Free-precession decay of two-photon–induced coherence in Ca vapor

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Calcium vapor has been excited by two-photon absorption of a mode-locked dye-laser pulse, and emission from a coherent superposition state of the $4s^2\,{}^1S_0$ and $4s3d\,{}^1D_2$ levels has been observed using a delayed off-resonant interrogation pulse in the picosecond regime. The emission signal intensity was measured as a function of the delay time of the interrogation pulse, and showed a peak well delayed from that of the dye-laser pulse. This reflects the fact that the induced coherence is expressed by a time integral of the excitation. The emission signal was then found to decay as a result of the Doppler effect. The above process is analyzed by Bloch and Maxwell equations. This is the first reported observation of the coherence of an atomic system induced by two-photon absorption, a coherence which persists after the passage of the excitation pulse and decays inhomogeneously as a result of thermal motion. In carrying out the experiment, deformation of the input dye-laser pulse during propagation through the two-photon resonant medium was carefully avoided.

Two-photon absorption is now an important tool for studying various properties of electronic excited levels or bands of atoms, molecules, or solids. By the absorption of two photons of laser light, a system of identical atoms is brought into a coherent linear superposition state of the ground and the electronic two-photon excited levels, and the coherence remains within a characteristic relaxation time in the absence of the driving field after the excitation. This coherence, however, has not been well investigated so far. This coherent state is analogous to a superradiant state,[1] or to the state of free precession of a macroscopic dipole moment of two-level atoms, if we consider the precession of a three-dimensional vector constructed by the amplitudes of the ground and the two-photon excited levels.[2] We have for the first time observed this coherence and its inhomogeneous decay in the absence of the driving field. We may call this a free-precession decay (FPD) of two-photon induced coherence. We had to avoid carefully the deformation of the exciting pulse during propagation in the medium in order to see this effect.

We observed this coherence by an anti-Stokes Raman process using an off-resonant interrogation pulse. Its pulse width was about 60 psec, so that observation of FPD in the picosecond regime was allowed. This is a unique feature which is difficult in a one-photon FPD.

Coherence of vibrational or phonon levels after the excitation by the stimulated Raman process was observed in organic liquids and in calcite.[3] In the infrared regime, and where the Stark effect can be used, coherence of two initially degenerate levels was observed in CH_3F.[4] The decay of the coherence in these two cases was determined mostly by the homogeneous transverse relaxation.

In the commonly used alkali-metal vapors it is generally difficult to observe coherence in the absence of a driving field after their excitation by two-photon absorption.[5] The difficulty is due, first, to the modulation of the coherent states by the hyperfine splittings of these atoms. Secondly, strong resonant emissions through energetically lower levels may disturb the coherent states. We avoided these two difficulties by using the singlet system of the calcium atoms.

A partial energy-level diagram of this atom is

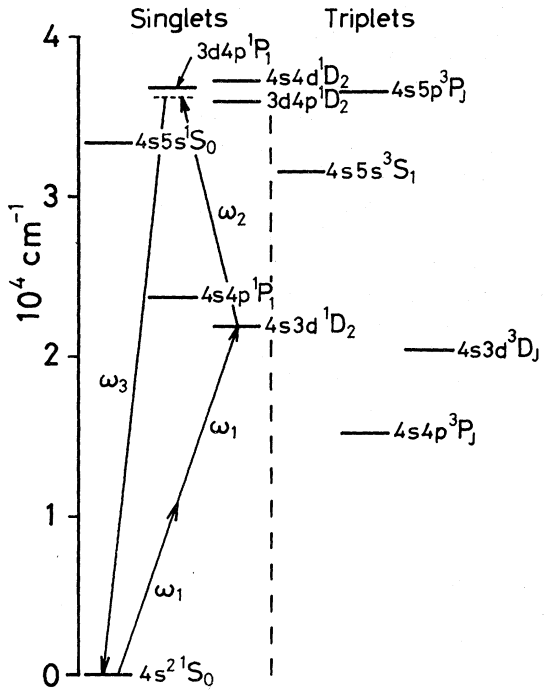

FIG. 1. Energy levels and transitions in calcium.

shown in Fig. 1. The two-photon transition from the ground level $4s^2\,{}^1S_0$ to the lowest singlet excited level $4s3d\,{}^1D_2$ was made via an intermediate level $4s4p\,{}^1P_1$ by absorbing two photons of 9153 Å radiation (frequency ω_1) of a dye laser. A radiative intercombination transition from the $4s3d\,{}^1D_2$ to $4s4p\,{}^3P_J$ level is forbidden. A transfer of the energy from the $4s3d\,{}^1D_2$ to $4s3d\,{}^3D_J$ level by inelastic collision with helium atoms, which were used as a buffer gas, should be also negligible.[6]

Let us denote the $4s^2\,{}^1S_0$, $4s4p\,{}^1P_1$, and $4s3d\,{}^1D_2$ levels by a, b, and c, respectively. The coherence of the linear superposition state of the $4s^2\,{}^1S_0$ and $4s3d\,{}^1D_2$ levels is represented by an off-diagonal matrix element ρ_{ac} of the density operator. The system of these atoms with nonzero ρ_{ac} can have a macroscopic nonlinear electric dipole moment oscillating at a frequency $\omega_3 = 2\omega_1 + \omega_2$ when the interrogation pulse at ω_2 is applied. By examining a phase-matching relation $\vec{k}_3 = 2\vec{k}_1 + \vec{k}_2$, we can determine whether or not the emission is due to the coherence,[7] where $\vec{k}_1$, $\vec{k}_2$, and $\vec{k}_3$ are the corresponding wave vectors. Actually, the signal intensity of this emission is proportional to the squared absolute value of ρ_{ac}. Its magnitude increases from zero and oscillates with increasing excitation. In the gaseous atoms, the phases of the atoms in different locations mix in time due to thermal motion, and ρ_{ac} is decreased. The emitted signal in turn is the summation of Doppler-shifted components, and should show a Gaussian decay with a characteristic decay constant of an inverse Doppler width, if a homogeneous transverse relaxation time is longer than this decay time.

A dye laser with 1, 1′-diethyl-2, 2′-quinotricarbocyanine iodide[8] was pumped by a part of the output of a mode-locked ruby laser. It was tuned to 9153 Å by three Fabry-Perot interferometers. The spectra consisted of 3 to 4 lines with 2.5 cm^{-1} separations. The pulse train of the dye laser consisted of about 15 pulses with 4-nsec separations. The width of a single pulse was estimated to be about 60 psec (FWHM) by the present experiments. The remaining part of the pulse train of the ruby laser, with variable time delay, was used as the interrogation pulse, the peak power of which was about 10 MW. Both beams were focused concentrically on the center of the Ca sample through a 30-cm focal length lens. The beam cross sections at the focus were, however, as large as 0.05×0.02 cm^2 due to large divergence of the laser outputs. The sample was contained in a 55-cm-long heat-pipe oven with the He buffer gas which was at a pressure of about 30 Torr. The temperature of the oven was kept low enough to avoid the deformation of the dye-laser pulse, and was about 986 °K. The central Ca region was about 7 cm long. The signal at 2758 Å (ω_3) was observed with the help of a dispersing prism, a Nikon G250 monochromator and a HTV R106UH photomultiplier, and was integrated over the pulse train. The signal was only observable in the narrow forward direction. The block diagram of the experimental arrangement was similar to the one in Ref. 7 except for the addition of a delay line for the interrogation pulse.

In order to determine a zero-delay position of the interrogation pulse against each dye-laser pulse, and to observe the shape of these pulses, we performed another experiment, simply replacing the Ca oven by a potassium dihydrogen phosphate (KDP) crystal. In this crystal the signal (ω_3) is simply determined by the instantaneous fields.

The detected signal intensities $I_3(t_D)$ of Ca as well as KDP are plotted versus the delay time t_D in Fig. 2, where Ca density N was 2.0×10^{15} cm^{-3}, and the power of the dye laser was about 1 MW. This power roughly corresponds to a $\frac{1}{2}\pi$ pulse of the two-photon transition (I_3 was observed to be proportional to the square of the dye-laser power within this power level). The peak of the Ca signal is well delayed with respect to that of the KDP

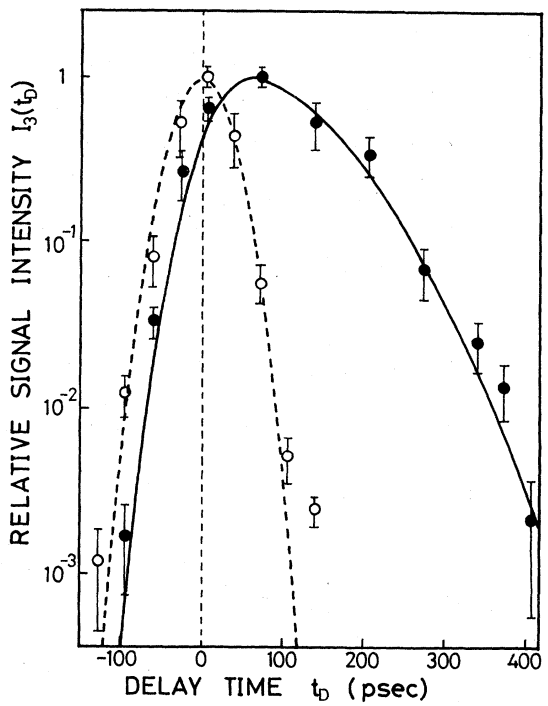

FIG. 2. Measured signal intensities $I_3(t_D)$ of Ca (closed circles) and KDP (open circles) versus the delay time t_D, where the circles and bars are average values and the mean square deviations, respectively, of 2 to 6 shots. Also shown are the theoretical curves for Ca (solid curve) and KDP (dashed curve) by Eqs. (3) and (4). All are normalized at the peaks.

which indicates the peak of the dye-laser pulse. The Ca signal then remains long after the KDP signal has decayed; for example, when the former remains within a factor of 3, the latter has decayed by four orders of magnitude. We performed other experiments, for example at the dye-laser power of about 0.25 MW, but at an increased density of 1.8×10^{16} cm^{-3}. The results were the same within the experimental accuracy.

The signal of Ca vapor is analyzed on the basis of the Bloch-Maxwell equations. The three fields may be expressed by slowly varing amplitudes $E_l(t, z)$ and phases $\phi_l(t, z)$ for each frequency of ω_l ($l=1, 2, 3$). We have to check first of all that the variations in $E_{1,2}$ and $\phi_{1,2}$ during propagation through the medium are negligible when they are seen from a coordinate system moving with the velocity $\omega_1/k_1 \simeq \omega_2/k_2$. These variations play an essential role, however, in propagation effects such as two-photon self-induced transparency.[9,10] Assuming that the length of the medium is L, it is found that E_1 does not change more than a factor of

$$\exp(\pm 2\pi N\omega_1 p_{ab} p_{bc} L/\hbar c\Delta_1),$$

since the imaginary part of ρ_{ac} does not exceed $\mp\frac{1}{2}$, where $\Delta_1 = \Omega_b - \Omega_a - \omega_1$, and $\hbar\Omega_i$ is the energy of the level i, and p_{ij} is the matrix element of the electric dipole moment operator between the levels i and j. E_2 is, however, constant in the moving coordinate system. Since the signal intensity I_3 is proportional to E_1^4, we know that I_3 does not change more than a factor of 1.43(+) or 0.70(−), depending on the sign of the exponential when the density N is 2.0×10^{15} cm^{-3}. This change is negligible compared with the difference between the experimental points of Ca and KDP in Fig. 2. A similar argument shows that the variations in $\phi_{1,2}$ are less than 0.26 rad. We may thus conclude that the decay of the Ca signal is not due to the deformation of the exciting pulse (at least at this density of 2.0×10^{15} cm^{-3}). Then the Maxwell equations give the signal integrated over the width of a single interrogation pulse as[11]

$$I_3(t_D) = \frac{\pi N^2\omega_3^2 L_{\rm eff}^2 A}{2\hbar^2 c}\left|\sum_i \frac{p_{ai}p_{ic}}{\Delta_3}\right|^2 \int_{-\infty}^{\infty} dt\, E_2^2(t-t_D-z/c)\left|\frac{1}{\sqrt{\pi}\,u}\int_{-\infty}^{\infty} dv \exp(-v^2/u^2)\rho_{ac}(v, t-z/c)\right|^2, \quad (1)$$

where $\rho_{ac}(v, t-z/c)$ is an approximation to $\rho_{ac}(v, t, z)$, and represents a component of the density matrix for a group of atoms moving with velocity v along z, $L_{\rm eff}$ is the effective coherence length near the focal point,[12] A is the beam cross section at the focus, and $\Delta_3 = \Omega_i - \Omega_a - \omega_3$. The speed parameter u is expressed by $(2k_BT/m)^{1/2}$, where k_B, T, and m are the Boltzmann constant, the sample temperature, and the atomic mass, respectively.

In order to understand the behavior of the signal with respect to t_D, we found a simple analytic solution of ρ_{ac}. To do this, we took a weak-pulse limit so that $\rho_{aa}=1$, and $\rho_{cc}=0$.[13] Assuming a Gaussian pulse envelope,

$$E_{1,2}(t-z/c) = E_{1,2}(0)\exp\left(-\frac{(t-z/c)^2}{2\tau_{p1,p2}^2}\right),$$

where $\tau_{p1,p2}$ are parameters of the pulse width, we obtain

$$\rho_{ac}(v, t-z/c) = -\frac{ip_{ab}p_{bc}}{4\hbar^2\Delta_1}\int_{-\infty}^{t} dt' E_1(0)^2 \exp\left(-\frac{(t'-z/c)^2}{\tau_{p1}^2}\right)\exp[-2ik_1v(t'-t)]. \quad (2)$$

Equation (1) with Eq. (2) gives the dependence of the signal on t_D as

$$I_3(t_D) \propto \int_{-\infty}^{\infty} dt \exp[-(t-t_D)^2/\tau_{p2}^2]\left(\int_{-\infty}^{t} dt' \exp[-t'^2/\tau_{p1}^2 - k_1^2u^2(t'-t)^2]\right)^2. \quad (3)$$

This result shows that the signal first increases, and then decays as $\exp(-k_1^2u^2t_D^2)$ when $t_D \gg \tau_{p1}, \tau_{p2}$, where $2k_1u$ is the Doppler width at $2\omega_1$. The signal of KDP, on the other hand, is proportional to a simple Gaussian function,

$$I_3(t_D) \propto \exp[-2t_D^2/(\tau_{p1}^2+2\tau_{p2}^2)]. \quad (4)$$

The curves of these two expressions are also shown in Fig. 2 with the parameters $\tau_{p1}=\tau_{p2}=35$ psec, and $T=986$ °K. We have also found that the computer solutions of ρ_{ac} with and without restriction of the weak-pulse limit give no important difference in the shape of the curve. The experimental results are hence in good agreement with our understanding, both in the rise and in the decay of the signal. In addition, it follows that the homogeneous transverse relaxation time is longer than our Doppler decay time, as we have postulated in the beginning.

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[11]The phase ϕ_3 consists of two terms: One is linearly dependent on z, but is small; the other can be large, but is a function of $t-z/c$ (hence the signal is chirped). Equation (1) holds under these conditions.
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