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Correlation and dynamic effects in coherent backscattering of light by optically dense ensembles of cold atoms

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Abstract. **Coherent backscattering of light by optically dense atomic ensembles is considered. The spectrum of scattered radiation is studied. The dynamics of the total intensity and enhancement factor is considered in the case of scattering of pulsed radiation. The spectral and temporal characteristics of coherent backscattering are analysed depending on the observation conditions. As an example, calculations are performed for an ensemble of ^{85}Rb atoms in a magnetooptical trap. It is shown that analysis of the correlation and dynamic properties of scattered radiation makes it possible to separate contributions from different orders of scattering and thereby to study the process of radiation trapping in dense media in more detail.**

Keywords: *coherent backscattering, cold atoms, enhancement factor*.

1. Introduction

Atomic ensembles of cold and ultracold atoms, in particular, Bose – Einstein condensates featuring a number of unique physical properties have been studied in many recent experimental and theoretical papers. Aside from fundamental physical reasons, ultracold atomic systems and their interaction with light attract attention due to a wide scope of their possible applications, in particular, in the problems of quantum information, where light is used for information transfer, while an atomic ensemble is employed as a physical object for its storage.

In this paper, we discuss theoretically some interference phenomena taking place upon multiple scattering of light in atomic ensembles at sufficiently low temperatures in magnetooptical traps. It is known [1 – 4] that interference upon multiple scattering in disordered rarefied systems results in the so-called effect of coherent backscattering (CBS) (which is also called sometimes weak localisation of light). This effect is manifested in the form of a drastic anisotropic addition to the scattering cross section, which is zero only within a narrow cone near the direction opposite to the wave vector of a plane wave incident on a medium. CBS is usually caused by the constructive interference of two waves, one of which is formed due to multiple successive scattering from a chain of atoms (inhomogeneities), and another – due to scattering from the same chain, but upon the reverse sequence of scattering events.

Many aspects of CBS of light by ensembles of cold atoms in magnetooptical traps were studied in detail both experimentally and theoretically in recent papers [5 – 8]. In particular, the polarisation and angular dependences of the scattering cross section (backscattering cone) and its dependence on the laser radiation frequency were thoroughly analysed.

The difficulties involved in the theoretical interpretation of CBS experiments appear because the characteristics of light scattered in an optically dense medium are determined by the partial contributions of radiation that experienced a different number of single-scattering events. In this paper, we proposed and analysed two variants of the improvement of the method for CBS observation, which allow a partial separation of scattering of different orders. The first variant is based on the analysis of the spectrum of scattered light and the study of its difference from the spectrum of the incident radiation. We showed that the Doppler effect causes the difference between the spectra of light scattered in different orders, which can be found in experiments. The second variant is based on the possible separation of these contributions during the observation of emission of an atomic cloud after shutoff of a probe light source. Photons, which have experienced a different number of scattering events inside a dense medium, have different delay times, thereby making contributions at different afterglow times.

2. CBS spectrum

The spectral characteristics of coherent radiation backscattered by ensembles of cold atoms are usually studied by analysing the dependence of the integrated intensity of scattered light on the incident light frequency, the probe radiation being assumed monochromatic [5 – 8]. The enhancement factor, which is the basic quantitative characteristic of CBS (integrated enhancement factor), was determined in papers [5 – 8] as the ratio of the integrated total intensity of light to the integrated intensity of the incoherent component (i.e., the ratio of the amplitude of the backscattering cone to that of the quasi-isotropic component of scattered light). At the same time, it is clear that analysis of the enhancement factor for a separated spectral component of scattered light characterises CBS in

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more detail and can give additional information both on the properties of a scattering medium and internal characteristics of multiple scattering. We will define the ratio of the spectral density of backscattered light to the spectral density of its non-interference component as the spectral enhancement factor.

The difference between the integrated and spectral enhancement factors is caused by the change in the spectrum of radiation upon scattering. If an elementary scattering event is the Rayleigh or elastic Raman process during which the photon frequency does not change (an atom undergoes transition to the same or another Zeeman level having the same energy) and the spectrum of the incident light is sufficiently narrow, then the spectrum changes mostly due to the Doppler frequency shift after scattering of light by moving atoms. The atomic cloud in a trap is cooled down to $T \sim 50$ μK. However, after the shutoff of cooling lasers this temperature can increase for different reasons up to ~ 50 mK. In this case, the characteristic Doppler shifts do not exceed one – two natural widths of the level, which severely complicates a direct spectral analysis of scattered light because it is necessary to resolve very narrow spectral lines in the radio-frequency range. This problem can be solved by the methods of light-beam spectroscopy (LBS), whose possibilities were earlier demonstrated by studying the velocity distributions of atoms and molecules in liquids and gases [9]. Note also that the related methods of correlation spectroscopy were earlier successfully used to analyse the properties of atomic ensembles in magneto-optical traps at low temperatures [10 – 13]. In [10 – 13], the methods of homo- and heterodyning were applied to study the scattering spectra of laser radiation forming a trap, which provided a non-destructive control of its parameters, for example, temperature and also allowed one to obtain some information on forces acting on atoms in the trap.

Unlike papers [10 – 13], we assume that the laser radiation forming a trap is switched off, and our aim will be the description of CBS of probe radiation. We will perform the spectral analysis of different scattering orders forming both the non-interference (quasi-isotropic) and interference (anisotropic) components of backward scattering. The most natural practical scheme is the LBS method based on the heterodyne detection. In this case, the spectrum of photocurrent intensity fluctuations coincides in shape with the spectrum of the scattered field [14]. The difference between these spectra is caused only by the shift of the emission spectrum to the radio-frequency region due to heterodyne detection and by the appearance of the spectrally independent shot-noise addition. This allows us in fact to calculate directly the intensity spectrum of the scattered light. This spectrum is determined by the spectral expansion of the correlation function $D^{(E)}_{\mu_1\mu_2}(\boldsymbol{r}_1, t_1; \boldsymbol{r}_2, t_2)$ expressed in terms of the negative-frequency $[E^-_\mu(\boldsymbol{r}, t)]$ and positive-frequency $[E^+_\mu(\boldsymbol{r}, t)]$ components of the Heisenberg operators of the electric field strength (μ is the polarisation index):

$$D^{(E)}_{\mu_1\mu_2}(\boldsymbol{r}_1, t_1; \boldsymbol{r}_2, t_2) = \langle E^-_{\mu_1}(\boldsymbol{r}_1, t_1) E^+_{\mu_2}(\boldsymbol{r}_2, t_2)\rangle. \quad (1)$$

This correlation function can be calculated based on the general theory of multiple scattering of light by atoms (see, for example, [5 – 7]). It is convenient to use the diagram technique of Konstantinov – Perel – Keldysh for nonequilibrium systems to construct the perturbation theory expansions [15 – 18]. By omitting the details of calculations, we consider the main results.

We calculated the spectrum of scattered light for a spherically symmetric Gaussian distribution of the concentration $n(\boldsymbol{r})$ of atoms in a cloud of radius $r_0 = 1$ mm and the concentration of atoms at its centre $n_0 = 3.2 \times 10^{10}$ cm^{-3}:

$$n(\boldsymbol{r}) = n_0 \exp\left(-\frac{r^2}{2r_0^2}\right). \quad (2)$$

The velocity distribution of atoms was assumed Maxwellian. The most probable velocity of atoms in a trap $v_0 = (2T/m)^{1/2}$ was determined by the condition $kv_0 = \gamma$, where k is the modulus of the wave vector of probe radiation and γ is the natural width of the excited state of the atom. Note that for typical experimental parameters (see, for example, [5]), the dynamic heating of the atomic cloud up to the considered temperatures occurs for a few milliseconds. The time required to record a backscattered signal is substantially shorter. Thus, the stationary scattering regime in the atomic clouds was established for the time of the order of $5\gamma^{-1} \sim 150$ ns (see below). This means that we can assume that the cloud temperature is invariable during spectral measurements and consider stationary scattering.

The polarisation scheme, for which the calculation was performed, corresponds to the analysis of scattering of linearly polirised light in the polarisation-preserving channel (L‖L). The spectrum of the input probe radiation was assumed monochromatic and its carrier frequency ω varied within several natural widths ($\gamma \approx 6$ MHz) in the vicinity of the resonance hyperfine $F = 3 \to F' = 4$ transition of the D_2 line of the ^{85}Rb atom. The shape of the backscattered cone was not analysed and only backward scattering was considered.

Figure 1 shows the partial contributions to the spectrum of the non-interference component of the first three scattering orders. These dependences are constructed for the case when the incident radiation is shifted to the blue wing of the $F = 3 \to F' = 4$ transition by $\delta\omega = \omega - \omega_0 = +\gamma = +kv_0$, where ω_0 is the atomic transition frequency. However, it follows from these dependences that scattered radiation is predominantly characterised, on the contrary, by negative detunings $\Delta' = \omega' - \omega_0$ from the atomic resonance, i.e., its frequency falls into the red wing of the line. The line shape is

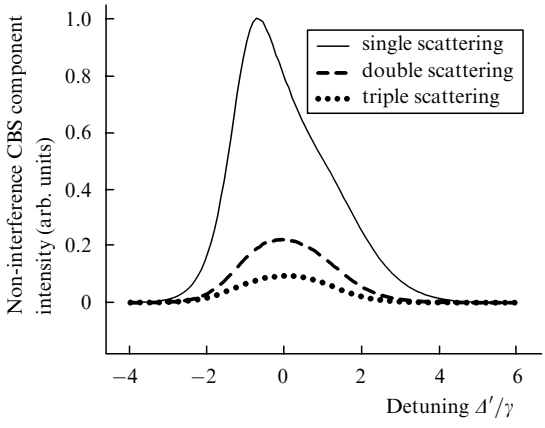

Figure 1. Spectral intensity profiles of the non-interference CBS component corresponding to the first three scattering orders. Atoms are irradiated by monochromatic light at the frequency $\omega = \omega_0 + \gamma$.

as if 'reflected' with respect to the resonance transition frequency. This circumstance is caused by the fact that the atom whose velocity v satisfies the condition $\omega - \boldsymbol{kv} = \omega_0$ has the maximum probability to scatter a photon, but upon backward scattering this atom emits a photon at a doubly shifted frequency $\omega' = \omega - 2\boldsymbol{kv}$. In the case of multiple scattering, such a shift is also present, although it is manifested to a lesser extent. Even for triply scattered light, the maximum of the spectral density corresponds to zero detunings, which corresponds to the average Doppler shift of the order of γ. A decrease in the shift and pulling of the line frequency to the region of the atomic transition frequency is caused by the isotropic scattering of light by extended atomic chains. Upon summation of contributions from all the scattering orders existing in the problem under study, the effect is preserved. The corresponding spectral dependences for different detunings $\delta\omega$ ae presented in Fig. 2a.

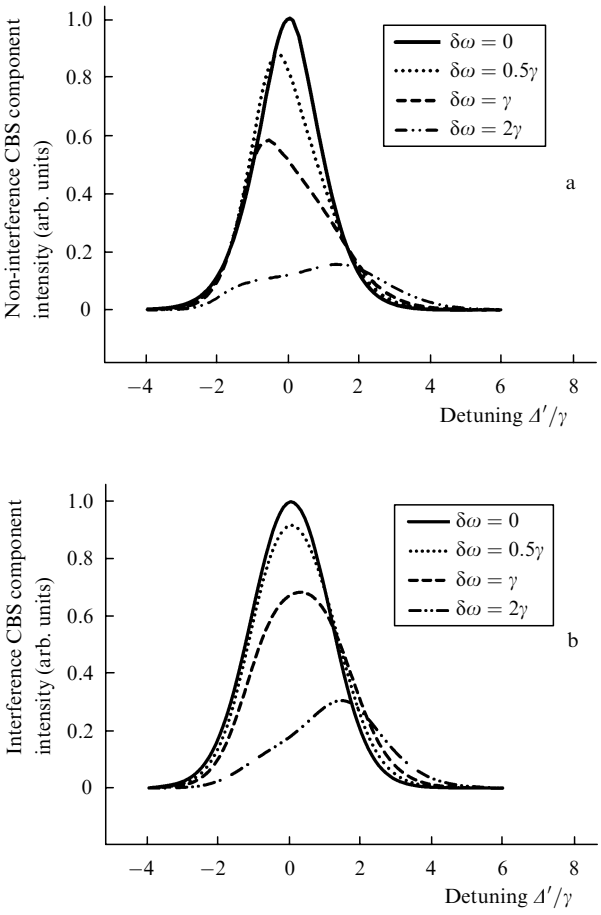

Figure 2. Spectra of the non-interference (a) and interference (b) CBS components for different probe light detunings $\delta\omega$.

Figure 2b shows the spectrum of the interference component of CBS. The Doppler shift of the interference component is substantially smaller than that of the non-interference component. This result appears natural mainly due to the absence of a large and strongly shifted first-order contribution and also because scattering by moving atoms leading to the frequency shift is accompanied by phase shifts which additionally suppress the interference component. If the detuning $\delta\omega$ of incident radiation is selected substantially larger than the Doppler width, the shift will be smaller due to a rapid decrease in the number of scattering atoms moving at velocities satisfying the resonance condition $\omega - \boldsymbol{kv} = \omega_0$.

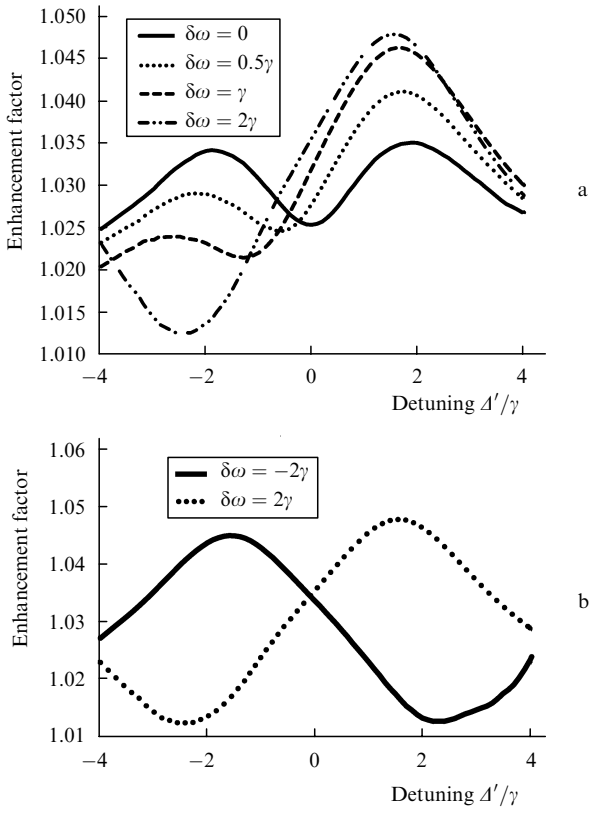

Figure 3. Spectral enhancement factor for different probe light detunings $\delta\omega$.

Figure 3 shows the calculated spectral enhancement factors. The curves presented in this figure have a number of peculiarities. When probe radiation is tuned exactly in resonance with the $F = 3 \rightarrow F' = 4$ transition, i.e. when $\delta\omega = 0$, an almost symmetric dependence with two distinct maxima is observed. The maxima are observed because the spectrum of singly scattered light is somewhat narrower than the spectrum of doubly scattered light, which results in the increasing role of multiple scattering in the formation of the spectral wings, thereby enhancing the enhancement factor.

If the probe frequency is detuned from the resonance, then along with the discrepancy between the widths of the spectra of a ladder and interference component, the above-mentioned Doppler shift is also observed. This results in the shift of the minimum of the enhancement factor to the region where the maximum of the non-interference component is located. In this case, two maxima are also observed, although one of them is substantially weaker (for detuning $\delta\omega = 2/\gamma$, it is located outside the region in Fig. 3). The spectral enhancement factor at the main maximum is substantially higher than the integrated enhancement factor at the same temperature. For comparison, we present in Fig. 3a the data corresponding to different detunings $\delta\omega$ of probe radiation. For detunings of the same magnitude but with opposite signs, the curves are almost symmetric (Fig. 3b). A small asymmetry is caused by the influence of nonresonance transitions.

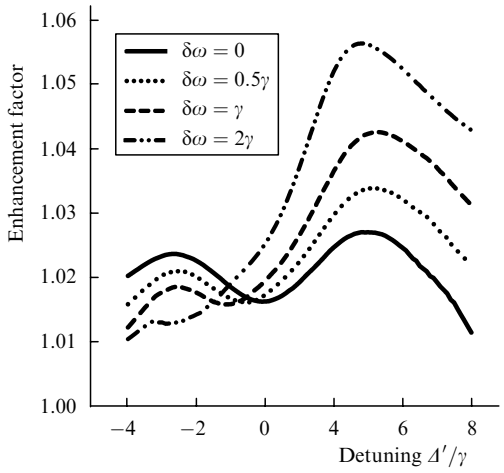

Figure 4. Spectral enhancement factor for different probe light detunings. The H||H polarisation channel.

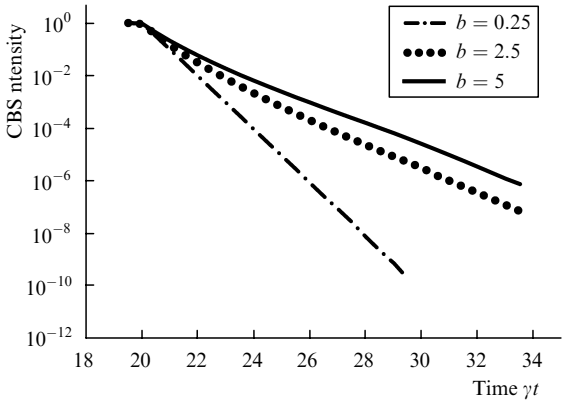

Figure 5. Time dependences of the total CBS intensity upon pulsed excitation for different optical densities b. The L||L polarisation channel, the pulse duration is $20\gamma^{-1}$.

The data presented above correspond to the L||L polarisation channel. For other polarisation channels, the dependences are qualitatively the same. However, some quantitative difference takes place for the scheme with preserving radiation spirality (H||H) due to a relatively small role of single scattering (Fig. 4).

3. Pulsed CBS

The majority of the papers on scattering of light by atoms in magnetooptical traps is devoted to analysis of the stationary scattering regime. At the same time, transient processes and emission of atoms caused by the radiation trapping upon multiple scattering are of interest from several points of view. First, the radiation trapping substantially restricts the cooling conditions of atoms [19, 20]. Second, the diffusion of photons in an optically dense medium of cold atoms substantially differs from well-known radiation transfer in the case of strong Doppler broadening [21–23]. This diffusion also differs from diffusion of light in a medium of classical scatterers. Thus, experiments performed by now show that the light diffusion velocity in a dense air mixture of TiO_2 particles with the average diameter 220 nm is an order of magnitude lower than the speed of light in vacuum [24]. For atoms cooled in magnetooptical traps, this velocity is lower than the speed of light already by four–five orders of magnitude [25].

The general CBS theory developed in [5–7] can be also used in the case of pulsed probe radiation. We calculated the interference and non-interference backscattering components for a rectangular pulse. The calculation was performed for a spherically symmetric atomic ensemble with Gaussian distribution (2) with the radius $r_0 =$ 1 mm. The atoms were assumed immobile during the action of a light pulse. Figure 5 shows the time dependences of the backscattered radiation intensity calculated for atomic clouds of different optical densities. One can see that within some time after the pulse end, the radiation decay is described by the nearly exponential law. The decay rate is substantially lower than that for a single atom, which is the manifestation of the trapping effect. The data in Fig. 5 are presented for one possible L||L polarisation channel. Our calculations showed that the radiation decay rates are the same for all polarisation components. The decay rates

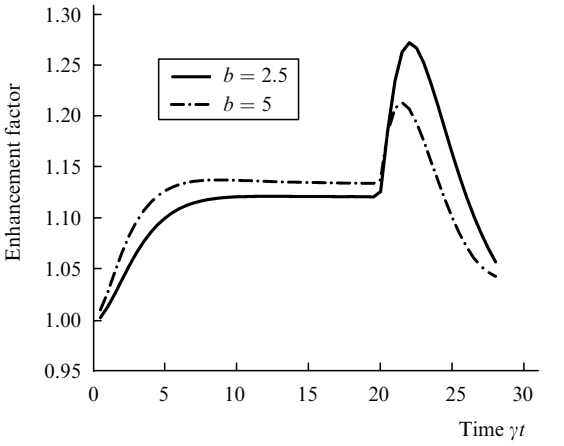

Figure 6. Time dependences of the enhancement factor for different optical densities b. The L||L polarisation channel, the pulse duration is $20\gamma^{-1}$.

for the interference and non-interference components are also coincident. This result appears reasonable because the time delays are determined in fact by the decay time of the excited-state population of atoms in an ensemble.

Figure 6 shows the temporal dynamics of the enhancement factor for two optical thicknesses of the atomic ensemble for the L||L polarisation channel. Note that the enhancement factor increases immediately after the end of the exciting pulse. This is caused by a substantially more rapid decay of single scattering, which provides the better conditions for CBS. Therefore, the radiation trapping by an atomic ensemble is manifested in a nontrivial way in the temporal dynamics of the enhancement factor. The relative role of different scattering orders changes with time. For high scattering orders, the relative number of constructively interfering channels tends to decrease, which is reflected in a decrease in the enhancement factor at long decay times (Fig. 6).

4. Conclusions

We have considered the spectral characteristics and dynamics of CBS. It has been shown that two possible approaches exist for partial separating scattering orders both for the interference and non-interference CBS

components. We hope that a combination of theoretical calculations with experimental data can provide important spectroscopic information on the state of an atomic ensemble in the case of radiation trapping. In particular, the difference between the spectra of the interference and non-interference components makes it possible to find experimental conditions under which the CBS enhancement can be observed even for an ensemble of atoms heated to temperature at which $kv_0 > \gamma$. At the same time, the thermal motion of atoms suppressing the interference component results in the spectral separation of different orders of scattering, which can be used in experiments.

We have analysed the properties of the CBS formation dynamics for pulsed radiation. The temporal parameters of multiply scattered delayed radiation have been calculated. It has been shown that due to diffusion of multiply scattered light, the radiation intensity decays in a complicated way, and the enhancement factor substantially exceeds its stationary value during a certain time. The properties found in the study can be used for separating partial contributions of individual scattering orders.

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