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Mechanism for Two-Roton Raman Scattering from Liquid Helium

J. Woods Halley*
Department of Physics, University of California, Berkeley, California 94720
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We study couplings between the phonon-roton excitations of liquid helium and an electromagnetic field. By including the electronic excitations of the helium atoms as intermediate states, we find a second-order two-roton Raman scattering amplitude which may be large enough to be observable. The roton scattering cross section as a function of energy transfer is calculated from the experimentally known roton spectrum and the pair-correlation function. The lowest-order infrared-absorption process involves $(qa)^2$ and the quadrupole-dipole interaction and is too small to be observed.

I. INTRODUCTION

In several many-particle systems, Raman scattering has been a useful tool for studying elementary excitations, complementing inelastic neutron scattering. In liquid helium, Brillouin scattering[1] has permitted the study of sound waves. It is of interest to ask if the rest (roton part) of the vibrational excitation spectrum observed by neutron scattering could also be observed in light scattering experiments. One might expect, however, that only a small coupling would arise between the phonon-roton spectrum of liquid helium and an electromagnetic field for two reasons: First, the requirements of energy and momentum conservation would lead only to production of single excitations of very small energy (Brillouin scattering). Second, none of the excitations carry an electric or magnetic dipole moment in lowest order of approximation, so no coupling seems possible. We point out here that one can get around the first, momentum conservation, difficulty by considering the creation of two excitations simultaneously. Then the peak absorption or scattering cross section occurs when the two-excitation density of states is largest, i.e., when the energy transfer to the fluid is such that rotons at one of the two flat places in the ω versus k curve for the excitations are produced in the Raman scattering or infrared-absorption process. Because the liquid is isotropic these peaks in the density of states occur at the same energy in all directions in k space and a larger peak is expected than in the analogous situation in most solids.[2] The experimental Raman-scattering (infrared-absorption) peaks would come at convenient energy transfers (infrared photon energies) of 12 and 19 cm^{-1} so that the experiment might be quite easy to perform. A Raman-scattering experiment is also simpler than the corresponding Brillouin-scattering experiment because the energy and momentum conservation rules do not fix the angle of the scattered photon as a function of energy transfer in the two-excitation case.[3]

The second, dipole-moment, difficulty is resolved if one considers that in an excitation of large k and small wavelength, the helium atoms are mutually polarized by the van der Waals interactions. This polarization provides a means by which the excitation can couple to the electromagnetic field. The magnitude of this polarization clearly increases with increasing k. The result is that the two-roton process, which involves high-k excitations, might have a substantial cross section compared with the one-phonon (Brillouin) process (small k) even though the latter is of first order in the density fluctuation associated with the phonon while the two-roton process is of second order in the density fluctuation associated with the roton. In this paper, we calculate the Raman-scattering cross section for two-roton scattering on the basis of these ideas. We find that the dipole-dipole term in a multipole expansion of the Coulomb interaction between helium atoms gives a two-roton Raman-scattering cross section which may be observable. The first contribution to infrared absorption comes from the quadrupole-dipole term and is too small to be observable. If the Raman scattering is observed, an easy way to investigate the effects of vortices on the roton spectrum may present itself. This is briefly discussed.

II. MODEL HAMILTONIAN AND REDUCTION TO PHONON AND EXCITON VARIABLES

The model which we consider is described by the following Hamiltonian:

$$\mathcal{H} = \mathcal{H}_{\text{atoms}} + \mathcal{H}_{\text{atom-atom}} + \mathcal{H}_{\text{atom-field}}, \tag{1}$$

$$\text{where} \quad \mathcal{H}_{\text{atoms}} = \sum_{l=1}^{N} \left(\frac{-\hbar^2}{2M} \right) \nabla_l^2 + \sum_{l=1}^{N} \left[\sum_{i=1}^{2} \left(\frac{-\hbar^2}{2m_e} \nabla_{li}^2 - \frac{2e^2}{|\vec{r}_{li}|} \right) + \frac{e^2}{|\vec{r}_{l1} - \vec{r}_{l2}|} \right],$$

$$\mathcal{H}_{\text{atom-atom}} = \sum_{l<m} \left[\frac{4e^2}{|\vec{R}_{lm}|} + \sum_{i,j=1}^{2} \frac{e^2}{|\vec{R}_{lm} + \vec{r}_{li} - \vec{r}_{mj}|} - 2e^2 \sum_{i=1}^{2} \left(\frac{1}{|\vec{R}_{lm} + \vec{r}_{li}|} + \frac{1}{|\vec{R}_{lm} - \vec{r}_{mi}|} \right) \right], \tag{2}$$

$$\mathcal{H}_{\text{atom field}} = \frac{-e}{mc} \sum_{l=1}^{N} \sum_{i=1}^{2} \vec{p}_{li} \cdot \vec{A}(\vec{r}_{li}, t).$$

Here M is the mass of the helium nucleus, m_e is the electron mass, $\vec{R}_{lm} = \vec{R}_l - \vec{R}_m$ where $\vec{R}_l$ is the position of the lth nucleus, $\vec{r}_{li} = \vec{x}_{li} - \vec{R}_l$, and $\vec{x}_{li}$ is the coordinate of the ith electron on the lth nucleus (we neglect exchange effects). To express this Hamiltonian in terms of phonon and exciton variables we write an effective atom-atom interaction

$$\mathcal{H}^{\text{eff}}(\{\vec{R}_l\}) = \langle \text{gnd} | \mathcal{H}_{\text{atom-atom}} | \text{gnd} \rangle , \tag{3}$$

where $|\text{gnd}\rangle$ refers only to the electronic coordinates and is defined in the Appendix. Then in the spirit of the Born-Oppenheimer approximation we consider the part

$$\mathcal{H}_{\text{phonons}} = \sum_{l=1}^{N} \left(\frac{-\hbar^2}{2M} \right) \nabla_l^2 + \mathcal{H}^{\text{eff}}(\{\vec{R}_l\}) \tag{4}$$

of the Hamiltonian and find vibrational modes from it. These we will identify with the experimental phonon-roton spectrum. To get the vibrational modes in the present approximation scheme we choose a lattice of nuclear positions $\{R_l^{(0)}\}$. Later we will average over lattices in a way which is consistent with the observed pair-correlation function for the fluid. We expand $\mathcal{H}^{\text{eff}}$ to second order in atomic displacements $\delta\vec{R}_l = \vec{R}_l - \vec{R}_l^{(0)}$ from this lattice and, restricting attention to lattices for which $\mathcal{H}^{\text{eff}}$ is a local minimum, we have to second order

$$\mathcal{H}_{\text{phonons}} \approx \sum_{l=1}^{N} \left(\frac{-\hbar^2}{2M} \right) \nabla_l^2 + \tfrac{1}{2} \sum_{l,m} \sum_{\mu,\nu} \delta R_{lm}{}^{\mu} \delta R_m{}^{\nu} G_{lm}{}^{\mu\nu} , \tag{5}$$

leading in the usual way[4] to

$$\mathcal{H}_{\text{phonons}} = \sum_{\vec{k}\mu} \hbar\omega_{\vec{k}\mu} a_{\vec{k}\mu}^{\dagger} a_{\vec{k}\mu} , \tag{6}$$

where $a_{\vec{k}\mu}^{\dagger}$ are phonon operators obeying boson commutation relations and related to the displacements $\delta R_l{}^{\mu} = R_l{}^{\mu} - (R_l{}^{\mu})_0$ by

$$\delta\vec{R}_l = \sum_{\vec{k}\mu} \hat{e}_{\vec{k}\mu} (\hbar / 2\rho\omega_{\vec{k}\mu} V)^{1/2} [a_{\vec{k}\mu} \exp(-i\vec{k} \cdot \vec{R}_l^{(0)}) + \text{H.c.}] , \tag{7}$$

where $\hat{e}_{\vec{k}\mu}$ is the polarization vector and $\omega_{\vec{k}\mu}$ is the frequency of the phonon.

Now because we want to represent the liquid in some approximate fashion by this model, we assume that G has the property that it gives no transverse modes so that the sums on μ in the last two equations are dropped. The Hamiltonian is then rewritten

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I , \quad \mathcal{H}_0 = \mathcal{H}_{\text{phonons}} + \mathcal{H}_{\text{excitons}} ,$$

$$\mathcal{H}_{\text{excitons}} = \sum_l \sum_{\nu} E_{\nu} b_{\nu}^{(l)\dagger} b_{\nu}^{(l)} = \sum_{\nu,\vec{k}} E_{\nu} \beta_{\vec{k}}^{(\nu)\dagger} \beta_{\vec{k}}^{(\nu)} , \tag{8}$$

$$\mathcal{H}_I = \mathcal{H}_{\text{atom-atom}} - \mathcal{H}^{\text{eff}}(\{R_l\}) + \sum_{l=1}^{N} \sum_{i=1}^{2} \left(\frac{-2e^2}{|\vec{x}_{li} - \vec{R}_l|} + \frac{2e^2}{|\vec{x}_{li} - \vec{R}_l^{(0)}|} \right) ,$$

where $\mathcal{H}_{\text{excitons}}$ accounts for the electronic states of the helium atoms in a way detailed in the Appendix. The remaining interactions can now be written in terms of exciton and phonon variables. Expanding $\mathcal{H}_{\text{atom-atom}}$ in a series in $|\vec{r}_{li}|/|\vec{R}_{lm}|$ in the usual way[5] gives a multipole moment expansion:

$$\mathcal{H}_{\text{atom-atom}} = \sum_{l<m} \sum_{i,j=1}^{2} \Bigg(\frac{-e^2}{R_{lm}^{3}} (2z_{li}z_{mj} - x_{li}x_{mj} - y_{li}y_{mj}) + \frac{3}{2}\frac{e^2}{R_{m}^{4}} [r_{li}^{2}z_{mj} - z_{li}r_{mj}^{2} + (2x_{li}x_{mj} + 2y_{li}y_{mj} - 3z_{li}z_{mj})(z_{li} - z_{mj})] + \frac{3}{4}\frac{e^2}{R_{lm}^{5}} [r_{li}^{2}r_{mj}^{2} - 5r_{li}^{2}z_{mj}^{2} - 5r_{mj}^{2}z_{li}^{2} - 15z_{li}^{2}z_{mj}^{2} + 2(4z_{li}z_{mj} - x_{li}x_{mj} - y_{li}y_{mj})^{2}] + \cdots \Bigg). \tag{9}$$

Using this, we express $\mathcal{H}_I$ in terms of local exciton operators $b_l^{(\nu)}$ and local displacements δR_{lm}^{μ}. We find

$$\begin{aligned}
\mathcal{H}_I = {} & \sum_{l<m} \left(\frac{-e^2}{|\vec{R}_{lm}^{(0)}|^3} [\mathfrak{M}^{(1)}(\nu,g;\nu',g) b_l^{(\nu)\dagger} b_m^{(\nu')} + \mathfrak{M}^{(1)}(\nu,g;g,\nu') b_l^{(\nu)\dagger} b_m^{(\nu')} + \text{H.c.}] \right) \\
& + \frac{3}{2} \sum_{\nu} \sum_{l<m} \left(\frac{e^2}{|R_{lm}^{(0)}|^4} [\mathfrak{M}^{(2)}(g,g;g,\nu)(b_m^{(\nu)} - b_l^{(\nu)}) + \mathfrak{M}_2^{(2)}(g,g;g,\nu)(b_l^{(\nu)} - b_m^{(\nu)}) + \text{H.c.}] \right) \\
& + \sum_{l<m} \sum_{\nu,\nu'} \left[\frac{-e^2}{|R_{lm}^{(0)}|^3} \left(\sum_j M_j^{(1;3)}(l,m) \frac{\delta R_{lm}^{\ j}}{R_{lm}^{(0)}} + \sum_{i,j} M_{ij}^{(2;3)}(l,m) \frac{\delta R_{lm}^{\ i} \delta R_{lm}^{\ j}}{|\vec{R}_{lm}^{(0)}|^2} \right) \right. \\
& \left. \times [\mathfrak{M}^{(1)}(\nu,g;\nu'g) b_l^{(\nu)\dagger} b_m^{(\nu')\dagger} + \mathfrak{M}^{(1)}(\nu,g;g,\nu') b_l^{(\nu)\dagger} b_m^{(\nu')} + \text{H.c.}] \right] \\
& + \frac{3}{2} \sum_{\nu} \sum_{l<m} \left[\frac{e^2}{|\vec{R}_{lm}^{(0)}|^4} \left(\sum_j M_j^{(1;4)}(l,m) \frac{\delta R_{lm}^{\ j}}{|\vec{R}_{lm}^{(0)}|} + \sum_{i,j} M_{ij}^{(2;4)}(l,m) \frac{\delta R_{lm}^{\ i} \delta R_{lm}^{\ j}}{|\vec{R}_{lm}^{(0)}|^2} \right) \right. \\
& \left. \times [\mathfrak{M}_1^{(2)}(g,g;g,\nu)(b_m^{(\nu)} - b_l^{(\nu)}) + \mathfrak{M}_2^{(2)}(g,g;g,\nu)(b_l^{(\nu)} - b_m^{(\nu)}) + \text{H.c.}] \right].
\end{aligned} \tag{10}$$

Here we have kept terms to fourth order in the b and δR in the dipole-dipole interaction. Some terms have been dropped on the basis of parity selection rules. In the quadrupole-dipole term we show only terms linear in the b since these give rise to the only second-order infrared absorption. In (10) the matrix elements are defined as follows

$$\begin{aligned}
M_i^{(1;3)}(l,m) &= \frac{-3R_{lm}^{(0)i}}{R_{lm}^{(0)}}, \quad M_{ij}^{(2;3)}(l,m) = \frac{3}{2}\left(\frac{5R_{lm}^{(0)i}R_{lm}^{(0)j}}{R_{lm}^{(0)2}} - \delta_{ij} \right), \\
M_i^{(1;4)}(l,m) &= \frac{-4R_{lm}^{(0)i}}{R_{lm}^{(0)}}, \quad M_{ij}^{(2;4)}(l,m) = 2\left(\frac{6R_{lm}^{(0)i}R_{lm}^{(0)j}}{R_{lm}^{(0)2}} - \delta_{ij} \right);
\end{aligned} \tag{11}$$

$$\mathfrak{M}^{(1)}(\nu,\nu';\nu'',\nu''') = 2Z(\nu,\nu')Z(\nu'',\nu''') - X(\nu,\nu')X(\nu'',\nu''') - Y(\nu,\nu')Y(\nu'',\nu'''),$$

$$\mathfrak{M}_1^{(2)}(\nu,\nu';\nu'',\nu''') = R^2(\nu,\nu')Z(\nu'',\nu'''), \quad \mathfrak{M}_2^{(2)}(\nu,\nu';\nu'',\nu''') = -3Z^2(\nu,\nu')Z(\nu'',\nu''');$$

$$Z(\nu,\nu')=\int\int\psi_\nu^{(l)*}\left(\sum_{i=1}^{2}z_{li}\right)\psi_{\nu'}^{(l)}d\tau_1 d\tau_2,\qquad X(\nu,\nu')=\int\int\psi_\nu^{(l)*}\left(\sum_{i=1}^{2}x_{li}\right)\psi_{\nu'}^{(l)}d\tau_1 d\tau_2,$$

$$Y(\nu,\nu')=\int\int\psi_\nu^{(l)*}\left(\sum_{i=1}^{2}Y_{li}\right)\psi_{\nu'}^{(l)}d\tau_1 d\tau_2,\qquad R^2(\nu,\nu')=\int\int\psi_\nu^{(l)*}\left(\sum_{i=1}^{2}r_{li}^2\right)\psi_{\nu'}^{(l)}d\tau_1 d\tau_2,$$

$$Z^2(\nu,\nu')=\int\int\psi_\nu^{(l)*}\left(\sum_{i=1}^{2}z_{li}^2\right)\psi_{\nu'}^{(l)}d\tau_1 d\tau_2.$$

In Eq. (10), the first two sets of terms involve only exciton operators. When they are added to $\mathcal{H}_{\text{excitons}}$, the resultant Hamiltonian can be diagonalized by a canonical transformation (to remove the linear terms) and a Bogoliubov transformation. The modified exciton Hamiltonian will again be of form (8) but E_ν will depend on $\vec{K}$. The width of the resultant exciton band is expected to be small compared with the mean energy of the exciton, and we will ignore it since it leads to no effects of essential interest here. The term in $M_j^{(1,3)}$ leads to light scattering with production of one phonon-roton excitation. For such a scattering the maximum momentum transfer is $\sim 2K$ where K is the wave number of the scattered photon or $10^5\ \text{cm}^{-1}$. This is much smaller than the roton wave number of $10^8\ \text{cm}^{-1}$, and the excitations produced are thus phonons near the center of the zone. We thus have a mechanism for Brillouin scattering. The kinematics of this are discussed in Ref. 3 and the process has been observed in liquid helium. The terms in $M_{ij}^{(2;3)}(l,m)$ give the two-roton Raman scattering (Fig. 1) mechanism of interest here. The term in $M_{ij}^{(2;3)}(l,m)$ gives infrared absorption. It is not hard to show, by procedures like those used below, that the matrix element for this mechanism goes as $(1-e^{ika})$ where k is the photon wave number and a is of the order of a few tens of lattice spacings at most. Thus the two-roton infrared absorption is expected to be $\sim(ka)^2\ll 1$ times less intense than expected from the value of $M_{ij}^{(2;4)}(l,m)$ without proper consideration of kinematical effects.

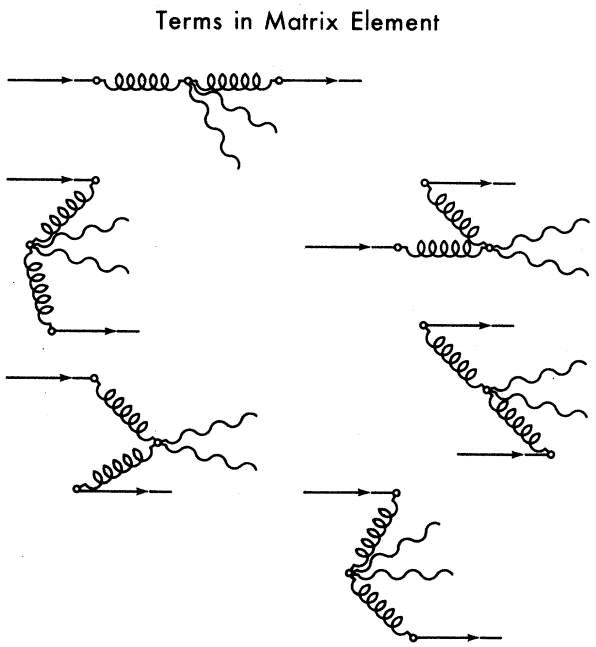

FIG. 1. Diagrams describing mechanism leading to two-roton Raman scattering.

These remarks indicate that the term of interest is that containing $M_{ij}^{(2;3)}(l,m)$. We express it in terms of the Fourier transformed operators $\beta_{\vec{k}}^{(\nu)}$, $a_{q\mu}$. Denoting this part of $\mathcal{H}_I$ by $\mathcal{H}_I^{(2-2)}$ we have

$$\mathcal{H}_I^{(2-2)}=\sum_{\vec{k},\vec{k}'}\sum_{\vec{q},\mu}\sum_{\mu',\nu',\nu}\Big(\{\beta_{\vec{k}}^{(\nu)\dagger}\beta_{\vec{k}}^{(\nu')}\mathfrak{M}^{(1)}(\nu,g;\nu',g)$$

$$\times[T_{\mu\mu'}(\vec{k}',\vec{q},\vec{k}+\vec{k}'-\vec{q})a_{q\mu}a_{\vec{k}+\vec{k}'-\vec{q},\mu'}+T_{\mu\mu'}(\vec{k}',\vec{q},-\vec{k}-\vec{k}'+\vec{q})a_{q\mu}a^{\dagger}_{\vec{k}+\vec{k}'-\vec{q},\mu'}+\text{H.c.}]+\text{H.c.}\}$$

$$+\{\beta^{(\nu)\dagger}_{\vec{k}}\beta^{(\nu')}_{\vec{k}}\mathfrak{M}^{(1)}(\nu,g;g,\nu')[T_{\mu\mu'}(-\vec{k}',\vec{q},\vec{k}-\vec{k}'-\vec{q})a_{\vec{q}\mu}a_{\vec{k}-\vec{k}'-\vec{q},\mu'}$$

$$+T_{\mu\mu'}(-\vec{k},\vec{q},-\vec{k}+\vec{k}'+\vec{q})a_{\vec{q}\mu}a^{\dagger}_{-\vec{k}+\vec{k}'+\vec{q},\mu'}+\text{H.c.}]+\text{H.c.}\}\Big)\,. \tag{12}$$

Here

$$T_{\mu\mu'}(\vec{k}',\vec{q},\vec{q}')=-\frac{e^2\hbar}{4\rho V}\sum_{i,j}\sum_{l-m}\frac{M_{ij}^{(2;3)}(l,m)}{(R_{lm}^{(0)})^5}D(l,m;-\vec{k}',\vec{q},\vec{q}')e_{q\mu}{}^{i}e_{q'\mu'}{}^{j}(\omega_{q\mu}\omega_{q'\mu'})^{\frac{1}{2}}]\,, \tag{13}$$

in which

$$D(l,m;-\vec{k}',\vec{q},\vec{q}')=e^{-i\vec{k}'\cdot\vec{R}_{ml}^{(0)}}(1-e^{i\vec{q}\cdot\vec{R}_{ml}^{(0)}})(1-e^{i\vec{q}'\cdot\vec{R}_{ml}^{(0)}})\,. \tag{14}$$

To calculate the Raman amplitude we also need to express $\mathcal{H}_{\text{atom field}}$ in terms of photon operators $\alpha_{\vec{k}\lambda}$ and exciton operators $\beta_{\vec{k}}^{(\nu)}$. We find[6]

$$\mathcal{H}_{\text{atom field}}=\sum_{k,\lambda,\nu}[m(\vec{k},\lambda,\nu)(\beta_{\vec{k}}^{(\nu)\dagger}\alpha_{\vec{k}\lambda}+\beta_{-\vec{k}}^{(\nu)\dagger}\alpha_{\vec{k}\lambda}^{\dagger})+\text{H.c.}]\,, \tag{15}$$

$$\text{where}\quad m(\vec{k},\lambda,\nu)=ie(N/V)^{-1/2}(2\pi\hbar/ck)^{1/2}[\vec{\mathfrak{X}}(\nu,g)\cdot\hat{\epsilon}_{\vec{k}\lambda}](E_\nu/\hbar)\,, \tag{16}$$

$$\text{and}\qquad \vec{\mathfrak{X}}(\nu,g)=\int\int\psi_\nu^{(l)*}\sum_{i=1}^{2}\vec{x}_{li}\psi_g^{(l)}d\tau_1 d\tau_2\,. \tag{17}$$

III. CALCULATION OF RAMAN AMPLITUDE

The scattering rate for the process

$$(1\text{ photon at }\vec{q})\rightarrow(2\text{ rotons at }\vec{k},\ \vec{k}')+(1\text{ photon at }\vec{q}') \tag{18}$$

via the mechanism described by Fig. 1 is given by

$$\sum_{\vec{k},\vec{k}'}(2\pi/\hbar)|M(\vec{q},\vec{k},\vec{k}',\vec{q}')|^2\delta(\hbar c(|q|-|q'|)-\hbar\omega_{\vec{k}}-\hbar\omega_{\vec{k}'})\,, \tag{19}$$

$$\text{where}\quad M(\vec{q},\vec{k},\vec{k}',\vec{q})=\sum_{m',m''}\frac{\langle\vec{q}|\mathcal{H}'|m'\rangle\langle m'|\mathcal{H}'|m''\rangle\langle m''|\mathcal{H}'|\vec{k},\vec{k}',\vec{q}'\rangle}{(\hbar c|\vec{q}|-E_{m'})(\hbar c|\vec{q}|-E_{m''})}\,, \tag{20}$$

$$\text{and}\quad \mathcal{H}'=\mathcal{H}_I^{(2-2)}+\mathcal{H}_{\text{atom field}}\,. \tag{21}$$

The six diagrams[7] corresponding to the sum (20) are shown in Fig. 1. Writing out $M(\vec{q};\vec{k},\vec{k}',\vec{q}')$ using (12) and (18) then gives

$$M(\vec{q};\vec{k},\mu,\vec{k}',\mu',\vec{q}')$$

$$=\sum_{\nu,\nu'}\{m^*(\vec{q},\nu,\lambda)\mathfrak{M}^{(1)}(\nu,g;\nu',g)T^{*}_{\mu\mu'}(\vec{q}-\vec{k}-\vec{k}',\vec{k},\vec{k}')m^*(\vec{q}-\vec{k}-\vec{k}',\nu',\lambda')$$

$$\times[(E_{-\vec{q}}{}^{\nu}+E_{\vec{q}-\vec{k}-\vec{k}'}{}^{\nu'}+\hbar\omega_{\vec{k}}^{\text{ph}}+\hbar\omega_{\vec{k}'}^{\text{ph}})^{-1}(\hbar\omega_{q\lambda}+E_{-q}{}^{\nu})^{-1}$$

$$+(E_{-\vec{q}}{}^{\nu}+E_{\vec{q}-\vec{k}-\vec{k}'}{}^{\nu'}+\hbar\omega_{\vec{k}\mu}^{\text{ph}}+\hbar\omega_{\vec{k}'\mu'}^{\text{ph}})^{-1}(-\hbar\omega_{\vec{q}\lambda}+E_{\vec{q}-\vec{k}-\vec{k}'}{}^{\nu}+\hbar\omega_{\vec{k}}^{\text{ph}}+\hbar\omega_{\vec{k}'}^{\text{ph}})^{-1}]$$

$$+m(\vec{q},\nu,\lambda)\mathfrak{M}^{(1)}(g,\nu;g,\nu')T^{*}_{\mu\mu'}(\vec{k}+\vec{k}'-\vec{q},\vec{k},\vec{k}')m(\vec{q}-\vec{k}-\vec{k}',\nu',\lambda')$$

$$\times[(h\omega_{\vec{q}\lambda}-h\omega_{\vec{k}\mu}-h\omega_{\vec{k}'\mu'}+E_{\vec{k}+\vec{k}'-\vec{q}}^{\nu})^{-1}(E_{\vec{q}}^{\nu}+E_{\vec{k}+\vec{k}'-\vec{q}}^{\nu'}-h\omega_{\vec{k}\mu}^{\mathrm{ph}}-h\omega_{\vec{k}'\mu'}^{\mathrm{ph}})^{-1}$$

$$+(-h\omega_{\vec{q}\lambda}+E_{\vec{q}}^{\nu})^{-1}(-h\omega_{\vec{k}\mu}^{\mathrm{ph}}-h\omega_{\vec{k}'\mu'}^{\mathrm{ph}}+E_{\vec{k}+\vec{k}'-\vec{q}}^{\nu'}+E_{\vec{q}}^{\nu})^{-1}]$$

$$+m(q,\nu,\lambda)\mathfrak{M}^{(1)}(\nu',g;g,\nu)T_{\mu\mu'}{}^{*}(\vec{k}+\vec{k}'-\vec{q},\vec{k},\vec{k}')m^{*}(\vec{q}-\vec{k}-\vec{k}',\nu',\lambda')$$

$$\times(h\omega_{\vec{q}}-E_{\vec{q}}^{\nu})^{-1}(h\omega_{\vec{q}}-E_{\vec{q}-\vec{k}-\vec{k}'}^{\nu'}-h\omega_{\vec{k}\mu}^{\mathrm{ph}}-h\omega_{\vec{k}'\mu'}^{\mathrm{ph}})^{-1}$$

$$+m(q,\nu,\lambda)\mathfrak{M}^{(1)}(\nu,g;g,\nu')T_{\mu\mu'}{}^{*}(\vec{q}-\vec{k}-\vec{k}',\vec{k},\vec{k}')m^{*}(\vec{q}-\vec{k}-\vec{k}',\nu',\lambda')$$

$$\times[(-h\omega_{\vec{q}}+h\omega_{\vec{k}'\mu'}^{\mathrm{ph}}+h\omega_{\vec{k}\mu}^{\mathrm{ph}}-E_{\vec{k}+\vec{k}'-\vec{q}}^{\nu'})^{-1}(-h\omega_{\vec{q}\lambda}-E_{-\vec{q}}^{\nu})^{-1}]\,. \tag{22}$$

To simplify this, we note that the E^{ν} are weakly k dependent and that the m depend on $\vec{q}$ only through the polarization of the photons. In the situation of interest, $|\vec{k}|$, $|\vec{k}'|\gg\vec{q}$ so that $\vec{k}\simeq-\vec{k}'$. Further $E^{\nu}\gg h\omega_{\vec{k}}^{\mathrm{ph}}$. With these approximations the k dependence of M is in the factors T which, in this approximation, are the same for each term in (22). Keeping only the longitudinal μ as discussed before, T becomes

$$T(\vec{k}+\vec{k}'-\vec{q},\vec{k},\vec{k}')=\frac{-e^2\hbar}{4\rho V\omega_{\vec{k}}}\sum_{l-m}\frac{3}{2}\left(\frac{5(\vec{R}_{lm}^{(0)}\cdot\hat{k})^2}{|R_{lm}^{(0)}|^2}-1\right)\frac{4}{R_{lm}^{(0)5}}\sin^2(\tfrac{1}{2}\vec{k}\cdot\vec{R}_{lm}^{(0)})\,. \tag{23}$$

Writing $\theta(\hat{k};l,m)$ for the angle between $\vec{k}$ and $\vec{R}_{lm}^{(0)}$ one has

$$T(\vec{k}+\vec{k}'-\vec{q},\vec{k},\vec{k}')=\frac{-3e^2\hbar}{2\rho V\omega_{\vec{k}}}\sum_{l-m}\left(\frac{5\cos^2\theta(\hat{k};l,m)-1}{R_{lm}^{5}}\right)\sin^2(\tfrac{1}{2}kR_{lm}\cos\theta(\hat{k};l,m))\,, \tag{24}$$

and replacing $$\sum_{l-m}(\cdots)\to 2\pi\int_{-1}^{+1}d\mu\int_{0}^{\infty}dR_{lm}g(R_{lm})(\cdots)\,, \tag{25}$$

where $g(R_{lm})$ is the pair-correlation function and $\mu=\cos\theta(k;l,m)$, one has, after doing the angular integrals, that

$$T(\vec{k}+\vec{k}'-\vec{q},\vec{k},\vec{k}')=\frac{-6\pi e^2\hbar}{V\omega_{\vec{k}}}\int_{0}^{\infty}\frac{g(R_{lm})dR_{lm}}{R_{lm}^{5}}\left(\frac{1}{3}-2\frac{\sin kR_{lm}}{kR_{lm}}-5\frac{\cos kR_{lm}}{(kR_{lm})^2}+5\frac{\sin kR_{lm}}{(kR_{lm})^3}\right)\equiv T(k)\,. \tag{26}$$

If we neglect the wave-vector dependence of the exciton energies, we have

$$M(\vec{q};\vec{k},\mu;\vec{k}',\mu',\vec{q}')\simeq T(k)\sum_{\nu,\nu'}\{m^{*}(\vec{q},\nu,\lambda)m^{*}(\vec{q}',\nu',\lambda')\mathfrak{M}^{(1)}(\nu,g;\nu';g)$$

$$\times[(E^{\nu}+E^{\nu'})^{-1}(E^{\nu})^{-1}+(E^{\nu}+E^{\nu'})^{-1}(E^{\nu'}-h\omega_{\vec{q}})^{-1}]$$

$$+m(\vec{q},\nu,\lambda)m(\vec{q}',\nu',\lambda')\mathfrak{M}^{(1)}(g,\nu;g,\nu')$$

$$\times[(E^{\nu}+h\omega_{\vec{q}})^{-1}(E^{\nu}+E^{\nu'})^{-1}+(E^{\nu}-h\omega_{\vec{q}})^{-1}(E^{\nu}+E^{\nu'})^{-1}]$$

$$+m(q,\nu,\lambda)m^{*}(q',\nu',\lambda')\mathfrak{M}^{(1)}(\nu',g;g,\nu)/(h\omega_{\vec{q}}-E^{\nu})(h\omega_{\vec{q}}-E^{\nu'})$$

$$+m(\vec{q},\nu,\lambda)m^{*}(\vec{q}',\nu',\lambda')\mathfrak{M}^{(1)}(\nu,g;g,\nu')/(h\omega_{\vec{q}}+E^{\nu'})(h\omega_{\vec{q}}+E^{\nu})\}\,. \tag{27}$$

The dependence of this on energy transfer is entirely contained in $T(k)$. To get the transition rate from M we have

$$W(\hbar\Delta\omega \equiv \hbar\omega_{\vec{q}} - \hbar\omega_{\vec{q}'}) = (2\pi/\hbar)\sum_{\vec{k}} |M(\vec{k},\vec{q},\vec{q}')|^2 \delta(2\hbar\omega_{\vec{k}}^{\mathrm{ph}} - \hbar\omega_{\vec{q}} + \hbar\omega_{\vec{q}'})$$

$$= \frac{V}{\pi\hbar^2} \sum_{i=1}^{3} \left(\frac{k_i^2}{|d\omega_{ki}^{\mathrm{ph}}/dk_i|} |M(k_i,\vec{q},\vec{q}')|^2 \right)_{2\omega_{ki}^{\mathrm{ph}} = \Delta\omega} , \tag{28}$$

where k_1, k_2, k_3 are the three solutions to $2\omega_i^{\mathrm{ph}} = \Delta\omega$. To evaluate this, one needs $T(k)$ which can be found numerically from the experimental pair-correlation function and the spectrum ω_k^{ph}. The result is shown in Fig. 2, where the experimental data[8,9] for g and ω_k^{ph} have been used. Using $T(k)$ we compute $W(\Delta\omega)$ as shown in Fig. 3. $W(\Delta\omega)$ is related to the Raman-scattering cross section by the relation

$$d^2\sigma/d(\hbar\omega_{\vec{q}'})d\Omega_{\vec{q}'} = W(\Delta\omega)V^2q'^2/\hbar c^2(2\pi)^3 . \tag{29}$$

We estimate the magnitude of the scattering rate as follows. From (22) the scattering rate is of order

$$W \sim (1/\hbar c)[(e\mathcal{E}a_b)^2(\delta R)^2 e^2 a_b^2/R^5E^2] \times (\text{density of states}) ,$$

where $\mathcal{E}$ is the electric field, a_b the Bohr radius, R the interatomic distance, δR the displacement associated with the roton, and E a typical exciton energy. The density of states is

$$\sim Vk^2\Delta k/\Delta E , \tag{30}$$

if one observes a range Δk of wave numbers and ΔE of energies. We put $V \sim 1$ (cm)^3, $k \sim \Delta k \sim 1$ (Å)$^{-1}$,

$$\mathcal{E} \sim (2\pi\hbar c/k)^{1/2}\omega_{\vec{k}}/c\sqrt{V} \sim 10^{-5} \text{ statV/cm} , \quad E \sim 10 \text{ eV}, \quad \delta R/R \sim 10^{-2} .$$

Then we find

$$W \sim 10^{-18} \left(\begin{matrix} \text{scattered photons with energy transfer} \\ \text{at the two-roton energy} \end{matrix} \right) \Big/ (\text{incident photon}) . \tag{31}$$

If one can increase the path length of the light in the helium by a factor of 10^2 one has 10^{-16}. This is weak but might be observable.

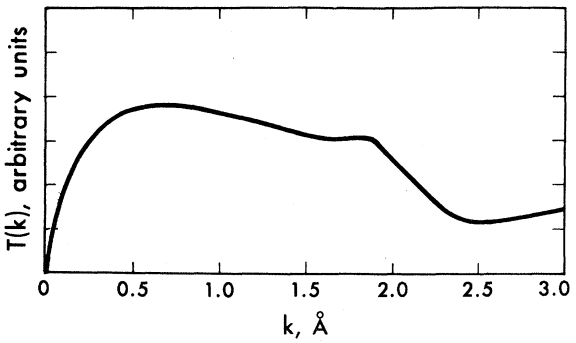

FIG. 2. The function $T(k)$.

IV. DISCUSSION

The model used only represents real liquid helium rather crudely. This is particularly the case because we have used a rather literal lattice model[10] for the liquid. Because we are not trying to calculate phonon spectra, however, this crudeness will not affect the results very much. In particular, the dependence of the dipole-dipole interaction on the wave vectors of the interacting

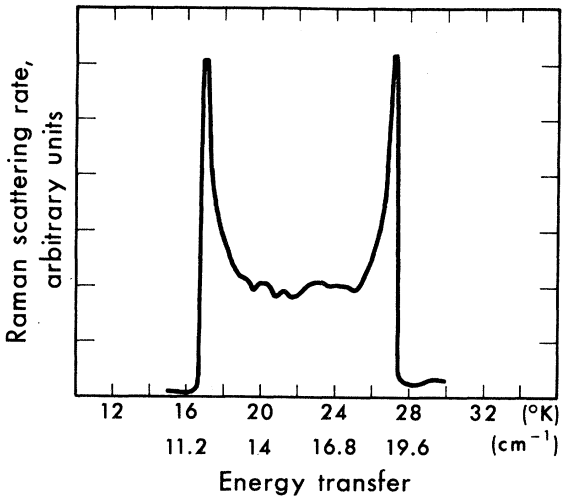

FIG. 3. Computed scattering rate.

excitations is not expected to depend in any important way on the lattice model. It is this wave-vector dependence which gives the form of the predicted cross section. We have assumed that

the physical origin of the exciton-phonon interaction is the same as that of the deformation-potential interaction in solids and that this interaction can be treated in perturbation theory. The first assumption seems plausible, as does the second at least for the very small cross-section we have calculated. It should be noted though, that we have proved neither assumption.

The excitons we have used are only Fourier transformed for mathematical convenience, and the question of whether the excitons in liquid helium are localized or bandlike will not affect the conclusions reached here. An extension of the present results to include other kinds of excitons,[11] for example molecular excitations of the sort, He_2^*, would not affect our conclusions in any important way.

The present results do not seem to contradict any sum rules[12] on $S(q,\omega)$ for the following reason. The effective coupling which we derive depends on the distance between the two displaced atoms labeled l and m with displacements δR_l and δR_m [Eq. (15)]. If we now refer to the standard derivation[13] of the conclusion that the scattering cross section for neutron or light scattering is proportional to $S(q,\omega)$ we see that it depends on the assumption of a coupling between exciting field and many-body system which is of form $\sum_i V(\vec{x} - \vec{x}_i)$. If one tries to replace this by the form

$$\sum_{i,j} V(\vec{x}-\vec{x}_i,\ \vec{x}-\vec{x}_j,\ \vec{x}_i-\vec{x}_j)$$

appropriate to this situation, then the derivation fails. Physically, the matter can be put this way: The large coupling at large wave vectors of the excited rotons occur because the roton is assumed to polarize the helium atoms through the accompanying very short wavelength density fluctuation. This polarization cannot be included in $S(\vec{q},\omega)$ which contains no reference to the wave vectors of the rotons.

We note that the two-roton Raman-scattering experiment would be interesting both because it could be used to check the present theory and because it would provide a simpler way of looking at the excitation spectrum of liquid helium than neutron scattering and therefore might be useful in the study of questions of temperature, pressure, and rotation dependence of the roton spectrum. With regard to checking the present theory, it should be possible to divide the experimental results by the roton-phonon density of states determined from neutron scattering and thus obtain a measure of $T(k)$. With regard to study of the spectrum, one interesting question concerns what happens to the spectrum in rotating helium. It is suggested[14] by weakly interacting models that the phonon-roton spectrum will lose its l degeneracy in the presence of a vortex. Though neutron studies[15] have shown no evidence of this effect, it would be interesting to study this question further. The treatment of light scattering from a rotating fluid containing vortices is a somewhat different theoretical problem which we will consider later.

Finally we note that the present machinery can be used to study the mechanisms of uv absorption of light with the production of excitons, a process of some current experimental interest.

V. ACKNOWLEDGMENTS

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APPENDIX; EXCITON NOTATION

Define $\psi_\nu^{(l)}(1,2)$ such that

$$[\mathcal{H}_1^{(l,0)}(1)+\mathcal{H}_1^{(l,0)}(2)+V^{(l)}(1,2)]\psi_\nu^{(l)}(1,2)=E_\nu\psi_\nu^{(l)}(1,2), \quad \psi_\nu^{(l)}(1,2)=-\psi_\nu^{(l)}(2,1).$$

1, 2 include spin and space. Also

$$\mathcal{H}_1^{(l,0)}(i)=(-\hbar^2/2m_e)\nabla_{li}^2-2e^2/|\vec{x}_{li}-\vec{R}_l^{(0)}|\ , \quad V^{(l)}(1,2)=e^2/|\vec{x}_{l1}-\vec{x}_{l2}|.$$

Let $\psi_g^{(l)}(1,2)$ be the ground state of the set $\psi_\nu^{(l)}(1,2)$. The ground state for the system described by the Hamiltonian

$$\mathcal{H}_{\text{excitons}}=\sum_{l=1}^{N}[\mathcal{H}_1^{(l,0)}(1)+\mathcal{H}_1^{(l,0)}(2)+V^{(l)}(1,2)]$$

is $|\text{gnd}\rangle=\mathfrak{a}\left\{\prod_{l=1}^{N}\psi_g^{(l)}(\vec{x}_{l1},\sigma_{l1};\vec{x}_{l2},\sigma_{l2})\right\}$,

where $\mathcal{A}\{\cdots\} = (N!)^{-\frac{1}{2}} \sum_P (-1)^P \{\cdots\}$

permutes the electron coordinates. Now define

$$b^{(\nu)\dagger}|\mathrm{gnd}\rangle = \mathcal{A}\left\{\psi_\nu^{(l)}(\vec{x}_{l1},\sigma_{l1};\vec{x}_{l2},\sigma_{l2}) \prod_{l'\neq l} \psi_g^{(l')}(\vec{x}_{l'1},\sigma_{l'1};\vec{x}_{l'2},\sigma_{l'2})\right\}.$$

Then for low temperatures

$$\mathcal{H}_{\mathrm{excitons}} = \sum_{l=1}^{N} \sum_{\nu\neq g} E_\nu b^{(\nu)\dagger} b^{(\nu)} + \mathrm{const.}$$

Fourier transforming, $\beta^{(\nu)} = (N)^{-\frac{1}{2}} \sum_{l=1} e^{-i\vec{k}\cdot\vec{R}_l^{(0)}} b^{(\nu)}$,

gives $\mathcal{H}_{\mathrm{excitons}} = \sum_{\vec{k}} \sum_\nu E_\nu \beta_{\vec{k}}^{(\nu)\dagger} \beta_{\vec{k}}^{(\nu)}$.

Now from our definitions

$$\mathcal{H}_{\mathrm{excitons}} = \mathcal{H}_{\mathrm{atoms}} + \sum_{l=1}^{N} \sum_{i=1}^{2} \left(\frac{-2e^2}{|\vec{x}_{li}-\vec{R}_l|} + \frac{2e^2}{|\vec{x}_{li}-\vec{R}_l^{(0)}|}\right) + \sum_{l=1}^{N} \frac{\hbar^2}{2m_e} \nabla_l^2,$$

so that $\mathcal{H}_{\mathrm{atoms}} = \mathcal{H}_{\mathrm{excitons}} - \sum_{l=1}^{N} \frac{\hbar^2}{2m_e} \nabla_l^2 - \sum_{l=1}^{N} \sum_{i=1}^{2} \left(\frac{-2e^2}{|\vec{x}_{li}-\vec{R}_l|} + \frac{2e^2}{|\vec{x}_{li}-\vec{R}_l^{(0)}|}\right)$.

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*Present address: School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota.

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[2]An exception is the case of two spin-wave processes in rutile antiferromagnets. For a review see R. Loudon, Advan. Phys. 17, 243 (1968).

[3]The conservation laws give

$$K_0 = 2k_s n \sin\theta$$

for one-excitation scattering but only that the two outgoing excitations produced have equal and opposite momenta for two-excitation scattering. Here K_0 is the wave vector of the excitation, n is the index of refraction, and 2θ is the scattering angle. It is particularly notable that in the two-excitation case there is no dependence on θ.

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