

Quantum correlations in the optical characteristics of low-temperature H↓ and D↓ gases

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Nonlocal quantum correlations should be manifested in the optical characteristics of low-temperature gases of spin-polarized hydrogen (H↓) and deuterium (D↓). Temperatures $T \sim 0.1$ K, which have already been achieved experimentally and at which the gas remains a Boltzmann gas, would be sufficient.

1. The production of gases of spin-polarized atomic hydrogen (H↓) and deuterium (D↓) at low temperatures T corresponding to the condition $kR_0 \ll 1$ (k is the thermal momentum of the particles, and $R_0 \approx 3.5$ Å is the effective range of the interatomic interaction) opens up the possibility of observing quantum correlations in such gases. Previous work has established that local quantum correlations should have a substantial influence on the probabilities for recombination and relaxation processes and should correspondingly be manifested in the decay kinetics of a gas.^{1,2} The specific spin waves which are observed in gaseous H↓ are related in an essential way to local quantum correlations.^{3–5}

In the present letter it is shown that nonlocal quantum correlations should be manifested in the optical characteristics of sufficiently dense H↓ and D↓ gases. The absorption of photons at the wings of the spectral lines of atomic transitions occurs when a pair of particles close on each other to a distance r_* , where the resonant interaction of an atom with an excited atom coincides with the frequency deviation $\Delta\omega = \omega - \omega_0$, where ω is the photon frequency, and ω_0 is the frequency of the atomic transition. The distance r_* may be substantially greater than R_0 while remaining vastly shorter than the average distance between particles. The possibility of a manifestation of nonlocal quantum correlations in the frequency and temperature dependence of the absorption coefficient turns out to be related to the condition $kr_* \lesssim 1$, which is satisfied even in Boltzmann H↓ and D↓ gases over a wide range of $\Delta\omega$.

2. Let us consider the interaction of an atomic gas of density n with resonant electromagnetic radiation of wavelength λ under the condition

$$n\lambda^3 \gg 1. \quad (1)$$

In this case the broadening of the spectral line due to the resonant interaction of the excited atom with the unexcited atom, $U(r) \sim d^2/r^3$ (d is the reduced dipole moment of the atomic transition), prevails over the natural broadening. Let us assume

$$|\Delta\omega| \gg nd^2 \quad (2)$$

(we are using atomic units). The absorption of the photon then occurs in a pair

interaction of particles at distances $r \ll n^{-1/3}$. The relative motion of the atoms in the initial state is free ($r > R_0$), while that in the final state is dictated by their resonant interaction. We write the Hamiltonian of the interaction responsible for the absorption in the form

$$H' = \frac{1}{2} \tilde{d} \omega_0 \int d\mathbf{r}_1 d\mathbf{r}_2 \hat{\psi}(\mathbf{r}_1) \hat{\psi}(\mathbf{r}_2) \hat{\mathbf{A}}(\mathbf{R}) \hat{\mathbf{D}}^\dagger(\mathbf{r}_1, \mathbf{r}_2) + \text{H.a.} \quad (3)$$

Here $\tilde{d} = \sqrt{2}d$ is the reduced dipole moment of the transition in the quasimolecule, $\hat{\psi}$ is the field operator of the gas of unexcited atoms, and $\hat{\mathbf{A}}$ is the operator of the electromagnetic field. In the long-wave limit, $\lambda \gg r = |\mathbf{r}_1 - \mathbf{r}_2|$, which clearly prevails under condition (1) and at $r \ll n^{-1/3}$, we can set the argument of this operator equal to $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$. The field operator of the excited quasimolecules is

$$\hat{\mathbf{D}}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{\mathbf{q} \alpha s} e^{i\mathbf{q}\mathbf{R}} \mathbf{e}_\alpha(\mathbf{r}) \chi_{\alpha s}(\mathbf{r}) \hat{b}_{\mathbf{q} \alpha s}, \quad (4)$$

where $\hat{b}_{\mathbf{q} \alpha s}$ is the operator which annihilates a quasimolecule with the momentum of the center of mass, $\mathbf{q}; \mathbf{e}_\alpha(\mathbf{r})$ is the polarization vector ($\alpha = 1, 2, 3$); and $\chi_{\alpha s}(\mathbf{r})$ is the wave function of the relative motion of the atoms, which is the solution of the Schrödinger equation with the potential $U_\alpha(\mathbf{r})$.

We write the probability for the absorption of a photon per unit time as follows:

$$W = 2\pi \sum_{i, f} \rho_i |\langle f | \hat{H}' | i \rangle|^2 \delta(E_i - E_f) = \int_{-\infty}^{\infty} dt \sum_i \rho_i \langle i | \hat{H}'(0) \hat{H}'(t) | i \rangle, \quad (5)$$

where ρ_i is the equilibrium density matrix. Making use of the explicit expressions for the operators $\hat{\mathbf{A}}$ and $\hat{\mathbf{D}}$, we find the following expression for the absorption coefficient $K_\omega = W/c$:

$$K_\omega = \frac{\pi \omega_0 d^2}{c} \int_{-\infty}^{\infty} dt \int d\mathbf{r} d\mathbf{r}' d\mathbf{R} d\mathbf{R}' \sum_{\mathbf{q} \alpha s} e^{-i(\Delta\omega - q^2/4m - \epsilon_{\alpha s})t} \times e^{i(\mathbf{p} - \mathbf{q})(\mathbf{R} - \mathbf{R}')} (\mathbf{e}_\alpha(\mathbf{r}') \mathbf{e}_{\mathbf{p}\lambda}^*) (\mathbf{e}_\alpha^*(\mathbf{r}) \mathbf{e}_{\mathbf{p}\lambda}) \chi_{\alpha s}(\mathbf{r}') \chi_{\alpha s}^*(\mathbf{r}) \times \times \langle \hat{\psi}^\dagger(\mathbf{R}' + \frac{\mathbf{r}'}{2}, 0) \hat{\psi}^\dagger(\mathbf{R}' - \frac{\mathbf{r}'}{2}, 0) \hat{\psi}(\mathbf{R} + \frac{\mathbf{r}}{2}, t) \hat{\psi}(\mathbf{R} - \frac{\mathbf{r}}{2}, t) \rangle, \quad (6)$$

where \mathbf{p} is the momentum of the photon, $\mathbf{e}_{\mathbf{p}\lambda}$ is its polarization vector, and $\epsilon_{\alpha s}$ is the energy of the final state of the quasimolecule in the c.m. frame.

The time scale of the variations of the correlation function in (6) would be $1/T$ in the case of an ideal gas. Since we are interested in low temperatures, we adopt the condition

$$|\Delta\omega| \gg T. \quad (7)$$

We can then set $t = 0$ in the correlation function, and we can also ignore the term $q^2/4m \sim T$ in the argument of the exponential function. The summation over \mathbf{q} in (6) allows us to set $\mathbf{R}' = \mathbf{R} = 0$ in the correlation function; an integration over dt converts the t -dependent exponential function into $\delta(\Delta\omega - \epsilon_{\alpha s})$. For low initial momenta (certainly under the condition $k \leq 1/r_*$), the correlation function in (6) is a smooth function of the coordinates \mathbf{r} and \mathbf{r}' . At a sufficiently large value of $\epsilon_{\alpha s}$, on the other hand,

$$|\epsilon_{\alpha s}| \approx |\Delta\omega| \sim d^2/r_*^3 \gg 1/(mr_*^2), \quad (8)$$

the semiclassical approximation can be used for the final-state wave function $\chi_{\alpha s}(\mathbf{r})$, and the integral over $d\mathbf{r}$ and $d\mathbf{r}'$ will be built up near the caustic: the surface on which the relation $\epsilon_{\alpha s} = U_\alpha(\mathbf{r})$. Effectively, the intervals $|\mathbf{r} - \mathbf{r}_*|, |\mathbf{r}' - \mathbf{r}_*| \ll r_*$ are at work in integral (6) [the function $\chi_{\alpha s}$ begins to oscillate markedly at a distance on the order of r_* [($1/mr_*^2$)/(d^2/r_*^3)]^{1/3} $\ll r_*$ from the caustic]. In the case of transitions to states of the continuum, we can thus approximate $\epsilon_{\alpha s}$ by $U_\alpha(\mathbf{r})$ in the argument of the δ -function, and we can sum over s . Using the completeness relation $\sum_s \chi_{\alpha s}^*(\mathbf{r})\chi_{\alpha s}(\mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')$, and integrating over $d\mathbf{r}'$, we finally find

$$K_\omega = \frac{2\pi^2 d^2 \omega_0 n^2}{c} \sum_\alpha \int d\mathbf{r} |\mathbf{e}_\alpha(\mathbf{r}) \mathbf{e}_{\mathbf{p}\lambda}^*|^2 Z(\mathbf{r}) \delta(\Delta\omega - U_\alpha(\mathbf{r})); \quad (9)$$

$$Z(\mathbf{r}) = \frac{1}{n^2} \langle \hat{\psi}^+(\mathbf{r}) \hat{\psi}^+(0) \hat{\psi}(\mathbf{r}) \hat{\psi}(0) \rangle. \quad (10)$$

Condition (8) also ensures an adiabatic separation of the relative motion of the atoms and the rotation of the polarization vector which was adopted in (4).

Under the condition $kr_* \gg 1$, the correlation function is $Z(r_*) = 1$ and expression (9) becomes the result of the standard quasistatic theory (Ref. 6, for example), in which condition (7) is not necessary.

If we consider the sufficiently remote wing of the line, where the condition $|\Delta\omega| \sim (d^2/r_*^3) \gg (\mu_B \mathcal{H})$ holds, we can ignore the presence of the external magnetic field \mathcal{H} in the $\mathbf{H}\downarrow$ and $\mathbf{D}\downarrow$ gases. This field is required to maintain the polarization of the spins. In this case we have $U_\alpha(\mathbf{r}) = (1 - 3\delta_{\alpha 1})(d^2/r^3)$ (one of the polarization vectors is oriented parallel to the axis of the quasimolecule, while the two others are perpendicular to it). Condition (7) predetermines a pronounced asymmetry of the lineshape. At frequencies $\Delta\omega > 0$, corresponding to transitions only to states of the continuum in terms of the relative motion of the quasimolecule, we find from (9) and (10)

$$K_\omega = \tilde{K}(\Delta\omega) Z(r_*); \quad d^2/r_*^3 = \Delta\omega, \quad (11)$$

where $\tilde{K}(\Delta\omega) = (16\pi^3 \omega_0 / 9c)(nd^2/\Delta\omega)^2$ is the known expression of the quasistatic theory for the absorption coefficient in the wing of a line (Ref. 7, for example). Under the condition $\Delta\omega < 0$, there can be only sharp peaks corresponding to transitions to bound states of the excited molecule.

3. Under the condition $kr_* \gg 1$, we naturally have the classical result $K_\omega = \tilde{K}(\Delta\omega)$. In a Bose gas, the absorption coefficient in the wing of a line, (11), increases monotonically with decreasing T , and at $kr_* \ll 1$ it approaches the value $2\tilde{K}(\Delta\omega)$. In a Fermi gas, K_ω falls off monotonically with decreasing T . Under the condition $kr_* \ll 1$ we have $K_\omega \sim T$. The quantum correlations are thus clearly manifested in the temperature dependence of the absorption coefficient of a purely Boltzmann gas (cf. Refs. 3-5).

In H \downarrow and D \downarrow gases, condition (1) holds at usual densities, $n \sim 10^{16}-10^{17}$ cm $^{-3}$. In the temperature interval $T \sim 0.1-0.8$ K, which is characteristic of today's experiments,^{8,9} we could span the range from $kr_* \gtrsim 1$ to $kr_* \lesssim 1$ through a suitable choice of $\Delta\omega$.

The formation of a Bose condensate in a gas of H \downarrow would have fundamental consequences for the absorption coefficient. In the Bose condensation region, the condition $kr_* \ll 1$ would definitely hold. Therefore setting $r_* = 0$ in correlation function (10), we find, in the approximation of an ideal gas,

$$Z(0) = 2 - (n_0/n)^2, \quad (12)$$

where n_0 is the density of the condensate. It can thus be seen that K_ω begins to fall off smoothly from the value $2\tilde{K}(\Delta\omega)$ as the relative number of particles in the condensate increases, returning to the classical value $\tilde{K}(\Delta\omega)$ at $n = n_0$. This effect indicates that it would be possible to optically detect the appearance of a condensate in the system; this capability is particularly attractive in light of the effort which is required to reach the region of Bose condensation.

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