Recovering the homogeneous absorption of inhomogeneous media

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The resonant absorption of light by an ensemble of absorbers decreases when the resonance is inhomogeneously broadened. Recovering the lost absorption cross-section is of great importance for various applications of light-matter interactions, particularly in quantum optics, but no recovery mechanism has yet been identified and successfully demonstrated. Here, we formulate the limit set by the inhomogeneity on the absorption, and present a mechanism able to circumvent this limit and fully recover the homogeneous absorption of the ensemble. We experimentally study this mechanism using two different level schemes in atomic vapors and demonstrate up to 5-fold enhancement of the absorption above the inhomogeneous limit. Our scheme relies on light shifts induced by auxiliary fields and is thus applicable to various physical systems and inhomogeneity mechanisms.

Inhomogeneous broadening of spectral lines is a prevalent limiting factor in experiments and applications involving light-matter interactions in ensembles. This common impediment occurs for various atomic and atom-like absorbers, including quantum dots, diamond color centers, rare-earth ions in crystals, hot atoms, and particularly with Rydberg excitations. The broadening originates from a distribution of resonant frequencies of the individual absorbers. Common sources of inhomogeneity are nonuniform magnetic fields or local crystal strains, or thermal distributions of atomic velocities.

The inhomogeneous broadening dominates when the width of the resonance-frequency distribution $2\sigma$ is larger than the homogeneous linewidth $2\gamma$. This is accompanied by a decrease in absorption, which scales as $\gamma/\sigma$. Standard techniques for circumventing inhomogeneous broadening rely on selective methods, such as hole burning and Doppler-free configurations, which resonantly address only a small fraction of the ensemble. Another class of techniques is dynamical decoupling, such as spin echo, which suppresses decoherence while engaging the entire ensemble. Additionally, state population transfer can be made efficient for a large fraction of the ensemble using pulsed light-shifts. However, these techniques do not recover the bare absorption lost due to the inhomogeneity.

It is known that far off-resonance operation is mostly insensitive to inhomogeneous broadening, and so are processes relying on slow light, such as light storage. Scattering effects, however, such as absorption or fluorescence, are directly affected by inhomogeneity. Indeed, the effective absorption cross-section and the optical depth decrease in typical systems by 1-3 orders of magnitude. This is particularly important for extreme nonlinear optics, such as the effective photon-photon interaction mediated by Rydberg atoms, which require high effective optical-depth within an interaction range of only a few micrometers. As the atomic density is practically limited, the effective cross-section controls the strength of the interaction.

In atomic gases, inhomogeneous broadening results from the Doppler effect for different atomic velocities. It was originally suggested by Cohen-Tannoudji et al. that velocity-dependent light-shifts due to an additional field can be utilized to counteract the broadening due to the Doppler effect. This idea has been analyzed and realized in several configurations. Inhomogeneous light-shifts were also used for generating narrow features with multi-photon excitations of hot Rydberg atoms, and for increasing the coherence time of laser-trapped atoms. Several proposals focused on counteracting the decrease in amplitude due to the inhomogeneity. However, and despite its importance, no significant enhancement of the effective absorption cross-section has been experimentally realized so far.

In this work, we first define the ‘inhomogeneous limit’ of the effective cross-section, for a probe field coupled to inhomogeneously-broadened transitions in either two-level or three-level systems. We find that the same limit applies to both cases, even when a coupling field in the three-level case generates a narrow absorption feature for the probe. The latter occurs, for example, in Raman transitions between two sub-states of a ground level. We then show that this limit can be surpassed in a four-level system, by utilizing light-shifts due to an additional ‘recovery’ field. Consequently, the probe field can resonantly interact with the majority of the ensemble, and in the ideal case the absorption is fully recovered. We use a hot atomic medium to study this mechanism and demonstrate a significant increase in the effective cross-section, $\times 5$ higher than the inhomogeneous limit. This is the first demonstration of any absorption effect significantly exceeding the inhomogeneous limit. Finally, to establish the generality of the recovery mechanism, we implement it in a ladder-type level system involving Rydberg states and demonstrate a similar increase of absorption beyond the inhomogeneous limit.

The inhomogeneous limit.— Consider a two-level system with an optical transition $|g\rangle \leftrightarrow |e\rangle$ of homogeneous (half) linewidth $\gamma$. To introduce inhomogeneity, assume that the resonance frequency of this transition is shifted by a parameter $\delta$ which has a Gaussian distribution with a standard deviation $\sigma \gg \gamma$. Consequently, the absorp-
tion line broadens, and its amplitude reduced by the factor \( \beta_0 = \sqrt{2/\pi} \sigma/\gamma \). A typical example is the \( D_1 \) transition in hot rubidium atoms, where \( \gamma = 2.875 \, \text{MHz} \). The Doppler width at 50°C is \( \sigma = kv_T = 220 \, \text{MHz} \), where \( k \) is the probe wavevector and \( v_T \) the RMS thermal velocity, and therefore the absorption is reduced by a factor of \( \beta_0 \approx 60 \) with respect to that of stationary atoms. In the solid state, an important example are silicon-vacancy (SiV−) centers in diamond at a few K \([32, 33]\). These centers have a homogeneous width of \( \gamma_{\text{SiV}} \approx 50 \, \text{MHz} \) and an inhomogeneous width of \( \sigma_{\text{SiV}} \approx 5 \, \text{GHz} \) due to varying local strains in the surrounding lattice, which reduces the absorption by a factor of \( \beta_0 \approx 80 \).

Now consider a three-level system \(|g⟩\leftrightarrow|e⟩\rightarrow|s⟩\), as depicted in Fig.1(a), and assume that the frequency difference between \(|g⟩\) and \(|s⟩\) is unaffected by the inhomogeneity. This is common for Raman transitions in \( \Lambda \)-systems where \(|g⟩\) and \(|s⟩\) are within the ground-level manifold, such that the resonance frequencies of the two optical transitions \(|g⟩\leftrightarrow|e⟩\) and \(|e⟩\leftrightarrow|s⟩\) share the same broad distribution of shifts \( \delta \). When a coupling field drives the \(|e⟩\leftrightarrow|s⟩\) transition and is detuned from resonance by \( \Delta \gg \sigma \), it can induce the absorption of the probe on the two-photon transition \(|g⟩\leftrightarrow|s⟩\). The homogeneous width of the two-photon transition is the sum of its bare width \( \gamma_{sg} \) and the scattering rate \((\Omega/\Delta)^2\gamma\), where \( \Omega \) is the Rabi frequency of the coupling field. Notably, both contributions can be substantially smaller than \( \sigma \), and thus two-photon transitions are often thought of as circumventing inhomogeneity.

Nevertheless, the absorption of the probe at these two-photon transitions is never higher than that of the broadened one-photon transition. For weak coupling fields, this limit is explained by the inefficient coupling \((\Omega/\Delta)^2 \ll \gamma_{sg} \) to \(|e⟩\). For the less obvious case of strong coupling \((\Omega/\Delta)^2 \gg \gamma_{sg} \), it results from inhomogeneous light-shifts: The strong coupling field shifts the state \(|s⟩\) by \( \sim \Omega^2/\Delta \), thereby shifting the two-photon resonance. In an inhomogeneous medium, the light-shift depends on \( \delta \) and is given by \( \Omega^2/(\Delta - \delta) \approx \Omega^2/\Delta + (\Omega/\Delta)^2\delta \). It follows that the dressed two-photon transition is inhomogeneously shifted, as shown by the shifted Raman-absorption lines in Fig.1(b). Replacing \( \delta \) by its standard deviation \( \sigma \), we find an induced inhomogeneous broadening \( \sim (\Omega/\Delta)^2 \sigma \). The ratio between this broadening and the homogeneous width \( \sim (\Omega/\Delta)^2 \gamma \) is \( \sigma/\gamma \), which is the same ratio as in the two-level case. The amplitude of the two-photon line decreases by the same ratio, and it is therefore bounded by the same inhomogeneous limit. Full density-matrix calculations of an inhomogeneous ensemble confirm that the reduction of absorption is at least \( \beta_0 \) over the whole spectrum for all choices of \( \Omega \) and \( \Delta \).

**Inhomogeneous compensation.**— Our approach for recovering the absorption cross-section is to counteract the induced inhomogeneous broadening by adding another excited state \(|r⟩\) and a recovery light-field that drives the optical \(|g⟩\leftrightarrow|r⟩\) transition [Fig.1(c)]. The additional transition is inhomogeneously broadened as well, bearing frequency shifts \( \delta_r \) that are correlated with \( \delta \) and are distributed with standard deviation \( \sigma_r \). The far-detuned recovery field, with detuning \( \Delta_r \) and Rabi frequency \( \Omega_r \), shifts the state \(|g⟩\) by \( \Omega_r^2/(\Delta_r - \delta_r) \). If the light-shifts of \(|s⟩\) and \(|g⟩\) are tailored to satisfy the compensation condition

\[
\frac{\Omega^2}{\Delta - \delta} = \frac{\Omega_r^2}{\Delta_r - \delta_r},
\]

then the resonance frequency of the two-photon transition will coincide for the entire ensemble, overcoming the inhomogeneity [Fig.1(d)].

The homogeneous linewidth of the two-photon resonance determines the sensitivity of the recovered absorption to deviations from Eq. (1). This linewidth, averaged over the ensemble, is given by \( \Gamma = \Gamma_r + \gamma_{sg} \), where \( \gamma_{sg} \) is the scattering rate due to the coupling and recovery fields are approximately \( \Gamma = (\Omega^2/(\Delta^2 + \sigma^2))\gamma_r \) and \( \Gamma_r = (\Omega_r^2/(\Delta_r^2 + \sigma_r^2))\gamma_r \).

Note that, as long as \( \gamma \ll \sigma \), the expressions comprising Eq. (1) hold for a large fraction of the ensemble, even
when the fields are tuned to within the inhomogeneously-broadened line $|\Delta| \lesssim \sigma$. This near-resonant regime features a wide transmission window surrounding the enhanced peak, whose width of $2\Omega + 2\Omega_r$ is explained by Autler-Townes splittings.

In hot atoms with co-propagating fields, $\delta = kv$ and $\delta_r = k_r v$ are both linear in the atomic velocity $v$ and therefore correlated; here $k$ and $k_r$ are the wavenumbers of the coupling and recovery fields. The compensation condition is met for $\Omega_r = \Omega \sqrt{k_r/k}$ and $\Delta_r = \Delta (k_r/k)$. Other physical systems often exhibit similar correlations. For example, in color-centers such as SIV$_r^-$, both $\delta$ and $\delta_r$ depend on the same local crystal strain $\Delta$.

**Enhancement resource.**— The compensation condition guarantees that the resonance frequencies of the two-photon absorption-lines of all the absorbers are aligned. The height of these lines determines the height of the enhanced absorption peak. We define the enhancement of absorption $\beta$ as the ratio between absorption on the two-photon and one-photon resonances. The absorption cross-section can be recovered only up to its homogeneous value, hence the maximal enhancement is $\beta_0$. Generally, each four-level absorber contributes at its two-photon resonance only a fraction of the homogeneous cross-section. This fraction is determined by the competition between the coupling-field scattering rate $\Gamma$ and the total linewidth of the two-photon transition $\Gamma + \Gamma_r + \gamma_{sg}$. It follows that the enhancement of absorption is given by $\beta = \beta_0 \Gamma / (\Gamma + \Gamma_r + \gamma_{sg})$. This intuitive formula agrees extremely well with full density-matrix calculations.

It is instructive to consider the simple case where both excited states ($|e\rangle,|r\rangle$) share the same inhomogeneous shifts $\delta = \delta_r$, and consequently $\sigma = \sigma_r$, $\Omega = \Omega_r$, and $\Delta = \Delta_r$. The resulting enhancement is

$$\beta = \frac{\beta_0 \gamma}{\gamma + \gamma_r} \frac{\mu^2}{1 + \mu^2}. \quad (2)$$

Here $\mu^2 = \Omega^2(\gamma + \gamma_r)/[(\Delta^2 + \sigma^2)\gamma_{sg}]$ is a saturation parameter that scales linearly with the coupling and recovery fields intensities, and $\beta \to \beta_0 \cdot \gamma / (\gamma + \gamma_r)$ at saturation. Therefore, full recovery of the homogeneous absorption cross-section requires both a narrow transition for the recovery field $\gamma_r \ll \gamma$ and a high intensity of the coupling and recovery fields $\mu^2 \gg 1$.

It is important to distinguish between the mechanism we study and those developed for enhancing the homogeneous absorption in degenerate multilevel systems [44, 38]. These mechanisms circumvent the multilevel structure of the transition and thus can only increase the absorption up to that originating from the strongest transition in the manifold [40]. In contrast, our recovery mechanism applies even when the strongest transition is directly used from the onset, as in the experiments we report here.

**Recovery of absorption for atomic vapor.**— We experimentally study the inhomogeneous compensation mechanism using $^{87}$Rb vapor at $33-42$ °C. First, we employ an $N$-type system [38, 39, 11], as depicted in Fig. 2, in a 75-mm-long cell of natural abundance Rb. The probe beam waist is 375 $\mu$m, while the coupling and recovery beams are $\sim 4$ times larger. We use a ring-shaped beam for Zeeman optical pumping. The probe propagates inside the dark center of the pump beam, thus avoiding undesired light shifts from the pump. Finite transit-time of atoms crossing the probe beam and noise in the phase-lock between the lasers govern the bare width of the two-photon transition $\gamma_{sg} = 0.35$ MHz. The excited states decay rates are $\gamma = 2.875$ MHz and $\gamma_r = 3.033$ MHz.

Figure 2 shows absorption spectra for a bare three-level system (blue) and with the additional recovery field (green). Once the recovery field is turned on, the two-photon absorption line becomes narrow and high, considerably surpassing the inhomogeneous limit.

In order to characterize the mechanism, we measure multiple spectra at a fixed coupling-field detuning of $\Delta = -270$ MHz, and scan the intensities of the coupling and recovery fields, as well as the detuning $\Delta_r$ of the recovery field. We extract the enhancement $\beta$ for each spectrum from the ratio between the peak absorption at the two-photon and one-photon resonances. Figure 3(a) shows the dependence of $\beta$ on $\Omega_r$ for several values of $\Omega$, where for each $\Omega$ we choose $\Delta_r$ that maximizes the enhancement. We find, as expected from condition (1), that the maximal enhancement is obtained when $\Omega_r \approx \Omega$. 

Fig. 2. Experimental demonstration of absorption enhancement with inhomogeneous compensation. The absorption spectra of the probe field exhibit both the one-photon resonance $|g\rangle \to |e\rangle$ near zero detuning, and the two-photon (Raman) resonance $|g\rangle \to |s\rangle$ induced by the coupling field around $\Delta = -270$ MHz. Without the recovery field (blue curve), the absorption on both resonances is bounded by the inhomogeneous limit (dashed red). With the recovery field (green curve), the absorption peak grows substantially, exceeding the inhomogeneous limit by a factor of $\beta = 4.8 \pm 0.4$. In this measurement, $\Omega = 29$ MHz, $\Omega_r = 29.6$ MHz, and $\Delta_r = -300$ MHz.
In Fig. 3(b), we show the dependence of the maximal enhancement on $\Delta_r$. The enhancement is maximal at $\Delta_r \approx \Delta$, and is sensitive to deviations of order $\pm 10$ MHz around this condition. This sensitivity agrees well with the requirement that Eq. 7 is satisfied up to the homogeneous linewidth of the two-photon resonance.

Figure 4 presents the dependence of $\beta$ on the intensity of the coupling and recovery fields. Ideally, according to Eq. 2 with $\mu^2 \gg 1$ and $\gamma \approx \gamma_r$, the enhancement should saturate at $\beta \to \delta_0/2 \approx 30$, as indeed obtained in a calculation of the simple four-level system (green curve). In our experimental system, the main deviations from ideal conditions are an additional hyperfine level in the excited manifold and the nonuniform intensity of our beams due to their finite sizes. A full calculation accounting for these with no fit parameters quantitatively reproduces the measured trends and the maximal enhancement [dashed blue line in Figs. 3(b) and 4].

To confirm the generality of the recovery mechanism, we also apply it to a four-level ladder scheme of $^{87}$Rb in an isotopically-pure 5-mm-long cell (Fig. 5). The probe beam, with a waist of 85 $\mu$m, counterpropagates the coupling and recovery beams, with waists of 750 $\mu$m and 220 $\mu$m, respectively. Here, the decay rates $\gamma_{sk} \approx 1.25$ MHz and $\gamma_r \approx 1$ MHz include the radiative lifetimes of the 5D and 3F levels, respectively. In this ladder system, the two-photon transition $|g\rangle \leftrightarrow |s\rangle$ exhibits a small residual Doppler broadening $\sigma = 1$ MHz, satisfying $\sigma < \sigma_s$. As shown in Fig. 5, the recovery mechanism applies equally well, yielding a substantial enhancement of the absorption $\beta = 4.6 \pm 0.3$ above the inhomogeneous limit.

Conclusions.— We presented a limit to the absorption enhancement in an inhomogeneous medium and showed how light shifts arising from strong driving fields can compensate for the inhomogeneity, circumvent this limit, and recover a substantial part of the absorption cross-section. We experimentally studied the recovery mechanism in atomic vapor and demonstrated an enhancement of the absorption cross-section up to $\beta = 4.8 \pm 0.4$ times higher than the inhomogeneous limit. The attainable recovery is limited by the available intensities of the driving fields (via the saturation parameter $\mu$) and by the ratio $\gamma/\gamma_r$ between the homogeneous widths of the employed transitions. Ideal conditions $\mu, \gamma/\gamma_r \gg 1$ allow for full recovery of the homogeneous absorption.

The mechanism is general and can be applied to various inhomogeneous systems in the gas and solid phases. Our experiments show that both spin and orbital transitions can be utilized (in either $N$-type or ladder-type configurations), with the recovery field coupled to either populated or unpopulated states, and that the mechanism prevails even for small, nonzero, two-photon inhomogeneous broadening $\sigma \ll \sigma_s$. The more extreme case, when $\sigma_2$ approaches $\sigma$, are beyond of the scope of this work [42]. The key requirement is that the inhomogeneous shifts $\delta$ and $\delta_t$ are correlated, such that condition 7 can be satisfied.

The enhancement of the effective absorption cross-section is essential for single-photon nonlinearities via the Rydberg blockade [43]. In this context, the ladder system employed here attracted much recent attention [10, 44, 45]. Here most crucial is the optical depth of the $\sim 10 \mu$m$^3$ blockade volume, termed $OD_B$. In cold atoms,
ODB > 1 is routinely achieved [22, 45–47], whereas in hot atoms, the inhomogeneous broadening severely reduces ODB, making it extremely challenging to realize the strong blockade regime. Our recovery mechanism can alleviate this deficiency, leading to better scalability and higher fidelity of future quantum technology in room-temperature media.

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Fig. 5. Inhomogeneous compensation for a ladder system with a Rydberg level. The absorption spectrum in the absence of the recovery field (blue) is bounded by the inhomogeneous limit (dashed red). The addition of the recovery field significantly enhances the absorption of the probe (green) above this limit. In this measurement, the driving lasers with $\Omega = 55$ MHz and $\Omega_r = 45$ MHz are tuned to resonance.

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