Antilocalization in Coherent Backscattering of Light in a Multi-Resonance Atomic System

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Theoretical prediction of antilocalization of light in ultracold atomic gas samples, in the weak localization regime, is reported. Calculations and Monte-Carlo simulations show that, for selected spectral ranges in the vicinity of atomic $^{85}$Rb hyperfine transitions, quantum coherence in optical transitions through nondegenerate hyperfine levels in multiple light scattering generates destructive interference in otherwise reciprocal scattering paths. This effect leads to enhancement factors less than unity in a coherent backscattering geometry, and suggests the possibility of enhanced diffusion of light in ultracold atomic vapors.

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I. INTRODUCTION

An area of developing interest in fundamental atomic physics research is the influence of disorder on atomic or on mesoscopic dynamics. As an example, recent theoretical results \cite{1} have indicated that disorder-induced transitions from a Bose-Einstein Condensate to Bose or to Anderson glass phases is possible, and within current experimental capability. Recent studies by Krug, et al. \cite{2} have shown how disorder within the eigenenergy spectrum of a single atom can explain anomalies in multiphoton microwave ionization cross-sections. Another area of considerable current research interest is localization of light, which may be described as a disorder-induced phase transition in the transport properties of electromagnetic radiation in strongly scattering random media. Investigations in this area, originally stimulated by Anderson localization of electrons, have resulted in two demonstrations of light localization in condensed matter systems \cite{3, 4}.

Recently, coherent multiple light scattering has been experimentally observed in samples of ultracold \cite{5} and Sr atoms \cite{6, 7}. Corresponding theoretical results \cite{6, 7, 8} have revealed fundamental mechanisms responsible for the observations, and numerical simulations have had considerable success in describing the experimental results. In all cases, the essential physical mechanisms are due to interferences in multiple wave scattering from the constituent atoms of the medium. For ultracold atoms, and under otherwise not stringent conditions, the interferences survive configuration averaging, thus generating macroscopic observables. For radiation incident on a disordered system, the effect manifests itself as a spatially narrow (∼1 mrad) cusp-shaped intensity enhancement in the nearly backwards direction \cite{11, 12}; this is referred to as the coherent backscattering (CBS) cone. In the present case, the disorder is principally due to the random locations of the atoms as found in a typical ultracold atomic gas sample. The detailed angular profile and magnitude of the enhancement depends on the polarization of the incident and the detected light, and on the physical size of the sample under study. For classical radiation scattering from a $^1S_0 \rightarrow ^1P_1$ atomic transition, the largest possible interferometric enhancement is to increase the intensity by a factor of two \cite{5, 11, 13} over the incoherently scattered background light.

Earlier \cite{11} we have shown that energetically remote atomic resonances can influence the spatial distribution of backscattered intensity in the cone region. For near-resonance scattering, these transitions appear as a spectral asymmetry, in certain polarization channels, in the interferometric enhancement. In addition, for CBS from a completely oriented atomic gas, we have found, in a double scattering limit, a near-resonance enhancement approaching the classical limit of two \cite{2}. In the present paper, we present a surprising theoretical result which is due to the presence of off-resonance transitions. We find, for non resonant scattering, but over a quite wide spectral range, destructive interference in certain polarization channels of the coherently backscattered light. This results in so-called antilocalization \cite{12} in the optical regime, with a persistent enhancement less than unity. Such effects in electron transport have been widely studied in a range of physical systems where spin-orbit interaction is important \cite{14, 15}. Theoretical study of polarization effects in coherent backscattering of $s = 1/2$ massive particles by Gorodnichev, et al. \cite{17} have shown related novel effects.

In the following sections we present a brief overview of the physical system, including interferences generated by off resonance light scattering from multilevel atoms. This is followed by a summary of the essential theoretical ideas, and our approach to simulate coherent multiple...
scattering in an ultracold atomic gas. We then present theoretical results, in a double scattering limit, illustrating the antilocalization effect and its physical origin.

II. OVERVIEW OF THE PHYSICAL SYSTEM

A. Interference in nonresonant atomic light scattering

The amplitude for quasielastic scattering of light which is detuned several natural widths from any atomic resonance has significant contributions from all energetically nearby atomic transitions. The scattering cross section is essentially given by the Kramers-Heisenberg equation. To illustrate this point, consider light scattering from a single ground level, and two non degenerate excited levels. As the dispersive part of the scattering amplitude changes sign when tuning through a resonance, there will be a minimum (near-zero) in the scattering amplitude in the spectral region between the two levels \[ I_0 \] . The scattering amplitude changes sign again as the incident radiation is tuned through this minimum. When we consider the complexity of real atomic transitions, the spectral location of the minimum depends on the polarization of the incident radiation, and on the multipole distribution within the ground state of the scattering atom. This behavior of the dispersion, when selected in one of two otherwise reciprocal multiple scattering trajectories is what is directly responsible for the existence of antilocalization in atomic CBS.

B. Brief overview of the theoretical treatment

A theory of the CBS process in an ultracold atomic gas has been developed recently by several groups \[ R \] , and takes into account the influence of the optical depth and sample size on the character of the CBS cone. Our treatment \[ R \] also accounts for spectral variations on the CBS cone, and on the influence of far-off-resonance transitions. More recently, we have generalized the theoretical treatment to include effects associated with an angular-momentum-polarized atomic medium as discussed in \[ R \] . In both our earlier theoretical approach \[ R \] , and in the present report, the general analytical development was realized by a Monte-Carlo simulation of coherent multiple scattering in an ultracold \( T < 50 \mu K \), Gaussian-shaped sample of gaseous \(^{85}\)Rb atoms confined to a magneto optical trap. These simulations, which included experimental parameters to characterize the sample, were in excellent agreement with our experimental results \[ R \].

The most important modification of the simulation procedure, comparing with that which we made earlier, is inclusion of mesoscopically averaged polarization effects for light propagating in the atomic sample. The problem of light propagation through a sample consisting of atoms with arbitrary polarization in their angular momentum was discussed first by Cohen-Tannoudji and Laloe \[ 22 \]. In those papers, such well known effects of optical anisotropy of an atomic vapor as birefringence, gyrotropy and dichroism were connected with the formalism of the irreducible tensor components of atomic polarization, i.e. with the orientation vector and the alignment tensor. In the Green’s function formalism, the problem of light propagation through the polarized atomic vapor was later discussed in Ref. \[ 23 \]. The technique developed there allows one to obtain an analytical solution for the Green’s function in many practically important cases. Basically, the retarded-type Green’s function can be found as a solution of a certain type of Dyson equation, which reveals the normal vacuum wave equation modified by the contribution of the polarization operators on the right-hand side. These operators include all the polarization sensitive effects for forward propagating light. In turn, that lets us make a self-consistent Monte-Carlo simulation of the CBS process, including all the important polarization-sensitive effects. For more details we address the reader to \[ 20 \].

Finally we emphasize that in all cases the simulations are made for conditions quite close to those in the experiment. These conditions include sample size, temperature, shape and density, and the characteristic intensity of the CBS laser beam. These conditions are such that recurrent scattering may be neglected, and such that saturation of the atomic transition is also negligible. In simulations of thermal effects, and of the influence of atomic magnetization on the coherent backscattering enhancement, more severe conditions are used in order to illustrate the possible range of influence of these effects.

III. RESULTS

A. Spectral variation of the CBS enhancement and antilocalization

In this communication, we consider the helicity of the incident radiation to be antiparallel to the orientation of the atomic vapor. This geometry requires special preparation since, because of optical pumping, there is a tendency to reorient the collective spin vector of the atomic ensemble along the beam, especially following a long interaction with the probe light, and after accumulation of enough Raman-type transitions. However, at this stage we can neglect the optical pumping mechanism and assume that most of the atoms populate the \( |F_0, m = -F_0 | \) Zeeman state. In addition, we assume that the ensemble is located in an external magnetic field directed along the light beam. Thus the photons scattered via Raman channels are generally Zeeman frequency shifted. We assume this splitting to be quite small, and comparable or
transitions from the which are shown here, repopulate atoms via Raman transition. The two interfering channels, also of positive helicity. The two interfering channels, of positive helicity incoming photon on a system consisting of two and selected in experiment.

In Fig. 1 we show double backscattering of a positive helicity incoming photon on an ensemble of $^{85}$Rb atoms oriented opposite to the helicity vector of the probe beam. In this example, double scattering is a combination of Rayleigh- and Raman-type transitions. The solid and dashed lines indicate the interfering direct and reciprocal scattering paths for probing between $F_0 = 3 \rightarrow F = 4$ and $F_0 = 3 \rightarrow F = 3$ hyperfine transitions.

However, because of typically very slow relaxation in the ground state, such splitting is enough to consider any Raman-type transition as an inelastic channel in the scattering process. In the following section we will explain how such quasi-elastic scattered modes can be resolved and selected in experiment.

In Fig. 1 we show double backscattering of a positive helicity incoming photon on a system consisting of two $^{85}$Rb atoms; the exit channel consists of detection of light scattered in the backward direction. The respective angular dependence with respect to the location of the atomic scatterers. For an optically thin sample the processes shown in Fig. 1 dominates for atoms located preferably along the probe beam direction. The respective angular factor is proportional to the probability to initiate the $\sigma_+$ or $\sigma_-$ transition on the second atoms by the photon also emitted on the $\sigma_+$ or $\sigma_-$ transition by the first atom.

In our example, as shown in Fig. 2, there are two competing channels of double Raman-type scattering which can interfere constructively. For equidistantly split Zeeman sublevels these processes contribute in the helicity preserving channel of the backscattering with the same frequency shift for the outgoing photon as for the diagrams shown in Fig. 1. In an experiment, possible selection of either destructively or constructively interfering channels can be done by focussing attention to their angular dependence with respect to the location of the atomic scatterers. For an optically thin sample the processes of Fig. 1 dominates for atoms located preferably along the probe beam direction. The respective angular factor is proportional to the probability to initiate the $\sigma_+$ or $\sigma_-$ transition on the second atoms by the photon also emitted on the $\sigma_+$ or $\sigma_-$ transition by the first atom.
This probability is given by

\[ P_{++}(\theta) = P_{--}(\theta) \propto \frac{1}{4}(\cos^2 \theta + 1)^2. \] (1)

In turn the processes shown in Fig. 2 are dominant if atoms are located preferably in the plane orthogonal to the probe beam. The respective angular factor is proportional to the probability to initiate the \( \pi \) transition on the second atom by the photon emitted on the \( \pi \) transition by the first atom:

\[ P_{\pi\pi}(\theta) \propto \sin^4 \theta. \] (2)

By comparing the angular distributions 1 and 2, one can expect that, for a cigar-type atomic cloud, stretched along the probe beam direction and squeezed in orthogonal directions, the destructively interfering channels should give the dominant contribution.

A more general way to select the destructively interfering channels can be achieved if the Zeeman sublevels are non-equidistantly split by applied fields. For heavy alkali atoms, as in the current example of \( ^{85}\text{Rb} \), this can be done by admixture of an external electric field. But it would be even more straightforward to accomplish this for lighter alkali atoms, where the splitting is always non-equidistant because of quadratic Zeeman effect. In this case the contribution of the processes shown in Fig. 2 can be cancelled out and in double scattering the destructive interference can drop the output intensity down to near the zero level.

However, as can be verified, the effect of destructive interference can be perfectly selected only for double scattering. The destructive interference associated with the diagram of Fig. 1 can be constructive for more general diagrams, including higher orders of scattering. We can expect that for a dense sample, because of accumulation of multiple scattering contributions of different orders, the effect of the destructive interference in the backscattered light still survives but can be suppressed. Realistic estimation of the remaining anti-enhancement factor can be done only numerically.

In Fig. 3 we show the results of numerical simulations of the enhancement factor in the case of pure double scattering for a spherically symmetric dense Gaussian-type cloud and for the Zeeman splitting in the ground state of \( ^{85}\text{Rb} \). In the graphs plotted in Fig. 3 the optical depth has the same value of \( b = 1 \) for each detuning laser frequency \( \omega_L \) or corresponding detuning \( \Delta = \omega_L - \omega_{43} \). The solid curve shows the contribution of the processes depicted in Fig. 1. The frequency of the probe laser \( \omega_L \) scans the whole upper hyperfine manifold from the \( F_0 = 3 \) ground hyperfine level, and there are two evident off-resonance spectral domains associated with antilocalization. In these spectral ranges, the enhancement factor is negative. It is interesting that even in such an ideal situation, the anti-enhancement is less than 100%. This is because of the polarization dependence of the refractive index along the light ray, which is characterized by the properties of the Green propagation function; this essentially modifies the results of the ideal conditions required for complete destructive interference. The chain curve in Fig. 3 shows how the enhancement factor would change if both the destructive and constructive interfering channels shown in 1 and 2 were taken into account. Comparison of these dependencies suggests to us that antilocalization would be more readily observed in a system with non-equidistant Zeeman sublevels.

An essential and important point is that maximal constructive interference in the CBS process is normally associated with time reversed symmetry of the scattering amplitude. Indeed, the reciprocal path is described by an amplitude which, in certain cases, can be similar to a time reverse amplitude of the direct path. That is the situation if there are only two interfering channels and Rayleigh type scattering. But that is not the case for processes appearing in the Raman channels and assisted by a magnetic type interaction such as hyperfine interaction of nuclear and electronic spins. The destructively interfering amplitudes shown in Fig. 1 are not reversed in time. The complete time symmetry needs that the sign of the field to be changed for the time reverse amplitude, leading to a sign change in angular momentum projectors. In our case the transition amplitude is in fact determined by the matrix elements of only the operators acting on electronic degrees of freedom and nuclear spin plays a role similar to an external magnetic field with respect to such an amplitude. In turn this means that the processes discussed for isolated atoms will not be seen without internal magnetic interactions as a hyperfine interaction, which is in fact obvious as far as the ground state of atom would be a spin one half state in this case.

Finally, we point out an interesting possibility sug-
gested by our results. It is well known \cite{11,12} that constructive interference in recurrent scattering can reduce the effective diffusion constant in a randomly scattering medium. Alternately, this means that optical diffusive transport through the medium suffers from an enhanced resistance to the diffusive current. In direct analogy with our results, destructive interferences in recurrent scattering should lead to modified diffusion of light in the medium. Although the regime of recurrent scattering has not yet been reached in experiments on ultracold atomic gases, it is well within the current capabilities of experimental ultracold atomic physics. A useful observable in that case would be the analogue of the recent experiments of Labeyrie, et al. \cite{24}, where the time dependence of light emerging from an ultracold cloud of $^{85}$Rb was measured. The rate of emergence of difusive light should be enhanced in the antilocalization spectral regime.

\section*{B. Proposed experimental investigation using light-beating spectroscopy}

In the previous section it was indicated that the calculations were done in a small static magnetic field on the order of a few gauss. This produces a slight ground state Zeeman shift which may be exploited to select different quasi-elastic Raman channels contributing to the multiple scattering process. The essential tool is the light beating method, which is a common technique of high resolution spectroscopy. In an application of this method to observation of antilocalization, the scattered light should be mixed with a local oscillator in a balanced heterodyne detector. This coherent and high quality stable mode could be a portion of the monochromatic probe light. Then any Raman scattering channel can be observed in the resultant photocurrent spectrum because of the relatively low frequency of the beat note associated with the Zeeman shift compared with the local oscillator. With this approach, the Raman scattering could be observed as a resonant feature in the photocurrent spectrum at the reference frequency equal to the Zeeman splitting between sublevels $|F_0, m = -F_0 + 2\rangle$ and $|F_0, m = -F_0\rangle$.

Here we mention only that the photocurrent spectrum reproduces the spectral profile of the scattered light emerging the sample and responding to monochromatic incident light. While probing the sample in an off-resonant domain, the spectral distribution is described by the effect of any Raman shift and also by broadening of the originally monochromatic coherent wave as a result of atomic motion. For the sake of simplicity, let us consider an optically thin atomic sample (in the frequency range associated with antilocalization) and ignore effects associated with optical depth and anisotropy; these features may be quite important under real experimental conditions. Then the contribution of single scattering is described by the following spectral profile

$$I_1(\omega) \propto \int d\tau e^{i(\omega - \omega_R)\tau} \left\langle \exp \left[ \frac{2\omega_L}{c} (z(\tau) - z(0)) \right] \right\rangle$$  \hspace{1cm} (3)$$

where $\omega_L$ is the probe light laser frequency and $\omega_R$ is the carrier frequency for light scattered via the Raman channel. The angle brackets denote averaging over the velocity distribution for any atom randomly moving in space. For steady state conditions, the stochastic function $z(\tau) = z(0) + v_\tau$, where $v_\tau$ is a random atomic velocity projected on the $z$-direction, and shows the atomic displacement along the $z$-direction for a short time increment $\tau$. The spectral profile for double scattering can be expressed as

$$I_2(\omega) \propto \int d\tau e^{i(\omega - \omega_R)\tau} \left\langle \exp \left[ \frac{i\omega_L}{c} (s_{12}(\tau) - s_{12}(0)) \right] \right\rangle$$  \hspace{1cm} (4)$$

where $s_{12}(\tau)$ denotes the displacement of the scattering loop for any pair of atomic scatterers. This displacement is associated with the random motion of each atom, and includes all the spatial coordinates with no preference for any particular $z$-direction. The averaging in \cite{13} is extended over the velocity distribution of two atoms. In this expression we ignore Sagnac-type retardation effects for reciprocal paths; we are neglecting the small displacement of atoms during the time of light propagation between the scatterers.

From these equations, we see that the spectral profiles of single and double scattering are similar but not identical. Thus it would be possible to resolve these terms in a sufficiently precise spectral analysis. The most critical experimental aspect would be to resolve the Raman components in comparison with the Doppler broadening, which should then be much less than Zeeman splitting. This means that the atoms should be quite cold and heating effects associated with the probe light interaction would be undesirable. Such conditions are readily achievable using techniques of ultracold atomic physics; similar conditions applied to our previous measurements of coherent backscattering from ultracold atomic Rb \cite{14,15} As a preliminary estimate, we compared the total intensities given by the integrals in \cite{13} and \cite{14} and of similar expressions for higher orders of multiple scattering over $\omega$. This gives us the enhancement factor for such a Raman-type scattering channel, which could be expressed in terms of cross sections, as

$$X_{EF} = \frac{d\sigma^S + d\sigma^L_1 + d\sigma^L_2}{d\sigma^S + d\sigma^L_1}$$  \hspace{1cm} (5)$$

where the denominator contains the contributions of single scattering and ladder terms of all scattering orders and the numerator has an additional interference term. For a sample where the spectrally-dependent optical depth $b$ in the range of the enhancement minimum is varied near unity, the enhancement factor changes in the interval of a few percent and has a rather stable minimum value near $-0.03$. An effect of such size is readily
within current experimental capability. The relatively small value can be explained by the fact that double scattering cannot fully compete with the contributions from single scattering. As we have verified by numerical simulations, in a dense sample with $b$ increased to ten, and including higher orders of scattering, the effect of destructive interference appearing in double scattering also survives in the level of a few percent, but is partially washed out by the losses of light via other scattering channels and by effects of optical anisotropy.

IV. SUMMARY

A theoretical study of spectral variations in the coherent backscattering enhancement factor, for a multiresonance atomic gas, has been reported. Numerical simulations of the variations, which consider the influence of hyperfine interferences in multiple light scattering, and atomic magnetization, show that antilocalization, where there is destructive interference in the coherent backscattering cone, can occur in the optical regime. This, in turn, suggests the possibility of enhanced diffusion of light in an ultracold atomic gas. Finally, an experimental approach to observation of the predicted effects, using light-beating spectroscopy, is proposed.

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