Enhancing electromagnetically-induced transparency in a multilevel broadened medium

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Electromagnetically-induced transparency has become an important tool to control the optical properties of dense media. However, in a broad class of systems, the interplay between inhomogeneous broadening and the existence of several excited levels may lead to a vanishing transparency. Here, by identifying the underlying physical mechanisms resulting in this effect, we show that transparency can be strongly enhanced. We thereby demonstrate a 5-fold enhancement in a room-temperature vapor of alkali-metal atoms via a specific shaping of the atomic velocity distribution.

I. INTRODUCTION

Among the capabilities enabled by electromagnetically induced transparency (EIT) [1, 2] or related phenomena are high precision magnetometery, lasing without inversion, and slowing [3] and stopping of light pulses [4, 5]. These possibilities opened new avenues for optical information storage and quantum information processing. Recent experiments based on dynamic EIT have demonstrated the reversible mapping of single-photons or qubits [6–8] and of quantum continuous variables [9–11]. 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, most relevant for quantum memory schemes [2]: 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 [12, 13]. 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 [11, 14] and can eventually disappear for large broadening [15]. The broadening also leads to a narrowing of the transparency window [16–18]. 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 the present paper, we report measurements that provide a detailed picture of EIT in a Doppler broadened 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.

II. EFFECT OF INHOMOGENEOUS BROADENING ON THE EIT FEATURES

In a Λ-type 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 exhibits two symmetrical absorption peaks as a function of its detuning from the resonance, defining a transparency window at resonance. In an inhomogeneously broadened medium, the absorption spectrum of the various atoms differs from this description as they are involved in off-resonant processes, drastically modifying the susceptibility and the EIT spectrum.

Specifically, in a Doppler broadened medium, the atoms are 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 $2\pi.\Delta_{Doppler} = -\mathbf{k}\cdot\mathbf{v}$ where $\mathbf{k}$ is the wave-vector of the fields. The two-photon detuning, defined as $2\pi.\Delta_{2ph} = \omega_{probe} - \omega_{control} + \omega_{gs}$ where ω_{probe} and $\omega_{control}$ are the field frequencies and ω_{gs} the splitting between the two ground states, does not depend on the velocity either. 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. We have considered the case of velocities opposite to the laser propagation direction ($\Delta_{Doppler} > 0$). 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 symmet-

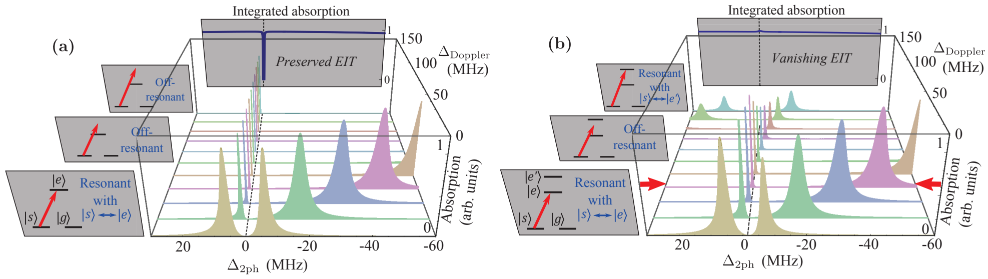

FIG. 1: EIT features in a Λ and in a multi-level system. The calculated probe absorptions are displayed for atoms with different velocities given by the Doppler detunings $\Delta_{Doppler}$, as a function of the detuning Δ_{2ph}. Panel (a) corresponds to a Λ-scheme and shows a preserved transparency. Panel (b) takes into account an additional excited state, leading to a vanishing transparency. The dotted line indicates the center of the EIT window for atoms with $\Delta_{Doppler}$=0. The integrated absorption is obtained for a Gaussian velocity distribution of 160 MHz half-width (thermal distribution for Cesium at 300 K). The control Rabi frequency is $\Omega = 2.3\gamma$, where γ is the natural linewidth.

rical 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 $\Delta_{2ph} = 0$ without reaching this point. As a result, transparency is preserved at the zero two-photon detuning for all the velocity classes. The integration over the whole distribution thus preserves the transparency at resonance but leads to a reduction of the transparency window width.

When one takes into account the presence of other levels, additional Stark shifts appear that move the Raman resonance from the position described in the Λ configuration. Based on the model developed in [21], Fig. 1(b) shows the case of two excited levels, $|e\rangle$ and $|e'\rangle$. The strong modification of the susceptibility can be understood as follows. In this 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 $\omega_{e'e}$. These two classes exhibit a quasi-symmetrical Autler-Townes doublet centered close to 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 transparency window of atoms with zero Doppler shift, 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. We find that in our case the atomic velocity classes to be removed for optimal EIT recovery correspond to Doppler detunings 35MHz$\lesssim \Delta_0 \lesssim$45MHz (Appendix A). By excluding these specific atoms from the interaction process, the EIT can be recovered, as we will show.

III. EXPERIMENTAL SETUP AND RESULTS

The experiment is sketched in Fig. 2. The optically dense medium is obtained from a 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 shield. The scheme of the 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 $|6S_{1/2}, F = 3, m = 3\rangle$ separated by 1.25 MHz. The excited state $|e\rangle$ is the Zeeman state $|6P_{3/2}, F = 2, m = 2\rangle$. 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 $|s\rangle \rightarrow |e\rangle$ for non-moving atoms, while the probe field is σ^--polarized and addresses the transition $|g\rangle \rightarrow |e\rangle$.

The experiment is performed in the continuous-wave regime. 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 is maintained in $|6S_{1/2}, F = 3\rangle$ [11].

Control and probe fields are collimated (5mm diameter) and with respective powers of 200 mW and 150 nW. They are combined on a polarizing beam splitter and then pass through a $\lambda/4$ to enter the cell with circular polarization. The probe field is then extracted using a $\lambda/4$ 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.

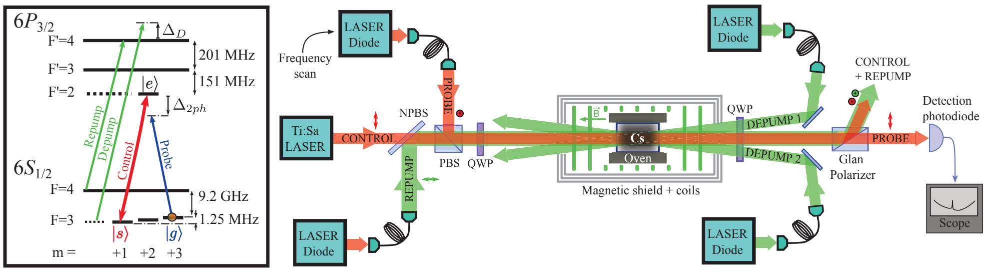

FIG. 2: The transmission of a cesium vapor is probed by scanning the frequency of a weak σ^--polarized probe field. The strong σ^+-polarized control field is kept on resonance with the $|s\rangle$ to $|e\rangle$ transition. A repumper beam enables to efficiently prepare the atoms in the $|g\rangle$ ground 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. PBS and NPBS : polarizing and non-polarizing beam splitter, QWP: $\lambda/4$ plate.

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 detuned by Δ_D from the $|F=3\rangle \rightarrow |F'=4\rangle$ transition for non-moving atoms. They enable to burn specific holes in the velocity distribution. For experimental convenience, they are contra-propagative with the control and probe fields, with an 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 $\Delta_D = \Delta_0$. The atoms are then efficiently pumped into the hyperfine ground state F=4 and do not contribute anymore to the interaction.

Transmission spectra are obtained by scanning the probe detuning, which corresponds to scanning the two-photon detuning Δ_{2ph}. Figure 3(a) gives the 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. We then send a depumping beam. A broad peak, corresponding to the hole burnt in the velocity distribution, appears in the transmission spectrum (curve (2)). As can be seen, the EIT peak is significantly enhanced. The best transparency is obtained for detunings close to the predicted value of $\Delta_D = 40$ MHz, confirming that the EIT is recovered when the detrimental effect of these atoms is suppressed. The depth of the hole created in the distribution by the depumping beam at 40 MHz being saturated for a power of about 6mW, we tested the effect of a second depumping beam, detuned from the optimal frequency and corresponding to atoms with Δ_D =85 MHz. As shown by curve (3), the transparency is slightly increased, since this beam removes some more atoms which have non-zero absorption in the EIT window.

IV. DISCUSSION AND CONCLUSION

These results are in very good agreement with the transmission curves calculated from the model developed in Ref. [21] and given 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, as shown in the inset of Fig. 3(b). Even in the absence of depumping beams, the distribution is non Gaussian, since it is strongly modified by the control and the repump beams [22–24]. It can be seen that the effect of one and two depumping beams on the EIT is correctly reproduced. Because of the distorted velocity distribution, this model also correctly predicts a small EIT feature in the absence of depumping, in contrast to the model of Fig. 1(b).

Finally, in order to quantify the EIT enhancement, we introduce an EIT contrast C defined as the ratio $C = (t_{max} - t_{min})/(1 - t_{min})$ with t_{max} and t_{min} being the probe transmittance at the maximum and on the side of the EIT peak. In our experiment, C is increased by a factor 5, which can be shown to yield a decrease of the group velocity by a factor of the same order of magnitude. This shows the potential of our method for efficiently improving EIT-based processes. We note that the full theoretical model [21] predicts an enhancement factor around 8. The main limitation in our experimental demonstration is coming from the contrast of the hole burned in the velocity distribution. A detailed study of the hole burning dynamics is expected to bring further enhancement of the EIT feature [22, 23] .

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 causing the suppression of transparency. We have proposed a procedure to remove these atoms from the interaction 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

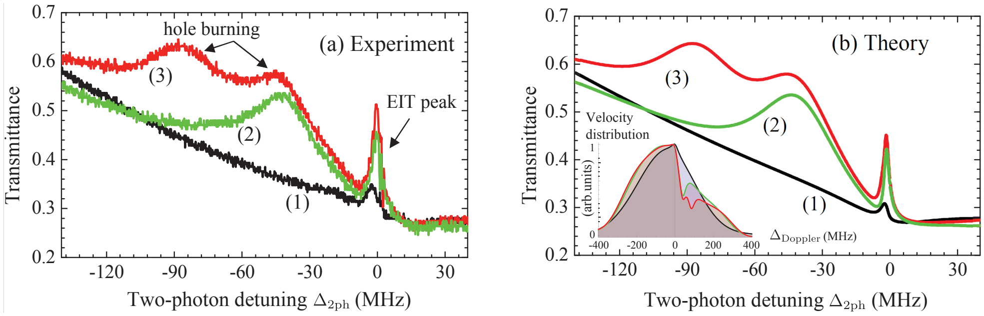

FIG. 3: Transparency enhancement by reshaping of the velocity distribution. (a) Transmission of the probe field as a function of the detuning Δ_{2ph}. Curve (1): without depumping beams, Curve (2): one depumping beam with Δ_D=40 MHz and Curve (3): two depumping beams with Δ_D=40 MHz and 85 MHz. The transmission is normalized to the transmission for large detunings. The experimental powers are 6 mW for the depumpers, 4.5 mW for the repumper without depump, 5.5 mW with one depumps and 7.5 mW with two depumps (see text). (b) Theoretical predictions calculated from Ref. [21]. $\Omega = 2.3\gamma$, corresponding to the experimental value, with $\gamma = 2\pi \times 5.2$ MHz. We include the ground state decoherence due to the dephasing between control and probe lasers experimentally estimated to be $\gamma_{sg} = 0.077\gamma$. This value corresponds to twice the linewidth of these independent lasers. The inset gives the velocity distribution extracted from the data and used for the model.

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.

Appendix A: Determination of velocity classes leading to absorption

The simplified model given in the text enables to identify the atoms which strongly modify the EIT and to calculate their Doppler shift Δ_0. With d and d' the dipole elements for $|s\rangle \rightarrow |e\rangle$ and $|s\rangle \rightarrow |e'\rangle$ transitions and $\omega_{e'e}$ the splitting in the excited state, Δ_0 can be obtained from:

$$\frac{|d|^2}{\Delta_0} + \frac{|d'|^2}{(\Delta_0 - \omega_{e'e})} = -\frac{|d'|^2}{\omega_{e'e}}.$$

This equation states that the Raman absorption resonance, displaced by the Stark shifts due to the two excited levels (left hand side) is centered at the same frequency as the EIT window for atoms with zero Doppler shift, itself shifted due to level $|e'\rangle$ (right hand side). This equation has two solutions that do not depend on the control field Rabi frequency. The solution that introduces the largest absorption is Δ_0=48MHz, marked by arrows in Fig. A1.

With a full model [21] taking into account the hyperfine structure of atomic cesium, and the variation of the height of the Raman absorption resonance with Doppler detuning, we find that in our case the atomic velocity classes to be removed for optimal EIT recovery correspond to Doppler detunings 35MHz$\lesssim \Delta_0 \lesssim$45MHz, which is consistent with the simplified

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