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Theory of Inelastic Neutron Scattering from Superfluid He^4 with a Free Surface

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It is shown that one can take advantage of total neutron reflection at a free surface to eliminate volume-proportional terms in inelastic neutron scattering. When this is done, the neutron inelastic-scattering cross section for superfluid He^4 with a free surface is found, within an ideal-fluid model, to contain only ripplon peaks distinctly separated from a smooth background due to nonresonant single-phonon processes occurring near the surface of the liquid. Some numerical results are given for the predicted cross section.

I. INTRODUCTION

In view of the recent interest in the physics of liquid-helium surfaces[1,2] it is useful to consider various means of discovering the nature of the elementary excitations associated with these surfaces. That these excitations should be quantized capillary waves (ripplons) was first proposed by Atkins in 1953.[3] However, to date the only experimental tests of this hypothesis have involved measurements of the temperature dependence of the surface tension, and these measurements do not completely rule out other possibilities.[4] It would be best to have more-direct measurements of the excitation spectrum. In this paper, we first develop the general theory of inelastic neutron scattering from a free plane surface of a bulk material. From this theory emerges the interesting result that if either the angle θ_i between the incident neutron beam and the surface or the angle θ_f between the surface and the scattered beam is less than the critical angle θ_c for total reflection, then inelastic scattering can be a consequence only of absorption or emission of excitations near the surface of the material. In this case, the scattering is proportional only to the surface area exposed to the beam and contains no terms proportional to the volume of the target.

As usual, the scattering cross section is proportional to a dynamical structure factor for the system. In Sec. II we obtain an approximate structure factor from the equations of motion for superfluid He^4 with a free surface. For the case where at least one of the angles θ_i and θ_f is less than θ_c, this structure factor contains sharp peaks, corresponding to the creation or annihilation of ripplons, separated from a broad background owing to the creation or annihilation of phonons near the surface of the liquid. If both θ_i and θ_f are greater than θ_c, giant peaks owing to the creation or annihilation of phonons in the bulk appear in the background. Their contribution is proportional to the volume of the target. These results should be valid for momentum transfers (in cm^{-1}) small compared to the inverse interatomic spacing and energy transfers less than a few degrees Kelvin.

The nature of the ripplon spectrum for large wave numbers is unknown, but there is conjecture[2] that it may have a rotonlike minimum. A neutron scattering experiment is, in principle, capable of providing important information here.

Finally, in Sec. II we give some numerical results for the scattering cross section for a representative experimental arrangement.

II. DERIVATION OF THE CROSS SECTION

In the ensuing derivation we will, purely for convenience, make specific reference to liquid helium. The development will, in fact, be quite general in that, for example, it is applicable to solids as well.

A neutron with coordinate $\vec{r}$ interacts with helium via the potential

$$H_{\text{int}}(\vec{r}) = (2\pi\hbar^2 b/mm_4)\rho_{\text{tot}}(\vec{r}) \equiv V_0\rho_{\text{tot}}(\vec{r}). \tag{1}$$

Here m is the neutron mass, m_4 the mass of a He^4 atom, and b the neutron-He^4 scattering length. For the He^4 mass density $\rho_{\text{tot}}(\vec{r})$, we write

$$\rho_{\text{tot}}(\vec{r}) = \rho(\vec{r})\,\Theta(-z-\zeta(x,y)). \tag{2}$$

The unit step function $\Theta(-z-\zeta(x,y))$ locates the liquid, having mass density $\rho(\vec{r})$, below the surface at $z=\zeta(x,y)$. We expand (2) for small $\zeta(x,y)$ and small deviations $\delta\rho(r)$ of $\rho(\vec{r})$ from its bulk equilibrium value ρ_0; this gives

$$\rho_{\text{tot}}(\vec{r}) \approx \rho_0\Theta(-z) + \rho_0\delta(z)\zeta(x,y) + \delta\rho(\vec{r})\Theta(-z)$$
$$\equiv \rho_0\Theta(-z) + \delta\rho_{\text{tot}}(\vec{r}). \tag{3}$$

It is convenient to rewrite (1) as

$$H_{\text{int}}(\vec{r}) = H_1(z) + H_2(\vec{r}), \tag{4}$$

with

$$H_1(z) = V_0\rho_0\Theta(-z); \quad H_2(r) = V_0\delta\rho_{\text{tot}}(\vec{r}). \tag{5}$$

Now, $H_1(z)$ is responsible for the important effect of elastic total reflection and must be dealt with exactly. $H_2(\vec{r})$ causes inelastic scattering and may be treated in perturbation theory using the eigenfunctions for neutron motion in the presence of $H_1(z)$ as the zero-order states. With these points in mind, the calculation of the differential inelastic scattering cross section per unit solid angle per unit energy proceeds in the standard fashion[5] leading to

$$\frac{d^2\sigma}{d\Omega d(\hbar\omega)} = \frac{b^2}{2\pi\hbar m_4^2}\left(\frac{E_f}{E_i}\right)^{1/2}\int_{-\infty}^{\infty} dt\, e^{i\omega(t-t')} \times \int d^3r\, d^3r'\, \psi_i^{(+)*}(\vec{r})\psi_f^{(-)}(\vec{r})\psi_f^{(-)*}(\vec{r}')\psi_i^{(+)}(\vec{r}') \times \langle \delta\rho_{\text{tot}}(\vec{r}t)\delta\rho_{\text{tot}}(\vec{r}'t')\rangle . \quad (6)$$

This cross section does not, of course, account for the elastically reflected or transmitted neutrons, which leave the surface in definite and simply determined directions. Here E_i is the incident neutron energy, E_f is the final neutron energy, $\hbar\omega = E_i - E_f$, and $\langle \delta\rho_{\text{tot}}(\vec{r}t)\delta\rho_{\text{tot}}(\vec{r}'t')\rangle$ is the density-fluctuation correlation function for the system. $\psi_i^{(+)}$ is, in the language of formal scattering theory,[6] an incoming-neutron-state solution to the problem in the presence of $H_1(z)$ and $\psi_f^{(-)}$ is an outgoing state. These states are quite easily determined for $H_1(z)$, which is in essence just a one-dimensional step-function potential.

We now specialize to the most interesting situation where the incoming neutron beam is incident on the surface from above the liquid and the outgoing beam is detected above the liquid. In this case,

$$\psi_i^{(+)}(\vec{r}) = e^{i\vec{k}_{\parallel i}\cdot\vec{r}} \times \begin{cases} e^{-ik_i z} + Be^{ik_i z} & \text{for } z > 0, \\ \psi_i^{(+)}(0)\, e^{-iq_i z} & \text{for } z \le 0; \end{cases} \quad (7a)$$

$$\psi_f^{(-)}(\vec{r}) = e^{i\vec{k}_{\parallel f}\cdot\vec{r}} \times \begin{cases} De^{-ik_f z} + e^{ik_f z} & \text{for } z > 0, \\ \psi_f^{(-)}(0)\, e^{iq_f z} & \text{for } z \le 0. \end{cases} \quad (7b)$$

In (7), $\vec{k}_{\parallel_i}$ and $\vec{k}_{\parallel_f}$ are the initial and final wave vectors parallel to the surface, and k_i and k_f are the initial and final z components of the wave vectors. Further,

$$q_i = \left[\frac{2m}{\hbar^2}\left(\frac{\hbar^2 k_i^2}{2m} - \rho_0 V_0\right)\right]^{1/2}; \qquad q_f = \left[\frac{2m}{\hbar^2}\left(\frac{\hbar^2 k_f^2}{2m} - \rho_0 V_0\right)\right]^{1/2}. \quad (8)$$

The constants B, $\psi_i^{(+)}(0)$, D, and $\psi_f^{(-)}(0)$ are determined by requiring that the wave functions and their derivatives be continuous at $z = 0$.

Inserting Eqs. (7) into (6), noting that $\delta\rho_{\text{tot}}(\vec{r}t) = 0$ for $z > 0$ and that $\langle \delta\rho_{\text{tot}}(\vec{r}t)\delta\rho_{\text{tot}}(\vec{r}'t')\rangle$ must be a function only of coordinate differences in the $x-y$ plane, gives

$$\frac{d^2\sigma}{d\Omega d(\hbar\omega)} = \frac{Ab^2}{2\pi\hbar m_4^2}\left(\frac{E_f}{E_i}\right)^{1/2} |\psi_f^{(-)}(0)\,\psi_i^{(+)}(0)|^2 \times S_{\text{tot}}(\vec{k}_{\parallel}, \kappa, \omega), \quad (9)$$

where the dynamical structure factor $S_{\text{tot}}(\vec{k}_{\parallel}, \kappa, \omega)$ is defined by

$$S_{\text{tot}}(k_{\parallel}, \kappa, \omega) \equiv \int_{-\infty}^{\infty} dx\, dy \int_{-\infty}^{\infty} dt \int_{-\infty}^{0} dz \int_{-\infty}^{0} dz'\, e^{i\omega(t-t')} \times e^{-i\vec{k}_{\parallel}\cdot(\vec{r}-\vec{r}')} e^{\kappa^* z} e^{\kappa z'} \langle \delta\rho_{\text{tot}}(\vec{r}t)\delta\rho_{\text{tot}}(\vec{r}'t')\rangle . \quad (10)$$

In (9), A is the surface area, while in (10),

$$\vec{k}_{\parallel} = \vec{k}_{\parallel_i} - \vec{k}_{\parallel_f}; \quad \kappa = -iq_i - iq_f . \quad (11)$$

It should be noted that as a consequence of the lack of translational invariance of $\langle \delta\rho_{\text{tot}}(\vec{r}t)\,\delta\rho_{\text{tot}}(\vec{r}'t')\rangle$ in the z direction, the z component of the momentum (of the neutron plus the He^4 excitations involved) will not be conserved in the scattering.

We shall be particularly interested in the case when κ has a positive real part. For example, when $\hbar^2 q_i^2/2m < \rho_0 V_0$—corresponding to elastic total reflection of the (unperturbed) incident beam—then the electric-dipole matrix element is [see Eq. (8)]

$$q_i = -i\left[\frac{2m}{\hbar^2}\left(\rho_0 V_0 - \frac{\hbar^2 k_i^2}{2m}\right)\right]^{1/2} \equiv -i\kappa_i . \quad (12)$$

In this case, we see from (10) that the z and z' integrations cut off at a distance of order $[\text{Re}\kappa]^{-1}$ from the liquid surface. Consequently, only fluctuations near the liquid surface are probed.

We now turn to an approximate calculation of $S_{\text{tot}}(\vec{k}_{\parallel}, \kappa, \omega)$ for superfluid He^4.

III. STRUCTURE FACTOR

In order to obtain a form for the structure factor, expected to be valid for wave numbers small compared to the inverse interatomic spacing and frequencies (in temperature units) small compared to, say, the bulk roton frequency, we will calculate the retarded density-density response function $\chi(\vec{k}_{\parallel}, \kappa, \omega)$ from the equations of motion for a superfluid with a free surface and then make use of the identity[7]

$$S_{\text{tot}}(\vec{k}_{\parallel}, \kappa, \omega) = [2\hbar/(1 - e^{-\beta\hbar\omega})]\,\text{Im}\,\chi(\vec{k}_{\parallel}, \kappa, \omega + i\epsilon); \quad \epsilon = 0^+ . \quad (13)$$

To commence, we recall[7] that the response of the density to an external potential $\delta U(\vec{k}_{\parallel}, z', \omega)$ coupled to the density is given by

$$\delta\rho_{tot}(\vec{k}_\parallel, z, \omega) = \int_{-\infty}^{\infty} dz\, \chi(\vec{k}_\parallel, z, z', \omega)\delta U(\vec{k}_\parallel, z', \omega). \tag{14}$$

Taking $\delta U(\vec{k}_\parallel, z', \omega)$ to be of the form

$$\delta U(\vec{k}_\parallel, z', \omega) = \delta U(\vec{k}_\parallel, \kappa, \omega)\, e^{\kappa z'}\, \Theta(-z'), \tag{15}$$

allows us to write

$$\frac{\delta\rho_{tot}(\vec{k}_\parallel, z, \omega)}{\delta U(\vec{k}_\parallel, \kappa, \omega)} = \int_{-\infty}^{0} dz' e^{\kappa z'} \chi(\vec{k}_\parallel, z, z', \omega). \tag{16}$$

The left-hand side of (16) is the quantity most conveniently calculated from the equations of motion. To obtain $\chi(\vec{k}_\parallel, \kappa, \omega)$ we use

$$\chi(\vec{k}_\parallel, \kappa, \omega) = \int_{-\infty}^{0} dz \int_{-\infty}^{0} dz' e^{\kappa^* z} e^{\kappa z'} \chi(\vec{k}_\parallel, z, z', \omega) = \frac{\delta\rho_{tot}(\vec{k}_\parallel, \kappa^*, \omega)}{\delta U(\vec{k}_\parallel, \kappa, \omega)}. \tag{17}$$

The superfluid equations of motion are, in linearized form,

$$\frac{\partial\rho(\vec{r}t)}{\partial t} + \rho_0 \nabla\cdot\vec{v}(\vec{r}t) = 0, \tag{18a}$$

$$\rho_0 \frac{\partial \vec{v}(\vec{r}t)}{\partial t} + \nabla P(\vec{r}t) = \rho_0 \delta U(\vec{r}t). \tag{18b}$$

$P(\vec{r}t)$ is the local pressure and $\vec{v}(\vec{r}t)$ is the local superfluid velocity. We have ignored dissipation and assumed temperatures sufficiently low that the normal component of the fluid may be neglected. Equations (18) are supplemented by boundary conditions at the surface given by[8]

$$v_z(\vec{r}t)|_{z=0} = \frac{\partial\zeta(x, y, t)}{\partial t}, \tag{19a}$$

$$\delta P(\vec{r}t)|_{z=0} = -\alpha\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\zeta(x, y, t), \tag{19b}$$

where δP is the pressure deviation in the liquid and α is the surface tension. Since the flow is irrotational, it is convenient to introduce a velocity potential $\varphi(\vec{r}t)$ by

$$\vec{v}(\vec{r}t) = \nabla\varphi(\vec{r}t). \tag{20}$$

Combining Eqs. (18) with (20) and using

$$\delta P(\vec{r}t) = s^2 \delta\rho(\vec{r}t), \tag{21}$$

one finds

$$\frac{\partial^2\varphi(\vec{r}t)}{\partial t^2} - s^2\nabla^2\varphi(\vec{r}t) = + \frac{\partial U(\vec{r}t)}{\partial t}. \tag{22}$$

Here s is the zero-temperature sound velocity in bulk He^4. Fourier transformed in time and the spacial coordinates x and y, (22) becomes

$$\frac{\partial^2\varphi(\vec{k}_\parallel, z, \omega)}{\partial z^2} - \left(k_\parallel^2 - \frac{\omega^2}{s^2}\right)\varphi(\vec{k}_\parallel, z, \omega) = \frac{i\omega}{s^2}\delta U(\vec{k}_\parallel, \kappa, \omega) e^{\kappa z}, \tag{23}$$

where we have used (15).

The general solution to (23) is

$$\varphi(\vec{k}_\parallel, z, \omega) = + \frac{i\omega/s^2}{\kappa^2 - (\vec{k}_\parallel^2 - \omega^2/s^2)}\delta U(\vec{k}_\parallel, \kappa, \omega) e^{\kappa z} + \varphi_1(\vec{k}_\parallel, \kappa, \omega) e^{\kappa_1 z}, \tag{24}$$

with

$$\kappa_1^2 = k_\parallel^2 - \omega^2/s^2. \tag{25}$$

The term $\varphi_1 e^{\kappa_1 z}$ is a solution to the homogeneous version of (23) with the coefficient φ_1 yet to be determined. Fourier transformed, Eqs. (19), with (20) and (21), become

$$\left.\frac{\partial\varphi(\vec{k}_\parallel, z, \omega)}{\partial z}\right|_{z=0} = -i\omega\zeta(\vec{k}_\parallel, \omega), \tag{26}$$

$$s^2\delta\rho(\vec{k}_\parallel, z, \omega)|_{z=0} = \alpha k^2 \zeta(\vec{k}_\parallel, \omega). \tag{27}$$

Putting together (18b), (20), and (21) gives

$$s^2\delta\rho(\vec{k}_\parallel, z, \omega) = \rho_0\delta U(\vec{k}_\parallel, \kappa, \omega) e^{\kappa z} + i\omega\rho_0\varphi(\vec{k}_\parallel, z, \omega). \tag{28}$$

Equations (24)–(28) are easily solved, and after some algebra one finds

$$\frac{\delta\zeta(\vec{k}_\parallel, \omega)}{\delta U(\vec{k}_\parallel, \kappa, \omega)} = -\frac{\kappa\kappa_1 + k_\parallel^2}{(\kappa + \kappa_1)(\omega^2 - \omega_r^2)}, \tag{29}$$

$$\frac{\delta\rho(\vec{k}_\parallel, z, \omega)}{\delta U(\vec{k}_\parallel, \kappa, \omega)} = -\frac{\rho_0 e^{\kappa z}}{s^2}\left(\frac{k_\parallel^2 - \kappa^2}{\kappa^2 - \kappa_1^2}\right) - \frac{\rho_0 e^{\kappa_1 z}}{s^2}\left(\frac{\kappa_1\kappa + k_\parallel^2}{\kappa(\kappa + \kappa_1)(\omega^2 - \omega_r^2)} + \frac{\kappa^2 - k_\parallel^2}{\kappa^2 - \kappa_1^2}\right). \tag{30}$$

Here

$$\omega_r = \left(\frac{\alpha}{\rho_0}\kappa_1 k_\parallel^2\right)^{1/2} \tag{31}$$

is the classical frequency for a capillary wave in a compressible liquid.

To proceed, we note from (3) that

$$\delta\rho_{tot}(\vec{k}_\parallel, z, \omega) = \rho_0\delta(z)\zeta(\vec{k}_\parallel, \omega) + \Theta(-z)\delta\rho(\vec{k}_\parallel, z, \omega). \tag{32}$$

Hence, we combine Eqs. (29)–(32) with (17) to obtain, after some rearrangement,

$$\chi(\vec{k}_\parallel, \kappa, \omega) = -\rho_0\kappa_1\left(\frac{\omega^2}{s^2}\frac{1}{\kappa_1(\kappa_1 + \kappa^*)} + 1\right)\left(\frac{\omega^2}{s^2}\frac{1}{\kappa_1(\kappa_1 + \kappa)} + 1\right)(\omega^2 - \omega_r^2)^{-1} + \frac{\rho_0}{s^2(\kappa + \kappa^*)}\left[1 + \frac{\omega^2/s^2}{(\kappa^* + \kappa_1)(\kappa + \kappa_1)}\left(\frac{\kappa + \kappa^*}{\kappa_1} + 1\right)\right]. \tag{33}$$

In order to obtain the required structure factor, we need only insert (33) into (13). The results must be discussed in terms of several cases: (i)
(i) For $\kappa=\kappa'+i\kappa''$, κ' and κ'' real, and $k_\parallel^2>\omega^2/s^2$, one has

$$S_{\text{tot}}(\vec{k},\kappa,\omega)=\frac{\pi\rho_0\hbar\kappa_1}{\omega(1-e^{-\beta\hbar\omega})}\left|\frac{\omega^2}{s^2}\frac{1}{\kappa_1(\kappa_1+\kappa)}+1\right|^2 \times[\delta(\omega-\omega_r)+\delta(\omega+\omega_r)]. \quad (34)$$

(ii) For $\kappa=\kappa'+i\kappa''$, κ' and κ'' real, $\kappa'>0$, and $k_\parallel^2\leqslant\omega^2/s^2$, one has

$$S_{\text{tot}}(\vec{k}_\parallel,\kappa,\omega)=\frac{2\hbar\rho_0}{(1-e^{-\beta\hbar\omega})}\frac{\omega^4 g_1}{(\omega^4+\omega_q^4)|\kappa^2+q_1^2|^2} \times\left|(\kappa^2-k_\parallel^2)+\frac{\sigma\kappa k^2}{\rho_0 s^2}\right|^2. \quad (35)$$

(iii) For $\kappa=-ik_z$, k_z real, and $k_\parallel^2\leqslant\omega^2/s^2$,

$$S_{\text{tot}}(\vec{k}_\parallel,\kappa,\omega)=\frac{2\hbar\rho_0}{(1-e^{-\beta\hbar\omega})}\left(\frac{\pi Lk^2}{2\omega}[\delta(\omega-sk)+\delta(\omega+sk)] + \frac{\omega^2 q_1}{(\omega^4+\omega_q^4)(\omega^2-s^2k^2)^2}[(sk)^4+\omega_{k_z}^4]\right). \quad (36)$$

In the above, L is the depth of the liquid and

$$q_1=\left(\frac{\omega^2}{s^2}-k_\parallel^2\right)^{1/2}\operatorname{sgn}\omega,\quad \kappa_1=\left(k_\parallel^2-\frac{\omega^2}{s^2}\right)^{1/2},$$
$$\omega_r=\left(\frac{\alpha\kappa_1 k_\parallel^2}{\rho_0}\right)^{1/2},\quad \omega_q^2=\frac{\alpha q_1 k_\parallel^2}{\rho_0}, \quad (37)$$
$$\omega_{k_z}^2=\frac{\alpha k_z k_\parallel^2}{\rho_0},\quad k^2=k_\parallel^2+k_z^2.$$

We conclude this section with a discussion of these results. First, it is quite easy to prove $\omega_r < sk_\parallel$ for all finite $k_\parallel$. Hence, when κ has a positive real part, the structure factor [Eqs. (34) and (35)] consists of δ-function peaks, corresponding to ripplon creation or annihilation, separated from broad backgrounds, starting at $\omega=\pm sk_\parallel$, corresponding to creation or annihilation of phonons near the surface. This behavior is sketched in Fig. 1. When κ is imaginary, there appear δ-function peaks [Eq. (36)] in these backgrounds corresponding to creation or annihilation of single phonons in the bulk. In this case, both the initial and final neutron wave functions extend throughout the bulk, and the cross section is proportional to the total volume of the sample. This scattering due to creation or annihilation of phonons in the bulk has been thoroughly studied experimentally.[9]

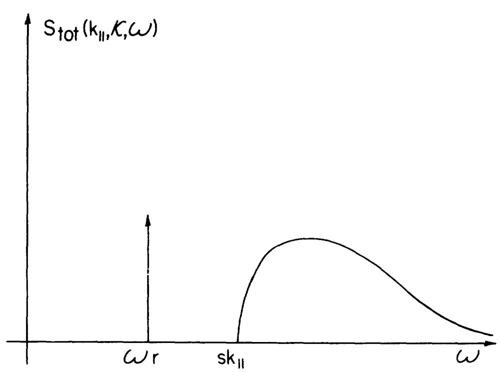

FIG. 1. Sketch of $S_{\text{tot}}(\vec{k}_\parallel,\kappa,\omega)$ for $\omega>0$ when $\text{Re}\kappa>0$.

IV. NUMERICAL RESULTS FOR THE CROSS SECTION

In this section, we investigate the predictions of our formalism for a representative experimental arrangement. We assume temperatures low enough so that the scattering involves only the creation of excitations. Further, we consider only the energy-integrated cross section. It turns out that this cross section, for appropriately chosen beam energies and incoming and outgoing angles, is quite sensitive to the form of the ripplon dispersion relation, the quantity of primary interest here.

We choose the angle of the incoming beam with the surface to be just below the angle for critical reflection (~0.5 deg for neutrons with energies of the order of 4*K*). The number of neutrons detected per second per unit solid angle at θ_f, the angle between the scattered beam and the surface (in this section we assume that the incident and scattered beams lie in a plane perpendicular to the liquid surface), is

$$N=I_0\int_0^{E_i}d(\hbar\omega)\frac{d^2\sigma}{d\Omega\, d(\hbar\omega)}\equiv I_0\frac{d\sigma}{d\Omega}, \quad (38)$$

where I_0A is the number of neutrons per second incident on a surface of area A. For the moment, we assume that only the ripplon peak contributes to (38), a point to which we will return shortly. In this case, we obtain the cross section simply by integrating (9) over the positive energy ripplon peak in (34), a procedure which yields

$$\frac{1}{A}\frac{d\sigma}{d\Omega}=\frac{b^2}{2\pi m_4^2}\left(\frac{E_f}{E_i}\right)^{1/2}\frac{\pi\rho_0\hbar\kappa_1}{\omega_r}|\psi_f^{(-)}(0)\psi_i^{(+)}(0)|^2 \times\left|\frac{\omega^2}{s^2}\frac{1}{\kappa_1(\kappa_1+k)}+1\right|^2 \Big/ \left|1-\frac{\partial\omega_r}{\partial\omega}\right|_{\omega=\omega_r} \quad (39)$$

From (7) one easily finds

$$|\psi_i^{(+)}(0)|^2=\frac{4E_i\sin^2\theta_i}{V_0}, \quad (40)$$

$$|\psi_f^{(-)}(0)|^2 = \frac{4E_f \sin^2\theta_f}{[(E_f \sin^2\theta_f)^{1/2} + (E_f \sin^2\theta_f - V_0)^{1/2}]^2}. \tag{41}$$

Now, since $V_0 = 1.95\times10^{-4}K$ is so small, and the critical angle for total reflection θ_{ic} is given by

$$\theta_{ic} = \left(\sin^{-1}\frac{V_0}{E_i}\right)^{1/2} \approx \left(\frac{V_0}{E_i}\right)^{1/2}, \tag{42}$$

the expression (39) may be simplified to

$$\frac{1}{A}\frac{d\sigma}{d\Omega} = 2.58\times10^{-11}\frac{\kappa_1 s}{\omega_r}\left(\frac{E_f}{E_i}\right)^{1/2}\left(\frac{\theta_i}{\theta_{ic}}\right)^2 \times \left|\frac{\omega_r^2}{s^2}\frac{1}{\kappa_1(\kappa_1+\kappa)}+1\right|^2 \Big/ \left|1-\frac{\partial\omega_r}{\partial\omega}\right|_{\omega=\omega_r}. \tag{43}$$

Here we have used the numbers $\rho_0 = 0.145$ g/cm^3, $\alpha = 0.378$ ergs/cm^2, $b = 3.0\times10^{-13}$ cm, and $s = 2.38\times10^4$ cm/sec appropriate to He^4. To continue, we note that

$$k_\parallel = (2m/\hbar^2)^{1/2}[(\sqrt{E_i})\cos\theta_i - (\sqrt{E_f})\cos\theta_f]. \tag{44}$$

Using (44) in conjunction with (25) and (31) gives

$$\left(1-\frac{\partial\omega_r}{\partial\omega}\right)_{\omega=\omega_r} = 1 + \frac{\alpha k_\parallel^2}{2\rho_0 s^2\kappa_1} - \left(\frac{\alpha^2 m}{2\rho_0^2}\right)^{1/2}\frac{k_\parallel}{\kappa_1\omega_r\sqrt{E_f}}\left(\frac{k_\parallel^2}{2}+\kappa_1^2\right)\cos\theta_f. \tag{45}$$

By combining (25) and (31) we may find one equation for ω_r in terms of $k_\parallel$. A useful form for the result is

$$\frac{\hbar\omega_r(x)}{k_B} = \frac{\hbar\rho_0 s^3}{\alpha k_B}x^{3/2}\left\{\left[1+\left(\frac{x}{2}\right)^2\right]^{1/2} - \frac{x}{2}\right\}^{1/2}, \tag{46}$$

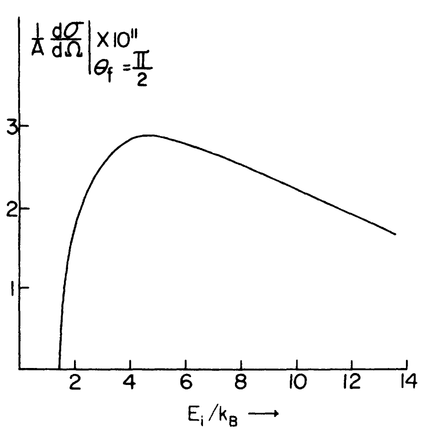

FIG. 2. Plot of $(1/A)\ d\sigma/d\Omega$ for $\theta_f = \frac{1}{2}\pi$, $\theta_i = 0.9\theta_{ic}$, and $E_i/k_B < 13.6K$.

$$x \equiv \alpha k/2\rho_0 s^2. \tag{47}$$

Since $E_f = E_i - \hbar\omega_r$, we may combine (44) and (47) to find a second relation

$$x = \frac{\alpha}{2\rho_0 s^2}\left(\frac{2mk_B}{\hbar^2}\right)^{1/2}\left[\left(\frac{E_i}{k_B}\right)^{1/2}\cos\theta_i - \left(\frac{E_i - \hbar\omega_r(x)}{k_B}\right)^{1/2}\cos\theta_f\right]. \tag{48}$$

The kinematical problem is thus reduced to solving (46) and (48) for x and $\omega_r(x)$ in terms of E_i, $\cos\theta_f$, and $\cos\theta_i$. E_f, κ_1, and $k_\parallel$ are then easily found. Equations (46) and (48) are most easily solved via graphical techniques.

Let us now examine the domain of validity of (43). Clearly, one requirement is that ω_r be greater than zero. The consequences of this are most easily understood when $\theta_f = \frac{1}{2}\pi$ and when x may be considered small. Using (46) and (48) our requirement then becomes ($\theta_i \ll 1$)

$$\frac{E_i}{k_B} \geq \hbar^4\frac{\alpha}{\rho_0 k_B}\left(\frac{2m}{\hbar^2}\right)^3 = 1.66K. \tag{49}$$

Removal of the restriction that x be small only slightly changes this result; the correct minimum being $1.5K$. For a general θ_f one then expects that $d\sigma/d\Omega$ be zero for energies less than some minimum value. The other requirement is that E_i be less than $\hbar s k_\parallel$ so that the phonon background does not contribute to the scattering. For the simple case $\theta_i < \theta_{ic} \ll 1$ and $\theta_f = \frac{1}{2}\pi$ we have

$$k_\parallel = [(2m/\hbar^2)E_i]^{1/2}. \tag{50}$$

In this case, then, for

$$E_i/k_B < 2ms^2/k_B = 13.6K, \tag{51}$$

the background will not contribute. For angles θ_f near $\frac{1}{2}\pi$, then, there is a considerable range of energies over which (43) is valid.

In Fig. 2, we plot $(1/A)\ d\sigma/d\Omega$ for $\theta_f = \frac{1}{2}\pi$, θ_i

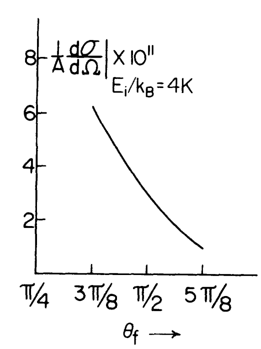

FIG. 3. Plot of $(1/A)d\sigma/d\Omega$ for $E_i/k_B = 4K$, $\theta_i = 0.9\theta_{ic}$, and $\frac{3}{8}\pi \leq \theta_f \leq \frac{5}{8}\pi$.

$=0.9\theta_{ic}$, and $E_i/k_B < 13.6K$. The cross section rises from the low-energy cutoff (which is quite sensitive to the form of the ripplon spectrum), and then bends over. The bending is a consequence of the fact that taking $\theta_i = 0.9\theta_{ic}$ makes θ_i a function of E_i. Were θ_i to be held constant, the cross section would rise monotonically with E_i. Finally, in Fig. 3, we plot the cross section for fixed $E_i/k_B = 4K$, $\theta_i = 0.9\theta_{ic}$, and $\frac{3}{8}\pi \leq \theta_f \leq \frac{5}{8}\pi$.

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Quantum Theory of Nonlinear Optical Processes with Time-Dependent Pump Amplitude and Phase: Frequency Conversion*

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The behavior of a simple theoretical model of a quantum-frequency converter with a time-dependent pump amplitude and phase is analyzed quantum mechanically. A sufficient condition ensuring periodic frequency conversion between the modes is found and exact solutions are given in these cases. The complete time-dependent density matrices which give the most complete statistical description of the system are presented for a variety of initial states of the system. In the general case where the sufficient condition is not satisfied, time-dependent-perturbation-theory results give corrections to the amplitude and the period of the energy exchange between the modes. Comparisons are made between these results and those of other authors.

I. INTRODUCTION

It is well known that nonlinear optical effects arise as a result of the nonlinear response of a medium to intense light fields obtainable in laser beams.[1] An important class of these phenomena involve nonlinear coupling between electromagnetic waves, usually referred to as parametric interactions.[2,3] The fundamental physical process underlying nonlinear parametric interactions has come to play a central role in several physical phenomena of interest. These include Raman and Brillouin effects, Stokes and anti-Stokes generations, etc. All these effects involve nonlinear coupling between various types of boson excitations such as phonons, spin waves, plasmons, rotons, polaritons, etc., as well as electromagnetic waves.[4]

In the optical regime two of the most important nonlinear parametric interactions are frequency conversion and parametric amplification where three electromagnetic modes are coupled. A quantum-mechanical model suitable for discussions of these effects was proposed some time ago by Louisell, Yariv, and Siegman.[5] The proposed model is macroscopic in that one introduces a