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Theory of Optical Free-Induction Decay and Two-Photon Superradiance*

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The Stark-pulse technique of Brewer and Shoemaker allows one to observe phenomena such as optical free-induction decay, optical nutation, and two-photon superradiance. In this paper, we develop a theoretical treatment of these phenomena under the appropriate experimental conditions. The calculations of the free-induction decay and the corresponding optical nutation are shown to be in excellent agreement with experiment. We also discuss the two-photon superradiance phenomenon of Brewer and Shoemaker as a Raman effect appearing in first-order perturbation theory. The analysis illustrates the essential features of the experimental observation.

INTRODUCTION

A recent series of experiments by Brewer and Shoemaker[1-3] have opened up new areas in spectroscopy and have provided a simple means of measuring relaxation times. In these experiments, molecules are Stark shifted in and out of resonance with a laser, and their resulting emission heterodyne detected. The present paper offers a theoretical analysis of the observations.

In the experiment, a cw laser is incident on a Doppler-broadened gaseous medium. A Stark field is applied moving either all or part of the atomic ensemble in (or out) of resonance with the laser. The Stark shift thus enables the laser to interact with molecules belonging to different velocity groups.

In the case of a simple two-level system, there are two separate physical processes which take place when the Stark field is applied (taken to be at $t=0$). Those molecules which interacted with the laser for $t<0$ are switched out of resonance and exhibit free-induction decay.[4,5] Those molecules which are switched into resonance at $t=0$ exhibit optical nutation.[6] This separation of the problem into two parts depends on the frequency shift $\delta\nu$ caused by the Stark field being large enough so that the two velocity groups do not overlap.

The experiment of Brewer and Shoemaker is the first observation of free-induction decay in the optical regime. The molecules which radiate in this manner are coherently excited by the laser for times $t<0$. After the Stark shift, these molecules are far off resonance and the macroscopic dipole radiates at a frequency determined by the amount of Stark shift they experience. The exponential decay of the amplitude has the usual T_2 term and a term describing the coherent dephasing of the frequency spread in the ensemble of initially excited molecules.

The optical-nutation effect[6] is well known and our calculation will concentrate on the free-induction decay. A brief discussion of optical nutation is included so that we can compare our results with the experimental data.

For most of the molecular transitions studied by Brewer and Shoemaker, we are justified in considering only a two-level system. In the special case where the Stark shift removes a molecular degeneracy, many levels are brought into play and an additional process appears. This process, which was called two-photon superradiance,[2] is charac-

terized by a lifetime longer than that of the free-induction decay. As originally suggested, it may be interpreted in terms of a Raman effect. Since atomic velocities do not enter into the Raman shift, the Doppler broadening does not contribute any dephasing and as a consequence, the radiation has a longer lifetime than in the case of free-induction decay.

In a later section of this paper, we will discuss two-photon superradiance in enough detail to bring out the essentials of this process. In this section, however, we concentrate on developing the theoretical basis for the treatment of the free-induction decay and optical nutation.

The density-matrix equations of motion for a two-level atom[7] of atomic frequency ω interacting with a classical electric field are given by

$$\dot{\rho}_{aa}(t,z,\omega) = -\rho_{aa}\frac{1}{T_1} - i\frac{\wp}{\hbar}E(t,z)(\rho_{ab}-\rho_{ba}) \quad , \tag{1}$$

$$\dot{\rho}_{bb}(t,z,\omega) = (1-\rho_{bb})\frac{1}{T_1} + i\frac{\wp}{\hbar}E(t,z)(\rho_{ab}-\rho_{ba}), \tag{2}$$

$$\dot{\rho}_{ab}(t,z,\omega) = -i\omega\rho_{ab} - \rho_{ab}\frac{1}{T_2} - i\frac{\wp}{\hbar}E(t,z)(\rho_{aa}-\rho_{bb}) \quad , \tag{3}$$

where $\wp$ is the dipole matrix element. The laser field is taken as $\mathcal{E}_0\cos(kz-\nu t)$, and we write the total field as

$$E(t,z) = \mathcal{E}(t,z)\cos(kz-\nu t) \quad .$$

The slowly varying envelope $\mathcal{E}(t,z)$ consists of two parts: the constant laser amplitude $\mathcal{E}_0$ and a contribution $\mathcal{E}_1$ from the medium. $\mathcal{E}_1$ in turn has two contributions. One is from the free-induction decay process ($\mathcal{E}_{\rm FID}$) and the other from the optical nutation ($\mathcal{E}_{\rm ON}$).

In our calculation, both $\mathcal{E}_{\rm FID}$ and $\mathcal{E}_{\rm ON}$ are small compared to the laser amplitude $\mathcal{E}_0$, and we are therefore in a position to replace the total electric field in the density-matrix equations by the laser field alone. This approximation, known as thin-medium perturbation theory,[6,8] is valid under the experimental conditions of Brewer and Shoemaker.

At the detector, the field is $(\mathcal{E}_0+\mathcal{E}_{\rm FID}+\mathcal{E}_{\rm ON})\times\cos(kz-\nu t)$. Since $\mathcal{E}_0$ is much larger than the other terms, the intensity averaged over the optical period is

$$\tfrac{1}{2}\mathcal{E}_0^2 + (\mathcal{E}_{\rm FID}+\mathcal{E}_{\rm ON})\mathcal{E}_0 \quad .$$

The signal in which we are interested, $\mathcal{E}_{\rm FID}+\mathcal{E}_{\rm ON}$, is measured relative to the dc contribution, $\frac{1}{2}\mathcal{E}_0^2$. We also note that by means of optical heterodyne detection, the magnitude of the term $\mathcal{E}_{\rm FID}+\mathcal{E}_{\rm ON}$ has been amplified by a factor of $\mathcal{E}_0$.

Our system of equations is completed with Maxwell's equations coupling $\mathcal{E}(t,z)$ to the polarization. With the slowly varying amplitude and phase approximation, the wave equation becomes

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathcal{E}(t,z) = -\frac{N_0\nu\wp}{2c\epsilon_0}\int d\omega\,\sigma(\omega) \times \mathrm{Re}[2ie^{-i(kz-\nu t)}\rho_{ab}(t,z,\omega)] \quad . \tag{4}$$

We have used N_0 for the density of atoms and $\sigma(\omega)$ for the Doppler distribution.

FREE-INDUCTION DECAY

We assume that those molecules which exhibit free-induction decay have reached a steady state when the Stark field is applied (at $t=0$). In this case, we have from Eqs. (1)–(3)

$$\rho_{ab}(0^-,z,\omega) = \tfrac{1}{2}ie^{ikz}\left(\frac{T_2\tilde{\mathcal{E}}_0(1-i\Delta T_2)}{\Delta^2T_2^2+T_1T_2\tilde{\mathcal{E}}_0^2+1}\right) \quad , \tag{5}$$

$$\rho_{bb}(0^-,z,\omega) - \rho_{aa}(0^-,z,\omega) = 1 - \frac{T_1T_2\tilde{\mathcal{E}}_0^2}{\Delta^2T_2^2+T_1T_2\tilde{\mathcal{E}}_0^2} \quad , \tag{6}$$

with $\tilde{\mathcal{E}}_0 = (\wp/\hbar)\mathcal{E}_0$ and $\Delta = \omega-\nu$. Once the Stark field is applied, these same molecules move to a frequency $\omega+\delta\nu$ and no longer interact with the laser (they are too far off resonance). These coherently excited molecules simply decay and oscillate:

$$\rho_{ab}(t\geq 0,z,\omega+\delta\nu) = -\tfrac{1}{2}ie^{i(kz-\nu t)}e^{-t/T_2}e^{-i(\omega+\delta\nu-\nu)t}C \quad ,$$

C being an integration constant (complex) to be determined from the boundary conditions. The boundary condition we impose is that the contribution to ρ_{ab} from each group of molecules be continuous across $t=0$. Thus, we write for ρ_{ab}

$$\rho_{ab}(t\geq 0,z,\omega+\delta\nu) = \tfrac{1}{2}ie^{i(kz-\nu t)}e^{-t/T_2}e^{-i(\omega+\delta\nu-\nu)t} \times\frac{\tilde{\mathcal{E}}_0(1-i\Delta T_2)/T_2}{\Delta^2+(T_1/T_2)\tilde{\mathcal{E}}_0^2+(1/T_2^2)} \quad . \tag{7}$$

Rewriting the resonance denominator in Eq. (5) as $\Delta^2+\Gamma^2$, where

$$\Gamma = [(T_1\tilde{\mathcal{E}}_0^2/T_2)+(1/T_2^2)]^{1/2} \quad ,$$

we see that the Stark shift $\delta\nu$ must be greater than Γ in order to resolve different velocity groups. This is well satisfied under typical experimental conditions.[3]

The contribution to $\mathcal{E}_1$ from the free-induction-decay process we call $\mathcal{E}_{\rm FID}$. It is obtained by use of the integral form of Eq. (4):

$$\tilde{\mathcal{E}}_{\rm FID} = -\tfrac{1}{2}c\alpha'\int_{\mu}^{\mu+2L/c}d\eta\int d\omega\,\sigma(\omega-\delta\nu) \times\mathrm{Re}[2i\exp\{-ikc[(\eta-\mu)/2-(\eta+\mu)/2]\} \times\rho_{ab}(\tfrac{1}{2}(\eta+\mu),\tfrac{1}{2}c(\eta-\mu),\omega)] \quad ,$$

where $\eta\equiv t+z/c$, $\mu\equiv t-z/c$, L is the length of the Stark cell (10 cm), and $\alpha' = \wp^2N_0\nu/2\hbar c\epsilon_0$.

The phased array of dipoles which are shifted out of resonance at $t=0$ have their frequencies changed, but not their wave vectors. A phase mismatch then occurs when these dipoles begin to radiate. If we do the integration over η, we pick up the phase matching term $\sin(\delta\nu L/2c)/\delta\nu(L/2c)$ which, under the prevailing experimental conditions, is nearly equal to one ($\delta\nu L/2c \ll 1$). We can neglect the phase matching by evaluating the integrand at $\eta=\mu$ and multiplying by $2L/c$. The integration over ω may then be done if we assume a slowly varying Doppler curve (i.e., $1/T_2\Delta\omega_{\text{Doppler}} \ll 1$), since in that case the function $\sigma(\omega)$ can be removed from the frequency integral, and the result is just a Fourier transform of a Lorentzian. In a straightforward manner, one then obtains

$$\mathcal{E}_{\text{FID}} = + c\alpha' \frac{L}{T_2 c}\, \mathcal{E}_0 \sigma(\nu)\pi \times\left(\frac{1 - T_2[(T_1/T_2)\tilde{\mathcal{E}}_0^2 + (1/T_2^2)]^{1/2}}{[(T_1/T_2)\tilde{\mathcal{E}}_0^2 + (1/T_2^2)]^{1/2}}\right) \times\exp\left\{-\left[\frac{1}{T_2} + \left(\frac{T_1}{T_2}\,\tilde{\mathcal{E}}_0^2 + \frac{1}{T_2^2}\right)^{1/2}\right]\mu\right\}\cos(\delta\nu)\mu \quad . \tag{8}$$

The free-induction decay is therefore a sinusoidal oscillation at a frequency $\delta\nu$ with a decay rate $1/\tau$ given by

$$\frac{1}{\tau} = \frac{1}{T_2} + \left(\frac{T_1}{T_2}\,\tilde{\mathcal{E}}_0^2 + \frac{1}{T_2^2}\right)^{1/2} \quad . \tag{9}$$

We notice that there are two contributions to this decay rate. The first is the usual $1/T_2$ term. The second comes from the frequency spread in the phase array of dipoles set up by the laser field for $t \le 0$. This can be seen from the denominator in Eq. (7).

OPTICAL NUTATION

The electric field at the detector results from a superposition of the free-induction decay and the optical nutation.[6] This latter process is somewhat more complicated algebraically and we make some simplifying assumptions. In Eqs. (1)–(3), we take $T_1 = T_2 = T$, and again use thin-medium perturbation theory to replace E by E_0.

In optical nutation, we are talking about molecules which are initially far off resonance and which, at $t=0$, are suddenly shifted into resonance with the laser. For $t \le 0$, we take steady-state values to be $\rho_{ab}=0$ and $\rho_{bb} - \rho_{aa} = 1$. Because the electric field amplitude is constant, the density-matrix equations (1)–(3) (after making the rotating-wave approximation) become a set of three simple linear differential equations which can be solved by standard techniques. The algebra is somewhat complicated, however, and we just write the answer here. The off-diagonal element of the density matrix is then

$$\rho_{ab} = i\,\frac{\tilde{\mathcal{E}}_0\, e^{i(kz-\nu t)}}{2(\Delta^2 + \tilde{\mathcal{E}}_0^2 + 1/T_2^2)}\,\frac{1}{T}\Big((1 - i\Delta T) - e^{-t/T} \times((1 - i\Delta T)\cos(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2} t - \{(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2} T + i[\Delta/(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2}]\} \times\sin(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2} t)\Big) \quad . \tag{10}$$

The contribution to E from the optical nutation we write as $\mathcal{E}_{\text{ON}}$ and it may be found by integrating Eq. (4). This expression for the optical nutation is complicated by the fact that this experiment imposes an initial condition at $t=0$ rather than at $\mu=0$, as in more conventional optical-pulse experiments. This introduces a negligible contribution provided the atoms respond slowly in the transit time of the cell [$(2L/c)\tilde{\mathcal{E}}_0 \ll 1$]. This is well satisfied and we write our result as if the initial conditions were imposed at $\mu=0$:

$$\tilde{\mathcal{E}}_{\text{ON}}(\mu) = \frac{c\alpha'}{2}\,\frac{2L}{c}\int_{-\infty}^{\infty}\sigma(\omega - \delta\nu)\,\frac{\tilde{\mathcal{E}}_0/T}{\Delta^2 + \tilde{\mathcal{E}}_0^2 + 1/T^2} \times\{1 - e^{-\mu/T}[\cos(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2}\mu - T(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2}\sin(\tilde{\mathcal{E}}_0^2 + \Delta^2)^{1/2}\mu]\}\, d\omega \quad . \tag{11}$$

This integral cannot be computed analytically. If, however, we assume $\tilde{\mathcal{E}}_0 \gg 1/T$, and that the Doppler line is very broad, then we can do the integration:

$$\mathcal{E}_{\text{ON}} = \frac{c\alpha'}{2}\,\sigma(\nu - \delta\nu)2\pi\,\frac{L}{Tc} \times[1 + e^{-\mu/T}(\tilde{\mathcal{E}}_0 T)\, J_0(\tilde{\mathcal{E}}_0\mu)]\,\frac{\hbar}{\wp} \quad .$$

Thus we recover the usual Bessel-function behavior of optical nutation.[6]

The limiting case $\tilde{\mathcal{E}}_0 \gg 1/T$ is not what is encountered experimentally. Consequently, we have done a numerical integration to find $\mathcal{E}_{\text{ON}}$. When $\tilde{\mathcal{E}}_{\text{ON}}$ is comparable to $1/T$, the optical nutation behaves like a critically damped harmonic oscillator instead of a Bessel function. As a result, the optical nutation appears mainly as a decaying background. This is shown in Fig. 1, where we have plotted $\mathcal{E}_{\text{ON}} + \mathcal{E}_{\text{FID}}$. The fast ripple due to free-induction decay (at a frequency $\delta\nu$) rides on top of the slowly varying optical-nutation contribution.

For Fig. 1, we have chosen parameters consistent with the available experimental data. Certain information, such as the initial position of the laser frequency in the Doppler line, was not known. This will have an effect on the relative amplitudes of the free-induction decay and the optical nutation. In our numerical calculation, we used a value of

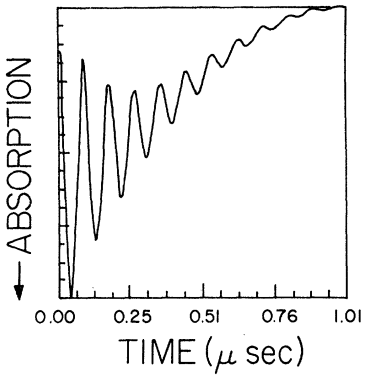

FIG. 1. Computer generated plot of the sum of optical nutation and free-induction decay effects. For this graph, $T_1 = T_2 = 1.4$ μsec. $\tilde{\mathcal{E}}_0 = 4$ MHz, $\delta\nu = 70$ MHz. All frequencies quoted are in angular units.

40 MHz off the center of the Doppler line in a direction so as to enhance the amplitude of the free-induction decay. This amplitude is then about 15% greater than it would be if the laser were initially at the center of the Doppler distribution.

In the experiment, both the Stark pulse and the detector have a finite rise time. As a result, there is a slight discrepancy between our predictions and the results of Brewer and Shoemaker for very short times (~40 nsec). With this proviso, Fig. 1 is in very good agreement with the experimental results.

TWO-PHOTON SUPERRADIANCE

As mentioned in the Introduction, the two-photon superradiance of Brewer and Shoemaker is basically a different process from those we have been considering up to now. While optical nutation and free-induction decay can be obtained from pulsing the atoms (with a Stark field) or pulsing the radiation (the more conventional approach), the two-photon superradiance effect can only be obtained from the Stark-pulse technique.

This effect was observed when a degenerate transition, initially in resonance with the laser, was subsequently split by interaction with the Stark field. Under these circumstances, there are several transition frequencies and several free-induction-decay signals which quickly disappear due to mutual destructive interference. Brewer and Shoemaker observed these signals, but in addition, they found another rapidly oscillating signal with a lifetime on the order of T_2 rather than that given in Eq. (9).

Since the magnetic sublevels in the upper (and lower) state are equally split by the Stark shift, Raman scattering from these levels can take place at one frequency only. As the atomic velocities do not enter into the Raman shift,[9] there is no dephasing due to the Doppler distribution as was the case in Eq. (9). This led to the suggestion that the radiation was a superradiant Raman effect because it occurred when the upper and lower Raman levels were equally populated (i.e., it was neither spontaneous nor stimulated) and it arose due to the coherence between the levels. It will be shown in the present section of this paper that this viewpoint is in accord with the theoretical analysis.

Because we are now dealing with many levels rather than just two, we will need a somewhat expanded notation over the one used in the previous sections. We will use a and b as before to refer to the upper and lower states, and m and m' to refer to the magnetic sublevels. Thus, $\rho_{am,bm'}$ refers to $\langle am|\,\rho\,|bm'\rangle$, where ρ is the density operator. With this notation, the general density-matrix equations (using the fact that there are no dipole matrix elements between the magnetic quantum states of a given level) are

$$\dot{\rho}_{am,am'} = -i(\omega_{am} - \omega_{am'})\,\rho_{am,am'} - \gamma_{am,am'}\,\rho_{am,am'} + \frac{i}{\hbar}E(t,z)\sum_{m''}(\wp_{mm''}\rho_{bm'',am'} - \rho_{am,bm''}\wp_{m'',m'}) \quad , \tag{12}$$

$$\dot{\rho}_{bm,bm'} = -i(\omega_{bm} - \omega_{bm'})\,\rho_{bm,bm'} - \gamma_{bm,bm'}\,\rho_{bm,bm'} + \frac{i}{\hbar}E(t,z)\sum_{m''}(\wp_{mm''}\rho_{am'',bm'} - \rho_{bm,am''}\wp_{m'',m'}) \quad , \tag{13}$$

$$\dot{\rho}_{am,bm'} = -i(\omega_{am} - \omega_{bm'} - kv)\,\rho_{am,bm'} - \frac{1}{T_2}\rho_{am,bm'} + \frac{i}{\hbar}E(t,z)\sum_{m''}(\wp_{mm''}\rho_{bm'',bm'} - \rho_{am,am''}\wp_{m'',m'}) \quad . \tag{14}$$

We have not included any "pump" terms of the sort appearing in Eq. (2) since they cannot give rise to the effect we are looking for and merely complicate the analysis. Another change from Eqs. (1)–(3) is that rather than taking the medium to be inhomogeneously broadened, we have explicitly written the velocity v in these equations. In our final expression for the radiation, this term will cancel out, emphasizing that there is no dephasing.

The axis of quantization for our equations is chosen to be the direction of the Stark field. Since the experiment is carried out with the Stark field orthogonal to the polarization of the laser, the selection rules for electric dipole transitions are $\Delta m = \pm 1$. The dipole matrix element is then given by

$$\wp_{mm'} = e\langle am|x|bm'\rangle \ .$$

The constant laser field for times $t \le 0$ establishes a steady-state situation in the atoms and gives a density operator $\rho\ (t=0)$ which will serve as an initial condition for the response of the atoms after the shift. The zeroth-order solution to the density-matrix equations is then

$$\rho^{(0)}_{am,am'}(t) = \rho_{am,am'}(0)\exp[-i(\omega_{am}-\omega_{am'})t-\gamma_{am\,am'}t]\ , \tag{15}$$

$$\rho^{(0)}_{bm,bm'}(t) = \rho_{bm,bm'}(0)\exp[-i(\omega_{bm}-\omega_{bm'})t-\gamma_{bm\,bm'}t]\ , \tag{16}$$

$$\rho^{(0)}_{am,bm'}(t) = \rho_{am,bm'}(0)$$

$$\times\exp[-i(\omega_{am}-\omega_{bm'}-kv)t-(1/T_2)t]\ . \tag{17}$$

Using Eq. (4), we can calculate the electric field radiated after the Stark shift exactly as we did in our previous discussion. In this case, however, the levels are split apart and there are many allowed values of $\omega_{am}-\omega_{bm'}$ which cause the free-induction decays to destructively interfere and the signal to quickly disappear. (We are neglecting the optical nutation since, as we have shown, it is an easily identifiable background.)

Since the effect we are looking for is a Raman effect, we expect that it will only turn up in higher-order perturbation theory. Substituting Eqs. (15)–(17) back into the density-matrix equations, we find

$$\rho^{(1)}_{am,bm'}(t) = \rho_{am,bm'}(0)\exp[-i(\omega_{am}-\omega_{bm'}-kv)t-(1/T_2)t] - \frac{i}{\hbar}\mathcal{E}_0\int_0^t dt'\cos(kz-\nu t')$$
$$\times\exp[-i(\omega_{am}-\omega_{bm'}-kv)(t-t')]e^{-(1/T_2)(t-t')}\sum_{m''}\{\wp_{m''m'}\rho_{am,am''}(0)\exp[-i(\omega_{am}-\omega_{am''})t'-\gamma_{am\,am''}t']$$
$$-\wp_{mm''}\rho_{bm'',bm'}(0)\exp[-i(\omega_{bm''}-\omega_{bm'})t'-\gamma_{bm''bm'}t']\}\ . \tag{18}$$

When the integration is performed, the polarization will decompose into three types of terms. The first oscillates as $\exp[-i(\omega_{am}-\omega_{bm'}-kv)t]$ and gives no net contribution since, as noted before, the frequencies $\omega_{am}-\omega_{bm'}$ are distributed over a wide range and will destructively interfere. The second type of term goes as $\exp[-i(\omega_{bm'}-\omega_{bm''}-\nu)t]$ with $m'=m''\pm 2$. Since the Stark effect is linear, $|\omega_{bm}-\omega_{bm\pm2}|$ is the same for all m, and there is no cancellation due to mutual interference. We also note that the velocity-dependent exponentials have canceled out, and there is no contribution to the dephasing from the Doppler broadening. This is exactly what we were looking for, and represents, as noted by Brewer and Shoemaker, the Raman emission from the two-photon transition.

This process contributes to the density matrix a term given by

$$\rho^{2\phi}_{am,bm'}(t) = -\frac{i}{2\hbar}\mathcal{E}_0 e^{i(kz-\nu t)}\sum_{m''}\left(\wp_{m''m'}\rho_{am,am''}(0)\right.$$
$$\times\frac{e^{-i(\omega_{am}-\omega_{am''})t}e^{-\gamma_{aa}t}}{i(\omega_{am''}-\omega_{bm'}-kv-\nu)+[(1/T_2)-\gamma_{am''am}]}$$
$$-\wp_{mm'}\rho_{bm'',bm'}(0)$$
$$\left.\times\frac{e^{-i(\omega_{bm''}-\omega_{bm'})t}e^{-\gamma_{bb}t}}{i(\omega_{am}-\omega_{bm''}-kv-\nu)+(1/T_2-\gamma_{bm''bm'})}\right)\ . \tag{19}$$

Here, we use the symbol γ_{aa} to refer to the decay rate of the density-matrix element $\rho_{am,am\pm2}$, which, in the absence of collisions, is $\frac{1}{2}(\gamma_{am}+\gamma_{am\pm2})$. Similar considerations apply, of course, to γ_{bb}. We use this symbol to differentiate such a decay from the lifetime $1/\gamma_a$ or the usual phase-memory time T_2. The word "intracoherence" time[10] has been used for γ_{aa} and γ_{bb} to emphasize that they are coherence times and hence, strongly influenced by collisions.

The radiation from this term, $\mathcal{E}_{2\phi}$, may be found by summing over the product of $\wp_{mm'}$ and $\rho^{2\phi}_{am,bm'}$, and integrating over the distribution of velocities. Since the velocities appear only in the resonance denominator and implicitly in the term $\rho_{bm'',bm'}(0)$. This integral contributes only to the amplitude of the signal:

$$\mathcal{E}_{2\phi} = -\frac{N_0\nu}{2\epsilon_0}\frac{\mathcal{E}_0}{\hbar}\frac{L}{c}\mathcal{A}_a e^{2i\Delta\omega_a\mu-\gamma_{aa}\mu}$$
$$-\frac{N_0\nu}{2\epsilon_0}\frac{\mathcal{E}_0}{\hbar}\frac{L}{c}\mathcal{A}_b e^{2i\Delta\omega_b\mu-\gamma_{bb}\mu}+\text{c. c.} \tag{20}$$

As in the case of free optical nutation, there are small factors coming from the application of boundary conditions at $t=0$ rather than $\mu=0$. We ignore such terms in this case as well. The amplitudes of the two-photon radiation are then

$$\mathcal{A}_a = \frac{1}{2}\sum_m\int\sigma(kv)\left(\frac{\wp_{m,m+1}\wp_{m+2,m+1}}{i(\omega_{am+2}-\omega_{bm+1}-kv-\nu)+(1/T_2-\gamma_{aa})}+\frac{\wp_{m+1,m+2}\wp_{m+1,m}}{-i(\omega_{am}-\omega_{bm+1}-kv-\nu)+(1/T_2-\gamma_{aa})}\right)\rho_{am,am+2}\,d(kv),$$

$$\alpha_b = -\frac{1}{2}\sum_m \int \sigma(kv)\left(\frac{\wp_{m,m+1}\wp_{m,m-1}}{i(\omega_{am}-\omega_{bm-1}-kv-\nu)+(1/T_2-\gamma_{bb})}+\frac{\wp_{m-1,m}\wp_{m+1,m}}{-i(\omega_a-\omega_{bm+1}-kv-\nu)+(1/T_2-\gamma_{bb})}\right)\rho_{bm-1,bm+1}\,d(kv)\,.$$

We have pointed out before that there is no dephasing to contribute to the decay time of this radiation. We should, however, note the fact that it decays with the intracoherence[10] time γ_{aa} (or γ_{bb}). This is the same intracoherence time measured in the Hanle effect.[11] Since measurements reveal[2] that this decay rate is on the order of $1/T_2$, the resonance denominators give a large contribution to the amplitude, and we expect the signal to be quite strong. This has been found experi-

In Eq. (20), we see that there are actually two contributing terms: one at a frequency $2\Delta\omega_a$ and the other at a frequency $2\Delta\omega_b$. These frequencies are very close, but a spectrum analysis[2] reveals the two components of Eq. (20).

As in the case of the free-induction decay, this signal appears on top of the optical nutation for the atoms switched into resonance. Since many levels are involved, it becomes tedious to solve the problem completely. For this reason, we have attempted only a qualitative discussion rather than a precise fit to the data as in the previous sections.

CONCLUSION

In this paper, we have discussed two new observations that have come from the work of Brewer and Shoemaker. These are the optical free-induction decay and the two-photon superradiance. Free-induction decay has a complicated decay time as it involves the dephasing of a set of molecules with a finite spread in spectral width which are simultaneously decaying owing to collisional dephasing. The signal, which beats against the laser at a frequency determined by the magnitude of the Stark shift, rides on top of the optical nutation of the atoms switched into resonance. A specific calculation was made which showed excellent agreement between experiment and theory.

Two-photon superradiance occurs when a resonant degenerate line, interacting with the laser, is split by the Stark field. We have shown that it is a Raman effect which shows up in first-order perturbation theory and comes about due to the coherence established by the laser among the magnetic sublevels. The radiation is emitted without a dephasing contribution and decays according to the intracoherence times of the sublevels. Since these times are comparable to the ordinary coherence times, the resonance denominators can become nearly zero, causing large-amplitude signals. All these properties are in accord with the experimental observations.

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