

Hot electrons on liquid helium

Richard S. Crandall

RCA Laboratories, Princeton, New Jersey 08540

(Received 10 February 1975)

Using a model of electrons trapped in image-potential-induced surface states, I have calculated the mobility of electrons moving parallel to the liquid surface in high electric fields. To obtain agreement with experiment, it is necessary to include a large number of quantum levels of the electron motion perpendicular to the liquid surface. As the electrons are heated by the electric field they populate these higher states.

I. INTRODUCTION

The unique system of mobile electrons on the surface of liquid ${}^4\text{He}$ has received extensive study in the past few years.¹ This system, whose motion perpendicular to the liquid surface is quantized in hydrogen-atom-like levels, was predicted² to be free-electron-like. Most of the experiments^{3,4} are consistent with this view. The first calculation of the mobility of electrons moving parallel to the liquid surface indicated that gas atoms would be the dominant electron scatterers above about 1.2 K,⁵ whereas electron-surface-wave scattering would dominate at lower temperature. A more recent calculation⁶ showed that electron-surface-wave scattering was unimportant above about 0.85 K. Early mobility measurements^{3,7} showed only gas-atom scattering, whereas recent measurements⁸ indicate that there is an additional scattering mechanism below about 1.5 K. An increase in the mobility in high electric fields was predicted⁹ but it has not been observed. However, a decrease in the mobility with increasing electric field has been observed.^{10,11}

In this paper, I shall discuss a calculation of non-Ohmic electron transport on liquid ${}^4\text{He}$. In an earlier calculation⁹ of the non-Ohmic transport, I assumed the electrons to be in the ground state of the motion perpendicular to the liquid surface and remain there in high electric fields. This assumption ignores the increase in the population of higher states as the electrons absorb energy from the electric field. Therefore, it can not explain the commonly observed decrease in the mobility with increasing electric field. However, by including the population of these higher quantum states, it is possible to obtain good agreement with the experimental data. The main reason for the mobility decrease as higher quantum states become populated is because the density of states increases with increasing quantum level. Even though the scattering cross section decreases for the higher quantum states, the relaxation time increases for higher states because the density of states increases faster than the cross section decreases.

Therefore, as electrons populate higher quantum states, the momentum relaxation time and, hence, the mobility decrease.

In Sec. II, I will sketch a modification of the earlier two-dimensional theory⁹ that includes higher quantum levels of the perpendicular motion and also discuss the method used to calculate the scattering cross sections in these levels. In Sec. III, I shall compare the results of calculations with some recent non-Ohmic transport measurements.¹¹

II. THEORY

A. Mobility

The image-potential-induced surface-state model¹ has great appeal because of its simplicity. An electron is attracted to the liquid or other surface by the image-potential attraction arising from the polarization of the surface. If this polarization is not strong enough to overcome the repulsive exclusion-principle- interaction with the surface, the electron will be localized outside the liquid surface. The energy spectrum of this motion is nearly hydrogenic because the image potential varies inversely with the distance z from the surface. However, the finiteness of the potential barrier at the surface causes deviations from the hydrogenic energy spectrum.¹

One assumes the motion parallel to the liquid surface is free-electron-like. Theory¹² shows that electron-electron interactions cause the electrons to crystallize into a two-dimensional lattice, but the bulk of the experimental evidence¹ points to free-electron-like motion. Nevertheless, these measurements are not necessarily sensitive to the short-range correlations that certainly exist because of electron-electron interactions. With this reservation, I shall describe the parallel motion as free-electron-like.

The image-potential model has been widely discussed.¹ Therefore, I will not elaborate on the model except to say that the total energy E is the sum of the energy E_n for motion quantized perpendicular to the surface and the free-electron-like

energy E_K for the motion parallel to the surface; i. e., $E = E_n + E_K = -E_0/n^2 + \hbar^2 K^2/2m$, where E_0 is the binding energy, n is the quantum number, and \vec{K} is the wave vector parallel to the surface.

If more than one energy level is considered, the Boltzmann transport equation is no longer two dimensional. Nevertheless, its solution proceeds along the lines of my early calculation,⁹ which I shall henceforth refer to as I. Because energy and momentum changes are small in a collision, the distribution function $f(E_n, \vec{K})$ is

$$f(E_n, \vec{K}) = f(E) + \vec{g}(E) \cdot \vec{K}, \quad (1)$$

where $f(E)$ is the part of the distribution function that is isotropic in wave-vector space and depends only on the total energy of the electron. The vector $\vec{g}(E)$ depends on the energy level E_n and is in the direction of, and proportional to, the electric field \vec{F} . The Boltzmann equation then factors into two equations which are the same as Eqs. (7) and (8) in I with f_s replaced by $f(E)$. The energy E that appears in I now represents the total energy. The collision integral $\hat{C}f(E_n, \vec{K})$ [Eq. (15) in I] reduces as in I [Eqs. (18)–(23)], with the provision that when converting from the summation over quantum states to an integration over energy, the density of states $N(E)$ per unit energy per unit area is used. It is defined as

$$\begin{aligned} N(E) &= \frac{m}{2\pi\hbar^2} \sum_n \Theta(E - E_n) \\ &= N_0 \sum_n \Theta(E - E_n) = N_0 P(E). \end{aligned} \quad (2)$$

Here $\Theta(E - E_n)$ is the unit step function. The summation on n runs over all levels.

Because I am interested in analyzing measurements made above 1 K, only gas-atom scattering will be considered. This simplifies the calculations somewhat because the relaxation time τ does not depend on E_K . In the Appendix, I shall outline how to include other scattering mechanisms. Because the energy change in a collision is a small fraction of the electron energy, scattering events in which the quantum level changes are neglected.

The solution of the Boltzmann equation then proceeds along the lines in I. Details are in the Appendix. The momentum relaxation time is a function of the total energy; i. e.,

$$\tau(E) = \tau_G [G_n P(E)]^{-1}, \quad (3)$$

where τ_G is defined by Eq. (26) in I. The function G_n is the ratio of the scattering cross section in each quantum level n to the ground state $n = 1$. Similarly the energy-loss-rate function $W(E)$, defined by Eq. (27) in I, is now

$$W(E) = 3(m/M) \tau_G^{-1} G_n P(E), \quad (4)$$

where M is the mass of a gas atom. This expression corrects a numerical error in I. For the ground state, $P(E) = G_n = 1$, and τ and $W(E)$ are the same as in I.

The unnormalized electron distribution function $f(E)$ is

$$f(E) = \exp\left(-\frac{1}{k_B T} \int_{E_1}^E \frac{dE'}{1 + e^2 F^2 \tau(E')/k_B T m W(E')}\right). \quad (5)$$

Here e is the electron charge and k_B is Boltzmann's constant. This expression is the same form as Eq. (13) in I. However, as notated above, $\tau(E)$ and $W(E)$ are functions of the particular quantum level, and the integration is over the total energy. If the integration variable in Eq. (5) is changed from E' to E'_K , $f(E)$ can be readily evaluated. It is, using Eqs. (3) and (4),

$$f(E) = \exp\left(-\frac{1}{k_B} \sum_{n=1}^N \int_0^{E-E_n} \frac{dE'_K}{T_n}\right) \quad (6)$$

$$= f_N \exp[(-E + E_N)/k_B T_N], \quad (7)$$

$$f_N = \exp\left(-\sum_{n=1}^{N-1} \frac{E_{n+1} - E_n}{k_B T_n}\right), \quad (8)$$

and

$$\begin{aligned} \frac{T_n}{T} &= 1 + \frac{e^2 F^2 \tau_G^2 M}{3k_B T m^2 n^2 G_n^2} \\ &= 1 + \left(\frac{V_D}{V_R}\right)^2 \frac{1}{n^2 G_n^2}, \end{aligned} \quad (9a)$$

$$V_D = e\vec{F}\tau_G/m. \quad (9b)$$

The quantum number N is defined by

$$E_{N+1} > E > E_N. \quad (10)$$

As an example, $f(E)$ is plotted versus E in Fig. 1 at two values of V_D/V_R ; $T = 2$ K. To amplify the density-of-states effect, $G_n = 1$. A Maxwellian is shown by the dashed line. For $V_D/V_R = 3$, the distribution shows significant deviation from a Maxwellian. It is, however, of Maxwellian form in

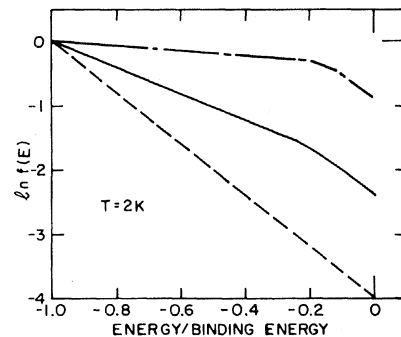


FIG. 1. Logarithm of $f(E)$ [Eq. (7)] vs. E/E_0 at 2 K for different values of V_D/V_R . For the dashed lines $V_D/V_R = 0$; solid line $V_D/V_R = 1$; broken line $V_D/V_R = 3$.

each level. In the ground state $T_1 = 10T$. In higher levels, T_n decreases markedly. $T_2 = 3.25T$, $T_3 = 2T$; $T_4 = \frac{25}{16}T$. This decrease in T_n is due solely to the increase in the density of states. For comparison purposes, the $f(E)$ were normalized to unity at $E = E_1$.

The transport coefficients are obtained in the usual manner. The mobility is

$$\mu = -\frac{e}{m} \int_{E_1}^0 dE E_K \tau(E) N(E) \frac{\partial f(E)}{\partial E} / \int_{E_1}^0 dE N(E) f(E). \quad (11)$$

After substituting Eqs. (2), (3), and (7) into Eq. (11) and performing the integration over energy, the mobility can be expressed as a summation over all electron-bound states. It is

$$\mu = \frac{\mu_0 \sum_{n=1}^{N_0} (T_n/G_n)(f_n - f_{n+1})}{\sum_{n=1}^{N_0} T_n n(f_n - f_{n+1})}, \quad (12)$$

where $\mu_0 = e\tau_C/m$ and N_0 is the quantum number of the highest-energy-bound state. Since I am omitting the continuum states, $f_{N_0+1} = 0$.

Once the G_n have been evaluated, Eq. (12) can be used for μ . The evaluation of the G_n is the subject of Sec. III. If all the G_n are the same, it is apparent from the structure of Eq. (12) that an increase in \bar{F} and hence, an increase in T_n leads to a decrease in μ . If there is a single level, then Eq. (12) shows that μ is independent of electric field.

To proceed further with a quantitative evaluation of the mobility expression, we must calculate the scattering cross section in each quantum level to determine G_n .

B. Electron wave functions and scattering cross section

The electron mobility measurements that I will analyze were made with an electric field F_1 normal to the liquid surface. This is in addition to the field \bar{F} parallel to the surface. The purpose of F_1 is to insure that the electrons remain near the surface at all temperatures. In the absence of F_1 , the states are hydrogenic and therefore subject to spontaneous ionization above about 0.5 K.^{1,13} This happens because as n increases, the level spacing goes to zero giving a divergent density of states and hence, a divergent partition function. This level degeneracy can be reduced by applying a small electric field perpendicular to surface. This will then allow the electrons to thermalize into the ground state by decreasing the statistical weight of the excited states. This fact was realized by some experimenters but apparently not by others and has led to a failure to observe the surface states in some cases.⁷

Therefore, it is necessary to consider the effect of this field on the density-of-states function, as well as the electron wave functions. The potential energy of a single electron is

$$V(z) = \begin{cases} -Ze/z + eF_1 z - V_s, & z > 0 \\ V_0 - V_s, & z = 0, \end{cases} \quad (13)$$

where V_s is the potential due to F_1 at $z = 0$, Z is the magnitude of the image charge,¹ and V_0 is the surface barrier.¹ If F_1 is small, it can be treated as a perturbation of the first few hydrogenic energy levels. Conversely, for the high quantum levels or in a large field, the image term can be treated as a perturbation. In this case, the unperturbed wave functions¹⁴ are Airy functions. For the energy levels in between these limits, the WKB method can be used to find the wave functions and energy levels.

It remains to calculate the matrix elements for gas-atom scattering. For gas-atom scattering, it is convenient to write

$$M_G^2 = \int d\bar{R}_G d\bar{R}'_G d\bar{R}_e d\bar{R}'_e |\Phi_G(\bar{R}_G)|^2 |\Phi_G(\bar{R}'_G)|^2 \times |\Phi_n(\bar{R}_e)|^2 |\Phi_n(\bar{R}'_e)|^2 V(\bar{R}_G, \bar{R}_e) V(\bar{R}'_G, \bar{R}'_e). \quad (14)$$

Here the subscripts G and e refer to the gas atom and electron, respectively. \bar{R} is the radius vector, and Φ_G and Φ_n are the wave functions of the gas atom and electron, respectively. The perturbation is

$$V(\bar{R}_G, \bar{R}_e) = \hbar^2 2\pi a_s m^{-1} \delta(\bar{R}_e - \bar{R}_G), \quad (15)$$

where a_s is the gas atom scattering length. The gas atom wave functions are plane waves as are the wave functions for electron motion parallel to the liquid surface. Substitution of Eq. (15) into Eq. (14) and subsequent integration gives

$$M_G^2 = \frac{(\hbar^2 \pi a_s)^2}{m} \int dz_e dz'_e e^{i\Delta G_z(z_e - z'_e)} \times |\Phi_n(z_e)|^2 |\Phi_n(z'_e)|^2. \quad (16)$$

Averaging M_G^2 over all momentum transfers ΔG_z inserts a $\delta(z_e - z'_e)$ in the integral which allows the integration over z'_e to be performed. Therefore,

$$M_G^2 = \frac{2\pi^2 (\hbar^2 a_s)^2}{m} \int dz_e |\Phi_n(z)|^4, \quad (17)$$

which is a convenient form since the integral is just the average value of the square of the electron charge density. The function G_n is, therefore,

$$G_n = \int dz |\Phi_n(z)|^4 / \int dz |\Phi_1(z)|^4. \quad (18)$$

This expression for G_n and the energy levels found by using the appropriate wave functions can be used with Eq. (12) to obtain μ . This will be done in Sec. III to analyze some recent experimental data.

III. COMPARISON BETWEEN THEORY AND EXPERIMENT

Non-Ohmic or hot-electron effects were first observed by Sommer and Tanner.³ They measured

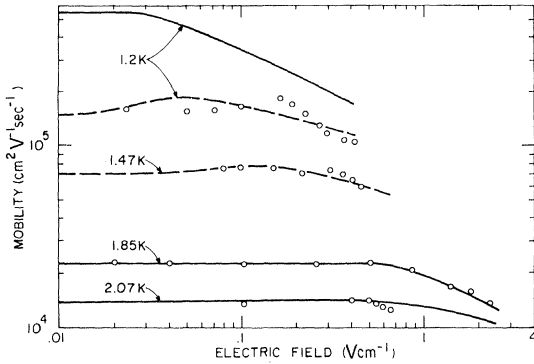


FIG. 2. Mobility vs the electric field parallel to the liquid surface and different temperatures. Field F_{\perp} perpendicular to the liquid surface is 20 V/cm. Circles are the data of Ref. 11. Solid curves from Eq. (12) with only gas-atom scattering. Dashed curves include a phenomenological surface scattering.

a mobility decrease with increasing applied electric field.¹⁰ This result can not be explained by the strictly two-dimensional theory in I, since τ_G is independent of energy. Therefore, even if the electrons are heated by the applied electric field, the mobility would not change. However, as Eq. (12) indicates, the population of higher quantum states of the electron motion perpendicular to the liquid, can lead to a decrease in μ .

Recent measurements of the electron-drift mobility were made by McGill and Bridges.⁸ They also measured the field dependence of the electron mobility in the temperature range between 1.2 and 2.1 K. Their data¹¹ are shown in Fig. 2. An electric field F_{\perp} of 20 V/cm was applied perpendicular to the liquid surface. The solid curves are calculated using Eq. (12) for the mobility. Expression (7) is used for the distribution function. I treated the field F_{\perp} as a perturbation to obtain the wave functions and energy-level spacing of the first three quantum levels. The next 17 levels were obtained using the WKB method. For the remaining levels, the image potential was omitted, making the wave functions Airy functions. Using these wave functions, Eq. (17) was integrated numerically to find M_G^2 and thus G_n . Only at the highest fields was it necessary to include more than the first 20 levels to obtain better than 1% precision. Both the magnitude and field dependence are in substantial agreement with experiment above 1.85 K. The agreement is surprisingly good considering that there are no adjustable parameters in the theory. However, one should note that the high-field behavior arises from an average of the relaxation time over many quantum levels and thus may not be sensitive to fine details of the wave functions.

On the other hand, the theory, if only gas-atom

scattering is considered, is at variance with experiment below 1.85 K. Both the Ohmic and non-Ohmic values of μ are higher than the experimental values. This is shown by the solid curve at 1.2 K. The result that the calculated mobility values are larger than the experimental values means that an additional scattering mechanism becomes important at low temperature. It is presumably the helium surface. The scattering can be described by the electron-ripplon interaction.^{5,6} However, because there is still some controversy over the correct formulation of this interaction and, furthermore, because the experimental data is limited at low temperature, I feel that a calculation using this interaction may not be justified at this time. In the Appendix, however, I show how to include this scattering mechanism.

Nevertheless, I shall attempt to analyze the data using a phenomenological energy- and temperature-independent scattering cross section to represent surface scattering. Furthermore, I assume that surface scattering is elastic. In this case, the momentum relaxation time is written as

$$1/\tau(E) = P(E)(G_n/\tau_G + R_n/\tau_s), \quad (19)$$

where τ_s is a constant that is determined by the best fit of μ to the experimental data at zero electric field. The energy-loss function $W(E)$ is still given by Eq. (4). R_n has the same meaning as G_n but applies to the surface-scattering mechanism. It is determined by the best fit to the non-Ohmic data.

The dashed curves in Fig. 2 were obtained in this manner with $R_n = n^{-4/3}$. While the fit could be improved by a different choice of R_n , there is not much point in going any further with this method of analysis. The real test of the non-Ohmic theory is at the higher temperatures where gas atom scattering predominates and, here, theory agrees well with experiment.

An idea of how each quantum level contributes to

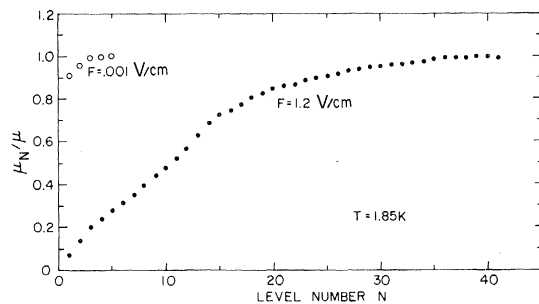


FIG. 3. Integrated mobility up to level N normalized to the total mobility vs level number at low and high fields at 1.85 K.

the current or mobility is given in Fig. 3 where μ_N/μ is plotted versus the level index N . μ_N is defined as the fractional contribution to the mobility of all the levels up to and including level N . The circles correspond to low electric field where the electrons are in thermal equilibrium. The dots correspond to a field of 1.2 V/cm. The liquid temperature is 1.85 K in both cases. The high-field results show that a large number of levels give a significant contribution to the current. Because the electric field F_\perp increases the level spacing, the higher levels are significantly above the hydrogenic binding energy E_0 . In fact, 50% of the current is carried by energy levels that are greater than $2.5E_0$ above ground state.

$$\hat{C}f(E, \vec{K}) = \frac{m}{2\pi\hbar^3} \sum \int d\theta dE' P(E') M_R^2(E') \{ [f(E', \vec{K} - \vec{q}) n_R - f(E, \vec{K})(n_R + 1)] \delta(E' - E + \hbar\omega) + [f(E', \vec{K} + \vec{q})(n_R + 1) - f(E', \vec{K}) n_R] \delta(E' - E - \hbar\omega) \}, \quad (A1)$$

where $M_R^2(E)$ is the absolute square of the matrix element for electron-rippion scattering, n_R is the ripplon occupation density, and $\delta(E)$ is the energy-conserving δ function. Since I am considering only scattering events in which the electron quantum number n remains unchanged, the summation over n' contains only the term $n' = n$. The δ functions in Eq. (A1) are expanded as in I and then $\hat{C}f(E, \vec{K})$ separates into two terms; one that is anisotropic in wave vector space:

$$\hat{C}(\vec{g} \cdot \vec{K}) = \vec{g} \cdot \vec{K} / \tau(E) = \frac{mA}{2\pi\hbar^3} \int d\theta \vec{g} \cdot (\vec{K}' - \vec{K}) M_R^2(E) P(E) (2n_R + 1), \quad (A2)$$

and one that is isotropic in wave-vector space

$$\hat{C}f(E) = \frac{mA}{2\pi\hbar^3} \frac{\partial}{\partial E} \int d\theta M_R^2(E) P(E) \hbar\omega \times \left(f(E) + \hbar\omega (n_R + \frac{1}{2}) \frac{\partial f(E)}{\partial E} \right) \quad (A3)$$

$$\approx \frac{\partial}{\partial E} \left[E W(E) \left(f(E) + k_B T \frac{\partial f(E)}{\partial E} \right) \right] \quad (A4)$$

ACKNOWLEDGMENTS

I am grateful to James McGill and Frank Bridges for supplying me with their data prior to publication.

APPENDIX

To show in some detail how the Boltzmann equation can be reduced from an integro-differential equation to a first-order differential equation, I shall consider liquid-surface-wave scattering. The interaction with the surface is conveniently treated by quantizing the surface waves. In their quantized form, they are called riplons. The collision integral, Eq. (18) in I, becomes for this electron-rippion interaction

Equations (A3) and (A4) define the energy-loss function $W(E)$. Similarly, Eq. (A2) defines the momentum relaxation time $\tau(E)$. The distribution function $f(E)$ can be found by equating Eq. (A4) to Eq. (11) in I, which is

$$\hat{C}f(E) = -\frac{e^2}{m} F^2 \frac{\partial}{\partial E} \left(E \tau(E) \frac{\partial f(E)}{\partial E} \right) \quad (A5)$$

and solving the resulting differential equation.

If fact, because the equations are linear, gas-atom and ripplon scattering can be combined to give

$$1/\tau(E) = 1/\tau_G(E) + 1/\tau_R(E), \quad (A6)$$

and

$$W(E) = W_G(E) + W_R(E), \quad (A7)$$

where the subscripts G and R refer to gas-atom and ripplon scattering, respectively. The differential equation resulting from equating Eqs. (A4) and (A5) can readily be solved for the distribution function. The momentum relaxation time $\tau_R(E)$ and the energy-loss function $W_R(E)$ can be calculated once the form of $M_R^2(E)$ is known.

¹M. W. Cole, Rev. Mod. Phys. **46**, 451 (1974).

²M. W. Cole and M. H. Cohen, Phys. Rev. Lett. **23**, 1238 (1969); V. B. Shikin, Zh. Eksp. Teor. Fiz. **58**, 1748 (1970) [Sov. Phys. -JETP **31**, 936 (1970)].

³W. J. Sommer and D. J. Tanner, Phys. Rev. Lett. **27**, 1845 (1971); T. R. Brown and C. C. Grimes, *ibid.* **29**, 1233 (1972).

⁴C. C. Grimes and T. R. Brown, Phys. Rev. Lett. **32**, 280 (1974).

⁵M. W. Cole, Phys. Rev. B **2**, 4239 (1970).

⁶V. B. Shikin, Zh. Eksp. Teor. Fiz. **60**, 713 (1971) [Sov. Phys. -JETP **33**, 387 (1971)]; *ibid.* **61**, 2053 (1971) [Sov. Phys. -JETP **34**, 1095 (1972)].

⁷R. M. Ostermeier and K. W. Schwarz, Phys. Rev. Lett. **29**, 25 (1972).

⁸J. McGill and F. Bridges, Bull. Am. Phys. Soc. **19**, 284 (1974).

⁹R. S. Crandall, Phys. Rev. A **6**, 790 (1972).

¹⁰W. T. Sommer (private communication).

¹¹J. McGill, PhD. thesis (University of California, Santa

- Cruz, 1975) (unpublished).
- ¹²R. S. Crandall and R. Williams, *Phys. Lett. A* 34, 404 (1971); R. S. Crandall, *ibid. A* 9, 2136 (1973); P. M. Platzman and H. Fukuyama, *Phys. Rev. B* 10, 3150 (1974).
- ¹³R. S. Crandall and R. Williams, *Phys. Rev. A* 5, 2183 (1972).
- ¹⁴L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon Press, London, 1958), p. 70.