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Enhancing electromagnetically-induced transparency in a multilevel broadened medium

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We show that electromagnetically-induced transparency in a room-temperature vapor of alkali-metal atoms can be enhanced via a specific shaping of the atomic velocity distribution. In such a medium, the interplay between doppler broadening and the existence of several excited levels may lead to a vanishing transparency, which cannot be predicted in a generic Λ -type three-level model. Here we identify the underlying physical mechanisms resulting in this phenomenon and evidence some velocity classes turning transparency into absorption. By excluding these specific atoms from the interaction process, an enhancement of the transparency contrast by a factor 5 is thereby demonstrated, with potential applications for metrology and quantum information processing.

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Quantum interference effects in the amplitudes of optical transitions in atomic medium can lead to strong modifications of its optical properties. For instance, a light field resonant with an atomic transition is usually strongly absorbed in a dense medium but, in the presence of an additional driving field, it can be totally transmitted. This coherent effect, well-known as electromagnetically induced transparency (EIT) [1–3], has become an important tool to control the optical properties of dense media. Among the capabilities enabled by EIT or related phenomena are high precision magnetometry [4], lasing without inversion [5], and slowing [6] and stopping of light pulses [7, 8]. These possibilities opened new avenues for optical information storage and quantum information processing. Recent experiments based on dynamic EIT have demonstrated reversible mapping of single-photons or qubits [9–11] and of quantum continuous variables [12–14]. These seminal demonstrations spurred intense experimental and theoretical efforts to improve the efficiency of such processes and extend them to new enabling photonic technologies.

An important effort concerns indeed the modeling of the EIT process in the non-ideal case. The EIT configuration is usually modeled by a generic Λ -type three-level system [3]: two atomic ground states are connected to an excited state via two optical fields, a probe and a control field. However, in many optically dense media, the relevant energy structure is more complex and can strongly modify the EIT features. A typical case is the use of ensembles of alkali-metal atoms in which experiments have been most performed [15, 16]. The hyperfine interaction in the excited state introduces several levels, which can simultaneously participate in the coherent interaction. The deviation from the Λ -type approximation can be very significant when the inhomogeneous broadening is comparable with the separation between these excited levels, such as for example in the D_2 -line of atomic cesium at room temperature. The observed transparency

is generally lower than predicted [14, 17] and can eventually disappear for large broadening as observed in [18]. Various numerical analysis have investigated particular regimes, such as double- Λ system [19] or off-resonant Raman transition in a broadened medium [20]. However, to date no full study of EIT in inhomogeneously broadened medium with multiple excited levels has been performed.

In this paper, we report measurements that provide a detailed picture of EIT in such a medium. In agreement with the general model recently developed in Ref. [21], we evidence the process leading to a reduced transparency and we experimentally demonstrate how to mitigate this effect. Our observations are made possible by identifying atoms from specific velocity classes that absorb the light in the process and by then reshaping accordingly the atomic velocity distribution. This procedure enables to recover a significant transparency.

We begin by discussing the effect of inhomogeneous broadening on the EIT features. In a Λ -type three-level system, the susceptibility of the medium is strongly modified when a driving field is applied to one of the transition. In the absence of broadening, the transmission of a probe field through the atomic medium exhibits two symmetrical absorption peaks as a function of its detuning from the resonance. This feature corresponds to the well-known Autler-Townes doublet [22], defining a so-called "transparency window" at resonance. In an inhomogeneously broadened medium, the absorption spectrum of the various atoms differs from the previous description as they are involved in off-resonant processes. As the susceptibility of the medium is obtained by integrating over all the atoms, the EIT spectrum can be drastically modified.

Specifically, in a Doppler broadened medium, the atomic population is distributed over a wide range of velocity classes. For an atom moving with velocity \mathbf{v} and copropagating control and probe fields, the two laser frequencies are Doppler shifted in the atom rest frame by

approximately the same detuning $2\pi \cdot \Delta_{Doppler} = -\mathbf{k} \cdot \mathbf{v}$ where \mathbf{k} is the wave-vector of the fields. The two-photon detuning, defined as $2\pi \cdot \Delta_{2ph} = \omega_{probe} - \omega_{control} + \omega_{gs}$ where ω_{probe} and $\omega_{control}$ are the field frequencies and ω_{gs} the splitting between the two atomic ground states, does not depend on the velocity class. Figure 1(a) gives the absorption as a function of the two-photon detuning for different Doppler shifts when the control field is resonant for atoms at rest. In this figure, we have considered the case of atomic velocities opposite to the laser propagation direction: $\Delta_{Doppler}$ is then positive. While for atoms at rest one can observe the usual Autler-Townes doublet, the spectrum is modified when the Doppler shift increases, i.e. the two absorption peaks are not symmetrical anymore. The peak with $\Delta_{2ph} < 0$ corresponds to the one-photon absorption resonance and the second peak ($\Delta_{2ph} > 0$) corresponds to the Raman absorption process, and it is Stark shifted from the zero two-photon detuning. When the Doppler detuning increases, this Raman peak gets closer to $\Delta_{2ph} = 0$. However, it is important to note that it never reaches this specific point. As a result, transparency is preserved at the zero two-photon detuning for all the velocity classes. The integration over the whole velocity distribution thus preserves the transparency at resonance but leads to an important reduction of the transparency window width.

When one takes into account multiple excited levels, the susceptibility in the driven system is very different. Indeed, the presence of other levels induces additional Stark shifts that move the Raman resonance from the position described in the Λ configuration. Based on the model developed in [21], Figure 1(b) shows for instance the case of two excited levels, $|e\rangle$ and $|e'\rangle$. The strong modification of the susceptibility can be understood as follows. In this multiple level scheme, two velocity classes now see the control field on resonance but for different levels: $|e\rangle$ for zero Doppler shift and $|e'\rangle$ for a shift equal to the separation between the two levels. These two classes exhibit a quasi symmetrical Autler-Townes doublet centered on their respective atomic transition, i.e. on the zero two-photon detuning shown on Fig. 1(b). For atoms with intermediate velocities, the Raman absorption undergoes Stark shifts due to the two excited states and eventually crosses the $\Delta_{2ph} = 0$ line, as illustrated in Fig. 1(b). While transparency was always preserved at resonance in the Λ model, here there is no longer any value of the detuning for which atoms are transparent independently of their velocity. Consequently, if the broadening is comparable with the hyperfine splitting, the integration over all the velocity classes can result in a total disappearance of the EIT, as shown here.

Our model also enables to identify the atoms which strongly modify the EIT feature. By taking into account the dipole matrix elements and hyperfine splittings in the excited state, the Doppler shift corresponding to the atoms absorbing in the center of the expected trans-

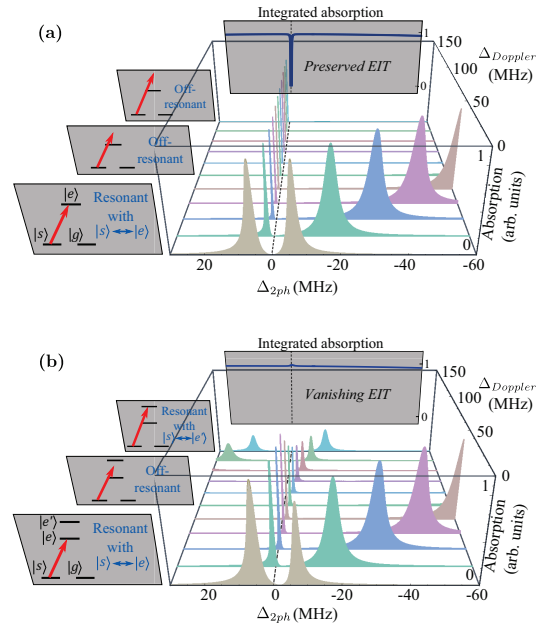


FIG. 1: (color online). Calculated probe absorption for atoms with different velocities, given here by the Doppler detunings $\Delta_{Doppler}$, as a function of the detuning Δ_{2ph} between the control and the probe fields. Panel (a) corresponds to a Λ -scheme interaction with two ground states and one excited state while panel (b) takes into account an additional excited state. For both cases, the integrated absorption is obtained for a Gaussian velocity distribution of 160 MHz half-width, which corresponds to the thermal distribution for Cesium atoms at $T=300$ K. While the 3-level model predicts a narrow but preserved transparency window at zero two-photon detuning, the 4-level model leads to a vanishing transparency. $|s\rangle$ and $|g\rangle$ denote levels $|6S_{1/2}, F=3, m=1\rangle$ and $|6S_{1/2}, F=3, m=3\rangle$, while $|e\rangle$ and $|e'\rangle$ correspond to levels $|6P_{3/2}, F=2, m=3\rangle$ and $|6P_{3/2}, F=3, m=3\rangle$ in atomic cesium. The control Rabi frequency is equal to $\Omega = 2\pi \times 12$ MHz.

parency window can be determined. In our configuration, it corresponds to a Doppler detuning $\Delta_0 \simeq 45$ MHz, as can be seen in Fig. 1(b). By excluding these specific atoms from the interaction process, the effect can be mitigated, as we will show.

We now turn to our experiment. The setup is illustrated in Fig. 2. The optically dense medium is obtained from a warm vapor of ^{133}Cs heated at 35°C in a paraffin-coated cylindrical glass cell (3 cm long and 3 cm in diameter). At this temperature, the Doppler broadening reaches a half-width equal to 160 MHz. The cell is placed in a longitudinal magnetic field produced by sets of coils and the system is enclosed into a magnetic shielding composed of three layers of μ -metal. Residual magnetic fields are smaller than 0.2 mG, with a homogeneity better than 1:700. The scheme of the light-atom interaction is given in the inset of Fig.2. The two ground states $|s\rangle$ and $|g\rangle$ are the two Zeeman states $|6S_{1/2}, F=3, m=1\rangle$ and $|6S_{1/2}, F=3, m=3\rangle$ sepa-

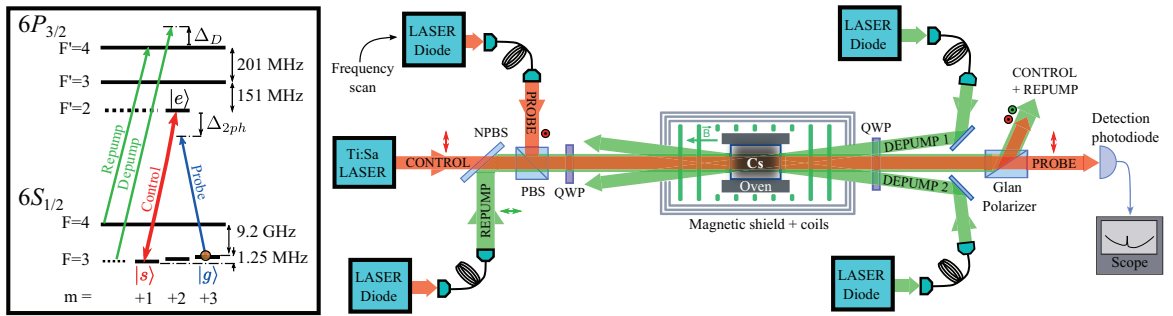


FIG. 2: (color online). Experimental measurement of induced transparency. The transmission of a cesium vapor is probed by scanning the frequency of a weak σ^- -polarized probe field. The strong σ^+ -polarized control field is kept on resonance with the $|s\rangle$ to $|e\rangle$ transition. A repumper beam enables to efficiently prepare the atoms in the $|g\rangle$ ground state. Two σ^+ -polarized depumpers beams can be used to burn holes in the velocity distribution in order to exclude atoms with specific Doppler shifts from the interaction process. The inset gives the relevant energy diagram for the $6S_{1/2} \rightarrow 6P_{3/2}$ transition in atomic cesium, as well as the associated light fields. PBS and NPBS : polarizing and non-polarizing beam splitter, QWP: quarter-wave plate.

rated by 1.25 MHz. The excited state is the Zeeman state $|e\rangle = |6P_{3/2}, F = 2, m = 3\rangle$. As explained previously, this simple Λ system is strongly influenced by the other excited states, i.e. $|6P_{3/2}, F = 3\rangle$ and $|6P_{3/2}, F = 4\rangle$, which are respectively 151 MHz and 352 MHz from the $|e\rangle$ state. The control field is σ^+ -polarized and resonant with the transition $|g\rangle \rightarrow |e\rangle$ for non-moving atoms, while the probe field is σ^- -polarized and addresses the transition $|s\rangle \rightarrow |e\rangle$. Transmission spectra are obtained by scanning the detuning of the probe field, which identifies in this configuration to the two-photon detuning Δ_{2ph} .

The experiment is performed in the continuous-wave regime, with all the fields on. The control field tends to pump all the atoms into the Zeeman sublevel of maximum m of the $F = 3$ level. However, off-resonant pumping through the excited state $|6P_{3/2}, F = 3\rangle$ in the presence of large Doppler shifts can eventually drive all the atomic population into the $|F = 4\rangle$ dark state. To prevent this depumping, an additional σ^+ repump field is used on the transition $|F = 4\rangle \rightarrow |F' = 4\rangle$. In this way, a significant fraction of the atoms are maintained in $|6S_{1/2}, F = 3\rangle$, with 90% in $m = 3$ [14].

Control and probe fields are collimated, with a diameter of 5 mm, and respective powers of 200 mW and 150 nW. They are first combined on a polarizing beam splitter and then pass through a quarter-wave plate to enter the cell with circular polarization. The probe field is then extracted using a quarter-waveplate and a Glan polarizer with high extinction ratio (10^6). The repump beam has a slightly larger beam size and its power is adjusted during the experiment to keep the optical density constant independently of the other experimental conditions.

As explained above, the disappearance of the EIT is predicted to be due to some specific velocity classes. In order to exclude these atoms from the interaction, we use one or two additional depumping beams close to the $|F = 3\rangle \rightarrow |F' = 4\rangle$ transition. They enable to burn specific holes in the velocity distribution. For experimental

convenience, they are contra-propagative with the control and probe fields, with a small angle around 3° . In order to remove atoms that see the probe field frequency shifted by Δ_0 , a counterpropagating depumping beam must have a frequency detuned by $\Delta_D = \Delta_0$ from the $|F = 3\rangle \rightarrow |F' = 4\rangle$ transition for non-moving atoms. The atoms are then efficiently pumped into the hyperfine ground state $F = 4$ and they do not contribute anymore to the interaction.

Figures 3(a) and 3(b) present the experimental measurements and theoretical simulations of the transmission spectrum, with and without reshaping of the atomic velocity distribution. Curve (1) shows a very weak EIT peak near $\Delta_{2ph} = 0$ as expected in such a broadened medium with several excited states. To confirm our model and recover the transparency, we performed two experiments with one and two depumping beams. The results are given in Fig. 3(a), curve (2) and (3) respectively. With such depumping beams, broad peaks appear in the Doppler broadened absorption spectrum. They are centered on the beam detuning and correspond to the holes burnt in the velocity distribution. Spectrum (2) is obtained with a single depumping beam with a detuning $\Delta_D = 40$ MHz, close to the predicted velocity class leading to the strongest absorption. As can be seen, the EIT peak is in this case significantly enhanced, confirming that the EIT is recovered when the detrimental effect of these atoms is suppressed. Adding a second depumping beam ($\Delta_D = 85$ MHz) increases again the transparency but its effect is not as strong as the previous one, which confirms that the absorbing atoms are in a narrow zone of the atomic velocity distribution.

These results are in very good agreement with the transmission curves calculated from the model developed in Ref. [21], as shown in Fig. 3(b). In this case, we have used the velocity distributions extracted from the experimental data to compute the susceptibility of the atomic ensemble. It can be seen that the effect of one and two

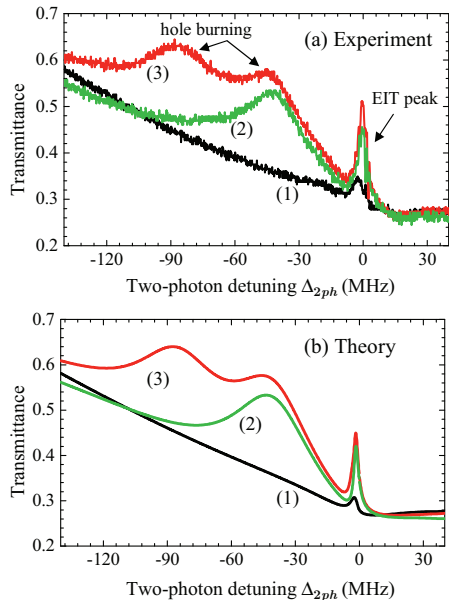


FIG. 3: (color online). (a) Experimental transmission of the probe field as a function of the detuning Δ_{2ph} . The line (1) is obtained without depumping beams, while the lines (2) and (3) correspond respectively to the use of one depumping beam ($\Delta_D = 40\text{MHz}$) and two depumping beams ($\Delta_D = 40\text{MHz}$ and 85MHz). The transmission is normalized to the transmission for large detunings. The two depumpers have equal power (6 mW), while the repumper power is adjusted for each measurement to keep the same optical depth (4.5 mW without depump, 5.5 mW for one depumper and 7.5 mW for two). (b) Theoretical predictions calculated from Ref. [21]. The control Rabi frequency is 2.3γ , corresponding to the experimental value, and the excited state natural linewidth is $\gamma = 2\pi \times 5.2\text{ MHz}$. We include the ground state decoherence due to the dephasing between control and probe lasers experimentally estimated to be $\gamma_{sg} = 0.077\gamma$. To calculate the transmission, the susceptibility is convoluted with the velocity distribution extracted from the experimental data.

depumping beams is correctly reproduced.

Let us note also that in the absence of depumping beams, the experimental EIT peak shown as curve (1) in Fig. 3(a) and (b), does not completely disappear, while it is predicted to go to zero in Fig.1(b). This feature can be explained by the combined effect of the control and the repump that more efficiently brings back atoms with zero Doppler shift from the $|F = 4\rangle$ ground state.

Finally, in order to quantify the EIT enhancement, we can introduce an EIT contrast C defined as the ratio $C = (t_{max} - t_{min}) / (1 - t_{min})$ with t_{max} and t_{min} being respectively the probe transmittance at the maximum and on the side of the EIT peak. In our experiment, C is increased by a factor 5, thus confirming the efficiency of the procedure.

In summary, we have reported a detailed experimental characterization of the EIT properties of a medium with Doppler broadening and multiple excited levels. It can be shown that some specific velocity classes are mainly caus-

ing the suppression of transparency. We have proposed a procedure to remove these atoms from the interaction process via a well-designed reshaping of the atomic velocity distribution. Our observations confirmed the general mechanism and enabled to demonstrate a strong enhancement of the transparency. This study may bring new applications and offer significant improvements in various settings based on EIT or related effects, including in quantum information science and metrology. Moreover, beyond the specific medium used here, i.e alkali-metal atoms at room temperature, our method, which allows an efficient engineering of the EIT properties of an inhomogeneously broadened medium, can be extended to various atom-like physical systems presenting simultaneously large broadening and multiple levels, e.g. in rare-earth doped crystals, quantum dots or nitrogen-vacancy centers in diamonds.

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