Evidence for electromagnetically induced transparency in a solid medium

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Strong evidence indicating the occurrence of electromagnetically induced transparency (EIT) in a solid medium was found. Fluorescence reduction due to EIT was observed in solids. The excitation intensity dependence of the fluorescence reduction was found to be consistent with theoretical predictions for EIT. It was experimentally demonstrated that the essential condition for the observation of EIT in solids is that the Rabi frequency of excitation must be larger than the inhomogeneous width of the transition between the two states coupled to the third state by the pair of optical fields. [S1050-2947(98)00411-9]

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I. INTRODUCTION

The distinctive characteristics of electromagnetically induced transparency (EIT) are marked changes in absorbance [1] and refractive index [2]. They occur as a result of quantum interference and are unrelated to common bleaching phenomena, saturation effects, or hole burning. Many interesting studies have been reported on potential applications based on these characteristics, including lasing without inversion (LWI) [3–5], quantum noise reduction [6], and sum frequency generation [7].

However, most of the experimental studies on EIT have been carried out in gases. We have come across only two reports on the observation of EIT in solids [8,9], and these were published recently during preparation of this paper. One of the reports describes microwave-induced absorbance changes in ruby [8], while the other describes transmittance changes induced by visible light in Pr^{3+}:Y_2SiO_5 [9], which coincidentally is similar to the material studied in this paper (the selected energy levels and experimental methods are, however, different). As spectral changes thought to be due to EIT in solids have only recently been observed, there is relatively little information about EIT in solids and no convincing evidence as yet to conclusively prove that the observed signals were actually due to EIT. There are also some reports by the Australian National University group on dynamic Stark splitting in a two-level system [10–12] and Autler-Townes splitting in a three-level system [12,13] by the application of radio-frequency fields to an N-defect center in diamond. The observed phenomena, particularly Autler-Townes splitting in a three-level system, are thought to be closely related to EIT even though the destructive interference between the two components of the absorptive profile of the Autler-Townes doublet, which is one of the characteristics of EIT, was not observed. In their experiments, the spectral changes in the radio-frequency region were investigated. From the viewpoint of optical-device applications such as LWI, however, EIT in the optical region for solids is desired.

Using two laser beams whose frequencies differ by the transition frequency of the two lower levels in a three-level system, the system can be pumped into a superposition state of the two levels where the population is trapped due to destructive interference between the two different absorption paths, resulting in suppression of the total absorption. This type of suppression of absorption is the origin of EIT. It is known that a short dephasing time in a medium breaks the atomic coherence of the superposition state (population trapping state) and inhomogeneous broadening reduces the number of the trapped atoms or ions that contribute to EIT. In solids, the inverse of the dephasing time (dephasing rate) and/or inhomogeneous broadening are generally so large that it has been difficult to observe EIT signals clearly.

In this paper we present strong evidence for the occurrence of EIT and population trapping (PT) in a solid medium based on the first observation of fluorescence reduction due to PT in solids. The excitation intensity dependence of the observed fluorescence reduction also clearly revealed that the essential condition for achieving an obvious EIT in a solid medium is that the Rabi frequency of excitation must be larger than the inhomogeneous width of a two-photon transition.

II. EXPERIMENTS

The sample was a $2.5\times3.0\times3.5$ mm Y_2SiO_5 crystal doped with 0.05 at. % Pr^{3+} ions (Scientific Material Corp.). It was set in a continuous gas-flow cryostat (Oxford, Optistat CF) maintained at 3.8 K during the measurement.

Figure 1 shows the energy levels for the $^3H_4$ ground-state hyperfine sublevels and $^1D_2$ excited-state sublevels of Pr^{3+}-doped Y_2SiO_5. The angular frequencies of the incident laser beams (ω_1,ω_2) are shown in the figure. It is known that the Pr^{3+} ions in Y_2SiO_5 occupy two nonequivalent crystallographic sites, site 1 and site 2 [14]. The ions at site 1 have a transition energy of $16\,502.3\ \mathrm{cm}^{-1}$ for the $^3H_4(1)$-$^1D_2(1)$ transition, where $^3H_4(1)$ and $^1D_2(1)$ are, respectively, the lowest crystal-field components of $^3H_4$ and $^1D_2$. The corresponding transition energy for site 2 is $16\,449.2\ \mathrm{cm}^{-1}$. Since the energy difference between these two transitions ($53.1\ \mathrm{cm}^{-1}$) is much larger than the linewidth of each transition [14], it was possible to excite the ions at site 1 independently of the ions at site 2.

A schematic of the experimental setup used for observing transmission and fluorescence related to EIT is shown in Fig. 2. A laser beam from a ring-dye laser (Coherent, model 699-29) pumped by an argon-ion laser (Coherent, Innova

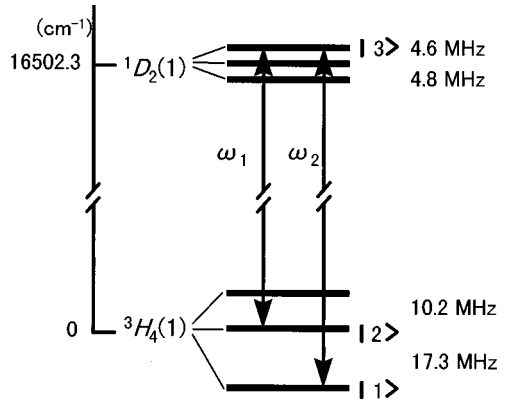

FIG. 1. Energy levels for the $^3H_4$ ground-state sublevels and $^1D_2$ excited-state sublevels for site 1 in Pr^{3+}:Y_2SiO_5. $|1\rangle$ and $|2\rangle$ are the two ground-state sublevels split by 17.3 MHz, and $|3\rangle$ is one of the excited-state sublevels.

400-20) was split into two components by a partially reflecting mirror.

To bring the difference between the two laser frequencies $\omega_1/(2\pi)$ and $\omega_2/(2\pi)$ into resonance with the ground-state splitting frequency of 17.3 MHz, the frequencies of the transmitted (beam 1) and reflected (beam 2) beams were, respectively, shifted by 60 and 77.3 MHz using acoustooptic (AO) modulators (Isomet, D322B). Half-wave plate rotators were inserted into the beam paths, and the orientations of the two linearly polarized beams were adjusted to be parallel to the axis where the sample exhibited strongest absorption. The intensity of beam 1 was varied between 0.2 and 10 W/cm^2, while that of beam 2 was kept constant at 0.1 W/cm^2.

The two beams were overlapped in the sample at an angle of intersection of 2°. The diameter of the two beams in the sample was approximately 450 μm. The intensities of the transmitted light (beam 1) and fluorescence were each detected by two coupled sets of photomultipliers and monochromators. The frequency of the ring-dye laser was resonant with the transition between $^3H_4(1)$ and $^1D_2(1)$ (16502.3 cm^{-1}, 605.98 nm). Monochromators were employed as narrow band filters to eliminate unnecessary light. In the fluorescence measurement, a Stokes line was detected at 611 nm. This line corresponded to the transition from the lowest level of the $^1D_2$ crystal-field components to the third level of the $^3H_4$ components.

Due to frequency jitter, the linewidth of the ring-dye laser was approximately 500 kHz. However, the frequency difference between the two laser beams was set accurately by the AO modulators, and the resolution of the observed spectra was higher than 10 kHz.

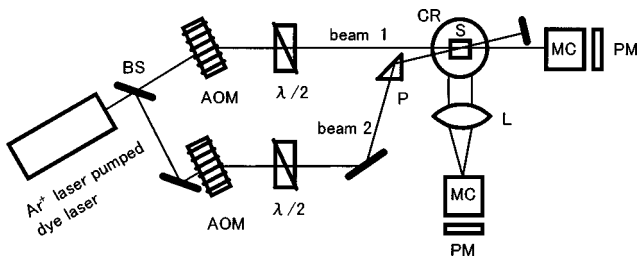

FIG. 2. Schematic of the experimental setup. Abbreviations: BS, partially reflecting mirror; AOM, acoustooptic modulator; λ/2, half-wave plate; *P*, prism; CR, cryostat; *S*, sample; *L*, lens; MC, monochromator; and PM, photomultiplier.

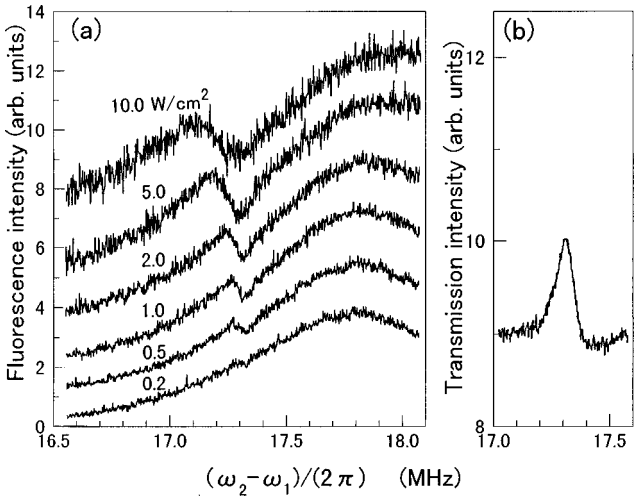

FIG. 3. (a) Fluorescence spectra for six different light intensities. The spectra are arranged by moving baselines for easy comparison of the hole shapes. (b) The transmission spectrum for an excitation light intensity of 1.0 W/cm^2.

To obtain transmission and fluorescence spectra, the frequency of beam 2 $[\omega_2/(2\pi)]$ was scanned using an AO modulator controlled by an rf source (Isomet, 1205C-2), while the frequency of beam 1 was fixed. The observed spectra were detected with a scan speed of 2 MHz/s and were typically averaged 64 times.

III. RESULTS AND DISCUSSION

The fluorescence spectra for six different light intensities of beam 1 are shown in Fig. 3(a). The dip at 17.3 MHz indicates that under the two-photon-resonance condition, the population was trapped in a coherent superposition state. This is the first observation of EIT (or PT) in fluorescence spectra in a solid medium.

In the transmitted spectra, enhancement of transmittance due to EIT was also clearly observed as shown in Fig. 3(b). We would like to stress here that the observation of the change in fluorescence directly indicates the reduction of the higher-state population excited from the ground-state sublevels. It seems to be difficult to separate the contribution of EIT from other effects such as the stimulated emission of Raman signals by measuring only absorbance or transmittance changes in the transmitted light intensity.

Pr^{3+}:Y_2SiO_5 is known as a material which shows hole burning [15]. Therefore, with the long hole lifetimes in the material, one may think that absorption and fluorescence are reduced and the experiment is difficult. Actually the absorbance and fluorescence measured using only beam 1 were reduced due to hole burning in the sample. However, weak absorption and fluorescence could be observed, probably due to the relaxation between ground-state sublevels. When beam 2 was applied in addition to beam 1, the absorption and the fluorescence were enhanced in the vicinity of frequencies that were separated from the beam 2 frequency by the frequency differences of the ground-state sublevels. This hap-

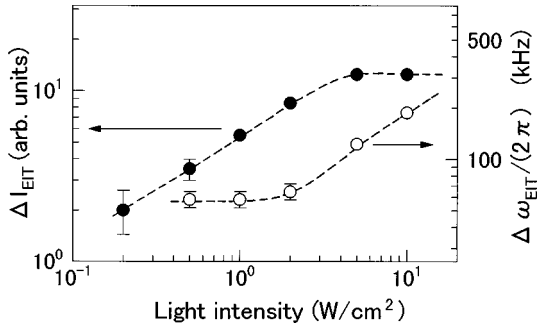

FIG. 4. Light intensity dependence of the hole width (open circles) and depth (closed circles). The broken-line curves have been inserted to guide the reader's eye.

pened because beam 2 pumped a part of the population back to the initial states of excitation by beam 1. The absorption and fluorescence due to beam 2, on the other hand, were enhanced by beam 1. In this enhanced fluorescence spectrum, the dip due to EIT was clearly observed.

The broad background structure in the spectra shown in Fig. 3(a) may be due to hole burning; that is, population redistribution between the ground-state sublevels described above. The approximately 1-MHz width of the broad structure probably corresponds to the homogeneous width of the optical transition under laser irradiation. At present, the reason why the broad structure shows a peak at 17.8 MHz rather than 17.3 MHz is not clear.

Another dip was also observed at 10.2 MHz. Since the dip at 17.3 MHz was larger and clearer than that at 10.2 MHz, our study focused on the former.

Figure 4 shows the dependence of the dip depth and width on the light intensity. The depth was defined here as the fluorescence intensity difference between the maximum at the low-energy side of the dip and the bottom of the dip. The width was defined as the width at half the depth of the dip.

In the low-intensity region below 1.0 W/cm^2, the width did not change but the depth increased with the increase in light intensity. In the high-intensity region above 5.0 W/cm^2, on the other hand, the depth did not change but the width increased with the increase in light intensity. This behavior has great significance, because it provides strong evidence of EIT in a solid medium as explained below.

Pr^{3+} ions that are trapped in the coherent superposition state and exhibit EIT satisfy the two-photon-resonance condition

$$\omega_2-\omega_1=\omega_{12}, \tag{1}$$

where ω_{12} is the angular frequency difference between the two lower states. The limits of the resonance condition can be estimated at the Rabi frequency of the excitation light. This condition is represented as

$$|\omega_2-\omega_1-\omega_{12}|<\Omega/2, \tag{2}$$

where Ω is the Rabi frequency. Here, the excitation light is defined as the beam that has a higher Rabi frequency, and in this report beam 1 corresponds to the excitation light. The estimates of the limits were obtained from experiments [16] and theory [4,17]. They indicate that the dip width of EIT in the absorption and fluorescence spectra is approximately equal to the Rabi frequency of excitation when inhomogeneous broadening can be ignored. Judging from the limits of the resonance condition, when the Rabi frequency of beam 1 (Ω_1) is sufficiently smaller than the inhomogeneous width of the transition $|1\rangle$-$|2\rangle$ ($\Delta\omega_{12\text{inhomo}}$), the dip width of EIT ($\Delta\omega_{EIT}$) measured by scanning ω_1 or ω_2 is expected to be $\Delta\omega_{12\text{inhomo}}$. When $\Delta\omega_{12\text{inhomo}}\ll\Omega_1$, the dip width is expected to be Ω_1. Here, $|1\rangle$, $|2\rangle$, and $|3\rangle$ are the hyperfine sublevels shown in Fig. 1.

The resonance condition (2) is valid when the rate of dephasing of the coherence between $|1\rangle$ and $|2\rangle$ is sufficiently smaller than Ω_1. Therefore, the Ω_1 dependence of $\Delta\omega_{EIT}$ described above is expected only when the dephasing rate is sufficiently smaller than Ω_1.

Let us consider the dip depth ΔI_{EIT} in the fluorescence spectra. Assuming that the fluorescence is almost saturated by beam 1, ΔI_{EIT} is proportional to the number of Pr^{3+} ions that are in the PT state. Therefore, when $\Omega_1\ll\Delta\omega_{12\text{inhomo}}$, ΔI_{EIT} is proportional to Ω_1, and when $\Delta\omega_{12\text{inhomo}}\ll\Omega_1$, ΔI_{EIT} is constant.

The Rabi frequency is proportional to the square root of the intensity of the excitation light. This can be expressed as

$$\Omega_1=2\pi\mu_{23}E/h\propto I^{1/2}, \tag{3}$$

where μ_{23} is the transition dipole moment of the $|2\rangle$-$|3\rangle$ transition, E is the electric field of beam 1, h is the Planck constant, and I is the light intensity of beam 1. The dependence of the dip width and depth on the light intensity can therefore be expressed as

$$\Delta\omega_{EIT}=\Delta\omega_{12\text{inhomo}}(\Omega_1\ll\Delta\omega_{12\text{inhomo}}), \tag{4}$$

$$\Delta\omega_{EIT}=\Omega_1\propto I^{1/2}(\Delta\omega_{12\text{inhomo}}\ll\Omega_1), \tag{5}$$

$$\Delta I_{EIT}\propto I^{1/2}(\Omega_1\ll\Delta\omega_{12\text{inhomo}}), \tag{6}$$

and

$$\Delta I_{EIT}=\text{const}(\Delta\omega_{12\text{inhomo}}\ll\Omega_1). \tag{7}$$

The logarithmic plots of the light intensity dependence in Fig. 4 show that $\Delta\omega_{EIT}$ is constant for low light intensities [i.e., in the low Rabi frequency region ($I<1$ W/cm^2)], but changes along a line of slope 1/2 at high light intensities [i.e., in the high Rabi frequency region (2 W/cm$^2<I$)]. The Rabi frequency $[\Omega_1/(2\pi)]$ where $\Delta\omega_{EIT}$ begins to increase is approximately 100 kHz, which is comparable to $\Delta\omega_{12\text{inhomo}}/(2\pi)$ of 70 kHz reported by Holliday *et al.* [15]. The dip depth ΔI_{EIT}, on the other hand, changes along a line of slope 1/2 in the low light intensity region but is constant in the high-light-intensity region. A likely reason for the slope of the observed ΔI_{EIT} curve being slightly steeper than the theoretical value of 1/2 is that the fluorescence was not saturated completely.

Equation (4) indicates that the dip width in the low-intensity region should agree with the inhomogeneous width $\Delta\omega_{12\text{inhomo}}$. The observed dip width of 60 kHz in the region is actually consistent with the inhomogeneous width of 70 kHz.

Equation (5) predicts that, in the high-intensity region, $\Delta\omega_{EIT}$ should be approximately equal to Ω_1. The observed

dip width $[\Delta\omega_{EIT}/(2\pi)]$ of 200 kHz in the region is comparable to the calculated Rabi frequency $[\Omega_1/(2\pi)]$ of 300 kHz. The Rabi frequencies were calculated using an oscillator strength of 3×10^{-7} [14], a measured beam diameter of 450 μm, and measured light intensities.

The dip depth in the high-light-intensity region can be estimated as follows. The inhomogeneous width of the $^3H_4(1)$-$^1D_2(1)$ transition is much larger than the energy separation of the sublevels. Therefore, when the sample is excited by beam 1 and beam 2 and detuning from the two-photon-resonance condition is larger than $\Omega_1/2$ (i.e., $\Omega_1/2 < |\omega_2-\omega_1-\omega_{12}|$), all nine transitions between the three $^3H_4(1)$ sublevels and the three $^1D_2(1)$ sublevels contribute to absorption at ω_1 and ω_2. Therefore there are eighteen combinations of incident light and resonant transitions that cause absorption. However, when condition (2) is satisfied, the six combinations that include the two sublevels satisfying the condition do not contribute to the absorption because of PT. Assuming that the nine transition probabilities are the same, the reduction of absorption and fluorescence in the case of two-photon resonance is expected to be six divided by eighteen; that is, 33%. This estimate is independent of the ratios of the intensities of beam 1 and beam 2, because each contribution of beam 1 and beam 2 to the absorption becomes one-third under condition (2).

The estimate of 33% is valid when population redistribution (hole burning) can be ignored. Population redistribution in the actual sample may increase the contribution of the six combinations to the absorption when $\Omega_1/2 < |\omega_2-\omega_1-\omega_{12}| < \Delta\omega_{homo}$, where $\Delta\omega_{homo}$ is the homogeneous broadening of the $^3H_4(1)$-$^1D_2(1)$ transition. This is because beam 1, which connects levels $|2\rangle$ and $|3\rangle$, tends to increase the population in $|1\rangle$ of the ions that have $|1\rangle$-$|3\rangle$ transitions resonant with beam 2 by pumping the population via $|3\rangle$, and beam 2 tends to increase the population in $|2\rangle$ of the ions that have $|2\rangle$-$|3\rangle$ transitions resonant with beam 1. Since these six combinations cause the dip under condition (2), the value of 33% represents the minimum expected dip.

The actually observed reduction of fluorescence was approximately 10%. This discrepancy is thought to be caused by transition probability differences among the nine transitions and by fluorescence from excited parts of the sample where the two beams did not overlap each other.

The excitation intensity dependence of the observed dip width and depth is quantitatively well described by Eqs. (4) to (7) for different ratios of Ω_1 to $\Delta\omega_{12\text{inhomo}}$. These results clearly show that the observed dip was due to EIT.

The obtained results are also significant from another point of view. They experimentally confirm that the essential condition for achieving large EIT in a solid medium that has an inhomogeneously broadened spectrum is

$$\Delta\omega_{12\text{inhomo}} < \Omega. \tag{8}$$

It is important to note that it is possible to achieve large EIT with any inhomogeneous width for transitions that are resonant with the incident light, as long as condition (8) is satisfied. Under this condition, the solid medium exhibits a transition-energy distribution equivalent to the effective transition-energy distribution of atomic vapors to which the Doppler-free technique using two-photon resonance [18] is applied.

The dephasing time of the coherence between $|1\rangle$ and $|2\rangle$ could be estimated as follows. The rate of dephasing is expected to be sufficiently smaller than Ω_1 during our experiment, because the data in Fig. 4 are well explained by Eqs. (4) to (7). At the low excitation intensity of 0.5 W/cm^2, Ω_1 and the dephasing rate should be smaller than the dip width of 60 kHz. Therefore, the dephasing time of the ground-state sublevels should be longer than 3 μs because $1/(2\pi \times 60$ kHz) is approximately 3 μs. Equall, Cone, and Macfarlane observed the excitation intensity dependence of the homogeneous line width of the transition between $^3H_4(1)$ and $^1D_2(1)$ in Pr^{3+}:Y_2SiO_5 [14]. According to their results, the homogeneous width at the intensity of 0.5 W/cm^2 is approximately 2.5 kHz, and this reflects the dephasing time of the sublevels under their experimental condition. The concentration of Pr^{3+} ions in their sample (0.02%) was smaller than that in ours (0.05%), and the temperature of their sample (1.4 K) was lower than that of ours (3.8 K). Therefore, the dephasing time of our sample is shorter than the 70 μs expected from the 2.5-kHz width $[1/(2\pi\times2.5\text{ kHz})\cong70\ \mu\text{s}]$. At the high excitation intensity of 10 W/cm^2, the dip width that was observed in our study was 200 kHz, and Equall, Cone, and Macfarlane report a homogeneous width of 5 kHz at that intensity. This indicates that the dephasing time in our sample at a high excitation intensity is likely to be between 1 and 30 μs. Therefore, during our experiment, the dephasing time of the ground-state sublevels was probably around a few tens of μs.

Single crystals doped with rare-earth ions were employed as candidate materials meeting condition (8) because they exhibit extremely small inhomogeneous broadening of transitions between the hyperfine sublevels in a common crystal-field component. Before this study, a few rare-earth-ion doped samples had been investigated to obtain EIT signals at liquid helium temperature. In fact, three-level systems selected from six $^3H_4(1)$ and $^1D_2(1)$ hyperfine levels in Pr^{3+}:$YAlO_3$ were studied using the same experimental setup as this study. However, no clear EIT signals were observed. The dephasing times of the hyperfine levels in the samples other than Pr^{3+}:Y_2SiO_5 were probably too short to maintain a coherent superposition state, particularly under laser irradiation.

IV. CONCLUSION

EIT was observed in a solid medium using the fluorescence spectrum. Unlike the transmission spectrum, a dip in the fluorescence spectrum directly indicates the suppression of excitation from ground-state sublevels to the excited state. The dependence of the dip shape on the light intensity was observed, and the behavior could be explained quantitatively by theoretical predictions related to EIT in a solid medium. This clearly shows that EIT and PT were achieved in a solid medium. It was also experimentally confirmed that for a solid medium with inhomogeneous broadening that cannot be canceled by the multiphoton Doppler-free technique, the essential condition for achieving a large EIT is $\Delta\omega_{12\text{inhomo}} < \Omega$.

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