Diffusion-Induced Ramsey Narrowing

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Diffusion-induced Ramsey narrowing is characterized and identified as a general phenomenon, in which diffusion of coherence in and out of an interaction region such as a laser beam induces spectral narrowing of the associated resonance line shape. Illustrative experiments and an intuitive analytical model are presented for this spectral narrowing effect, which occurs commonly in optically interrogated atomic systems and may also be relevant to quantum dots and other solid-state spin systems.

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The lifetime of an atomic coherence is often limited by the finite interaction time between the atoms and resonant radiation: e.g., by atomic motion through a laser beam. For atoms constrained to diffuse in a buffer gas, this interaction time is usually estimated by the lowest-order diffusion mode, which leads to the typical Lorentzian line shape, but implicitly assumes that atoms diffuse out of the laser beam and do not return [1–3]. However, when other decoherence effects are small, atoms can diffuse out of the interaction region and return before decohering. That is, atoms can evolve coherently in the dark (outside of the laser beam) between periods of interaction (inside the laser beam), in analogy to Ramsey spectroscopy [4]. In many cases of interest, diffusing atoms can spend a majority of their coherence lifetime in the dark, which induces a significant spectral narrowing of the center of the atomic resonance line shape.

In the present Letter, we identify this ‘‘diffusion-induced Ramsey narrowing’’ as a general phenomenon relevant to a wide variety of physical systems including atomic frequency standards [5] and dynamic light-matter interactions such as slow and stored light in atomic vapor [6]. An analogous spectral narrowing may also occur because of spin diffusion in solid-state systems [7] such as quantum dots [8]. Here, we characterize diffusion-induced Ramsey narrowing through demonstration experiments using electromagnetically induced transparency (EIT) in warm Rb vapor, and with an intuitive analytical model of the repeated diffusive return of atomic coherence to the laser beam (see Fig. 1). Previously, such spectral narrowing had only been treated in a few special cases [9].

EIT results from optical pumping of atoms into a noninteracting ‘‘dark’’ state for two optical fields that are in two-photon Raman resonance with a pair of metastable ground states of the atomic system [10]. EIT gives rise to a narrow transmission resonance for the optical fields, with a minimum spectral width set by the rate of decoherence between the two ground states constituting the dark state. To characterize diffusion-induced Ramsey narrowing using EIT, we employed a diode laser operating at 795 nm on the ^{87}Rb $D1$ transition to drive the atoms into EIT resonance between the $F = 2$ and $F = 1$ hyperfine levels of the electronic ground state. The beam passed through an enriched ^{87}Rb vapor cell (2.5 cm diameter, 5 cm length, Ne buffer gas) which was heated to approximately 45 °C to create optically thin Rb vapor ($n \sim 10^{11}$ cm^{-3}). The cell was mounted within three layers of magnetic shields to screen external fields. Sets of coils were used as needed to provide a homogeneous longitudinal magnetic field (B_z) and/or a transverse gradient in the longitudinal field

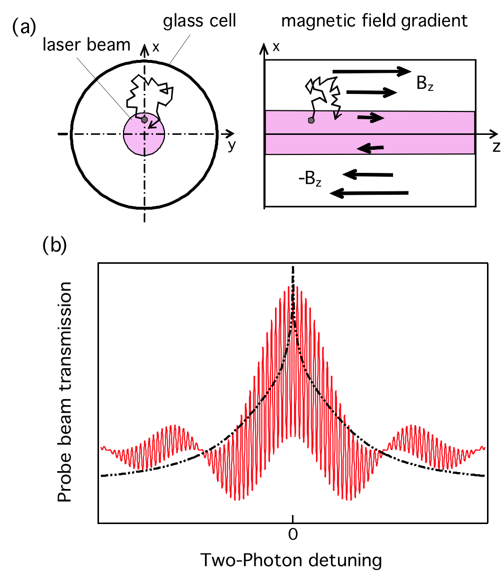

FIG. 1 (color online). (a) *Left*: example path for atoms diffusing in and out of the laser beam. *Right*: in some measurements, a transverse gradient in the longitudinal magnetic field was applied to decohere atoms that diffused far from the beam. (b) Calculated EIT line shapes for diffusive return of atomic coherence to the laser beam. Solid red curve: an example for a particular diffusion history (Ramsey sequence), with equal time spent in the laser beam $t_{\rm in} = \tau_D$ before and after diffusing in the dark for time $t_{\rm out} = 20\tau_D$, where τ_D is the mean time to leave the beam given by the lowest-order diffusion mode. Intrinsic and power-broadened decoherence rates $\Gamma_0 = 0.003\tau_D^{-1}$ and $\Gamma = 0.09\tau_D^{-1}$ are used in this calculation. Dashed black curve: weighted average over all Ramsey sequences.

($\partial B_z/\partial x$) as shown in Fig. 1(a). A photodetector measured the total light intensity transmitted through the vapor cell.

In a first set of experiments, we employed a vertical cavity surface emission laser with a transverse Gaussian intensity profile, which was current modulated at 3.4 GHz to form the two optical fields necessary for EIT [11]. We used a collimating lens and iris, and measured EIT line shapes for various laser beam diameters in a cell with 3 Torr Ne buffer gas (Rb diffusion coefficient $D \approx 50\ \mathrm{cm}^2/\mathrm{s}$). We set $B_z = 0$ and $\partial B_z/\partial x = 0$, such that EIT occurred for all relevant combinations of m_F sublevels. As shown in Fig. 2(a), the measured EIT resonance for a 1.5 mm diameter beam has a full-width half-maximum (FWHM) of 740 Hz, whereas the calculated FWHM $\approx$ 20 kHz if one makes the common assumption that the coherence lifetime is set by the lowest-order diffusion mode out of the beam (e.g., see [3]). As also shown in Fig. 2(a), the measured EIT line shape for the 1.5 mm diameter beam is spectrally narrower near resonance than a Lorentzian: this sharp central peak is the characteristic signature of diffusion-induced Ramsey narrowing. In contrast, the measured EIT resonance for a 10 mm diameter beam is well fit by a Lorentzian line shape with FWHM $\approx$ 400 Hz [see Fig. 2(b)], which is in good agreement with the calculated FWHM using the lowest-order diffusion mode, and is consistent with the small fraction of atoms that leave this relatively large diameter beam and return during the maximum coherence lifetime (set by buffer gas collisions and diffusion to the cell walls).

In a complementary set of experiments, we measured the EIT line shape as a function of buffer gas pressure, thereby altering the Rb diffusion coefficient and changing the fraction of atomic coherence that evolves in the dark. (We used a slightly different apparatus than that described above. See [12] for details.) In Fig. 3 we compare measured EIT line shapes for 5 and 100 Torr Ne buffer gas ($D \approx 30$ and $1.5\ \mathrm{cm}^2/\mathrm{s}$ respectively), with a 0.8 mm laser beam diameter. Fits to the data are shown both for our analytical "repeated interaction model" (outlined below) and a Lorentzian line shape. The repeated interaction model provides a good fit at both high and low buffer gas pressure, demonstrating its ability to account for the physics of diffusion-induced Ramsey narrowing. In contrast, the Lorentzian fit deviates significantly from the measurements even for 100 Torr Ne buffer gas—i.e., even under conditions of relatively slow Rb diffusion and reduced

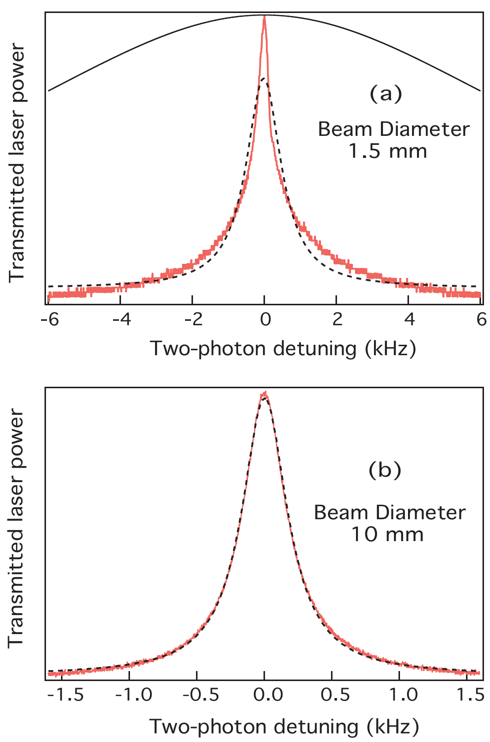

FIG. 2 (color online). Measured Rb EIT line shapes (in red) and fits using a Lorentzian line shape (dashed lines), with laser beam diameters of approximately (a) 1.5 mm and (b) 10 mm in a 3 Torr Ne cell with total incident laser intensity of 1 $\mu\mathrm{W/mm}^2$. EIT resonance contrasts (as defined in [5]) are (a) 1.2% and (b) 10%. Fitted Lorentzian parameters are the amplitude, off-resonant background, and full widths of approximately (a) 1400 and (b) 400 Hz. Broad solid curve in (a) is a 20 kHz FWHM Lorentzian, the expected line shape for a coherence lifetime set by the lowest-order diffusion mode out of the beam, with amplitude set equal to the peak measured amplitude for illustrative purposes.

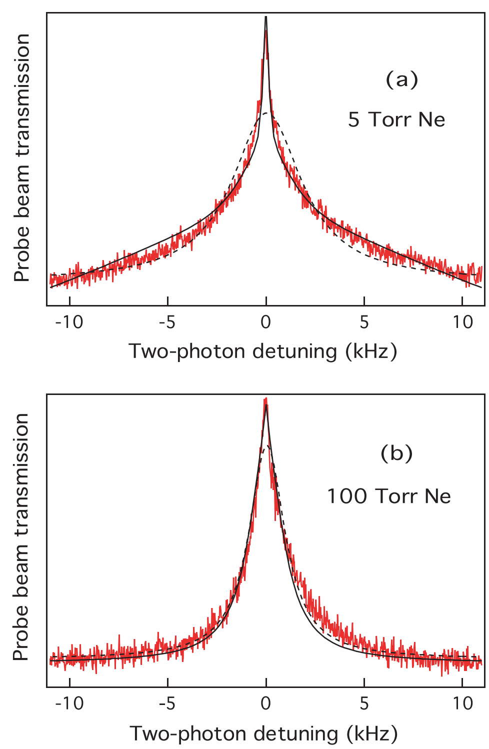

FIG. 3 (color online). Measured Rb EIT line shapes (in red) at (a) 5 and (b) 100 Torr Ne buffer gas pressure, with 22 μW laser power and approximately 0.8 mm beam diameter. Dashed lines are fits using a Lorentzian line shape. Fitted parameters are the amplitude, off-resonant background, and full widths of approximately (a) 4.3 and (b) 2.3 kHz. Black solid lines are fits using the repeated interaction model, as described in text.

coherence evolution in the dark—which indicates the inadequacy of the traditional approach of assuming that atoms diffuse out of the laser beam and do not return.

Destroying the coherence of atoms that leave the laser beam should eliminate diffusion-induced Ramsey narrowing. As a partial demonstration of this behavior, we measured magnetic-field-sensitive EIT spectra (coupling the $F = 2, m_F = 1$ and $F = 1, m_F = 1$ levels) in the presence of both a homogeneous longitudinal magnetic field $B_z \approx 43$ mG and a transverse gradient in the longitudinal magnetic field, $\partial B_z/\partial x$ [Fig. 1(a)], using a 0.8 mm diameter laser beam and 5 Torr Ne buffer gas [12]. An appropriately chosen magnetic field gradient ($\approx$2 mG/cm) induced modest inhomogeneous broadening of the EIT resonance ($\approx$ 400 Hz) for atoms within the laser beam [12], but caused atoms that diffused sufficiently far from the beam to lose phase coherence because of the extended period such atoms spend in the dark experiencing different magnetic fields. As shown in Fig. 4, the characteristic sharp peak in the EIT spectrum is effectively eliminated by application of the magnetic field gradient. The EIT line shape is otherwise not affected because atoms that remain in or near the beam are not significantly dephased by the applied gradient.

We found good agreement between our measurements and numerical calculations of the Maxwell-Bloch equations, which describe the atom-light interaction, coupled to the diffusion equation, which describes the atomic motion. We also developed and successfully applied a more intuitive and analytically soluble approach—the repeated interaction model mentioned above. In this model, the atomic resonance line shape is calculated for an atom having a specific history (“Ramsey sequence”) of alternating interactions with the laser beam and evolution in the dark. The equilibrium line shape for the atomic ensemble is then determined by a weighted average of the line shapes from different Ramsey sequences, using the distributions of times spent in and out of the laser beam (t_{in} and t_{out}) as determined from the diffusion equation; see Fig. 5. With this approach, the atomic motion and the atomic response to laser fields are decoupled, which dramatically simplifies the calculation and allows for an analytical solution.

For example, for a Ramsey sequence in which atoms spend time t_{in} in the laser beam and t_{out} in the dark, and assuming a large difference in intensity between the two EIT optical fields, the leading-order analytical expression for the weak field's transmission T as a function of two-photon Raman detuning Δ is given by:

$$\mathrm{T}(\Delta) = \mathrm{T}_0 + \frac{\kappa|\Omega_d|^2}{\Delta^2+\Gamma^2}(-\Gamma + \sqrt{\Delta^2+\Gamma^2}\{e^{-\Gamma t_{in}}\cos[\Delta t_{in} + \phi_\triangle] - e^{-\Gamma t_{in}-\Gamma_0 t_{out}}\cos[\Delta(t_{out}+t_{in})+\phi_\triangle] + e^{-2\Gamma t_{in}-\Gamma_0 t_{out}}\cos[\Delta(2t_{in}+t_{out})+\phi_\triangle]\}). \quad (1)$$

Here T_0 is the background transmission far from two-photon Raman resonance through the optically thin cell; $\kappa = \frac{3\pi}{16} n\lambda^2 L\gamma_r/\gamma^2$, where n is the atomic density, λ is the optical wavelength, L is the cell length, and γ_r and γ are the radiative decay rate and the total relaxation rate of the excited state, respectively; Ω_d is the Rabi frequency for the strong optical field; $\Gamma = \Gamma_0 + |\Omega_d|^2/2\gamma$ is the power-broadened EIT linewidth in the absence of atomic motion, where Γ_0 is the intrinsic relaxation rate of the ground-state coherence (set by buffer gas collisions, etc.); and $\tan\phi_\triangle = \Delta/\Gamma$. The above expression also assumes $\gamma \gg \Delta, \Gamma_0, \Gamma$ and $\gamma\Gamma \gg \Delta^2$, which are typically satisfied for EIT in warm Rb vapor. The first term in brackets in Eq. (1) is the contribution from atoms that interact with the laser beam only once. The second and third terms account for returning atoms. For a general Ramsey sequence, the time spent inside the laser beam before and after evolution in the dark may differ. Also, atoms may return to the beam more than once; each additional diffusive return will produce two extra terms similar to the last two lines in Eq. (1).

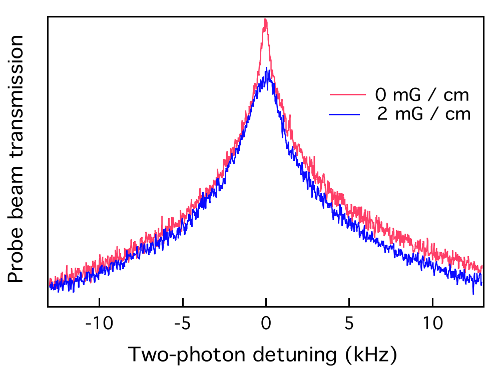

FIG. 4 (color online). Measured modification of the Rb EIT line shape by application of a transverse magnetic field gradient $\partial B_z/\partial x = 2$ mG/cm in a 5 Torr Ne buffer gas cell with approximately 0.8 mm laser beam diameter. The field gradient suppresses the sharp peak caused by diffusion-induced Ramsey narrowing, but leaves the broader EIT line shape largely unaffected.

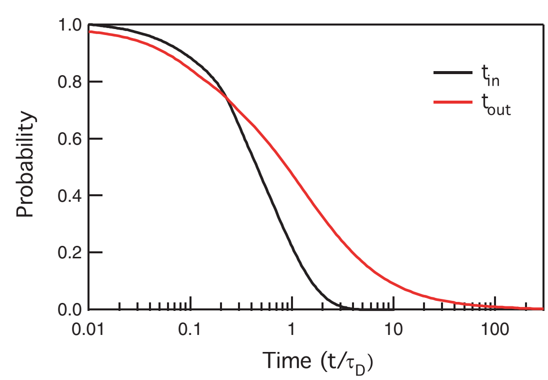

FIG. 5 (color online). Probability distributions, calculated from the diffusion equation, for an atom to spend time t_{in} inside a laser beam with a steplike transverse intensity profile, and then to spend time t_{out} outside of the laser beam before returning. Times are expressed relative to τ_D, the mean diffusion time to leave the beam given by the lowest-order diffusion mode.

Figure 1(b) shows the EIT line shape for an example Ramsey sequence described by Eq. (1), as well as the ensemble line shape determined from the weighted average over all Ramsey sequences.

We achieved good fits of the repeated interaction model to measured EIT line shapes, particularly the sharp central peak; e.g., see Fig. 3. Fitted parameters were the overall transmission amplitude and the off-resonant background transmission. For these fits we used a steplike laser profile in the transverse direction, which simplifies the calculations and is a reasonable approximation when the effective two-photon Rabi period is longer than the average time atoms spend in the laser beam, so that an atom averages over the transverse Gaussian distribution of laser intensity. We set the model laser power and steplike beam diameter near the nominal experimental values to optimize fits. A thorough discussion of the repeated interaction model and a detailed comparison with experiment (including the role of laser beam profile) will be presented in a future publication.

The width of the line shape envelope for an individual Ramsey sequence, such as that shown in Fig. 1(b), scales inversely with the time the atom spends in the laser beam ($t_{\rm in} \approx \tau_D \propto w^2/D$, where D is the Rb diffusion coefficient and w is the beam diameter); whereas the width of the Ramsey fringes is inversely proportional to the free-evolution time in the dark $t_{\rm out}$. The sharp central peak, indicative of diffusion-induced Ramsey narrowing, emerges intuitively in this model, since only the central fringe adds constructively for all Ramsey sequences with different diffusion times outside of the beam. The narrow width of this central peak is limited by other effects (atomic collisions, magnetic field gradients, wall collisions, etc.) which set an upper bound on $t_{\rm out}$. Since the atoms contributing most to the sharp central peak spend the majority of their time in the dark, the width of this peak is relatively insensitive to power broadening.

In general, when the laser beam diameter is small, reshaping and narrowing of the line shape are strong, since a large fraction of the atoms participate in the diffusion-induced Ramsey process, and the free-evolution time between interactions with the laser beam can be long. For larger laser beam diameters, the Ramsey narrowing gradually disappears since a smaller fraction of the atoms can diffuse out of the beam and return before decohering due to other effects. In particular, when the laser beam diameter approaches the cell diameter, atoms diffusing out of the beam rapidly decohere due to wall collisions. In ongoing work we are investigating the effect of coherence-preserving wall coatings, as well as the application of diffusion-induced Ramsey narrowing to the optimization of atomic clocks and stored light.

We note that non-Lorentzian EIT line shapes can appear in other circumstances. For instance, a well-known form of linewidth narrowing occurs in optically thick media due to frequency-selective absorption [13]. Alternatively, for an optically thin medium in the limit of high buffer gas pressure, atoms can reach equilibrium locally. An inhomogeneous laser intensity distribution then produces a spatial variation of the power broadening and an inhomogeneously broadened (and non-Lorentzian) line shape [14]. In the limit of low buffer gas pressure, atoms can be pumped at one intensity and probed at another, leading to a non-Lorentzian line shape dependent on the effusive time of flight of atoms across the sample cell [15]. These effects are qualitatively and quantitatively distinct from diffusion-induced Ramsey narrowing.

In conclusion, we identified a novel form of spectral narrowing arising from the diffusion of coherence in and out of an interaction region, such as a laser beam. We characterized this diffusion-induced Ramsey narrowing with measurements on EIT in warm Rb vapor, and found good agreement with an intuitive analytical model based on a weighted average of distinct atomic histories in the light and the dark. This repeated interaction model and the spectral narrowing effects studied here are relevant to spectroscopy, quantum optics, and emerging solid-state applications [8] based on long-lived coherences.

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