Four-dimensional theory of interaction of two decaying states

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A unified method is used to describe quasiclosed (decaying) two-level systems that interact with electromagnetic radiation. In contrast to the known vector model, the analysis imposes no restrictions on the relaxation constant. An evolution equation that is invariant to the Lorentz group is obtained for the pseudospin. In limiting cases, the equation reduces to the Frenkel' equation for a relativistic spin in an electric or a magnetic field, and to the Bloch optic equation. The exact solution of a four-dimensional equation is obtained under conditions when radiative and ionization broadenings of the transition dominate. An approximate quasistationary solution of the problem is also obtained, with account taken of dephasing collisions, and the range of its validity is indicated.

1. It is known that in optics a two-level system can be set in correspondence with a simple and lucid vector model.[1,2] The problem of resonant interaction of a monochromatic field with an atom reduces then to an investigation of the "pseudospin" precession in "energy space." When relaxation is taken into account, however, a limit is imposed on the validity of this model. We analyze here a four-dimensional vector model that generalizes Ref. 1 to include the case of an arbitrary ratio of the relaxation constants of the states.

Consider an ensemble of two-level atoms in a light field described by the equation for the density matrix ρ (see, e.g., Ref. 3):

$$d\rho/dt+\Gamma\rho=-i(V\rho-\rho V^{+})+Q. \tag{1}$$

The decay of the states is specified by the matrix

$$\Gamma\rho=\begin{pmatrix}\Gamma_1\rho_{11} & \Gamma\rho_{12}\\ \Gamma\rho_{21} & \Gamma_2\rho_{22}-A\rho_{11}\end{pmatrix}, \tag{2}$$

where Γ_1 and Γ_2 are the total widths of the levels 1 and 2, Γ is the polarization relaxation constant, A is the Einstein coefficient, and Q is the excitation function. The interaction of the atom with the light field is specified by vector V, not necessarily Hermitian. We consider the case of a monochromatic wave of amplitude $\vec{\mathscr{E}}$ and frequency ω. We obtain

$$V_{12}=(G-i\gamma)e^{-i\Omega t},\quad V_{21}=(G-i\gamma)e^{i\Omega t},$$
$$G=\vec{\mathscr{E}}\mathbf{d}_{12}/2\hbar,\quad \Omega=\omega-\omega_{12}, \tag{3}$$

where $\mathbf{d}_{12}$ and ω_{12} are the dipole-moment operator and the frequency of the 1–2 transition, G can be chosen to be pure real, and a non-Hermitian part of the Hamiltonian appears, for example, when account is taken of the interaction of the discrete levels via a continuum[4] or in nonadiabatic molecule collision.[5,6] The initial equation (1) takes thus into account a number of relaxation processes: radiative broadening, broadening by collisions in the relaxation-constant model, ionization broadening, and interactions of the states 1 and 2 with the continuum.

Equations (1) can be set in correspondence with the 4-vector equation $a^\mu=(a^0,\mathbf{a})$. To this end, we expand the density matrix in terms of Pauli matrices $\sigma_\mu=(1,\boldsymbol{\sigma})$:

$$\rho=\sigma_\mu a^\mu=a_0-\boldsymbol{\sigma}\mathbf{a},\quad a_0={}^1/_2\,\mathrm{Sp}\,\rho,\quad \mathbf{a}=-{}^1/_2\,\mathrm{Sp}\,\boldsymbol{\sigma}\rho, \tag{4}$$

where a_0 is variable in the general case. Equation (1) is then transformed into

$$\dot{a}^\mu+\Gamma^{\mu\nu}a_\nu=F^{\mu\nu}a_\nu+f^\mu, \tag{5}$$

where $\mu,\ \nu=0,\ 1,\ 2;\ f^\mu=(f^0,f)$ is the level-excitation 4-vector,

$$\Gamma^{\mu\nu}=\begin{vmatrix}\Gamma_+-A/2 & 0 & 0 & -A/2\\ 0 & -\Gamma & 0 & 0\\ 0 & 0 & -\Gamma & 0\\ -A/2 & 0 & 0 & -\Gamma_+-A/2\end{vmatrix},$$

$$F^{\mu\nu}=\begin{vmatrix}0 & -2\gamma & 0 & -\Gamma_-\\ 2\gamma & 0 & -\Omega & 0\\ 0 & \Omega & 0 & 2G\\ \Gamma_- & 0 & -2G & 0\end{vmatrix},\qquad \Gamma_\pm={}^1/_2(\Gamma_1\pm\Gamma_2). \tag{6}$$

The indices are raised and lowered by the diagonal metric tensor $g_{\mu\nu}=\mathrm{diag}(1,-1,-1,-1)$. The relaxation tensor $\Gamma^{\mu\nu}$ is symmetric and $F^{\mu\nu}$ is antisymmetric. Just as any antisymmetric 4-tensor, $F^{\mu\nu}$ can be expressed in terms of the components of two three-dimensional vectors: $F^{\mu\nu}(-\mathbf{E},\mathbf{H})$ (see Ref. 7, §6), where

$$\mathbf{E}=(2\gamma,0,\Gamma_-),\quad \mathbf{H}=(-2G,0,\Omega). \tag{7}$$

The four-dimensional equation (5) coincides at $\Gamma^{\mu\nu}=0, f^\mu=0$ with the Frenkel[1] equation (see Ref. 8, §41), which describes the spin or a relativistic electron that moves quasiclassically in an external electric or magnetic field. In quantum electrodynamics the 4-vector a^μ describes the electron polarization, and the differentiation in the equations of

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motion is with respect to the proper invariant time. This analogy enables us to use the relativistic-theory formalism to analyze four-level systems.

The four-dimensional character of Eq. (5) is essentially due to the term $\mathbf{E}\cdot\mathbf{a} - Aa_z/2$, and Eq. (5) does not reduce to three-dimensional even at $A=0$. If $\Gamma_- - A/2 = \gamma = 0$, the equations for a_0 and $\mathbf{a}$ become independent and the vector $\mathbf{a}$ precesses, with damping, around $\mathbf{H}$. For example, at $A=\gamma=0$ and $\Gamma_1=\Gamma_2$ we obtain

$$\dot{a}_z=[\mathbf{a}\times\mathbf{H}]_z-(a_z-\bar{a}_z)/T_1,$$
$$\dot{\mathbf{a}}_\perp=[\mathbf{a}\times\mathbf{H}]_\perp-\mathbf{a}_\perp/T_2, \quad (8)$$

where $T_1=1/\Gamma_1$, $T_2=1/\Gamma$ are the "longitudinal" and "transverse" relaxation times, and $\bar{a}_z=f_z/\Gamma_1$ is the stationary value of a_z. Equation (8) coincides with the Bloch equation that describes the precession of the nuclear spin $\mathbf{a}$ in a magnetic field $\mathbf{H}$. Similar equations describe in optics the motion of a pseudospin vector in energy space.

2. It is natural to name the four-dimensional description (5) the generalized vector model. If $\Gamma_1 - A \neq \Gamma_2$ or $\gamma\neq 0$, Eq. (5) does not reduce the Bloch's equation (8). Thereis, however, a case of a non-Bloch two-level systems that is nonetheless describable simply enough. This is the case of the relaxation isotropic tensor:

$$\Gamma^{\mu\nu}=\Gamma g^{\mu\nu}. \quad (9)$$

The condition (9) is satisfied if processes that dephase the atomic oscillator are neglected. In particular $\Gamma=\Gamma_+$ when the dominant relaxation mechanism is spontaneous decay or ionization broadening. The substitution $a^\mu = b^\mu e^{-\Gamma t}$ allows us then to rewrite (5) in the simple vector form

$$\dot{b}_0=\mathbf{bE}, \quad \dot{\mathbf{b}}=b_0\mathbf{E}+[\mathbf{b}\times\mathbf{H}]. \quad (10)$$

The square of the 4-vector "length" b^μ is conserved in this case:

$$b_\mu b^\mu=\text{const}. \quad (11)$$

It follows therefore that the end point of the 4-vector b^μ traces an orbit on a pseudosphere surface in our four-dimensional space. In the particular case of a pure initial state (e.g., if only one of the levels is populated at $t=0$), the constant in (11) is zero and the pseudosphere is transformed into a cone.

By virtue of the Lorentz-invariance of (5) and (10), it is possible to make up of the $F^{\mu\nu}$ components two invariant scalars:

$$I_1=\mathbf{H}^2-\mathbf{E}^2, \quad I_2=\mathbf{EH}. \quad (12)$$

Thus, for a complete analysis of the time behavior of the solution (10) it suffices to consider three cases: $I_1\neq 0$, $I_2\neq 0$; $I_1\neq 0$, $I_2\neq 0$; $I_1=I_2=0$. In the first case there exists a Lorentz transformation with parameter $\mathbf{L}$, such that $\mathbf{E}$ becomes parallel to $\mathbf{H}$. In that case $\mathbf{L}$ is given by

$$\mathbf{L}/(1+L^2)=[\mathbf{E}\times\mathbf{H}]/(E^2+H^2). \quad (13)$$

If $E_y=H_y=0$, the system decay probability per unit time, $W=-2a_0$, takes the form

$$W={}^1/_2\beta\{(\beta-\cos\varphi)(\Gamma-E')e^{-(\Gamma-E')t}+(\beta+\cos\varphi)(\Gamma+E')e^{-(\Gamma+E')t}$$
$$+Le^{-\Gamma t}[(\Gamma\sin\varphi-L\beta H')\sin H't-(H'\sin\varphi+L\beta\Gamma)\cos H't]\}, \quad (14)$$

where E' and H' are quantities transformed to a common direction, φ is the angle between their common direction and the z axis, and $\beta=(1-L^2)^{-1/2}$. Expression (14) is characterized by two decay times $\tau_\pm=(\Gamma\pm E')^{-1}$, with $\tau_-\gg\tau_+$ possible. The oscillating term in (14) is proportional to L, and consequently the general condition for the onset of oscillating decay is that (13) be nonzero.

If the vectors $\mathbf{H}$ and $\mathbf{E}$ are perpendicular ($I_1\neq 0$), it is possible to choose a coordinate system in which $\mathbf{E}'=0$ ($I_1>0$) or $\mathbf{H}'=0$ ($I_1<0$). If $I_1>0$, then (10) reduces to a three-dimensional equation such as (8) with $\mathbf{H}'=\mathbf{H}(H^2-E^2)^{1/2}/H$. If $I_1<0$, the system decays with two characteristic times $\tau_\pm=(\Gamma\pm E')^{-1}$, where

$$\mathbf{E}'=(\mathbf{E}/E)(E^2-H^2)^{1/2}, \quad \mathbf{L}=[\mathbf{E}\times\mathbf{H}]/E^2.$$

Let now both invariants (12) be zero. The following conditions are imposed on the interaction parameters: $\Omega=\mp 2\gamma$, $2G=\mp\Gamma_-$. The system decay is not exponential in this case:

$$1-W(t)=({}^1/_2E^2t^2+\Gamma_-t+1)e^{-\Gamma t},$$

where $W(t)$ is the decay probability by the instant of time t. Actually, the vanishing of both invariants I_1 and I_2 means equality, in the radiation field, of quasienergy levels whose complex energies differ by a quantity $R=(I_1+4iI_2)^{1/2}$ that vanishes at $I_1=I_2=0$. A similar situation arises in the case of overlapping nuclear resonances.[9]

3. In the general case of anisotropic relaxation, when (9) does not hold, Lorentz transformations do not simplify Eqs. (5) substantially. We consider therefore an approximate analysis of Eqs. (5).

We assume that a_0 varies slowly with time, and that the components of $\mathbf{a}$ can attune themselves to the variation of a_0. It suffices therefore to consider in the equations for $\mathbf{a}$ only the stationary regime. Assuming that $\dot{\mathbf{a}}=0$ ($\Gamma_{ij}t\gg 1$), we have

$$(\Gamma_{ij}+e_{ijk}H_j)a_k=E_ia_0; \quad i,j,k=1,2,3, \quad (15)$$

where $\Gamma_{ij}=\mathrm{diag}(\Gamma,\Gamma,\Gamma_+)$ is a three-dimensional relaxation tensor ($A=0$) which is anisotropic, and e_{ijk} is a unit antisymmetric tensor.

We write the solution of (15) in the form

$$a_i=B_{ik}(\mathbf{H})E_ka_0. \quad (16)$$

It follows from (15) that the components $B_{ik}(\mathbf{H})$ are subject to the relations

$$B_{ik}(\mathbf{H})=B_{ki}(-\mathbf{H}), \tag{17}$$

which are analogous to the properties of the conductivity tensor of a conductor in a magnetic field.[10] Resolving B into symmetric and antisymmetric parts, we get

$$a_i/a_0=S_{ik}E_k+[\mathbf{E}\times\mathbf{A}]_i, \tag{18}$$

where the symmetric part is

$$S_{ik}=\frac{1}{\Delta}(\Gamma_{ik}^{-1}\|\hat{\Gamma}\|+H_iH_k), \quad \Delta=\|\hat{\Gamma}\|+\Gamma_{ij}H_iH_j, \tag{19}$$

and the axial vector

$$A_i=-\frac{1}{\Delta}\Gamma_{ik}H_k \tag{20}$$

is equivalent to the antisymmetric part of B. Since $\hat{\Gamma}$ is diagonal, the term $\mathbf{E}\times\mathbf{A}$ leads to the appearance of a component of $\mathbf{a}$ that is perpendicular to $\mathbf{E}$ and is proportional to $\mathbf{H}$. Using (18), we find the solution of the equation of a_0:

$$2a_0=1-W(t)=\exp[-(1-\delta)\Gamma_+t],$$

where $\delta=S_{ik}E_iE_k/\Gamma_+$. The condition that a_0 varies slowly will be satisfied if $1-\delta\ll1$, i.e.,

$$\Gamma_+-\frac{(\mathbf{EH})^2+E_x^2\Gamma\Gamma_++E_z^2\Gamma^2}{\Gamma^2\Gamma_++\Gamma H_x^2+\Gamma_+H_z^2}\ll\Gamma_+. \tag{21}$$

Expression (21) determines the validity of the quasistationary approximation in the general case when inelastic collisions (the difference between Γ and Γ_+) are taken into account and at an arbitrary magnitude of the level interaction.

In the simplest case $E_x=0$ ($\gamma=0$), i.e., when, e.g., there is no interaction of the discrete state via the continuum, (21) leads to the conditions

$$1-E_z^2/\Gamma_+^2\ll1, \quad \varkappa/(1+\varkappa+\Omega^2/\Gamma^2)\ll1, \tag{22}$$

where $\varkappa=4G^2/\Gamma\Gamma_+$ is the saturation parameter of the discrete transition. The first of the conditions means a substantial difference between the level widths, and the second means smallness of or a large detuning compared with the homogeneous linewidth. At $\Gamma_1\gg\Gamma_2$ Eq. (22) leads to the results of Ref. 11 for the probability of resonant photoionization of atoms.

Another simple example is the case of isotropic relaxation $\Gamma_{ij}=\Gamma\delta_{ij}$. It is easy to show that the decay probability W takes the form

$$\dot{W}=2\Gamma\left\{1-\frac{E^2}{\Gamma^2}+\frac{1}{\Gamma^2}\frac{[\mathbf{E}\times\mathbf{H}]^2}{\Gamma^2+H^2}\right\}\exp[-(1-\delta)\Gamma t]. \tag{23}$$

Fano's result[12] corresponds to the case $\Gamma_1\gg\Gamma_2$, G and is valid, according to (23), for times $t\ll\Gamma_1/G^2$. Let us determine the range of the parameters $H_{x,z}$ (Ω,G) for which the quasistationary regime is realized. To this end we align the z axis with the vector $\mathbf{E}$ by rotating the coordinate system through an angle $\varphi=\arccos E_z/E$. Then, putting $1-\delta=\alpha$, where $\alpha\ll1$, we obtain

$$\tilde{\Omega}^2(1-E^2/\Gamma^2-\alpha)+4\tilde{G}^2\leqslant(\alpha-1+{}^2/\Gamma^2)\Gamma^2. \tag{24}$$

Here $\tilde{\Omega}$ and $\tilde{G}$ are the deviation from resonance and the interaction matrix element in the coordinate frame rotated through the angle φ. It follows from (24) that a nonstationary regime exists only at $\alpha\geqslant1-E^2/\Gamma^2$. This is natural, in as much as the quantity $1-E^2/\Gamma^2$ determines the degree of interference between the transitions to the continuum from levels 1 and 2, and the system lifetime cannot be longer than $[\Gamma(1-E^2/\Gamma^2)]^{-1}$. Thus, the region of the values of $\tilde{\Omega}$ and $\tilde{G}$ is bounded by the hyperbola

$$\frac{\tilde{G}^2}{a^2}-\frac{\tilde{\Omega}^2}{b^2}=1, \quad a=\frac{\Gamma}{2}\left(\alpha-1+\frac{E^2}{\Gamma^2}\right)^{1/2}, \quad b=\Gamma.$$

An error has crept into Eqs. (5.2) and (5.9) of Ref. 4. When this is allowed for, the corresponding inequality of Ref. 9 will coincide with (24).

The strictly stationary regime of Eqs. (5) is possible at a nonzero excitation 4-vector f^μ. The components f_0 and f_z take into account the population of the levels 1 and 2, while f_x and f_y take the polarization transfer into account.

We note in conclusion that the four-dimensional approach is fully equivalent to the density-matrix formalism, but offers a number of advantages due to its clarity and to the inclusion of the decay (ionization) probability $\dot{W}=-2\dot{a}_0$ as an independent variable in the equations. Obviously, the generalized vector model is valid not only for optical processes, but also for two-level systems of any type.

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