Nonlinear effects due to a cascade of coherence in probe-field spectroscopy

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The transfer of coherence of states is important to nonlinear saturation effects. Incorporating the transfer of coherence may lead to not only substantial changes in nonlinear interference effects but also changes in level splitting.

Radiative (spontaneous) cascades between excited atomic states are well known. They can play an extremely important role in many problems, including problems in saturation spectroscopy. In general, one speaks in terms of spontaneous transitions of particles when the result of the transition is a change in the numbers of particles in the combining levels, m and n. For example, it is because of cascade transitions that the first Einstein coefficient A_{mn} appears in the factor $(\Gamma_m - A_{mn} + \Gamma_n)/\Gamma_m$ in the saturation parameter (Γ_m and Γ_n are the decay rates of levels m and n), and the change in the population of the lower level, n, due to the external field is proportional to the factor $1 - A_{mn}/\Gamma_m$ (Refs. 1 and 2, for example).

Important to several phenomena are not only "cascades of particles" but also "cascades of coherence," i.e., a transfer to a lower level of a coherence between magnetic sublevels which exists in the upper state. This is the situation in optical orientation,[3] the Hanle effect,[4–6] nonlinear Faraday rotation,[7] and the polarization of cascade fluorescence.[5] To the best of our knowledge, a cascade of coherence has not been considered in discussions of nonlinear phenomena in probe-field spectroscopy. Our purpose in this letter is to analyze possible effects of this sort.

We consider the simple case of a transition between levels m and n with total angular momenta $J_m = J_n = 1$. A strong, resonant monochromatic field, linearly polarized along the z axis, causes transitions with $\Delta M = 0$ (the solid arrows in Fig. 1; the transition with $M = 0$ is forbidden). We assume that the probe field $\mathbf{E}_\mu$ is linearly polarized and is perpendicular to the strong field $\mathbf{E}$. The probe field induces transitions with $\Delta M = \pm 1$ (the wavy arrows in Fig. 1, shown for $\Delta M = 1$). It is easy to see that in both states, m and n, the fields $\mathbf{E}$ and $\mathbf{E}_\mu$ induce a correlation (a coherence) between magnetic sublevels. This correlation is described by the density-matrix elements $\rho_m(M,M')$ and $\rho_n(M,M')$; it is represented by the arcing arrows in Fig. 1. Formally, a cascade of coherence is manifested by the onset of a coherence in the lower state, n, at a rate given, in general, by (see, for example, Ref. 1, § 2, Ref. 2):

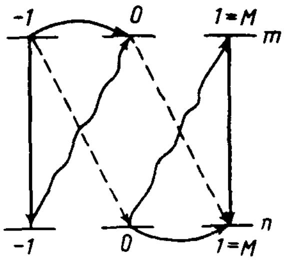

FIG. 1.

$$A_{mn}\sum_{M_1M_1'\sigma}\langle J_nM1\sigma|J_mM_1\rangle\langle J_nM'1\sigma|J_mM_1'\rangle\rho_m(M_1,M_1'). \qquad (1)$$

Here $\langle ...|...\rangle$ are Clebsch–Gordan coefficients, and $\sigma=0,\pm 1$. In the particular case $J_m=J_n=1$, the coherences $\rho_m(0,-1)$ and $\rho_m(1,0)$ (Fig. 1) induce $\rho_n(1,0)$ and $\rho_n(0,-1)$, and the proportionality factors in Eq. (1) turn out to be equal to $A_{mn}/2$. A spontaneous transition of coherence is depicted by the two dashed arrows in Fig. 1.

In the relaxation-constant model, the system of equations for the density-matrix elements $\rho_m(M,M')$, $\rho_n(M,M')$, and $\rho_{mn}(M,M')$ is[1,2]

$$\begin{gathered}(\Gamma-i\Omega_\mu)\rho_{mn}(1,0)-iG_1\rho_n(1,0)=iG_{10}^\mu[\rho_n(0,0)-\rho_m(1,1)],\\ -iG_1^*\rho_{mn}(1,0)+(\Gamma_n-i\epsilon)\rho_n(1,0)=-iG_{10}^\mu\rho_{nm}(1,1)+\tfrac{1}{2}A_{mn}\rho_m(0,-1);\end{gathered} \qquad (2)$$

$$\begin{gathered}(\Gamma-i\Omega_\mu)\rho_{mn}(0,-1)+iG_{-1}\rho_m(0,-1)=iG_{0-1}^\mu[\rho_n(-1,-1)-\rho_m(0,0)],\\ iG_{-1}^*\rho_{mn}(0,-1)+(\Gamma_m-i\epsilon)\rho_m(0,-1)=iG_{0-1}^\mu\rho_{nm}(-1,-1);\end{gathered} \qquad (3)$$

$$\begin{gathered}\Omega_\mu=\omega_\mu-\omega_{mn},\ \Omega=\omega-\omega_{mn},\ \epsilon=\Omega_\mu-\Omega,\\ G_1=-G_{-1}=dE_0/2\sqrt{6}\hbar,\ G_{10}^\mu=G_{0-1}^\mu=-dE_\mu/2\sqrt{6}\hbar.\end{gathered} \qquad (4)$$

Here ω_{mn} is the Borovskiĭ frequency, ω and ω_μ are the frequencies of the strong and probe fields, E_0 is the z component of the strong field, E_μ is the circular component of the probe field, Γ is the polarization relaxation constant, and d is the reduced dipole matrix element for the $m-n$ transition.

On the right sides of Eqs. (2) and (3) are the populations of the sublevels, $\rho_m(M,M)$ and $\rho_n(M,M)$, and the polarizations $\rho_{nm}(M,M)$ formed by the strong field. Since the equations for these quantities and the corresponding solutions are well known (Refs. 1 and 2, for example), we will not reproduce them here. In the second equation of system (2), the term on the right side with A_{mn} describes the cascade of coherence in which we are interested in this letter. Equations with the other circular component of the probe field are similar in form and need not be written out.

According to qualitative considerations regarding the manifestation of a cascade of coherence, system of equations (3) is independent, and its solution enters the right side of system (2). Using the solutions of Eqs. (2) and (3), we can write the following expression for the work performed by the probe field,

$$P_\mu = 2\hbar\omega_\mu \mathrm{Re}\, i \sum_{MM'} G^\mu_{MM'} \rho_{nm}(M',M)$$

$$= 4\hbar\omega_\mu \mathrm{Re}\left\{ \left| G^\mu_{0-1} \right|^2 \frac{(\Gamma_m - i\epsilon)[\rho_n(-1,-1) - \rho_m(0,0)] - iG_{-1}\rho_{nm}(-1,-1)}{(\Gamma - i\Omega_\mu)(\Gamma_m - i\epsilon) + |G_{-1}|^2} \right.$$

$$+ \left| G^\mu_{10} \right|^2 \frac{(\Gamma_n - i\epsilon)[\rho_n(00) - \rho_m(1,1)] - iG_1\rho_{nm}(1,1)}{(\Gamma - i\Omega_\mu)(\Gamma_n - i\epsilon) + |G_1|^2} + G^{\mu *}_{10} G^\mu_{0-1} G_1 G^*_{-1}$$

$$\left. \times \frac{A_{mn}}{2} \frac{\rho_n(-1,-1) - \rho_m(0,0) + \dfrac{\Gamma - i\Omega_\mu}{\Gamma + i\Omega}[\rho_n(1,1) - \rho_m(1,1)]}{[(\Gamma - i\Omega_\mu)(\Gamma_m - i\epsilon) + |G_{-1}|^2][(\Gamma - i\Omega_\mu)(\Gamma_n - i\epsilon) + |G_1|^2]} \right\}. \qquad (5)$$

The terms in curly brackets on the first and second lines are standard terms for the absorption (or intensification) spectrum of the probe field in three-level systems. Serving as these triplets of levels in our case are $jM = m-1$, $m0, n-1$ and $m1$, $n0$, $n1$, as discussed previously. The difference from the expressions derived in the model of nondegenerate states[1,2] is basically that here two of the three levels in each triplet are magnetic sublevels of the same state. The third term in (5) is due to a cascade of coherence; it has some unusual properties. We first note the product of different MM' components of a perturbation, instead of the standard square moduli. Admittedly, by virtue of (4) we have

$$G^{\mu *}_{10} G^\mu_{0-1} G_1 G^*_{-1} = -|G^\mu_{10}|^2 |G_1|^2,$$

but this simplification arises because of the particular properties of the $J_m = J_n = 1$ transition. The combinations of matrix elements of the field perturbation written above emphasize the interference nature of the effects which stem from a cascade of coherence: The component G^μ_{10} performs work on the polarization induced by the other component, G^μ_{0-1}.

The spectral properties of the "cascade term" (its resonances) are governed by its denominator, which is the product of the denominators of the first two terms. The work performed by the probe field, P_μ, can behave in various ways as a function of its frequency ω_μ, depending on the relations among relaxation constants, on the values of the static magnetic field, on the orientations of the wave vectors, on the role played by the Doppler effect, and on other circumstances (the Doppler frequency shift leads to the replacements $\Omega_\mu \to \Omega_\mu - \mathbf{k}_\mu \cdot \mathbf{v}$, $\Omega \to \Omega - \mathbf{k} \cdot \mathbf{v}$). We will not discuss that question in this letter; we will simply point out that the integral absorption (integrated over ω_μ) due to the cascade of coherence is zero. In this regard, the effect is similar to nonlinear interference effects.[1,2,8–10] Let us take a closer look at this similarity.

According to our theory,[1,2,10] nonlinear effects of probe-field spectroscopy arise for three fundamental reasons: a field-induced splitting of levels, a field-induced change in populations, and nonlinear interference effects. This classification is based on general properties of systems of equations like (2) or (3): A splitting of levels is determined by the determinant of the matrix of the system of equations. The change in populations appears on the right side of the equation for the polarization of an allowed transition. The nonlinear interference effects arise from the right side of the equation for the polarization of a forbidden transition. From this standpoint, the cascade term in (5) is a typical nonlinear interference effect [see system (2)]. There is a distinction from other nonlinear interference effects: The latter are caused by the polarization of an optically allowed adjacent transition induced by the strong field [the terms with $\rho_{nm}(-1,-1)$ and $\rho_{nm}(1,1)$ in (5)], while the cascade nonlinear interference effect is generated by a cascade of coherence on a forbidden transition.

In order of magnitude, the cascade nonlinear interference effect in expression (5) differs from the first two terms by a factor A_{mn}/Γ_m, which may be small, but it may also be on the order of one.

In a sense, the case $J_m=J_n=1$, with orthogonal polarizations $\mathbf{E}$ and $\mathbf{E}_\mu$, is simplistic. Manifestations of a cascade of coherence also exist at other values of J_m and J_n and for other field polarizations. In addition to giving rise to a nonlinear interference effect, a cascade of coherence can influence the splitting of levels. In the case $J_m=J_n=1/2$, with orthogonal linear polarizations of the fields $\mathbf{E}$ and $\mathbf{E}_\mu$, for example, the off-diagonal elements of the density matrix for forbidden and allowed transitions are related by a fourth-order system of equations. There is no cascade nonlinear interference effect in this case, but the cascade of coherence does make a contribution to the determinant of the system. It turns out to be

$$(\Gamma-i\Omega_\mu)[\Gamma-i(\epsilon-\Omega)](\Gamma_m-i\epsilon)(\Gamma_n-i\epsilon)+2(\Gamma-i\epsilon)(\Gamma_m+\Gamma_n-A_{mn}/3-2i\epsilon)|G|^2, \tag{6}$$

where the term $A_{mn}/3$ reflects the role played by the cascade of coherence between the magnetic sublevels $M=\pm 1/2$.

In general, a cascade of coherence also makes contributions to the determinant of the corresponding system of equations (i.e., to the splitting of levels) and to the formation of cascade nonlinear interference effects.

We recall in conclusion that the discussion above applies to a coherence on an optically forbidden transition (a correlation between magnetic sublevels of one stationary state). There is the possibility in principle of a corresponding cascade of coherence between levels for which a dipole transition is allowed. The equation for the density-matrix element $\rho_{nn'}(M,M')$ is (we are omitting field-perturbation terms)

$$\left(\frac{d}{dt}+\Gamma_{nn'}\right)\rho_{nn'}(M,M')=\sqrt{A_{n_1n}A_{n_1'n'}}\sum_{M_1M_1'\sigma}\rho_{n_1n_1'}(M_1,M_1')$$
$$\times\langle J_nM1\sigma|J_{n_1}M_1\rangle\langle J_{n'}M'1\sigma|J_{n_1'}M_1'\rangle$$
$$\times\exp[-i(\omega_{n_1n}-\omega_{n_1'n'})t]. \tag{7}$$

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