Radiation trapping in a cold atomic gas

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We experimentally study radiation trapping of near resonant light in a cloud of laser cooled rubidium atoms. Unlike in most previous studies, dealing with hot vapors, collisional braodening is here negligible and Doppler broadening due to the residual atomic velocity is narrower than the homogenous broadening. This is an interesting new regime, at the boundary between coherent and incoherent radiation transport. We analyze in detail our low temperature data (quasi elastic regime), then provide some experimental evidence for Doppler-based frequency redistribution. The data are compared with an analytical model valid for coherent transport and a Monte-Carlo simulation including the Doppler effect.

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I. INTRODUCTION

Ultra-cold atomic vapors have recently come to the attention of researchers in the field of mesoscopic physics [1]. Indeed, cold atoms can be manipulated with a high degree of control and mesoscopic samples with unprecedented purity and tunability can be assembled. An emerging field of study deals with the transport of near resonant light in such media. Using cold atoms, one can realize a monodisperse ensemble of strongly resonant point scatterers, free of defects and absorption, where the light mean free path can be varied continuously, simply by tuning the laser frequency without manipulation of the sample. These properties are very attractive to study localization of light in strongly scattering media. However, coherent transport requires that the light remains monochromatic, which is possible only if scattering is fully elastic. In the limit of low saturation of the atomic transition (e.g., at low light intensity for resonant light), the scattering by an atom at rest can be considered essentially elastic and no frequency redistribution is present. However, even at low temperature, the Doppler effect on the slowly moving atoms makes the frequency of the scattered photon to be slightly different from the frequency of the incoming photon. Moreover, this frequency change depends on the relative orientations of the atomic velocity and the incoming and outgoing wave vectors of the photons. Altogether, some frequency redistribution occurs because of the residual atomic motion (Doppler effect). For an atomic vapor which is laser cooled to approximately 80μK, the Doppler broadening is only a small fraction of the natural (homogenous) linewidth. Frequency redistribution is thus small, which indeed allows to observe interferential effects in the multiple scattering regime such as coherent backscattering [2, 3]. However, redistribution may become important if many scattering events are chained i.e., for samples of large optical thicknesses. A convenient mean to observe this is to study radiation trapping (RT). Trapping of resonant radiation in atomic vapors has been the focus of numerous studies dating back to the beginning of the 20th century. Holstein used in 1947 an hypothesis of complete frequency redistribution (CFR) at each scattering event – where the frequency of the outgoing photon is completely uncorrelated with the frequency of the incoming photon – to derive decay rates in various situations [4]. Since then, many theoretical approaches beyond CFR where employed to deal with experimental observations (see e.g. [5–10]). With the advent of laser cooling in the eighties, new experimental regimes became accessible towards the coherent propagation of light in atomic gases. However, it was only at the end of the 20th century that studies of light transport in optically thick clouds of cold atoms started to appear [11, 12]. In a previous paper [12], we focussed on transport in the quasi-elastic regime and showed that, owing to the strongly resonant character of atomic scatterers, the transport velocity which characterizes the propagation of energy in the diffuse light was more than 4 orders of magnitude smaller than the vacuum speed of light. In the present work, we discuss in more details the role of frequency redistribution in laser-cooled vapors, which offers the unique opportunity to study the transition from coherent to incoherent transport. A typical coherent effect is the existence of a speckle pattern when light is scattered by a disordered medium with frozen disorder. Although this has not been observed with atomic clouds in the multiple scattering regime, it is likely to exist if the atoms are sufficiently cold (coherent transport regime). With not-so-cold atoms, the scattered light has a relatively broad frequency distribution and interference effects responsible for speckle are likely to be washed out (incoherent transport). Another well-known interference effect is coherent backscattering, which is observed [13] to be quite sensitive to the residual atomic velocity. We first give in section II a very brief description of the principles of the experiment and numerical simulation. We then present in section III experimental data in the regime

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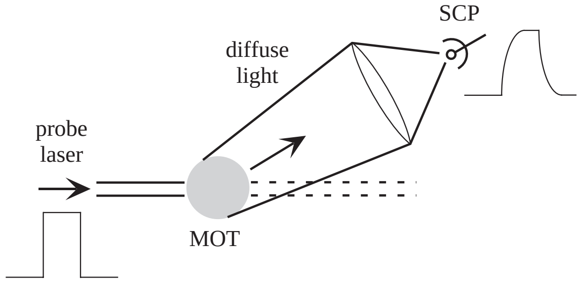

FIG. 1: Experimental setup. A cloud of laser cooled atoms released from a magneto-optical trap (MOT) is illuminated by a rectangular pulse of near resonant light. Part of the transmitted diffuse light is detected as a function of time by a single channel plate (SCP).

of quasi-coherent transport, corresponding to our lowest temperature of 80μK. The comparison between resonant and detuned excitation allows us to emphasize some peculiarities of RT in ultracold vapors. We then focus in section IV on the role of Doppler frequency redistribution in our experimental situation.

II. DESCRIPTION OF EXPERIMENT AND SIMULATION

In this section, we first briefly recall the main features of our experiment which was described in more details elsewhere [12]. We also shortly discuss the Monte-Carlo simulation used to analyze our results. Radiation trapping experiments in atomic gases are immensely simplified by the fact that the time step for the random walk of photons inside such media, the so-called *transport time* [14] in the regime of coherent transport, is of the order of the lifetime of the excited state (typically a few tens of nanoseconds). Even for samples of relatively low opacity, trapping times in excess of a microsecond are obtained. The requirements for pulsed excitation and time-resolved detection are thus much less stringent than in the case of non or weakly resonant samples [15].

The results presented in this paper are obtained using the experimental configuration shown in Fig. 1. A cloud of up to 10^{10} Rb^{85} atoms is collected in a magneto-optical trap (MOT), then released and further cooled down to $T = mv_{\rm rms}^2/(3k_B) = 80\mu$K using optical molasses. Here, $v_{\rm rms} = 0.15$m/s is the *rms* velocity modulus, yielding a corresponding Doppler shift $kv_{\rm rms}/\Gamma \approx 1/31$ (where $\Gamma/2\pi = 5.9$MHz is the homogenous width of the atomic transition, the inverse of the excited state lifetime). We then turn off all trapping light and illuminate the sample with a short pulse (typical duration a few μs, switch-off time 100ns) of low intensity near resonant light. The on-resonance saturation parameter is typically below 5×10^{-2}, which means that the scattering process is mainly elastic. Also, with a probe pulse duration of typically a few μs, less than ten photons are exchanged per atom and hyperfine pumping or mechanical effects can be neglected. The probe beam, whose diameter is typically 1-2 mm, passes through the center of the spherical, roughly Gaussian-shaped atomic cloud. We then collect the *diffuse* light transmitted by the sample in a solid angle 0.1 srd, as a function of the time elapsed after the probe is switched off. The diffuse light is detected by a SCP (Single Channel Plate). By adjusting the frequency of the molasses beams, we can tune the temperature of the cloud i.e., the width of the atomic velocity distribution. Using this method, we can vary the ratio of the Doppler width to the homogenous (natural) width from 5×10^{-2} to $\simeq 1$. Attempts to further heat the gas resulted in a drastic reduction of the steady-state atomic population in the MOT. The total number N of atoms in the cloud can be adjusted independently of temperature by switching off the repumper laser shortly before the trapping beams. The time-resolved fluorescence signal is averaged over 512 experimental cycles. The resulting signal-to-noise ratio allows us to observe the RT decay up to approximately 10^{-2} of the initial, steady-state value of the diffuse transmission. We compare our experimental data to a Monte-Carlo simulation which was initially developed to explain our coherent backscattering signals [16]. The features of this numerical simulation are detailed in [17]. The new ingredient is the atomic velocity. For all atoms, we assume independent isotropic Gaussian velocity distributions, identical at all spatial positions. During propagation of a photon in the atomic medium, the probability of the photon being scattered by an atom is chosen according the local atomic density (not uniform in the Gaussian-shaped atomic cloud), the atomic velocity distribution and the frequency of the incoming photon. The atomic velocity along the incoming wave vector and the scattering probability are of course correlated: a photon with frequency red-detuned from resonance is more likely to be scattered by an atom with velocity opposite to the wave vector of the photon, so that the frequency in the atomic rest frame is closer to resonance. This in turn introduces a correlation between the frequency of the incoming photon and the frequency of the scattered photon. At very low velocity, the frequency change at each scattering event is very small – much smaller than the natural linewidth Γ – making the photon frequency slowly changing along a multiple scattering path. In contrast, if the atomic velocity is large enough – roughly when the Doppler shift $kv_{\rm rms}$ is comparable or larger than Γ – the frequency shift at each scattering event will typically be of the order of Γ and the frequency of the scattered photon will be significantly different from the incoming frequency. Note however that, even when the Doppler broadening is by far dominant, correlations between the incoming and the scattered frequencies cannot be neglected. This is the so-called partial frequency redistribution (PFR) regime [7]. In the

Monte-Carlo calculation for a medium with uniform density like a slab, the step length until the next scattering is randomly chosen according to an exponential distribution, using the mean free path evaluated at the scattered photon frequency, as obtained from the effective scattering cross section, which is the "natural" Lorentzian convolved with the Gaussian velocity distribution. The global jump distribution of photons in the medium results from the superposition of various exponentials with various lengths, and is thus non exponential for non-zero temperatures; it is typically a Levy distribution [18]. In a medium with non-uniform atomic density, even at fixed frequency, the jump distribution is not universal, but depends on the optical depth along the propagation path. How to take into account this effect has been explained in [17].

Finally, we monitor the number n of scattering events undergone by the photon before exiting the medium. As discussed in [12], for large n, the time spent by the photon inside the medium is simply $t \approx n \times \tau_{\rm nat}$ where $\tau_{\rm nat} = 27$ns is the excited state lifetime. Note that this property is true independently of the frequency of the photon, provided the medium is optically thick. This is far from obvious and results from the combination of the Wigner time delay at each scattering event and the effect of the index of refraction of the atomic medium which changes the effective velocity of the photon. Both effects depend on the detuning from resonance, but the global effect does not. This is valid only on average; for very short paths, large fluctuations may exist between various photons. This is however not relevant in our case, as we are interested in relatively large scattering orders. This approach was quite successful in reproducing our earlier results on radiation trapping from a cold atomic cloud [12].

III. QUASI-ELASTIC REGIME: SHAPE OF THE RADIATION TRAPPING DECAY

This section is devoted to the discussion of RT signals obtained at our lowest temperature of 80μK. There, Doppler frequency redistribution is *a priori* expected to play a rather insignificant role. We thus refer to this situation as the "quasi-elastic regime".

A. Resonant excitation

In the situation of elastic (or coherent) transport and monochromatic excitation, the RT decay is fully determined by the sample's geometry and optical thickness $b = -\log(T_c)$, where T_c is the "coherent" transmission i.e., the proportion of incident light that has experienced no scattering. One of the most remarkable characteristic of cold atomic vapors is their extremely resonant behavior. Indeed, the scattering cross-section for an atom at rest is:

$$\sigma(\delta) = g\frac{3\lambda^2}{2\pi}\frac{1}{1+4(\delta/\Gamma)^2} \tag{1}$$

Here, g is the degeneracy parameter which depends on the specific atomic transition. Assuming a uniform distribution among the Zeeman sub-states of the $F = 3 \to F' = 4$ transition of the D2 line of Rb^{85} (and no optical pumping), one finds $g = 3/7$. $\delta = \omega - \omega_0$ is the light detuning from the resonance, where ω is the angular frequency of the light and ω_0 corresponds to the atomic resonance. For atoms at rest, the optical thickness seen by a weak monochromatic probe beam of detuning δ passing through a diameter of the cloud (assumed isotropic) is determined by three parameters:

$$b(N, \delta, w) = \sqrt{\pi/2}\sigma(\delta)\frac{N}{w^2} \tag{2}$$

where N is the total number of atoms, w the $1/e^2$ radius of the Gaussian density profile. Note that for non-zero temperatures the optical thickness will depend in addition on $v_{\rm rms}$, since the Lorentzian cross-section has to be convolved by the Gaussian velocity distribution. Because of the sharp resonance, the width and shape of the incident light's spectrum will have to be accounted for in the analysis of RT signals, as will be discussed later. In the following, we first consider the case of monochromatic incident light. For a simple geometry like a slab, the impulsional response (for the diffuse transmitted intensity) of a sample of optical thickness b can be analytically found by solving the diffusion equation with appropriate boundary conditions (see e.g. [19]). Indeed, for purely monochromatic excitation and propagation, the photons follow a random walk inside the atomic medium. Thus the density of electromagnetic energy has a diffusive behavior. For simple geometries, this equation can be solved. The impulsional response of the system is a decreasing function of time if the medium is optically thin $b < 1$, because the diffuse transmission is dominated by single scattering. For an optically thick medium, the impulsional response in the transmitted direction is peaked around a most probable time $t_{\rm diff} \propto b^2$, the typical time needed to diffuse throughout a sample of optical thickness b. In any case, the long time decay of the diffuse intensity is exponential with a time constant τ_0 characteristic of the so-called fundamental Holstein mode [12]:

$$\tau_0^{\rm el} = \frac{3}{\alpha\pi^2}b^2\tau_{\rm nat} \tag{3}$$

where α is a geometry-dependent parameter (1 for a slab, 4 for a homogenous sphere and 5.35 for a Gaussian-density sphere). In this specific case of purely monochromatic photons, the Holstein mode is simply the solution of the time-dependent Milne equation [7] with the smallest decay rate. Expression (3) is only valid in the diffusive approximation, i.e., for rather large b's. A more

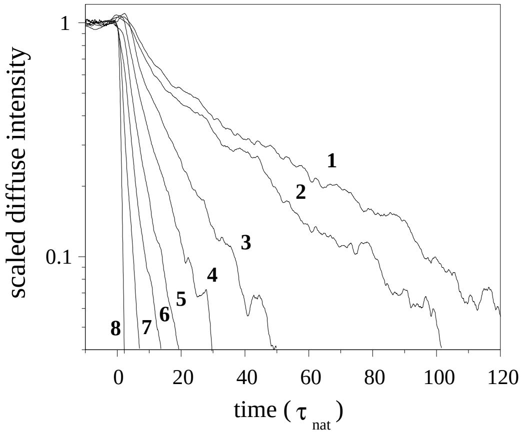

FIG. 2: Experimental radiation trapping decays for resonant excitation. We plot the diffuse transmitted intensity for resonant excitation $\delta = 0$ and different optical thicknesses: $b = 34$ (**1**), $b = 28$ (**2**), $b = 19$ (**3**), $b = 13$ (**4**), $b = 9$ (**5**), $b = 6$ (**6**) and $b = 2$ (**7**). Trace **8** corresponds to the switching off of the probe. In all cases, an approximately exponential decay is observed at long times.

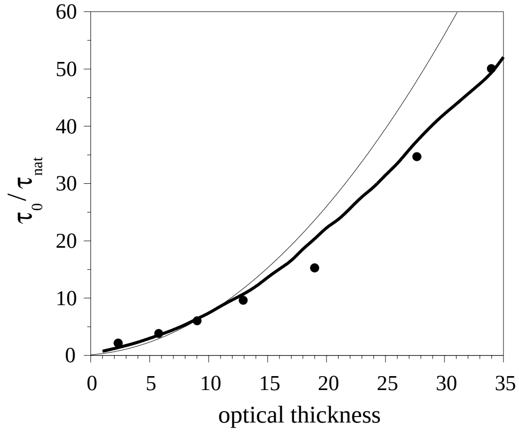

FIG. 3: Decay time constant for resonant excitation. We plot (symbols) the late decay constants – in units of the lifetime of the atomic excited state – extracted from the data of Fig. 2. The thin line is the prediction from eq. (3), valid at zero temperature. The bold line is a Monte-Carlo simulation taking into account the atomic *rms* velocity $v_{\rm rms} = 0.15$m/s, the shape of the atomic medium and the finite linewidth of the laser excitation.

accurate expression requires to take into account properly the boundary conditions; this can be done by replacing the optical thickness b by $b + 2z_0$ in Eq. (3), where $z_0 \simeq 0.7$ is the so-called extrapolation length.

However, our experiments are not performed in the impulsional regime, but instead using a rectangular probe pulse long enough so that a steady-state is reached for the diffuse intensity at the end of the pulse (pulse duration $\gg t_{\rm diff}$). Throughout this paper, we will refer to this steady-state value as the "diffuse transmission" T_d. The measured RT signal is thus the convolution of the impulsional response by the excitation profile. Equivalently, it can also be seen as the decay of electromagnetic energy initially trapped in the atomic medium. We present in Fig. 2 some experimental decay signals obtained for resonant light ($\delta = 0$) and different optical thicknesses (obtained by varying N), at our lowest temperature of about 80μK. As can be seen, the RT decay is almost purely exponential after an initial short period of faster decay ($t < 10\tau_{\rm nat}$), due mostly to the finite linewidth of our probe laser (see discussion at the end of the section). The time constant of the exponential decay, plotted in Fig. 3, is in decent agreement with the prediction of expression (3) (line). Note however the increasing discrepancy at large b's, which originates from the non-zero temperature of the vapor. The analysis of the role of temperature is the object of section IV.

B. Detuned excitation

Expression (2) suggests two ways of varying the optical thickness, either by tuning N or δ. In the absence of frequency redistribution, one would expect equivalent behaviors for the RT decay. We thus recorded RT signals using both methods to vary the optical thickness and observed markedly different results. The most striking observation is a clear non exponential decay in the case of detuned excitation, in contrast with what is observed in the resonant situation. An illustration is given in Fig. 4, where both thick curves are experimental data obtained with the same sample but different excitation frequencies: $\delta = 0$ (**A**) and $\delta = 1.1\Gamma$ (**B**). The on-resonance optical thickness is $b = 33$. Although the initial decay is much faster in the detuned case, owing to the smaller optical thickness $b(\delta = 1.1\Gamma) \approx 5$, it can be clearly seen that the late time decay is quite identical in both cases. We will discuss the various MC simulations (thin lines) in the following. A more complete characterization of this behavior can be carried out by measuring the evolution of the earliest and latest identifiable decay constants in the signal, as a function of the light detuning for a fixed number of atoms in the cloud. This is done in Fig. 5, where we plot the early constant τ_i (open symbols) and the latest constant τ_l (solid circles) for a cloud of on-resonance optical thickness $b(\delta = 0) = 33$.

We observe that the early time constant τ_i closely behaves like a Lorentzian of width Γ (bold line in Fig. 5).

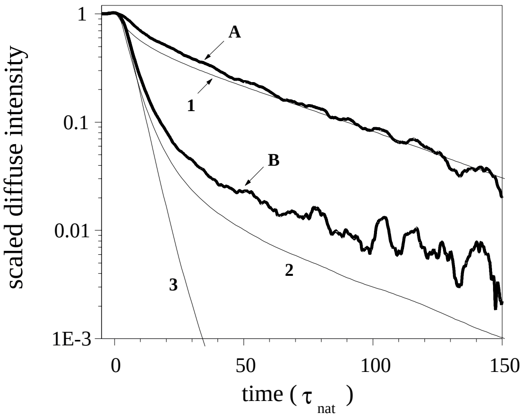

FIG. 4: Radiation trapping for a detuned excitation. We plot (bold line **B**) the diffuse transmitted intensity for a detuning $\delta = 1.1\Gamma$, showing a clear non-exponential decay. For comparison we also show (**A**) the signal from the same cloud but $\delta = 0$ ($b(\delta = 0) = 33$). The *rms* atomic velocity is 0.15m/s. Note the identical decay rates at large times. The thin lines are Monte-Carlo simulations (see discussion at end of section): $\delta = 0$, $v_{\rm rms} = 0.15$m/s, $\Delta\nu = 2.35$MHz (**1**); $\delta = 1.1\Gamma$, $v_{\rm rms} = 0.15$m/s, $\Delta\nu = 2.35$MHz (**2**); $\delta = 1.1\Gamma$, $v_{\rm rms} = 0.15$m/s, $\Delta\nu = 0$ (**3**). The observed non-exponential decay is clearly dominated by the laser spectrum for these parameters. Frequency redistribution induced by the residual Doppler effect on the atoms plays only a minor role at such a low temperature.

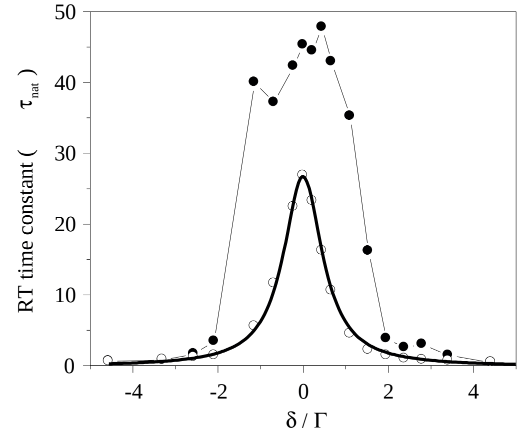

FIG. 5: Radiation trapping decay rate for a detuned excitation. This graph shows the earliest (open circles) and latest (solid circles) decay constants which can be identified in the diffuse transmitted intensity, in an experiment where δ is varied for a fixed number of atoms in the cloud ($b(\delta = 0) = 33$). The bold line is a Lorentzian fit of the early time constant, with FWHM$\approx \Gamma$.

However, we expect this early decay to be determined by the optical thickness at the laser detuning δ. Indeed, we will see in section IV A that frequency redistribution is in our case a slow process which requires many scattering events and thus some time to become significant. Assuming that the fundamental Holstein mode corresponding to $b(\delta)$ is reached rapidly, one would expect from eq. (3) $\tau_i \propto b(\delta)^2 \propto b(0)^2/[1 + 4(\delta/\Gamma)^2]^2$, thus to behave as a *squared* Lorentzian. The observed discrepancy originates from two reasons: firstly, at small b, the photon performs a random walk in the atomic medium, but – because it undergoes only a small number of scattering events before leaving the medium – such a random walk is not well approximated by a diffusive motion. It is only when the number of scattering events is large that the diffusion approximation is a good description of the random walk. As a consequence, at small b, the diffusion approximation breaks down and the decay rate is not quadratic but rather linear in b as can be seen on the experimental points and the Monte-Carlo simulation in Fig. 3. Secondly, the laser is not purely monochromatic, but has some finite linewidth. This finite excitation bandwidth is also responsible for the faster initial decay observed on the curves of Fig. 2, due to the escape of detuned photons from the wings of the laser line. As a result, the initial decay constant for $\delta = 0$ is almost a factor 2 smaller than τ_0. Coming back to Fig. 5, we see that τ_l exhibits a different behavior: it is nearly constant at $\simeq 45\tau_{\rm nat}$ (which is consistent with the data of Fig. 3 and $b = 33$) for a detuning range of roughly 3Γ centered on resonance, then abruptly decreases for larger δ's. This sudden drop is a consequence of our limited signal to noise ratio $SN \approx 10^2$: the cross-over between different decay constants occurs at times where the signal has dropped below the noise level. Thus one may conclude that given an arbitrarily large SN, the late decay constant is always $\tau_0(\delta = 0)$ regardless of the value of the laser detuning. The reason for this is discussed below.

¿From the previous analysis, we conclude that for the same optical thickness, photons last longer in the sample for a detuned excitation. This is further emphasized in Fig. 6, where we compare the data of Figs. 2 and 5, by plotting the area under the normalized RT decay curves (which is a measure of how long photons are trapped inside the medium on average) as a function of the optical thickness, varied either by tuning N at $\delta = 0$ (stars) or δ at fixed N ($\delta < 0$: open circles; $\delta > 0$: solid circles). We observe that the area in the detuned case can be more than twice that of the resonant case for b of the order of 10 ($\delta \approx 0.8\Gamma$). The observed difference between resonant and detuned excitation could have two possible causes: frequency redistribution or finite laser linewidth. As discussed in section IV A, due to the residual velocity spread in our atomic vapor, photons experiencing enough scattering events will diffuse also in frequency space, some of

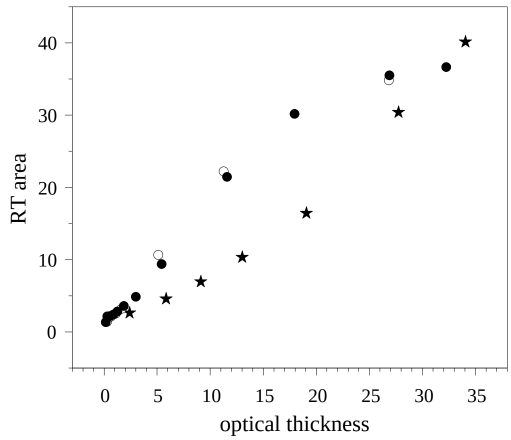

FIG. 6: Resonant versus detuned excitation. We plot the area under the RT decay signal (steady state scaled to 1), as a function of the optical thickness varied using two methods: a) by varying N at a fixed detuning $\delta = 0$ (stars); b) by varying δ at fixed N ($\delta < 0$: open circles; $\delta > 0$: solid circles). The fact that the two sets of results differ significanty proves that the behaviour of the system does not depend only on the optical thickness at the center of the laser line, and thus that non-monochromatic effects must be taken into account. Photons with different frequency may either come directly from the non-monochromatic laser or may be dynamically created in the medium by frequency redistribution induced by the residual Doppler effect.

them getting closer to the resonance and thus remaining trapped longer inside the cloud. On the other hand, a noticeable feature in our highly resonant cloud of cold atoms is the sensitivity of RT to the linewidth of the exciting radiation. Let us consider e.g., a Lorentzian lineshape for the laser whose central frequency is detuned from the atomic resonance by δ. Incoming photons at this frequency experience an optical thickness as given by expressions (2) and (1), and will leave the cloud at a rate given by (3). However, a few photons in the wings of the laser spectrum are resonant with the atomic transition: their abundance relative to that of center-of-line photons is $p(\delta) = 1/(1+4(\delta/\Delta\nu)^2)$ where $\Delta\nu$ is the laser linewidth (FWHM). These photons "see" the maximal optical thickness $b(\delta = 0)$ and will thus decay very slowly. Considering only these two frequency components, the RT signal will consist of two exponential decays:

$$RT(t) \propto T_d(\delta)e^{-t/\tau_0(\delta)} + p(\delta)T_d(0)e^{-t/\tau_0(0)} \qquad (4)$$

where T_d is the diffuse transmission which depends on the optical thickness and thus on the light frequency.

The diffuse transmission is a linearly increasing function of b at $b \ll 1$, exhibits a maximum at $b \simeq 2$, and decreases as $1/b$ at large b [20]. For typical experimental situations where both $b(0)$ and $b(\delta)$ are substantially larger than unity, the amplitude of the second exponential component in the expression above is much smaller than that of the first one. If we choose $b(0) = 30$, $\delta = \Gamma$ and a laser linewidth $\Delta\nu = 2$ MHz, we find that the crossover between the two exponentials occurs at a time $t \simeq 11\tau_{\mathrm{nat}}$, when the RT signal has fallen to 3×10^{-3} of its initial value. This is of course a very rough estimate since we didn't account for all the other frequency components of the laser line. Curve **2** in Fig. 4 shows the actual RT profile simulated using the experimental parameters and a laser linewidth $\Delta\nu = 2.35$MHz (a value compatible with the result of heterodyne measurements). As can be seen, the simulated profile is quite similar to the experimental one (**B**) and we may conclude that the spectrum of the laser is probably responsible for most of the observed non-exponential behavior at this temperature. This is confirmed by curve **3** where the atomic velocity spread was included but not the finite spectrum of the excitation (monochromatic laser). To look for signatures of frequency redistribution in our sample, we must thus directly study the effect of the vapor temperature on radiation trapping.

IV. EFFECT OF CLOUD TEMPERATURE ON RADIATION TRAPPING

In this section, we concentrate on the role of Doppler frequency redistribution in our regime of an ultracold gas. We present some RT experiments performed as a function of temperature, which allow us to obtain some evidence of frequency redistribution.

A. Frequency redistribution in laser-cooled gases

As already mentioned, in the standard operating mode of the MOT, the residual Doppler broadening is much smaller (by a factor of about 30) than the natural width Γ of the transition (homogenous broadening). This is of course a situation where CFR fails, since the outgoing photon has almost the same frequency than the incoming one and frequency correlation is high. In this regime, frequency redistribution is a slow process which can be viewed as a diffusion in frequency space, characterized by a diffusion coefficient (for resonant incoming photons) $D_\omega = 2/3(kv_{\mathrm{rms}})^2\Gamma$. This means that with our 80μK temperature, about 250 scattering events are needed for the frequency distribution of the diffuse light to spread over a width of Γ after an initial monochromatic excitation. For the description to be complete, one also needs to mention the recoil effect associated with both absorption and emission of photons by atoms. These processes give rise to both a systematic red shift of the scattered light (a drift term, linear in the number n of scattering events) and also a diffusion term scaling like $\sqrt{n}$ (of magnitude smaller than the drift for large n's). However, the recoil-induced drift of the frequency

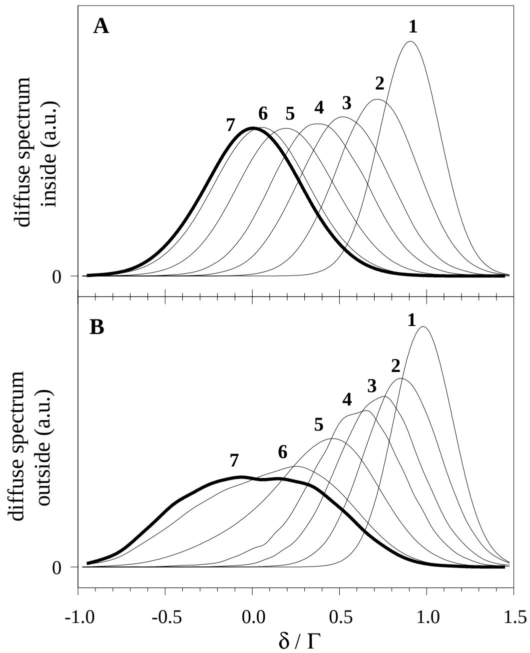

FIG. 7: Frequency redistribution in a laser-cooled vapor (simulation). We plot the evolution of the frequency distribution as a function of time, for an initially detuned excitation $\delta = \Gamma$: t= $10\tau_{\rm nat}$ (**1**), t= $20\tau_{\rm nat}$ (**2**), t= $30\tau_{\rm nat}$ (**3**), t= $40\tau_{\rm nat}$ (**4**), t= $60\tau_{\rm nat}$(**5**), t= $100\tau_{\rm nat}$ (**6**) and t= $160\tau_{\rm nat}$ (**7**). The on-resonance optical thickness is 33, the atomic *rms* velocity 0.3m/s and the excitation is monochromatic. Plot **A** refers to the photons trapped in the bulk of the atomic cloud, while **B** refers to the photons in the diffuse transmitted intensity. In the bulk, the initial diffusive broadening is clearly visible, together with a progressive migration of photons towards resonance. As off-resonant photons escape more easily the medium, the frequency distribution in the diffuse transmitted intensity is somewhat different. Photons close to resonance are less numerous, and off-resonance photons – both blue and red detuned – are important at late times. For the latest time (bold line), a steady-state spectrum is reached.

distribution is usually much slower than the Doppler-induced spreading: the relative drift per scattering event is only $kv_{\rm recoil}/\Gamma \approx 10^{-3}$ (the recoil velocity for rubidium is $v_{\rm recoil} = 6 \times 10^{-3}$m/s).

It thus is negligible for the typical experimental parameters we are dealing with, although it is visible in our Monte-Carlo simulations and could become important for lower temperatures or larger optical thicknesses. One expects the impact of frequency redistribution to be quite different for resonant or detuned excitation. In the resonant case, frequency redistribution can only result in a gradual detuning of the scattered photons and thus in a faster RT decay rate. For detuned excitation, photons can diffuse closer to resonance thus experiencing an increased optical thickness and decaying at a slower rate. Given enough time, some photons will eventually reach resonance and the late decay rate should be identical to that of resonant excitation. Thus, frequency redistribution results in a non exponential decay for detuned excitation, a signature superficially similar to that of polychromatic excitation as discussed in the previous section. This description is illustrated in Fig. 7 where we follow, using the Monte-Carlo simulation, the frequency spectrum of the trapped and emerging radiations as a function of time. The trapped photons, initially monochromatic, display a diffusive broadening of the frequency distribution, together with a drift of the mean frequency towards resonance (Fig. 7(**A**)). At late time, an equilibrium between the diffusive broadening and the drift towards resonance is reached; the resulting frequency distribution is close to a Gaussian whose width depends on the average atomic velocity and on the optical thickness of the medium. If the recoil effect is included in the Monte-Carlo simulation, the situation is essentially identical, but the final frequency distribution is not centered around the resonance frequency, but very slightly red detuned. Following the ideas exposed in [21], adapted to the specificities of ultra-cold atoms, it is possible to calculate more or less analytically these behaviors. This is however beyond the scope of this paper and will be published elsewhere [22]. The frequency spectrum of the diffuse transmitted intensity is also shown, in Fig. 7(**B**). It is significantly different from the spectrum of the photons trapped in the bulk of the atomic cloud. The reason is easy to understand: far-detuned photons see an optically thinner atomic medium and consequently escape more easily from the cloud. Thus the spectrum of the diffuse transmitted intensity is more important in the wings. If the atomic velocity is larger, and consequently the frequency spectrum broader, one can even observe the “line reversal” phenomenon – well known in stellar atmospheres – where the spectrum of the diffuse transmitted intensity has a minimum at resonance and two symmetric bumps off-resonance. This result is in marked contrast with the situation of ref [23], where a gradual flip between the excitation band and the inhomogeneous line was observed. The origin of this difference, the fact that at the absorption edge the line shape is dominated by homogenous broadening, was already recognized by the authors.

B. Experiments: resonant excitation

We performed a series of experiments where we varied the temperature of our atomic vapor by applying a near resonant optical molasses. The 3D velocity distribution was inferred by monitoring the ballistic expansion of the cloud after the heating phase. Due to the heating, the recapture efficiency by the MOT was decreased which yielded a temperature-dependent number of atoms in the cloud. We first present in Fig. 8 results for a resonant excitation. Note that in this case we always observe an

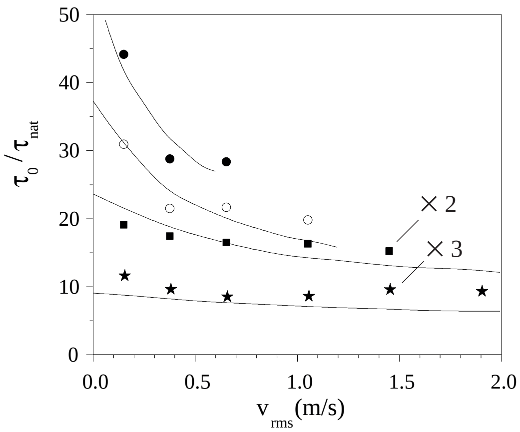

FIG. 8: Effect of temperature on RT decay ($\delta = 0$). We plot the experimental RT decay constants as a function of the atomic *rms* velocity, for various on-resonance optical thicknesses: $b = 30$ (solid circles); $b = 25$ (open circles); $b = 13$ (solid squares, $\times 2$); $b = 5$ (stars, $\times 3$). The lines correspond to the MC simulations.

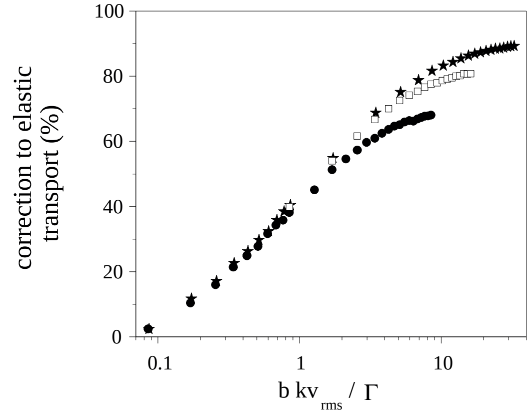

FIG. 9: Different regimes of frequency redistribution (MC simulations, slab geometry). We plot the deviation to elastic decay $1 - \tau_0/\tau_0^{\mathrm{el}}$ as a function of the product $bkv_{\mathrm{rms}}/\Gamma$ (where v_{rms} is varied) for different values of the optical thickness: $b = 10$ (circles), $b = 20$ (squares) and $b = 40$ (stars). We find a universal behavior (the deviation depends only on $bkv_{\mathrm{rms}}/\Gamma$) for $kv_{\mathrm{rms}}b/\Gamma < 1$ (frequency diffusion regime), while for $kv_{\mathrm{rms}}b/\Gamma \gg 1$ the deviation depends only on b (Doppler regime).

exponential decay for the RT signal.

We plot the measured decay constants as a function of the *rms* velocity of the atoms, for different on-resonance optical thicknesses. For a given curve in Fig. 8, the optical thickness is maintained *fixed* by tuning N while the temperature is varied. Clearly, τ_0 decreases with increasing temperature. This is a clear effect of frequency redistribution: resonant photons can be brought out of resonance by Doppler effect, and more easily escape the medium. An important parameter is the ratio of the typical Doppler shift kv_{rms} to the natural linewidth Γ. When it is larger than unity, the Doppler effect is dominant. If frequency redistribution were complete, the decay rate would be given, for large on-resonance optical thickness b [4, 7], by:

$$\tau_0 = \frac{b\sqrt{\log{(b/2)}}}{1.05}\ \tau_{\mathrm{nat}} \tag{5}$$

which is much smaller than the purely monochromatic expression, Eq. (3). Although CFR is known not to be true in the case of pure Doppler broadening, this expression gives a fairly good approximation of the decay rate in the pure Doppler case: the true value differs by less than 10% [7]. Where does the transition between the monochromatic regime and the Doppler regime take place? Obviously, from the experimental results, this depends on the optical thickness b. The larger the optical thickness, the faster the decay of τ_0 with velocity.

This can be understood with the following arguments: at small atomic velocity, the photon transport can be described by a diffusive motion *both* in space and frequency. This is of course not valid strictly speaking as first shown in [4], but we think that it is a reasonable assumption at least in our lowest temperature range. The frequency distribution of the photons trapped in the medium is then a Gaussian which spreads at a rate $(D_\omega t)^{1/2}$. A resonant photon typically performs b^2 scattering events before escaping the medium. Thus, one expects the frequency redistribution to be small and the decay rate unaffected by the residual Doppler effect if the width of the Gaussian at time $b^2\tau_{\mathrm{nat}}$ is smaller than Γ, i.e.:

$$kv_{\mathrm{rms}}b \leq \Gamma \tag{6}$$

This equation defines the regime where frequency redistribution is a small effect. On the other hand, the pure Doppler regime, where Eq. (5) is approximately valid, is reached only at much higher velocity, $kv_{\mathrm{rms}} \geq \Gamma$. In the whole range $\Gamma/b < kv_{\mathrm{rms}} < \Gamma$, the decay rate smoothly decreases towards the Doppler limit. This analysis is supported by the curves in Fig. 9, where we plot the deviation from elastic decay $1 - \tau_0/\tau_0^{\mathrm{el}}$ as a function of v_{rms} for different values of the optical thickness (along a given curve, the number of atoms is varied with v_{rms} to maintain b fixed). For $kv_{\mathrm{rms}}b < \Gamma$, the decay constant is determined only by the value of $kv_{\mathrm{rms}}b/\Gamma$ (frequency diffusion regime). On the contrary, for $kv_{\mathrm{rms}}b > \Gamma$ the curves at different b's split and saturate; this is the Doppler regime where τ_0 depends only on b (Eq. (5). For low temperature – at fixed atomic velocity – the change from the monochromatic regime where τ_0 increases quadratically with b to the Doppler regime where it increases

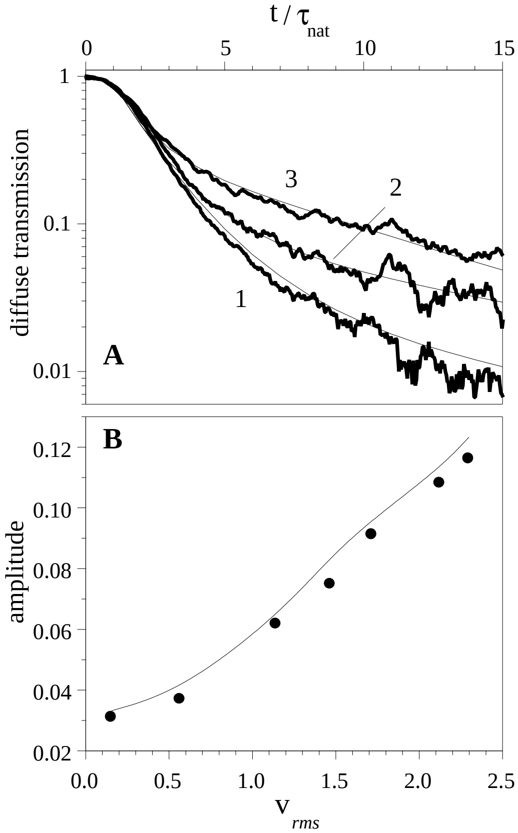

FIG. 10: Effet of temperature on radiation trapping decay ($\delta \neq 0$). We plot in **A** the measured radiation trapping decays for a fixed detuning $\delta = 1.5\Gamma$ and different velocity spreads in the atomic vapor: **1** $v_{\rm rms} = 0.15$m/s; **2** $v_{\rm rms} = 0.42$m/s; **3** $v_{\rm rms} = 1.72$m/s. For each temperature, the total number of atoms has been adjusted to maintain a fixed optical thickness $b = 2.2$. We plot in **B** the measured RT amplitude at time t= $8\tau_{\rm nat}$ for a light detuning $\delta = 1.5\Gamma$ and various temperatures. As temperature is increased, more photons remain trapped inside the cloud after a given time, indicating an increased RT efficiency due to frequency redistribution.

only linearly (forgetting the small logarithmic correction in Eq. (5)) is readily visible in Fig. 3 around $b = 30$. This is perfectly compatible with the measured atomic velocity, as we have then $kv_{\rm rms}b/\Gamma = 1$.

C. Experiments: detuned excitation

We now turn to experiments with a detuned excitation. We monitor the RT signal for a fixed detuning $\delta = 1.5\Gamma$ and different sample temperatures. To allow for a direct comparison between the data, we adjusted N for each temperature to maintain a fixed optical thickness of the sample [24]. The corresponding decay curves are shown in Fig. 10(**A**). As can be seen, the decay is now clearly non exponential. The overall decay is slower as the gas temperature is increased, which reflects the fact that photons can diffuse faster in frequency space and thus get closer from resonance. This is illustrated in Fig. 10(**B**), where we plot the value of the normalized RT signal measured after a time t= $8\tau_{\rm nat}$ for a laser detuning $\delta = 1.5\Gamma$. The steady increase of the RT amplitude with temperature is a clear signature of frequency redistribution. The typical velocity for which frequency redistribution plays an important role is of the order of 1 m/s – between curves 2 and 3 in Fig. 10(**A**) – although the single scattering typical Doppler effect $kv_{\rm rms}$ is only 20% of the linewidth. This is again a clear effect of multiple scattering and in good agreement with Eq. (6).

V. CONCLUSION

We presented a detailed study of radiation trapping of near-resonant light in a cloud of laser-cooled atoms. Tuning the temperature of the vapor allowed us to explore a new boundary between coherent and incoherent transport. At our lowest temperature of 80μK (quasi-elastic regime), our findings are consistent with the theory of (coherent) multiple scattering. The study of RT for detuned excitation allowed us to highlight the intrinsic sensitivity of this strongly resonant system to the excitation spectrum. We then studied the impact of the gas temperature on RT and found some indirect evidence of frequency redistribution due to the residual atomic velocity spread. This approach is complementary to another work [13] where we look how frequency redistribution affects *coherent* properties of transport such as coherent backscattering.

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