Depolarized Rayleigh scattering in helium II

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Depolarized Rayleigh scattering in helium due to the fluctuations of the phonon distribution function is considered. The spectral and total extinction coefficients are calculated in the case of scattering of a linearly polarized wave.

In Rayleigh light scattering in liquids and dense gases, the narrow "unshifted" line that appears as the result of fluctuations of thermodynamic quantities, with "wavelengths" of the order of the wavelength of the incident light, has been well studied theoretically. The much more diffuse wing of the Rayleigh line contains, in addition, a scalar part also a symmetric part and corresponds to scattering from anisotropy fluctuations, elastic strains, and so on. The corresponding (Maxwell, Debye) relaxation times cannot be calculated phenomenologically, and require consideration of the motion of the particles over atomic distances and introduction of model representations.

The scalar scattering in superfluid ^{4}He with formation of scattering doublets from first and second sound has been well studied.[1,2] As is shown in the present work, a complete description of the symmetric part is also possible in ^{4}He. Such scattering arises because of fluctuations of the distribution function of phonons with wavelength less than the wavelength of the light, but larger than the interatomic distance.[3,4] Thus, the problem arises of the description of the relaxation of the fluctuations in the phonon gas. The situation is complicated by the existence of several relaxation processes. In the case $s\tau_\parallel > \lambda/2\sin(\theta/2)$ (s is the speed of sound, $\tau_\parallel$ is the characteristic time of longitudinal relaxation, λ is the wavelength of light, θ is the scattering angle), we have purely Doppler collisionless broadening. In the opposite limiting case, a state of incomplete thermodynamic equilibrium is realized. Equalization of the values of the phonon temperature and the velocity, which correspond to different directions in **k** space, is effected by the superdiffusion operator.[5] It is easy to estimate the values of the scattering angles corresponding to the different directions. Thus, at $\lambda\sim 5\times 10^{-5}$ cm and $T\sim 0.6$ K we have $\tau_\parallel^{-1}\sim 5\times 10^{7}\,\mathrm{s}^{-1}$, which gives the "limiting" value of the angle $\theta_\Gamma = 2\arcsin(\lambda/2s\tau_\parallel)\sim 5°$. Thus, the "superdiffusion" relaxation takes place in a narrow cone near scattering at zero angle.

1. Having the aim of calculating the spectral extinction coefficients in the case of superdiffusion relaxation, we determine the time correlator of the generalized temperature. As has alrady been shown,[5] under normal pressures, in the region of phonon temperatures, the transverse relaxation is due to processes of three-phonon collisions and is described by a differential equation of fourth order. We introduce a random force in it. We start out from the kinetic equation for the phonon distribution function with a random external force y_k:

$$\frac{\partial N_k}{\partial t}+\frac{\partial \omega_k}{\partial k_i}\frac{\partial N_k}{\partial r_i}-\frac{\partial \omega_k}{\partial r_i}\frac{\partial N_k}{\partial k_i}=I\{N_k\}+y_k. \tag{1}$$

We multiply (1) by $sk^3dk/(2\pi)^3$ and, using the phonon of the equilibrium distribution function with respect to directions in **k** space, we integrate over dk. Neglecting the dependence of the speed of sound on the coordinates, we obtain

$$\frac{\partial E}{\partial t}+sn_i\frac{\partial E}{\partial r_i}=(2\hat{l}_i\hat{l}_jM\hat{l}_i\hat{l}_j-\hat{l}^2M\hat{l}^2)\frac{1}{\Theta}+Y. \tag{2}$$

Here we have "redefined" the random force

$$Y=\int y_k sk^3\,dk/(2\pi)^3$$

and the following notation is used: E is the angular density of phonon energy, Θ is the generalized temperature, n_i is a unit vector, $l_i = e_{ijm}n_j\partial/\partial n_m$ is the operator of an infinitely small rotation, and M is a parameter dependent on the temperature. We shall need the linearized equation (2). We represent the generalized temperature in the form

$$\Theta=T(1+Z(\mathbf{n})),$$

where T is the equilibrium phonon temperature, Z is a dimensionless function of the direction. Denoting by E_0 and M_0 the values of the phonon energy and of parameter, determined at the equilibrium temperature T, we obtain

$$4E_0\left\{\frac{\partial Z}{\partial t}+sn_i\frac{\partial Z}{\partial r_i}\right\}=-\frac{M_0}{T}\hat{l}^2(\hat{l}^2+2)Z+Y. \tag{3}$$

Following the general theory of quasistationary fluctuations, we represent the rate of increase of the entropy density of the phonon system as a quadratic function of the quantities Y and Z. The change in the entropy density as a consequence of collisions is

$$\frac{d\sigma}{dt}=\int(I\{N_k\}+y_k)\ln\frac{1+n_k}{n_k}\,d\tau. \tag{4}$$

The symbol $d\tau$ denotes integration over phase space. Separating in the integration over $d\tau$ the integrations over the modulus of the wave vector and over the solid angle do, and taking into account, as in (1), the equilibrium of the distribution function along the direction in **k** space, we have

$$\frac{d\sigma}{dt}=\int\frac{1}{\Theta}(2\hat{l}_i\hat{l}_jM\hat{l}_i\hat{l}_j-\hat{l}^2M\hat{l}^2)\frac{1}{\Theta}\,do+\int Y\frac{do}{\Theta}. \tag{5}$$

With account of (4), and after linearization, we have

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$$\frac{d\sigma}{dt}=\frac{M_0}{T}\int Z\hat{l}^2(\hat{l}^2+2)Z\,do-\frac{1}{T}\int YZ\,do. \qquad (6)$$

By virtue of the spherical symmetry of the problem it is convenient to carry out an expansion in spherical functions. We are interested in fluctuations which do not change the mean temperature and velocity of the phonon system. Then the expansion of Y and Z begins with the second spherical harmonic

$$Z=\sum_{l=2}^{\infty}\sum_{m=-l}^{l}A_l{}^mY_l{}^m,\qquad Y=\sum_{l=2}^{\infty}\sum_{m=-l}^{l}B_l{}^mY_l{}^m, \qquad (7)$$

Y_l^m is the spherical harmonic.[6]

Substituting (7) in (6), we obtain

$$\frac{d\sigma}{dt}=\frac{M_0}{T^2}\sum_{l,l'}\sum_{m,m'}A_l{}^mA_{l'}^{m'}\int Y_l{}^m\hat{l}^2(\hat{l}^2+2)Y_{l'}^{m'}do$$
$$-\frac{1}{T}\sum_{l,l'}\sum_{m,m'}A_l{}^mB_{l'}^{m'}\int Y_l{}^mY_{l'}^{m'}\,do. \qquad (8)$$

With account of the orthogonality conditions

$$\int Y_l{}^mY_{l'}{}^{m'}\,do=2\pi\frac{1+\delta_{0m}}{2l+1}\frac{(l+|m|)!}{(l-|m|)!}\delta_{ll'}\delta_{mm'}$$

we have

$$\frac{d\sigma}{dt}=\sum_{l=2}^{\infty}\sum_{m=-l}\frac{2\pi(1+\delta_{0m})}{2l+1}\frac{(l+|m|)!}{(l-|m|)!}\frac{A_l{}^m}{T}\left\{\frac{M_0LA_l{}^m}{T}-B_l{}^m\right\}, \qquad (9)$$

where $L=l(l+1)(l^2+l-2)$ is the eigenvalue of the superdiffusion operator. In fluctuation theory[7] the increment in the entropy is customarily represented in the form

$$\frac{d\sigma}{dt}=-\sum_a\dot{x}_aX_a, \qquad (10)$$

where the quantities $\dot{x}_a$ and X_b are connected by the equation

$$\dot{x}_a=-\sum_b\gamma_{ab}X_b+y_a \qquad (11)$$

with the correlation function for the random force

$$\langle y_ay_b\rangle=\gamma_{ab}+\gamma_{ba}. \qquad (12)$$

Comparing Eqs. (10–12) with (9), we have

$$\dot{x}_l{}^m=\frac{M_0}{T}LA_l{}^m-B_l{}^m, \qquad (13)$$

$$X_l{}^m=\frac{2\pi(1+\delta_{0m})}{2l+1}\frac{(l+|m|)!}{(l-|m|)!}\frac{A_l{}^m}{T}, \qquad (14)$$

$$\dot{x}_l{}^m=\left\{\frac{2l+1}{2\pi(1+\delta_{0m})}\frac{(l-|m|)!}{(l+|m|)!}\right\}M_0LX_l{}^m+B_l{}^m, \qquad (15)$$

$$\langle B_l{}^mB_{l'}^{m'}\rangle=\frac{2l+1}{\pi(1+\delta_{0m})}\frac{(l-|m|)!}{(l+|m|)!}M_0L\delta_{ll'}\delta_{mm'}. \qquad (16)$$

As was explained above, of practical interest in our case is Rayleigh scattering at angles close to zero. This corresponds to the calculation of $\langle A_l^mA_{l'}^{m'}\rangle$ for the spatially homogeneous problem. The spherical harmonics are the eigenfunctions of Eq. (2), and the calculations are considerably simplified. We multiply (2) by Y_l^m and integrate over the solid angle

$$4E_0\frac{\partial A_l{}^m}{\partial t}=-\frac{M_0}{T}LA_l{}^m+B_l{}^m. \qquad (17)$$

Taking the Fourier transform, we obtain for the spectral correlator

$$\langle A_l{}^mA_{l'}^{m'}\rangle_\omega=\frac{\langle(B_l{}^m)^2\rangle_\omega\delta_{ll'}\delta_{mm'}}{(M_0L/T)^2+(4E_0\omega)^2}, \qquad (18)$$

where ω is the frequency of the fluctuation wave. The combination of (16) and (18) yields

$$\langle A_l{}^mA_l{}^m\rangle_\omega=\frac{2l+1}{\pi(1+\delta_{0m})}\frac{(l-|m|)!}{(l+|m|)!}\frac{T^2}{M_0L}\times\left\{1+\left(\frac{4E_0\omega T}{M_0L}\right)^2\right\}^{-1}. \qquad (19)$$

Equation (19) indicates that upon establishment of complete thermodynamic equilibrium in the considered case, each spherical harmonic relaxes independently with its own relaxation time.

We make use of the expression for the fluctuation increment obtained in Ref. 4 to the dielectric tensor:

$$\delta\varepsilon_{ij}=-\frac{\rho}{\varepsilon}\left(\frac{\partial\varepsilon}{\partial\rho}\right)^2\int d\tau\frac{k_ik_j}{\omega_k}N_k, \qquad (20)$$

where ρ is the density of the liquid, ε is the scalar dielectric constant of helium. We write out the tensor product and average over the fluctuations:

$$\langle\delta\varepsilon_{ij}\,\delta\varepsilon_{lm}\rangle=\frac{\rho^2}{\varepsilon^2}\left(\frac{\partial\varepsilon}{\partial\rho}\right)^4\left\langle\int d\tau\,d\tau'\frac{k_ik_jk_l{}'k_m{}'}{\omega_k\omega_{k'}}\delta N_k\,\delta N_{k'}\right\rangle. \qquad (21)$$

Here δN_k is the deviation of the distribution function from its equilibrium value, which can be expressed in terms of the function $Z(\mathbf{n})$:

$$\delta N_k=-\frac{\partial N_k}{\partial\omega_k}\omega_kZ(\mathbf{n}), \qquad (22)$$

which gives

$$\langle\delta\varepsilon_{ij}\,\delta\varepsilon_{lm}\rangle=\frac{9\rho_n{}^2\rho^2}{\varepsilon^2}\left(\frac{\partial\varepsilon}{\partial\rho}\right)^4\left\langle\int do\,do'\,n_in_jn_l{}'n_m{}'Z(\mathbf{n})Z(\mathbf{n}')\right\rangle \qquad (23)$$

Forward scattering with change in the polarization is described by the $xyxy$ component of the tensor. Carrying out the expansion of $Z(\mathbf{n})$ in spherical functions and integrating with account of the orthogonality condition

$$\int n_yn_x\sum_{l,m}A_l{}^mY_l{}^m\,do=\frac{8\pi}{5}A_2{}^2$$

we obtain (performing the Fourier transformation)

$$\langle(\delta\varepsilon_{xy})^2\rangle=\frac{576}{25}\pi^2\frac{\rho_n{}^2\rho^2}{\varepsilon^2}\left(\frac{\partial\varepsilon}{\partial\rho}\right)^4\langle(A_2{}^2)^2\rangle. \qquad (24)$$

We note that, although introduction of an infinite set of relaxation times is necessary for the calculation of the damping of an arbitrary perturbation, forward scattering "picks out" the relaxation of only the second spherical harmonic. The spectral extinction coefficient at a specified angle, R_ω, is given by the formula

$$R_\omega=\frac{1}{16\pi^2}\left(\frac{\Omega}{c}\right)^4 a_i a_l b_j b_m\langle\delta\varepsilon_{ij}\,\delta\varepsilon_{lm}\rangle_{\omega,q}, \tag{25}$$

where the unit vectors **a** and **b** indicate the directions of polarization of the incident and scattered waves, respectively. In our case, (25) is rewritten in the form

$$R_\omega=\frac{1}{16\pi^2}\left(\frac{\Omega}{c}\right)^4\langle(\delta\varepsilon_{xy})^2\rangle_\omega \tag{26}$$

and, substituting Eq. (19) at $l=m=2$, $L=24$ in (24), and also in (26), we finally obtain

$$R_\omega=\frac{1}{80\pi}\left(\frac{\Omega}{c}\right)^4\left(\frac{\partial\varepsilon}{\partial\rho}\right)^4\frac{\rho_n^2\rho^2T^2}{M_0\varepsilon^2}\left\{1+\left(\frac{E_0T\omega}{6M_0}\right)^2\right\}^{-1}. \tag{27}$$

In liquid helium, the dielectric constant is close to unity and the derivative $\partial\varepsilon/\partial\rho$ is equal to $(\varepsilon-1)/\rho$ with a high degree of accuracy. Using this together with the thermodynamic expression for the phonon energy in terms of the density of the normal component, we obtain

$$R_\omega=\frac{1}{80\pi}\left(\frac{\Omega}{c}\right)^4(\varepsilon-1)^4\frac{\rho_n^2T^2}{M_0\rho^2}\left\{1+\left(\frac{\rho_nTs^2\omega}{8M_0}\right)^2\right\}^{-1}, \tag{28}$$

and also the extinction coefficient integrated over the frequencies,

$$R=\frac{1}{20\pi}\left(\frac{\Omega}{c}\right)^4(\varepsilon-1)^4\frac{\rho_nT}{\rho^2s}. \tag{29}$$

It is not difficult to carry out the numerical estimate of the resultant expression. Thus, for characteristic light wavelengths $\sim$5000 Å and phonon temperatures $\sim$0.6 K, the dielectric constant has the value $\varepsilon-1\sim0.05$, the normal density is $\sim1\times10^{-6}$ g/cm^3. This leads to the value $R\sim0.5\times10^{-12}$ cm^{-1}.

2. In the collisionless limit $s\tau_\parallel>\lambda/2\sin(\theta/2)$ weakly attenuated fluctuation waves are formed by the phonons moving at different angles with respect to the wave vectors of the waves. It is clear that these waves have different phase velocities. The electromagnetic waves, being scattered independently by the various fluctuation waves, obtain different frequency shifts as a result of the Doppler effect. It is easy to calculate the scattering intensity as a function of the frequency. In the collisionless case, the perturbation of the phonon distribution function propagates with the speed of sound, whence it follows that the Fourier component of the time correlation function has the following form

$$\langle\delta n_k\delta n_{k'}\rangle_{\omega,q}=2\pi N_k(N_k+1)\,\delta(k-k')\,\delta(\omega-s\mathbf{qn}), \tag{30}$$

where ω, $q=(2/\lambda)\sin(\theta/2)$ are the frequency and the wave vector of the fluctuation wave. Substituting this expression in the Fourier component (21) and carrying out integration over the wave vector of the phonons, we obtain

$$\langle\delta\varepsilon_{ij}\,\delta\varepsilon_{lm}\rangle=\frac{6\pi\rho^2\rho_nT}{\varepsilon^2s^2}\left(\frac{\delta\varepsilon}{\partial\rho}\right)^4F(\mathbf{q},\omega), \tag{31}$$

where the symbol $F(\mathbf{q},\omega)$ denotes a symmetric tensor of fourth rank, which depends on the wave vector and the frequency of the fluctuation wave:

$$F(\mathbf{q},\omega)=\left\langle\int do\left\{n_in_jn_ln_m\delta(\omega-s\mathbf{qn})\right\}\right\rangle. \tag{32}$$

The presence of the δ function in the integrand means that the averaging over the solid angle reduces to averaging over the circle formed by the intersection of the unit sphere and the plane $(\omega-s\mathbf{q}\cdot\mathbf{n})=0$. It is convenient to represent the vector **n** in the form of the sum of two vectors parallel and perpendicular to the direction **q**:

$$\mathbf{n}=\frac{\omega}{sq}\mathbf{e}+\left[1-\left(\frac{\omega}{sq}\right)^2\right]^{1/2}\mathbf{e}_\perp, \tag{33}$$

where **e** and $\mathbf{e}_\perp$ are unit vectors that are respectively parallel and perpendicular to **q**. After rather cumbersome calculations, substituting (33) and (32) and using the formulas for averaging the products of unit vectors perpendicular to a given vector,

$$\langle e_{\perp i}e_{\perp j}\rangle={}^1/_2\delta_{ij}^\perp={}^1/_2(\delta_{ij}-e_ie_j),$$

$$\langle e_{\perp i}e_{\perp j}e_{\perp l}e_{\perp m}\rangle={}^1/_8(\delta_{ij}^\perp\delta_{km}^\perp+\delta_{ik}^\perp\delta_{jm}^\perp+\delta_{im}^\perp\delta_{jk}^\perp),$$

we obtain

$$\begin{aligned}F(\mathbf{q},\omega)=\frac{\pi}{4sq}\Bigg\{&\left(1-\left(\frac{\omega}{sq}\right)^2\right)^2(\delta_{ij}\delta_{lm}+\delta_{il}\delta_{jm}+\delta_{im}\delta_{jl})\\&+\left(3-30\left(\frac{\omega}{sq}\right)^2+35\left(\frac{\omega}{sq}\right)^4\right)e_ie_je_le_m\\&-\left(1+6\left(\frac{\omega}{sq}\right)^2-5\left(\frac{\omega}{sq}\right)^4\right)(\delta_{ij}e_le_k+\delta_{il}e_je_m+\delta_{im}e_je_l\\&+\delta_{jl}e_ie_m+\delta_{jm}e_ie_l+\delta_{lm}e_ie_j)\Bigg\}.\end{aligned} \tag{34}$$

This expression is, as it should be, symmetric relative to permutation of the indices. We then have for the spectral extinction coefficient (25), by transforming as in (28) the derivative of the dielectric constant of helium with respect to the density:

$$\begin{aligned}R_\omega=\frac{3}{32}\left(\frac{\Omega}{c}\right)^4\frac{T\rho_n}{\rho^2s^3q}(\varepsilon-1)^4\Bigg\{&\left(1-\left(\frac{\omega}{sq}\right)^2\right)^2(1+2(\mathbf{ab})^2)\\+&\left(3-30\left(\frac{\omega}{sq}\right)^2+35\left(\frac{\omega}{sq}\right)^4\right)(\mathbf{ae})^2(\mathbf{be})^2-\left(1-6\left(\frac{\omega}{sq}\right)^2\right.\\&\left.+5\left(\frac{\omega}{sq}\right)^4\right)((\mathbf{be})^2+(\mathbf{ae})^2+4(\mathbf{ae})(\mathbf{be})(\mathbf{ab}))\Bigg\}.\end{aligned} \tag{35}$$

For greater clarity, we write out the obtained expression in a chosen set of coordinates. Let the wave vector of the incident wave **k** be directed along the z axis. The wave vector of the scattered wave **k′** makes the angles φ, ψ, θ with the axes z, y, z, respectively. (The chosen angles are not independent but satisfy the relation $\cos^2\varphi+\cos^2\psi+\cos^2\theta=1$.) The polarization vector of the incident wave is directed along the x

axis, that of the scattered wave is perpendicular to the plane formed by the vectors **k**′ and **a** (the appearance of just this component of the polarization is a characteristic of symmetric scattering and is convenient for experimental verification). Thus the unit vectors have the coordinates

$$\mathbf{a}=(1,0,0),$$
$$\mathbf{e}=\left(\cos\frac{\theta}{2}\frac{\cos\varphi}{\sin\theta},\ \cos\frac{\theta}{2}\frac{\cos\psi}{\sin\theta},\ -\sin\frac{\theta}{2}\right),\tag{36}$$
$$\mathbf{b}=\left(0,\ \frac{\cos\theta}{\sin\varphi},\ -\frac{\cos\psi}{\sin\varphi}\right),$$

which gives

$$R_\omega=\frac{3}{32}\left(\frac{\Omega}{c}\right)^4\frac{T\rho_n}{\rho^2s^3q}(\varepsilon-1)^4\left\{\left(1-\left(\frac{\omega}{sq}\right)^2\right)^2\right.$$
$$+\left(3-30\left(\frac{\omega}{sq}\right)^2+35\left(\frac{\omega}{sq}\right)^4\right)\frac{\cos^2\psi\operatorname{ctg}^2\varphi}{16\sin^4(\theta/2)}$$
$$\left.-\left(1-6\left(\frac{\omega}{sq}\right)^2+5\left(\frac{\omega}{sq}\right)^4\right)\frac{\sin^2 2\varphi+4\cos^2\psi}{16\sin^2\varphi\sin^2(\theta/2)}\right\}\tag{37}$$

In the particular case of scattering in the direction of the vector **a**.

$$R_\omega=\frac{3}{32}\left(\frac{\Omega}{c}\right)^4\frac{T\rho_n}{\rho^2s^3q}(\varepsilon-1)^4\left(1-\left(\frac{\omega}{sq}\right)^2\right)^2\ .\tag{38}$$

Thus, in the considered collisionless limit, the entire depolarized scattering is in the frequency range $|\omega|\leqslant(2s/\lambda)\sin(\theta/2)$. For the calculation of the integrated extinction coefficient, we integrate (37) with respect to the frequency. The terms containing the angular dependence drop out, as should be the case for symmetric scattering, and we get back to Eq. (29).

In the roton temperature region, the following formula, which is similar to (20), can be used

$$\delta\varepsilon_{ij}=-\frac{(\varepsilon-1)^2}{\rho}a\int d\tau n_in_jN_k,\tag{39}$$

where $\mathbf{n}=\mathbf{k}/k$, N_k is the roton distribution function, and a is some phonomenological constant. Similarly to the derivation in the phonon range of temperatures, we get from (39) for the simultaneous correlation of the permittivity tensor

$$\langle\delta\varepsilon_{ij}\,\delta\varepsilon_{lm}\rangle=(\varepsilon-1)^4\frac{a^2T\rho_n}{5\rho^2p_0^2}(\delta_{ij}\delta_{lm}+\delta_{il}\delta_{jm}+\delta_{im}\delta_{jl}),\tag{40}$$

where ρ_n is the roton normal density, p_0 is the characteristic momentum of the rotons. In the case of symmetric scattering, the integrated extinction coefficient is

$$R=\frac{1}{80\pi^2}\left(\frac{\Omega}{c}\right)^4(\varepsilon-1)^4\frac{a^2T\rho_n}{\rho^2p_0^2}.\tag{41}$$

It is not difficult to estimate (41) by taking it into account that, in order of magnitude, $a\sim p_0^2/\Delta$, where Δ is the characteristic energy of the rotons. Then, in the case $T\sim1$ K, $\rho_n\sim\rho$, $\Omega/c\sim10^5$ cm^{-1}, $\varepsilon-1\sim0.05$, we get $R\sim10^{-9}$ cm^{-1}.

As has been shown, the distribution-function fluctuations lead to the appearance of a depolarization part in the scattered light. The obtained spectral extinction coefficients do not contain such vaguely defined quantities as $\tau_\parallel$ and $\tau_\perp$, so that an accurate comparison with experiment is possible. Evidently the value of the extinction coefficient in the region of phonon temperatures lies at the borderline of experimental possibility, whereas the extinction coefficient in the roton region can be measured.

It should also be noted that a mechanism similar to that considered leads to the appearance of a depolarization component even in the scattering of a linearly polarized wave from acoustical phonons in solids.

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