Dip in time-varying saturated absorption in spectra of homogeneously broadened systems with bleaching

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A theory is derived for nonlinear resonances of a new type: dips in the absorption spectra of the intense pulsed emission from a three-level system and from vibrational-rotational transitions of molecules. These dips form against the background of homogeneously broadened lines and may have widths less than the radiative width. Under certain conditons, a peak forms within a dip.

In typical situations in nonlinear laser spectroscopy of gaseous media, dips form in the saturated absorption against the background of Doppler-broadened lines because Bennett holes are burned in the velocity distribution of the level population as intense monochromatic light acts on the working transition. Spectral holes are detected by means of probing fields on the same transition or on an adjacent one. Depending on the experimental approach, one may observe nonlinear resonances of a variety of types,[1] common features of which are an inhomogeneous broadening of the lines and the presence of a probing field which reveals the dip. The width of the resonances is related to the radiative (or collisional) width of the line; only in individual cases, because of particular features of the experiment, will the resonances contain a finer, subradiative structure, which is a consequence of an interference of states[2] or of the exciting and probing fields.

Note in this connection that dips have been detected in radiation-broadened lines in measurements with beams of Rb atoms[4,5] and He atoms[6] and also a vapor of Na atoms.[7] These dips were caused by the limited duration of the incoherent interaction of the atoms with the field under absorption saturation conditions.

In this letter we offer a physical interpretation of the mechanism for the formation of dips in the time-varying absorption against the background of homogeneously broadened lines. We demonstrate that dips of this sort can be observed in a very simple layout for measuring the pulsed absorption of a traveling wave in a gas-filled cell, without the use of probing fields. We find that under certain condtions a peak forms at the center of the dip; this peak is similar to the inverted Lamb dip in nonlinear spectroscopy.[1] We propose a new system, specifically, vibrational–rotational transitions in molecular gases, in which it would be possible to observe dips of an identical physical nature.

To discuss the mechanism for the formation of the dips, we use the example of a three-level system corresponding to the measurements of Refs. 4, 5, and 7. The ground state of the system is state 0, the upper level is 1, and the metastable intermediate level is 2. We assume that the field acts on the $0 \to 1$ resonant transition. The equations for the density matrix of the immobile atom can then be written as follows in the standard approximation:

$$\begin{cases} \dot{\rho}_0 + \gamma_2\rho_0 - (\gamma_{10} - \gamma_2)\rho_1 = \gamma_2 - 2\mathrm{Re}\, iV\rho \\ \dot{\rho}_1 + \gamma_1\rho_1 = 2\mathrm{Re}\, iV\rho \\ \dot{\rho} + (\gamma - i\Omega)\rho = iV(\rho_1 - \rho_0) \end{cases} \quad \left| \begin{array}{l} \rho_0(0) = 1 \\ \rho_1(0) = 0 \\ \rho(0) = 0 \end{array} \right. \qquad (1)$$

$$\rho_0 + \rho_1 + \rho_2 = 1, \qquad \gamma_1 = \gamma_{10} + \gamma_{12}, \qquad V = d_{10}E(t)/2\hbar.$$

Here ρ_j is the population of level j ($j = 0,1,2$), ρ is an off-diagonal element of the density matrix corresponding to the $0 \to 1$ transition, Ω is the deviation of the frequency of the laser light from the frequency of the resonant transition, γ_j ($j = 1,2$) are the decay constants of the levels, γ_{10} and γ_{12} are the rates of decay of level 1 to states 0 and 2, and γ is the rate of decay of the induced dipole moment on the $0 \to 1$ transition. An important feature of (1) is that it incorporates the radiative (collisional) relaxation of the metastable level. We treat the work performed by the field $\mathcal{P}(\Omega,\tau) = 4\,\mathrm{Re}\, iV\rho(\Omega,\tau)$, and the population of the upper level at the end of the pulse, $\rho_1(\Omega,\tau)$, as unknowns. The absorption coefficient for the exciting light on the resonant transition is proportional to the first of these unknowns; the intensity of the spontaneous emission on the $1 \to 0$ and $1 \to 2$ transitions, integrated over the spectrum, is proportional to the second of these unknowns. It follows that the experiment could be carried out in two ways: to detect absorption or to detect fluorescence. If a time sweep is carried out, or if a shutter is used to cut out the last part of the pulse, one can do without probing fields. To simplify the calculations, we assume that the pulse $E(t)$ is square. Numerical calculations of ρ_1 and $\mathcal{P}$, with variations in all the parameters involved, have shown that the shape of the spectrum is the same within 1% in the two cases. Figures 1–3 show some calculated results which characterize the behavior of the lineshape upon a change in the field intensity V, the pulse length τ, and the decay rate of the metastable level, γ_2. We see from these figures that at certain values of V, τ, and

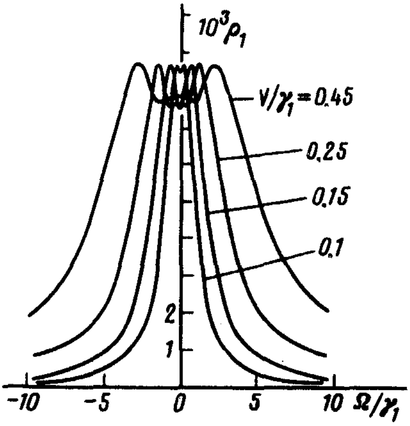

FIG. 1. Lineshape of time-varying saturated absorption (integrated over the fluorescence spectrum) at the end of the pulse (τ) for various values of the saturation parameter V/γ_1. $\tau\gamma_1 = 120$; $\gamma_{10} = \gamma = 0.5\gamma_1$; $\gamma_2 = 0.0004\gamma_1$.

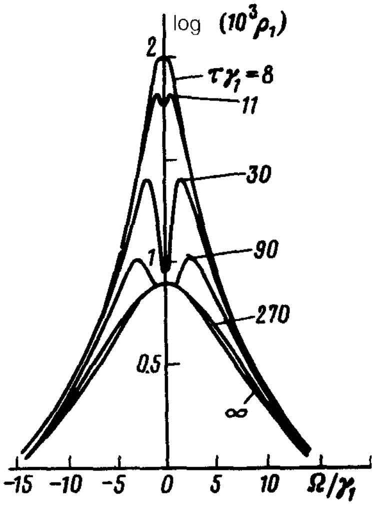

FIG. 2. Shape of the dip, in semilogarithmic scale, for various ratios of the pulse length τ to the lifetime of the upper level, γ_1^{-1}. $V/\gamma_1 = 0.5$; $\gamma_{10} = \gamma = 0.5\gamma_1$; $\gamma_2 = 0.004\gamma_1$.

γ_2 there is a dip in the homogeneously broadened line, and there is a peak in the dip. An analytic solution of the balance equations found from (1) with $\rho = 0$ agrees with the results shown in Fig. 2 within a maximum error $\lesssim 1\%$ for $\tau\gamma_1 \gtrsim 100$. As $\tau\gamma_1$ is reduced to 8, the error increases to 15%. In other words, population effects dominate the formation of the dip. A numerical solution of (1) with a pulse shape $V(t) = at^5 \exp(-\alpha t)$ has shown that there are similar formations in the line in the case of a smooth pulse; in fact, they are slightly more pronounced.

The qualitative behavior of the shape of the dip which can be seen from Figs. 1 and 3, along with the simplicity of the observation arrangement, leads unambiguously to the following mechanism for the formation of the dip and for the formation of the

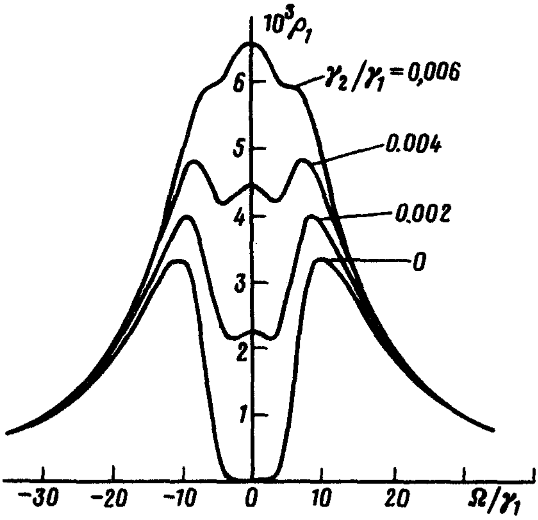

FIG. 3. Shape of the dip and the peak in the dip for various values of the decay rate of the metastable level, γ_2. $V/\gamma_1 = 1$; $\tau\gamma_1 = 120$; $\gamma_{10} = 0.1\gamma_1$; $\gamma = 0.5\gamma_1$.

peak in the dip. Dips of this type form as a result of a competition between the time over which the field acts and the duration of the cycle of a complete transfer of population from the ground state to the metastable level (or its equivalent). To demonstrate the point, we assume that the lifetime of the metastable level is infinitely long. When a long pulse is applied to the system, the ground state will then be emptied, and there will be no absorption at the end of the pulse. If the frequency of the light is tuned away from the center of the line, the cross section for the interaction of the field with the system will decrease, so the rate of transfer of population will also decrease. The result will be a nonvanishing absorption at the end of the pulse; this circumstance will give rise to side peaks in the absorption line. If the frequency deviation is increased further, the absorption will fall to zero, and wings will form on the line, in the standard fashion. As the pulse length is reduced below a certain value which depends on the relaxation constants and the field intensity, the depth of the dip starts to decrease, and ultimately the dip disappears, since the decay of the upper level to the metastable state and thus the bleaching of the medium do not have time to occur. If we now start the relaxation of the metastable level, the absorption of the resonant light at large values of τ will no longer be zero and will be determined by that fraction of the population which percolates through the bottleneck of the metastable level to the ground state. A peak will thus appear at the center of the dip; it will be more prominent, the closer the conditions are to a steady state. A steady state will evidently be reached as the pulse length, the field intensity, and the decay rate of the metastable level are all increased.

The picture drawn here of the mechanism for the formation of a dip naturally leads us to look at other systems with a bottleneck, in particular, vibrational-rotational transitions of molecules. We have carried out a solution of the corresponding equations for the density matrix written in the model of strong rotational-inelastic collisions. For square pulses of length $\tau > 10\gamma_{\text{rot}}^{-1}$, where γ_{rot}^{-1} is the rotational relaxation time, we found an approximate analytic expression for the work performed by the field. That expression leads to a dependence of the line shape V, τ, and the ratio $\gamma_{\text{vib}}/\gamma_{\text{rot}}$ which are essentially the same as the behavior in Figs. 1–3 (γ_{vib}^{-1} is the vibrational relaxation time, which plays the role of the decay time of the metastable level). In the time units which we have adopted here, γ_1^{-1} for the three-level system and γ_{rot}^{-1} for vibrational-rotational transitons of molecules, the characteristic values of the parameters V, τ, and γ_2 (γ_{vib}) are identical in order of magnitude for the two systems.

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