Gauge invariance of phenomenological models of the interaction of quantum dissipative systems with electromagnetic fields

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We discuss specific features of the electrodynamic characteristics of quantum systems within the framework of models that include a phenomenological description of the relaxation processes. As is shown by W. E. Lamb, Jr., R. R. Schlicher, and M. O. Scully [Phys. Rev. A **36**, 2763 (1987)], the use of phenomenological relaxation operators, which adequately describe the attenuation of eigenvibrations of a quantum system, may lead to incorrect solutions in the presence of external electromagnetic fields determined by the vector potential for different resonance processes. This incorrectness can be eliminated by giving a gauge-invariant form to the relaxation operator. Lamb, Jr., *et al.* proposed the corresponding gauge-invariant modification for the Weisskopf-Wigner relaxation operator, which is introduced directly into the Schrödinger equation within the framework of the two-level approximation. In the present paper, this problem is studied for the von Neumann equation supplemented by a relaxation operator. First, we show that the solution of the equation for the density matrix with the relaxation operator correctly obtained "from the first principles" has properties that ensure gauge invariance for the observables. Second, we propose a common recipe for transformation of the phenomenological relaxation operator into the correct (gauge-invariant) form in the density-matrix equations for a multilevel system. Also, we discuss the methods of elimination of other inaccuracies (not related to the gauge-invariance problem) which arise if the electrodynamic response of a dissipative quantum system is calculated within the framework of simplified relaxation models (first of all, the model corresponding to constant relaxation rates of coherences in quantum transitions). Examples illustrating the correctness of the results obtained within the framework of the proposed methods in contrast to inaccuracy of the results of the standard calculation techniques are given.

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I. INTRODUCTION

A known example of the problem of gauge invariance in the medium-radiation interaction is the multiyear discussion related to the comparison of the results obtained within the framework of different expressions for the electron-electromagnetic wave interaction operator, namely, for $-\boldsymbol{dE}$ or $-e\boldsymbol{A}\hat{\boldsymbol{p}}/mc$ (here, $\boldsymbol{d}$ is the dipole moment, $\hat{\boldsymbol{p}}=-i\hbar\boldsymbol{\nabla}$ is the momentum operator, $\boldsymbol{A}$ is the vector potential, $\boldsymbol{E}(t)=-\dot{\boldsymbol{A}}/c$ is the corresponding homogeneous electric field, and $e<0$ is the electron charge). See, e.g., [1] and references therein (Ref. [2] in particular) and [3].

For problems admitting the first-principles-based formulation, the "apparent" nonequivalence of the results, which appears sometimes in different approaches, is certainly only a matter of correctness of the approximations or interpretations of calculation results. This conclusion is evident by virtue of the gauge invariance of the initial equations of the theory [4]. As concerns the phenomenological models, the problem of gauge invariance of the results obtained with their use is much more significant since, essentially, it is a matter of correctness of the phenomenological model itself.

An important result in this relation was obtained in [2], where the resonant interaction of the two-level transition with a homogeneous rf field was discussed within the framework of the Weisskopf-Wigner approximation, where the model relaxation operator is introduced directly in the Schrödinger equation. It is shown in [2] that the probability of transition remains unchanged with the gauge transformation from the electrostatic potential $\varphi=-\boldsymbol{rE}(t)$ to the vector potential $\boldsymbol{A}=-\frac{1}{c}\int_{t_0}^{t}\boldsymbol{E}(\tau)d\tau$ if the Weisskopf-Wigner relaxation operator is subjected to a certain unitary transformation.

Note that within the framework of the classical description, the friction force $\boldsymbol{F}_{fr}$, which is a phenomenological relaxation term, can certainly be introduced in the equation of motion,

$$\dot{\boldsymbol{r}}=\frac{\partial H}{\partial \boldsymbol{p}}=\frac{\boldsymbol{p}}{m}-\frac{e\boldsymbol{A}}{mc},\quad \dot{\boldsymbol{p}}=-\frac{\partial H}{\partial \boldsymbol{r}}+\boldsymbol{F}_{fr},$$

where $H(\boldsymbol{r},\boldsymbol{p})$ is the classical Hamiltonian. In this case, the gauge-invariant form for the relaxation term is obtained by itself under the assumption that such a force depends on the particle velocity, i.e., $\boldsymbol{F}_{fr}(\dot{\boldsymbol{r}})\Rightarrow\boldsymbol{F}_{fr}(\frac{\boldsymbol{p}}{m}-\frac{e\boldsymbol{A}}{mc})$. The objective of this paper is seeking an appropriate recipe for the quantum system, whose interaction with some dissipative reservoir is described by a method which currently is recognized as most effective for analyzing a wide scope of problems of radiation interaction with dissipative quantum media (see, e.g., [1,5,6]). We mean the kinetic equation, i.e., the von Neumann equation for the density matrix with relaxation operator $\hat{R}$ on the right-hand side,

$$i\hbar\dot{\rho}-[\hat{H},\rho]=\hat{R}\rho. \tag{1}$$

In principle, Eq. (1) can be obtained by averaging the exact equation for the density matrix of two interacting sub-

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systems over the coordinates of one of them, namely, the so-called reservoir. However, this inevitably leads to simplifications, the main of which is the assumption that the interaction of the subsystems is weak (see, e.g., [1,5,6]). Thus, by virtue of the approximate nature of Eq. (1), the gauge invariance itself of the initial relationships of quantum mechanics does not guarantee, in general, the "automatic" gauge invariance when Eq. (1) is used. So much this is true for a phenomenological determination of the relaxation operator.

Certainly, Eq. (1)—although it is very popular—still is not the only known form of describing relaxation processes in quantum systems. Significant progress in the theory of quantum dissipative systems was reached, in particular, by using the Heisenberg-Langevin method (see, e.g., [1] or [7]) and the Lagrangian or spatiotemporal approach to quantum mechanics[1] [8], which was developed by Feynman. Nevertheless, the problem of loss of gauge invariance during idealizations or in a phenomenological description of relaxation processes, which we discuss in our paper, has not been explored within the framework of these methods.

In some papers, their authors developed a description of dissipation in quantum systems by using a certain modification of the standard Hamiltonian of a conservative system (instead of adding a relaxation operator into the standard Schrödinger or von Neumann equations) (see, e.g., [9,10] and references therein). Obviously, there are problems with the validation of such techniques. A discussion of these issues can be found in, e.g., [9,10]. One variant of such a "modified" Hamiltonian was used in [9] to demonstrate the capabilities of quantization in extended phase space [11] by gauge-invariant calculation of the conductivity of an ensemble of noninteracting quantum particles in a homogeneous electric field. In this relation, we should also mention some papers devoted to analogies between conservative and dissipative systems. In [12], it is shown that the gauge-invariant Lagrangian, which models the interaction between an electromagnetic field and a complex scalar field in the classical limit, corresponds to the bilinear Lagrangian for two uncoupled classical linear dissipative oscillators with the positive and negative dampings, respectively. In [13], it is shown that the quantization procedure proposed by G. 't Hooft [14] led to identical expressions for a damped harmonic oscillator and some deterministic quantum systems. Thus, the results obtained in the above-mentioned papers do not remove the posed problem of seeking a common recipe for the transformation of the phenomenological relaxation operator into the correct (gauge-invariant) form for the density-matrix equation of a multilevel system.

The paper is organized as follows. In Sec. II we discuss the basic initial relationships. In Sec. III we establish the gauge-invariant form of the relaxation operator. Also, the results obtained by other authors (in particular, in [2,9]) in the solution of similar problems are discussed here in more detail than in the introduction. In Sec. IV we show that the relaxation operator correctly obtained "from the first principles" ensures gauge invariance when the observables are determined by Eq. (1). The reason for the possible "loss" of gauge invariance is the use of simple models of a relaxation operator which is either independent of the external field or allows for the external-field effect within the framework of a simplified gauge-noninvariant model. In Sec. V we propose a phenomenological recipe for the transformation of the relaxation operator obtained without allowance for the external-field effect into the gauge-invariant form. Section VI and VII are devoted to examples illustrating the correctness of the physical results obtained by using the relaxation operator transformed into the gauge-invariant form. In particular, we obtained a gauge-invariant equation for the current excited in a three-dimensional harmonic quantum dissipative oscillator affected by a nonstationary electric field in the presence of a homogeneous magnetic field and established a fairly universal relationship between the Hermitian and non-Hermitian components of the dielectric permittivity tensor of an ensemble of such oscillators. We also obtained a fairly general gauge-invariant expression for the response of a multilevel dissipative system to the action of an electric field with arbitrary spatiotemporal dependence (in the linear approximation). In Sec. VIII we discuss the demands imposed on the "two-level" model by the gauge invariance requirement.

II. INITIAL RELATIONSHIPS

Consider the motion of a "no-spin" particle with charge e in the force field specified by the potential $U(\boldsymbol{r})$ and "external" electromagnetic fields specified by the vector potential $\boldsymbol{A}(\boldsymbol{r},t)$ and the scalar potential $\varphi(\boldsymbol{r},t)$. The Hamiltonian of the considered system is determined by the standard expression [4]

$$\hat{H} = \hat{H}_0 + \hat{h}, \quad \hat{H}_0 = \frac{\hat{\boldsymbol{p}}^2}{2m} + U(\boldsymbol{r}), \tag{2a}$$

$$\hat{h} = -\frac{e}{2mc}[\hat{\boldsymbol{p}}\boldsymbol{A}(\boldsymbol{r},t) + \boldsymbol{A}(\boldsymbol{r},t)\hat{\boldsymbol{p}}] + \frac{e^2}{2mc^2}\boldsymbol{A}^2(\boldsymbol{r},t) + e\varphi(\boldsymbol{r},t),$$

(hereafter, we use hats to denote the operators of only the physical quantities whose action in the coordinate space is not reduced to simple multiplication by the coordinate function). Besides the dipole-moment operator $\boldsymbol{d}=e\boldsymbol{r}$, we will use the current operator[2] [4]

$$\hat{\boldsymbol{j}} = \frac{e}{m}\left(\hat{\boldsymbol{p}} - \frac{e\boldsymbol{A}}{c}\right), \tag{2b}$$

where $\hat{\boldsymbol{p}}/m - e\boldsymbol{A}/mc = \hat{\boldsymbol{v}}$ and $\hat{\boldsymbol{v}}$ is the velocity operator.

Along with the often-used representation of the density matrix ρ_{mn} in the basis of eigenfunctions $\psi_k(\boldsymbol{r})$ of the unperturbed energy operator $\hat{H}_0$ (usually, $\psi_k E_k = \hat{H}_0\psi_k$, where E_k are the eigenvalues of the energy operator $\hat{H}_0$), we also employ the coordinate representation $\rho(\boldsymbol{r},\boldsymbol{r}')$ since many of the

[1] Note that in [8] it is directly indicated that within the framework of the Hamiltonian formulation of quantum mechanics, "…many results may be obtained more simply than by the Lagrangian method…"

[2] Namely, the values of the dipole moment and/or current are the key values for most of the material-radiation interaction problems.

identities used in this paper are most easily proved, namely, in this representation. Correspondingly, for the commutator of the Hamiltonian and the density matrix $[\hat{H},\rho]$ in Eq. (1) we have [4]

$$[\hat{H},\rho]=[\hat{H}(\boldsymbol{r})-\hat{H}^{*}(\boldsymbol{r}')]\rho(\boldsymbol{r},\boldsymbol{r}')\rightarrow H_{mj}\rho_{jn}-\rho_{mj}H_{jn}, \tag{3a}$$

where $H_{mn}=\delta_{mn}E_m+h_{mn}$. The subscripts $\perp$ and $\|$ denote the transverse and longitudinal components of the density matrix formed by its off-diagonal and diagonal elements, respectively,

$$\rho=\rho_{\perp}+\rho_{\|},\quad \rho_{\perp}=\sum_{m\neq n}\rho_{mn}\psi_m(\boldsymbol{r})\psi_n^{*}(\boldsymbol{r}'),$$

$$\rho_{\|}=\sum_{m=n}\rho_{mn}\psi_m(\boldsymbol{r})\psi_n^{*}(\boldsymbol{r}').$$

We will use an overbar to denote the average values of the physical quantities and the corresponding averaging operation determined in a standard way for any operator $\hat{\boldsymbol{g}}$,

$$\bar{\boldsymbol{g}}=\overline{\hat{\boldsymbol{g}}\rho}=\int_{\infty}[\hat{\boldsymbol{g}}(\boldsymbol{r})\rho(\boldsymbol{r},\boldsymbol{r}')]_{\boldsymbol{r}'=\boldsymbol{r}}d^3r=\boldsymbol{g}_{nm}\rho_{mn}, \tag{3b}$$

[it is assumed in Eqs. (3a) and (3b) and elsewhere below that the summation, as usual, is performed over recurrent indices and the cases in which this operation is absent are obvious from the context]. In particular, we assume that the dipole-moment operator is off diagonal, i.e., $\boldsymbol{d}_{nn}=0$, such that $\overline{\boldsymbol{d}\rho_{\|}}=0$.

Differentiating the expression for the average value $\bar{\boldsymbol{g}}$ with respect to time and using Eq. (1) together with the identity $\hat{\boldsymbol{g}}[\hat{H},\rho]=-(\hat{H}\hat{\boldsymbol{g}}-\hat{\boldsymbol{g}}\hat{H})\rho$, we obtain an expression for the operator $\dot{\hat{\boldsymbol{g}}}$ of the rate of variation in the physical quantity,

$$\dot{\hat{\boldsymbol{g}}}=\frac{\partial}{\partial t}\hat{\boldsymbol{g}}+\frac{i}{\hbar}(\hat{H}\hat{\boldsymbol{g}}-\hat{\boldsymbol{g}}\hat{H})-\frac{i}{\hbar}\hat{\boldsymbol{g}}\hat{R}, \tag{4a}$$

where the operator $\hat{\boldsymbol{g}}$ is determined by the "natural" relationship

$$\dot{\bar{\boldsymbol{g}}}=\overline{\dot{\hat{\boldsymbol{g}}}\rho}. \tag{4b}$$

It is seen from Eq. (4a) that in the case where the dissipative system is described by Eq. (1), the standard (see [4]) expression for the operator of the derivative $\dot{\hat{\boldsymbol{g}}}=\dot{\hat{\boldsymbol{g}}}+i(\hat{H}\hat{\boldsymbol{g}}-\hat{\boldsymbol{g}}\hat{H})/\hbar$ is adequate only under the condition $\hat{\boldsymbol{g}}\hat{R}\rho=0$, but this condition is not always fulfilled. The appearance of an "addition" in the expressions for the momentum and energy evolution operators is objectively due to processes in which physical quantities are lost during interaction with the reservoir. At the same time, the additional term in the equation for the operator of the dipole-moment derivative

$$\dot{\hat{\boldsymbol{d}}}=\frac{e}{m}\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)-\frac{i}{\hbar}\boldsymbol{d}\hat{R} \tag{5}$$

appeared (under the condition $\overline{\boldsymbol{d}\hat{R}\rho}\neq 0$) due to the approximate description of a quantum system within the framework of Eq. (1). In relation with this, consider a simple example which is important for the further analysis, namely, the case where relaxation of the off-diagonal elements of the density matrix is determined by the often-used (see [1,5,6]) relationship

$$(\hat{R}\rho_{\perp})_{kn}=-i\hbar\gamma_{kn}\rho_{kn}, \tag{6a}$$

where $k\neq n$ and γ_{kn} are the inverse times of "transverse" relaxation for the corresponding transitions. In the absence of external fields, we obtain the following equation for the off-diagonal elements of the density matrix:

$$\dot{\rho}_{kn}+(i\Omega_{kn}+\gamma_{kn})\rho_{kn}=0, \tag{6b}$$

where $\Omega_{kn}=(E_k-E_n)/\hbar$ are the transition frequencies. Solution of Eq. (6b) has the form $\rho_{kn}=\rho_{0kn}e^{-(i\Omega_{kn}+\gamma_{kn})t}$, $\rho_{0kn}=$const. For the dipole moment, we obtain the expression

$$\bar{\boldsymbol{d}}(t)=\boldsymbol{d}_{kn}\rho_{0nk}e^{(i\Omega_{kn}-\gamma_{kn})t}, \tag{6c}$$

[it was taken into account in Eq. (6c) that $\Omega_{kn}=-\Omega_{nk}$ and $\gamma_{kn}=\gamma_{nk}$]. Differentiating Eq. (6c) with respect to time, we obtain an expression corresponding to the particular case of Eq. (5),

$$\dot{\bar{\boldsymbol{d}}}\rightarrow(\dot{\boldsymbol{d}})_{kn}=i\Omega_{kn}\boldsymbol{d}_{kn}-\gamma_{kn}\boldsymbol{d}_{kn}=e\boldsymbol{v}_{kn}-\gamma_{kn}\boldsymbol{d}_{kn}. \tag{6d}$$

This expression comprises the relaxation parameter γ_{mn} in explicit form. The last term in Eq. (6d) is a consequence of the relaxation model which is described by Eq. (6a) and corresponds to the "death" (occurring with frequency γ_{kn}) of a particle with nonzero dipole moment followed by an instantaneous "birth" of a particle with zero dipole moment. The first particle is in the so-called "coherent" state, in which $\rho_{k\neq n}\neq 0$, and the second particle corresponds to the state with $\rho_{k\neq n}=0$. In other words, such a relaxation model corresponds to a "leak" of the dipole moment through the reservoir due to diffusion or other relaxation processes "hidden" within the framework of such a description.

The inequality $(\dot{\boldsymbol{d}})_{kn}\neq e\boldsymbol{v}_{kn}$, which follows from Eq. (6d), leads to an ambiguity when the electric current of the system in the Maxwell equation is determined.[3] For monochromatic

[3]Note that this ambiguity is not related to the particle-number conservation law since the conditions $\overline{\boldsymbol{d}\hat{R}\rho}\neq 0$ and $\overline{\hat{R}\rho}=0$ can be fulfilled together, which is exactly the case that corresponds to relaxation operator (6a).

rf fields[4] $\boldsymbol{E}=\mathrm{Re}\,\tilde{\boldsymbol{E}}e^{-i\omega t}$ with a sufficiently high frequency ω, the above-mentioned ambiguity obviously does not affect the Hermitian component of the complex electric polarizability tensor of the system. However, the choice of a variant, as is shown in Sec. V, can appear to be of fundamental importance for the correct calculation of the anti-Hermitian polarizability component.

In order to find the expression for the total-current operator, we make use of Eq. (4) for the kinematic-momentum operator $m\hat{\boldsymbol{v}}=\hat{\boldsymbol{p}}-e\boldsymbol{A}/\boldsymbol{c}$ and the energy operator

$$\hat{W}=\frac{m\hat{\boldsymbol{v}}^2}{2}+U(\boldsymbol{r}),$$

[the operator $\hat{W}$ in the case $\varphi(\boldsymbol{r},t)\neq 0$ is not identical to the Hamiltonian $\hat{H}$]. As a result, we obtain

$$m\frac{\partial}{\partial t}\overline{\hat{\boldsymbol{v}}\rho}+\overline{\boldsymbol{\nabla}U\rho}-\overline{\hat{\boldsymbol{F}}_{fr}\rho}=e\overline{\boldsymbol{E}\rho}+\frac{e}{2c}\overline{(\hat{\boldsymbol{v}}\times\boldsymbol{B}-\boldsymbol{B}\times\hat{\boldsymbol{v}})\rho},\tag{7a}$$

$$\frac{\partial}{\partial t}\overline{\hat{W}\rho}+\overline{\hat{G}\rho}=\frac{e}{2}\overline{(\hat{\boldsymbol{v}}\boldsymbol{E}+\boldsymbol{E}\hat{\boldsymbol{v}})\rho},$$

where $\boldsymbol{B}=\mathrm{rot}\,\boldsymbol{A}$ and $\boldsymbol{E}=-\boldsymbol{\nabla}\varphi-\frac{1}{c}\frac{\partial\boldsymbol{A}}{\partial t}$ are the magnetic and electric fields, respectively, and $-\hat{\boldsymbol{F}}_{fr}=\frac{i}{\hbar}m\hat{\boldsymbol{v}}\hat{R}$ and $\hat{G}=\frac{i}{\hbar}\hat{W}\hat{R}$ are the loss operators due to the outflow of the momentum and energy of the system into the dissipative reservoir. We then make use of the energy and momentum conservation law for the volume V, into which the considered quantum system is placed,

$$m\frac{\partial}{\partial t}(\overline{\hat{\boldsymbol{v}}\rho})+\frac{\partial}{\partial t}\int_V\mathbf{P}dV-\overline{\hat{\boldsymbol{F}}_{fr}\rho}=\oint_S[(\hat{\sigma}+\hat{T})\cdot\boldsymbol{s}]dS,$$

$$\frac{\partial}{\partial t}(\overline{\hat{W}\rho})+\frac{\partial}{\partial t}\int_V W_{E,B}dV+\overline{\hat{G}\rho}=-\oint_S(\boldsymbol{\Pi}\cdot\boldsymbol{s})dS,\tag{7b}$$

where $W_{E,B}=\frac{E^2+B^2}{8\pi}$ is the energy density of the electromagnetic field, $\boldsymbol{\Pi}=\frac{c}{4\pi}(\boldsymbol{E}\times\boldsymbol{B})$ is the Poynting vector, $\mathbf{P}=\frac{\boldsymbol{\Pi}}{c^2}$ is the vector of the field momentum flow density, $\hat{\sigma}\equiv\sigma_{ij}=\frac{1}{4\pi}[E_iE_j+B_iB_j-1/2\,\delta_{ij}(E^2+B^2)]$ (here, $i,j=x,y,z$) is the Maxwell stress tensor, which is equal to the tensor of the field momentum flow density taken with the inverse sign (see, e.g., [17]), $\hat{T}\equiv T_{ij}$ is the stress tensor of external forces that keep the immobile positive charges creating the electrostatic potential $e^{-1}U(\boldsymbol{r})$ in equilibrium, S is the surface enveloping the volume V, and $\boldsymbol{s}$ is the unit vector of the normal to the surface element dS, which is directed outward from the volume V. Taking into account the known relationships which follow from the Maxwell equations for the momentum and energy variation in the classical electromagnetic field [17]

$$\frac{\partial}{\partial t}\int_V\mathbf{P}dV+\int_V(q_e+q_i)\boldsymbol{E}dV+\frac{1}{c}\int_V(\boldsymbol{j}\times\boldsymbol{B})dV=\oint_S(\hat{\sigma}\cdot\boldsymbol{s})dS,\quad\frac{\partial}{\partial t}\int_V W_{E,B}dV+\int_V\boldsymbol{j}\boldsymbol{E}dV=-\oint_S(\boldsymbol{\Pi}\cdot\boldsymbol{s})dS,\tag{7c}$$

and the condition of equilibrium of immobile positive charges

$$\overline{\boldsymbol{\nabla}U\rho}+\int_V q_i\boldsymbol{E}dV+\oint_S(\hat{T}\cdot\boldsymbol{s})dS=0\tag{7d}$$

(q_e, q_i, and $\boldsymbol{j}$ are the macroscopic densities of the electron and ion charge and the current, respectively), we obtain, using Eqs. (7a), (7b), and (7d), the following expressions:

$$e\overline{\boldsymbol{E}\rho}+\frac{e}{2mc}\overline{\left[\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)\times\boldsymbol{B}-\boldsymbol{B}\times\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)\right]\rho}=\int_V q_e\boldsymbol{E}dV+\frac{1}{c}\int_V(\boldsymbol{j}\times\boldsymbol{B})dV,$$

$$\frac{e}{2m}\overline{\left[\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)\boldsymbol{E}+\boldsymbol{E}\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)\right]\rho}=\oint_V\boldsymbol{j}\boldsymbol{E}dV.$$

It is therefore implied that the Maxwell equations should include the current determined by the standard operator (2b). It follows from Eqs. (2b) and (5) that the expression for the total current can be represented as

$$\bar{\boldsymbol{j}}=\dot{\bar{\boldsymbol{d}}}+\frac{i}{\hbar}\overline{\boldsymbol{d}\hat{R}\rho}\tag{8}$$

(of course, if the spin is neglected). Equation (8) can be interpreted as the division of the total current into the polarization current and the conductivity current, which is conventional in the electrodynamics of continuous dissipative media (see, e.g., [18]).

The above conclusion on the total current of the system corresponds, in particular, to the known relationships describing the polarization of a two-level system in an external homogeneous electric field (see [5]),

$$\ddot{\bar{\boldsymbol{d}}}+2\gamma_{21}\dot{\bar{\boldsymbol{d}}}+(\gamma_{21}^2+\Omega_{21}^2)\bar{\boldsymbol{d}}=2(N_1-N_2)(\boldsymbol{E}\boldsymbol{d}_{21})\frac{\boldsymbol{d}_{12}\Omega_{21}}{\hbar},$$

[4] As concerns electrostatic (quasielectrostatic) fields, the permittivity of an ensemble of "coupled" particles in constant fields can be determined from purely thermodynamic considerations [15,16] and should not depend at all on characteristic relaxation times. This means that the use of a relaxation operator that does not satisfy the condition $\overline{\boldsymbol{d}\hat{R}\rho}=0$ [specified, e.g., in form (6a)] needs, in the case of constant fields, special discussion for particular situations. Such an approach can be adequate in, e.g., a system through which direct current can pass.

$$\hbar\Omega_{21}\left(\frac{(\dot{N}_2-\dot{N}_1)}{2}+\Gamma_{21}\frac{(N_2-N_1-\Delta N_0)}{2}\right)=\boldsymbol{E}(\dot{\bar{\boldsymbol{d}}}+\gamma_{21}\bar{\boldsymbol{d}}),$$

where $\boldsymbol{d}_{21}$ is the matrix element of the dipole-moment operator, $N_{1,2}$ are the populations of the lower and the upper levels, respectively, $(N_1+N_2=\text{const})$, Ω_{21} is the transition frequency, γ_{21} and Γ_{21} are the relaxation frequencies of the off-diagonal elements of the density matrix and the population difference, respectively, and ΔN_0 is the "equilibrium" population difference, which is established in the absence of an external field. It is seen from the second equation that the energy exchange between the material and the electromagnetic field is determined by the work of the electric field over the total current corresponding to Eq. (8).

Considering an ensemble of space-distributed identical oscillators, where each oscillator is described by Eq. (1),[5] it is convenient to normalize the density matrix not to unity but to the number of oscillators in a unit volume (number density) N. In this case, the results of averaging for the dipole moment and current operators will be equal to the macroscopic (i.e., averaged over a physically small volume) values of the polarization $\boldsymbol{P}=\overline{\boldsymbol{d}\rho}$ and the current density $\boldsymbol{J}=\overline{\hat{\boldsymbol{j}}\rho}$, respectively (see, e.g., [19]). Within the framework of the approach in which the dissipative processes are taken into account by the relaxation operator in Eq. (1), the polarization current $\dot{\boldsymbol{P}}$, as was shown above, can differ from the total current $\boldsymbol{J}$. It was already mentioned that this case is typical for the electrodynamics of dissipative media. In particular, the "effective" polarization $\boldsymbol{P}_{eff}=\int_{-\infty}^{t}\boldsymbol{J}(\tau)d\tau$ and, correspondingly, the "effective" electric displacement vector $\boldsymbol{D}_{eff}=\boldsymbol{E}+4\pi\boldsymbol{P}_{eff}$ should be introduced when complex dielectric permittivity is determined [18].

III. TRANSFORMATION OF THE RELAXATION OPERATOR DURING GAUGE TRANSFORMATION OF ELECTROMAGNETIC FIELDS

Consider the gauge transformation of electromagnetic potentials [17]

$$\boldsymbol{A}_2=\boldsymbol{A}_1+\nabla f,\quad \varphi_2=\varphi_1-\frac{1}{c}\frac{\partial f}{\partial t},\tag{9}$$

where $f(\boldsymbol{r},t)$ is an arbitrary scalar function, Hereafter, we will assign the subscripts "1" or "2" to the operators and quantities specified for potential combinations $\boldsymbol{A}_1,\varphi_1$ and $\boldsymbol{A}_2,\varphi_2$, respectively.

As the gauge transformation is applied, the solution of the Schrödinger equation is transformed as $\psi_2=\psi_1\exp[ief(\boldsymbol{r},t)/\hbar c]$ (see [4]). This transformation of the ψ function follows from the corresponding transformation of the Schrödinger equation, which can be represented as $\hat{S}\psi=0$,

$$\hat{S}_2=e^{ief/\hbar c}\hat{S}_1e^{-ief/\hbar c},$$

where $\hat{S}=i\hbar\partial/\partial t-\hat{H}$. A similar procedure (preserving the gauge invariance) of the ψ function using the Schrödinger equation with the phenomenological relaxation operator $(\hat{S}-\hat{\Gamma})\psi=0$ can probably take place only after the corresponding transformation of the relaxation operator

$$\hat{\Gamma}_2=e^{ief/\hbar c}\hat{\Gamma}_1e^{-ief/\hbar c}.$$

This procedure was found in [2] for the particular case corresponding to the Wiesskopf-Wigner operator $\hat{\Gamma}_1$ for a two-level system with a homogeneous rf electric field, where $\varphi_1=-\boldsymbol{rE}(t),\boldsymbol{A}_1=0$, and $\varphi_2=0,\boldsymbol{A}_2=-c\int_{t_0}^{t}\boldsymbol{E}(\tau)d\tau$.

Considering gauge transformation for the density matrix of a quantum system in arbitrary electromagnetic fields, it is convenient to represent Eq. (1) as $(\hat{L}-\hat{R})\rho=0$, where

$$\hat{L}\rho=i\hbar\frac{\partial}{\partial t}\rho-[\hat{H},\rho].\tag{10a}$$

The operator $\hat{L}$ is transformed with the gauge transformation of electromagnetic potentials as

$$\hat{L}_2=e^{i\chi}\hat{L}_1e^{-i\chi},\tag{10b}$$

where

$$\chi(\boldsymbol{r},\boldsymbol{r}',t)=\frac{e}{\hbar c}[f(\boldsymbol{r},t)-f(\boldsymbol{r}',t)].\tag{10c}$$

From Eqs. (10a)–(10c) it follows that the solution of the density-matrix equation in the case of gauge transformation of the fields without the relaxation is transformed as

$$\rho_2=\rho_1e^{i\chi}.\tag{10d}$$

Taking the relationships

$$(e^{i\chi})_{\boldsymbol{r}'=\boldsymbol{r}}=1,\quad \hat{\boldsymbol{p}}e^{i\chi}\rho=e^{i\chi}\left[\hat{\boldsymbol{p}}+\frac{e}{c}(\boldsymbol{A}_2\text{-}\boldsymbol{A}_1)\right]\rho\tag{10e}$$

into account, it can easily be verified that transformation (10d) preserves, in particular, the average dipole moment, the current, and the energy of the system

$$\overline{\boldsymbol{d}\rho_1}=\overline{\boldsymbol{d}\rho_2},\quad \overline{\hat{\boldsymbol{j}}_1\rho_1}=\overline{\hat{\boldsymbol{j}}_2\rho_2},\quad \overline{\hat{W}_1\rho_1}=\overline{\hat{W}_2\rho_2}.\tag{11}$$

With allowance for the relaxation processes, density-matrix transformation (10d) that ensures gauge invariance can take place only if during the gauge transformation of the fields the relaxation operator is transformed in exactly the same way as the operator $\hat{L}$,

$$\hat{R}_2=e^{i\chi}\hat{R}_1e^{-i\chi}.\tag{12}$$

Proof of the existence of property (12) in the case of correct derivation of the relaxation operator is presented in Sec. III.

[5] In other words, oscillators either do not interact with each other or such a process can be described by introducing a phenomenological relaxation operator in the density-matrix equation of a separate oscillator.

Note that property (12) for the relaxation parameter $\hat{R}$ certainly ensures gauge invariance of the expression determined by Eq. (8) for the difference between the total current and the polarization current $\bar{\boldsymbol{j}}-\dot{\bar{\boldsymbol{d}}}$ since $\overline{\boldsymbol{d}\hat{R}_2\rho_2}=\overline{\boldsymbol{d}\hat{R}_1\rho_1}$ in view of Eqs. (10d), (10e), and (12).

Finalizing this section, we will discuss, in greater detail than we did in the introduction, another approach for a gauge-invariant description of dissipative quantum systems. This approach consists in quantization of a time-dependent Hamiltonian, which, in the classical limit, correctly describes the motion of a particle in a medium with linear friction. The corresponding Hamiltonian was proposed in [20] for a damped harmonic oscillator. In [21][6] and [9], this approach was used for analyzing the motion of particles in a homogeneous electric field. In general, such a Hamiltonian can be represented as

$$H_\gamma=\frac{1}{2m}e^{-\gamma t}\left(\boldsymbol{p}-\frac{e}{c}\int_{-\infty}^{t}e^{\gamma\tau}\frac{\partial\boldsymbol{A}(\boldsymbol{r},\tau)}{\partial\tau}d\tau\right)^2+e^{\gamma t}e\varphi(\boldsymbol{r},t).$$

The second-order equation for the coordinate

$$\ddot{\boldsymbol{r}}=-\gamma\dot{\boldsymbol{r}}+\frac{e}{m}\left\{\left[-\frac{1}{c}\frac{\partial\boldsymbol{A}(\boldsymbol{r},t)}{\partial t}-\boldsymbol{\nabla}\varphi(\boldsymbol{r},t)\right]+\left[\frac{\dot{\boldsymbol{r}}}{c}\mathrm{rot}\left(\int_{-\infty}^{t}e^{\gamma\tau}\frac{\partial\boldsymbol{A}(\boldsymbol{r},\tau)}{\partial\tau}d\tau\right)\right]\right\},$$

which corresponds to this Hamiltonian under the condition $\mathrm{rot}\,\boldsymbol{A}=0$, obviously, correctly describes the motion of a classical particle in a medium with the linear friction force $\boldsymbol{F}_{fr}=-\gamma m\dot{\boldsymbol{r}}$. Then one can make use of the standard quantization procedure by making the replacement $\boldsymbol{p}\to\hat{\boldsymbol{p}}=-i\hbar\boldsymbol{\nabla}$ and substituting the corresponding Hamiltonian into the Schrödinger equation $i\hbar\partial\psi/\partial t=\hat{H}_\gamma\psi$. It can easily be verified that in this case, the velocity operator determined by the relationship $\partial\langle\psi|\boldsymbol{r}|\psi\rangle/\partial t=\langle\psi|\hat{\dot{\boldsymbol{r}}}|\psi\rangle$ will have the form $\hat{\dot{\boldsymbol{r}}}=e^{-\gamma t}\{\hat{\boldsymbol{p}}-(e/c)\int_{-\infty}^{t}e^{\gamma\tau}[\partial\boldsymbol{A}(\boldsymbol{r},\tau)/\partial\tau]d\tau\}m^{-1}$ and the ψ function will be transformed as $\psi_2=\psi_1\exp\{(ie/c\hbar)\int_{-\infty}^{t}e^{\gamma\tau}[\partial f(\boldsymbol{r},\tau)/\partial\tau]d\tau\}$ by gauge transformation (9). This, in particular, ensures gauge invariance of the average velocity $\langle\psi|\hat{\dot{\boldsymbol{r}}}|\psi\rangle$. It is exactly what was shown in [9] for the case of a homogeneous electric field[7] given by $E_x=\mathrm{Re}\,\tilde{E}_xe^{i\omega t}$.

We note that the limitation of such a method of taking dissipative effects into account was mentioned as early as in startup paper [20]. For example, using the nonstationary Hamiltonian $\hat{H}_\gamma$, it is impossible to describe the transition of the system to the state of equilibrium with the reservoir, which is the basis of correct models of quantum-system relaxation (see, e.g., [5,16]). Moreover, as we have just shown, such a Hamiltonian yields an erroneous result even in the classical limit if the magnetic field is nonzero (when $\mathrm{rot}\,\boldsymbol{A}\neq0$).

IV. CORRECT DERIVATION OF THE RELAXATION OPERATOR

We will give a fairly general proof that the correct derivation of the relaxation operator "from the first principles" ensures the properties required for gauge invariance. Consider two examples.

(i) Dynamic system in a classical noise field. Let the system described in Sec.I (we will call it the e system), which is characterized by the coordinates $\boldsymbol{r}$ and Hamiltonian (2a), interact with the classical noise field specified by the vector potential $\boldsymbol{a}_q(\boldsymbol{r},t)$. Averaging over noise (which can, depending on the particular formulation of the problem, be averaging over a physically small volume, correlation time, or an ensemble of realizations) will be denoted by an overbar with index q. In particular, we assume $\overline{\boldsymbol{a}_q}^q=0$.

Represent the Hamiltonian of the total (i.e., not averaged) system $\hat{H}_\Sigma$ in the form

$$\hat{H}_\Sigma=\hat{H}+\hat{I}_q,\quad \hat{I}_q=\hat{h}_q+\frac{e^2\boldsymbol{a}_q^2}{2mc^2},\quad \hat{h}_q=-\frac{e}{2c}(\hat{\boldsymbol{v}}\boldsymbol{a}_q+\boldsymbol{a}_q\hat{\boldsymbol{v}}),\tag{13a}$$

where $\hat{\boldsymbol{v}}=\hat{\boldsymbol{p}}/m-e\boldsymbol{A}(\boldsymbol{r},t)/mc$ is the particle velocity operator. The equation for the not averaged density matrix ρ_Σ,

$$i\hbar\dot{\rho}_\Sigma=[\hat{H}_\Sigma,\rho_\Sigma].\tag{13b}$$

We now single out the averaged and "noise" components of the density matrix and the Hamiltonian,

$$\rho_\Sigma=\overline{\rho}_\Sigma^q+\delta\rho,\quad \overline{\rho}_\Sigma^q=\rho,\tag{14a}$$

$$\hat{H}_\Sigma=\overline{\hat{H}_\Sigma}^q+\delta\hat{I},\quad \overline{\hat{H}_\Sigma}^q=\hat{H}+\frac{e^2\overline{\boldsymbol{a}_q^2}^q}{2mc^2},\tag{14b}$$

$$\delta\hat{I}=\hat{I}_q-\frac{e^2\overline{\boldsymbol{a}_q^2}^q}{2mc^2}=\hat{h}_q+o(\boldsymbol{a}_q^2).$$

Substitute Eq. (14) into Eq. (13b) for the density matrix and average over the noise field, assuming that $\delta\rho\propto\boldsymbol{a}_q$ and the noise intensity is small. As a result, we obtain Eq. (1) for the averaged density matrix along with the equations for the relaxation operator,

$$\hat{R}\rho=\frac{e^2}{2mc^2}[\overline{\boldsymbol{a}_q^2}^q,\rho]+\overline{[\hat{h}_q,\delta\rho]}^q,\tag{15a}$$

where the "noise" components of the density matrix $\delta\rho$ is related to the averaged one ρ by the following equation:

[6] In [21], it is stated that quantum-mechanical solutions for different gauges are significantly different even in the particular case of a homogeneous constant field in the absence of dissipation. However, this statement is very controversial: the authors of [21] give an explicit expression for the middle coordinate $\bar{x}(t)$ of the "wave packet" $|\psi|^2(x,t)$ only in the case of $\boldsymbol{A}$ gauge, and they did not obtain the corresponding expression for φ gauge in closed form.

[7] In [9], the same expression for the average velocity $\langle\psi|\dot{x}|\psi\rangle=\mathrm{Re}\{e\tilde{E}_xe^{i\omega t}/[m(\gamma+i\omega)]\}$ was obtained for two gauges ($E_x=-\partial\varphi/\partial x$ and $E_x=-c\,\partial A_x/\partial t$) with the use of the extended phase-space formulation of quantum mechanics [11].

$$\hat{L}\delta\rho = [\hat{h}_q, \rho], \tag{15b}$$

[the result of averaging in Eq. (15a) depends on the statistical properties of the noise field]. The first term on the right-hand side of Eq. (15a) describes the averaged force action of the noise field on the e system. A classical counterpart of this effect is the effect of averaged ponderomotive action of the high-frequency field on charged particles [22]. By direct checking, one can establish the following property of the operator $\hat{h}_q$: $[\hat{h}_{q_2}, \rho] = e^{i\chi}[\hat{h}_{q_1}, e^{-i\chi}\rho]$. It can easily be verified that the simultaneous fulfillment of the last condition and the same condition (10b) for the operator $\hat{L}$ leads to the property $\delta\rho_2(\rho) = e^{i\chi}\delta\rho_1(e^{-i\chi}\rho)$ for the solution of Eq. (15b). As a result, one can obtain the property (12) for the relaxation operator $\hat{R}$.

(ii) Interaction of two subsystems. Let the e system, which is characterized by the Hamiltonian $\hat{H}(\boldsymbol{r})$, interact with the Q system ("reservoir") described by a set of coordinates $\boldsymbol{q}$ and energy operator $\hat{Q}(\boldsymbol{q})$. The Hamiltonian of the total system has the form

$$\hat{H}_\Sigma(\boldsymbol{r},\boldsymbol{q}) = \hat{H}(\boldsymbol{r}) + \hat{Q}(\boldsymbol{q}) + \hat{I}(\boldsymbol{r},\boldsymbol{q}), \tag{16a}$$

where $\hat{I}(\boldsymbol{r},\boldsymbol{q})$ is the generalized (i.e., the linear dependence from velocity is possible) gauge-invariant interaction potential. In a fairly general form the interaction operator in Eq. (16a) can be presented as $\hat{I} = \frac{1}{2}[\hat{\boldsymbol{a}}(\boldsymbol{r},\boldsymbol{q})\hat{\boldsymbol{v}} + \hat{\boldsymbol{v}}\hat{\boldsymbol{a}}(\boldsymbol{r},\boldsymbol{q})] + \hat{V}(\boldsymbol{r},\boldsymbol{q})$, where the action of the vector and scalar operators $\hat{\boldsymbol{a}}(\boldsymbol{r},\boldsymbol{q})$ and $\hat{V}(\boldsymbol{r},\boldsymbol{q})$ on the coordinate function $\boldsymbol{r}$ reduces to simple multiplication [see, e.g., Eq. (13a)]. Represent the density matrix of the total system $\rho_\Sigma(\boldsymbol{r},\boldsymbol{r}',\boldsymbol{q},\boldsymbol{q}')$ as

$$\rho_\Sigma = \rho(\boldsymbol{r},\boldsymbol{r}')\rho_Q(\boldsymbol{q},\boldsymbol{q}') + \rho_I(\boldsymbol{r},\boldsymbol{r}',\boldsymbol{q},\boldsymbol{q}'), \tag{16b}$$

where $\rho = \overline{\rho_\Sigma}^q = \int_\infty \rho_\Sigma(\boldsymbol{r},\boldsymbol{r}',\boldsymbol{q},\boldsymbol{q}')_{\boldsymbol{q}'=\boldsymbol{q}} d^3q$, $\rho_Q = \overline{\rho_\Sigma} = \int_\infty \rho_\Sigma(\boldsymbol{r},\boldsymbol{r}',\boldsymbol{q},\boldsymbol{q}')_{\boldsymbol{r}'=\boldsymbol{r}} d^3r$ (here, the overbar with index q denotes averaging over the coordinates of the reservoir $\boldsymbol{q}$, whereas the overbar without the index denotes—as before—averaging over the coordinates $\boldsymbol{r}$ of the e system). The system is described by Eq. (13b) for the density matrix ρ_Σ. Substitute Eq. (16b) into Eq. (13b) and average over the coordinates of the Q system and e system, respectively. Allowing for the properties $\bar{\rho}_I = \bar{\rho}_I^q = 0$ and $[\hat{Q}, \rho_{Q,I}]^q = [\hat{H}, \rho] = [\hat{H}, \rho_I] = 0$, we obtain the equations for density matrices ρ and ρ_Q,

$$i\hbar\dot{\rho} = [\hat{H}, \rho] + \overline{[\hat{I}, \rho_I]}^q + \overline{[\hat{I}, \rho\rho_Q]}^q,$$

$$i\hbar\dot{\rho}_Q = [\hat{Q}, \rho_Q] + \overline{[\hat{I}, \rho_I]} + \overline{[\hat{I}, \rho\rho_Q]}.$$

Multiplying the last two equations by matrices ρ_Q and ρ, respectively, and subtracting them from Eq. (13b), one can obtain the equation for the matrix ρ_I,

$$i\hbar\dot{\rho}_I - [(\hat{H} + \hat{Q}), \rho_I] - [\hat{I}, \rho\rho_Q] + \overline{[\hat{I}, \rho\rho_Q]}^q\rho_Q + \overline{[\hat{I}, \rho\rho_Q]}\rho$$
$$= [\hat{I}, \rho_I] - \overline{[\hat{I}, \rho_I]}^q\rho_Q - \overline{[\hat{I}, \rho_I]}\rho = o(\hat{I}^2).$$

Assuming that $\rho_I \propto \hat{I}$ and the interaction energy is small compared with the characteristic values of the "intrinsic" energies, we obtain Eq. (1), in which the relaxation operator $\hat{R}$ is determined by the following system of equations:

$$\hat{R}\rho = [\hat{\phi}, \rho] + \overline{[\hat{I}, \rho_I]}^q, \tag{17a}$$

$$\hat{L}\rho_I - [\hat{Q}, \rho_I] = [\hat{I}, \rho\rho_Q] - \overline{[\hat{I}, \rho\rho_Q]}^q\rho_Q - \overline{[\hat{I}, \rho\rho_Q]}\rho, \tag{17b}$$

where the form of the density matrix of the reservoir ρ_Q is usually determined from thermodynamic considerations, the operator $\hat{L}$ is determined by Eq. (10a), and $\hat{\phi}(\boldsymbol{r}) = \overline{\hat{I}\rho_Q}^q$. The first term on the right-hand side of Eq. (17a) describes the possible modification of the Hamiltonian (2a) due to the averaged force action of the Q system. This term can be eliminated by overdetermining the Hamiltonian $\hat{H} \to \hat{H} + \hat{\phi}$ in Eq. (2a). The property (10b) of the operator $\hat{L}$ certainly ensures the existence of property (12) for the operator $\hat{R}$. The proving is exactly the same as in the previous paragraph (i). The above-derived interaction operator $\hat{I}$ has the properties $[\hat{I}_2, \rho] = e^{i\chi}[\hat{I}_1, e^{-i\chi}\rho]$ and $[\hat{I}_1, e^{-i\chi}\rho\rho_Q] = [\hat{I}_2, \rho\rho_Q]$, which can easily be checked, so, one can obtain the relation $\rho_{I2}(\rho) = e^{i\chi}\rho_{I1}(e^{-i\chi}\rho)$ from Eq. (17b) and, as a result, the relation $\hat{R}_2\rho = e^{i\chi}\hat{R}_1(e^{-i\chi}\rho)$ from Eq. (17a). Generalization of the above proof to the case of a set of Q systems, where the Hamiltonian of the reservoir and the interaction operator are given by $\hat{Q} \equiv \Sigma_k\hat{Q}_k(\mathbf{q}_k)$ and $\hat{I} \equiv \Sigma_k\hat{I}_k(\mathbf{r}, \mathbf{q}_k)$, is trivial. Interaction of the e system and quantized wave field can be represented in such a form [5].

Thus, the solution of Eq. (1) with relaxation operator $\hat{R}$ specified by systems of Eqs. (15) and (17) satisfies condition (10d) which ensures gauge invariance for the observables. It is important to mention that the latter statement is far from being a trivial consequence of the corresponding property of the exact von Neumann Eq. (13b) since when Eq. (1) supplemented by Eqs. (15) and (17) determining the relaxation operator was derived from Eq. (13b), we neglected terms of the order of $\hat{I}^3$ and $\boldsymbol{a}_q^3$, respectively.

It is a key factor that condition (12) turns out to be fulfilled in general if Eq. (15b) or, correspondingly, Eq. (17b) are solved for the operator $\hat{L}$ dependent on a "regular" external field specified by the potentials $\boldsymbol{A}$ and φ in general form. The corresponding procedure is possible, in principle, but is very cumbersome (see, e.g., [5,23]). As a rule, the relaxation operator is calculated for the unperturbed system (or within the framework of another simplified gauge-noninvariant model, as in [23]) and cannot ensure gauge invariance of the solution of Eq. (1) since this operator is independent of the field potentials.

V. PHENOMENOLOGICAL GAUGE-INVARIANT RELAXATION OPERATOR

Assume we know the relaxation operator $\hat{R}_0$ determined in the absence of an external electromagnetic field (or within the framework of another model assuming, as in [23], that $\boldsymbol{A} = 0$, which is not significant in this treatment). In the case $\mathrm{rot}\,\boldsymbol{A} = 0$, for obtaining a gauge-invariant representation of

the relaxation operator $\hat{R}_A$ dependent on the vector potential of the field, it suffices to make use of Eqs. (12) and (10c), putting $\hat{R}_1 \to \hat{R}_0, \hat{R}_2 \to \hat{R}_A$, and $\nabla f \to \boldsymbol{A}$ Thus, we obtain

$$\hat{R}_A = e^{iu_0}\hat{R}_0 e^{-iu_0}, \tag{18a}$$

where

$$u_0 = \frac{e}{\hbar c}\int_{r'}^{r} \boldsymbol{A}(\boldsymbol{\xi},t)d\boldsymbol{\xi}. \tag{18b}$$

The applicability range of Eq. (18a) for different amplitudes and scales of spatiotemporal inhomogeneity of the vector potential is fully determined by the sort of the electric fields for which the expression for the operator $\hat{R}_0$ (which we assume known) is valid. In the case $\mathrm{rot}\,\boldsymbol{A} \neq 0$, the corresponding modification of Eq. (18a) is only possible for weak magnetic fields $\boldsymbol{B} = \mathrm{rot}\,\boldsymbol{A}$ since the operator $\hat{R}_0$ by definition neglects the effect of the magnetic field on the relaxation process. Confining ourselves to the approximation of weak magnetic fields (the corresponding criterion should be formulated for each particular case), we propose the following generalization of Eq. (18a):

$$\hat{R}_A = e^{iu}\hat{R}_0 e^{-iu}, \tag{19a}$$

where

$$u = \frac{e}{\hbar c}\int_{\Phi} \boldsymbol{A}(\boldsymbol{\xi},t)d\boldsymbol{\xi}. \tag{19b}$$

Here, $\Phi \equiv \Phi(\boldsymbol{r},\boldsymbol{r}')$ is the contour connecting the points $\boldsymbol{r}$ and $\boldsymbol{r}'$. The algorithm for specifying this contour defines the function $u(\boldsymbol{r},\boldsymbol{r}',t)$ in the case $\mathrm{rot}\,\boldsymbol{A} \neq 0$. The operator $\hat{R}_A(\rho)$ determined by Eqs. (19a) and (19b) satisfies condition (12) for the gauge transformation of field potentials. Indeed, allowing for Eqs. (9) and (10c), we obtain

$$u_2 = \frac{e}{\hbar c}\int_{\Phi} (\boldsymbol{A}_1 + \nabla f)d\boldsymbol{\xi} = u_1 + \chi.$$

The class of admissible contours $\Phi(\boldsymbol{r},\boldsymbol{r}')$ can be limited by requiring that when the characteristic relaxation time tends to zero the current in the system also tends to zero and the energy of the system tends to $E_n\rho_{0nn}$, where ρ_0 is a certain diagonal matrix corresponding to thermodynamic equilibrium with the reservoir. We now will show that the contours satisfying this condition should collapse for $\boldsymbol{r} \to \boldsymbol{r}'$, i.e., should satisfy the condition

$$\lim_{r \to r'} \int_{\Phi} dl = 0, \tag{20a}$$

where dl is a differentially small element of the length of the curve along the $\Phi(\boldsymbol{r},\boldsymbol{r}')$ contour. Indeed, let the operator $\hat{R}_0 = -i\hbar\gamma\hat{\tilde{R}}_0$ describe relaxation with characteristic rate γ to some diagonal matrix ρ_0, which satisfies the conditions $\hat{\tilde{R}}_0\rho_0 = [\hat{H},\rho_0] = 0$. In this case, Eq. (1) with relaxation operator (19) can be represented as

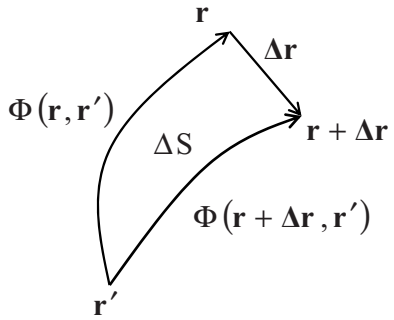

FIG. 1. The domain of integration in calculating the difference $u(\boldsymbol{r}+\Delta\boldsymbol{r},\boldsymbol{r}',t) - u(\boldsymbol{r},\boldsymbol{r}',t)$.

$$\gamma\hat{\tilde{R}}_0(e^{-iu}\rho) + e^{-iu}\{\dot{\rho} + (i/\hbar)[\hat{H},\rho]\} = 0.$$

In the limit where the characteristic relaxation rate γ tends to ∞, the solution of this equation corresponds to the asymptotic form $\rho = \rho_0 e^{iu} + o(\gamma^{-1})$. We now take into account that the following relationships are valid under condition (20a):

$$(e^{iu})_{r'=r} = 1, \quad \lim_{r \to r'}[\hat{\boldsymbol{p}}u(\boldsymbol{r},\boldsymbol{r}',t)] = -i\frac{e}{c}\boldsymbol{A}(\boldsymbol{r},t),$$

$$\lim_{r \to r'}[\hat{\boldsymbol{p}}^2 u(\boldsymbol{r},\boldsymbol{r}',t)] = -\frac{e\hbar}{c}\mathrm{div}\,\boldsymbol{A}(\boldsymbol{r},t). \tag{20b}$$

The first relationship in Eq. (20b) is obvious, and the other two can easily be proved by using the expression for the differentially small increment of the function u,

$$u(\boldsymbol{r}+\Delta\boldsymbol{r},\boldsymbol{r}',t) - u(\boldsymbol{r},\boldsymbol{r}',t) = \frac{e}{\hbar c}\left[\boldsymbol{A}(\boldsymbol{r},t)\cdot\Delta\boldsymbol{r} + \int_{\Delta S}(\boldsymbol{B}\cdot\boldsymbol{s})dS\right] + o((\Delta\boldsymbol{r})^2) + \ldots,$$

where the second term in the parentheses is the flux of the vector $\boldsymbol{B}(\boldsymbol{r},t) = \mathrm{rot}\,\boldsymbol{A}$ via the surface $\Delta S \equiv \Delta S(\boldsymbol{r},\boldsymbol{r}',\Delta\boldsymbol{r})$ spanned on the closed contour passing through the points $\boldsymbol{r}'$, $\Delta\boldsymbol{r}+\boldsymbol{r}$, and $\boldsymbol{r}$, which is formed by the contours $\Phi(\Delta\boldsymbol{r}+\boldsymbol{r},\boldsymbol{r}')$ and $\Phi(\boldsymbol{r},\boldsymbol{r}')$ and a segment of the straight line between the points $\Delta\boldsymbol{r}+\boldsymbol{r}$ and $\boldsymbol{r}$ (see Fig. 1). For the smooth curve $\Phi(\boldsymbol{r},\boldsymbol{r}')$, it suffices to consider the case of small values of $|\boldsymbol{r}-\boldsymbol{r}'|$ where this closed contour forms a triangle,

$$\Delta S \approx \frac{1}{2}|\Delta\boldsymbol{r} \times (\boldsymbol{r}-\boldsymbol{r}')|.$$

It follows from Eq. (20b) that

$$\lim_{r \to r'}\left\{\left[\hat{\boldsymbol{p}} - \frac{e}{c}\boldsymbol{A}(\boldsymbol{r},t)\right]e^{iu}\rho\right\} = (\hat{\boldsymbol{p}}\rho)_{r'=r}, \tag{20c}$$

$$\lim_{\boldsymbol{r}\to\boldsymbol{r}'}(\hat{W}e^{iu}\rho)=(\hat{H}_0\rho)_{\boldsymbol{r}'=\boldsymbol{r}}.$$

For the density matrix $\rho\to\rho_A=\rho_0 e^{iu}$, by virtue of Eqs. (20b) and (20c), we have[8]

$$\bar{\boldsymbol{d}}_{\gamma\to\infty}=\overline{\boldsymbol{d}(e^{iu}\rho_0)}=\overline{\boldsymbol{d}\rho_0}=0,\quad \bar{\boldsymbol{j}}_{\gamma\to\infty}=\overline{\frac{e}{m}\left[\hat{\boldsymbol{p}}-\frac{e}{c}\boldsymbol{A}(\boldsymbol{r},t)\right](e^{iu}\rho_0)}$$
$$=\overline{\hat{\boldsymbol{p}}\rho_0}=0,$$

$$\bar{W}_{\gamma\to\infty}=\overline{\hat{W}(e^{iu}\rho_0)}=\overline{\hat{H}_0\rho_0}=E_n\rho_{0nn}.$$

We note that when the characteristic relaxation time tends to zero, the "initial" relaxation operator $\hat{R}_0$ corresponds to the asymptotic solution $\rho\to\rho_0$, which leads, despite the correct relationship for the dipole moment $\bar{\boldsymbol{d}}_{\gamma\to\infty}=\overline{\boldsymbol{d}\rho_0}=0$, to physically meaningless relationships for the electric current and energy

$$\bar{\boldsymbol{j}}_{\gamma\to\infty}=\overline{\frac{e}{m}\left[\hat{\boldsymbol{p}}-\frac{e}{c}\boldsymbol{A}(\boldsymbol{r},t)\right]\rho_0}=-\frac{e^2}{mc}\boldsymbol{A}_{nn}\rho_{0nn},$$

$$\bar{W}_{\gamma\to\infty}=\overline{\hat{W}\rho_0}=\left[E_n+\frac{e^2}{2mc^2}(A^2)_{nn}\right]\rho_{0nn}.$$

Equation (1) with relaxation operator (19a) certainly permits one to pass to the electric-dipole approximation if the inhomogeneity scale l_E of an external electromagnetic field significantly exceeds a, the size of the region of electron motion ("atom" size). Making use of gauge transformation (9) for $f=-\boldsymbol{r}\boldsymbol{A}_1(\boldsymbol{r}=0,t)$, the density-matrix equation can be reduced to

$$i\hbar\dot{\rho}-[\hat{H}_0,\rho]=[\hat{h}_{dE},\rho]+\hat{R}_0(\rho)+o(a/l_E),\qquad(21)$$

where $\hat{h}_{dE}=-\boldsymbol{d}\boldsymbol{E}(\boldsymbol{r}=0,t)$ is the standard operator of the field-particle interaction energy within the framework of the electric-dipole approximation and the terms $o(a/l_E)$, beginning with the first one, are of higher orders of smallness with respect to the parameter (a/l_E).

With allowance for the field nonpotentiality, the relaxation operator $\hat{R}_A$ depends, generally speaking, on the choice of the $\Phi(\boldsymbol{r},\boldsymbol{r}')$ contour in Eq. (19b). The problem of the influence of this arbitrariness on the final expressions for the average values was not clarified for the general case in this paper. However, the equations describing the evolution of the average values of the dipole moment, current, and energy of the system do not depend on the choice of the $\Phi(\boldsymbol{r},\boldsymbol{r}')$ trajectory for the class of contours satisfying condition (20a), at least within the framework of an approximation in which the equations of the dynamics of average quantities form a closed system.

[8]In this case, the correction to the polarization current in a dissipative system, which is described by Eq. (8), is not significant since $\hat{R}_0\rho_0=\hat{R}_A\rho_A=0$.

VI. APPROXIMATION OF CONSTANT RELAXATION TIMES: EQUATIONS FOR AVERAGE QUANTITIES OF A HARMONIC DISSIPATIVE OSCILLATOR IN EXTERNAL FIELDS

Consider the case where the relaxation processes can be divided into "transverse" and "longitudinal" components of the density matrix in the absence of external fields and the relaxation of the "off-diagonal" and "diagonal" components are determined by characteristic times γ^{-1} and Γ^{-1}, respectively. In this approximation, the relaxation operator $\hat{R}_0$ can be represented as

$$\hat{R}_0\rho=-i\hbar[\gamma\rho_\perp+\Gamma(\rho_\parallel-\rho_0)],\qquad(22)$$

where ρ_0 is the diagonal matrix corresponding to the equilibrium state in the absence of external electromagnetic fields: $[\hat{H},\rho_0]=0$. From Eqs. (22) and (19a), we obtain the following relaxation operator $\hat{R}_A$:

$$\hat{R}_A\rho=-i\hbar[(\gamma+\Gamma)\rho-e^{iu}(\gamma\tilde{\rho}_\parallel+\Gamma\tilde{\rho}_\perp+\Gamma\rho_0)],\qquad(23)$$

where $\tilde{\rho}_{\parallel,\perp}=(e^{-iu}\rho)_{\parallel,\perp}$. We now multiply Eq. (1), which has a relaxation operator determined by Eq. (23), by the operator of the dipole moment $\boldsymbol{d}$. Using averaging, we find the operator of the derivative of the dipole moment determined by the relationship $\dot{\bar{\boldsymbol{d}}}=\overline{\hat{\dot{\boldsymbol{d}}}\rho}$,

$$\hat{\dot{\boldsymbol{d}}}=\frac{e}{m}\left(\hat{\boldsymbol{p}}-\frac{e\boldsymbol{A}}{c}\right)-\gamma\boldsymbol{d}.\qquad(24a)$$

Then we perform a similar procedure with the polarization-current operator $\hat{\dot{\boldsymbol{d}}}$ found above and the energy operator $\hat{W}$. As a result, allowing for Eqs. (4a), (4b), (20b), and (24a), we obtain the following equations for the average values:[9]

$$\ddot{\bar{\boldsymbol{d}}}+2\gamma\dot{\bar{\boldsymbol{d}}}+\gamma^2\bar{\boldsymbol{d}}+\frac{e}{m}\overline{\boldsymbol{\nabla}U\rho}=\frac{e^2}{m}\overline{\boldsymbol{E}\rho}$$
$$+\frac{e}{mc}\overline{\left[\frac{1}{2}(\hat{\dot{\boldsymbol{d}}}\times\boldsymbol{B}-\boldsymbol{B}\times\hat{\dot{\boldsymbol{d}}})+\gamma(\boldsymbol{d}\times\boldsymbol{B})\right]\rho}.\qquad(24b)$$

$$\dot{\bar{W}}+\Gamma(\bar{W}-W_0)=\overline{\left[\frac{1}{2}(\hat{\dot{\boldsymbol{d}}}\boldsymbol{E}+\boldsymbol{E}\hat{\dot{\boldsymbol{d}}})+\gamma\boldsymbol{d}\boldsymbol{E}\right]\rho},\qquad(24c)$$

$$\bar{\boldsymbol{j}}=\dot{\bar{\boldsymbol{d}}}+\gamma\bar{\boldsymbol{d}},\qquad(24d)$$

where $\bar{W}=\overline{\hat{W}\rho}$ and $W_0=\overline{\hat{H}_0\rho_0}=E_n\rho_{onn}$. Taking into account the relationship $\ddot{\bar{\boldsymbol{d}}}+2\gamma\dot{\bar{\boldsymbol{d}}}+\gamma^2\bar{\boldsymbol{d}}=\frac{\partial\bar{\boldsymbol{j}}}{\partial t}+\gamma\bar{\boldsymbol{j}}$, which follows from Eq. (24d), it can easily be verified that Eqs. (24a), (24b), and (24d) correspond to general Eqs. (5), (7a), and (8) for the

[9]In this case, the quantity $\mathrm{Sp}\,\rho\equiv\bar{\rho}$ is described by the equation $\dot{\bar{\rho}}=\Gamma(\bar{\rho}_0-\bar{\rho})$, i.e., the normalization $\mathrm{Sp}\,\rho=1$ is preserved in time if this is true at the initial instant, namely, if $\mathrm{Sp}\,\rho(t=0)=\mathrm{Sp}\,\rho_0=1$.

considered relaxation model, within the framework of which

$$\frac{i}{\hbar}\overline{\hat{d}\hat{R}\rho} = \gamma \boldsymbol{d}, \quad \overline{\hat{\boldsymbol{F}}_{fr}\rho} = -\gamma\overline{\left(\hat{\boldsymbol{p}} - \frac{e\boldsymbol{A}}{c}\right)\rho} = -\gamma\frac{m}{e}\bar{\boldsymbol{j}},$$

$$\overline{\hat{G}\rho} = \Gamma(\bar{W} - W_0).$$

It is important to mention that if, instead of the modified operator $\hat{R}_A$, we used the "initial" relaxation operator $\hat{R}_0$ in the course of such a procedure, then physically meaningless terms not disappearing in the limit $\boldsymbol{A}=\text{const}$ and equal to $-(e^2/mc)[\Gamma\overline{\boldsymbol{A}\rho_0}+(\gamma-\Gamma)\overline{\boldsymbol{A}\rho_{\|}}]$ and $(e^2/2mc^2)[\Gamma\overline{\boldsymbol{A}^2\rho_0}+(\gamma-\Gamma)\overline{\boldsymbol{A}^2\rho_{\perp}}]$, respectively, would appear on the right-hand sides of Eqs. (24b) and (24c).

Within the framework of the approximation of homogeneous fields $\boldsymbol{E}$ and $\boldsymbol{B}$, as well as for a parabolic three-dimensional potential well, where $U=\frac{1}{2}(\alpha_x x^2+\alpha_y y^2+\alpha_z z^2)$, from Eqs. (24b) and (24c) we obtain the equations (of the type of Bloch equations) for the dipole moment and the energy of a three-dimensional dissipative oscillator in magnetic and electric fields,

$$\ddot{\bar{\boldsymbol{d}}} + 2\gamma\dot{\bar{\boldsymbol{d}}} + \gamma^2\bar{\boldsymbol{d}} + \hat{\Theta}\bar{\boldsymbol{d}} + \omega_H\boldsymbol{b}\times(\dot{\bar{\boldsymbol{d}}} + \gamma\bar{\boldsymbol{d}}) = \frac{e^2}{m}\boldsymbol{E}, \quad (25a)$$

$$\dot{\bar{W}} + \Gamma(\bar{W} - W_0) = (\dot{\bar{\boldsymbol{d}}} + \gamma\bar{\boldsymbol{d}})\boldsymbol{E}, \quad (25b)$$

where $\boldsymbol{B}=\boldsymbol{b}B$, $\boldsymbol{b}$ is a unit vector that specifies the magnetic-field direction, $\omega_H=eB/mc$ is the gyrofrequency, and $\hat{\Theta}$ is the diagonal tensor, such that $\hat{\Theta}\equiv\Theta_{ij}=\delta_{ij}\alpha_j/m$ (here, $i,j=x,y,z$). The average value of the total current $\bar{\boldsymbol{j}}$ is related to the dipole moment of the system by Eq. (24d). Equations (25a) and (25b) are completely independent of the choice of the integration path $\Phi(\boldsymbol{r},\boldsymbol{r}')$ in Eq. (19b).

Note that Eqs. (25a) and (25b) under the condition $\omega_H=0$ differ from the known equations for the polarization and energy of a two-level system, which are presented in Sec. II (see also [5]), only by the expression for the "exciting force" on the right-hand side of the equation for the dipole moment $[e^2\boldsymbol{E}/m$ instead of $2(N_1-N_2)(\boldsymbol{E}\boldsymbol{d}_{21})(\boldsymbol{d}_{12}\Omega_{21}/\hbar)]$. This difference is natural since by summing over the levels of a multilevel system, we obtain[10] $\Sigma_{n>k}2(N_k-N_n)(\boldsymbol{E}\boldsymbol{d}_{nk})(\boldsymbol{d}_{kn}\Omega_{nk}/\hbar)=e^2\boldsymbol{E}/m$.

With allowance for relationship (24d), Eq. (25a) can be reduced to the equation for the current

$$\frac{\partial\bar{\boldsymbol{j}}}{\partial t} + \gamma\bar{\boldsymbol{j}} + \omega_H\boldsymbol{b}\times\bar{\boldsymbol{j}} + \hat{\Theta}\left(e^{-\gamma t}\int_{-\infty}^{t} e^{\gamma\tau}\bar{\boldsymbol{j}}d\tau\right) = \frac{e^2}{m}\boldsymbol{E}. \quad (25c)$$

Equation (25c), obviously, corresponds to the classical limit in the absence of relaxation processes[11] (i.e., for $\gamma\to 0$). At the same time, in the presence of dissipation, Eq. (25c) does not coincide with the equation for the classical particle in a medium with linear friction force[12] $\boldsymbol{F}_{fr}=-\gamma m\dot{\boldsymbol{r}}$. A particular case of a "free" particle, where[13] $\hat{\Theta}=0$, is the exception.

We then obtain the constitutive equation for the medium formed by an ensemble of such oscillators distributed in space.[14] In this case, as was mentioned in Sec. II, the density matrix can conveniently be normalized to the number of elementary oscillators in a unit volume, namely, $N=\Sigma_n\rho_{nn}$. In this case, the expression for the macroscopic polarization $\boldsymbol{P}$ of the medium is obtained from Eq. (25a) by the replacements $\bar{\boldsymbol{d}}\to\boldsymbol{P}$ and $e^2/m\to\omega_p^2/4\pi$ (here, $\omega_p=\sqrt{4\pi e^2N/m}$ is the plasma frequency). It should be born in mind that in a condensed medium, the average macroscopic field $\boldsymbol{E}$ is different from the acting field $\boldsymbol{E}_a$. In terms of the Lorentz-Lorentz model [17], where $\boldsymbol{E}_a=\boldsymbol{E}+4\pi\boldsymbol{P}/3$, this feature leads to the replacement of the tensor $\hat{\Theta}$ by the tensor $\hat{\tilde{\Theta}}$ determined by the expression

$$\tilde{\Theta}_{ij} = \Theta_{ij} - \delta_{ij}\frac{\omega_p^2}{3} = \delta_{ij}\left(\frac{\alpha_j}{m} - \frac{\omega_p^2}{3}\right).$$

As a result, we obtain the following equation for the polarization:

$$\ddot{\boldsymbol{P}} + 2\gamma\dot{\boldsymbol{P}} + \gamma^2\boldsymbol{P} + \hat{\tilde{\Theta}}\boldsymbol{P} + \omega_H\boldsymbol{b}\times(\dot{\boldsymbol{P}} + \gamma\boldsymbol{P}) = \frac{\omega_p^2}{4\pi}\boldsymbol{E}. \quad (26a)$$

Relationship between the polarization $\boldsymbol{P}$ and the total-current density $\boldsymbol{J}$ follows from Eq. (24d):

$$\boldsymbol{J} = \dot{\boldsymbol{P}} + \gamma\boldsymbol{P}. \quad (26b)$$

In the case of a constant magnetic field and monochromatic electric field $\boldsymbol{E}=\text{Re}\,\tilde{\boldsymbol{E}}e^{-i\omega t}$, the electrodynamic properties of the medium are characterized by the complex dielectric permittivity tensor $\hat{\varepsilon}(\omega)$, which is determined by the relationship $\boldsymbol{D}_{eff}=\boldsymbol{E}+4\pi\int_{-\infty}^{t}\boldsymbol{J}(\tau)d\tau=\text{Re}\,\hat{\varepsilon}(\omega)\tilde{\boldsymbol{E}}e^{-i\omega t}$ (see, e.g., [18]). In the weak-damping limit, where $\omega\gg\gamma$, a universal (i.e., independent of the values of the coefficients in the equation) relationship between the Hermitian and anti-Hermitian com-

[10]For proving, it is convenient to use the rule of sums for the oscillator forces (see, e.g., [15,24]): $\sum_k\Omega_{kn}|(\boldsymbol{d}\cdot\boldsymbol{a})_{nk}|^2=e^2\hbar/2m$ for any number n, where $\boldsymbol{a}$ is an arbitrary unit vector.

[11]This is natural for a harmonic oscillator (see, e.g., [4]), and a constant magnetic field is not important in this case since the equidistancy of the energy spectrum is retained in the nonrelativistic limit when this field is present.

[12]Probably, this is the manifestation of a fundamental feature of the correct quantum model of relaxation, which assumes that the off-diagonal density-matrix elements "die" as the matrix tends to some diagonal matrix corresponding to an equilibrium with the reservoirs [5,6,16].

[13]For the particular case $\hat{\Theta}=\omega_H=0$ and a monochromatic electric field, the result obtained in [9] for the conductivity of an ensemble of noninteracting particles follows from Eq. (25c).

[14]This model is important, in particular, for the theory of quantum dots in magnetic fields (see, e.g., [25]).

ponents of the tensor $\hat{\varepsilon}(\omega)\equiv\varepsilon_{ij}(\omega)=\varepsilon_{ij}^{H}+\varepsilon_{ij}^{aH}$ (hereafter, $i,j=x,y,z$) follows from Eqs. (26a) and (26b):

$$\varepsilon_{ij}^{aH}=i\frac{\gamma}{\omega}\frac{\partial}{\partial\omega}[\omega(\varepsilon_{ij}^{H}-\delta_{ij})], \tag{26c}$$

where $\varepsilon_{ij}^{H}=\varepsilon_{ij}(\gamma\to 0)$. Condition (26c) guarantees the positive dissipation in the case of the positive energy of an electromagnetic field in the medium. Note that if we determined the complex dielectric permittivity through a relationship of the electric field with the vector $\boldsymbol{D}=\boldsymbol{E}+4\pi\boldsymbol{P}$ rather than the vector $\boldsymbol{D}_{eff}$, then from Eq. (26a) we could obtain—instead of Eq. (26c)—the expression $\varepsilon_{ij}^{aH}=i\gamma\partial(\varepsilon_{ij}^{H}-\delta_{ij})/\partial\omega$, which admits a physically meaningless (in this case) negative dissipation, as well. In particular, for the circular polarization of the rf field[15] at $\boldsymbol{E}\perp\boldsymbol{b}\|\boldsymbol{z}_0$ and $\alpha_x=\alpha_y$, it can be verified that the latter relationship corresponds to the negative dissipation in the frequency range $\omega<\omega_H/2$. This fact confirms once again that the total current in the system should be determined namely in accordance with Eq. (8).

Thus, arbitrariness of choosing the integration path in Eq. (19b) does not lead to physically different or incorrect results, at least for the considered simple model, as well as in the "fast" relaxation limit. Thus, within the framework of the phenomenological approach, we can choose the integration path in Eq. (19b) leading to the simplest result for a particular symmetry of the vector field $\boldsymbol{A}(\boldsymbol{r},t)$ and/or basis functions $\psi_k(\boldsymbol{r})$.

VII. LINEAR RESPONSE OF A MULTILEVEL SYSTEM

As another example of application of general equations obtained for the gauge-invariant relaxation operator in Sec. V, we consider the linear response of a multilevel system, in which the relaxation of the off-diagonal elements of the density matrix in the absence of an external field is described by operator (6a). Consider for simplicity a potential electric field $\boldsymbol{E}(\boldsymbol{r},t)$ specified by two sets of electrodynamic potentials

$$\boldsymbol{A}_1=-c\int_{-\infty}^{t}\boldsymbol{E}(\boldsymbol{r},\tau)d\tau,\quad \varphi_1=0;\quad \boldsymbol{A}_2=0, \tag{27}$$

$$\varphi_2=-\int_0^r\boldsymbol{E}(\boldsymbol{\xi},t)d\boldsymbol{\xi}$$

[we assume that the field is "switched on" for $t\to-\infty$, i.e., $\boldsymbol{E}(t\to-\infty)\to 0$]. The initial density matrix is assumed to be diagonal, such that $\rho_{m\neq n}(t\to-\infty)\to 0$. Within the framework of the linear approximation with respect to the field $\boldsymbol{E}$, the populations in the system are not perturbed, i.e., $\rho_{nn}=N_n=\text{const}$.

The matrix element of the operator of particle-field interaction specified by the vector potential is determined by the following relationship which is valid in the case $\text{rot}\,\boldsymbol{A}=0$:

$$\langle k|\frac{\hat{\boldsymbol{p}}\boldsymbol{A}+\boldsymbol{A}\hat{\boldsymbol{p}}}{2m}|n\rangle=i\Omega_{kn}\left[\int_0^r\boldsymbol{A}(\boldsymbol{\xi},t)d\boldsymbol{\xi}\right]_{kn}. \tag{28a}$$

Equation (28a) follows directly from the definition of the eigenfunctions of the unperturbed Hamiltonian $\psi_{k,n}E_{k,n}=\hat{H}_0\psi_{k,n}$. If $\boldsymbol{A}$ is an arbitrary unit vector, then from Eq. (28a) we obtain the known expression for the matrix element of the momentum operator,

$$\frac{\boldsymbol{p}_{kn}}{m}=i\Omega_{kn}\boldsymbol{r}_{kn}=\frac{i}{e}\Omega_{kn}\boldsymbol{d}_{kn}. \tag{28b}$$

Let us make use of the relaxation operator specified by Eq. (18a), in which the "initial" operator $\hat{R}_0$ corresponds to Eq. (6a). Within the framework of the linear approximation, one can easily obtain the following relationship for the off-diagonal elements of the relaxation term in the density-matrix equation:

$$-\frac{i}{\hbar}(\hat{R}_A\rho)_{mn}\approx-\gamma_{mn}[(1-iu_0)\rho]_{mn}\approx-\gamma_{mn}\left\{\rho_{mn}-\frac{ie}{c\hbar}\left[\int_0^r\boldsymbol{A}(\boldsymbol{\xi},t)d\boldsymbol{\xi}\right]_{mn}(N_n-N_m)\right\} \tag{28c}$$

[the function u_0 is given by Eq. (18b)]. Then, using Eqs. (27), (28a), and (28c), from Eq. (1) for the density matrix we obtain the following linearized equations for the off-diagonal elements of the density matrix for different gauges of electromagnetic potentials:

$$\dot{\rho}_{1mn}+i(\Omega_{mn}-i\gamma_{mn})\rho_{1mn}=\frac{e(\Omega_{mn}-i\gamma_{mn})}{\hbar}\int_{-\infty}^{t}\left[\int_0^r\boldsymbol{E}(\boldsymbol{\xi},\tau)d\boldsymbol{\xi}\right]_{mn}d\tau(N_n-N_m), \tag{29a}$$

$$\dot{\rho}_{2mn}+i(\Omega_{mn}-i\gamma_{mn})\rho_{2mn}=\frac{ie}{\hbar}\left[\int_0^r\boldsymbol{E}(\boldsymbol{\xi},t)d\boldsymbol{\xi}\right]_{mn}(N_n-N_m). \tag{29b}$$

Integrating Eq. (29b), we obtain

$$\rho_{2mn}=\frac{ie}{\hbar}(N_n-N_m)e^{-i(\Omega_{mn}-i\gamma_{mn})t}\int_{-\infty}^{t}e^{i(\Omega_{mn}-i\gamma_{mn})\tau}\left[\int_0^r\boldsymbol{E}(\boldsymbol{\xi},\tau)d\boldsymbol{\xi}\right]_{mn}d\tau. \tag{30a}$$

Integrating Eq. (29a) in a similar way and transforming the result after integration by parts, we arrive at

[15] If the rotation direction of the electric-field vector coincides with the direction of cyclotron rotation of an electron.

$$\rho_{1mn} = \rho_{2mn} - \frac{ie}{\hbar}(N_n - N_m)\int_{-\infty}^{t}\left[\int_0^r \boldsymbol{E}(\boldsymbol{\xi},\tau)d\boldsymbol{\xi}\right]_{mn} d\tau. \tag{30b}$$

From Eq. (30b) it follows that $\boldsymbol{d}_{nm}(\rho_{2mn}-\rho_{1mn})=0$ for any pair of levels, i.e., not only the total dipole moment but also the partial dipole moments for separate transitions are preserved after the gauge transformation.

Generally speaking, the gauge invariance of the expression for the dipole moment $\bar{\boldsymbol{d}}$ should certainly ensure gauge invariance for the system current $\bar{\boldsymbol{j}}=\hat{\boldsymbol{j}}\rho$, as well. However, we will present the calculation of the current by the method of direct averaging of the corresponding operator (this is important for the subsequent discussion of features of the gauge-invariant description of a two-level system).

Matrix elements of the derivative of the dipole moment and the total current (with allowance for dissipation, these quantities, as is shown in Sec. II, are not identical) are obtained from general Eqs. (5), (8), and (28b),

$$(\dot{\boldsymbol{d}})_{mn} = (i\Omega_{mn} - \gamma_{mn})\boldsymbol{d}_{mn} - \frac{e^2}{mc}\boldsymbol{A}_{mn}, \quad \boldsymbol{j}_{mn} = (\dot{\boldsymbol{d}})_{mn} + \gamma_{mn}\boldsymbol{d}_{mn}$$

$$= i\Omega_{mn}\boldsymbol{d}_{mn} - \frac{e^2}{mc}\boldsymbol{A}_{mn}. \tag{31}$$

From Eq. (30b) we obtain the condition $\gamma_{mn}\boldsymbol{d}_{nm}(\rho_{2mn}-\rho_{1mn})=0$ for any pair of levels, which implies that, as was mentioned in Sec. III, the difference between the total current and the derivative of the dipole moment is gauge invariant, i.e., $\bar{\boldsymbol{j}}_1-\dot{\bar{\boldsymbol{d}}}_1=\bar{\boldsymbol{j}}_2-\dot{\bar{\boldsymbol{d}}}_2$. With allowance for the properties $\Omega_{mn}=-\Omega_{nm}$ and $\gamma_{mn}=\gamma_{nm}$, from Eqs. (30b) and (31) we find

$$\bar{\boldsymbol{j}}_1 = \bar{\boldsymbol{j}}_2 + \boldsymbol{G}, \quad \boldsymbol{G} = \frac{e^2}{m}N_n\int_{-\infty}^{t}\boldsymbol{E}_{nn}(\tau)d\tau - i\Omega_{mn}\boldsymbol{d}_{nm}(\rho_{1mn}-\rho_{2mn}). \tag{32a}$$

Using Eq. (30b), we transform the second term in the expression for the factor $\boldsymbol{G}$. As a result, we arrive at the relationship

$$\boldsymbol{G} = \frac{e^2}{m}N_n\int_{-\infty}^{t}\boldsymbol{E}_{nn}(\tau)d\tau - \frac{2e}{\hbar}N_n\Omega_{mn}\boldsymbol{d}_{nm}\int_{-\infty}^{t}\left[\int_0^r \boldsymbol{E}(\boldsymbol{\xi},\tau)d\boldsymbol{\xi}\right]_{mn} d\tau. \tag{32b}$$

For further analysis, it is convenient to make use of the identity which generalizes the known rule of sums for the oscillator forces[16] and can be proved in a similar way,

$$e\sum_k \Omega_{kn}\boldsymbol{d}_{nk}\zeta_{kn} = \frac{e^2\hbar}{2m}(\boldsymbol{\nabla}\zeta)_{nn}, \tag{33}$$

where ζ is an arbitrary differentiable scalar function of the coordinates. In view of identity (33), the factor $\boldsymbol{G}$ turns out to be equal to zero, i.e., the current is preserved after the gauge transformation, as follows from Eqs. (11).

VIII. DESCRIPTION FEATURES OF A TWO-LEVEL SYSTEM

We note that identity (33) cannot be fulfilled if all numbers n and k, except for the isolated pair of levels, are neglected. Indeed, for a two-level system and a homogeneous (as is assumed for simplicity) electric field, it follows from Eq. (32b) that

$$\boldsymbol{G} = (N_1+N_2)\frac{e^2}{m}\int_{-\infty}^{t}\boldsymbol{E}(\tau)d\tau - (N_1 - N_2)\frac{2e}{\hbar}\Omega_{21}\boldsymbol{d}_{12}\left[\boldsymbol{d}_{21}\int_{-\infty}^{t}\boldsymbol{E}(\tau)d\tau\right] \neq 0.$$

Thus, within the framework of the two-level approximation, the condition of gauge invariance of the current specified by a standard operator is violated, even in the absence of dissipation. However, the gauge invariance of the expression for the dipole moment of the system remain intact.[17] This contradiction arises from the fact that the matrix elements of the operator for the derivative of the dipole moment $(\dot{\boldsymbol{d}})_{mn}$ in the case $\boldsymbol{A}\neq 0$ satisfies the condition $\boldsymbol{d}_{nm}\dot{\rho}_{mn}=(\dot{\boldsymbol{d}})_{nm}\rho_{mn}$ during summation over all levels of the "complete" system, but not for any pair of levels in separate. In other words, this is due to the fact that the basis formed by the choice of only one pair among the total spectrum of eigenfunctions of the unperturbed Hamiltonian $\hat{H}_0$ is incomplete.[18] In this case, the matrix elements of the operator for the derivative of the dipole moment[19] should probably be determined not from general equations (assuming that the derived basis is complete), but directly from the density-matrix equations within the framework of the "two-level" approximation,

$$\boldsymbol{d}_{21}\dot{\rho}_{12} + \boldsymbol{d}_{12}\dot{\rho}_{21} = (\dot{\boldsymbol{d}})_{21}\rho_{12} + (\dot{\boldsymbol{d}})_{12}\rho_{21} + (\dot{\boldsymbol{d}})_{22}\rho_{22} + (\dot{\boldsymbol{d}})_{11}\rho_{11}. \tag{34}$$

Then, using Eq. (8) for determining the matrix elements of the current operator, we easily obtain a gauge-invariant current for a gauge-invariant dipole moment. It is interesting to note, however, that in the case $\boldsymbol{A}=0$ the corresponding operators coincide in form with the standard one. In this sense, the gauge corresponding to $\boldsymbol{A}=0$ (in the case of a potential field) turns out to be "correct" for a two-level system.

Thus, within the framework of the two-level approximation, the procedures of overdetermining the standard current

[16]The usual rule of sums for the oscillator forces is obtained from Eq. (33) by the replacement $\zeta=(\boldsymbol{a}\cdot\boldsymbol{r})$, where $\boldsymbol{a}$ is an arbitrary unit vector.

[17]This fact was not mentioned in [2] since the authors of that paper studied the gauge invariance of the expression for the probability transition in a two-level system, which is determined, namely, by the dipole moment.

[18]Of course, this is not true for the systems for which a two-level description can be complete, e.g., for transitions between different spin states in a system having only two "allowed" spin projections on the quantizing axis.

[19]Including, in general, the operators of the derivatives of all quantities.

operator can, in principle, be avoided by representing the average current as a function of the average dipole moment using Eq. (8). In particular, for the model considered in the previous section, we obtain $\bar{j}=\dot{\bar{d}}+\gamma_{21}\bar{d}$.

IX. CONCLUSIONS

In this paper, we have shown that the solution of the density-matrix equation, in which the relaxation operator is correctly obtained "from the first principle," ensures gauge invariance for the observables. The reason for the possible "loss" of gauge invariance is the use of simple models of a relaxation operator which is either independent of the external field or allows for the external-field effect within the framework of a simplified gauge-noninvariant model. We propose a recipe for the transformation of the phenomenological relaxation operator in the density-matrix equation to a form that ensures gauge invariance of the solution. Examples illustrating the physical correctness of the results obtained using the proposed procedure are presented.

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