Slow Diffusion of Light in a Cold Atomic Cloud

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We study the diffusive propagation of multiply scattered light in an optically thick cloud of cold rubidium atoms illuminated by a quasiresonant laser beam. In the vicinity of a sharp atomic resonance, the energy transport velocity of the scattered light is almost 5 orders of magnitude smaller than the vacuum speed of light, reducing strongly the diffusion constant. We verify the theoretical prediction of a frequency-independent transport time around the resonance. We also observe the effect of the residual velocity of the atoms at long times.

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When light propagates through a multiply scattering medium, two components can be distinguished in the light emerging from the sample: the "transmitted" component, i.e., the light emerging in the incident mode, and the "diffuse" component corresponding to photons redistributed into other modes by the scattering processes. The temporal propagations of these two components are very different. The group velocity, whose behavior is determined by the index of refraction, describes the transmitted (and absorbed) component. The possibility to manipulate the refractive index allowed the spectacular observation of slow light [1], or of apparent supraluminal propagation [2] due to a negative group velocity. On the other hand, the diffuse light does not propagate ballistically, but shows a diffusive behavior for optically thick media. In this situation, it is the energy transport velocity v_E that accounts for the propagation of energy by the scattered wave [3,4]. In the present paper, we use the versatility of cold atomic vapors to study the temporal propagation of the diffuse light in the interesting situation of strongly resonant scattering, a previously unreachable regime where v_E is 5 orders of magnitude smaller than the vacuum speed of light c. This is the first observation of such a dramatic reduction of the energy transport velocity, and thus of the diffusion constant, in multiply scattering media. It is also a first and important step toward the possible strong (Anderson) localization of light in an atomic gas, a regime where interference effects further reduce the diffusion constant until it vanishes at the localization border [5]. Besides the mean-free path, which characterizes the stationary properties of multiple scattering, the diffusion constant is a crucial parameter for its dynamical properties [6,7].

The propagation of quasiresonant light in atomic vapors has been investigated for a long time under the name of radiation trapping (RT), in a regime where the atomic motion (Doppler effect) or collisions induce frequency redistribution, which completely blurs the effects linked to the atomic resonance [8,9]. In most cases, the inhomogeneous broadening dominated over the homogeneous one, since vapors with very small velocity spread could not be produced before the advent of laser cooling [10]. Following the pioneering work [11], we present the first systematic study of RT in cold atoms. We measure the light diffusion coefficient and transport velocity and analyze the role of the number of atoms and of the light frequency. We present some experimental evidence for the independence of the transport time with frequency, a nontrivial prediction of the theory for point scatterers. Finally, we show that the residual motion of the atoms noticeably affects the propagation of light in our sample.

The experimental setup and procedure are similar to those employed to observe coherent backscattering [12]. The magneto-optical trap (MOT) producing the cold atomic sample is turned off for 2 ms every 20 ms to allow for RT probing. We obtain a cold cloud of quasi-Gaussian shape (rms radius $r_0 = 2$–3 mm) containing up to 7×10^9 atoms, with a maximal optical thickness along a diameter $b = \sqrt{2\pi} r_0/\ell \approx 40$ where ℓ is the minimum scattering mean-free path of the light at the center of the atomic cloud. The rms velocity of the atoms is 0.15 m/s. We use a probe light resonant with the $F = 3 \rightarrow F' = 4$ transition of the $D2$ line of Rb^{85} (wavelength $\lambda = 780$ nm, natural lifetime $\tau_{nat} = 1/\Gamma = 27$ ns). The optical thickness is determined by measuring the spectral width of the transmission curve [13]. Together with a measurement of the density profile of the cloud by fluorescence imaging, this gives access to the scattering mean-free path ℓ. We can adjust the effective number of atoms in the cloud by turning off the MOT repumping laser shortly before the trapping laser. The principle of the RT measurement is shown in Fig. 1. A weak probe beam pulse generated by an acousto-optic modulator is sent through the center of the cloud (beam diameter 2 mm, linewidth 2 MHz FWHM, pulse width typically 4 μs, 90%–10% fall time $1.5\tau_{nat}$). The diffuse light is collected in a solid angle of about 0.1 srd at 17° from the forward direction and is detected by a photomutiplier. The RT signal is averaged over 512

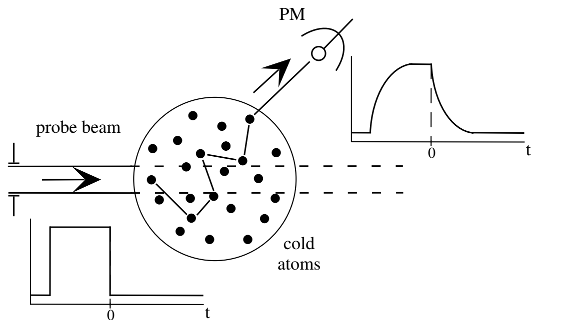

FIG. 1. Radiation trapping (RT) experimental scheme. A pulsed probe beam is sent through the center of a laser-cooled atomic cloud. The transmitted diffuse light is collected as a function of time in a solid angle close to the forward direction.

trap cycles. The signal-to-noise ratio allows a reliable measurement of the diffuse intensity down to about 1% of the initial, steady-state value.

Our analysis of the experimental data relies on the theory of multiple elastic scattering by resonant point scatterers [6,7] adapted to the case of cold atoms [14]. The propagation along a multiple scattering path can be decomposed into elementary steps involving a scattering event and the propagation in the effective medium up to the next scattering. The transport time τ_{tr} associated with this elementary step is the sum of the Wigner time delay τ_W for the scattering process and the propagation time with the group velocity. In our experimental conditions, the Wigner time (of the order of τ_{nat}) is much longer than the free propagation time ($\ell/c = 1$ ps for $\ell = 300\ \mu$m). The dynamics is thus "scattering dominated," in contrast to the usual media where it is "free propagation dominated."

The Wigner time delay, the group velocity, and the mean-free path vary rapidly in the vicinity of the atomic resonance. Even for a dilute gas (the case we consider here), where the index of refraction n_r of the medium is close to unity, the group velocity v_g—related to the derivative $dn_r/d\omega$—can be much smaller than c. The propagation time depends on the distance between two consecutive scattering events; its average value is obtained when it is equal to the mean-free path. For strongly resonant point scatterers, although τ_W, v_g, and ℓ depend on the detuning from resonance $\delta_L = \omega_L - \omega_0$, the average transport time per scattering event

$$\tau_{tr} = \tau_W + \frac{\ell}{v_g} \approx 1/\Gamma = \tau_{nat} \qquad (1)$$

is predicted to be equal to the natural lifetime, independently of the detuning from resonance [6]. Equivalently, the transport velocity can be written $v_E(\delta_L) = \ell/\tau_{tr} \approx \ell(\delta_L)/\tau_{nat}$. For an on-resonance mean-free path $\ell(0) = 300\ \mu$m, the transport velocity is 4 to 5 orders of magnitude smaller than c. For short scattering paths, the fluctuations of the path length induce fluctuations in the transport time. For more than a few scattering events, such fluctuations self-average, and the total transport time is the product of the scattering order by τ_{nat}.

The decay of the scattered light after the incoming laser beam is switched off can be written as a sum of decaying exponentials, each corresponding to an "eigenmode" of the RT, usually called a "Holstein mode" [8]. At late times, it is dominated by the lowest mode: the signal decays exponentially with a time constant τ_0. In the elastic diffusion theory, τ_0 is a function of the diffusion coefficient $D = v_E\ell/3 = \ell^2/3\tau_{tr}$ and of the thickness L of the medium, scaling like L^2/D. The detailed expression depends on the geometry of the medium. For large optical thickness b, one gets

$$\tau_0 \simeq \frac{3}{\alpha\pi^2}\tau_{nat}b^2, \qquad (2)$$

where α is a numerical factor whose value is 1 for a slab, 4 for a sphere, and 5.35 for an inhomogeneous sphere with Gaussian density [15]. The observation of τ_0 requires one to measure the signal at late times, once the higher-order modes have decayed.

In order to analyze the experimental results, we developed a Monte Carlo (MC) simulation of multiple scattering by the atomic cloud, described in [16]. In short, the calculation follows the propagation of a photon in the effective medium, with proper random choices of positions and directions of scattering events. We take into account the inhomogeneous density of the sample, the residual velocity of the atoms (Doppler effect), the recoil frequency shift, as well as the finite linewidth of the incoming laser. We compute the distribution of the scattering orders in the diffuse transmitted light and affect to the N^{th} scattering order the time delay $N\tau_{nat}$. This procedure yields the impulsional response of the system, which is convolved by the excitation to get the temporal profile of the transmitted intensity. This approach neglects interference effects between different multiple scattering paths responsible for the speckle pattern of the scattered light and, e.g., coherent backscattering; this is legitimate since we are in a dilute medium ($k\ell \gg 1$) and the speckle is averaged out in a large solid angle.

Examples of raw decay curves for resonant light ($\delta_L = 0$) are shown in Fig. 2(a) for different numbers of atoms in the cloud, i.e., for different optical thicknesses. Each curve has been normalized to the steady-state value ($t < 0$). After a transient at short time, an exponential decay is clearly visible. The dashed curves correspond to the MC simulations without an adjustable parameter. The values of τ_0 are obtained by fitting the signals to an exponential decay between roughly 10% and 1% of the steady-state value and displayed in Fig. 2(b) (circles). The solid line is the analytic prediction, Eq. (2). The dashed line is the result of the MC simulation. The different behavior of these two curves at large optical thickness is discussed below (Doppler effect included in the MC).

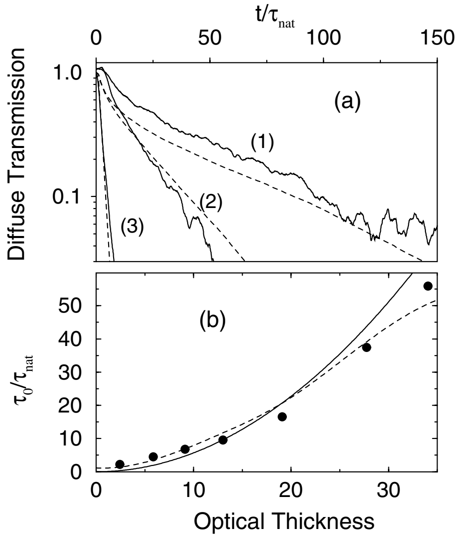

FIG. 2. Decay of resonant light trapped in the atomic cloud. We record the decay as we vary the number of atoms in the cloud. (a) shows the data for three optical thicknesses (solid lines): $b = 34$ (1), 19 (2), and 2.4 (3), and the Monte Carlo results (dashed lines). The measured decay constant τ_0 is plotted in (b) as a function of the optical thickness b (circles). It is compared to the prediction of Eq. (2) for a sample with Gaussian density (solid line), and to the MC calculation (dashed line).

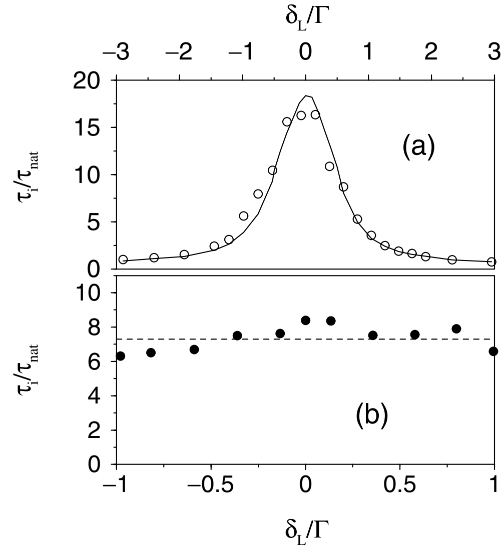

FIG. 3. Variation of transport time with frequency. We measure the "early" time ($2 < t/\tau_{\mathrm{nat}} < 10$) decay constant τ_{i} (in all these experiments, after an initial transient over $\approx 2\tau_{\mathrm{nat}}$ due to the higher Holstein modes, an exponential decay is observed for at least $10\tau_{\mathrm{nat}}$, as in Fig. 2) as a function of the detuning δ_{L} from resonance, in two separate experiments. (a) The number N_{at} of atoms in the cloud is fixed, with an optical thickness at $\delta_{\mathrm{L}} = 0$ of 22.4. The circles correspond to the experimental data and the solid line to the MC simulation. (b) For each value of δ_{L}, we adjust N_{at} to maintain a fixed optical thickness $b \approx 10.7 \pm 1.0$. τ_{i} is then nearly constant.

We observe, for an optical thickness $b = 34$, a decay constant $\tau_0 = 56\tau_{\mathrm{nat}}$. From this value and the experimental measurement of the mean-free path, we deduce (using an analysis based on the Holstein modes of a Gaussian cloud) the values of the minimum diffusion coefficient and transport velocity at the center of the atomic cloud: $D \simeq 0.66\ \mathrm{m}^2/\mathrm{s}$ and $v_{\mathrm{E}} \simeq 3.1 \times 10^{-5} c$.

The physical picture employed so far relies on the prediction that the transport time is frequency independent [6]. To check this nonintuitive assertion, we measured the RT decay as a function of detuning. To minimize the effect of frequency redistribution (see below), we measured the "early" decay constant τ_{i} at relatively short time, i.e., before frequency redistribution takes place, but after higher Holstein modes have decayed. Two distinct experiments were performed. First, we used a sample containing a fixed number of atoms N_{at} whose optical thickness varies as a Lorentzian with δ_{L}. The measured early decay constant (circles), plotted in Fig. 3(a), is maximum at resonance and has a width of roughly Γ. From Eq. (2), one expects $\tau_{\mathrm{i}}(\delta_{\mathrm{L}}) \propto b^2(\delta_{\mathrm{L}})\tau_{\mathrm{tr}}$. A small but significant difference with the expected b^2 behavior is due to the nonmonochromatic excitation, as confirmed by the MC simulation (solid line). In a second experiment, we adjusted N_{at} for each detuning value to maintain a constant optical thickness ($b = 10.7 \pm 1.0$). This cancels the trivial change of optical density with detuning. Since b is now constant, τ_{i} is directly proportional to the transport time. As shown in Fig. 3(b), this quantity is nearly constant (20% rms fluctuations, mean value $7.3\tau_{\mathrm{nat}}$), while, e.g., the scattering cross section varies by a factor of 5. This observation confirms the theoretical prediction of a frequency-independent transport time for resonant point scatterers, at least in the explored range. Note that these observations do not rely on the validity of the diffusion approximation, Eq. (2): indeed, the geometrical properties and the mean-free path—and thus the multiple scattering paths—are identical for all points in Fig. 3(b). The observation of a constant decay time is thus a direct proof of the constancy of τ_{tr}.

The main physical ingredient which distinguishes our MC approach from the elastic scattering theory of Eq. (2) is the Doppler effect. It may appear surprising that the velocity of the atoms plays a role in a laser cooled sample, where the Doppler width is negligible compared to the natural width (typical velocity spread v_{rms} such that $kv_{\mathrm{rms}} = \Gamma/30$). This is however a single scattering argument. In the course of multiple scattering, the frequency of the scattered wave performs a random walk resulting in frequency redistribution. For a large number N of scattering events, the frequency spreading is such that $\langle \delta\omega^2 \rangle \simeq (2/3)N(kv_{\mathrm{rms}})^2$. For an initially monochromatic excitation, the width of the frequency distribution will reach Γ after $N_0 \approx 250$ scattering events. Thus, frequency redistribution is expected to play a significant role for optical thicknesses larger than $b \approx \sqrt{N_0} \approx 16$ and cannot

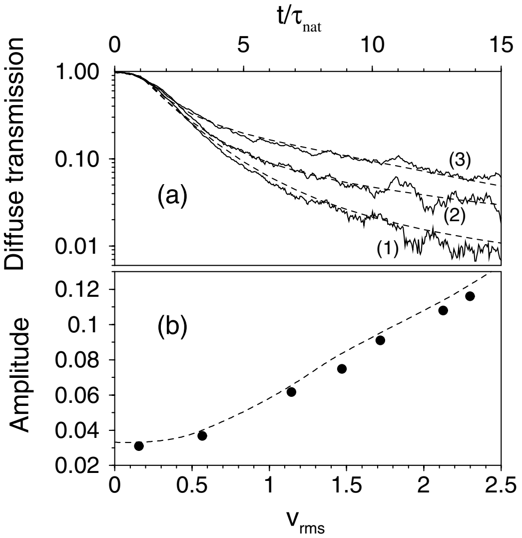

FIG. 4. Role of the temperature of the atomic sample. We measure RT as a function of the residual velocity v_{rms} of the atoms in the cloud, for detuned laser light $\delta_{\mathrm{L}} = +1.5\Gamma$. The optical thickness is fixed, $b = 2.2$. (a) shows three experimental decay curves, clearly nonexponential because of the occurrence of some frequency redistribution, for $v_{\mathrm{rms}} = 0.15$ (1), 0.42 (2), and 1.72 (3) m/s. We plot in (b) the amplitude of the decay signal after a time $t = 8\tau_{\mathrm{nat}}$, as a function of v_{rms}. Dashed lines are the MC results without adjustable parameters.

be neglected in our experiment. The effect of frequency redistribution on the RT decay depends on the laser detuning. For $\delta_{\mathrm{L}} = 0$, it results in a decrease of the effective optical thickness yielding a reduction of τ_0. In other words, on-resonance photons are trapped longer by colder clouds. For a detuned excitation $\delta_{\mathrm{L}} \neq 0$, the consequence is more subtle: at short times, when the frequency spread is small compared to $|\delta_{\mathrm{L}}|$, the light probes the atomic cloud with the initial detuning and the early decay constant τ_{i} is determined by the optical thickness at δ_{L} (see Fig. 3). However, given enough time, a fraction of the scattered light eventually reaches resonance where it feels the larger, on-resonance optical thickness. Thus, for detunings small enough so that a subsequent redistribution takes place before the photons leave the medium, the late-time decay constant τ_0 is independent of the initial detuning. We indeed observed this behavior in our experiment, which especially manifests itself as a nonexponential decay of RT as visible in Fig. 4. To verify the validity of our analysis, we recorded the RT decay as a function of the temperature of the sample (varied by applying an intense red-detuned molasses) for a detuned excitation. Figure 4(a) shows three experimental decay curves at increasing temperatures. As expected, the decay is slower for the higher temperatures. This is further illustrated in Fig. 4(b) where we plot the measured value of the decay curve at $t = 8\tau_{\mathrm{nat}}$ as a function of v_{rms}. In both (a) and (b), the dashed curves correspond to the MC results. The quantitative agreement is very good, confirming the role of temperature in the diffusion of light inside our sample.

In conclusion, we reported an experimental study of diffusive light transport in an optically thick sample of cold atoms. We measured a diffusion constant $D \simeq 0.66\ \mathrm{m}^2/\mathrm{s}$, corresponding to a transport velocity $v_{\mathrm{E}} \simeq 3.1 \times 10^{-5} c$. We confirm that the transport time is independent of frequency around resonance. We show that, even at temperatures in the 100 μK range, the frequency redistribution due to the residual Doppler effect influences the transport of light in optically thick samples. These results emphasize the interest of cold atoms both to reach new regimes, e.g., strongly resonant and monodisperse samples, and as a model system to check fundamental aspects of light scattering theory.

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