Effect of collisions on large-angle neutron scattering in monatomic gases

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(Submitted 29 December 1985)
Zh. Eksp. Teor. Fiz. **90**, 1701–1706 (May 1986)

The asymptotic behavior of the dynamical form factor of a monatomic gas is investigated in the limit $\Omega \gg qv_T$. The result can be expressed in terms of the atom-atom scattering cross section, and is proportional to Ω^{-6} in the case of a gas composed of identical atoms and Ω^{-4} in the case of a gas mixture.

There are two essentially different limiting cases in the theory of inelastic neutron scattering in gases. For relatively small energy and momentum transfers $\hbar\Omega$ and $\hbar q$, i.e., in the limiting case

$$ql \ll 1, \quad \Omega \lesssim qv_T \tag{1}$$

(l is the mean free path of the atoms and $v_T = (T/m_a)^{1/2}$ is their thermal velocity), the hydrodynamic regime is obtained, when the dynamics of the gas-density fluctuations, on which the scattering occurs, is governed by the equations of hydrodynamics. The spectral distribution of the scattered particles then exhibits peaks corresponding to the excitation of sound and temperature oscillations. In the opposite limiting case

$$\Omega \gg \nu, \quad \Omega \gtrsim qv_T$$

(ν is the effective collision rate) we have the "collisionless regime." The scattering in this case occurs on the individual atoms (we shall henceforth have in mind scattering in a monatomic gas), and the line shape is determined by the velocity distribution of these atoms. (In the theory of light scattering this corresponds to Doppler broadening.) Naturally, in this case the intensity of the scattering involving large energy transfers (i.e., for which $\Omega \gg qv_T$) is exponentially small, in accordance with the fact that the number of particles with velocities $v \gg v_T$ is small, since the scattering is determined by the particles in the "tail" of the Maxwell distribution (see below). But in 1941 V. L. Ginzburg, investigating light scattering in gases, noticed that, at sufficiently high values of Ω/qv, specifically, for

$$\Omega \gg qv_T, \ \nu, \tag{2}$$

the scattering intensity will again be determined by the collisions, and will decrease with Ω according to a power law. This law is different for a gas composed of atoms of the same kind and for a gas mixture. In the first case the intensity will be $\propto \Omega^{-6}$; in the second, $\propto \Omega^{-4}$ (Refs. 1 and 2). Actually, it has not been possible to observe this effect in light scattering in gases. It turns out that it is masked by the stronger effect connected with the fact that the polarizabilities of the atoms in the course of a collision are time dependent because of the distortion of the electron shells of the atoms. This leads to the appearance of the so-called "collision-induced exponential limb" of the scattering line, which has been the subject of a large number of theoretical and experimental investigations (see, for example, Refs. 3 and 4). The Ginzburg effect is smaller under these conditions because of its relativistic character, i.e., because it is connected with the inhomogeneity of the electric field of the light wave. But the effect of the electron-shell distortion manifests itself in a homogeneous field as well. Therefore, the first effect can be observed only in scattering on objects whose scattering properties do not vary during collision. In fact we can speak of scattering by free plasma electrons, which is governed by the electron charge, and of neutron scattering in a monatomic gas. The latter is determined largely by the scattering of the neutrons by the atomic nuclei. And the amplitude of the scattering of a slow neutron by a nucleus does not, of course, depend on the positions of the neighboring atoms.

We will calculate the "collision" contribution to neutron scattering under the condition (2). As a model problem, we shall first consider the scattering of light in an electron-ion plasma under the same conditions, with v_T taken to be the thermal velocity of the electrons.

According to the general theory of light scattering involving small frequency changes (i.e., with $\Omega \ll \omega$, where ω is the frequency of the incident light), the extinction coefficient is given by the formula (119.6) in Ref. 5. For the case of scattering on plasma electrons this formula, after being averaged over the polarizations, takes the form

$$dh = \frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2 (1+\cos^2\theta) S(\Omega, q)\, do' \frac{d\Omega}{2\pi}. \tag{3}$$

Here θ is the scattering angle and $S(\Omega,q)$ is the dynamical form factor for the electrons, which is given in terms of the electron density fluctuation operator by

$$S(\Omega, q) = \int \langle \delta\hat{N}_e(t, \mathbf{r}) \delta\hat{N}_e(0, 0)\rangle e^{i(\mathbf{qr}-\Omega t)} dt d^3r. \tag{4}$$

To compute the form factor, we must, generally speaking, solve the kinetic equation allowing for the collisions. But under condition (2), the ions are virtually stationary, and the electron density fluctuations determine the total-charge-density fluctuations in the plasma. For the latter fluctuations there is a general macroscopic expression, on the basis of which we obtain for $S(\Omega,q)$ the formula (see Ref. 6, Chap. 3, §4)

 0038-5646/86/050997-03$04.00

$$S(\Omega, q)=\frac{\hbar q^2}{2\pi e^2}\frac{\varepsilon_l''(\Omega, q)}{|\varepsilon_l(\Omega, q)|^2}[1-e^{-\hbar\Omega/T}]^{-1},$$

where $\varepsilon_l(\Omega,q)$ is the longitudinal permittivity of the plasma.

Below we shall limit ourselves everywhere to the case of moderate energy transfers, specifically, to the case when $\hbar\Omega\ll T$. In this case the preceding formula reduces to

$$S(\Omega, q)=\frac{Tq^2}{2\pi e^2\Omega}\frac{\varepsilon_l''(\Omega, q)}{|\varepsilon_l(\Omega, q)|^2}. \quad (5)$$

The formulas (3) and (5) reduce the problem to that of obtaining the well-known plasma-theory expressions for ε_l''. If $\Omega\gg\Omega_e$ (Ω_e is the electron plasma frequency), then we can set $|\varepsilon_l|^2\approx 1$. When the collisions are ignored, ε_l'' is determined by the Landau damping:

$$\varepsilon_l''(\Omega, q)=\left(\frac{\pi}{2}\right)^{1/2}\frac{\Omega_e^2\Omega}{(qv_{Te})^3}\exp\left(-\frac{\Omega^2}{2q^2v_{Te}^2}\right),\quad v_{Te}=\left(\frac{T}{m}\right)^{1/2}.$$

As to the collision-related part of ε_l'', it is determined by the collisions between the electrons and the ions, and is equal to (see, for example, Eq. (44.8) in Ref. 7)

$$\varepsilon_l''=\frac{4\pi e^2N_eN_i}{3\Omega^3T}\langle v^3\sigma_{ei}^*(v)\rangle, \quad (6)$$

where σ_{ei}^* is the transport cross section for scattering of an electron by an ion:

$$\sigma_{ei}^*=\int(1-\cos\vartheta)\,d\sigma_{ei}. \quad (7)$$

Here ϑ is the electron-scattering angle, $d\sigma_{ei}$ is the differential scattering cross section, v is the electron velocity, and the angle brackets denote an average over the Maxwell distribution. Substitution into (5) yields finally

$$S(\Omega, q)={}^2/_3N_eN_i(q^2/\Omega^4)\langle v^3\sigma_{ei}^*(v)\rangle. \quad (8)$$

It can be seen from Eqs. (6) and (8) that the scattering is indeed proportional to Ω^{-4} (provided $\hbar\Omega\ll T$), as it should be for a system composed to two kinds of particles—electrons and ions. It can also be seen from Eq. (8) that the contribution in question to the electron form factor is proportional to q^2. Since $q^2\lesssim\omega^2/c^2$ in the case of light scattering, this contribution contains the square of the velocity of light in the denominator, which makes the effect even smaller, as noted above.

Turning to the investigation of the corresponding effect in the case of neutron scattering in monatomic gases, we first consider a gas composed of atoms of the same kind. According to the general theory of neutron scattering (see, for example, the problem at the end of §86 of Ref. 8), the probability for scattering by a unit volume of the gas in unit time is given by

$$dw=\frac{4\pi^2\hbar^2a^2}{M^2}S(\Omega, q)\frac{d^3p'}{(2\pi\hbar)^3}, \quad (9)$$

where a is the slow-neutron scattering length on an atom, M is the reduced mass of the atom and neutron, p' is the final neutron momentum, and $S(\Omega,q)$ is the gas's dynamical form factor, which is given by a formula identical with (4), except that $\delta\hat{N}_e$ is replaced by $\delta\hat{N}=\hat{N}-\overline{N}$, the density fluctuation operator for the atoms of the gas.

To find $S(\Omega,q)$ from the general theory of fluctuations would require the solution of the kinetic equation, this time for the gas atoms. Naturally this cannot be done in the general case. Under the condition (2), however, we can simplify the problem by assuming the collision integral to be small and using the method of successive approximations. The computations turn out in this case to be quite tedious. We shall use here another more intuitive, but quite rigorous method that allows us to obtain the answer effecting the corresponding changes in the formula (8) for the electron form factor.

We shall proceed from the exact quantum-mechanical formula for the form factor. According to this formula, the form factor is proportional to the following sum of squares of matrix elements[1)]:

$$\sum_n\left|\left\langle n\left|\sum_a\exp(i\mathbf{q}\mathbf{r}_a)\right|m\right\rangle\right|^2 \quad (10)$$

(where $\mathbf{r}_a$ is the coordinate operator for the ath particle). In the theory being developed, the momentum transfer $\hbar q$ is assumed to be small, and the matrix elements can be expanded in powers of it. For the case of collisions between different particles—between electrons and ions, for example—we can limit ourselves to the first term of the expansion, an approximation which is clearly equivalent to the dipole approximation in the theory of radiation. Then the square of the matrix elements in (10) reduces to

$$\left|\left\langle n\left|\sum_a(i\mathbf{q}\mathbf{r}_a)\right|m\right\rangle\right|^2. \quad (12)$$

Our problem now is to follow, proceeding from the formula (12), how the transport cross section for scattering of an electron on an ion arises in (8).

Let us first of all note that (8) corresponds to the situation in which only the binary collisions are considered. Accordingly, in (12) also we can consider the scattering of one electron on one ion and multiply the result by N_eN_i. Furthermore, since it is clear *a priori* that, in an isotropic medium, the answer can depend only on $|\mathbf{q}|$, we can average over the directions of $\mathbf{q}$, so that, instead of (12), it is sufficient to compute

$$(q^2/3)N_eN_i|\langle n|\mathbf{r}_a|m\rangle|^2. \quad (13)$$

Let us now assume that the following relation is satisfied[2)]:

$$\Omega t_{\text{col}}\ll 1, \quad (14)$$

where t_{col} is the duration of a collision event. Such a requirement underlies the entire theory that uses the ordinary kinetic equations. The use of the condition (14) becomes more obvious if we assume that the collision can be described semiclassically (the final result does not depend on this assumption). Then, in the case of suitably normalized wave functions the matrix element is equal simply to the Fourier transform of the corresponding quantity:

$$\langle n|\mathbf{r}|m\rangle=\int_{-\infty}^{\infty}\mathbf{r}(t)e^{i\Omega t}\,dt=-\frac{1}{\Omega^2}\int_{-\infty}^{\infty}\dot{\mathbf{v}}e^{i\Omega t}\,dt,\qquad \Omega=\omega_{mn}.$$

The last formal transformation has been made in order to make the integrand vanish at infinity, where the electron acceleration is equal to zero. Now on account of the condition (14), we can set $\Omega=0$ under the integration sign, so that

$$\langle n|\mathbf{r}|m\rangle=-\Delta\mathbf{v}/\Omega^2,$$

where $\Delta\mathbf{v}=\mathbf{v}'-\mathbf{v}^0$ is the change that occurs in the electron velocity during scattering. Using the last expression, we can reduce the expression (13) to the form

$${}^2/_3 q^2 N_e N_i (v^2/\Omega^4)(1-\cos\vartheta). \tag{15}$$

The summation over the final states in the formula (11) reduces in the present case to integration over the scattering cross section de_{ei}, as a result of which the transport cross section σ_{ei}^* appears in (8).

Let us now turn to the case of a gas composed of identical atoms. As before, we can consider mutual scattering of a pair of atoms and multiply the result by the number of pairs, $N(N-1)/2\approx N^2/2$. As in the theory of radiation, however, the effect vanishes in the dipole approximation, since the sum $\mathbf{r}_1+\mathbf{r}_2$ does not possess off-diagonal matrix elements in view of the conservation of the velocity of the center of mass. Therefore, we must take account of the next "quadrupole" term, i.e., make the substitution

$$\sum_a e^{i\mathbf{qr}}\to-{}^1/_2[(\mathbf{qr}_1)^2+(\mathbf{qr}_2)^2]=-(\mathbf{qr})^2/4,$$

where we have introduced the relative radius vector $\mathbf{r}=2\mathbf{r}_1=-2\mathbf{r}_2$ for the two particles. Averaging (12) over the directions of $\mathbf{q}$ with the aid of the usual relation

$$\overline{q_iq_kq_lq_m}=q^4(\delta_{ik}\delta_{lm}+\delta_{il}\delta_{km}+\delta_{im}\delta_{kl})/15,$$

and allowing for the foregoing, we obtain in place of (13) the expression

$${}^1/_{60} q^4 N^2 |\langle n|x_ix_k|m\rangle|^2. \tag{16}$$

For the matrix element we have, as before,

$$\langle n|x_ix_k|m\rangle=\int_{-\infty}^{\infty} x_ix_k e^{i\Omega t}\,dt$$
$$=\frac{1}{i\Omega^3}\int_{-\infty}^{\infty}\frac{d^3}{dt^3}(x_ix_k)e^{i\Omega t}\,dt\approx\frac{2}{i\Omega^3}\Delta(v_iv_k).$$

Here $\mathbf{v}$ is the relative velocity of the atoms and $\Delta(v_iv_k)=v_i'v_k'-v_i^0v_k^0$ is the change that occurs in the tensor v_iv_k during scattering. We took into account the fact that when the atoms are far apart, $x_i=v_it$. Substituting into (16), we reduce this expression to the form

$$\frac{1}{15}\frac{q^4v^4N^2}{\Omega^6}(1-\cos 2\vartheta). \tag{17}$$

Now comparing the formulas (15) and (17), we arrive at the conclusion that the required expression for $S(\Omega,q)$ can be obtained from Eq. (7) by making the substitution

$$N_eN_i(1-\cos\vartheta)\to\frac{1}{10}N^2\frac{q^2v^2}{\Omega^2}(1-\cos 2\vartheta).$$

Thus, the final expression for $S(\Omega,q)$ has the form

$$S(\Omega,q)=\frac{1}{15}\frac{N^2q^4}{\Omega^6}\langle v^5\sigma^{**}(v)\rangle, \tag{18}$$

where

$$\sigma^{**}(v)=\int(1-\cos 2\vartheta)\,d\sigma.$$

The scattering cross section is proportional to Ω^{-6}, as it should be. Let us emphasize that, in the case of neutron scattering, the expression for the momentum transfer does not contain any relativistically small factor, so that the effect may not be small. We see that the observation of neutron scattering in this region provides direct information about the atom-atom scattering cross section.

To conclude, we discuss the scattering of neutrons in a mixture of two monatomic gases. We can easily show by carrying out calculations similar to those that led to (15) that the scattering probability can be obtained from Eq. (9) by making the substitution

$$\frac{a^2}{M^2}S(\Omega,q)\to\frac{2}{3}\mu^2\left(\frac{a_1}{M_1m_1}-\frac{a_2}{M_2m_2}\right)^2 N_1N_2\frac{q^2}{\Omega^4}\langle v^3\sigma_{12}^*(v)\rangle,$$

where μ is the reduced mass of the atoms of the first and second gases, M_1 is the reduced mass for an atom of the first gas and a neutron, m_1 is the mass of an atom of the first gas, etc. As it should be, the probability in this case is proportional to Ω^{-4}.

The authors are grateful to A. Ya. Shul'man, a discussion with whom motivated us to undertake this investigation. We are also grateful to A. F. Andreev and Ya. B. Zel'dovich for a discussion for the results obtained.

[1)] To obtain (10) from (3), it is sufficient to represent $\hat N(\mathbf{r},t)$ in the form

$$\hat N(\mathbf{r},t)=\sum_a\delta(\mathbf{r}-\mathbf{r}_a(t)). \tag{11}$$

[2)] We need to impose the condition (14) in order to be able to solve the problem in the general form without specifically computing the matrix elements. As to the condition $\hbar\Omega\ll T$ adopted earlier, in the majority of cases it is a consequence of (14). Only in the case of very low temperatures, when the de Broglie wavelength is much greater than the atomic dimensions, can we violate this condition without violating (14). In this case the atom-atom scattering cross section does not depend on the energy of the atoms, and it is again possible to derive a general relation. We shall, however, not dwell on this here.

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Translated by A. K. Agyei