Relaxation-Time Approximation for the Mobility of Electrons in Helium

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Kubo's theory of irreversible quantum statistics is applied to the problem of electron mobility in a hard-core gas. Without any assumptions regarding a relaxation time, the mobility is evaluated by cumulant techniques to first order in the gas density and second order in the scattering length. A momentum-dependent relaxation time appears in a natural manner, as does the criterion for validity of the classical Langevin theory.

I. INTRODUCTION

THE calculation of electron mobility in a hard-core gas was first carried out by Langevin who used classical transport theory.[1] It has been pointed out, however, that the kinetic equation presupposes in its derivation the existence of a single relaxation time and may not always be valid.[2–4]

Recently, significant advances have been made in the statistical theory of irreversible processes. In a problem closely related to the mobility, Greenwood[5] derived a formal expression which Edwards[6] evaluated to yield the usual solution to the Boltzmann equation for conductivity in a metal with a periodic lattice potential, Fermi–Dirac electron distribution, and randomly located impurities. Lax[7] has treated the same problem and also obtained the Boltzmann equation in the limit of weak coupling. Kohn and Luttinger[3] started from the equation of motion of the density matrix and, by adiabatically switching on an electric field, derived the Boltzmann equation for randomly distributed scattering centers. Kubo[4] has treated irreversible processes from first principles and derived an exact formal expression for the response to a weak force. Nakano[8] evaluated the Kubo expression and obtained the Grüneisen formula for conductivity in metals by assuming the existence of a relaxation time. Chester and Thellung[9] have shown that for a metal the Kubo formulation leads to the Boltzmann equation, with the sole assumption that the scattering is elastic.

In this paper, the mobility for a particular system is evaluated from Kubo's quantum-mechanical formulation of irreversible statistical mechanics without assuming the existence of a relaxation time, which, under certain conditions, is obtained as a *direct consequence* of the theory employed. The calculation is carried to second order in the scattering length by an extension of the techniques used by one of us (M.H.C.) in the evaluation of the free energy of an electron in a hard-core gas[10] (hereafter referred to as I).

II. STATEMENT OF THE PROBLEM

The mobility μ of an electron in a gas of density ρ under the influence of a weak external electric field E is evaluated under the following assumptions: (1) Interactions between electrons can be neglected, (2) an electron sees the gas as a system of randomly located, stationary, hard-core scatterers, and (3) collective properties of the gas enter the calculation through ρ. This model is applicable to helium where the atom mass is much greater than the electron mass and the s-wave scattering length is positive.

The total Hamiltonian H is

$$H=H_{\{N\}}+H_E, \tag{1}$$

$$H_{\{N\}}=T_e+V_{\{N\}}. \tag{2}$$

$H_{\{N\}}$ is the Hamiltonian of an electron in equilibrium with N hard-core scatterers, T_e the free-electron Hamiltonian, $V_{\{N\}}$ the interaction of an electron with N scattering centers, and H_E the Hamiltonian of an electron in an external field E.

$$T_e=p^2/2m, \tag{3}$$

$$V_{\{N\}}=\sum_{j=1}^{N} V(r_{je})\equiv\sum_{j=1}^{N} V_j, \tag{4}$$

$$V(r)=\infty, \quad r<a$$
$$=0, \quad \text{otherwise}, \tag{5}$$

$$H_E=-zeE. \tag{6}$$

V_j is the assumed hard-core interaction between the

* National Aeronautics and Space Administration graduate trainee during part of this work.

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electron and the jth atom, r_{je} is the distance between the electron and the jth atom, and a is the hard-core radius. The mobility, as derived by Kubo,[4] is given by

$$\mu=ie\ \mathrm{tr}\left\{\int_0^t \mathbf{R}(t')\,dt'\ p_z\right\} \Big/ \left(\hbar m\ \mathrm{tr}\{\exp(-\beta H_{\{N\}}/\hbar)\}\right), \tag{7}$$

where

$$\mathbf{R}(t)=\exp(-itH_{\{N\}}/\hbar) \times[z,\exp(-\beta H_{\{N\}})]\exp(itH_{\{N\}}/\hbar). \tag{8}$$

III. CUMULANT EXPANSION

An exact evaluation of μ would require the eigenfunctions and eigenvalues of $H_{\{N\}}$ which are not known. However, an approximate expression can be obtained by rewriting $\mathbf{R}(t)$ as a generalized exponential and using Kubo's cumulant techniques[11] to derive a cluster expansion for it. Putting $\hbar=m=1$, and using Eqs.(2) (3), (4), and (8), we have

$$\mathbf{R}=\exp\{-it(p^2/2+\sum_j V_j)\} \times[z,\exp\{-\beta(p^2/2+\sum_j V_j)\}]\exp\{it(p^2/2+\sum_j V_j)\}. \tag{9}$$

We now use the identity

$$\exp[\alpha(b+c)]=\exp(\alpha b) \times\exp\left\{\int_0^\alpha \exp(-bs)\,c\exp(bs)\,ds\right\}, \tag{10}$$

and define

$$g_j(x)=\exp\left\{-\int_0^x \exp(p^2s/2)\,V_j\exp(-p^2s/2)\,ds\right\} \equiv 1+f_j(x) \tag{11}$$

to rewrite Eq. (9) as follows:

$$\mathbf{R}=\exp\{-itp^2/2\}\prod_j\{1+f_j(it)\}[z,\exp\{-\beta p^2/2\} \times\prod_j\{1+f_j(\beta)\}]\exp\{itp^2/2\}\prod_j\{1+f_j^*(it)\}. \tag{12}$$

Following Kubo, we introduce a levelling operator $\mathbf{L}$ which levels (eliminates) all powers of f greater than one [e.g., $\mathbf{L}f_i(x)=f_i(x)$; $\mathbf{L}f_i^2(x)=0$; $\mathbf{L}f_i(x)f_j(x)=f_i(x)f_j(x)$, $i\neq j$; $\mathbf{L}f_i(x)f_i(y)=f_i(x)f_i(y)$, $x\neq y$]. We thus get

$$\mathbf{R}=\exp\{-itp^2/2\}\ \exp_{\mathrm{L}}\{\sum_j f_j(it)\}[z,\exp\{-\beta p^2/2\} \times\exp_{\mathrm{L}}\{\sum_j f_j(\beta)\}]\exp\{itp^2/2\}\ \exp_{\mathrm{L}}\{\sum_j f_j^*(it)\}. \tag{13}$$

Finally, introducing an ordering operator $\mathbf{O}$ we can write

$$\mathbf{R}=\exp_{\mathrm{O,L}}\{\sum_j[f_j(it)+f_j(\beta)+f_j^*(it)]\}. \tag{14}$$

The ordering operation means that proceeding from left to right, we have (1) $\exp(-itp^2/2)$, (2) all $f(it)$'s (3) $[z,\exp(-\beta p^2/2)\times$all $f(\beta)$'s$]$, (4) $\exp(itp^2/2)$, and (5) all $f^*(it)$'s.[12]

We now evaluate μ using the representation in which T_e is diagonal, as only the diagonal elements of $\mathbf{R}$, R_{kk}, are needed. R_{kk} is given by

$$R_{kk}=\langle\mathbf{k}\mid\mathbf{R}\mid\mathbf{k}\rangle =\langle\mathbf{k}\mid\exp_{\mathrm{O,L}}\{\sum_j[f_j(it)+f_j(\beta)+f_j^*(it)]\}\mid\mathbf{k}\rangle. \tag{15}$$

The average of an operator $\mathbf{B}$ is defined as

$$\langle\mathbf{B}\rangle\equiv C^{-1}\langle\mathbf{k}\mid\mathbf{B}_{\mathrm{O,L}}\mid\mathbf{k}\rangle, \tag{16}$$

where the normalization C is

$$C=\langle\mathbf{k}\mid\mathbf{1}_{\mathrm{O,L}}\mid\mathbf{k}\rangle \equiv\langle\mathbf{k}\mid\exp(-itp^2/2)[z,\exp(-\beta p^2/2)]\exp(itp^2/2)\mid\mathbf{k}\rangle =i\Omega(2\pi)^{-3}(\partial/\partial k_z)\exp(-\beta k^2/2) \tag{17}$$

and Ω is the volume. From Eqs. (15), (16), and (17), we get

$$R_{kk}=C\langle\exp\{\sum_j[f_j(it)+f_j(\beta)+f_j^*(it)]\}\rangle. \tag{18}$$

R_{kk} is now in a form which enables us to utilize generalized cumulant expansion techniques.[11] Thus,

$$R_{kk}=C\exp\langle\exp\{\sum_j[f_j(it)+f_j(\beta)+f_j^*(it)]\}-1\rangle_{\mathrm{cum}}, \tag{19}$$

where the symbol $\langle\ \rangle_{\mathrm{cum}}$ means cumulant average. In evaluating (19), the cumulant average must be taken before the levelling operation is performed.

The remainder of this paper will be concerned with a calculation of the mobility by expanding the cumulant to first order in the f's. This is equivalent to an expansion in powers of the density. The first term is given by

$$R_{kk}=C\exp\langle\sum_j[f_j(it)+f_j(\beta)+f_j^*(it)]\rangle_{\mathrm{cum}} =C\exp\{\sum_j[\langle g_j(it)\rangle+\langle g_j(\beta)\rangle+\langle g_j^*(it)\rangle-3]\}, \tag{20}$$

where the properties of cumulants and Eq. (11) have been employed. The averages of the g's are given by

$$\langle g_j(it)\rangle=C^{-1}\langle\mathbf{k}\mid\exp\{-it(p^2/2+V_j)\} \times[z,\exp(-\beta p^2/2)]\exp(itp^2/2)\mid\mathbf{k}\rangle, \tag{21}$$

$$\langle g_j(\beta)\rangle=C^{-1}\langle\mathbf{k}\mid\exp(-itp^2/2) \times[z,\exp\{-\beta(p^2/2+V_j)\}]\exp(itp^2/2)\mid\mathbf{k}\rangle, \tag{22}$$

$$\langle g_j^*(it)\rangle=C^{-1}\langle\mathbf{k}\mid\exp(-itp^2/2)[z,\exp(-\beta p^2/2)] \times\exp\{it(p^2/2+V_j)\}\mid\mathbf{k}\rangle. \tag{23}$$

[11] R. Kubo, J. Phys. Soc. Japan **17**, 1100 (1962).

[12] Following Kubo, we have used $\exp_{\mathrm{L,O}}$ to stand for $_{\mathrm{L,O}}\exp$.

Note that the ordering operation makes the average of the operator g a functional of its argument. As a result of the cumulant expansion, Eq. (20) for R_{kk} contains the Hamiltonian for N electrons each interacting with a single scattering center, instead of the more complicated Hamiltonian for one electron interacting with N scattering centers.

IV. MATRIX ELEMENTS

From Eqs. (7), (17), and (20), the mobility is

$$\mu=-\frac{e\Omega(2\pi)^{-3}}{\mathrm{tr}\{\exp(-\beta H_{\{N\}})\}}\int d\mathbf{k}\,k_z\frac{\partial}{\partial k_z}\exp(-\beta k^2/2)\int_0^t dt' \times\exp\sum_j[\langle g_j(it')\rangle+\langle g_j(\beta)\rangle+\langle g_j^*(it')\rangle-3]. \quad (24)$$

As was noted in I, the Gaussian factor in (24) limits the significant values of k to $k\lesssim\beta^{-1/2}$. As a consequence, matrix elements correct to second order in ka can be obtained using only the s-wave contribution to the wave function,

$$\psi_{\mathbf{k}}(\mathbf{r})=(2\pi)^{-3/2}\{\exp(i\mathbf{k}\cdot\mathbf{r})-\sin(ka)\times\exp[ik(r-a)]/kr\}. \quad (25)$$

We thus get

$$\begin{aligned}\langle g_j(it)\rangle&=C^{-1}\int d\mathbf{r}_e\int d\mathbf{r}_e'\,\langle\mathbf{k}\mid\mathbf{r}_e\rangle\\&\times\langle\mathbf{r}_e\mid\exp[-it(p^2/2+V_j)]\mid\mathbf{r}_e'\rangle\\&\times\langle\mathbf{r}_e'\mid\mathbf{k}\rangle\exp(itk^2/2)(\partial/\partial k_z)\exp(-\beta k^2/2)\\&=\exp(itk^2/2)\int d\mathbf{r}_e\int d\mathbf{r}_e'\int dK\,\psi_j^*(\mathbf{r}_{ej})\\&\times\exp(-itK^2/2)\psi_j(\mathbf{r}_{ej}')\exp[-i\mathbf{k}\cdot(\mathbf{r}_e-\mathbf{r}_e')]/\Omega\\&=1+\exp(itk^2/2)\int d\mathbf{r}\int d\mathbf{r}'\\&\times\exp[-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')]\{-a(r+r')+a^2[1+i(r+r')^2/t]\\&\times\exp[i(r+r')^2/2t]\}/[rr'\Omega(2\pi it)^{3/2}]\\&=1-2\pi iat/\Omega+2(2\pi)^{1/2}a^2\{[(itk^2-1)/k]\\&\times M(k(it)^{1/2})-it\exp(itk^2/2)\}/\Omega, \quad (26)\end{aligned}$$

where

$$\langle\mathbf{k}\mid\mathbf{r}\rangle=(2\pi)^{-3/2}\exp(i\mathbf{k}\cdot\mathbf{r}), \quad (27)$$

$$\mathbf{r}=\mathbf{r}_{ej},\qquad\mathbf{r}'=\mathbf{r}_{ej}', \quad (28)$$

and

$$M(x)=\int_0^x\exp(y^2/2)\,dy. \quad (29)$$

The integrations over $\mathbf{r}$ and $\mathbf{r}'$ are performed by first integrating over the solid angles and then making the change of variables, $s=r+r'$ and $q=r-r'$. Similarly,

$$\begin{aligned}\langle g_j(\beta)\rangle&=iC^{-1}(\partial/\partial k_z)\Big\{\int d\mathbf{r}_e\int d\mathbf{r}_e'\int dK\,\psi_j^*(\mathbf{r}_{ej})\\&\times\exp(-\beta K^2/2)\psi_j(\mathbf{r}_{ej}')\exp[-i\mathbf{k}\cdot(\mathbf{r}_e-\mathbf{r}_e')]\Big\}(2\pi)^3\\&=1-2\pi\beta a/\Omega-a^2\{[(1+2\beta k^2-\beta^2k^4)/\beta k^3]M(k\beta)\\&-[(1-\beta k^2)/\beta^{1/2}k^2)]\exp(\beta k^2/2)\}/[2(2\pi)^{3/2}\Omega], \quad (30)\end{aligned}$$

and

$$\langle g_j^*(it)\rangle=\langle g_j(it)\rangle^*. \quad (31)$$

Note that, as a result of integrating $\mathbf{r}_e$ and $\mathbf{r}_e'$ over all space, the coordinates of the scatterers no longer appear in the right-hand sides of Eqs. (26) and (30). The trace of $\exp(-\beta H_{\{N\}})$ was evaluated in I and was shown to be given by

$$\mathrm{tr}\{\exp(-\beta H_{\{N\}})\}=\Omega(2\pi\beta)^{-3/2}\exp(-2\pi\beta\rho a), \quad (32)$$

where $\rho=N/\Omega$. Substituting the above matrix elements into Eq. (20) and using Eqs. (24) and (32), we find the following expression for the mobility:

$$\mu=-e\beta(2\pi)^{-3/2}\int d\kappa\,\kappa_z\mathcal{I}(\kappa,\alpha,\mathrm{T})F(\kappa,\alpha)\times(\partial/\partial\kappa_z)\exp(-\kappa^2/2), \quad (33)$$

where

$$\mathcal{I}(\kappa,\alpha,\mathrm{T})=\int_0^{\mathrm{T}}I(\kappa,\alpha,\mathrm{T}')\,d\mathrm{T}', \quad (34)$$

$$\begin{aligned}I(\kappa,\alpha,\mathrm{T})&=\exp\{-4\pi^{1/2}\alpha[(\kappa^2\mathrm{T}+1)\mathcal{C}(\kappa\mathrm{T}^{1/2})\\&+(\kappa^2\mathrm{T}-1)\mathcal{S}(\kappa\mathrm{T}^{1/2})+\kappa\mathrm{T}^{1/2}(\cos(\kappa^2\mathrm{T}/2)\\&-\sin(\kappa^2\mathrm{T}/2))]/\kappa\}, \quad (35)\end{aligned}$$

$$\mathcal{S}(x)=\int_0^x\sin(y^2/2)\,dy, \quad (36)$$

$$\mathcal{C}(x)=\int_0^x\cos(y^2/2)\,dy, \quad (37)$$

$$\begin{aligned}F(\kappa,\alpha)&=\exp\{-\alpha M(\kappa)(1+2\kappa^2-\kappa^4)/\kappa^2\\&-\exp(\kappa^2/2)[(1-\kappa^2)/\kappa]/[2(2\pi)^{3/2}\kappa]\}. \quad (38)\end{aligned}$$

The following dimensionless parameters have been introduced:

$$\alpha\equiv\rho a^2\beta^{1/2}, \quad (39)$$

$$\mathrm{T}\equiv t/\beta, \quad (40)$$

$$\kappa\equiv\beta^{1/2}k. \quad (41)$$

Angular integration of Eq. (33) gives

$$\mu=[2e\beta/3(2\pi)^{1/2}]\int_0^\infty d\kappa\,\kappa^4\exp(-\kappa^2/2)\times\mathcal{I}(\kappa,\alpha,\mathrm{T})F(\kappa,\alpha). \quad (42)$$

V. RELAXATION TIME

In obtaining Eq. (42), no assumption was made concerning the existence of a relaxation time. Expansion of the Fresnel integrals[13] shows that for $\kappa^2 T \gg 1$,

$$I(\kappa, \alpha, T) \cong \exp(-T/\tau), \tag{43}$$

where

$$\tau = (4\pi\alpha\kappa)^{-1}. \tag{44}$$

The values of κ which contribute significantly to μ are restricted to $\kappa \sim 1$ by the factor $\kappa^4 \exp[-(\kappa^2/2)]$. For these κ's and experimentally obtainable values of the parameters, the condition that the time an electron spends under the influence of E be large ($\kappa^2 T \gg 1$) is equivalent to the condition that this time be large compared with the relaxation time. Thus, Eq. (34) can be written as

$$\begin{aligned} \mathcal{I}(\kappa, \alpha, T) &= \int_0^T I(\kappa, \alpha, T')\, dT' \\ &\cong \int_0^{T'' \gg \kappa^{-2}} I(\kappa, \alpha, T')\, dT' \\ &\quad + \int_{T''}^{T} \exp(-T'/\tau)\, dT' \\ &\cong \int_0^{T''} I(\kappa, \alpha, T')\, dT' + \int_{T''}^{\infty} \exp(-T'/\tau)\, dT'. \end{aligned} \tag{45}$$

τ can therefore be identified as a relaxation time in the sense that after a sufficient lapse of time, the approach to a steady-state condition is always given by an exponential decay, namely $\exp(-T/\tau)$.

Expression (42) is equivalent to the Langevin mobility formula provided $4\pi\alpha \ll 1$. With this restriction on α (low density or high temperature), we can approximate $\mathcal{I}$ and F by

$$\begin{aligned} \mathcal{I}(\kappa, \alpha, T = \infty) &= \int_0^{\infty} I(\kappa, \alpha, T)\, dT = \int_0^{\infty} \exp(-T/\tau)\, dT = \tau \end{aligned} \tag{46}$$

and

$$F(\kappa, \alpha) \cong 1. \tag{47}$$

Thus, μ reduces to

$$\begin{aligned} \mu &\simeq e\beta/[3(2\pi)^{1/2}\alpha] \int_0^{\infty} \kappa^3 \exp(-\kappa^2/2)\, d\kappa \\ &= 2e\beta/[3(2\pi)^{3/2} m^{1/2} \alpha]. \end{aligned} \tag{48}$$

In general, μ was found numerically as a function of ρ and T. The result is shown in Fig. 1 for $T = 3.96$°K and $a = 0.62$ Å (Ref. 14) along with the Langevin-theory value of μ. (The circles are the experimental results of Levine and Sanders[15] and show the very sharp drop in mobility which occurs at the onset of bubble formation.) The Langevin theory is seen to be accurate to within 5% for $\alpha < 4 \times 10^{-2}$, 10% for $\alpha < 5 \times 10^{-2}$, and 50% for $\alpha < 2 \times 10^{-1}$.

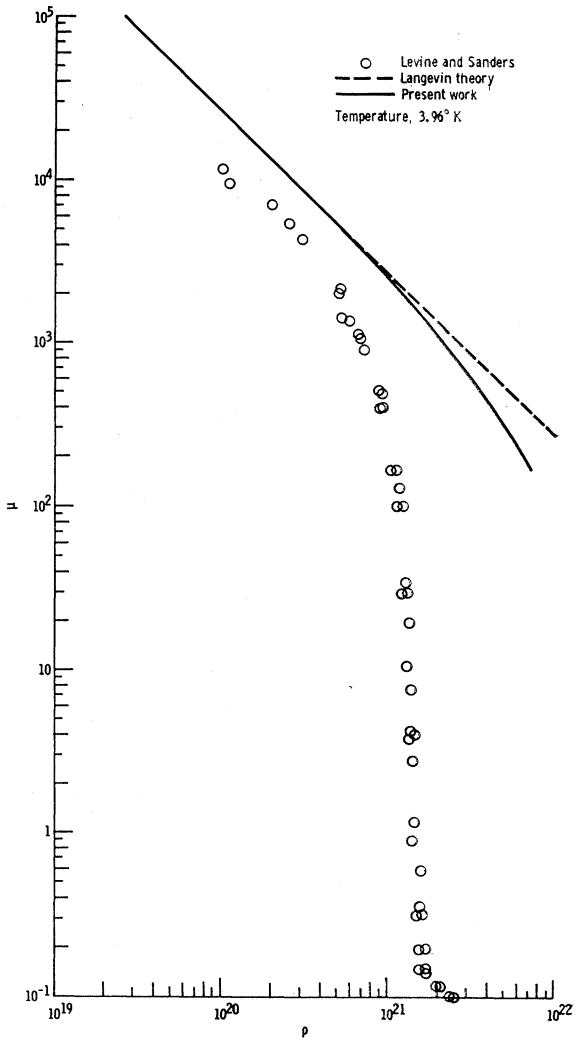

FIG. 1. Mobility μ (cm²/Vsec) versus number density ρ (atoms/cm³) at constant temperature for $a = 0.62$ Å.

Assuming an ideal-gas equation of state for the helium and an electron trapped in a 3-dimensional well of depth $2\pi\rho a\hbar/m$, the condition that there be no bubble is approximately $\alpha^3(\beta^{1/2}/a) \ll 10^{-2}$. In order to simultaneously have $\alpha \geq 1$ and $\alpha^3(\beta^{1/2}/a) \ll 10^{-2}$, one must go to small β (high temperature). However, this decreases α unless the fractional pressure increase equals the fractional temperature increase raised to the $\frac{3}{2}$ power. Thus, it is not feasible to construct an experi-

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ment in which the deviation from the Langevin theory calculated above could be seen.

VI. CONCLUSION

We have demonstrated that the Langevin theory for electron mobility in a hard-core gas can be derived from irreversible quantum statistics without postulating any relaxation time. Rather, a momentum-dependent relaxation time emerges as a *direct consequence* of the expansion procedure employed. This treatment also yields criteria for the limits of validity of the classical Langevin theory that help explain why it has been so successful.

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Radiation of Sound in He-II Films*

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Static He-II films which are locally disturbed are investigated with the two-fluid model. It is assumed that motion of the normal fluid is retarded by a viscous force per unit volume $r\omega\rho v_n$, in which r is a dimensionless viscosity parameter whose value in He-II films is shown to be $10^2 \leq r \leq 10^4$. A general surface wave and thermal disturbance is propagated through the film by two wave modes: a third-sound wave and a new highly attenuated, wave; these are first redescribed. For arbitrary initial conditions, the amounts of each mode produced are derived for a general experimental case. In particular, conditions which will produce third sound alone and the new mode alone are derived as functions of temperature and r. It is shown that pure third sound is difficult to produce, since the requirements on phase between the surface wave and temperature excitations are not experimentally natural ones. It is shown that, when the film is locally disturbed with a pure thermal excitation varying as $\exp i\omega t$, both modes are excited equally and are 180° out of phase, so that the net surface is undisturbed. When the film is locally disturbed with an isothermal pure pressure excitation, then for large viscous force on the normal fluid, almost all of the displacement is in the new mode. The motions of the film variables characteristic of each of the modes are analytically obtained in general, and described quantitatively for a particular case. In the new mode, the normal fluid and superfluid move with almost the same phases and velocities, and are 180° out of phase with the surface wave. In the third-sound mode, the superfluid moves in phase with the surface waves but the normal-fluid motion is more complicated. The results are compared with Lifshitz's results on first and second sound in bulk helium.

INTRODUCTION

THERE are two simple ways in which a static He-II film may be disturbed locally. The surface configuration itself may be changed, say by an indentation, and the temperature may be changed. This paper treats the question: What is the behavior of the film after arbitrary disturbances of these kinds? In general the answer is: Two, mixed thermal and pressure, wave modes propagate away from the source. The net velocity and attenuation of propagation in the film depend on how much of each of these two modes is excited. The principal purpose of this paper is to describe in detail the quantitative relations between initial conditions and the amplitudes and phases of the propagated waves. We shall also briefly redescribe both modes in terms of the two-fluid model of liquid helium.[1] Then the results will be applied to some interesting examples.

Actually if the normal fluid is totally immobile, as is often assumed, then only one kind of wave (third sound) is allowed in the film.[2] To explain recent observations of energy dissipation in He-II films it has been suggested that normal-fluid motion is merely retarded by large viscous forces in the film.[3] This extra degree of freedom brings with it into the problem some normal-fluid motion, energy dissipation, and a new overdamped wave mode.[4]

We intend then to complement Lifshitz's classic study on radiation of sound in He-II.[5] In Lifshitz's paper, which treats bulk helium, waves excited by thermal and pressure variations are considered, and the qualitative relations between initial conditions and propagated waves are found in a very elegant way and applied to some interesting examples. For that case, as is well known, the two modes are first and second sound. For our case the two modes are third sound and a new mode, not yet directly observed. Also, unfortunately,

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