reported results which show a marked deviation from the earlier work of Bennett, Chebotayev, and Knutson.[12]

The recent theory of Gyorffy, Borenstein, and Lamb[5] is most relevant to the present experiment. Therein, an expression for the power output of the type given in Eq. (3) above is further clarified. Their parameters γ_1 and γ_2 correspond to our γ' and γ, respectively. However, the theory predicts a nonlinear dependence on pressure for γ_2. From our determination of γ, this nonlinearity is apparently small for the 6328-Å transition. Estimates of line-shape asymmetry are in qualitative agreement with our observations.

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Atomic Relaxation in the Presence of a Coherent Optical Field

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Atomic relaxation in the presence of a strong coherent optical field is examined. Contrary to popular practice, we find that the Bloch equations do not apply when the applied field is strong enough to saturate the transition. The component of the induced polarization in phase with the field lives longer than the out-of-phase component. One important consequence is that pulse durations for adiabatic inversion of atomic levels need not be short relative to the transverse relaxation time of the atom if the pulse amplitude is sufficiently large.

ADIABATIC inversion of atomic levels by optical pulses has been proposed[1] as a technique for studying relaxation effects in atoms and molecules. Previous experiments involving light pulses and atomic coherence (e.g., photon echo[2] and self-induced transparency[3]) require pulse durations which are short compared to the transverse relaxation time T_2 of the atoms. Usually T_2 is several orders of magnitude smaller than the lifetime T_1 of the atom. Hence, this can be a serious limitation.

We find that adiabatic inversion is not subject to the same limitation on pulse duration. If the amplitude of the pulse is sufficiently large, pulse durations need only be short relative to T_1, not T_2. This feature, which is analogous to field-dependent relaxation[4] or spin locking[5] in NMR, suggests a broader range of applicability for adiabatic inversion than previously anticipated.

To exhibit this property we consider a simple model consisting of a two-level atom coupled to a coherent em field. The atom relaxes by virtue of a weak interaction with an external thermal bath, representing, for example, collisions with other gas atoms. The atomic states, labelled $|a\rangle$ and $|b\rangle$ with energy spacing $\epsilon_a-\epsilon_b=\hbar\omega_0$, are connected by an electric-dipole transition. We choose $l=1$, $m_l=1$ for level $|a\rangle$, and $l=0$ for level $|b\rangle$, where l,m_l are the usual angular momentum quantum numbers. The levels $|a\rangle$ and $|b\rangle$ are assumed to be nondegenerate.

Both the coherent applied field and the incoherent thermal bath are treated classically. The bath is described by a weak electric field whose components fluctuate randomly in time. The coherent wave oscillates with the frequency ω and is plane polarized in the x direction.

The Hamiltonian for the interacting atom is written $H=H_A+V_c(t)+V_I(t)$, where H_A is the Hamiltonian of the bare atom, and $V_c(t)$ and $V_I(t)$ are the coherent and incoherent interactions, respectively. Using the eigenstates $|a\rangle$, $|b\rangle$ of H_A as a basis for a matrix representation, we choose

$$V_c(t)=pE\cos\omega t\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},$$
$$V_I(t)=p\begin{pmatrix}\delta E_a(t) & \delta E_-(t)\\ \delta E_+(t) & \delta E_b(t)\end{pmatrix}, \tag{1}$$

where $p=ex_{ab}$ is the atomic dipole matrix element, E is the amplitude of the coherent field, $\delta E_\pm(t)=\delta E_x(t)\pm i\delta E_y(t)$ are the fluctuating field components

[1] E. B. Treacy, Phys. Letters **27A**, 421 (1968).
[2] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Letters **13**, 567 (1964); I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. **141**, 391 (1966).
[3] S. L. McCall and E. L. Hahn, Phys. Rev. Letters **18**, 908 (1967); C. K. N. Patel and R. E. Slusher, *ibid.* **19**, 1019 (1967).
[4] A. G. Redfield, Phys. Rev. **98**, 1787 (1955).
[5] S. R. Hartmann and E. L. Hahn, Phys. Rev. **128**, 2042 (1962).

of the bath representing "hard" (inelastic) collisions, and $p[\delta E_a(t)-\delta E_b(t)]$ represents the fluctuation in the atomic level spacing due to "soft" (elastic) collisions.

Each component of the incoherent field $\delta E_\alpha(t)$ ($\alpha=\pm, a, b$) is characterized by a correlation function. We choose the following simple exponential forms:

$$\begin{aligned}\langle\delta E_x(t)\delta E_x(t+\tau)\rangle_t&=\langle\delta E_y(t)\delta E_y(t+\tau)\rangle_t\\&=\tfrac{1}{3}\Delta E_h^2 e^{-|\tau|/\tau_h},\\ \langle[\delta E_a(t)-\delta E_b(t)][\delta E_a(t+\tau)-\delta E_b(t+\tau)]\rangle_t&\\=\Delta E_s^2 e^{-|\tau|/\tau_s},&\end{aligned}\tag{2}$$

where $\langle\ \rangle_t$ denotes a time average. Correlations between different field components are assumed to be nonexistent. In this simple model, the thermal bath is characterized by two correlation times τ_s and τ_h. In a gas, the correlation time for hard collisions τ_h is of order d/v_T where d is an atomic diameter and v_T is the thermal velocity of the gas. Typically, $\tau_h\sim10^{-13}$ sec. The correlation time for soft collisions is given by $\tau_s\sim1/\omega_c$, where ω_c is the collision frequency.

The presence of a strong coherent field significantly alters the relaxation of the atom by the bath. To understand this field dependence it is convenient to transform to a frame rotating with the frequency ω of the coherent field, viz., $|\psi(t)\rangle=\exp(-i\omega L_z t/\hbar)|\varphi(t)\rangle$, where $|\psi(t)\rangle$ is the state vector in the stationary frame, and L_z is the z component of the orbital angular momentum operator.

Under the rotation ω the Hamiltonian H assumes the new form $\mathcal{H}=\mathcal{H}_0+U(t)$, where

$$\begin{aligned}\mathcal{H}_0&\simeq H_A-\omega L_z+eEx/2=\hbar\begin{pmatrix}\Delta\omega & \Omega/2\\ \Omega/2 & 0\end{pmatrix},\\ U(t)&=p\begin{pmatrix}\delta E_a(t) & \delta E_-(t)e^{-i\omega t}\\ \delta E_+(t)e^{-i\omega t} & \delta E_b(t)\end{pmatrix}.\end{aligned}\tag{3}$$

Here, $\Delta\omega=\omega_0-\omega$ measures the departure of ω from resonance, and $\Omega=pE/\hbar$ is the Rabi flopping frequency. In the rotating-wave approximation $\mathcal{H}_0$ is time-independent. Viewed from the rotating frame only the interaction $U(t)$ with the thermal bath depends on the time.

The system $\mathcal{H}_0$ (not the bare atom) relaxes by virtue of the interaction $U(t)$. The two eigenstates of $\mathcal{H}_0$ are readily found:

$$\mathcal{H}_0|\pm\rangle=\hbar\omega_\pm|\pm\rangle;\quad \omega_\pm=\tfrac{1}{2}[\Delta\omega\pm(\Delta\omega^2+\Omega^2)^{1/2}],\tag{4a}$$

$$|\pm\rangle=\left(\omega_\pm^2+\frac{\Omega^2}{4}\right)^{-1/2}\begin{pmatrix}\omega_\pm\\ \Omega/2\end{pmatrix}.\tag{4b}$$

In this representation, the eigenstates $|+\rangle$ and $|-\rangle$ are two-component column vectors with eigenvalues ω_+ and ω_-, respectively.

Following the semiclassical theory of damping first suggested by Bloembergen, Purcell, and Pound[6] and later generalized by Redfield,[7] we write[8]

$$\begin{aligned}\frac{\partial\rho_{++}}{\partial t}&=-\frac{\partial\rho_{--}}{\partial t}=-\gamma_1(\rho_{++}-\rho_{--}),\\ \frac{\partial\rho_{+-}}{\partial t}&=-\gamma_2\rho_{+-},\end{aligned}\tag{5}$$

where

$$\gamma_1=\frac{2}{\hbar^2}\,\mathrm{Re}\int_0^\infty d\tau e^{-i(\omega_+-\omega_-)\tau}\times\langle\langle+|U(t)|-\rangle\langle-|U(t+\tau)|+\rangle\rangle_t,\tag{6a}$$

$$\gamma_2=\gamma_1+\frac{1}{\hbar^2}\int_{-\infty}^\infty d\tau\langle[\langle+|U(t)|+\rangle-\langle-|U(t)|-\rangle]\times[\langle+|U(t+\tau)|+\rangle-\langle-|U(t+\tau)|-\rangle]\rangle_t.\tag{6b}$$

Here, ρ is the density matrix ($\rho_{+-}=\langle+|\rho|-\rangle$, etc.) written in the interaction picture.

To describe atomic relaxation it is customary to assume the validity of the Bloch equations.[9] These phenomenological equations presume that the components of the polarization in phase and out of phase with the applied field decay at the same rate while the population difference between atomic levels decays at a different rate.

We find that the Bloch equations do not apply when Ω is large enough to saturate the transition. A transformation of the Redfield equations (5) from the basis $|\pm\rangle$ to the basis $|a\rangle$, $|b\rangle$ yields

$$\begin{aligned}\frac{d\mathbf{R}}{dt}=\boldsymbol{\Omega}\times\mathbf{R}&-\hat{\imath}\{[2\gamma_1\Omega^2+\gamma_2\Delta\omega^2]R_1\\&-(\gamma_2-2\gamma_1)\Delta\omega\Omega R_3\}/(\Delta\omega^2+\Omega^2)\\&-\hat{\jmath}\gamma_2R_2-\hat{k}\{[2\gamma_1\Delta\omega^2+\gamma_2\Omega^2]R_3\\&-(\gamma_2-2\gamma_1)\Delta\omega\Omega R_1\}/(\Delta\omega^2+\Omega^2).\end{aligned}\tag{7}$$

Here,

$$\mathbf{R}=R_1\hat{\imath}+R_2\hat{\jmath}+R_3\hat{k},\quad R_1=\rho_{ab}'+\rho_{ba}',$$
$$R_2=i(\rho_{ab}'-\rho_{ba}'),\quad R_3=\rho_{aa}'-\rho_{bb}',\quad \rho_{ab}'=\langle a|\rho'|b\rangle,$$

etc., where

$$\rho'=\exp(-i\mathcal{H}_0t/\hbar)\rho\,\exp(i\mathcal{H}_0t/\hbar).$$

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[6] N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948).

[7] A. G. Redfield, IBM J. Res. Develop. **1**, 19 (1957); in *Advances in Magnetic Resonance*, edited by J. S. Waugh (Academic Press Inc., New York, 1965), Vol. 1, Chap. 1.

[8] In the semiclassical theory, all diagonal components of the density matrix decay to the same value corresponding to an infinite temperature. As shown in Ref. 7, this deficiency is removed by recourse to a treatment which quantizes the bath. Then the density matrix decays to a Boltzmann distribution.

[9] As written, for example, by C. K. Rhodes, A. Szöke, and A. Javan, Phys. Rev. Letters **21**, 1151 (1968), Eq. (3).

Also, $\mathbf{\Omega}=\Omega\hat{\imath}+\Delta\omega\hat{k}$ and $\hat{\imath},\hat{\jmath},\hat{k}$ are orthogonal unit vectors. Equations (7) reduce to the usual Bloch equations in the limit of infinite temperature[8] if $\Omega\ll\Delta\omega$ or in the special case of isotropic relaxation $\gamma_2=2\gamma_1$ for arbitrary Ω, $\Delta\omega$.

Note that $\mathbf{R}\cdot\mathbf{\Omega}$ and $\mathbf{R}\times\mathbf{\Omega}$ relax according to $2\gamma_1$ and γ_2, respectively. This means that $2\gamma_1$ and γ_2 measure the relaxation of $\mathbf{R}$ parallel and transverse to the *precession* axis $\mathbf{\Omega}$ rather than to the $\hat{k}$ axis as in the Bloch equations.

Whether this distinction is important depends on the relative magnitude of $2\gamma_1$ and γ_2. Substitution of (2), (3), and (4) into (6) gives

$$\gamma_1=\left(\frac{p}{\hbar}\right)^2\left\{\frac{\Delta E_s^2}{2}\left(\frac{\Omega^2}{\Delta\omega^2+\Omega^2}\right)\left[\frac{\tau_s}{1+(\Delta\omega^2+\Omega^2)\tau_s^2}\right]\right.$$
$$+\frac{4}{3}\frac{\Delta E_h^2\tau_h}{(\Delta\omega^2+\Omega^2)}\left[\frac{\omega_+^2}{1+(\omega+[\Delta\omega^2+\Omega^2]^{1/2})^2\tau_h^2}\right.$$
$$\left.\left.+\frac{\omega_-^2}{1+(\omega-[\Delta\omega^2+\Omega^2]^{1/2})^2\tau_h^2}\right]\right\}, \quad (8a)$$

$$\gamma_2=\gamma_1+\left(\frac{p}{\hbar}\right)^2\left\{\Delta E_s^2\left(\frac{\Delta\omega^2}{\Delta\omega^2+\Omega^2}\right)\tau_s\right.$$
$$\left.+\tfrac{4}{3}\Delta E_h^2\left(\frac{\Omega^2}{\Delta\omega^2+\Omega^2}\right)\left[\frac{\tau_h}{1+\omega^2\tau_h^2}\right]\right\}. \quad (8b)$$

Viewed from the rotating frame, soft collisions appear partially as hard collisions and vice versa. This explains the admixture of both effects in the expressions for γ_1 and γ_2.

By way of comparison in the limit of vanishing applied field ($\Omega\rightarrow 0$), Eqs. (8) reduce to

$$\gamma_1\rightarrow\gamma_1^0=\frac{4}{3}\left(\frac{p\Delta E_h}{\hbar}\right)^2\left[\frac{\tau_h}{1+\omega_0^2\tau_h^2}\right], \quad (9a)$$

$$\gamma_2\rightarrow\gamma_2^0=\gamma_1^0+\Gamma_0, \quad (9b)$$

where $\Gamma_0=(p\Delta E_s/\hbar)^2\tau_s$ is the contribution due to soft collisions. Since the amplitude of the fluctuation in level width $p\Delta E_s/\hbar$ is of order ω_c and $\tau_s\sim 1/\omega_c$, we have $\Gamma_0\sim\omega_c$ as required.

Assuming γ_1^0 and γ_2^0 are known for the bare atom, we can obtain an order-of-magnitude assessment of γ_1 and γ_2. If $\omega\sim\omega_0\gg(\Delta\omega^2+\Omega^2)^{1/2}$, as is likely for optical frequencies, the term in (8a) due to hard collisions is of order γ_1^0. The remaining term due to soft collisions is of order $\Gamma_0/[1+(\Delta\omega^2+\Omega^2)\tau_s^2]$. The latter is small relative to γ_1^0 provided that $(\Delta\omega^2+\Omega^2)^{1/2}\tau_s$ is sufficiently large. This condition requires that the precession frequency of the vector $\mathbf{R}$ about the axis $\mathbf{\Omega}$ be large compared to the collision frequency $\omega_c\sim 1/\tau_s$. In this event, the longitudinal (along $\mathbf{\Omega}$) relaxation rate γ_1 is of order γ_1^0, which is generally small compared to the transverse relaxation rate $\gamma_2\sim\gamma_2^0$ given by (8b).

A specific example will serve to illustrate this result. For the 1.15-μ Ne transition in a 10:1 He-Ne mixture, Szöke and Javan[10] measure $\gamma_1^0\sim 7\times 10^6$ sec^{-1} and $\Gamma_0\sim 10^8 p_T$ sec^{-1}, where p_T is the partial pressure of Ne in torrs. For typical laser pulses and atomic dipole matrix elements, the Rabi flopping frequency lies in the range $\Omega\sim 10^8$–10^{10} sec^{-1}. Choosing $\Omega\sim 10^{10}$ sec^{-1} and $p_T=1$ Torr, we obtain $\Omega\tau_s\sim 10^2$. This gives, for the 1.15-μ Ne transition, $\gamma_1\sim\gamma_1^0\sim 7\times 10^6$ sec^{-1}, while $\gamma_2\sim\gamma_2^0\sim\Gamma_0\sim 10^8$ sec^{-1}. We conclude that for resonant Ne atoms ($\Delta\omega=0$) the in-phase component of the polarization R_1 will live more than ten times as long as the out-of-phase component R_2. Thus, operating conditions appropriate for a relatively long-lived in-phase component appear to be readily realizable in practice.

A characteristic feature of adiabatic inversion is that it generates a large in-phase component of the polarization. Since the projection of $\mathbf{R}$ along $\mathbf{\Omega}$ is preserved during $\sim T_1=1/\gamma_1^0$ (provided that $\Omega\tau_s\gg 1$), pulse durations need not be short relative to $T_2=1/\gamma_2^0$.

Preliminary experimental confirmation of adiabatic inversion in SF_6 at 10.6-μ wavelength has been obtained by Treacy and DeMaria.[11] The apparent persistence of the effect at anomalously high buffer gas pressures[12] prompted the present theoretical study. The relevance of our theory to SF_6 is not clear because of cross relaxation between neighboring rotational levels. We believe the theory is applicable to any system for which the distinction between T_1 and T_2 is large, a notable example being an impurity atom in a solid.

We wish to thank E. B. Treacy for suggesting this problem and for subsequent helpful conversations.

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[10] A. Szöke and A. Javan, Phys. Rev. **145**, 137 (1966).

[11] E. B. Treacy and A. J. DeMaria, Phys. Letters (to be published).

[12] E. B. Treacy, Second Conference on the Laser at New York Academy of Sciences, 1969 (unpublished).