Saturation induced coherence loss in coherent backscattering of light

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(Dated: September 23, 2003)

We use coherent backscattering (CBS) of light by cold Strontium atoms to study the mutual coherence of light waves 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. In our experiment, we observe a strongly reduced enhancement factor of the coherent backscattering cone when the intensity of the probe laser is increased, indicating a partial loss of coherence in multiple scattering.

PACS numbers: 42.25.Fx, 32.80.Pj

Wave coherence in the multiple scattering regime is a key ingredient to reveal the impact of interference on wave transport in strongly scattering media, with special interest in photonic crystals [1], random lasers [2], strong and weak localization [3, 4]. It has been shown that wave coherence has some robust features which survive the spatial configuration average. A clean illustration is given by the coherent backscattering phenomenon (CBS)[5, 6], a random *two-wave* zero path length interferometer. CBS manifests itself as an enhanced diffuse reflection peak in the backscattering direction. This 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 [7]. Hence the enhancement factor α (the peak to background ratio) provides a simple measure of spatial coherence properties of the disordered system after spatial averaging. For perfect contrast, α takes its maximal value of two, a symmetry property bearing on reciprocity [8]. In recent experiments, we have studied CBS in the elastic scattering regime with cold atomic vapours exposed to low intensity quasi-resonant monochromatic light. We have evidenced a loss of contrast due to the internal structure of atoms [9, 10, 11] and a full contrast restoration when non-degenerate atoms are used [12].

Studying optical wave transport and localization effects with cold atoms offer several advantages. Indeed, they act as ideal point-like scatterers, where the scattering matrix can be fully described with *ab initio* calculations and no adjustable parameters. Moreover, the presence of sharp resonances results in large scattering cross-sections, easily tunable by few orders of magnitude, and large associated time scales, making resonant scattering systems different from non-resonant multiple scattering studied so far. 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 non linearity 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. For resonantly driven atoms with a non-degenerate groundstate, a characteristic frequency width of this spectrum is Γ (inverse of the excited-state lifetime) [13]. This inelastic spectrum is a direct consequence of the vacuum-induced fluctuations of the driven atomic dipole. At very strong fields, this results in the celebrated Mollow triplet [13, 14]. An important question is whether the interference effects surviving the spatial average are affected by these non linearities and by these quantum fluctuations of the atomic dipole. Theoretical studies investigating the impact of $\chi^{(2)}$ [15] and $\chi^{(3)}$ [16] nonlinearities predict no CBS reduction. This seems to be supported by experimental work on CBS in gain medium [17]. On contrary, phase fluctuations during scattering are potential sources of a loss of coherence and a CBS contrast reduction. These *dephasing* phenomena, well known in electron transport [18], appear to be effective as soon as their correlation times are shorter than the wave transport time inside the medium. With resonant scatterers like atoms, the transport time can be very long [19], of the same order or even longer than the correlation time $\tau \simeq 1/\Gamma$ of vaccuum-induced dipole fluctuations. One may thus expect the same kind of decoherence mechanisms with atoms than for electrons with an added complexity coming from the resonant character of the scattering process. This new ingredient will induce frequency filtering [20] and tunability in strength and shape of the inelastic spectrum.

In this letter, we report the first experimental evidence of a reduced CBS contrast on an optically thick cold atomic strontium cloud when strong driving fields are used. The experimental setup has been described elsewhere [12]. Typical fluorescence measurements indicate that about 7 10^7 atoms, at a temperature of $1mK$, are trapped in a quasi-Gaussian spherical cloud with rms-size about $0.7mm$. 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 wavevector and ℓ the light scattering mean free path) and scattering occurs in the weak localization regime. The maximal 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 MOT is loaded during $28ms$ (93% of the duty cycle). Then the trapping beams and the magnetic gradient are switched off (typical falling time $1\mu s$ for the lasers and $100\mu s$ for the magnetic field). The residual magnetic field is less than $1G$ 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 $70\mu s$) to keep the maximum number of absorbed photons per atom below 400. In this way, mechanical effects will be negligible throughout the experiments since $400kv_{rec}/\Gamma \approx 0.3$, where v_{rec} 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 $2mm$) and the response function of our detection system yield an angular resolution well described by a Gaussian convolution with a width $\approx 0.06mrad$, sufficiently below the typical CBS angular width ($0.3mrad$). 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 two is predicted and observed at low light intensity [12]. However, since the channel isolation is not perfect in the experiment, single scattering will spoil 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 few percents.

The principle of the CBS detection scheme has been described in ref. [21]. 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 2D fitting procedure is then used to extract the main CBS cone parameters, its width and its enhancement factor. 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 [12]. 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. 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.

Beyond the complexity of the situation under consideration (multiple scattering with nonlinear and inelastic scatterers), one has to deal also with nonuniform scattering properties. Indeed, even in an 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 we compare it with the Lambert-Beer theoretical prediction taking into account the non-linear 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 s is the saturation parameter defined as:

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

where I_{sat} is the saturation intensity ($I_{sat} = 42mW/cm^2$ for Sr) and where δ is the laser detuning with respect to the atomic transition. The factor $1/(1+s)$ features the non-linear reduction of the scattering efficiency and one gets the normal Lambert-Beer law when $s \rightarrow 0$. 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 excellent agreement with the measured attenuation prooves 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.

Fig. 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 ($T \simeq 0.085$ in the data shown in Fig. 2). 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 two is in agreement with the presence of residual single scattering in the *forbidden* $h \| h$ polarization channel, as we previously discussed. The most striking feature is the rapid quasi-linear 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 up 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.

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 Fig.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. 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 *e.g.* responsible for an increase of the MOT volume in the multiple scattering regime [22]. Finally in our experiment, the ratio 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, but one has also to keep in mind that the dispersive aspect of the atomic response to a driving field implies that non linearities at propagation and at scattering are drastically different for on-resonant and detuned excitations.

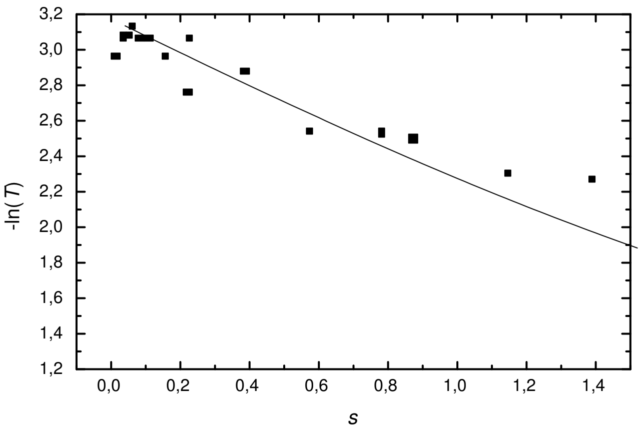

FIG. 1: Resonant ($\delta = 0$) coherent transmission T along a diameter of the cold strontium could as a function of the saturation parameter s. The squares are the experimental values and the solid line correspond to the theoretical Lambert-Beer prediction taking into account the nonlinear reduction of the scattering cross-section.

In summary, we observed that coherent backscattering is strongly reduced when the atomic transition is saturated, a signature of the wave coherence loss in the multiple scattering regime. We speculate that the origin of this reduction is due to the inelastic scattering of the light by the atoms, *i.e.* from the coupling of the atomic dipole to the vacuum modes of the field rather than to the non-linear response of the atomic dipole. The different behavior of the CBS enhancement factor for resonant and off-resonant excitations (see Fig. 2 and 3) indicates that the saturation parameter s alone does not allow for an universal scaling. It is important to realize that, in the quest of strong localization of light in disordered medium, large local build-up of the intensity can occur in the localized states. If the localization length is of the order of few optical wavelength, a single resonant photon could in principle saturate the atoms located in that region, in analogy with cavity QED effects [23]. The observations reported in this letter are thus important for the study of strong localization of light in atomic vapours.

The authors thank D. Delande and G. Labeyrie for fruitful discussions. This research is financially supported by the CNRS, the PACA region and the BNM.

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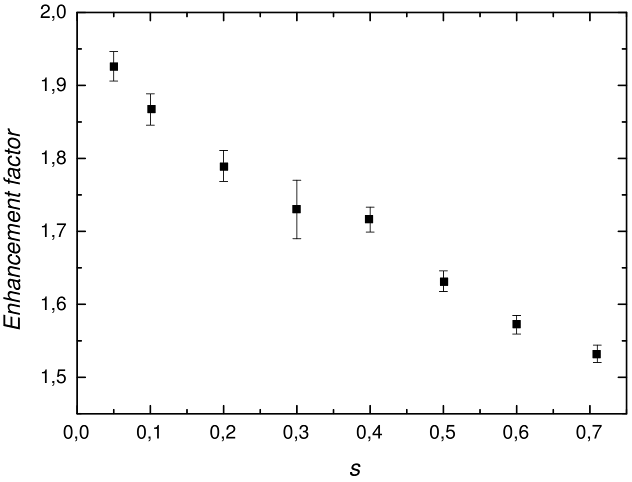

FIG. 2: Resonant ($\delta = 0$) CBS enhancement factor as a function of the incident saturation parameter s. The coherent transmission value is kept fixed to $T = 0.085$.

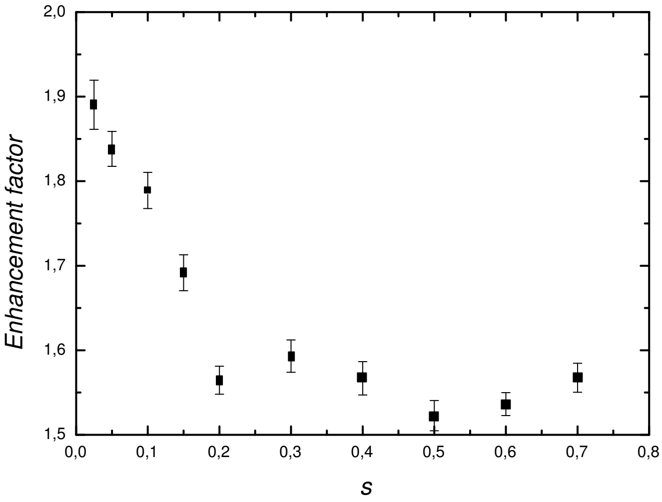

FIG. 3: Off-resonant ($\delta = \Gamma/2$) CBS enhancement factor as a function of the incident saturation parameter s. The coherent transmission value is kept fixed to $T = 0.19$. Compared to the resonant case, the overall behavior is different with a stronger dicrease at low s.

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