PHYSICAL REVIEW A VOLUME 10, NUMBER 4 OCTOBER 1974

Lifetime of surface-state electrons on liquid ^{4}He. II. Electron lattice

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(Received 21 March 1974)

Electrons trapped by their image potential on the surface of liquid ^{4}He can escape into the gas by thermal ionization. I have calculated the rate of ionization, assuming that the electron motion parallel to the liquid surface is described by the collective motion of an electron crystal. The first electrons to leave the surface extract their ionization energy from the kinetic energy of those that remain behind. This adiabatic cooling effect rapidly lowers the electron temperature to a small fraction of the liquid temperature. The ionization rate is substantially less for this electron-crystal model than for the free-electron model of the electron motion parallel to the surface. The rate also depends on the density of surface electrons.

I. INTRODUCTION

When an electron is near the surface of liquid helium, the short-range repulsive force due to the exclusion principle opposes the long-range attractive force due to electrostatic polarization; hence there is a potential minimum for the electron just outside the liquid. The electron motion is nearly hydrogenic when perpendicular to the liquid surface and nearly free when parallel to the surface. This model[1] of the two-dimensional surface state has stimulated much theoretical and experimental interest, which is the subject of a recent review by Cole.[2]

One of the early measurements on this system was that of the lifetime in the surface state.[3] This lifetime was about 10^{-4} sec in the region 1.1–3.0 K. This seems long since the binding energy is only 0.0007 eV.[1] Subsequently, Ostermeier and Schwartz[4] made a lifetime measurement and concluded that the lifetime was shorter than 10^{-5} sec. However, as has been pointed out by Cole,[2] in this measurement[4] the electrons were probably free in the gas instead of bound to the surface by the image potential. Therefore, they could not have been measuring the surface-state lifetime. In an attempt to explain this long lifetime, I calculated the thermal escape rate (reciprocal of the lifetime) from the surface state.[5] The theoretical lifetime was, however, about an order of magnitude shorter than the experimental value. Even though the model calculation predicted too large an escape rate, it did indicate why the electron lifetime is long. As Ref. 5 (henceforth referred to as Paper I) shows, the binding energy is of secondary importance in determining the ionization rate because neither the liquid nor the gas atoms in the vapor can transfer sufficient energy in a single scattering event to the electron to ionize it. Therefore, their role is to mix the electron motions parallel and perpendicular to the liquid surface and cause the transfer of energy from the parallel to the perpendicular motion. Only if there is sufficient energy in the parallel motion can the electron be ionized. Thus, the ability of the parallel motion to replenish its energy from the liquid and gas determines the ionization rate. The electron system is so weakly coupled to the liquid and gas that the ionization process, which removes the binding energy E_0 from the electron system, causes it to cool well below the liquid.

To improve on this calculation I shall use a different model of the electron motion parallel to the liquid surface. The assumption that electron motion is free-electron-like parallel to the liquid surface is strictly valid only for a single electron. If many electrons are on the surface, electron-electron interactions can cause the electrons to crystallize to lower their energy.[6-8] In this case, the motion is best described by the collective modes of a two-dimensional electron crystal.[7] Since the liquid and the gas atoms couple mainly to the electron motion parallel to the liquid surface, the escape rate depends on this motion. As the following calculation will show, electron crystallization reduces the escape rate. For this electron-crystal model, as well as for the free-electron model of the motion parallel to the liquid surface as discussed in Paper I, the surface state is ionized when energy is transferred from the motion parallel to the liquid surface to the motion perpendicular it. In the former case it is the lattice energy stored in the phonon modes that ionizes the surface state. The liquid and the gas atoms act as a catalyst for the ionization since they transfer little energy to the electrons. At high temperatures surface-state ionization removes sufficient energy from the electron lattice to reduce its temperature to a small fraction of the liquid temperature. Only below about 1 K is the ionization rate sufficiently small to allow the lattice to remain in thermal equilibrium. Section II outlines the surface-state model. In Sec. III, the ionization rate and the rate of energy exchange between the electron system and the liquid and gas are calculated and the energy balance is used to determine the electron temperature. In Sec. IV, I discuss the results in terms of the experiments.

II. SURFACE-STATE MODEL

A brief discussion of the surface-state model follows. More detailed discussions can be found in the original papers.[1,2,8] At the liquid surface the electron experiences a repulsive barrier of about 1 eV. Outside the liquid the electron is attracted to the liquid by its image charge. The electron, which is weakly bound, is far enough away from the liquid so that the surface can be assumed planar. If z is the distance above the liquid surface, the potential energy of a single electron is

$$\begin{aligned} V_i(z) &= -\alpha e^2/z, \quad z>0 \\ &= +V_b, \quad z \leq 0. \end{aligned} \tag{1}$$

The potential V_b is the liquid barrier. The strength of the image charge is

$$\alpha = (\epsilon - 1)/4(\epsilon + 1), \tag{2}$$

where ϵ is the static dielectric constant. For ^{4}He, $\alpha = 0.006$. The solid curve in Fig. 1(a) represents the potential energy $V_i(z)$. In the original surface-state model, $V_i(z)$ is the only potential depending on z. Any potential depending on the coordinates x and y parallel to the liquid surface serves only to give the electron an effective mass. However, if electron-electron interactions are included, then the above assumptions are no longer valid.

When many electrons are present on the liquid surface, the potential energy becomes[8]

$$V(x,y,z) = V_i(z) + \frac{1}{2}\sum_{i\neq j}\frac{e^2}{|\vec{r}_i - \vec{r}_j|} + V_p. \tag{3}$$

The first term is the image potential, which remains unchanged. The second term is the Coulomb potential between the electrons, where $\vec{r}_i$ is the coordinate of the ith electron. Since the system is neutral, the electron charge is balanced by an equal number of positive charges on the electrodes above and below the liquid surface. This is shown in Fig. 1(b). The potential energy of interaction between the electrons and these positive charges is represented by V_p.

The combination of the image potential and the electric field due to the plates A and B localizes the electrons near the surface; i.e., $\langle z\rangle \approx 110$ Å. Since the electron densities considered in experiments are less than 10^9 cm^{-2}, the mean electron spacing a_0 is much greater than $\langle z\rangle$. Hence, the characteristic frequencies for the motion perpendicular to the surface are much higher than those for the motion parallel to the surface. Thus the z motion can be ignored in calculating the x,y motion. Neglecting end effects makes V_p independent of x and y. Therefore, as long as the electron system remains within 10^3 Å of the surface, $V(x,y,z)$ separates, in lowest order, into a term depending on x and y and one depending on z. Thus,[8]

$$V(x,y,z) \approx V_\perp(z) + V_\parallel(x,y). \tag{4}$$

Because the electron system is dilute, the energy of Coulomb repulsion between the electrons exceeds the kinetic energy of localization. According to Wigner,[6] this Coulomb energy is minimized if the electrons are ordered. Similarly, we expect the surface electrons to form a two-dimensional electron crystal. In the potential $V_\parallel(x,y)$, the dynamics of an electron can be described[8] by phonons. The frequency spectrum of these states is shown in Fig. 2 for a square lattice of lattice constant R_0. The transverse branch has soundlike behavior at long wavelength. The dispersion curve for the longitudinal branch, however, does not exhibit soundlike behavicr at long wavelength;

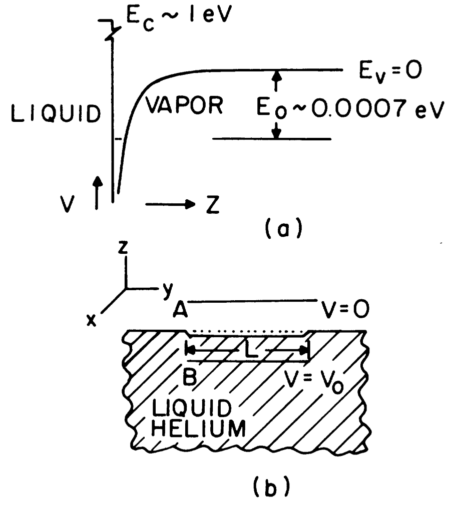

FIG. 1. (a) Sketch of the potential energy of an electron outside of liquid helium. Potential energy is plotted vertically and the z coordinate horizontally. E_0 is the binding energy. E_v is the vacuum level of energy. E_c is the conduction band in the liquid. (b) Schematic of the electron system on liquid helium. The metal plate B is immersed in the helium, and plate A, held at ground potential, is above the helium surface. The electrons are shown by dots on the helium surface. The length L, a rough measure of the size of the electron system, is given by the plate size. The z direction is perpendicular to the liquid surface.

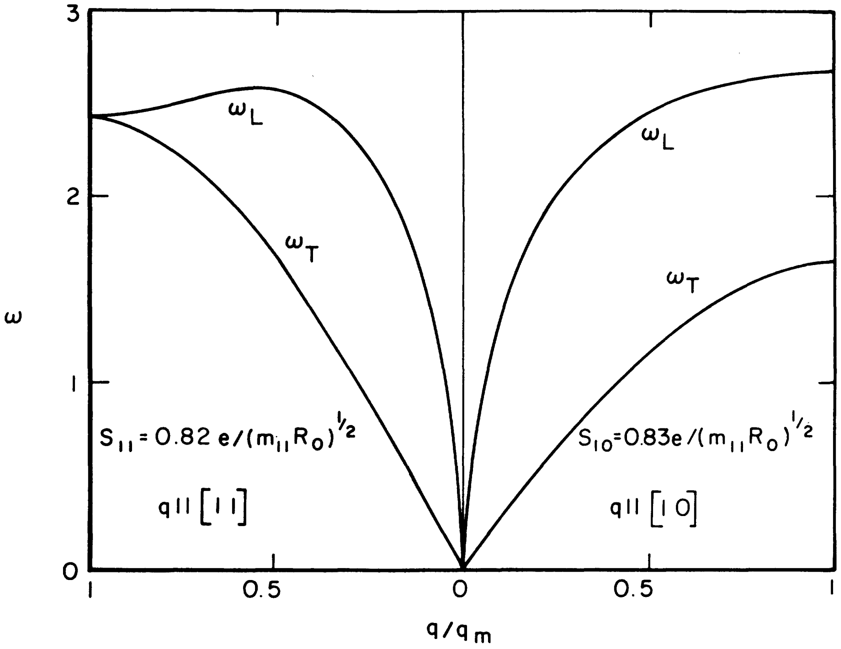

FIG. 2. Vibrational frequency $\omega(q)$ in units of $e/(mR_0^3)^{1/2}$ is plotted vs normalized wave vector $\vec{q}$ for the two principle directions of a square lattice. The maximum wave vector q_m is the value of $\vec{q}$ at the zone boundary. The transverse and longitudinal branches are labeled ω_T and ω_L, respectively. S is the sound velocity.

here the dispersion relation becomes the same as for the two-dimensional free-electron gas[9]; i.e., $\omega \propto q^{1/2}$, where q is the phonon wave vector.

For motion normal to the surface, the potential of an electron is

$$V_\perp = V_i(z) - z^2 \sum_{i \neq j} \frac{e^2}{4|\vec{R}_i - \vec{R}_j|^3} + V_p(z), \tag{5}$$

where the second term represents the electron-electron interaction; $\vec{R}_i$ is the coordinate of the ith electron-lattice point. This term is much less than $V_i(z)$ except when $z \gtrsim R_0$. This term causes tunneling of the electron out of its ground state. Because the tunneling distance is greater than 10^4 Å, this process is unimportant compared with thermal ionization. Henceforth, I shall omit this term.

The remaining term in Eq. (5), $V_p(z)$, arises from the positive charges on plate B in Fig. 1(b). This depends on the potential difference between plates A and B. For example, if we imagine that plate B is positive and plate A is at zero potential and, furthermore, that the maximum possible number[10] of electrons are on the helium surface, then $V_p(z)$ is zero for $z \gg R_0$. For a uniform electron density, $V_p(z) = 0$ for $z > 0$; it is $+eFz$ inside the liquid. On the other hand, for electrons in an ordered configuration, the field due to plates A and B cannot be completely screened at distances closer to the surface than R_0. For point charges, $V_p(z) \cong eFz$ for $z < R_0$. This situation for point charges is shown in Fig. 3(a). The solid curve is $V_\perp(z)$. The actual situation is different from this because the electrons are not point charges; they are spread about their lattice points and partially screen the applied field. If it were not for this electric field F holding the electrons on the surface, they would be free in the gas phase at temperatures above about 0.6 K.[2,3] The electric field increases the statistical weight of the bound states ensuring that the electrons are in the ground hydrogenic state. It was just this absence of a sufficient electric field F that affected the measurements of Ostermeier and Schwartz.[4] In what follows, I shall assume that the electrons are in this ground state.

In calculating the lifetime, plates A and B are assumed to be at the same potential. This approximates the experimental configuration of Ref. 3. The resulting potential is depicted in Fig. 3(b). At distances $>R_0$, the field due to the surface charge is $F_s = 2\pi e n_e$; at distances less than R_0, the field drops rapidly to zero. Therefore, $V_\perp(z)$ is nearly equal to the image potential for $z < R_0$. Since we require $V_\perp(z)$ only at distances closer than R_0, we shall set $V_\perp(z) = V_i(z)$. Actually, the main purpose of the last two terms in Eq. (5) is to ensure that an electron excited over the image potential barrier will diffuse into the gas region before it can be captured by the surface state.

Because $V(x, y, z)$ separates into components depending separately on the coordinates perpendicular and parallel to the liquid surface, the energy eigenvalues can be written as $E = E_\perp + E_\parallel$, where

$$E_\perp = -\alpha^2 m e^4/2\hbar^2 l^2 = -\hbar^2/2ml^2 z_c^2$$
$$= -E_0/l^2. \tag{6}$$

The free-electron mass is m, and z_c is the Bohr radius of this one-dimensional hydrogen atom. Since we let $V_b \to \infty$, l is an integer.[1] Because the electron motion parallel to the surface is described by an electron lattice, the values of $E_\parallel$ are quantized; i.e.,

$$E_\parallel = \hbar\omega, \tag{7}$$

where ω is the phonon frequency. To obtain an idea of the magnitude of these energies, we note that $E_0 \approx 7\times10^{-4}$ eV and that the highest phonon energy, for $R_0 = 10^4$ Å, is about 2.5×10^{-5} eV. Thus $E_\parallel \ll E_\perp$.

The wave functions for the motion perpendicular to the surface are identical to those of the s states of the hydrogen atom multiplied by z/z_c. These functions are given in Paper I. The lattice is treated by the usual second-quantization procedure.

III. THERMAL IONIZATION OF THE SURFACE STATE

The above model permits one to calculate the lifetime against thermal escape from the surface. The escape rate W_e is the inverse of this lifetime. To estimate W_e we calculate the rate for electronic transitions from the lowest-energy hydrogenic state, $l=1$, to a hydrogenic continuum state labeled by the wave vector k_z. This excitation rate is a good estimate of the escape rate if the electron diffuses to the right-hand side of the potential maximum in Fig. 3(b) before it makes a transition back to the ground state. This is usually the case because the recombination rate is small.

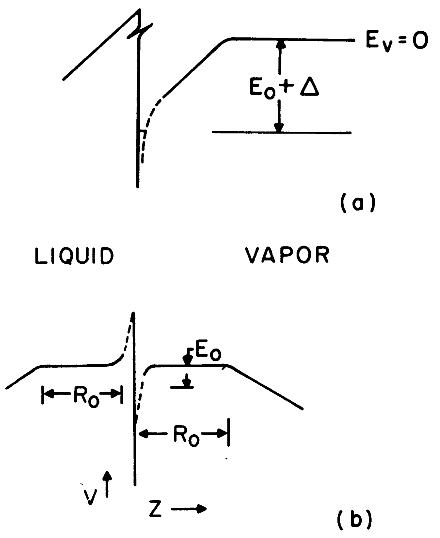

FIG. 3. Sketch of the potential energy of a single electron in the presence of many electrons at the surface of liquid helium. The dashed curves are due to the image potential. In (a) the metal plate in the liquid is positive. R_0 is the lattice constant of the electron lattice. E_v is the vacuum level of energy. The binding energy of the electron is increased to roughly $E_0 + \Delta$ because of the incomplete shielding of the electric field between the metal plates above and below the liquid. When the electrons are spread uniformly over the surface, $\Delta = 0$. In (b) both plates are at the same potential, raising the electron potential above that of the vacuum. Here the potential energy in the liquid is shown omitting the shift due to V_b.

Because little of the electron wave function lies inside the liquid,[1] the interaction with bulk liquid excitations is weak. The only remaining perturbations are collisions with the helium atoms in the vapor above the liquid or with liquid surface waves ("ripplons"). However, as shown in Paper I, the energy transfer in a collision with a gas atom or ripplon is a small fraction of the binding energy.

How, then, does the electron gain sufficient thermal energy from the liquid or gas? Since the parallel and perpendicular motions of the electron are uncoupled, it is possible for the total energy to be positive with the electron still bound to the surface, i.e., $\langle E_\parallel \rangle \geq E_0$. The surface state can then be ionized if the lattice energy is transferred to the perpendicular motion by gas atom or ripplon scattering. Because the energy transfer from these perturbations is small, their chief role is to serve as a catalyst and conserve momentum. This process will limit the rate of escape from the surface as long as the electron system remains in thermal equilibrium with the liquid and gas. This means that the liquid and gas must supply energy to the electron lattice rapidly enough to keep it in thermal equilibrium. If not, the energy exchange between the electrons and their surroundings will limit the ionization rate. In Paper I, I found that maintenance of thermal equilibrium was the rate-limiting step above 1 K (the electron motion parallel to the surface is free). Similarly, I shall show that it also limits the rate when the electrons are ordered.

First, I shall calculate the rate at which an electron is excited from the ground state of the perpendicular motion to a continuum state with the simultaneous absorption of several lattice phonons to conserve energy. Second, I shall calculate the rate of energy exchange between the electron lattice and the liquid or gas. Finally, I shall compare this energy-exchange rate with the energy-loss rate due to the ionization process which removes energy E_0 from the electron system.

A. Transition rate

Since the transitions involve a large number of quanta, it is convenient to use the time-dependent perturbation theory developed by Glauber[11] for neutron scattering. In this theory, the information about dynamical properties of the electron lattice is contained in the correlation function. This function is the thermal average of the expectation value of the product of the displacement components $u(\vec{R}_i, t)$ at different points separated by the distance $\vec{R}$ and time t. The correlation function is

$$C_T(\vec{R}, t) = \frac{\hbar}{2Nm} \sum_{\vec{q},p} \frac{1}{\omega} [\exp(-i\omega t + i\vec{q}\cdot\vec{R}) + 2n_\omega \cos(\vec{q}\cdot\vec{R} - \omega t)], \tag{8}$$

where N is the number of electrons, n_ω the phonon occupation density, and $p = 1, 2$ is the unit polarization vector. $C_T(\vec{R}, t)$ is diagonal because the lattice is assumed to be cylindrically symmetric about the z axis. The matrix elements for the perturbations were evaluated in Paper I; the interaction with a gas atom is assumed to be a contact interaction; i.e.,

$$V_G = v_0 \delta(\vec{r} - \vec{r}_G), \quad v_0 = ha_s/m, \tag{9}$$

where a_s is the gas-atom s-wave scattering length and $\vec{r}_G$ is the gas-atom coordinate. Initially, the electron is in its ground hydrogenic state and the gas atom is in a plane-wave state described by the wave vector $\vec{P} = \vec{P}_\parallel + \hat{z}P_z$. After the collision the electron is in a continuum state k_z. The gas atom changes to another plane-wave state described by $\vec{P}'$. Following Glauber, the time-dependent matrix element for the transition involving the gas atom and an electron is

$$M(t) = |V_G| \sum_j \exp(-i\{\Delta\vec{P}\cdot[\vec{R}_j + \vec{u}(\vec{R}_j, t)] - \Omega t\}), \tag{10}$$

where $\Delta\vec{P} = (\vec{P}_\parallel' - \vec{P}_\parallel)$. The difference between the initial and final energies of the gas atom and perpendicular motion of the electron is $\hbar\Omega$; $|V_G|$ is the absolute value of the matrix element for the perturbation as given by Eq. (9) in Paper I. The expression $M(t)$ is an operator capable of inducing all possible vibrational transitions of the electron lattice. Using this operator and applying the usual procedure,[11] the transition probability for an n-phonon process in the Born approximation becomes

$$w_n = \frac{|V_G|^2}{n!\hbar^2} \sum_j \exp(i\Delta\vec{P}\cdot\vec{R}_j) \exp[-\Delta P^2 C_T(0,0)] \times \int_{-\infty}^{\infty} e^{-i\Omega t} [\Delta P^2 C_T(\vec{R}_j, t)]^n \, dt. \tag{11}$$

The term $\exp[-\Delta P^2 C_T(0,0)]$ is the square of the familiar Debye-Waller factor. The transition rate W_e is given by the sum of w_n over all n and all final states. I find that, in the sum over $\vec{R}_j$, only the point $\vec{R}_j = 0$ contributes appreciably to w_n. In other words, the incoherent scattering determines the transition rate. The coherent effects represented by terms in the sum for $\vec{R}_j \neq 0$ are unimportant. Therefore, momentum conservation will not restrict the available phase space. Before reducing w_n to a manageable form, we note that the same expression holds for ripplon scattering with $|V_G|$ replaced by $|V_R|$ [Eq. (12) in Paper I]. Since both the energy change of the gas atom and the energy of a single ripplon are insignificant compared with the energy change of the electron, $\hbar\Omega$ is the same for both, i.e.,

$$\hbar\Omega \simeq E_0[1 + (k_z z_c)^2]. \tag{12}$$

After substituting Eq. (8) for $C_T(0, t)$ into Eq. (11) for w_n, interchanging the order of summation and integration, the time integration gives

$$w_n = (2\pi|V_G|^2/n!\hbar^2) \exp[-\Delta P^2 C_T(0,0)] (\hbar\Delta P^2/2Nm)^n \times \sum_{s=0}^{n} \frac{n!}{s!(n-s)!} \sum_{\vec{q}} \cdots \sum_{\vec{q}_n} \prod_{i=1}^{n-s} \frac{1+n_{\omega_i}}{\omega_i} \times \prod_{j=1}^{s} \frac{n_{\omega_j}}{\omega_j} \delta\left(\Omega - \sum_{i=1}^{n-s} \omega_i + \sum_{i=1}^{s} \omega_i\right). \tag{13}$$

This expression represents all processes consistent with the energy conservation. In s of the processes, a phonon of energy $\hbar\omega$ is absorbed by the electron, and in $n - s$ of these a phonon is emitted. For an Einstein lattice (all lattice frequencies are the same; i.e., $\omega = \omega_e$) the evaluation of Eq. (13) is trivial. The allowed transitions are those for which $\Omega = (2s - n)\omega_e$. However, an Einstein lattice is not a good approximation to the electron crystal owing to the importance of the low-energy modes. For a general phonon dispersion, the expression for w_n represents a complex summation beyond the scope of this paper. However, by applying the central limit theorem to Eq. (13), w_n can be readily evaluated. Sparks and Sham[12] have approximated similar sums involved in optical absorption by means of the central limit theorem. Using their approach, I define

$$\sigma_a(\xi) = \alpha_a^{-1} \sum_{\vec{q}} \frac{n_\omega}{\omega} \delta(\xi - \omega), \qquad \sigma_e(\xi) = \alpha_e^{-1} \sum_{\vec{q}} \frac{1+n_\omega}{\omega} \delta(\xi + \omega), \tag{14}$$

where the normalization constants are

$$\alpha_a = \sum_{\vec{q}} \frac{n_\omega}{\omega}, \quad \alpha_e = \sum_{\vec{q}} \frac{1+n_\omega}{\omega}. \tag{15}$$

Using these definitions, Eq. (13) reduces to

$$w_n = (2\pi |V_G|^2/\hbar^2 n!)\exp[-\Delta P^2 C_T(0,0)]\,(\hbar\Delta P^2/2Nm)^n$$
$$\times \sum_{s=0}^{n} \frac{n!}{(n-s)!\,s!}\,\alpha_e^s\,\alpha_a^{n-s} \prod_{i=1}^{n-s} \int d\xi_i\,\sigma_a(\xi_i)$$
$$\times \prod_{j=1}^{n} \int d\xi_j\,\sigma_e(\xi_j)\,\delta\Big(\Omega - \sum_{i=1}^{n-s}\xi_i + \sum_{j=1}^{s}\xi_j\Big). \qquad (16)$$

The product of integrals in the above expression replaces the product of sums in Eq. (13). According to the central-limit theorem, these convolution integrals lead to a Gaussian for large n. Therefore, w_n can be written as

$$w_n = [(2\pi)^{1/2}|V_G|^2/\hbar^2 n!]\exp[-\Delta P^2 C_T(0,0)]$$
$$\times (\hbar\Delta P^2/2Nm)^n \sum_{s=0}^{n} \frac{n!}{(n-s)!\,s!}\,\alpha_e^s\,\alpha_a^{n-s}\,\frac{1}{\sigma}$$
$$\times \exp[-(\Omega-\Omega_0)^2/2\sigma^2], \qquad (17)$$

where

$$\Omega_0 = (n-s)\int \xi\sigma_a\,d\xi + s\int \xi\sigma_e\,d\xi, \qquad (18)$$

$$\sigma^2 = (n-s)\Big[\int \xi^2\sigma_a\,d\xi - \Big(\int \xi\sigma_a\,d\xi\Big)^2\Big]$$
$$+ s\Big[\int \xi^2\sigma_e\,d\xi - \Big(\int \xi\sigma_e\,d\xi\Big)^2\Big]. \qquad (19)$$

Because of the central-limit theorem the multiple sum over $\vec{q}_1 \cdots \vec{q}_n$ has been replaced by a product of sums over single-phonon coordinates. This is a significant simplification of w_n when it is to be evaluated numerically. Since the transitions involve at least ten phonons, we expect the central-limit theorem to apply. For a lattice of infinite extent, the summations over $\vec{q}$ in Eq. (15) would diverge at finite temperature.[8] However, since the lattice is of finite extent, the lower limit of $\vec{q}$ is the inverse of the diameter of the lattice.[8]

The next step in the evaluation of the transition rate is to sum over the final states. For gas-atom scattering this sum is

$$\sum_f w_n = \frac{VL_G}{(2\pi)^4}\int d\theta\,\Delta P\,d\Delta P\,d\Delta P_z\,dk_z\,w_n, \qquad (20)$$

where $\Delta P_z = P_z' - P_z$. For ripplon scattering

$$\sum_f w_n = \frac{V}{(2\pi)^3}\int d\theta\,q_r\,dq_r\,dk_z\,w_n, \qquad (21)$$

where $\vec{q}_r$ is the ripplon wave vector, L_G is the length of the gas space above the liquid, and V is the system volume. For completeness we need the definitions of $|V_G|$ and $|V_R|$. They are given in Paper I by Eqs. (9) and (12), respectively. These are

$$|V_G|^2 = v_0^2(8\pi k_z z_c^2/V^2 L)[1+(k_z z_c + \Delta P_z z_c)^2]^{-3}, \qquad (22)$$

$$|V_R|^2 = 4\pi^2 k_z(\alpha_l n_l e^2)^2 kT(Vq_r^4 z_c^4\sigma_l)^{-1}, \qquad (23)$$

where α_l is the liquid polarizability, n_l the density of liquid atoms, and σ_l the surface tension. To obtain the matrix elements in the above form, I used the high-temperature approximation for the ripplon density and took the ripplon dispersion relation to be

$$\omega_r^2 \simeq \sigma_l q_r^3/d, \qquad (24)$$

which applies for ripplons with wavelengths less than about 0.1 cm. Here d is the liquid mass density and ω_r the ripplon frequency. Because the matrix elements are small for $k_z z_c \geq 1$, the limit $k_z z_c < 1$ is used for Eqs. (22) and (23). To account for this in the summation over final states I multiply w_n by $e^{-(k_z z_c)^2}$. I feel this is justified at this stage since a precise determination of the matrix elements for all $k_z z_c$ and subsequent summation over k_z would require an inordinate amount of numerical integration. This is not justified until the experimental situation is improved. Summing w_n over final states is straightforward. Thus

$$W_e = \sum_{n=0}^{\infty} w_n = \sum_{n=0}^{\infty} A \sum_{s=0}^{n} \frac{n!}{(n-s)!\,s!}\,\frac{\alpha_e^s\,\alpha_a^{n-s}}{(\alpha_e+\alpha_a)^n}$$
$$\times B\exp[-(1-\Omega_0/\Omega_1)^2 B^2]\,\mathrm{erfc}[(\Omega_1-\Omega_0)B(2\sigma^2)^{-1/2}], \qquad (25)$$

where erfc is the complimentary error function, $B = (1+2\sigma^2/\Omega_1^2)^{-1/2}$, $\Omega_1 = E_0/\hbar$, and A depends on the scattering mechanism. For ripplon scattering A is

$$A_R = 2P_R\langle u^2\rangle/n(n-1)z_c^2, \quad n>1; \qquad (26)$$

for gas-atom scattering,

$$A_G = P_G z_c^2/\langle u^2\rangle. \qquad (27)$$

The coefficients P_G and P_R are

$$P_G = 3mv_0^2 n_G/4\hbar^3 z_c, \qquad (28)$$

$$P_R = \pi(n_l\alpha_l e^2)^2 kT/4\hbar\sigma_l z_c^4 E_0, \qquad (29)$$

where n_G is the density of gas atoms.

The terms contained in the summation over s in Eq. (25) are independent of the scattering mechanism. Therefore, which scattering mechanism dominates in determining the lifetime depends on the parameter A. Above about 1 K, $P_G \gg P_R$.[5] For free electrons, as discussed in Paper I, this causes gas-atom scattering to determine the ionization rate above 1 K. Here, because the ratio $\langle u^2\rangle/z_c^2$ is much greater than unity for the electron lattice, ripplon scattering determines the ionization rate at much higher temperatures. The summation over s which represents a multitude of possible multiquantum transitions determines the temperature dependence and magnitude of W_e.

Because there is no restriction on the momentum transfer, as in the usual case of neutron scattering[11] or optical absorption,[12] processes in which phonons are both absorbed and emitted are equally probable. The summation over final states [Eqs. (20) and (21)] removes the $n!$ occurring in the denominator of Eq. (17). If $\Delta\vec{P}$ were fixed, as it usually is,[11,12] then the $n!$ would remain, ensuring the dominance of the lowest-order process, in which just enough phonons are absorbed to ionize the electron.

Before considering the mechanism of energy exchange between the electron system and the liquid and gas, I shall present the results of an evaluation of Eq. (25) for ripplon scattering. To numerically evaluate the sums over the phonon spectrum needed to determine Ω_0, σ, α_a, and α_e, I made an approximation to the phonon spectrum shown in Fig. 2. I assumed that the dispersion curve was cylindrically symmetric about z and used the dispersion curve shown for the [10] direction. This is a reasonable approximation because the ionization rate ought not to be sensitive to small changes in the shape of the dispersion curve as long as the long-wavelength behavior is retained and the highest lattice frequency ω_0 is unchanged. Figure 4 shows the ionization rate W_e as a function of temperature T for the lattice constants $R_0 = 10^4$ Å and 6300 Å. Even though it is not obvious from the exponential plot, $W_e \propto T^2$ in the high-temperature region, i.e., $T \gg T_0$ where $T_0 = \hbar\omega_0/k$. Since both $\langle u^2\rangle$ and P_R are both proportional to T in this region, the quadratic dependence on T implies that the summation over s, which represents the multiquantum nature of the transition, is independent of temperature. This occurs because the temperature dependence of the Debye-Waller factor just cancels the strong temperature dependence of $[C_T(0,t)]^n$ in Eq. (11). The Debye-Waller factor has a strong temperature dependence because of the large momentum transfer and the large magnitude of $C_T(0,0)$. The large momentum transfer also causes the coherent scattering to be much smaller than the incoherent scattering.

At low temperatures ($T < T_0$) the ionization rate decreases nearly exponentially with decreasing temperature. The slope of the exponential dependence is nearly equal to $-E_0/k$. However, the magnitude of the slope increases with decreasing temperature. This occurs because, as the temperature decreases, Ω_0, as defined by Eq. (18), decreases at fixed n and s. Therefore, a larger number of quanta are needed at lower temperature to maximize the Gaussian function in Eq. (17). Physically, more phonons are needed because the high-energy phonons are not excited. The larger n then leads to a stronger temperature dependence. One should note that W_e is less than the corresponding quantity for the free-electron case.[5]

B. Energy balance

When an electron escapes from the surface state, it removes, from the electron system, an energy E_0 which is greater than kT. If this energy is not replenished by the gas or liquid,[13] the electron system will be forced out of thermal equilibrium; i.e., it will cool. Since the energy-loss rate decreases with decreasing temperature, the electron system cools until the energy loss is balanced by the energy input. Many ripplon or gas-atom collisions are necessary to replace the energy E_0 removed by a single electron, because the energy exchange with a gas atom or ripplon is a small fraction of kT.

To find out how much the electron system cools, we assume that, after the initial cooling transient, there is a steady state with the energy loss just balanced by the energy input. In steady state the energy-balance equation is

$$\langle \dot{E}_l \rangle - \langle \dot{E}_i \rangle = 0 . \tag{30}$$

It balances the average of the energy-loss rate $\dot{E}_l$ against the average of the energy-input rate $\dot{E}_i$. The input is from gas atoms and ripplons and the loss is by ionization of the surface state. These averages are taken over the distribution

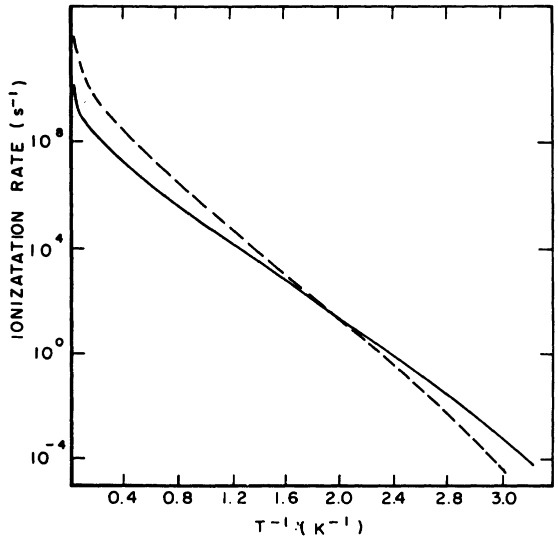

FIG. 4. Ionization rate plotted as a function of reciprocal temperature. The solid curve corresponds to a lattice constant R_0 of 10^4 Å; the dashed curve, to $R_0 = 6300$ Å. The liquid and electron temperature are the same.

function for the electron motion parallel to the liquid surface. Finding the exact distribution function requires the solution of the appropriate transport equation. Since this is beyond the scope of this paper, I shall make, as in Paper I, an *electron-temperature approximation* for the phonon distribution function; i.e., I assume the phonons are characterized by a Bose-Einstein distribution function at a temperature T_p.

The energy-input rate is defined by

$$E_t = \sum_f w_1 \hbar \Omega_t , \qquad (31)$$

where w_1 is the transition rate for a one-phonon process and $\hbar\Omega_t$ is the energy transferred between the electron lattice and gas atom or ripplon during the transition. Because we consider only the lowest-order process, the central-limit theorem is of no use. Fortunately, for $n=1$ the summation over $\vec{q}$ in Eq. (13) can readily be performed. For the $n=1$ transition, there are only two terms in the summation over s in Eq. (13). The term for $s=0$ represents absorption of energy by the lattice and the term $s=1$ represents energy loss.

For energy exchange, the matrix elements differ from Eqs. (22) and (23). Because the electron states quantized in the z direction are widely separated in energy, no energy is transferred between these levels. Therefore, the electron remains in its ground state along the z direction during the transition. The matrix elements for this type of transition were calculated in Paper I. For gas-atom scattering

$$|V'_G|^2 = |v_0/V|^2 [1 + \tfrac{1}{4}(\Delta P_z z_c)^2]^{-3}. \qquad (32)$$

For ripplon scattering,

$$|V'_R|^2 = \pi\hbar^2 P_R E_0 \left(\frac{\sigma_l}{d}\right)^{1/2} \frac{2kT/\hbar\omega_r + 1 + \delta N}{kTA_l q_r^{1/2}} . \qquad (33)$$

Here δN is +1 or −1 for ripplon emission and absorption, respectively, and A_l is the lattice area. The energy transfer in ripplon scattering is

$$\hbar\Omega_{tR} = \hbar\omega_r , \qquad (34)$$

while for gas-atom scattering it is

$$\hbar\Omega_{tG} = -\hbar^2[(\Delta P)^2 - 2\Delta P P_\parallel \cos\phi]/2M, \qquad (35)$$

where ϕ is the angle between $\vec{P}_\parallel$ and $\Delta\vec{P}$, and M is the mass of a gas atom. Since the hydrogenic state remains unchanged, there is no final-state summation over k_z. Upon performing the summations over $\vec{q}$ in Eq. (13), averaging over the initial states, and integrating over the final states, the energy input rate, correct to lowest order, is

$$\langle \dot{E}_{tG} \rangle = \left(1 - \frac{T_p}{T}\right) P_G \frac{3}{\pi 2^{9/2}} \left(\frac{m}{M}\right)^{1/2} \frac{(E_0^5 kT)^{1/2}}{\hbar^2 S_{10}^2 n_e^2} \left(\frac{z_c^2}{\langle u^2 \rangle}\right)^{5/2} \qquad (36)$$

for gas-atom scattering, and

$$\langle \dot{E}_{iR} \rangle = \left(1 - \frac{T_p}{T}\right) \frac{P_R(\frac{3}{4})!}{4\pi^2} \left(\frac{\sigma_l}{d}\right)^{1/2} \frac{E_0^2 z_c^2}{\hbar n_e S_{10}^2 (\langle u^2 \rangle)^{7/4}} \qquad (37)$$

for ripplon scattering. The ripplons and gas atoms are in thermal equilibrium at a temperature T. The lattice phonons are assumed to be in thermal equilibrium at T_p. These expressions show that for energy to be transferred to the electron lattice $T_p < T$. These expressions are both less than the corresponding expressions for the free-electron case [Eqs. (26) and (27) in Paper I]. In contrast to the ionization rate, the energy-input rate is determined by gas-atom scattering above about 1 K. Below 1 K the gas atoms are frozen out and ripplon scattering supplies energy to the electron lattice. This also occurs for the free-electron case.[5]

The energy-loss rate is given by the product of the ionization rate W_e and the energy lost per ionization event. Thus the energy balance becomes

$$W_e E_0 - \langle \dot{E}_{tG} \rangle - \langle \dot{E}_{iR} \rangle = 0. \qquad (38)$$

In this case the ionization rate is to be evaluated using a phonon distribution at the lattice temperature T_p, which is determined from the solution of Eq. (38).

IV. DISCUSSION

As in the free-electron case, the failure of the electron lattice to remain in thermal equilibrium with its surroundings is the main factor determining the electron lifetime above 1 K. This failure to achieve thermal equilibrium with the liquid arises because the energy transfer in a collision with a ripplon or gas atom is a small fraction of the electron binding energy. It is typically less than $10^{-3}E_0$. Because of this small energy exchange in a collision, the energy-exchanging collisions must be 10^3 times more frequent than the ionizing collisions if the lattice is to remain in thermal equilibrium with the liquid. As reference to Fig. 4 shows, the probability of ionizing collisions is large for temperatures above T_0. Thus, we might expect that the lattice would have to cool to a temperature near T_0 to obtain energy balance. This is substantiated by a numerical evaluation of Eq. (38) to find T_p as a function of T and hence the ionization rate.

In Fig. 5, the escape rate is plotted as a function of reciprocal temperature for a square lattice with $R_0 = 10^4$ Å and $A_l = 1$ cm^2. This lattice constant is roughly that corresponding to the electron density in the experiment.[3] The dashed curve ob-

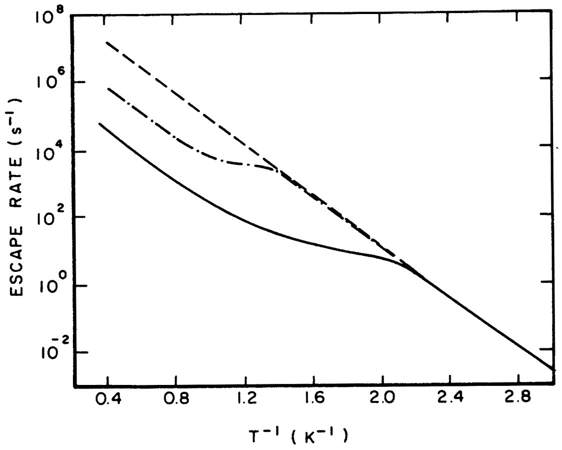

FIG. 5. Escape rate vs reciprocal temperature for a lattice with $R_0=10^4$ Å. The solid curve corresponds to an energy balance. The dashed curve is the $T_p=T$. The broken line corresponds to the energy-input rate being calculated with the free-electron model.

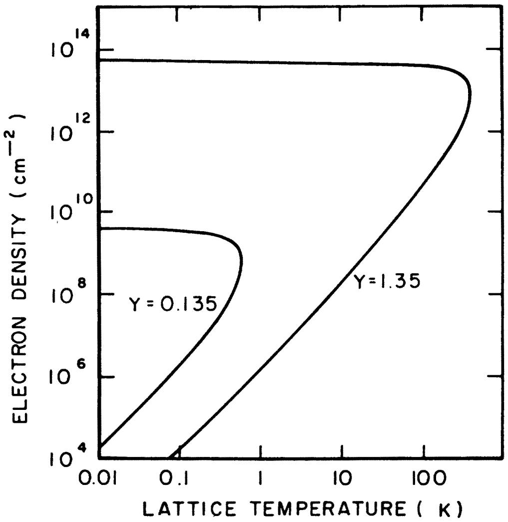

FIG. 6. Phase diagram of the electron crystal. The electron density is plotted vs the temperature. The solid region is inside the curves; the liquid region, outside. γ is the value of $\langle u^2\rangle/R_0^2$.

tains in the absence of an energy balance with $T_p=T$. This curve is also in Fig. 4. After demanding energy balance, I obtain the solid curve where T_p and T are the same only below 0.45 K. T_p decreases monotonically from a value of 0.9 K at $T=4.2$ K to 0.45 K at $T=0.45$ K. Thus, as I found for the free-electron case, there is significant electron cooling accompanied by a striking decrease in the escape rate. I made a preliminary estimate[14] of the escape rate using the Einstein approximation to the lattice to calculate W_e and the Debye approximation to calculate the energy-loss rate. The results are within a factor of 2 of the results presented here.

The values of W_e shown by the solid curve are significantly lower than the values calculated with the free-electron model (see Fig. 2 in Paper I). This reflects the weak coupling between the electron lattice and the liquid and gas. The results based on these two model calculations straddle the experimental values. For the potential configuration corresponding to Fig. 3(b) the measured[3] escape rate is about 10^4 sec between 1.2 and 3 K. This value is roughly midway between the values of W_e calculated with the free-electron model and with the electron-crystal model. One expects the lattice model to underestimate W_e because of the assumption of long-range order.

At this point, we must ask if long-range order exists in this two-dimensional lattice. The absence of long-range order would significantly affect the energy-input rate since it is determined by interactions with long-wavelength phonons. The ionization rate is less sensitive, however, because it is determined by scattering with short-wavelength phonons and we expect short-range order to hold.

To estimate the degree of crystalline order I use the usual melting criterion, which is to set $C_T(0,0)/R_0^2=\gamma$, where γ is a constant. Figure 6 is a plot of the melting line for two different values of γ. The line $\gamma=1.35$ corresponds to the melting criterion of Elgin and Goodstein as applied by Cole.[2] This estimate predicts the existence of the solid phase over a wide range of density and temperature. The line $\gamma=0.135$ is a more conservative estimate that presumably underestimates the solid region. Therefore, we expect the melting line to lie somewhere between the two.

To estimate the size of the effect of the loss of long-range order on the escape rate, I assume that the lattice contains only $\vec{q}$ vectors larger than $0.1q_m$ when calculating W_e. Even when the lattice has melted, the existing short-range order makes W_e much less than the corresponding quantity for the free-electron case.[5] For the energy-input rate we use the expressions for the free-electron case. This gives an upper limit to the energy-input rate. The dot-dash line in Fig. 5 represents the escape rate in this case. It lies higher than the solid curve because the energy-input rate is greater for free electrons than for the electron lattice. I expect the actual situation to be somewhere in between.

A lifetime measurement could be used to show the existence of the electron lattice. By varying the electron density on the surface, and, hence, the lattice constant, the ionization rate would change if the electrons were ordered.

ACKNOWLEDGMENTS

It is a pleasure to thank R. Williams, R. Klopfenstein, R. C. Alig, and S. Freeman for many helpful discussions.

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