approximation, which is frequently adopted in detailed spectral simulations, the diagonal matrix elements of the Liouville-space operator $\bar{\Delta}(x)$ are the line shifts associated with the individual transitions $a \to b$ and the diagonal matrix elements of the Liouville-space operator $\bar{\Gamma}(x)$ can be interpreted as the full widths at the half maxima.

The lowest-order nonvanishing contribution to the Liouville-space linewidth operator can be expressed as follows:

$$\bar{\Gamma}(x) = 2\pi\,\bar{P}\bar{V}\delta(x - \bar{L}_0)\bar{V}\bar{P} = 2\pi\,\mathrm{Tr}_E[\bar{V}\delta(x - \bar{L}_0)\bar{V}\rho^E]. \tag{43}$$

The Liouville-space line-shift operator can be evaluated as follows:

$$\bar{\Delta}(x) = \mathrm{Re}(\bar{V}) + \frac{\mathcal{P}}{2\pi}\int_{-\infty}^{\infty}\frac{dx'\bar{\Gamma}(x')}{x - x'}, \tag{44}$$

where Re denotes the real part and $\mathcal{P}$ indicates the Cauchy principal value.

C. Tetradic-matrix elements of the lowest-order nonvanishing contribution to the Liouville-space linewidth operator in the diagonal-resolvent approximation

In the diagonal-resolvent approximation, the tetradic matrix elements of the lowest-order nonvanishing contribution to the Liouville-space linewidth operator can be simply expressed as sums of the partial contributions that are associated with the environmental radiative (R) and collisional (C) interactions $\langle\langle ab\,|\,\bar{\Gamma}(x)\,|\,ab\rangle\rangle = \langle\langle ab\,|\,\bar{\Gamma}^R(x)\,|\,ab\rangle\rangle + \langle\langle ab\,|\,\bar{\Gamma}^C(x)\,|\,ab\rangle\rangle$.

The lowest-order radiative linewidth can be expressed as the sums of the separate components corresponding to the spontaneous (SR) and the induced (IR) radiation processes: $\langle\langle ab\,|\,\bar{\Gamma}^R(x)\,|\,ab\rangle\rangle = \langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{SR}}(x)\,|\,ab\rangle\rangle + \langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{IR}}(x)\,|\,ab\rangle\rangle$. We have evaluated the component from single-environmental-photon spontaneous emission processes to obtain the result

$$\begin{aligned}&\langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{SR}}(x)\,|\,ab\rangle\rangle\\ &= \left(\frac{2\pi}{\hbar^2}\right)\sum_{a''}\sum_{\vec{k}}\sum_{\lambda}|V_{aa''}(\vec{k}\lambda)|^2\delta(x - \omega_{a''} + \omega_b - \omega) + \left(\frac{2\pi}{\hbar^2}\right)\sum_{b''}\sum_{\vec{k}}\sum_{\lambda}|V_{bb''}(\vec{k}\lambda)|^2\delta(x - \omega_a + \omega_{b''} + \omega).\end{aligned} \tag{45}$$

The quantities $V_{aa''}(\vec{k}\lambda)$ are the linear-interaction matrix elements in Eq. (19). The component from single-environmental-photon absorption and induced emission processes is evaluated as

$$\begin{aligned}&\langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{IR}}(x)\,|\,ab\rangle\rangle\\ &= \left(\frac{2\pi}{\hbar^2}\right)\sum_{a''}\sum_{\vec{k}}\sum_{\lambda}|V_{aa''}(\vec{k}\lambda)|^2\delta(x - \omega_{a''} + \omega_b - \omega)\langle n_{\vec{k}\lambda}\rangle + \left(\frac{2\pi}{\hbar^2}\right)\sum_{a''}\sum_{\vec{k}}\sum_{\lambda}|V_{aa''}(\vec{k}\lambda)|^2\delta(x - \omega_{a''} + \omega_b + \omega)\langle n_{\vec{k}\lambda}\rangle\\ &+ \left(\frac{2\pi}{\hbar^2}\right)\sum_{b''}\sum_{\vec{k}}\sum_{\lambda}|V_{bb''}(\vec{k}\lambda)|^2\delta(x - \omega_a + \omega_{b''} + \omega)\langle n_{\vec{k}\lambda}\rangle + \left(\frac{2\pi}{\hbar^2}\right)\sum_{b''}\sum_{\vec{k}}\sum_{\lambda}|V_{bb''}(\vec{k}\lambda)|^2\delta(x - \omega_a + \omega_{b''} - \omega)\langle n_{\vec{k}\lambda}\rangle.\end{aligned} \tag{46}$$

The average environmental-photon occupation number denoted by $\langle n_{\vec{k}\lambda}\rangle$ can be related to the specific intensity $I(\vec{k},\lambda)$ corresponding to a Planck (thermal-equilibrium) distribution function.

The lowest-order partial contribution to the linewidth from the collisional (C) interactions can be expressed as the sums of the separate components associated with inelastic collisional transitions and elastic collisions involving the relevant (target) atomic system: $\langle\langle ab\,|\,\bar{\Gamma}^C(x)\,|\,ab\rangle\rangle = \langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{IC}}(x)\,|\,ab\rangle\rangle + \langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{EC}}(x)\,|\,ab\rangle\rangle$. We have evaluated the lowest-order component associated with inelastic collisional (IC) transitions to obtain the result

$$\begin{aligned}\langle\langle ab\,|\,\bar{\Gamma}^{\mathrm{IC}}(x)\,|\,ab\rangle\rangle &= \left(\frac{2\pi}{\hbar^2}\right)\sum_{a''}\sum_{\vec{p}}\sum_{\vec{p}''}\left|V^C_{a\vec{p},a''\vec{p}''}\right|^2\delta(x - \omega_{a''} + \omega_b - \omega_{\vec{p}''} + \omega_{\vec{p}})\rho_{\vec{p}}\\ &+ \left(\frac{2\pi}{\hbar^2}\right)\sum_{b''}\sum_{\vec{p}}\sum_{\vec{p}''}\left|V^C_{b\vec{p},b''\vec{p}''}\right|^2\delta(x - \omega_a + \omega_{b''} - \omega_{\vec{p}} + \omega_{\vec{p}})\rho_{\vec{p}}.\end{aligned} \tag{47}$$

The matrix elements $V^C_{a\vec{p},a''\vec{p}''}$ and $V^C_{b\vec{p},b''\vec{p}''}$ are to be evaluated in terms of the collisional interaction operator V^C, which is primarily the Coulomb interaction between the relevant (target) atomic system and the projectile atomic system. Note that the average over the initial relative momentum distribution is expressed in terms of the density operator $\rho_{\vec{p}}$, which can be represented by a Maxwellian (thermal-equilibrium) distribution function. The lowest-order component associated with elastic collisions (EC) is evaluated as follows:

$$\langle\langle ab\,|\,\bar{\Gamma}^{\rm EC}(x)\,|\,ab\rangle\rangle = \left(\frac{2\pi}{\hbar^2}\right)\sum_{\vec{p}}\sum_{\vec{p}'}\left|V^C_{a\vec{p},a\vec{p}'} - V^C_{b\vec{p},b\vec{p}'}\right|^2 \times \delta(x-\omega_a+\omega_b-\omega_{\vec{p}}+\omega_{\vec{p}'})\rho_{\vec{p}}. \quad (48)$$

The collisional contributions can also be expressed in terms of the matrix elements of the scattering operator S or in terms of the scattering cross sections and scattering amplitudes.

The total linewidth in the Markov (short-memory-time) approximation is obtained by setting $x = 0$. Our general expression for the Liouville-space self-energy operator $\bar{\Sigma}(z)$ can be evaluated beyond the lowest-order, diagonal-resolvent, and Markov approximations.

D. Tetradic-matrix elements of the lowest-order nonvanishing contribution to the Liouville-space line-shift operator in the diagonal-resolvent approximation

The individual lowest-order radiative and collisional contributions to the line shifts in the diagonal-resolvent and Markov approximations can be determined from Eq. (44) using the results obtained above for the individual lowest-order radiative and collisional contributions to the linewidths. Beyond the diagonal-resolvent approximation, the spectral line shapes must be described by an evaluation of the full set of tetradic matrix elements of the self-energy operator.

VI. ELECTROMAGNETIC-FIELD PROPAGATION IN THE SEMICLASSICAL DESCRIPTION

A wide variety of modified light-propagation characteristics, including slow light, fast light, left-handed light (associated with a negative value of the index of refraction), and stopped (or stored) light [41], can be systematically investigated by incorporating our analytical results for the linear and nonlinear macroscopic electromagnetic response as current-source terms in the macroscopic Maxwell field equations of classical electrodynamics [42]. The alternative microscopic Maxwell equations can be adopted simply by redefining the average (trace operation) that is associated with the Liouville-space inner product to correspond to the quantum-mechanical average alone, without reference to the quantum-statistical distribution. In a fully quantum-mechanical (QED) formulation, the equations of motion for the electromagnetic-field operators in the Heisenberg representation can serve as a starting point for an investigation of the propagation characteristics of the average electromagnetic fields. The quantized-electromagnetic-field formulation will be necessary for a fully consistent description of stopped (or stored) light, which is accomplished by the regulated transfer of the probe electromagnetic field into the coherently excited many-electron quantum system, giving rise to quasiparticle states of the light-matter system that have become know as dark-state polaritons [7,41].

A. Electromagnetic-field propagation in the space and time representation

In the Coulomb gauge, the inhomogeneous wave equation governing the electromagnetic vector potential $\vec{A}(\vec{r},t)$ can be expressed in the form

$$\nabla^2\vec{A}(\vec{r},t) - \left(\frac{1}{c^2}\right)\frac{\partial^2}{\partial t^2}\vec{A}(\vec{r},t) = -\left(\frac{4\pi}{c}\right)\vec{J}(\vec{r},t) + \left(\frac{1}{c}\right)\vec{\nabla}\frac{\partial}{\partial t}\Phi(\vec{r},t). \quad (49)$$

We have expressed the macroscopic electronic current density $\vec{J}(\vec{r},t)$ as a perturbation expansion in powers of either the entire classical electromagnetic field or the weak probe field alone.

We have emphasized that the semiclassical description of the electromagnetic interaction is most unambiguously implemented when the entire electrostatic interaction has been included in the zeroth-order Hamiltonian operator describing the many-electron quantum system corresponding to the optical medium. In this case, the scalar potential $\Phi(\vec{r},t)$ may be set equal to zero in the absence of free charges. However, we have pointed out that the electrostatic interactions among different atomic systems are not taken into account in the traditional atomic-structure approximation involving noninteracting atomic systems. In this case, the scalar potential should be determined by including the Poisson equation

$$\nabla^2\Phi(\vec{r},t) = -4\pi\rho(\vec{r},t). \quad (50)$$

In order to self-consistently incorporate these electrostatic interactions, it may be necessary to introduce a perturbation expansion for the macroscopic charge density $\rho(\vec{r},t)$ and also to take explicitly into account the Coulomb interaction operator $V_c(t) = \int d^3r\rho(\vec{r},t)\Phi(\vec{r},t)$ together with the relevant electromagnetic-interaction operator. The electric field will then be given, in terms of the transverse and longitudinal components, by $\vec{E}(\vec{r},t) = -(\frac{1}{c})\frac{\partial}{\partial t}\vec{A}(\vec{r},t) - \vec{\nabla}\Phi(\vec{r},t)$ and Eq. (49) will involve a coupling between these components.

For a complete analysis of the propagation characteristics of the electromagnetic field in the optical medium, it may be necessary to consider the differential continuity equations corresponding to the conservation of energy, linear momentum, and angular momentum in the interacting system of charged particles and electromagnetic fields. These differential conservation relationships are expressed in terms of the energy densities, the energy fluxes, the linear momentum and angular momentum densities, and the energy momentum and angular momentum tensors. Moreover, these quantities may be more conveniently defined in terms of the electric and magnetic fields rather than the electromagnetic vector and scalar potentials. The inhomogeneous wave equations can be reexpressed in terms of the electric and magnetic fields and also in terms of the polarization $\vec{P}(\vec{r},t)$ and the magnetization $\vec{M}(\vec{r},t)$ by introducing the relationships $\vec{J}(\vec{r},t) = \frac{\partial}{\partial t}\vec{P}(\vec{r},t) + c\vec{\nabla}\times\vec{M}(\vec{r},t)$ and $\rho(\vec{r},t) = -\vec{\nabla}\cdot\vec{P}(\vec{r},t)$, which are valid in the

absence of free charges and net currents passing through any cross-section area. However, it is well known that partition of the electronic current density is not unique.

The macroscopic formulation of the conservation relationships can be expressed in terms of the perturbation expansions for the macroscopic charge density $\rho(\vec{r},t)$ and the macroscopic electronic current density $\vec{J}(\vec{r},t)$. We have adopted a description of the semiclassical electromagnetic interaction based on perturbation expansions in terms of the combined (pump and probe) electromagnetic field. However, we have also pointed out that it might be advantages to include the electromagnetic interaction involving the intense pump field in the zeroth-order Hamiltonian operator, thereby providing a dressed-state representation corresponding to a nonperturbative and effectively nonlinear treatment of both the intense pump field and the external magnetic field. The perturbation expansions of the charge and current density operators would then be expressed in terms of the weak probe (or signal) field alone, while the macroscopic electromagnetic-response tensors occurring in these expansions will be implicit functions of the intense pump (or control) field and the external magnetic field.

B. Electromagnetic-field propagation in the frequency and wave-vector representation

After introducing the Fourier transformations and setting the scalar potential equal to zero, the wave equation for the electromagnetic vector potential can be expressed in the form

$$q^2\vec{A}(\vec{q},\omega)-\left(\frac{\omega^2}{c^2}\right)\vec{A}(\vec{q},\omega)=\left(\frac{4\pi}{c}\right)\vec{J}(\vec{q},\omega)$$
$$=\left(\frac{4\pi}{c}\right)\sum_{n=0}^{\infty}\vec{J}^n(\vec{q},\omega)=\left(\frac{4\pi}{c}\right)\sum_{n=0}^{\infty}\int d^3q_1\cdots\int d^3q_n\int d\omega_1\cdots$$
$$\int d\omega_n\boldsymbol{\sigma}^n(\vec{q},\vec{q}_1,\ldots,\vec{q}_n;\omega,\omega_1,\ldots,\omega_n)\cdot\vec{E}(\vec{q}_1,\omega_1)\times\cdots\times\vec{E}(\vec{q}_n,\omega_n). \quad (51)$$

The linear ($n=1$) and nonlinear ($n>1$) components of the macroscopic electronic current density $\vec{J}(\vec{q},\omega)$ can be analytically expressed using the compact Liouville-space operator forms that we have obtained for the corresponding conductivity tensors. The wave equation for the electromagnetic vector potential $\vec{A}(\vec{q},\omega)$ is a multiple-integral relationship. If the perturbation expansion for the macroscopic electronic current density is truncated at some order n, this wave equation can be solved and the dispersion relation giving the frequency ω as a function of the wave vector $\vec{q}$ can be obtained. Starting with a finite set of frequency and wave-vector variables, a set of coupled wave equations will be obtained governing the propagation of the fields associated with the various combinations of these frequency and wave-vector variables, which are generated as a result of the linear and nonlinear components of the macroscopic electronic current density. Since the solution of the general nonlinear and nonlocal form of the set of coupled wave equations is not expected to be obtainable in an analytical form, a numerical stimulation will be necessary. If we retain only the linear contribution and introduce the simple algebraic relationship pertaining to a single-mode electromagnetic field, the dispersion relation for a spatially uniform and stationary optical medium can be expressed in the familiar forms

$$\omega^2=\frac{(cq)^2}{1+4\pi i\sigma^1(\vec{q},\omega)/\omega}\rightarrow\frac{(cq)^2}{1+4\pi\chi^1(\vec{q},\omega)}=\frac{(cq)^2}{n(\vec{q},\omega)^2}. \quad (52)$$

The arrow indicates the simplified result that can be obtained by neglecting the magnetization and the index of refraction is denoted by $n(\vec{q},\omega)$.

In Eq. (51), the entire electrostatic interaction is assumed to be included in the zeroth-order Hamiltonian operator, so the scalar potential can be omitted. In order to systematically take into account any electrostatic interactions that are not included in the zeroth-order Hamiltonian operator, particularly those among different atomic systems of the optical medium, it will be necessary to develop a more general macroscopic formulation, in which separate perturbation expansions would be introduced for the charge and current densities and the scalar potential would be self-consistently determined, together with the electromagnetic vector potential.

The propagation characteristics of electromagnetic fields in pump-probe optical phenomena are usually determined by an analysis of the dispersion relation together with the various conservation relationships. In applications to EIT and related pump-probe optical phenomena, the primary propagation characteristic of interest is the group velocity $v_g=d\omega/dq$. As discussed in Sec. IV, these resonant nonlinear optical phenomena have been analyzed by evaluating the linear polarization within the nonperturbative and effectively nonlinear framework of a dressed-atom representation, by including the electromagnetic interaction involving the intense pump field in the zeroth-order Hamiltonian operator, together with the external-magnetic-field interaction. Consequently, the group velocity pertaining to the propagation of the weak probe electromagnetic field can be determined by introducing the dressed-atom linear susceptibility into Eq. (52). The dependence of the group velocity on the intense probe field provides the control mechanism for the slow and stopped light-propagation phenomena.

VII. CONCLUSION

A reduced-density-matrix description has been developed for linear and nonlinear (possibly coherent)

electromagnetic interactions of moving many-electron atomic systems. Time-domain (equation-of-motion) and frequency-domain (resolvent-operator) formulations have been developed in a unified and self-consistent manner. Atomic collisional and environmental radiative processes have been treated within the framework of a quantum-open-system approach and external magnetic fields have been taken into account together with the electromagnetic fields. The influence of the environment has thereby been described in terms of nonequilibrium kinetics (decoherence and relaxation) processes together with spectral line broadening mechanisms. We have emphasized that our general, nonperturbative, non-Markovian, and full tetradic-matrix formulations provide a rigorous foundation for the systematic introduction of the Born (lowest-order), Markov (short-memory-time), and diagonal-resolvent approximations.

A preliminary semiclassical description of the electromagnetic interaction has been adopted, based on a perturbation expansion of the electronic-system density operator in powers of either the combined (pump and probe) classical electromagnetic field or the probe field alone. Time-domain and frequency-domain sets (hierarchies) of coupled relationships have been obtained for the field components of this density operator. We have indicated how a self-consistent treatment of the electrostatic interaction among different atomic systems of the optical medium can be provided. We have emphasized that relativistic contributions to the electron-electron and electromagnetic interactions can be most systematically treated within the framework of a fully relativistic quantum-electrodynamics formulation, which would also lead to a completely consistent quantum-mechanical treatment of local-field and radiative corrections.

The tetradic-matrix elements of the time-domain and frequency-domain Liouville-space self-energy operators can be systematically evaluated, taking into account the dominant environmental interactions. In our semiclassical description, the subspace of the relevant projection operator has been defined as the subspace of either the zeroth-order many-electron-system eigenstates or the dressed many-electron-system eigenstates, taking into account the intense pump field. Furthermore, the Liouville-space self-energy corrections that we have taken into account describe the interaction of the quantized electronic system with the environment, unperturbed by the classical electromagnetic field. Consideration of the field-dependent modifications of the self-energy corrections is left to a future extension of this investigation.

In our semiclassical description of the electromagnetic interaction, the macroscopic electronic current density has been obtained as an expansion in powers of the classical electromagnetic field. The general form of the nth-order macroscopic electromagnetic response can be expressed in terms of the $(n+1)$th-rank conductivity tensor in either the space-time representation (applicable to a nonlocal and nonstationary medium) or the frequency and wave-vector representation (describing spatial and temporal dispersion).

We have emphasized that, for a self-consistent semiclassical description of the electromagnetic interaction, the expansion obtained for the macroscopic electronic current density should be introduced as a current-source term into the Maxwell field equations. A systematic treatment can thereby be provided for the induced-field and local-field corrections that are often discussed in the nonlinear-optics literature. The Maxwell field equations thus obtained govern the dynamics of the mean or macroscopic electromagnetic field. The field-induced and local-field corrections can be incorporated when the expansion of the macroscopic electronic current density in powers of the classical electromagnetic field is introduced into the Maxwell field equations. However, we have pointed out that this treatment of field-induced corrections, which is obtained at the level of a mean field, is not expected to be rigorously correct. Consequently, the semiclassical description should eventually be replaced by a fully quantum-mechanical description based on an open-system version of quantum electrodynamics. In the fully quantum-mechanical description, the Maxwell field equations would be replaced by the Heisenberg-Langevin equations of motion for the electromagnetic-field operators. This would provide a fundamental quantum-mechanical framework for a unified treatment of nonlinear electromagnetic interactions and quantum optical processes.

Using our preliminary semiclassical description of the electromagnetic interaction, compact analytical Liouville-space operator expressions have been derived for the linear and the general (nth-order) nonlinear macroscopic electromagnetic-response tensors. In these expressions, we have allowed for coherent initial electronic excitations and for the full tetradic-matrix form of the Liouville-space self-energy operator representing the environmental interactions either in the Markov approximation or with a model for non-Markovian behavior. We have pointed out that the compact analytical expression that we have obtained for the macroscopic electromagnetic-response tensors can be employed for the microscopic electromagnetic response simply by redefining the Liouville-space inner product to include the quantum-mechanical expectation value alone, without the quantum-statistical average.

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