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Saturation-induced coherence loss in coherent backscattering of light

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We use coherent backscattering of light by cold strontium atoms to study phase-breaking mechanisms in the multiple-scattering regime. As the probe light intensity is increased, the atomic optical transition starts to be saturated. Nonlinearities and inelastic scattering then occur. The latter induces a characteristic phase-breaking time that reduces the wave coherence. In our experiment, this leads to a strong reduction of the enhancement factor of the coherent backscattering cone. The results at different probe detuning are also presented.

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In mesoscopic physics, be it in photonic, atomic, or solid-state physics, most interesting phenomena are due to interference as exemplified by the weak localization corrections to the conductivity, universal conductance fluctuations, or even random lasers [1–4]. However, these subtle interference effects are very sensitive to phase-breaking mechanisms, like coupling of the waves to their environment. This imposes a characteristic phase-breaking time (or length) for the wave to maintain its coherence [5]. This is why mesoscopic samples have generally to be cooled down well below the Kelvin range to overcome the limitation of a short phase-breaking time [1]. Note, however, that contributions to phase-breaking mechanisms still exist even at zero temperature, for instance, spin-flip scattering [6].

Studying phase-breaking mechanisms in optical wave transport with cold atomic gases offer several advantages. Indeed, atomic samples are efficiently cooled down by laser techniques so that environment-induced mechanisms are under control and can be tuned at will. Moreover, atoms act as ideal pointlike scatterers, where the scattering tensor can be fully described with *ab initio* calculations and no adjustable parameters. The presence of sharp resonances results in large scattering cross sections, easily tunable by a few orders of magnitude, and large associated time scales, making resonant scattering systems very different from nonresonant multiple-scattering systems studied so far. Moreover, if strong driving fields are used, atoms exhibit unusual scattering properties. First, the atomic susceptibility shows up a dependence on the local field intensity. This nonlinearity alters both scattering (nonlinear reduction of the scattering cross section) and propagation (generation of a nonlinear refractive index for the effective medium). Second, in addition to the usual elastic component, atoms radiate an inelastic spectrum component. At very strong fields, this results in the celebrated Mollow triplet [7,8]. This inelastic spectrum is a direct consequence of the vacuum-induced fluctuations of the driven atomic dipole. At high intensity, the phase–breaking time is in the order of $1/\Gamma$, (excited-state lifetime) [9] and can be controlled via the intensity and/or the detuning of the incoming wave. This should have deep implications in the quest for Anderson localization of light [10] by atoms, since the field of one localized photon may saturate the atoms in its vicinity.

One of the most simple and efficient tools to investigate phase coherent effects is the coherent backscattering effect (CBS) [11], which appears when a wave illuminates a multiple-scattering sample. The CBS signal is related to the Fourier transform of the configuration-averaged two-field correlation function (the mutual coherence) at two space-time points at the surface of the medium. Putting it simply, the mesoscopic sample acts like a random two-wave zero path length interferometer and reveals an enhanced diffuse reflection peak in the backscattering direction. After spatial averaging, and for perfect interference, the enhancement factor α (the peak to background ratio) takes its maximum value of 2. It is deduced from a symmetry property bearing on reciprocity [12]. In recent experiments, CBS was studied in the elastic–scattering regime with cold atomic vapors exposed to low intensity quasiresonant monochromatic light. One has evidenced a loss of contrast due to the internal structure of atoms [13–15] and a full contrast restoration when nondegenerate atoms are used [16]. Theoretical studies investigating the impact of $\chi^{(2)}$ [17] and $\chi^{(3)}$ [18] nonlinearities predict no CBS reduction. This seems to be supported by experimental work on CBS in gain medium [19]. In contrast, phase fluctuations during scattering are potential sources of a loss of coherence and of a CBS contrast reduction. These phenomena appear to be effective as soon as the phase-breaking time is shorter than the wave transport time inside the medium. With resonant scatterers like atoms, the transport time can be very long [20], of the same order, or even longer than the correlation time of vacuum-induced dipole fluctuations. One may thus expect the same kind of decoherence mechanisms with intense electromagnetic radiation in cold gases than for electrons in condensed matter. However, with photons, there is an added complexity coming from the resonant character of the scattering process. This new ingredient will induce frequency filtering [21] and tunability in the strength and shape of the inelastic spectrum.

In this paper, we report experimental evidence of a reduced CBS contrast on an optically thick cold atomic strontium cloud when strong driving fields are used. The experi-

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mental setup has been described elsewhere [16]. Typical fluorescence measurements indicate that about 7 x10^7 atoms, at a temperature in the 1 mK range, are trapped in a quasi-Gaussian spherical cloud with a rms. size of about 0.7 mm. This corresponds to a typical atomic density at the center of the cloud about $n \simeq 10^{10}$ atoms/cm^3. With these parameters $k\ell \approx 10^4$ (k is the incoming wave vector and ℓ the light-scattering mean–free path) and scattering occurs in the weak localization regime. The maximum optical thickness achieved in our system, as deduced from coherent transmission measurements at low input intensity, is $b=3.5$, in reasonable agreement with the cloud size and number of atoms.

The CBS experiment procedure uses the following time sequence. First, the magneto-optical trap (MOT) is loaded during 28 ms (93% of the duty cycle). Then, the trapping beams and the magnetic gradient are switched off (typical falling time 1 μs for the lasers and 100 μs for the magnetic field). The residual magnetic field is less than 1 G, making the Zeeman splitting small compared to the linewidth Γ. Once the MOT is turned off, a resonant probe laser is switched on for a short period of time. In the present study, the probe laser parameters (intensity and frequency) are varied. The probe pulse duration is adjusted accordingly (typically from 5 to 70 μs) to keep the maximum number of absorbed photons per atom below 400. In this way, mechanical effects will be negligible throughout the experiments since $400kv_{\text{rec}}/\Gamma \approx 0.3$, where is the atomic recoil velocity associated with the absorption of a single photon. Finally, most of the atoms are recaptured during the next MOT sequence. The collimated CBS probe laser (beam waist 2 mm) and the response function of our detection system yield an angular resolution well described by a Gaussian convolution with a width ≈0.06 mrad, sufficiently below the typical CBS angular width (0.3 mrad). Wave plates and polarizing optical components are used to select the polarization of the incident probe beam and of the detected backward fluorescence. All measurements presented in this paper have been performed in the helicity preserving channel ($h\|h$). In this channel single scattering is rejected and an enhancement factor of 2 is predicted at low light intensity. However, since the channel isolation is not perfect in the experiment, single scattering will give an extra and unwanted incoherent contribution to the signal. This happens preferentially at low optical thickness because single scattering has the largest contribution to the total backscattered signal. For our experiments this effect leads to an enhancement factor reduction of a few percent.

The far-field backward fluorescence signal is collected on a cooled CCD camera. A mechanical chopper is placed between the MOT and the CCD. It is synchronized with the full time sequence in order to close the detection path when the MOT is operating and to open it when the probe beam is switched on. The total exposure time required for good signal-to-noise ratio is of the order of a few seconds. Once the full signal is collected, the acquisition is repeated during the same amount of time, maintaining the MOT magnetic gradient off, to obtain the background signal. This stray signal, corresponding to 15% of the total signal, is then subtracted to get the CBS signal. A two–dimensional fitting procedure is then used to analyzed the data. The theoretical shape of the CBS cone implemented in the fitting procedure is given by a Monte Carlo simulation performed at low saturation but taking into account the Gaussian distribution of atoms in a cloud [16]. Increasing the probe beam intensity did not reveal any significant change in the shape of the CBS cone, at least in the range of parameters used in our experiment. This is the reason why we treat all the data with the same cone shape, keeping the CBS width and the enhancement factor as free fitting parameters. The finite angular resolution of our apparatus has been taken into account by convolving the preceding theoretical CBS cone shape by a Gaussian having the measured apparatus angular width.

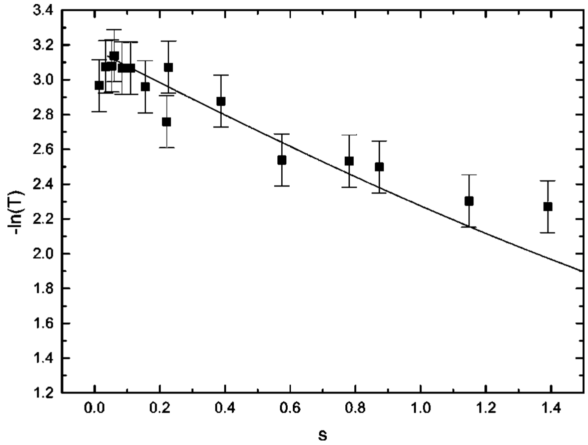

FIG. 1. Resonant ($\delta=0$) coherent transmission T along a diameter of the cold strontium cloud as a function of the saturation parameter s. The solid line corresponds to the theoretical Lambert-Beer prediction taking into account the nonlinear reduction of the scattering cross section. There is good agreement with the experimental data up to $s=1.2$.

Beyond the complexity of the situation under consideration (multiple scattering with nonlinear and inelastic scatterers), one also has to deal with nonuniform scattering properties. Indeed, even in a homogeneous slab geometry, the local intensity is not constant, as the incident coherent beam is attenuated when penetrating into the medium. Hence the atoms located deeper inside the medium will not be saturated in the same way as the atoms on the front part of the sample. Thus the saturation, and hence the scattering cross section, will not be constant along a given multiple-scattering path. The importance of the spatial variation of the saturation parameter can be estimated by looking at the attenuation of the coherent beam. In Fig. 1 we report the measured transmission and compare it with the Lambert-Beer theoretical prediction, taking into account the nonlinear reduction of the cross section. If one assumes that the local atomic saturation is dominated by the incident field and not by the scattered field, this theoretical curve is obtained by solving the following equation:

$$\frac{ds}{dz} = -\frac{s}{(1+s)\ell}. \tag{1}$$

Here the saturation parameter s is defined as

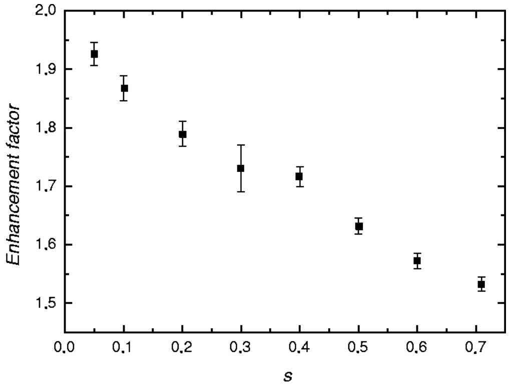

FIG. 2. Resonant ($\delta=0$) CBS enhancement factor as a function of the incident saturation parameter s. The coherent transmission value is kept fixed to $T=0.085$.

$$s = \frac{I/I_{\text{sat}}}{1 + (2\delta/\Gamma)^2}, \tag{2}$$

where I_{sat} is the saturation intensity ($I_{\text{sat}}=42\ \text{mW/cm}^2$ for Sr) and where δ is the laser detuning with respect to the atomic transition. The factor $1/(1+s)$ features the nonlinear reduction of the scattering efficiency and one gets the normal Lambert-Beer law when $s\to 0$. The low-intensity scattering mean–free-path ℓ reads

$$\ell(\delta) = \frac{1}{n\sigma(\delta)} = \frac{1 + (2\delta/\Gamma)^2}{n\sigma_0}, \tag{3}$$

with the resonant low-intensity scattering cross section $\sigma_0 = 3\lambda^2/2\pi$. The good agreement with the measured attenuation proves that saturation plays a role in our experimental conditions (since otherwise the transmission would not depend on s) and that the local atomic saturation is indeed dominated by the incident field.

Figure 2 shows the dependence of the CBS enhancement factor as a function of the incident saturation parameter s, with the probe maintained at resonance ($\delta=0$). In principle, one would like to vary the saturation parameter without modifying the relative weight of the various scattering orders involved in the CBS signal. This, however, proves to be difficult to assure because of the modification of the atom scattering properties when s increases. In order to minimize any effect relating to a modification of the distribution of scattering orders, we tried to keep the coherent beam profile throughout the sample as constant as possible. This is achieved by adjusting, for each value of s, the total number of cold atoms in the cloud in order to maintain the coherent transmission T as constant as possible. As shown in Fig. 2, we observe an enhancement factor of 1.93 ± 0.02 at low saturation. The small systematic reduction of α compared to the expected value of 2 is in agreement with the presence of residual single scattering in the *forbidden* $h\|h$ polarization channel, as previously discussed. The most striking feature is the rapid quasilinear decrease of the enhancement factor as s is increased. The slope derived from a rms. procedure is $(\delta\alpha/\delta s)\approx-0.6$. As the transmission is kept fixed, we estimate numerically the fraction of single scattering to increase by less than 10% when the saturation parameter is increased to $s=0.8$. The associated reduction of the enhancement factor should be of the order of 1%, negligible compared to the observed reduction. Thus the CBS reduction comes from the multiple-scattering signal.

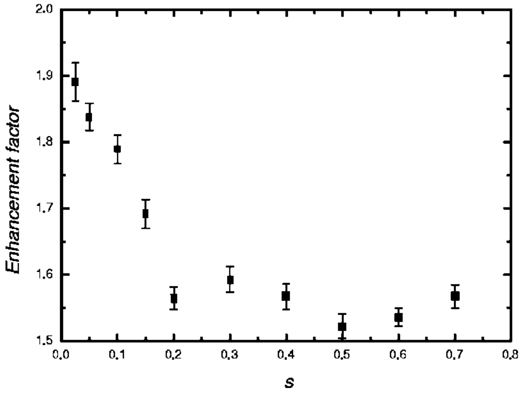

FIG. 3. Off-resonant ($\delta=\Gamma/2$) CBS enhancement factor as a function of the incident saturation parameter s. The coherent transmission value is kept fixed to $T=0.19$. Compared to the resonant case, the overall behavior is different with a stronger decrease at low s.

In order to see to what extent the resonance affects the coherence properties probed by CBS, we performed another experiment at $\delta=\Gamma/2$. The same experimental procedure has been used with a transmission now at $T=0.19$. As shown in Fig. 3, a different general behavior is observed. First, at low intensity, the linear decreasing is faster since $(\delta\alpha/\delta s)\approx-1.8$. Second, for larger saturation parameters ($0.3<s<0.8$) the decrease is then slowed down. The two sets of data in Figs. 2 and 3 are obtained with a different transmission value, but other studies show that the enhancement factor does not sensitively depend on the transmission value [22]. So, if we compare these data, it shows that s is not the only relevant parameter in our experiment. Indeed, the exact shape of the inelastic spectrum also depends on the detuning δ. In particular, for the detuned case, part of the inelastic spectrum will overlap the atomic resonance. This resonant inelastic light will thus be scattered again more efficiently then the off-resonant elastic part. This effect is, for example, responsible for an increase of the MOT volume in the multiple-scattering regime [23]. Finally in our experiment, the ratio of inelastic versus elastic multiple-scattered light may change with the detuning. We may then conclude that the CBS reduction is due to the inelastic spectrum. Preliminary calculations on simple toy models indicate that this is the case [24]. But one has also to keep in mind that the dispersive aspect of the atomic response to a driving field implies that nonlinearities at propagation and at scattering are drastically different for on-resonant and detuned excitations.

In summary, we observed that the CBS interference is strongly reduced when the atomic transition is saturated. This is a signature of the wave coherence loss associated with the shortening of the phase-breaking time compared to the transport time. The main origin of this reduction is probably related to the atomic dipole fluctuations induced by the vacuum field. The detuning dependence of the CBS enhancement factor (see Figs. 2 and 3) indicates that the saturation parameter s alone does not allow for a universal scaling. It is important to realize that, in the quest for strong localization of light in a disordered medium, large local buildup of the intensity can occur in the localized states. If the localization length is of the order of a few optical wavelengths, a single resonant photon could in principle saturate the atoms located in that region, in analogy with cavity QED effects [25]. The observations reported in this paper are thus important for the study of strong localization of light in atomic vapors.

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[22] Roughly speaking, inelastic events occur where the saturation is high, i.e., in the first atomic layers. Deeper inside the medium, where saturation is low, elastic events dominate. Thus, varying the coherent transmission in the multiple-scattering regime should not sensitively modify the inelastic mechanisms at work (and in turn the enhancement factor), at least in the conditions of the experiment.

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