Theory of Laser Saturation Spectroscopy

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A theory of the laser saturation spectroscopy experiments of Hänsch *et al.* is presented which is applicable at high values of the saturating laser beam. Phase- and velocity-changing collisions are taken into account.

Recent experiments by Hänsch, Shahin, and Schawlow[1-3] demonstrate the usefulness of the laser-saturated absorption method as a spectroscopic technique. In their work, laser waves of the same frequency traveling in opposite directions pass through an absorption tube containing the gas to be studied. One traveling wave, the saturator, is of high intensity; the other, the probe, is very weak. When the frequency of the laser is tuned near the frequency of the atomic transition, the probe-beam intensity exhibits a resonance with a width characteristic of the natural width of the atomic transition.

The purpose of this paper is to present a theory of the experiments of Refs. 1–3. The shape of the resonances is derived for the case of a three-level atom in which the two lower levels b and c are connected by the laser fields to a common upper level a. The problem is first solved iteratively in the saturating field strength; in addition to the resonances at the atomic frequencies ω_{ab} and ω_{ac}, there is a "cross-over" resonance at the average of these frequencies, which may[1] or may not[2] be inverted. A noniterative solution is then obtained for the case of well-separated resonances, i.e., for ω_{bc} large compared to the resonance widths; this solution is valid for large saturating field strengths. Phase- and velocity-changing collisions are also included.

The present method is similar to one used previously to calculate the output of a high-intensity gas laser.[4] The laser radiation is treated classically and the atoms quantum mechanically. The laser electric field is assumed to be in the x direction and of magnitude

$$\mathcal{E}(z,t)=E_1\sin(\omega t+\varphi_1-Kz)+E_2\sin(\omega t+\varphi_2+Kz),$$

where $E_1 \gg E_2$; E_1 is the saturating field and E_2 is the probe field. E_1 and E_2 are assumed to be slowly changing functions of z.[5] The problem consists of calculating the macroscopic polarization of the atoms produced by the field, $P(z,t)$, which can then be related to the gain of the probe beam: If

$$P(z,t)=\mathrm{Re}(iA_1e^{-i(\omega t-Kz)}+iA_2e^{-i(\omega t+Kz)}),$$

then Maxwell's equations give an intensity gain of $(K/E_2)\,\mathrm{Im}(A_2)$.

The problem is solved in the rest frame of an atom moving with axial velocity v_z. The time-dependent Schrödinger equation is

$$i\hbar\partial\Psi/\partial t=[H_0+e\mathcal{E}(z,t)x]\Psi.$$

Assume

$$\Psi=a(t)\varphi_a+b(t)\varphi_b+c(t)\varphi_c.$$

The time dependence of the density-matrix components $\rho_{aa}\equiv aa^*$, $\rho_{ab}\equiv ab^*$, etc., is given by

$$\dot\rho_{aa}=\lambda_a-(\gamma_a+\Gamma_a)\rho_{aa}+2\,\mathrm{Re}(iV_{ab}{}^*\rho_{ab})+2\,\mathrm{Re}(iV_{ac}{}^*\rho_{ac}), \tag{1}$$

$$\dot\rho_{bb}=\lambda_b-(\gamma_b+\Gamma_b)\rho_{bb}+\beta_{ab}\rho_{aa}-2\,\mathrm{Re}(iV_{ab}{}^*\rho_{ab}), \tag{2}$$

$$\dot\rho_{ab}=-(\gamma_{ab}+i\omega_{ab})\rho_{ab}+iV_{ab}(\rho_{aa}-\rho_{bb})-i\rho_{bc}{}^*V_{ac}, \tag{3}$$

$$\dot\rho_{bc}=-(\gamma_{bc}+i\omega_{bc})\rho_{bc}-i\rho_{ac}V_{ab}{}^*+i\rho_{ab}{}^*V_{ac}, \tag{4}$$

where $V_{aj}\equiv-\mathcal{P}_{aj}\mathcal{E}(z,t)/\hbar$, and $\mathcal{P}_{aj}$ is the matrix element of the electric dipole moment between states a and j. The phenomenological terms $-\gamma_i\rho_{ii}$, where γ_i is the natural width of level i, have been added to account for the spontaneous decay of level i to all other levels. Here, $\gamma_{ij}\equiv\frac{1}{2}(\gamma_i+\gamma_j)$. The equations for $\dot\rho_{cc}$ and $\dot\rho_{ac}$ are gotten by interchanging b and c in Eqs. (2) and (3). In order to account for the excitation of the atoms, e.g., by electron bombardment, λ_i has been added to $\dot\rho_{ii}$, where λ_i is the number of atoms per cm^3 sec excited to state i with velocity v_z. ρ_{ii} now stands for the number of atoms per cm^3 in state i with velocity v_z, rather than for the probability of state i. Velocity-changing collisions are included by adding a term $-\Gamma_i\rho_{ii}$ to $\dot{\rho}_{ii}$, where Γ_i is the probability per second for an atom in state i to change its axial velocity by collisions.[6] Atoms coming into the velocity range near v_z as a result of collisions with other atoms are neglected. Spontaneous emission from a to b and c is accounted for by adding $\beta_{ab}\rho_{aa}$ to $\dot{\rho}_{bb}$ and $\beta_{ac}\rho_{aa}$ to $\dot{\rho}_{cc}$, where β_{aj} is the spontaneous transition probability per second, $a\to j$. In the rotating wave approximation, we have

$$V_{ab}=i\mathcal{V}_{ab}e^{-i\omega t}(e^{iKz}+\epsilon e^{-iKz}), \tag{5}$$

where

$$\mathcal{V}_{ab}\equiv(-\mathcal{P}_{ab}E_1/2\hbar)\exp(-i\varphi_1),$$

and

$$\epsilon\equiv(E_2/E_1)\exp[-i(\varphi_2-\varphi_1)],$$

with a similar expression for V_{ac}. Here, z is the position of the moving atom and is thus time-dependent. If one substitutes

$$\rho_{ab}\equiv\mathcal{V}_{ab}e^{-i\omega t}(g_{ab}e^{iKz}+\epsilon h_{ab}e^{-iKz})$$

and

$$\rho_{bc}\equiv\mathcal{V}_{ab}{}^*\mathcal{V}_{ac}f_{bc},$$

then in the limit $\epsilon\to 0$, Eqs. (1)–(4) are satisfied by constant values of $\rho_{aa}-\rho_{bb}$, f_{bc}, g_{ab}, and h_{ab}, and can be solved iteratively (in E_1) to yield

$$h_{ab}\cong-\frac{i}{\Delta_{ab2}}\bigg\{n_{ab}-2|\mathcal{V}_{ab}|^2n_{ab}\left(\frac{1}{\gamma_a{}'}+\frac{1}{\gamma_b{}'}-\frac{\beta_{ab}}{\gamma_a{}'\gamma_b{}'}\right)\frac{\gamma_{ab}}{\Delta_{ab1}\Delta_{ab1}{}^*}$$
$$-2|\mathcal{V}_{ac}|^2n_{ac}\left(\frac{1}{\gamma_a{}'}-\frac{\beta_{ab}}{\gamma_a{}'\gamma_b{}'}\right)\frac{\gamma_{ac}}{\Delta_{ac1}\Delta_{ac1}{}^*}+\frac{|\mathcal{V}_{ac}|^2}{(\omega_{bc}+i\gamma_{bc})}\left(\frac{n_{ab}}{\Delta_{ab1}}-\frac{n_{ac}}{\Delta_{ac1}{}^*}\right)\bigg\}, \tag{6}$$

where

$$\Delta_{aj1}\equiv\omega-\omega_{aj}-Kv_z+i\gamma_{aj},\quad \Delta_{aj2}\equiv\omega-\omega_{aj}+Kv_z+i\gamma_{aj},\quad \gamma_i{}'\equiv\gamma_i+\Gamma_i,$$

$$n_{aj}\equiv n_a-n_j=\lambda_a/\gamma_a{}'-[\lambda_j+\beta_{aj}(\lambda_a/\gamma_2{}')]\gamma_j{}'^{-1},$$

with $j=b$ or c, and n_{aj} the zero-field inversion density between states a and j with velocity v_z. The expression for h_{ac} is Eq. (6) with $b\leftrightarrow c$.

The macroscopic polarization is[4]

$$P(z,t)=\int[2\,\mathrm{Re}(\rho_{ab}\mathcal{P}_{ab}{}^*)+2\,\mathrm{Re}(\rho_{ac}\mathcal{P}_{ac}{}^*)]dv_z$$

so that the intensity gain for the probe beam in the $-z$ direction is

$$G_2(\omega)=-(K/\hbar)[|\mathcal{P}_{ab}|^2\,\mathrm{Re}(\int h_{ab}dv_z+|\mathcal{P}_{ac}|^2\,\mathrm{Re}(\int h_{ac}dv_z)].$$

In the Doppler limit, integration over velocity gives the following expression for the gain at ω minus the gain for $\omega\to\infty$[7]:

$$G_2(\omega)-G_2(\infty)=-\frac{\pi^{3/2}E_1{}^2}{4\hbar^3u}\bigg\{N_{ab}|\mathcal{P}_{ab}|^4\frac{\gamma_a{}'+\gamma_b{}'-\beta_{ab}}{\gamma_a{}'\gamma_b{}'}\mathcal{L}(\omega_{ab},\gamma_{ab})+N_{ac}|\mathcal{P}_{ac}|^4\frac{\gamma_a{}'+\gamma_c{}'-\beta_{ac}}{\gamma_a{}'\gamma_c{}'}\mathcal{L}(\omega_{ac},\gamma_{ac})$$
$$+|\mathcal{P}_{ab}|^2|\mathcal{P}_{ac}|^2\left[N_{ac}\left(\frac{\gamma_b{}'-\beta_{ab}}{\gamma_a{}'\gamma_b{}'}\right)+N_{ab}\left(\frac{\gamma_c{}'-\beta_{ac}}{\gamma_a{}'\gamma_c{}'}\right)\right]\mathcal{L}\left(\frac{\omega_{ab}+\omega_{ac}}{2},\frac{\gamma_{ab}+\gamma_{ac}}{2}\right)$$
$$-\frac{1}{\pi}\frac{|\mathcal{P}_{ab}|^2|\mathcal{P}_{ac}|^2}{\omega_{bc}{}^2+\gamma_{bc}{}^2}\left[\frac{N_{ab}[(\omega-\omega_{ab})\omega_{bc}-\gamma_{ab}\gamma_{bc}]}{(\omega-\omega_{ab})^2+\gamma_{ab}{}^2}-\frac{N_{ac}[(\omega-\omega_{ac})\omega_{bc}+\gamma_{ac}\gamma_{bc}]}{(\omega-\omega_{ac})^2+\gamma_{ac}{}^2}\right]\bigg\}, \tag{7}$$

where

$$\mathcal{L}(\Omega,\gamma) \equiv \gamma/\pi[(\omega-\Omega)^2+\gamma^2],$$

and N_{aj} is the total zero-field inversion density (atoms per cm^3) between states a and j.

For $N_{ab}<0$ and $N_{ac}<0$, the resonances at ω_{ab} and ω_{ac} are positive, since γ_a' is always greater than β_{ab} or β_{ac}. However, the "cross-over" resonance can be positive or negative, depending on the sizes of β_{ab}/γ_b' and β_{ac}/γ_c'.[1] For example, if b and c are stable states, $\gamma_b'=\Gamma_b$ and $\gamma_c'=\Gamma_c$, so that the term is negative for $\beta_{ab}/\Gamma_b>1$ and $\beta_{ac}/\Gamma_c>1$ (as well as for other cases, depending on the relative sizes of N_{ab} and N_{ac}). The asymmetric term is small if ω_{bc}/γ_{ab} is large; however, it can be important for high-precision measurements of ω_{ab} (see Ref. 2). The fractional shift in the resonance center due to this term is of the order of $\gamma_{ab}^2/\omega_{ab}\omega_{bc}$.

If the resonances are well separated, then one can calculate each resonance separately, since atoms with a given v_z can participate in only one kind of transition, depending on the value of ω. It is then possible to calculate the shapes of the resonances noniteratively, for large values of E_1. For the case of ω near ω_{ab}, the Lorentzian factor in the first term in braces in Eq. (7) becomes

$$(1+8\alpha_{ab})^{-1/2}\mathcal{L}(\omega_{ab}, \tfrac{1}{2}\gamma_{ab}[1+(1+8\alpha_{ab})^{1/2}]),$$

where

$$\alpha_{aj} \equiv \frac{|\wp_{aj}|^2 E_1^{\,2}}{16\hbar^2\gamma_a'\gamma_j'}\,\frac{\gamma_a'+\gamma_j'-\beta_{aj}}{\gamma_{aj}}.$$

The power broadening of the cross-over term can be calculated by considering the case $\omega \cong \frac{1}{2}(\omega_{ab}+\omega_{ac})$. In this case, the Lorentzian factor in the part of the third term in braces in Eq. (7) proportional to N_{ac} becomes

$$(1+8\alpha_{ac})^{-1/2}\mathcal{L}(\tfrac{1}{2}(\omega_{ab}+\omega_{ac}), \tfrac{1}{2}[\gamma_{ab}+\gamma_{ac}(1+8\alpha_{ac})^{1/2}]),$$

and the part of that same term proportional to N_{ab} is multiplied by a similar expression with $c \rightarrow b$. Thus the cross-over resonance is a sum of two Lorentzians of different widths and heights.

Phase-changing collisions can be included by adding a term $d\mu_{aj}(t)/dt$ to ω_{aj}. By using an approximate averaging method,[4] one finds that the probe gain is given by the expressions derived previously if γ_{aj}, ω_{aj}, and α_{aj} are replaced by $\bar{\gamma}_{aj}$, $\bar{\omega}_{aj}$, and $\bar{\alpha}_{aj}$, respectively, where

$$\bar{\gamma}_{aj}=\gamma_{aj}+\gamma_p\langle 1-\cos\varphi_{aj}\rangle, \quad \bar{\omega}_{aj}=\omega_{aj}+\gamma_p\langle\sin\varphi_{aj}\rangle, \quad \bar{\alpha}_{aj}=\alpha_{aj}(\gamma_{aj}/\bar{\gamma}_{aj}),$$

with γ_p the reciprocal of the mean collision time, and φ_{aj} the phase change per collision for the $a\rightarrow j$ transition. Thus, for example, the resonance at ω_{ab} has a width of

$$\bar{\gamma}_{ab}[1+(1+8\bar{\alpha}_{ab})^{1/2}] = \bar{\gamma}_{ab}\left\{1+\left[1+\frac{|\wp_{ab}|^2E_1^{\,2}}{2\hbar^2}\left(\frac{1}{\gamma_a'}+\frac{1}{\gamma_b'}-\frac{\beta_{ab}}{\gamma_a'\gamma_b'}\right)\frac{1}{\bar{\gamma}_{ab}}\right]^{1/2}\right\}.$$

We have recently learned of a paper by Baklanov and Chebotaev[8] which treats the two-level version of our theory, with somewhat differing results. I should like to thank Dr. T. W. Hänsch for suggesting this problem to me.

[1]T. W. Hänsch, I. S. Shahin, and A. L. Schawlow, Phys. Rev. Lett. 27, 707 (1971).

[2]T. W. Hänsch, I. S. Shahin, and A. L. Schawlow, Nature (London) 235, 63 (1972).

[3]T. W. Hänsch, Appl. Opt. 11, 895 (1972).

[4]H. K. Holt, Phys. Rev. A 2, 233 (1970); see also S. Stenholm and W. E. Lamb, Jr., Phys. Rev. 181, 618 (1969), and B. J. Feldman and M. S. Feld, Phys. Rev. A 1, 1375 (1970), for equivalent theories.

[5]Effects due to the finite pulse widths and the noncollinearity of the two traveling waves in the experiments of Refs. 1–3 are neglected here. They will be considered in a later paper.

[6]Phase-changing collisions affect Eqs. (3) and (4); this is discussed at the end of the paper.

[7]$G_2(\omega)-G_2(\infty)$ gives the shapes of the resonances only in the limit $[G_2(\omega)-G_2(\infty)]Z \ll 1$, where Z is the length of the absorption region. Otherwise, the fractional change in the intensity is $\exp\{[G_2(\omega)-G_2(\infty)]Z\}-1$.

[8]E. V. Baklanov and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. 60, 552 (1971) [Sov. Phys. JETP 33, 300 (1971)].