

Multiple Scattering of Light by Atoms in the Weak Localization Regime

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Coherent backscattering is a multiple scattering interference effect which enhances the diffuse reflection off a disordered sample in the backward direction. Classically, the enhanced intensity is twice the average background under well chosen experimental conditions. We show how the quantum internal structure of atomic scatterers leads to a significantly smaller enhancement. Theoretical results for double scattering in the weak localization regime are presented which confirm recent experimental observations.

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Standard cooling techniques permit one to prepare optically thick media of cold atoms, like Bose-Einstein condensates [1]. In a sufficiently dense disordered atomic medium, light is expected to be localized by interference, in analogy to Anderson localization of electrons in disordered solid-state samples [2]. For multiple scattering of light, atoms are usually modeled as dipole point scatterers, their internal structure being ignored. In this Letter, we show that even far from strong localization, i.e., in a dilute medium, such an approximation is severely too optimistic, and we demonstrate that the degeneracy of the atomic ground state significantly reduces interference effects.

When monochromatic light is elastically scattered off a disordered medium, the interference between all partial waves produces strong angular fluctuations of the intensity distribution known as a speckle pattern. In the weak localization regime (a dilute disordered medium such that $k\ell \gg 1$ where $k = 2\pi/\lambda$ is the light wave number and ℓ the elastic mean free path)—the case we consider in the following—the phases associated with different scattering paths are essentially uncorrelated. Averaging over the positions of the scatterers then washes out interferences and produces a smooth reflected intensity. There is, however, an exception: the ensemble average cannot wash out the interference between a wave traveling along a scattering path and the wave traveling along the reverse path (where the same scatterers are visited in reverse order). Indeed, in the backscattering direction, the optical lengths of the direct and reverse paths are equal. Thus the two waves interfere constructively and enhance the average diffuse reflected intensity. This phenomenon is known as coherent backscattering (CBS), a hallmark of interference effects in disordered systems [3,4].

The average intensity I scattered at angle θ can be written as a sum of three terms, $I(\theta) = I_S(\theta) + I_L(\theta) + I_C(\theta)$ [5]. Here I_S is the single scattering contribution (for which the direct and reverse paths coincide), I_L the raw contribution of multiple scattering paths (the so-called ladder terms), and I_C the CBS contribution (the so-called maximally crossed terms). I_L and I_S do not contain any interference term and thus vary smoothly with θ . The contribution to I_L of a pair of direct and reverse paths is es-

entially $|T_{\text{dir}}|^2 + |T_{\text{rev}}|^2$ where T_{dir} and T_{rev} are the corresponding scattering amplitudes. The contribution to I_C is $2|T_{\text{dir}}||T_{\text{rev}}|\cos\phi$, with $\phi = (\mathbf{k} + \mathbf{k}') \cdot (\mathbf{r}' - \mathbf{r})$, where \mathbf{k}, \mathbf{k}' are the incident and scattered wave vectors and \mathbf{r}, \mathbf{r}' the positions of the first and last scatterers along the path. It follows that I_C is always smaller or equal than I_L . For a small scattering angle θ , the phase difference ϕ is essentially $\theta k\ell$. Thus, $I_C(\theta)$ is peaked around the backscattering direction $\theta = 0$ and rapidly decreases to zero over an angular width $\Delta\theta \sim 1/k\ell \ll 1$ [6].

In usual experiments, the incident light is polarized either linearly or circularly (with a given helicity h) and one studies the scattered light with the same or orthogonal polarization in four polarization channels: $\text{lin} \parallel \text{lin}$, $\text{lin} \perp \text{lin}$, $h \parallel h$ (helicity is preserved), and $h \perp h$ (helicity is flipped). The ratio of the average intensity in the backward direction $I(\theta = 0)$ to the background $I(1/k\ell \ll \theta \ll 1) = I_S + I_L$ defines the enhancement factor in each polarization channel:

$$\alpha = 1 + \frac{I_C(0)}{I_S + I_L}. \quad (1)$$

As $I_C \leq I_L$, its largest possible value is 2, reached if and only if $I_S = 0$ and $I_C = I_L$. For classical scatterers and exact backscattering [7], the first condition $I_S = 0$ is fulfilled in the $\text{lin} \perp \text{lin}$ and $h \parallel h$ channels if the scatterers have spherical symmetry. The second condition $I_C = I_L$ is fulfilled in the $\text{lin} \parallel \text{lin}$ and $h \parallel h$ channels provided reciprocity holds. Reciprocity is a symmetry property valid whenever the fundamental microscopic description of the system is time reversal invariant [8]. It assures one that

$$T_{\text{dir}}(\mathbf{k}\boldsymbol{\epsilon} \rightarrow \mathbf{k}'\boldsymbol{\epsilon}') = T_{\text{rev}}(-\mathbf{k}'\boldsymbol{\epsilon}'^* \rightarrow -\mathbf{k}\boldsymbol{\epsilon}^*), \quad (2)$$

where $(\mathbf{k}, \boldsymbol{\epsilon})$ and $(\mathbf{k}', \boldsymbol{\epsilon}')$ are the incident and scattered wave vectors and polarizations (the star indicates complex conjugation). In the backscattering direction ($\mathbf{k}' = -\mathbf{k}$) and parallel polarization channels ($\boldsymbol{\epsilon}' = \boldsymbol{\epsilon}^*$), the scattering amplitudes of any couple of direct and reverse paths are thus identical, leading to $I_C = I_L$ [9]. As a consequence, the maximum enhancement $\alpha = 2$ is expected for spherical scatterers in the $h \parallel h$ channel, a prediction confirmed by experiment [10].

Recently, CBS of light was observed with cold atoms with surprisingly low enhancement factors, with the lowest value in the $h \parallel h$ channel [11,12]. The quantum internal structure of the atoms can account for a major part of this astonishing observation. Two different reasons must be distinguished: (i) all polarization channels now contain a single scattering contribution, and (ii) the amplitudes interfering for CBS are in general not reciprocal which leads to $I_C < I_L$. This Letter is devoted to elucidating these points and presents a calculation of the CBS signal with atoms in the double scattering regime.

We consider a collection of atoms at rest exposed to monochromatic light, quasiresonant with an electric dipole transition between some ground state with angular momentum J and some excited state with angular momentum J' . For sufficiently weak light intensity, a perturbative description is in order: an atom with initial state $|Jm\rangle = |m\rangle$ (where m is the magnetic quantum number ranging from $-J$ to J) undergoes a transition into a final state $|Jm'\rangle = |m'\rangle$ while scattering an incoming photon ($\mathbf{k}, \boldsymbol{\epsilon}$) into an outgoing mode ($\mathbf{k}', \boldsymbol{\epsilon}'$). We assume that no magnetic field is present, so that the atomic ground state is $(2J + 1)$ -fold degenerate. Energy conservation then implies that the scattering process is purely elastic ($\omega = \omega'$).

The single scattering transition amplitude T_S is proportional to the matrix element of the resonant scattering ten-

sor [13] $t_S(\boldsymbol{\epsilon}, m; \boldsymbol{\epsilon}', m') = \langle m' | (\boldsymbol{\epsilon}'^* \cdot \mathbf{d}) (\boldsymbol{\epsilon} \cdot \mathbf{d}) | m \rangle$, where \mathbf{d} is the atomic dipole operator connecting the J and J' subspaces. This amplitude describes the absorption ($\boldsymbol{\epsilon} \cdot \mathbf{d}$) of the incoming photon, and the emission ($\boldsymbol{\epsilon}'^* \cdot \mathbf{d}$) of the final photon. Now not only transitions to the same $m' = m$ substate are allowed (Rayleigh transitions), but also to $m' \neq m$ (degenerate Raman transitions). It is then in general no longer possible to eliminate the single scattering contribution by polarization analysis. For example, a signal in the $h \parallel h$ channel in the backscattering direction is associated by angular momentum conservation with a transition $|m' - m| = 2$ and can be suppressed only if $J = 0, 1/2$.

Of more fundamental interest is the second reason for the enhancement reduction, $I_C < I_L$, and the role of reciprocity. As a general rule in quantum mechanics, only transition amplitudes which connect the *same* initial state to the *same* final state can interfere. Here the states of the complete system are the photon modes *and* the internal states $\{|m\rangle = |m_1, m_2, \dots\rangle$ of all atoms. Again, CBS originates from the interference between two amplitudes T_{dir} and T_{rev} associated with the direct and reverse scattering sequences of the same transition $|\mathbf{k}\boldsymbol{\epsilon}, \{m\rangle \rightarrow |\mathbf{k}'\boldsymbol{\epsilon}', \{m'\rangle$. But in this case, reciprocity—although it perfectly holds—fails to predict the enhancement factor. Indeed, the reciprocity relation now reads [14]

$$T_{\text{dir}}(\mathbf{k}\boldsymbol{\epsilon}, \{m\rangle \rightarrow \mathbf{k}'\boldsymbol{\epsilon}', \{m'\rangle) = (-1)^{\sum_i (m'_i - m_i)} T_{\text{rev}}(-\mathbf{k}'\boldsymbol{\epsilon}'^*, -\{m'\rangle \rightarrow -\mathbf{k}\boldsymbol{\epsilon}^*, -\{m\rangle). \quad (3)$$

In the backscattering direction ($\mathbf{k}' = -\mathbf{k}$) and parallel polarization channels ($\boldsymbol{\epsilon}' = \boldsymbol{\epsilon}^*$) these two reciprocal amplitudes will interfere and thus contribute to CBS if and only if $\{m'\rangle = -\{m\rangle$. So, among all pairs of interfering amplitudes, only a few are linked by reciprocity. Unless this condition is fulfilled, the reciprocal amplitudes are associated with different initial and final states of the system and cannot interfere. This is similar to the case of orthogonal polarizations where the reciprocity principle is still valid, but simply inapplicable. In the case of atomic scatterers with internal structure, this is true for all polarization channels, and stands out in sharp contrast to the classical case. We point out, however, that the condition $\{m'\rangle = -\{m\rangle$ is trivially true in the case of an $J = 0 \rightarrow J' = 1$ transition since the ground state then has no internal structure and $m = m' = 0$. Therefore, the enhancement factors for this transition will be the same as for classical point scatterers.

No fundamental reason is left for $I_C = I_L$ to hold. Let us illustrate why one expects now $I_C < I_L$ in general. Consider double scattering on two atoms without change of their internal states ($m'_1 = m_1; m'_2 = m_2$), for $J = 1/2 \rightarrow J' = 1/2$ in the $h \parallel h$ channel with positive incident helicity. The atoms are supposed to be initially prepared in the $|m_1 = -1/2\rangle$ and $|m_2 = +1/2\rangle$ substates (quantization axis parallel to the incoming light wave vector). In this configuration (see Fig. 1), atom 1 can scatter the incident photon. The intermediate photon can be scattered by

atom 2 to be detected in the backward direction with the required helicity. Along the reverse path, the photon must be scattered first by atom 2. But atom 2 cannot scatter the incident photon with positive helicity because it is in the maximum magnetic quantum number state $|m_2 = +1/2\rangle$. This simple example shows a situation where the reverse amplitude T_{rev} is strictly zero while the direct one T_{dir} is not. Consequently, this vanishing interference does not contribute at all to the CBS enhancement factor. More generally, a path and its reverse partner will have nonzero but different amplitudes $T_{\text{dir}} \neq T_{\text{rev}}$, resulting in a loss of contrast and an overall enhancement factor less than 2.

We now sketch the general lines of the complete calculation. Consider double scattering (higher orders can be calculated similarly) of a photon ($\mathbf{k}, \boldsymbol{\epsilon}$) into the mode ($\mathbf{k}', \boldsymbol{\epsilon}'$). The transition amplitudes can be calculated using standard diagrammatic techniques [13]. The dependence of the direct amplitude T_{dir} on the internal atomic structure factorizes into

$$t_{\text{dir}} = \langle m'_1 m'_2 | (\boldsymbol{\epsilon}'^* \cdot \mathbf{d}_2) (\mathbf{d}_2 \cdot \boldsymbol{\Delta} \cdot \mathbf{d}_1) (\boldsymbol{\epsilon} \cdot \mathbf{d}_1) | m_1, m_2 \rangle, \quad (4)$$

where $\boldsymbol{\Delta}_{ij} = \delta_{ij} - \hat{\mathbf{n}}_i \hat{\mathbf{n}}_j$ is the projector onto the plane transverse to the unit vector $\hat{\mathbf{n}}$ joining the two atoms. The internal part of T_{rev} is obtained by exchanging \mathbf{d}_1 and \mathbf{d}_2

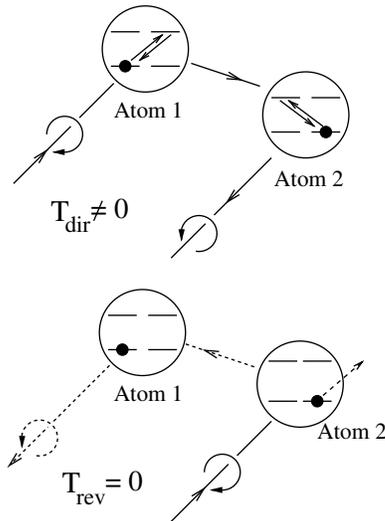


FIG. 1. Example of a direct and reverse scattering path having different amplitudes: double Rayleigh scattering on a $J = 1/2 \rightarrow J' = 1/2$ transition in the helicity preserving channel with positive incident helicity. The arrows show atomic transitions corresponding to absorption and emission of photons; the dashed lines show the process which has a vanishing amplitude.

in Eq. (4) (but *not* the magnetic quantum numbers):

$$t_{\text{rev}} = \langle m'_1 m'_2 | (\boldsymbol{\epsilon}^{\prime*} \cdot \mathbf{d}_1) (\mathbf{d}_1 \cdot \boldsymbol{\Delta} \cdot \mathbf{d}_2) (\boldsymbol{\epsilon} \cdot \mathbf{d}_2) | m_1, m_2 \rangle. \quad (5)$$

The average reflected intensity is proportional to $\langle |T_S|^2 \rangle + \langle |T_{\text{dir}} + T_{\text{rev}}|^2 \rangle$ where the brackets $\langle \dots \rangle$ indicate the ensemble average over the spatial distribution of the atoms and the average over the distribution of internal states $\{m\}$. We assume the atoms to be uniformly distributed in half-space, allowing comparison with results for classical point scatterers [15]. The initial distribution of internal states is supposed to be a complete statistical mixture, which is likely to be the case under usual experimental conditions.

Two different techniques have been used for the average over internal states: first, we have calculated the direct and reverse amplitudes for every possible initial and final state in terms of Clebsch-Gordan coefficients and performed the sum over all possible states using a standard calculation program (MATHEMATICA). Second, we derived analytical expressions [16] for the single and double scattering contributions using standard techniques of irreducible tensor operators [17]. The two approaches give the same result. Let us note that all results known for classical point scatterers [15] are recovered in the case of a $J = 0 \rightarrow J' = 1$ transition, for example, an enhancement factor equal to 2 in the $h \parallel h$ channel.

The normalized CBS angular intensity profile in the $h \perp h$ channel for a $J = 3 \rightarrow J' = 4$ transition is shown in Fig. 2 together with the experimental curve [11,12]. For the calculation, we used the measured value $k\ell \approx 14\,000$. The agreement is satisfactory, the shapes being similar,

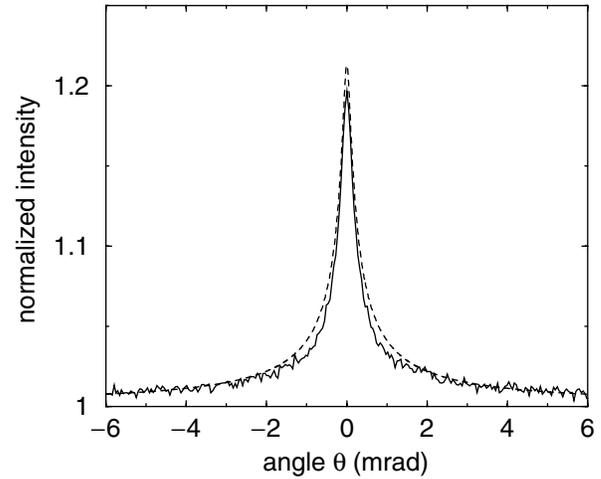


FIG. 2. Backscattered intensity normalized to the background as a function of the deviation θ from exact backscattering. The solid line is the experimental observation [11,12] in the $h \perp h$ channel on Rb atoms ($J = 3 \rightarrow J' = 4$ transition), while the dashed line is the result of the calculation taking into account single and double scattering and the internal structure of the atom. The reduction of the enhancement factor from 2 to 1.2 is due to both single scattering and the loss of contrast in the interference between direct and reverse scattering sequences because of the quantum internal structure of the atomic scatterers.

with a calculated FWHM $\Delta\theta = 0.57$ mrad, close to the experimental value of 0.50 mrad. The shapes in the various polarization channels are slightly different, but comparable to what is obtained for classical dipole scatterers, with widths $\Delta\theta$ of the order of $1/k\ell$.

In Table I, we show the enhancement factors predicted by our calculation for the transition $J = 3 \rightarrow J' = 4$ in the various channels together with the experimentally observed values. We also show the value of the enhancement factor when single scattering is not taken into account, that is, $1 + I_C(0)/I_L$; this is a direct measure of the effect of the internal structure of the atom, as this quantity is equal to 2 in all channels for classical spherical scatterers (and

TABLE I. Enhancement factor of the average intensity scattered in the backward direction for a $J = 3 \rightarrow J' = 4$ transition of atoms uniformly distributed in a semi-infinite medium, in the four polarization channels. In the first column, single scattering is not included in the calculation, so that deviations from a factor of 2 are entirely due to the imbalance of interfering amplitudes along the direct and reverse paths. For classical dipole point scatterers, this factor is 2 in the four channels. When single scattering is taken into account (second column), the enhancement factors come close to the experimental values of [11].

	Double scattering only	Single and double scattering	Experimental value
$h \perp h$	1.71	1.21	1.20
$\text{lin} \perp \text{lin}$	1.45	1.20	1.15
$\text{lin} \parallel \text{lin}$	1.60	1.20	1.12
$h \parallel h$	1.22	1.17	1.06

for a $J = 0 \rightarrow J' = 1$ transition). Clearly, in the $h \parallel h$ channel, the imbalance of interfering amplitudes appears as the key mechanism of the observed reduction of the enhancement factor. In the other channels, the single scattering contribution also plays an important role. Few words of caution are necessary: we have here neglected higher scattering orders and assumed a semi-infinite medium, whereas the experimental cloud has a Gaussian distribution of atoms. A detailed quantitative comparison should take both these effects into account and is under current study. It is thus not surprising that the calculated enhancement factors are only in fair agreement with the experimental observation in some channels. However, the fact that the calculation predicts correct results—especially the surprising observation that the maximum enhancement is obtained in the $h \perp h$ channel and the minimum one in the $h \parallel h$ channel—is a strong indication that we have caught the essential physical mechanisms.

It should be noticed that light scattered in a degenerate Raman transition does not interfere with the incident light simply because the respective final atomic states are orthogonal ($m' \neq m$). In contrast, degenerate Raman transitions do contribute to CBS for direct and reverse paths leading to the same final state (different from the initial state). There are even situations where they dominate in the CBS signal: in the $h \parallel h$ channel for a $J = 3 \rightarrow J' = 4$ transition, more than 80% of the double scattering CBS signal originates from degenerate Raman transitions.

In conclusion, we have presented a calculation of the CBS enhancement factor including single and double scattering contributions for an atomic transition $J \rightarrow J'$. The main result is the spectacular loss of contrast in the interference between direct and reverse scattering sequences because of the quantum internal structure of the scatterers. We think this could be of paramount importance in the understanding of light propagation in cold atomic media and in the search for strong localization of light.

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