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Electromagnetically induced inhibition of two-photon absorption in sodium vapor

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We report the observation of the electromagnetically induced inhibition of two-photon absorption with atomic sodium vapor as a test medium. A 60% reduction of two-photon resonant absorption was recorded at the 3S-4D transition when a cw coupling laser was applied at the 3P-5S transition.

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Electromagnetically induced transparency (EIT) is a modification of the optical properties of a medium by a strong coherent field. The studies on EIT have met with tremendous progress since the idea was proposed and demonstrated by Harris and co-workers [1,2]. EIT phenomena are closely related with lasing without inversion [3,4] and nonlinear optical processes [5–8]. A number of theoretical and experimental works have been devoted to various schemes where a single photon as well as multiple photons of EIT have been observed [5–13]. In this paper we demonstrate a scheme of EIT where a rather strong electromagnetically induced inhibition of two-photon absorption (EIITA) can be realized with a cw dye laser as the coupling field.

It was well known that two-photon absorption can be greatly enhanced whenever a single photon energy approaches that of an allowed atomic transition [14]. If more than one intermediate state exists, the two-photon absorption can be annihilated depending on the location of the intermediate states. This phenomenon is explained in terms of the destructive interference since there are two pathways leading to the same excited state. The annihilation of two-photon absorption can never occur when there is only one intermediate state [15]. In this case, a control laser can be applied to couple the intermediate state with another state to create two dressed states, states of the field-atom system. Then, the medium can be made transparent against two-photon absorption in the presence of multiple pathways [16]. The modification of two-photon absorption by one coherent field may have potential applications like two-photon lasing and enhancing $\chi^{(2)}$ in some media.

Considering the four-level system in Fig. 1, ω_1 , ω_2 are the frequencies of two laser fields whose sum equals the frequency seperation between state $|1\rangle$ and state $|3\rangle$, ω is the frequency of the coupling field between the transition $|2\rangle$ and $|4\rangle$, Δ_1 , Δ_2 , and Δ are corresponding detunings of the three fields from their resonance, respectively.

The equation of motion for the atomic density operator $\boldsymbol{\rho}$ in the interaction picture reads

$$\partial \rho / \partial t = -i\hbar [H, \rho] + \Lambda \rho, \tag{1}$$

where $\Lambda \rho$ indicates the irreversible contribution which includes the population and polarization decay terms due to the spontaneous emission. H is the Hamiltonian related to the

interaction of the two fields ω_1 , ω_2 and the strong coherent field ω with a single atom in the interaction picture

$$H = \Delta_1 |2\rangle\langle 2| + (\Delta_1 + \Delta_2)|3\rangle\langle 3| + (\Delta_1 + \Delta)|4\rangle\langle 4|$$

$$-G(|2\rangle\langle 4| + |4\rangle\langle 2|) - G_1(|1\rangle\langle 2| + |2\rangle\langle 1|)$$

$$-G_2(|2\rangle\langle 3| + |3\rangle\langle 2|), \tag{2}$$

where $G = \vec{\mu} \cdot \vec{E}/2\hbar$, $G_1 = \vec{\mu}_1 \cdot \vec{E}_1/2\hbar$, $G_2 = \vec{\mu}_2 \cdot \vec{E}_2/2\hbar$ are the Rabi frequencies of the coherent field and the two weak fields, respectively. Taking the Doppler broadening caused by the atomic motion into account, the frequencies of the three fields in the atomic frame are velocity dependent. In case of a geometrical arrangement where ω_1 and ω_2 are counterpropagating, and ω is copropagating with ω_2 , the frequency dependence is given by

$$\omega_1(v_z) = \omega_1(1 - v_z/c),$$

$$\omega_2(v_z) = \omega_2(1 + v_z/c),$$

$$\omega(v_z) = \omega(1 + v_z/c),$$
(3)

where v_z is the atomic velocity component along the propagating direction of field ω_1 . The entire population in state $|3\rangle$ can be calculated according to

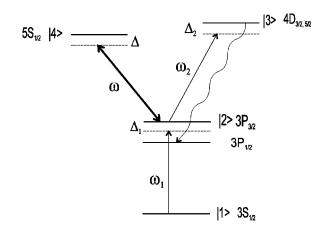


FIG. 1. Related energy-level scheme for electromagnetically induced inhibition of two-photon absorption in atomic sodium. Δ_1, Δ_2 , and Δ are the detunings of the fields ω_1, ω_2 , and ω from the corresponding transitions, respectively.

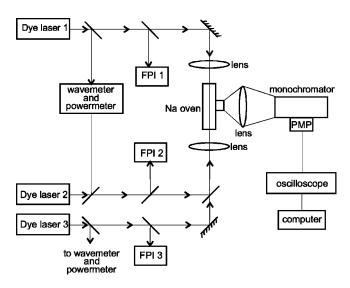


FIG. 2. Experimental setup.

$$\langle \rho_{33}(v) \rangle = \int \rho_{33}(v) P(v) dv$$
 (4)

where P(v) is the atomic velocity distribution function. In the numerical simulation, the population in the upper state $|3\rangle$, ρ_{33} , was monitored and its value, which is proportional to the two-photon absorption cross section, was calculated as a function of the frequency detuning Δ_2 while the two-photon absorption condition, $\Delta_1 + \Delta_2 = 0$, was kept fixed.

The experimental arrangement is shown in Fig. 2, two single-frequency cw dye lasers pumped by argon iron lasers passed in opposite directions through a 10-mm sodium vapor cell kept at temperature 230 °C. One of the dye lasers was Ametist SF-03 type with an automatic frequency scanning system. The maximal frequency scanning range is 15 GHz. The wavelengths of these two lasers were tuned close to 568.8 and 589.0 nm which correspond to the $3P_{3/2}$ - $4D_{5/2,3/2}$ and $3S_{1/2}$ - $3P_{3/2}$ transitions of atomic sodium, respectively. The power of each beam is about 5 mW at the entrance to the sodium vapor cell. The Rabi frequencies of ω_1 and ω_2 are 0.028 and 0.03 GHz, respectively. The coupling laser was another single-frequency dye laser of wavelength 616.0 nm with output power of 150 mW at the cell position, pumped by an argon iron laser (Inversion, Ar-6-100/SM). The coupling laser beam propagated in the same direction of the 568.8 nm laser. Two lenses with a focal length of 25 cm were applied to focus the laser beams onto the cell center. Interferometers were used to monitor the output of each dye laser to make sure that there were no mode jumps during the measurements. The linewidth of each dye laser is about 5 MHz.

Two-photon absorption from $3S_{1/2}$ to $4D_{3/2,5/2}$ was measured by detecting the fluorescence at 568.3 nm $(4D_{3/2} \rightarrow 3P_{1/2})$ using a RS666 photomultiplier as the detector. A monochromator was set in front of the detector in order to filter the spurious light. The signals from the photomultiplier were recorded by a TDS220 digital oscilloscope and processed by a computer.

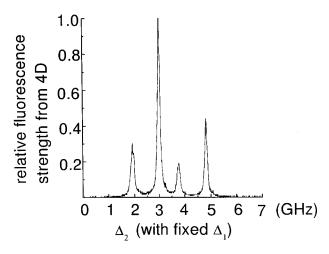


FIG. 3. Two-photon absorption spectra of $3S_{1/2}$ - $4D_{3/2,5/2}$ transitions with fixed ω_2 and scanned ω_1 . The four peaks correspond to the transitions $3S_{1/2}(F=2)$ - $4D_{5/2}$, $3S_{1/2}(F=2)$ - $4D_{3/2}$, $3S_{1/2}(F=1)$ - $4D_{5/2}$, and $3S_{1/2}(F=1)$ - $4D_{3/2}$, respectively.

In order to get the two-photon absorption signal, the frequency of the dye laser 2, ω_2 , was fixed and the frequency of dye laser 1, ω_1 was scanned to make $\omega_1 + \omega_2$ across the two-photon transition from $3S_{1/2}$ to $4D_{3/2,5/2}$. One of the two-photon absorption spectra from the osilloscope is shown in Fig. 3 where four two-photon absorption peaks were recorded. For our subsequent measurements we selected the left peak which corresponds to the transition $3S_{1/2}(F=2) \rightarrow 4D_{5/2}$. To demonstrate the reduction of the two-photon absorption, a number of runs similar to those shown in Fig. 3 were carried out, each with a different frequency ω_2 . The results are shown by the open circles in Fig. 4, where the power fluctuations of the three lasers have been taken into account during the data processing. It is clear that about 60% reduction of two-photon absorption was realized at line cen-

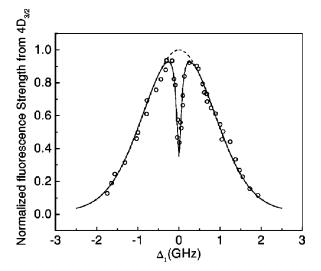


FIG. 4. Normalized two-photon absorption strengths vs the detuning of ω_2 , Δ_2 , when the two-photon condition $\Delta_1 + \Delta_2 = 0$ was kept. The circles are experimental and the open curves are theoretical. Parameters used in the theoretical simulation are $\Delta = 0$, $G_1 = G_2 = 0.03$; G = 0 for the dashed curve, G = 0.1 for the solid curve.

ter in the presence of coupling laser field. The results of the theoretical simulations according to equation (4) are also shown in Fig. 4, where the two-photon absorption strengths as a function of detuning Δ_1 are shown by the dashed and solid curves in the cases of absence and presence of driving laser field, respectively. Both theoretical curves and experimental data have been normalized to their own maximal values when there is no coupling field applied. In our simulations, the Doppler effect was taken into account. At temperature 230 °C, if ω_1 and ω_2 are copropagating, the theory result of the Doppler width of the two-photon transition is about 3.5 GHz. To reduce the Doppler effect, we make use of the two-photon nature of the transition to arrange these three beams as shown in Fig. 2. The residual Doppler width of the two-photon transition is about 60 MHz in this experiment, so it is possible to observe a clear reduction of two-photon absorption in a Doppler-broadened medium with a much smaller Rabi frequency of the coupling field compared to the Doppler width of the two-photon transition. In this experiment, the Rabi frequency of the coupling field is estimated to be 0.1 GHz.

In conclusion, we have reported that the two-photon absorption has been greatly reduced in this scheme with a cw tunable dye laser as the coupling field. We took the advantage of a Doppler-free configuration in the four-level system, so that the two-photon absorption reduction can be observed with the Rabi frequency of the coupling laser field much lower than the Doppler width of the two-photon transition.

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