Elementary Quantum Theory of Light Scattering in Liquid Helium

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The usual formalism of second quantization is generalized to include the case in which liquid helium contains atoms in electronic excited states. The formalism is applied to sccattering of the photon, where the dipolar coupling between excited and normal atoms is included as perturbation. The resulting cross sections for Brillouin and Raman scattering agree with those obtained by the semi-phenomenological theory of Stephen.

The relation with the recent theory of Iwamoto is also discussed in terms of Green functions.

§ 1. Introduction

With use of the laser, it is now experimentally possible to observe scattering of light in a liquid of small polarizability. Both liquid He^4 and liquid He^3 are of particular interest as one can apply a simple quasiparticle picture to each of them at low temperature. As was emphasized by Halley,[1] scattering of light will provide new information about elementary excitations in these liquids.

When a single excitation is created by scattering, its momentum and therefore its energy should be small (Brillouin scattering), since the wave-length of light is much longer than the interatomic distance. As Halley pointed out, however, two excitations of almost equal but opposite momenta can also be created, so that their energies need not be small in comparison with the characteristic energy of the atomic motion in the liquid. In the case of superfluid He^4, this means the creation of two rotons of almost equal but opposite momenta. Raman scattering of this type has been observed indeed by Greytak and Yan.[2]

A theoretical scheme to deal with Raman scattering in liquids has been given by Stephen,[3] who has extended ideas of the Lorentz-Lorenz theory of dispersion to include scattering from the density fluctuation. The relation between the electric field in the medium and the field of incident light is a nonlinear function of the density. When expanded in powers of the density fluctuation, the linear term gives rise to Brillouin scattering, whereas the quadratic term corresponds to scattering from a pair of excitations.

In the present paper, scattering of light in liquid helium will be formulated as quantum mechanical scattering processes. We ignore the multiple scattering of photons and concentrate ourselves on simple processes in which one photon

is absorbed and another photon is emitted. When absorption and emission are assumed to occur on a single atom, we obtain Brillouin scattering which corresponds to the linear term in Stephen's expansion in powers of the density fluctuation. In order to have terms of higher order, we need to take into account the possibility of excitonic motion of the Frenkel type. The excited state can move from one atom to another through the dipolar interaction as is well known in the theory of Frenkel excitons in crystals.[4] The dipolar interaction is tensorial, so that it can change the direction of polarization in accordance with the depolarization effect of Raman scattering observed by Greytak and Yan. Stephen's expansion corresponds to the perturbational expansion with respect to the dipolar interaction.

In dealing with quantum liquids, it is convenient as usual to make use of the second quantization method. The method is of course familiar when applied to liquid helium consisting of atoms all in the lowest electronic state. In the present paper, we shall first generalize the method to include the liquid containing excited atoms.

§ 2. Second quantization of a liquid containing excited atoms

It is almost trivial to generalize the usual method of second quantization to include liquid helium containing atoms in excited electronic states. Since no explicit derivation is given in standard books, we shall briefly describe it here.

We start from the description in configuration space. Suppose that the liquid contains N atoms and $2N$ electrons. Let $\boldsymbol{R}_1, \cdots, \boldsymbol{R}_N$ be nuclear coordinates including nuclear spin coordinates in the case of He^3. Let $\xi_1 \eta_1, \cdots, \xi_N \eta_N$ be electronic coordinates including spin coordinates. We write the time-dependent Schrödinger equation as

$$ i \frac{\partial \Psi}{\partial t} = (K_n + K_e + U)\Psi . \tag{1} $$

Here K_n and K_e are nuclear and electronic kinetic energies, respectively, and U is the whole interaction potential. We have taken $\hbar = 1$. We are going to integrate Eq. (1) over electronic coordinates by introducing the orthonormalized set of eigenfunctions χ_J of the Hamiltonian $K_e + U$ for the system with fixed nuclei.

$$ \Psi(\xi \eta R) = \sum_J \Phi(RJ) \chi_J(\xi \eta ; R) . \tag{2} $$

As usual, we apply the Born-Oppenheimer approximation and omit those terms in which K_n operates on χ_J. Then we obtain from (1)

$$ \left(i \frac{\partial}{\partial t} - K_n \right) \Phi(RJ) = \sum_{J'} U_{JJ'}(R) \Phi(RJ') , \tag{3} $$

$$U_{JJ'}(R) = \int d\xi d\eta \chi_J^*(K_e + U)\chi_{J'} . \qquad (4)$$

The diagonal element $U_{JJ}(R)$ is the so-called adiabatic potential.

In the case of liquid helium, electrons are well localized on each atom, so that we may assume the Heitler-London wave functions

$$\chi_J = \mathcal{A}\prod_{n=1}^{N} a_{jn}(\xi_n - R_n, \eta_n - R_n) . \qquad (5)$$

Here $a_j(\xi, \eta)$ is the wave function to describe the electronic state of an isolated atom, $j=0$ refers to the lowest s state and $j\neq0$ to excited states, and $\mathcal{A}$ means antisymmetrizing and normalizing the wave function with respect to electronic coordinates. We ignore overlap integrals between the a_j belonging to different atoms.

Let P be any permutation of $1\,2\cdots N$. We designate it as P_R when operating on $R_1, \cdots, R_N$ and as P_J when operating on $j_1, \cdots, j_N$. Clearly the operator $P_R P_J$ operating on (5) is equivalent to the inverse of P operating on electronic coordinates, so that $P_R P_J \chi = \chi$. On the other hand, $\varPsi$ on the left of (2) is either symmetric (He⁴) or antisymmetric (He³) against P_R. Hence

$$\pm \varPsi = \sum_J P_R \varPhi \cdot P_R \chi_J = \sum_J P_R P_J \varPhi \cdot P_R P_J \chi ,$$

so that

$$P_R P_J \varPhi = \pm \varPhi . \qquad (6)$$

Thus, in addition to nuclear coordinates $R_1, \cdots, R_N$, we have extra coordinates $j_1, \cdots, j_N$ which describe the internal electronic state of each atom. The wave function $\varPhi(R_1 j_1, \cdots, R_N j_N)$ in (3) should be either symmetric (He⁴) or antisymmetric (He³) under the permutation of atomic coordinates including j. It is then straightforward to transcribe the Hamiltonian in (3) into the second quantization scheme.

In what follows, we shall be concerned with the case of liquid He⁴ unless explicitly stated otherwise. Liquid He⁴ is described in terms of operators $\psi_j(R)$, and their hermitean conjugates $\psi_j^*(R)$, each of which is defined for each point R in space and for each value of the quantum number j. They satisfy the commutation rules

$$[\psi_j(R), \psi_i^*(R')] = \delta_{ji}\delta(R - R') ,$$

$$[\psi_j(R), \psi_i(R')] = [\psi_i^*(R), \psi_i^*(R')] = 0 . \qquad (7)$$

The operator $\psi_j(R)$ annihilates the atom in the j-th electronic state at point R in space and $\psi_j^*(R)$ is the corresponding creation operator. In the case of liquid He³, we should assume the anticommutation rules in place of the commutation rules.

The Hamiltonian in (3) can be expressed in terms of ψ-operators. For instance,

$$K_n = \sum_j \int d\boldsymbol{R} \frac{1}{2M} \frac{\partial \psi_j^*(\boldsymbol{R})}{\partial \boldsymbol{R}} \cdot \frac{\partial \psi_j(\boldsymbol{R})}{\partial \boldsymbol{R}}, \tag{8}$$

where M is the atomic mass. When all atoms are in the lowest electronic state, the diagonal element U_{00} in (3) gives the exchange repulsion between atoms. Let v be the potential of the usual interatomic force which is the sum of the exchange repulsion and the van der Waals attraction. Then

$$H_L = \int d\boldsymbol{R} \frac{1}{2M} \frac{\partial \psi_0^*}{\partial \boldsymbol{R}} \cdot \frac{\partial \psi_0}{\partial \boldsymbol{R}}$$

$$+ \frac{1}{2} \int\int d\boldsymbol{R}_1 d\boldsymbol{R}_2 v(R_{12}) \psi_0^*(\boldsymbol{R}_1) \psi_0^*(\boldsymbol{R}_2) \psi_0(\boldsymbol{R}_2) \psi_0(\boldsymbol{R}_1) \tag{9}$$

is the usual Hamiltonian of the liquid when there is no excited atom. Here $\boldsymbol{R}_{12} \equiv \boldsymbol{R}_1 - \boldsymbol{R}_2$. When a number of atoms are excited, we need first to include the excitation energy

$$H_A = \sum_{j \neq 0} E_j \int d\boldsymbol{R} \psi_j^*(\boldsymbol{R}) \psi_j(\boldsymbol{R}), \tag{10}$$

where E_j are electronic excitation energies of an isolated atom.

As is well known in the theory of the Frenkel exciton in solids, the excited state can move from one atom to another through the dipolar interaction. Hereafter, for the sake of simplicity, we take into consideration only first p excited states and denote their excitation energy by ω_0. We choose p state wave functions which have the same rotational symmetry as space coordinates $\mu = x, y, z$, so that the matrix elements of the electric dipole moment β_μ of an atom are real.

$$\beta = \int d\xi d\eta\, a_\mu^* \beta_\mu a_0.$$

Then the polarizability of a single atom is given by

$$\alpha = \frac{2\beta^2}{\omega_0}. \tag{11}$$

With use of this expression, the dipolar coupling can be written as

$$H_d = \frac{\alpha \omega_0}{4} \int\int d\boldsymbol{R}_1 d\boldsymbol{R}_2 \sum J_{\mu\nu}(\boldsymbol{R}_{12}) [2\psi_\mu^*(\boldsymbol{R}_1) \psi_0(\boldsymbol{R}_1) \psi_0^*(\boldsymbol{R}_2) \psi_\nu(\boldsymbol{R}_2)$$

$$+ \psi_\mu^*(\boldsymbol{R}_1) \psi_0(\boldsymbol{R}_1) \psi_\nu^*(\boldsymbol{R}_2) \psi_0(\boldsymbol{R}_2) + \psi_0^*(\boldsymbol{R}_1) \psi_\mu(\boldsymbol{R}_1) \psi_0^*(\boldsymbol{R}_2) \psi_\nu(\boldsymbol{R}_2)]. \tag{12}$$

Here

$$J_{\mu\nu}(\boldsymbol{R}) = \frac{1}{R^3} \left[\delta_{\mu\nu} - \frac{3}{R^2} R_\mu R_\nu \right]. \tag{13}$$

The dipolar approximation breaks down at short distances, where we need to deal with the full Coulomb interaction quantum mechanically. Instead of going into the detail, however, we take account of the short range correlation simply by cutting off (13) as $J_{\mu\nu}(\boldsymbol{R}) = 0$ for $R \lesssim 2a$, where a is the size of the atom.

As is well known, the virtual excitation caused by the second and third terms in the square bracket of (12) results in the van der Waals attraction between atoms.[4] As we shall see later on, it is essential to retain these terms in order to obtain the right expression for Raman scattering.

§ 3. Interaction with radiation and Brillouin scattering

The Hamiltonian of the interaction between atoms and radiation consists of two parts: one is linear and the other is quadratic with respect to the electromagnetic vector potential.

$$H_1 = \sum \left(\frac{\pi \alpha \omega_0^3}{Vck} \right)^{1/2} (b_{k\lambda} + b^*_{-k\lambda}) \int dR \, e^{i\boldsymbol{k}\cdot\boldsymbol{R}} \, i(\psi_\lambda^*(\boldsymbol{R}) \psi_0(\boldsymbol{R}) - \text{h.c.}), \qquad (14)$$

$$H_2 = \sum \left(\frac{\pi \alpha}{Vc} \right) \frac{\omega_0^2}{(kl)^{1/2}} (b_{k\lambda} + b^*_{-k\lambda})(b_{-l\lambda} + b^*_{l\lambda}) \rho_{l-k}. \qquad (15)$$

Here V is the normalization volume, $b_{k\lambda}$, $b^*_{k\lambda}$ are destruction and creation operators of photons, and c is the velocity of light. We have taken the dipole approximation since the wave-length of light is much longer than the atomic size a. It should be noted that the number of electrons per atom which appears in the usual expression for H_2 has been expressed in terms of matrix elements of the dipole moment by the use of the so-called f-sum rule.[5] In (15),

$$\rho_q = \int dR \, e^{-i\boldsymbol{q}\cdot\boldsymbol{R}} \rho(\boldsymbol{R}) \qquad (16)$$

is the Fourier transform of the total density of atoms. Since we assume that the number of excited atoms is small, we replace the total density approximately by the density of normal atoms.

$$\rho(R) \simeq \psi_0^*(\boldsymbol{R}) \psi_0(\boldsymbol{R}). \qquad (17)$$

For the same reason, we shall ignore the interaction between excited atoms.

Thus the total Hamiltonian of our system has the form

$$H = H_0 + H',$$

$$H_0 = H_L + H_A + H_R, \qquad H' = H_1 + H_2 + H_d. \qquad (18)$$

Here $H_R = \sum ckb^*_{k\lambda}b_{k\lambda}$ is the Hamiltonian of free photons. Strictly speaking, we should add the kinetic energy of excited atoms, but this is so small in comparison with H_A that we have ignored it.

We now regard the atomic polarizability α as a small parameter and introduce

the perturbational expansion with respect to H'. More precisely, the dimensionless expansion parameter will be $n\alpha$, where n is the density of atoms in the liquid. Since we have already included the van der Waals interaction in H_L, we should subtract this interaction from H'. This is equivalent, however, to ignoring the van der Waals interaction which arises from second order perturbation of H_a.

In the interaction representation defined by the unperturbed Hamiltonian H_0, ψ_μ, $b_{k\lambda}$ are proportional to $\exp(-i\omega_0 t)$, $\exp(-ickt)$ respectively. As for the operator $\psi_0(\boldsymbol{R}t) = \exp(iH_L t)\psi_0(\boldsymbol{R})\exp(-iH_L t)$, we cannot give an explicit expression unless we solve the many-body problem of liquid helium by a certain approximate method.

Hereafter we are concerned with scattering processes in which the incident photon of momentum $\boldsymbol{k}$ and polarization λ is absorbed and the photon of momentum $\boldsymbol{k}'$ and polarization λ' is emitted. For simplicity, we take the liquid at zero temperature, so that at the remote past $t=-\infty$ the liquid is in the eigenstate Φ_0 which belongs to the lowest eigenvalue E_0 of H_L. We seek for the transition probability of finding the liquid at $t=+\infty$ in the eigenstate Φ_f which belongs to a certain eigenvalue E_f of H_L.

The simplest is the first order process caused by H_2. The so-called scattering matrix can be obtained by inserting (15) into the S-matrix

$$-i \int_{-\infty}^{\infty} H_2(t)\,dt\,.$$

The resulting scattering matrix is

$$-i\left(\frac{2\pi\alpha}{Vc}\right)\frac{\omega_0^2}{(kk')^{1/2}}\delta_{\lambda'\lambda}\langle\Phi_f|\rho_{k'-k}|\Phi_0\rangle\,. \tag{19}$$

We obtain a similar term from the second order processes in

$$(-i)^2 \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt_1 H_1(t)\,H_1(t_1)\,.$$

The resulting scattering matrix has the form

$$i\left(\frac{\pi\alpha}{Vc}\right)\cdot\frac{\omega_0^3}{(kk')^{1/2}}\int d\boldsymbol{R}_1 d\boldsymbol{R}_2\, e^{i(\boldsymbol{k}\cdot\boldsymbol{R}_1-\boldsymbol{k}'\cdot\boldsymbol{R}_2)}$$

$$\times\langle\Phi_f|\psi_0^*(\boldsymbol{R}_2)\psi_{\lambda'}(\boldsymbol{R}_2)\frac{1}{\omega_0+H_L-ck-E_0}\psi_\lambda^*(\boldsymbol{R}_1)\psi_0(\boldsymbol{R}_1)$$

$$+\psi_0^*(\boldsymbol{R}_1)\psi_\lambda(\boldsymbol{R}_1)\frac{1}{\omega_0+ck'+H_L-E_0}\psi_{\lambda'}^*(\boldsymbol{R}_2)\psi_0(\boldsymbol{R}_2)|\Phi_0\rangle\,. \tag{20}$$

The two terms may be represented by the graphs in Fig. 1, in which the dotted line represents the photon and the full line the excited atom.

Now H_L-E_0 in the denominator of (20) is the change to occur in the energy of the atomic motion in the liquid when one atom is electronically excited. Except
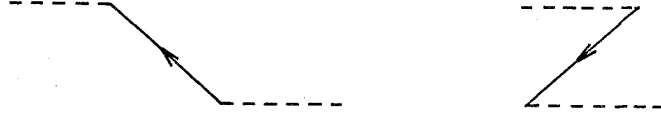
Fig. 1.

for the almost exact resonance $\omega_0 \cong ck$, it is so small compared with ω_0 that we may ignore it. Furthermore, since we have no excited atom in the state Φ_0,

$$\psi_\lambda(\mathbf{R})\psi_\mu^*(\mathbf{R}')\Phi_0 = \delta_{\lambda\mu}\delta(\mathbf{R}-\mathbf{R}')\Phi_0. \tag{21}$$

Finally, since all atoms go back to the lowest electronic state after scattering, the shift $\omega = ck - ck'$ in the photon energy should be equal to the energy change of the atomic motion in the liquid. Hence we may ignore ω in comparison with ck itself. Thus (20) may be replaced approximately by

$$i\frac{2\pi\alpha}{V}\cdot\frac{\omega_0^4}{\nu(\omega_0^2-\nu^2)}\delta_{\lambda'\lambda}\langle\Phi_f|\rho_{k'-k}|\Phi_0\rangle,$$

where $\nu = ck$. Adding (19), we obtain the scattering matrix for Brillouin scattering as

$$\langle f|T_1|0\rangle = \frac{2\pi i}{V}\delta_{\lambda'\lambda}\nu\alpha(\nu)\langle\Phi_f|\rho_{k'-k}|\Phi_0\rangle, \tag{22}$$

where

$$\alpha(\nu) = \alpha[1-(\nu/\nu_0)^2]^{-1} \tag{23}$$

is the atomic polarizability at the photon energy ν.

The differential cross section for the process, in which the photon with an energy between $ck' - d\omega$ and ck' is scattered into the element of solid angle $d\Omega$, is given by

$$\frac{d^2\sigma}{d\omega d\Omega} = \frac{V^2 k'^2}{(2\pi)^3 c^2}\sum_f 2\pi|\langle f|T|0\rangle|^2\cdot\delta(E_f-E_0-\omega). \tag{24}$$

In the case of (22), we have

$$\frac{d^2\sigma_1}{d\omega d\Omega} = \frac{\delta_{\lambda'\lambda}}{2\pi}\left(\frac{\nu}{c}\right)^4\alpha^2(\nu)S(q,\omega), \tag{25}$$

where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ is the momentum change of the photon and

$$S(q,\omega) = \int_{-\infty}^{\infty}\langle\Phi_0|\rho_q(t)\rho_{-q}(0)|\Phi_0\rangle, \tag{26}$$

$$\rho_q(t) = e^{iH_Lt}\rho_q e^{-iH_Lt}. \tag{27}$$

Our expression (25) agrees with the result obtained by Stephen when our $\alpha(\nu)$ is identified with his atomic polarizability α.

§ 4. Raman scattering of higher order

In the case of Brillouin scattering (25), absorption and emission of photons occur on a single atom, so that the direction of polarization does not change. In order to obtain the depolarization effect as was observed by Greytak and Yan, we need to take into consideration the dipolar interaction (12).

The lowest order contribution of H_d is contained in the S-matrix

$$(-i)^3 \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 [H_1(t) H_d(t_1) H_1(t_2)$$

$$+ H_1(t) H_1(t_2) H_d(t_2) + H_d(t) H_1(t_1) H_1(t_2)]. \tag{28}$$

Each of the three terms gives two scattering processes as shown by graphs in Fig. 2, in which the wavy line represents the dipolar interaction $\alpha \omega_0 J_{\mu\nu}$. It is
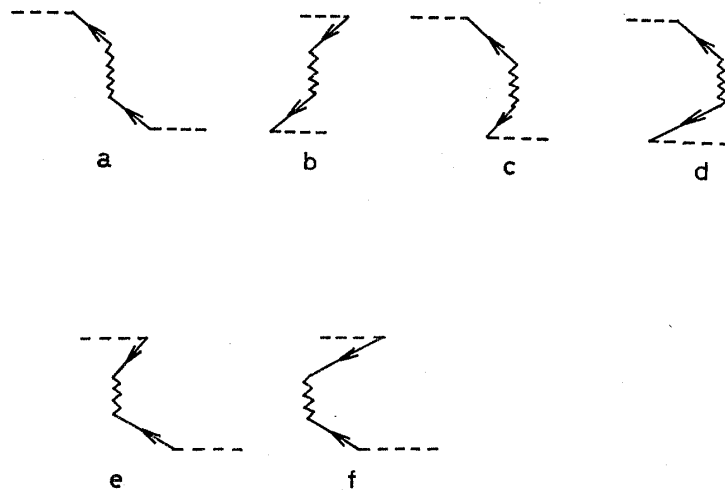

Fig. 2.

straightforward as before to carry out the time integral to find the scattering matrix. For instance, the graph a leads to the following scattering matrix.

$$-i \left(\frac{\pi \alpha^2}{2Vc} \right) \frac{\omega_0^4}{(kk')^{1/2}} \sum \int dR_1 \cdots dR_4 e^{i(k \cdot R_4 - k' \cdot R_1)} J_{\mu\nu}(R_{23})$$

$$\times \langle \Phi_f | \psi_0^*(R_1) \psi_{\lambda'}(R_1) \frac{1}{\omega_0 - ck - H_L + E_0} \psi_\mu^*(R_2) \psi_0(R_2)$$

$$+ \psi_0^*(R_3) \psi_\nu(R_3) \frac{1}{\omega_0 - ck - H_L + E_0} \psi_\lambda^*(R_4) \psi_0(R_4) | \Phi_0 \rangle.$$

We again ignore $H_L - E_0$ against ck in the denominator and make use of (21). Then the scattering matrix is simplified as

$$-i \left(\frac{\pi \alpha^2}{2V} \right) \frac{\omega_0^4}{\nu(\omega_0 - \nu)^2} \int dR_1 dR_2 e^{i(k \cdot R_2 - k' \cdot R_1)} J_{\lambda'\lambda}(R_{12}) \langle \Phi_f | \rho(R_1) \rho(R_2) | \Phi_0 \rangle. \tag{29}$$

Similarly the graph b leads to the scattering matrix, which we obtain from (29) by replacing $\omega_0 - \nu$ in the denominator by $\omega_0 + ck' \cong \omega_0 + \nu$.

On the other hand, the graphs c and d lead to the scattering matrix

$$i \left(\frac{\pi \alpha^2}{2Vc} \right) \frac{\omega_0^4}{(kk')^{1/2}} \cdot \frac{1}{2\omega_0} \left(\frac{1}{\omega_0 - ck} + \frac{1}{\omega_0 + ck} \right)$$

$$\times \int d\boldsymbol{R}_1 d\boldsymbol{R}_2 e^{i(\boldsymbol{k} \cdot \boldsymbol{R}_2 - \boldsymbol{k}' \cdot \boldsymbol{R}_1)} J_{\lambda'\lambda}(\boldsymbol{R}_{12}) \langle \Phi_f | \rho(\boldsymbol{R}_1) \rho(\boldsymbol{R}_2) | \Phi_0 \rangle .$$

When we replace $2\omega_0$ in the denominator by $2\omega_0 + c(k' - k) \cong 2\omega_0$, we obtain the scattering matrix corresponding to graphs e and f.

It is important that graphs c, d, e, f have the opposite sign and therefore tend to cancel graphs a, b. In fact, the sum of all these six graphs can be expressed in terms of (23) with no explicit dependence on ω_0 elsewhere. Thus the sum gives the scattering matrix

$$- i \left(\frac{2\pi}{V^2} \right) \nu \alpha^2(\nu) \sum_q L_{\lambda'\lambda}(\boldsymbol{q}) \langle \Phi_f | \rho_{\boldsymbol{k}'-\boldsymbol{q}} \rho_{\boldsymbol{q}-\boldsymbol{k}} | \Phi_0 \rangle , \tag{30}$$

where we have introduced

$$L_{\lambda'\lambda}(\boldsymbol{q}) = \int d\boldsymbol{R} e^{-i\boldsymbol{q} \cdot \boldsymbol{R}} J_{\lambda'\lambda}(\boldsymbol{R}) . \tag{31}$$

The sum over $\boldsymbol{q}$ in (30) contains the terms with $\boldsymbol{q} = \boldsymbol{k}$ and $\boldsymbol{q} = \boldsymbol{k}'$, which give

$$- i \left(\frac{2\pi}{V} \right) \nu \alpha(\nu) (n\alpha(\nu)) (L_{\lambda'\lambda}(\boldsymbol{k}) + L_{\lambda'\lambda}(\boldsymbol{k}')) \langle \Phi_f | \rho_{\boldsymbol{k}'-\boldsymbol{k}} | \Phi_0 \rangle .$$

This is the correction to the amplitude (22) of Brillouin scattering and may be ignored in the limit $n\alpha \ll 1$.

The genuine Raman amplitude of higher order is given by

$$\langle f | T_2 | 0 \rangle = - i \left(\frac{2\pi}{V^2} \right) \nu \alpha^2(\nu) \sum_q L_{\lambda'\lambda}(\boldsymbol{q}) \langle \Phi_f | \Delta\rho_{\boldsymbol{k}'-\boldsymbol{q}} \Delta\rho_{\boldsymbol{q}-\boldsymbol{k}} | \Phi_0 \rangle . \tag{32}$$

Here

$$\Delta\rho_q = \begin{cases} 0 , & q = 0 , \\ \rho_q , & q \neq 0 . \end{cases}$$

Inserting (32) in (24), we obtain the differential cross section

$$\frac{d^2\sigma_2}{d\omega d\Omega} = \left(\frac{\nu \alpha(\nu)}{c} \right)^4 \frac{1}{V^2} \sum_p \sum_q L_{\lambda'\lambda}(\boldsymbol{p}) L_{\lambda'\lambda}(\boldsymbol{q})$$

$$\times \int_{-\infty}^{\infty} \frac{dt}{(2\pi)} e^{i\omega t} \langle \Phi_0 | \Delta\rho_{\boldsymbol{k}-\boldsymbol{p}}(t) \Delta\rho_{\boldsymbol{p}-\boldsymbol{k}'}(t) \Delta\rho_{\boldsymbol{k}'-\boldsymbol{q}}(0) \Delta\rho_{\boldsymbol{q}-\boldsymbol{k}}(0) | \Phi_0 \rangle . \tag{33}$$

Thus the cross section is proportional to the Fourier transform of a four-body correlation function.

A simple, familiar approximation is, as Stephen did, to replace the four-body correlation function by the product of two-body correlation functions.

$$\langle \Phi_0 | \Delta\rho_{k-p}(t) \Delta\rho_{p-k'}(t) \Delta\rho_{k'-q}(0) \Delta\rho_{q-k}(0) | \Phi_0 \rangle$$

$$\cong (\delta_{pq} + \delta_{k+k'p+q}) \langle \Phi_0 | \Delta\rho_{k-p}(t) \Delta\rho_{p-k}(0) | \Phi_0 \rangle \langle \Phi_0 | \Delta\rho_{p-k'}(t) \Delta\rho_{k'-p}(0) | \Phi_0 \rangle .$$

$$\tag{34}$$

Furthermore, both k and k' are practically negligible compared with most of p. Thus the cross section (33) is approximately written as

$$\frac{d^2\sigma_2}{d\omega d\Omega} \cong 2 \left(\frac{\nu}{c}\right)^4 \left(\frac{\alpha^4(\nu)}{V}\right) \int_{-\infty}^{\infty} \frac{d\nu_1 d^3 q}{(2\pi)^4} L_{\lambda'\lambda}^2(q) S(q, \omega - \nu_1) S(q, \nu_1) . \tag{35}$$

This is essentially the same result as obtained by Stephen. Note that from the tensorial character of (31), this must have the form

$$L_{\mu\nu}(q) = g(q) \left(\frac{3q_\mu q_\nu}{q^3} - \delta_{\mu\nu}\right), \tag{36}$$

where

$$g(q) = \left(\frac{d^2}{dq^2} - \frac{1}{q}\frac{d}{dq}\right) \int \frac{e^{-iq \cdot R}}{R^5} dR . \tag{37}$$

As Stephen has shown, (35) together with (36) can account for the depolarization effect observed by Greytak and Yan.

§ 5. Two roton processes in liquid He⁴

In the case of superfluid He⁴, the liquid may be regarded as a gas of quasi-particles. Let B_p, B_p^* be destruction and creation operators of the quasiparticle of momentum p. For small p, the quasiparticle is the phonon, so that

$$\Delta\rho_p = f_p^{1/2}(B_p + B_{-p}^*), \tag{38}$$

where f_p is a positive constant. For larger p, where the quasiparticle is the roton, we need to add quadratic and higher order terms, which we shall ignore for simplicity, however. The approximation is equivalent to Stephen's hydro-dynamic one.

Now, since k, k' in (33) are negligible against most of p, q, we write this expression approximately as

$$\frac{d^2\sigma_2}{d\omega d\Omega} = n^2 \left(\frac{\nu\alpha(\nu)}{c}\right)^4 S_{\lambda'\lambda}(\omega) . \tag{39}$$

Under the approximation (38),

$$S_{\lambda'\lambda}(\omega) = \sum_p' \sum_q' f_p L_{\lambda'\lambda}(p) f_q L_{\lambda'\lambda}(q)$$

$$\times \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{i\omega t} \langle B_p(t) B_{-p}(t) B_{-q}^*(0) B_q^*(0) \rangle , \tag{40}$$

where $\langle \ \rangle$ is the expectation value with respect to $\varPhi_0$. As is well known,[6] the Fourier transform in (40) is equal to $-\pi^{-1}$ multiplied by the imaginary part of the retarded Green function. For $\omega > 0$, the latter is given by

$$K_{pq}(\omega) = -i \int_{-\infty}^{\infty} dt \, e^{i(\omega+i0^+)t} \langle TB_p(t) B_{-p}(t) B_{-q}^*(0) B_q(0) \rangle, \qquad (41)$$

where T means Wick's chronological ordering.

When the interaction between rotons is ignored, the Green function (41) is given by

$$K_{pq}(\omega) = (\delta_{pq} + \delta_{p-q}) D_p(\omega), \qquad (42)$$

$$D_p(\omega) = \frac{1}{\omega - 2E_p + i0^+}, \qquad (43)$$

where E_p is the energy required to create one free roton. Hence

$$S_{\lambda'\lambda}(\omega) = 2 \sum_p f_p^2 L_{\lambda'\lambda}^2(p) \delta(\omega - 2E_p). \qquad (44)$$

The spectrum is determined in this case by the density of states of the free roton in accordance with the conclusion of Halley and Stephen.

However, Iwamoto[7] has recently pointed out that the interaction between the two rotons created by light scattering plays an important role particularly when the group velocity of the roton vanishes. Following him, let us describe the interaction between rotons by an effective Hamiltonian

$$H_{rr} = \frac{1}{2V} \sum W_Q(p, p') B_{Q/2+p}^* B_{Q/2-p}^* B_{Q/2-p'} B_{Q/2+p'}. \qquad (45)$$

The approximation introduced by Iwamoto in dealing with this interaction is equivalent to the so-called ladder approximation in the method of Green functions. Thus the Green function (41) is approximately given by the solution of the integral equation

$$K_{pq}(\omega) = (\delta_{pq} + \delta_{p-q}) D_p(\omega) + \frac{2}{V} \sum D_p(\omega) W_0(p, p') K_{p'q}(\omega). \qquad (46)$$

Then (40) can be written as

$$S_{\lambda'\lambda}(\omega) = -\frac{1}{\pi V} \text{Im} \sum_p \sum_q f_p L_{\lambda'\lambda}(p) f_q L_{\lambda'\lambda}(q) \varPsi_{pq}(\omega), \qquad (47)$$

where $\varPsi_{pq}(\omega)$ is the solution of the integral equation

$$\varPsi_{pq}(\omega) = \frac{1}{\omega - 2E_p + i0^+} \left[V\delta_{pq} + \frac{1}{V} \sum_{p'} W_0(p, p') \varPsi_{p'q}(\omega) \right]. \qquad (48)$$

Iwamoto has already obtained a number of interesting results about the solution of (48). In particular, the spectrum (44) obtained in the zeroth order approximation should be modified in a drastic way by the final state interaction between rotons.

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