

Phase coherence times in the multiple scattering of photons by cold atoms

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We present an analysis of the dephasing present in the multiple scattering of photons by atoms with a quantum internal structure. The corresponding phase coherence times τ_ϕ are obtained as a function of the Zeeman degeneracy of the atomic dipole transition and the polarization state of the photons. These results allow for an explanation of the recent experiments on coherent backscattering of photons from a gas of cold rubidium atoms where the height of the backscattering cone depends on the atomic internal degrees of freedom coupled to the polarization of the photons. Some consequences of these results are presented, and analogies with the case of electronic systems are highlighted.

PACS:05.60.Gg, 32.80.-t, 42.25.Dd

Multiple scattering of waves in complex media accounts for a large number of coherent effects which are often presented under the shortcut of mesoscopic physics. These coherent effects result from interferences between the amplitudes associated to multiple scattering paths of the wave. In the weak scattering limit for which $kl_e \gg 1$, where k is the wave-vector and l_e the elastic mean free path (the average distance between successive collisions), the relevant physical quantities can be obtained from the probability $P(\mathbf{r}, \mathbf{r}', t)$ of quantum diffusion defined as the disorder averaged probability to propagate a wave packet between the points \mathbf{r} and \mathbf{r}' in a time t [1,2]. It allows to describe either weak localization effects in metals [1,3], spectral quantities or the coherent albedo and dynamical effects in multiple scattering of light by classical scatterers [1].

Interference effects are very sensitive to dephasing. Roughly speaking, a dephasing may originate either from an external field [3,4] or from additional degrees of freedom which affect in different ways each of the interfering amplitudes. An example is provided by the spin-flip scattering in metals where the spin of the electron rotates due to scattering by magnetic impurities [3].

In the presence of dephasing, the probability of quantum diffusion can be written as $P(\mathbf{r}, \mathbf{r}', t) \langle e^{i\phi(t)} \rangle$, where the random variable $\phi(t)$ is the relative phase of the two interfering paths. Its distribution depends on the origin of the dephasing and we denote by $\langle \dots \rangle$ the average over this distribution. In most cases we have $\langle e^{i\phi(t)} \rangle \simeq e^{-t/\tau_\phi}$ at least for long enough times t . The characteristic time τ_ϕ is the phase coherence or dephasing time. An exponential decrease of the probability of quantum diffusion does not necessarily describe a dephasing process. For

instance, the intensity of an electromagnetic wave which propagates in an absorbing medium decreases exponentially. But this is not a dephasing process since it affects equally both the coherent and incoherent contributions by a decrease of the overall intensity. We propose to speak of dephasing only when the coherent part is reduced with respect to the incoherent part.

Recently, the dephasing induced by the Coulomb interactions in metals has been reconsidered in detail in view of the recent experiments on the dephasing time $\tau_\phi(T)$ of the electrons at the Fermi level where an unexpected saturation at low temperature has been observed [5].

The propagation of photons in complex systems addresses similar questions and provides new sources of dephasing like the motion of the scatterers [6]. It also provides access to the type of scattering experienced by the photons (Rayleigh, Mie, resonant, Raman etc.) so that we have at our disposal both a theoretical description of the elementary scattering events and its generalization to multiple scattering.

Dephasing and decoherence in multiple scattering are sometimes perceived as a nuisance which prevents the observation of full interference effects. Instead, when the dephasing is well controlled, it should better be viewed as a unique tool in order to obtain new insights on the system that are usually out of reach in the very dilute and single scattering limits. A beautiful implementation of this has been demonstrated in the so-called diffusive wave spectroscopy [7]. There, the controlled dephasing between multiple scattering trajectories of the photons resulting from the dynamics of the scatterers allows to probe this dynamics on time scales unreachable otherwise.

Recently, the problem of multiple scattering of photons in a gas of cold atoms has been investigated in detail and coherent backscattering has been observed [8,9]. It has been shown that the existence of internal atomic degrees of freedom modifies significantly the coherent backscattering [10]. The purpose of this letter is to present an analysis of the dephasing induced by the internal atomic degrees of freedom and to derive corresponding expressions for the phase coherence times as a function of the polarization state of the photons and of the Zeeman degeneracy. We believe that the present analysis provides relevant spectroscopic tools in order to probe the dynamics of cold atoms.

We describe a gas of N atoms as two-level systems of characteristic transition frequency ω_0 [10]. The ground state defines the zero of energy and has total angular

momentum J . The excited state has a total angular momentum J_e and a natural width Γ due to coupling to the vacuum fluctuations. We shall assume, moreover, that the velocity v of the atoms is small compared to Γ/k (k is the light wave-vector) but large compared to $\hbar k/M$ (M being the mass of the atom), so that it is possible to neglect the Doppler and recoil effects. The external degrees of freedom of the atoms are therefore the classical assigned positions \mathbf{r}_α ($\alpha = 1, \dots, N$) uncorrelated with one another. The corresponding one-atom Hamiltonian

$$H_{at} = \omega_0 \sum_{m_e = -J_e}^{J_e} |J_e m_e\rangle \langle J_e m_e| \quad (1)$$

describes the internal quantum degrees of freedom, in units where $\hbar = c = 1$. No magnetic field is supposed to be present, so that the two levels are respectively $(2J+1)$ and $(2J_e+1)$ -fold degenerate.

The atom-photon interaction is described within the dipole approximation, namely using the Hamiltonian

$$V = - \sum_{\alpha=1}^N \mathbf{D}_\alpha \cdot \mathbf{E}(\mathbf{r}_\alpha) \quad (2)$$

where \mathbf{D}_α is the atomic dipole operator and $\mathbf{E}(\mathbf{r})$ is the quantized electric field operator (quantization volume \mathcal{V}) evaluated at the center of mass \mathbf{r}_α of each atom. The natural width of the excited atomic state is $\Gamma = d^2 \omega_0^3 / 3\pi \epsilon_0$ where $d = \langle J_e || \mathbf{D} || J \rangle / \sqrt{2J_e + 1}$ in standard notations. We define the dimensionless dipole operator $\mathbf{d} = \mathbf{D}/d$ with non vanishing matrix elements only between the two states J and J_e . The elastic scattering process between the two states $|\mathbf{k}\epsilon, Jm\rangle$ and $|\mathbf{k}'\epsilon', Jm'\rangle$, where $|\mathbf{k}\epsilon\rangle$ is a one-photon Fock state of the free transverse electromagnetic field in the mode \mathbf{k} of polarization ϵ , is described by the single scattering transition amplitude $t_{ij}(m, m', \omega) = t(\omega) \langle Jm' | d_i d_j | Jm \rangle$. Here, the resonant scattering amplitude is given by

$$t(\omega) = \frac{3}{2\pi\rho_0(\omega)} \frac{\Gamma/2}{\delta + i\Gamma/2} \quad (3)$$

where $\delta = \omega - \omega_0$ is the detuning of the probe light from the atomic resonance and $\rho_0(\omega) = \mathcal{V}\omega^2/2\pi^2$ is the free photon spectral density. The amplitude t_{ij} is a 3×3 -matrix which connects the incoming and outgoing polarizations of the scattering photon. It can be decomposed into the sum of a scalar, an antisymmetric and a traceless symmetric part. The scalar part is the only one which remains in the case of the Rayleigh scattering on a classical dipole ($J = 0, J_e = 1$).

In order to characterize the multiple scattering of a photon of frequency ω , we first calculate its average propagator $\overline{G}(\omega)$. The averaging is over the uncorrelated positions \mathbf{r}_α of the atoms and over the magnetic quantum numbers m_α of the atoms. The first, standard average restores the translation invariance. The

internal average, a trace with a scalar density matrix ρ assuming that the atoms are prepared independently and equally in their ground states, restores rotational invariance. The calculation of the average propagator reduces to that of the scalar self-energy $\Sigma_{ij}(\omega) = \Sigma(\omega)\delta_{ij}$. For a dilute enough atomic gas, it can be calculated within the self-consistent Born approximation [1] which neglects all possible interference originating from photon exchange between distinct atoms: $\Sigma(\omega) = NM_J t(\omega)$ where $M_J = \frac{1}{3}(2J_e + 1)/(2J + 1)$ such that $M_0 = 1$ [10]. The real and the imaginary parts of the self-energy give respectively the photon frequency shift and the elastic scattering mean free time $\tau_e = -[2\text{Im}\Sigma(\omega)]^{-1}$. The elastic mean free path $l_e = \tau_e$ (remember $c = 1$) measures the average distance between two successive collisions of the photons. It is worth emphasizing that l_e , up to the scalar factor M_J , is independent of the internal quantum structure of the atoms. Indeed, the optical theorem assures that the total cross section $\sigma = 1/nl_e$ equals the imaginary part of the diagonal matrix element of the collision amplitude $t_{ij}(\omega)$ which depends on the scalar component of this tensor only.

To go further, we need to calculate the (time integrated) average probability $P(\mathbf{r}_0, \mathbf{r})$ for a photon to propagate from a point source \mathbf{r}_0 to a point \mathbf{r} . We will calculate its Fourier transform

$$P_{\mathbf{k}\mathbf{k}'}(\mathbf{q}) \propto \overline{\left\langle \mathbf{k} - \frac{\mathbf{q}}{2} \left| G^A \right| \mathbf{k}' - \frac{\mathbf{q}}{2} \right\rangle \left\langle \mathbf{k}' + \frac{\mathbf{q}}{2} \left| G^R \right| \mathbf{k} + \frac{\mathbf{q}}{2} \right\rangle} \quad (4)$$

rewritten as $P_{\mathbf{k}\mathbf{k}'}(\mathbf{q}) \propto \overline{G^A G^R} + \overline{G^A G^R} \Gamma_{\mathbf{k}\mathbf{k}'}(\mathbf{q}) \overline{G^A G^R}$. The first term is the Drude-Boltzmann contribution. In the weak scattering limit $kl_e \gg 1$, the second term contains two contributions: the incoherent diffuson Γ_d for which the phase averages to zero, and the coherent cooperon Γ_c which still retains an overall phase. In the limit of dilute atomic gases, it is possible to write for both the diffuson and the cooperon an integral equation which generalizes the well-known scalar calculation [1] and takes into account the change of the photon polarization as a result of the existence of internal degrees of freedom of the atoms [11].

In order to describe multiple scattering, we need first to determine the atomic intensity vertex U (the differential cross section), the average square of the matrix element $\langle \mathbf{k}'\epsilon' | T(\omega) | \mathbf{k}\epsilon \rangle = (\overline{\boldsymbol{\epsilon}'} \cdot t(\omega) \cdot \boldsymbol{\epsilon}) e^{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{r}}$ of the transition operator. Averaging over the position and the internal quantum numbers yields [10]

$$\begin{aligned} U &= N \text{Tr} [\rho (\overline{\boldsymbol{\epsilon}}_4 \cdot t(\omega) \cdot \boldsymbol{\epsilon}_3) (\overline{\boldsymbol{\epsilon}}_2 \cdot t(\omega) \cdot \boldsymbol{\epsilon}_1)] \delta_{\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_2 + \mathbf{k}_4} \\ &= M_J |t(\omega)|^2 \epsilon_{1i} \overline{\epsilon}_{2j} \epsilon_{3k} \overline{\epsilon}_{4l} I_{il,jk} \delta_{\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_2 + \mathbf{k}_4}. \end{aligned} \quad (5)$$

The vertex U describes the scattering of two incoming photons $(\mathbf{k}_1, \boldsymbol{\epsilon}_1)$ and $(\mathbf{k}_3, \boldsymbol{\epsilon}_3)$ into the two outgoing states $(\mathbf{k}_2, \boldsymbol{\epsilon}_2)$ and $(\mathbf{k}_4, \boldsymbol{\epsilon}_4)$. The rank-four tensor $I_{il,jk} = M_J^{-1} \text{Tr} [\rho d_l d_k d_j d_i]$ can be decomposed into its three irreducible components [11]

$$I_{il,jk} = \sum_{K=0}^2 \lambda_K T_{il,jk}^{(K)} \quad (6)$$

where the eigenvalues

$$\lambda_K = 3(2J_e + 1) \left\{ \begin{matrix} 1 & 1 & K \\ J_e & J_e & J \end{matrix} \right\}^2 \quad (7)$$

are given in terms of $6j$ -Wigner symbols. The three basic tensors

$$T_{il,jk}^{(0)} = \frac{1}{3} \delta_{il} \delta_{jk} \quad (8)$$

$$T_{il,jk}^{(1)} = \frac{1}{2} [\delta_{ij} \delta_{kl} - \delta_{ik} \delta_{jl}] \quad (9)$$

$$T_{il,jk}^{(2)} = \frac{1}{2} [\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl}] - \frac{1}{3} \delta_{il} \delta_{jk} \quad (10)$$

project on the scalar, antisymmetric and symmetric traceless components, respectively, of the field intensity matrix $E_i E_j$. Note that energy conservation imposes $\lambda_0 = 1$ for all J, J_e . For the pure dipole scatterer $J = 0, J_e = 1$, the scattering tensor is proportional to the identity, implying that all eigenvalues are $\lambda_K = 1$.

Then, the propagation of the intensity between successive scattering events is described by the product of the average photon propagators, namely

$$G_{il,jk}(\mathbf{q}) = \frac{3}{8\pi l_e} \int d^3 r \frac{e^{-r/l_e}}{r^2} \Delta_{ij} \Delta_{kl} e^{i\mathbf{q}\cdot\mathbf{r}} \quad (11)$$

where $\Delta_{ij} = \delta_{ij} - \hat{r}_i \hat{r}_j$ describes the transverse projection of the polarization vector. In the diffusive limit $ql_e \ll 1$, the intensity propagator $G_{il,jk}$ can also be decomposed using the basic tensors (8-10) and we recover the eigenvalues $b_0 = 1, b_1 = \frac{1}{2}$ and $b_2 = \frac{7}{10}$ as for the classical dipolar case [12].

The summation of the geometric series built from the elementary scattering event, i.e. the tensorial product of $I_{il,jk}$ and $G_{il,jk}$, yields in the diffusive limit $ql_e \ll 1$ for the diffuson

$$\Gamma_d(\mathbf{q}) = \frac{3}{4\pi\rho_0 l_e \tau_{\text{tr}}} \sum_{K=0}^2 \Lambda_K(q) \epsilon_{1i} \bar{\epsilon}_{2j} \epsilon_{3k} \bar{\epsilon}_{4l} T_{il,jk}^{(K)} \quad (12)$$

where the propagators of the three eigenmodes are

$$\Lambda_K(q) = \frac{1}{b_K} \frac{1}{Dq^2 + \tau_d^{-1}(K)}. \quad (13)$$

Here, $D = l_e^2/3\tau_{\text{tr}}$ is the diffusion constant, involving the resonant transport time scale $\tau_{\text{tr}} = l_e + \Gamma^{-1}$ [11,13]. The polarization relaxation times

$$\tau_d(K) = \frac{b_K \lambda_K}{1 - b_K \lambda_K} \tau_{\text{tr}} \quad (14)$$

describe the depolarization of the initial light beam and are related to the depolarization factors calculated in the

classical Rayleigh case [14]. Among the three modes, we have $\tau_d^{-1}(0) = 0$, and the corresponding scalar mode $\Lambda_0(q)$ diverges for small q . It is the singlet Goldstone mode associated with the local conservation of the number of photons. The antisymmetric mode $K = 1$ is analogous to the triplet mode obtained in the spin-orbit scattering in electronic systems [3].

A finite time τ_d leads to an exponential attenuation of incoherent diffusion contribution. In order to identify unambiguously a dephasing time, we have to consider the cooperon contribution Γ_c . It accounts for the interference of amplitudes associated to two time-reversed multiple scattering paths of the photons and can be calculated along the same lines. The atomic intensity vertex has a form similar to (6), with the new eigenvalues defined in terms of $9j$ -symbols,

$$\chi_K = 3(2J_e + 1) \left\{ \begin{matrix} 1 & J_e & J \\ 1 & J & J_e \\ K & 1 & 1 \end{matrix} \right\} \quad (15)$$

The cooperon contribution reads

$$\Gamma_c(\mathbf{q}_c) = \frac{3}{4\pi\rho_0 l_e \tau_{\text{tr}}} \sum_K X_K(q_c) \epsilon_{1i} \bar{\epsilon}_{2j} \bar{\epsilon}_{4k} \epsilon_{3l} T_{il,jk}^{(K)} \quad (16)$$

where $\mathbf{q}_c = \mathbf{k} + \mathbf{k}'$ is the total momentum and the polarization vectors ϵ_3 and $\bar{\epsilon}_4$ have been exchanged. The propagators of the three eigenmodes are given by

$$X_K(q) = \frac{\chi_K}{Dq^2 + \tau_d^{-1}(K) + \tau_\phi^{-1}(K)} \quad (17)$$

The overall depolarization described by the times $\tau_d(K)$ affects the cooperon as well. But the cooperon involves an additional contribution, the dephasing time

$$\tau_\phi(K) = \frac{\lambda_K \chi_K}{\lambda_K - \chi_K} \tau_{\text{tr}} \quad (18)$$

This expression constitutes our main result. The limit of the classical Rayleigh scattering $J = 0$ corresponds to $\lambda_K = \chi_K = 1$ and thus to the absence of dephasing ($\tau_\phi(0) = \infty$). Of particular interest is the value of $\tau_\phi(0)$ associated to the intensity of the field. Explicitly, we find

$$\frac{\tau_\phi(0)}{\tau_{\text{tr}}} = \begin{cases} (J(2J+3))^{-1}, & J_e = J+1 \\ J^2 + J - 1, & J_e = J \\ (2J^2 + J - 1)^{-1}, & J_e = J-1 \end{cases} \quad (19)$$

An absence of dephasing only occurs for the classical dipole $J = 0$ and in the semi-classical limit $J = J_e \rightarrow \infty$.

In experiments on coherent backscattering (CBS) of light by atoms without an internal degeneracy ($J = 0$), the optimal CBS enhancement factor is found in the polarization channel of preserved helicity [15]. The present analysis allows to explain the unexpected experimental observation that the CBS enhancement factor for cold

rubidium atoms ($J = 3, J_e = 4$) in the channel of preserved helicity is lower than in the channel of flipped helicity [8]. Indeed, following [14], we define a CBS contrast function $\mathcal{C}(n) = \Gamma_c^{(n)}(0)/\Gamma_d^{(n)}(0)$ as the ratio of cooperon and diffuson contributions at the n th scattering order. For large n , only the largest eigenvalues $b_0 = \lambda_0 = 1$ of the diffuson mode expansion will contribute for both polarization channels. For the channel of flipped helicity (\perp), the contrast function behaves as $\mathcal{C}_\perp(n) \sim \frac{30}{7}(b_2\chi_2)^n$ and decreases exponentially. In the channel of preserved helicity (\parallel), all irreducible modes contribute *a priori*: $\mathcal{C}_\parallel(n) \sim \chi_0^n - 3(b_1\chi_1)^n + \frac{5}{7}(b_2\chi_2)^n$. In the case of classical dipole scatterers such that $\chi_K = 1$, the parallel contrast stays optimal, $\mathcal{C}_\parallel(n) \sim 1$. But in the case $J = 3, J_e = 4$, the product $b_2\chi_2 = \frac{19}{40}$ is much larger than $\chi_0 = \frac{1}{28}$ and therefore dominates. The contrast in the perpendicular channel then is higher than in the parallel channel, $\mathcal{C}_\perp/\mathcal{C}_\parallel \sim 6$, independently of the scattering order n .

In terms of the above definitions, $b_K\chi_K = [1 + \tau_{tr}/\tau_d(K) + \tau_{tr}/\tau_\phi(K)]^{-1}$. We can therefore conclude that the loss of CBS contrast in perpendicular polarization channels is not a dephasing process since it persists even for $J = 0$ where $\tau_\phi^{-1}(K) = 0$, but still $\tau_d^{-1}(2) > 0$. Since the cooperon and diffuson contributions probe different field correlators, it should rather be regarded as an effect of polarization decorrelation. On the contrary, an atomic internal degeneracy affects the contrast in channels of parallel polarizations and therefore appears as a particularly neat realization of a microscopic dephasing mechanism.

In summary, we have identified dephasing times in the weak localization of light by cold atoms. They depend in a remarkably simple manner on the internal atomic degeneracy and the field polarization mode. It is of interest to notice that the dephasing times can become negative (since for instance $\chi_0 = -\frac{1}{3}$ for $J_e = J = \frac{1}{2}$). Although this does not change the sign of the contribution to the coherent backscattering cone, it might change the sign of the correction to the bulk diffusion constant D [16], signalling an antilocalization contribution like for the spin-orbit correction to the conductivity in metals. This may lead to an unusual behavior at the Anderson localization transition edge. This is quite similar to the case of spin-orbit [3] in disordered metals (symplectic limit) except for the fact that here the problem is richer so that the critical exponents depend on the Zeeman degeneracy J and J_e of the atoms [17].

This research was supported in part by the Israel Academy of Sciences and by the Fund for Promotion of Research at the Technion as well as by the PRIMA research group of the CNRS in France. C(h+.A). M. wish to thank A. Buchleitner and D. Delande for their interest.

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