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## LOW-TEMPERATURE MOBILITY OF HEAVY IMPURITIES IN FERMI LIQUIDS\*

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The temperature dependence of the mobility  $\mu$  of a heavy impurity in an interacting Fermi liquid is found to be of the form  $1/\mu = A + BT^2 \ln(1/T) + O(T^2)$  for low  $T$ . The coefficients are calculated for the case of a large, hard-sphere impurity. The result is in qualitative agreement with recent experimental data for negative ions in  $\text{He}^3$ .

We present theoretical arguments for the temperature dependence of the mobility  $\mu$  of a heavy impurity in an interacting Fermi liquid. For a hard-sphere impurity of radius  $a$  ( $k_{\text{F}}a \gg 1$ ) we find

$$\frac{1}{\mu} = \frac{1}{3\pi} k_{\text{F}}^2 (k_{\text{F}}a)^2 [1 + (8/315)(k_{\text{F}}a)m^*k_{\text{F}}V(2k_{\text{F}}) \times (T/\epsilon_{\text{F}})^2 \ln(\epsilon_{\text{F}}/T) + O(T^2)] \quad (1)$$

for  $T \ll \epsilon_{\text{F}} \equiv k_{\text{F}}^2/2m^*$ , where  $k_{\text{F}}$  and  $m^*$  are, respectively, the Fermi momentum and the effective mass of the quasiparticles.  $V(2k_{\text{F}})$  represents the backward scattering amplitude for two quasiparticles at the Fermi surface with opposite momenta. The  $T^2 \ln T$  term is a consequence of the Friedel density oscillations around the impurity and would be absent for a noninteracting Fermi liquid to which the previous investigations have been directed.<sup>1-4</sup> Equation (1) agrees qualitatively with recent measurements<sup>5</sup> of negative-ion mobilities in liquid  $\text{He}^3$  as shown in Fig. 1 and discussed in the last paragraph. We only outline the derivation of (1) here; the details will be published elsewhere.

The Hamiltonian of the impurity-liquid system is

$$H = H_0 + \int d^3x \rho(\vec{x})U(\vec{x} - \vec{z}(t)), \quad (2)$$

where  $H_0$  is the Hamiltonian of the liquid without the impurity,  $U$  is the impurity-fermion interaction,  $\rho(x)$  is the density of the liquid, and  $\vec{z}(t)$  is the position of the impurity. Consider a small oscillation  $z = z_0 \sin \omega t$ . The mobility can be obtained from the formula  $P = \dot{z}^2/\mu$  in the limit of small  $\dot{z}$ , where  $P$  is the power dissipated by the impurity. We expand  $H$  to first order in  $z$  to obtain

$$H = H' - zF, \quad (3)$$

where

$$H' = H_0 + \int d^3x \rho(\vec{x})U(\vec{x}),$$

$$F = \int d^3x \rho(\vec{x})\dot{z} \cdot \nabla U(\vec{x}). \quad (4)$$

$P$  can be obtained from the Golden Rule and we find

$$1/\mu = \lim_{\omega \rightarrow 0} (1/2\omega) \int dt e^{i\omega t} \langle [F(t), F] \rangle, \quad (5)$$

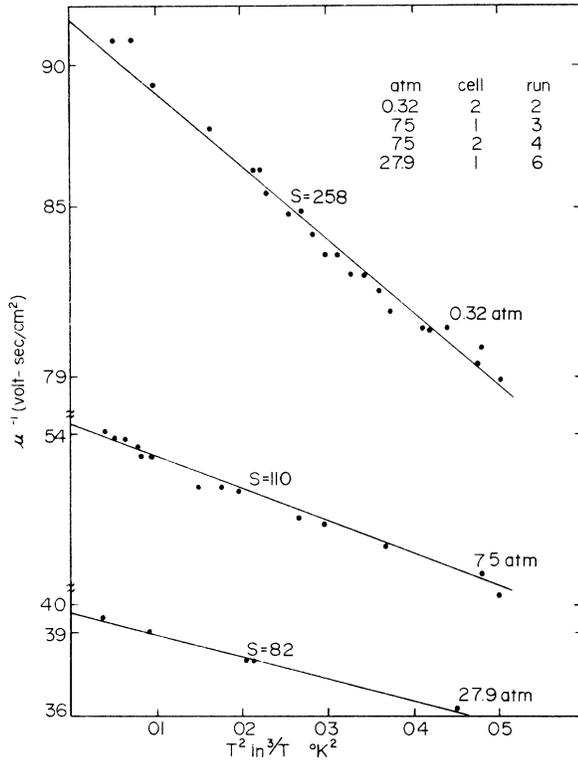


FIG. 1. Temperature and pressure dependence of the mobility of negative ions in liquid  $\text{He}^3$ . The experimental data are taken from Appendix 3 of Kuchmir's thesis (Ref. 5). The straight lines represent the best fit through the experimental points. Note units of  $1/\mu$ .

where  $F(t)$  is the Heisenberg operator  $e^{iH'T_F}e^{-iH't}$  and the average  $\langle \dots \rangle$  is taken over the equilibrium ensemble of the liquid with the impurity fixed at the origin. In the derivation of (5) the recoil of the impurity was not taken into account and (5) is therefore applicable only to sufficiently massive impurities.

The calculation of  $\mu$  now becomes the problem of finding the density correlation function in the presence of a static external field  $U(x)$ . For a noninteracting Fermi liquid the problem reduces to solving the one-particle Schrödinger equation with the potential  $U(x)$ . If  $U(x)$  is spherically symmetric, (5) can be written, after some algebra, in the form

$$1/\mu = R(0) + \int_0^\infty d\epsilon f(\epsilon) [R'(\epsilon) - R'(-\epsilon)], \quad (6)$$

where  $f(\epsilon) = [\exp(\epsilon/T) + 1]^{-1}$ ,

$$R(\epsilon) = (16m^2/3\pi k^2) \sum_l (l+1) |A_l(\epsilon)|^2, \quad (7)$$

$$A_l(\epsilon) = \int_0^\infty dr y_l(\epsilon, r) y_{l+1}(\epsilon, r) [\partial U(r)/\partial r], \quad (8)$$

$y_l$  satisfies the radial equation

$$[d^2/dr^2 + k^2 - 2mU(r) - l(l+1)/r^2] y_l = 0, \quad (9)$$

and

$$\lim_{r \rightarrow \infty} y_l = \sin(kr + \delta_l - \frac{1}{2}\pi l). \quad (10)$$

In the above,  $\epsilon$  is the particle energy  $k^2/2m$  measured from the Fermi energy and  $\delta_l$  is the  $l$ th phase shift for impurity-fermion scattering. At low temperatures we may expand  $R(\epsilon)$  in powers of  $\epsilon$  and obtain from (6)

$$1/\mu = R(0) + (\pi^2/6) T^2 R''(0) + O(T^4). \quad (11)$$

For a hard-core interaction of radius  $a$ ,  $A_l(\epsilon)$  is found to be

$$A_l(\epsilon) = (k^2/2m) \sin(\delta_l - \delta_{l+1}),$$

$$\delta_l = -j_l(ka)/n_l(ka), \quad (12)$$

which leads to the exact result

$$\frac{1}{\mu} = \frac{1}{3\pi} k_F^2 (k_F a)^2 \left[ 1 + \frac{1}{3} \pi^2 \left( \frac{T}{\epsilon_F} \right)^2 + O(T^4) \right] \quad (13)$$

for  $k_F a \gg 1$ . At this point we wish to clarify some confusion in the literature in regard to the behavior of  $\mu$  as  $T \rightarrow 0$ .

The authors in Refs. 1-4 have used the Boltzmann equation to determine the mobility of an impurity of mass  $M$ . An examination of the Boltzmann collision integral shows that the appropriate expansion parameter is  $(m/M)\epsilon_F/T$  as was mentioned in Ref. 1, and not  $m/M$  or  $T/\epsilon_F$  separately. In the limit  $(m/M)\epsilon_F/T \gg 1$  Clark, Abe and Aizu, and Schappert find  $\mu \sim 1/(MT)^2$ . The effective mass of the negative ion was estimated in Ref. 5 to be  $390M_{\text{He}^3}$  at a pressure of 0.26 atm, so that for  $T < 0.012^\circ\text{K}$  the infinite-mass approximation would fail and  $\mu$  would be expected to increase with decreasing  $T$ . A result similar to (13) was obtained in the limit  $(m/M)\epsilon_F/T \ll 1$  by Davis and Dagonnier.<sup>4,6</sup> Note that if one takes the limit  $m/M \rightarrow 0$  first as was done by Mott and Lekner,<sup>7</sup> the mobility would be a constant even at  $T=0$ . The effective mass of the positive ion is only  $40M_{\text{He}^3}$  at a pressure of 0.26 atm, so that for  $T < 0.12^\circ\text{K}$  the infinite-mass approximation will break down and  $\mu$  would be expected to increase with decreasing  $T$  as was observed qualitatively in Ref. 5.

Equation (6) shows that only the particles near the Fermi level contribute to  $1/\mu$ . For a normal

Fermi liquid, we take into account the interaction between the fermions by simply replacing the free-particle parameters in (7)-(9) by the corresponding quasiparticle parameters and  $U(x)$  by a self-consistent field. It is well known that around an impurity there is a long-range density oscillation<sup>8</sup> with wave number  $2k_F$ :

$$\Delta\rho(r) = -\frac{1}{2\pi^2 r^3} \sum_l (2l+1) \sin\delta_l \cos(2k_F r + \delta_l - l\pi) = -\frac{k_F}{2\pi^2 r^3} \text{Re}f(k_F, \pi) \exp(2ik_F r)$$

for large  $r$ , where

$$f(k, \theta) \equiv k^{-1} \sum_l (2l+1) \sin\delta_l \exp(2i\delta_l) P_l(\cos\theta).$$

$\Delta\rho(r)$  implies an oscillating part in the self-consistent field:

$$\Delta U(r) = \int d^3x' V(x') \rho(\vec{x} - \vec{x}') \approx -(k_F/2\pi^2 r^3) V(2k_F) \text{Re}f(k_F, \pi) \exp(2ik_F r), \quad (14)$$

where  $V$  is the effective quasiparticle interaction. Thus we expect that the backward scattering of the quasiparticles will be strongly modified and will be seen to lead to a term proportional to  $\epsilon^2 \ln \epsilon$  in the expansion of  $A_l(\epsilon)$ .

For simplicity we again consider the model of a hard-sphere impurity with radius  $a$ . The correction  $\Delta A_l$  due to the oscillating potential is found to be

$$\begin{aligned} \Delta A_l(\epsilon) &= k \int dr \Delta U(r) \cos(2kr - \pi l + \delta_l + \delta_{l+1}) \cos^2(\delta_l - \delta_{l+1}) \\ &= \frac{k_F}{4\pi^2} V(2k_F) \left\{ \frac{1}{2a^2} + (k - k_F)^2 \ln|k - k_F| + O[(k - k_F)^2] \right\} \\ &\quad \times \text{Re}f(k_F, \pi) \cos^2(\delta_l - \delta_{l+1}) (-)^l \exp -i(\delta_l + \delta_{l+1}). \end{aligned} \quad (15)$$

For  $ka \gg 1$  we make the semiclassical approximation for  $\delta_l$ :

$$\begin{aligned} \delta_l &= -ka + \frac{1}{2}\pi(l + \frac{1}{2}) + ka[1 - (1 - x^2)^{1/2} - x \sin^{-1}x] \quad \text{for } l + \frac{1}{2} < ka, \\ &= 0 \quad \text{for } l + \frac{1}{2} > ka, \end{aligned} \quad (16)$$

where  $x = (l + \frac{1}{2})/ka$ . Using (16), (15), and (6), we obtain Eq. (1).

The quantity  $V(2k_F)$  appearing in (1) is the only parameter yet unspecified. Since a realistic interaction generally contains a hard core,  $V(2k_F)$  must be interpreted as the backward-scattering  $T$  matrix for two quasiparticles at the Fermi surface with opposite momenta.

Several partial waves must be included in estimating  $V(2k_F)$  when  $k_F d$  is not small, where  $d$  is the core radius in  $V(\vec{x})$ . For liquid He<sup>3</sup> at intermediate pressures,  $d \sim 2.5 \text{ \AA}$  and  $k_F \sim 0.8 \text{ \AA}^{-1}$ . Thus  $k_F d \sim 2$  and the hard-core contribution to  $V(2k_F)$  was found by keeping five partial waves to be

$$V(2k_F)_{\text{hard core}} \approx -0.45(4\pi/m^*k_F). \quad (17)$$

The contribution of that attractive tail may be estimated by a direct Fourier transform. We write

$$V(2k_F) = -(4\pi/m^*k_F)b. \quad (18)$$

A complete calculation of the electron-bubble-helium interaction has never been done and is certainly beyond the scope of this paper. To interpret the negative-ion mobility data we shall simply treat the electron bubble as a hard sphere of effective radius  $a$  and include the additional term  $U_{\text{pol}}(r) = -\frac{1}{2}\alpha e^2/r^4$  due to the polarization of the liquid around the charged impurity. It can be shown that for a large sphere ( $k_F a \gg 1$ ) the constant term in (1) is multiplied by the factor  $[1 - (\alpha e^2/4a^4)\epsilon_F^{-1}]^2$  and the coefficient of the  $T^2 \ln T$  is unchanged. The magnitude of the polarization term is very sensitive to  $a$  due to its dependence on  $1/a^4$ . Taking  $\alpha = 0.22 \text{ \AA}^3$ ,  $\epsilon_F = 1.7^\circ \text{K}$ , the choice of  $a$  which gives the best agreement with the experimental value of  $1/\mu$  at  $T=0$  and  $0.32 \text{ atm}$  is  $a \approx 20 \text{ \AA}$ , and hence the polarization correction is negligible.

In the figure  $1/\mu$  is plotted against  $T^2 \ln(3/T)$ . By (1) and (18) the slope of the line is  $S = (128/945)m^* a^3 k_F b$ . The pressure dependence of  $a$

for negative ions is shown in Fig. 2 of Ref. 9 and gives  $a \approx 15.75$  and  $12.2 \text{ \AA}$  at pressures of 0.32 and 7.5 atm, respectively. The pressure dependence of  $S$  is dominated by  $a^3$  so that the corresponding ratio for  $S$  is  $(15.75/12.2)^3 = 2.15$ , which is in good agreement with the experimentally observed  $\sim 2.4$ . It is also seen that the predicted temperature dependence is qualitatively observed.

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<sup>1</sup>R. C. Clark, Proc. Phys. Soc. (London) 82, 785 (1963).

<sup>2</sup>R. Abe and K. Aizu, Phys. Rev. 123, 10 (1961).

<sup>3</sup>G. T. Schappert, Phys. Rev. 168, 162 (1968).

<sup>4</sup>H. T. Davis and R. Dagonnier, J. Chem. Phys. 44, 4030 (1966).

<sup>5</sup>A. C. Anderson, M. Kuchnir, and J. C. Wheatley, Phys. Rev. 168, 261 (1968); M. Kuchnir, thesis, University of Illinois, 1966 (unpublished).

<sup>6</sup>Notice that in Refs. 2-4, the impurity-fermion interaction was treated by a Born approximation or a pseudopotential, which is inadequate for an impurity of size larger than  $k_F^{-1}$ . Thus the overall numerical factor for  $\mu(T=0)$  may not be taken seriously.

<sup>7</sup>N. F. Mott and J. Lekner, "Ion Mobilities in Liquid He<sup>3</sup>" (to be published).

<sup>8</sup>For example, C. Kittel, Quantum Theory of Solids (John Wiley & Sons, Inc., New York, 1963), Chap. 18.

<sup>9</sup>B. E. Springett, M. H. Cohen, and J. Jortner, Phys. Rev. 159, 183 (1967).

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### EXCITONS IN AgCl†

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We report the absorption spectrum of AgCl sheet single crystals up to photon energies of about 5.5 eV. The present results do not confirm the existence of the peak reported in the literature previously and currently attributed to direct excitons. Instead, a structure similar to that already known to be present in the "tail" of the fundamental absorption of large AgCl single crystals is also found at energies up to about 4.1 eV.

The very existence of direct excitons in silver halide crystals is of considerable interest.<sup>1,2</sup> In this Letter we report the absorption spectrum of AgCl sheet crystals up to photon energies of about 5.5 eV. Our results do not confirm the existence of the peak, reported in the literature previously, that has currently been attributed to direct excitons. This peak appears to be characteristic of vacuum-deposited films, but not of single crystals. Instead, a structure similar to that already known to be present in the "tail" of the fundamental absorption of large AgCl single crystals is also found at energies up to about 4.1 eV.

The problem of direct excitonic transitions in the silver halides has been the object of several experimental investigations,<sup>3-5</sup> for which, however, vacuum-deposited films rather than single crystals were used. This was due to a high ab-

sorption constant, requiring the use of very thin films.

Large single crystals free from strains and very pure have been used by Brown *et al.*<sup>6,7</sup> for absorption measurements in the range from about 3.2 to about 3.4 eV. These authors have observed a structure in the "tail" of the fundamental absorption edge attributable to indirect excitonic transitions.

Spectral reflectivity response has been measured on different types and qualities of samples<sup>8,9</sup> showing some correlation with absorption spectra of vacuum-deposited films. A thorough investigation of reflectivity properties of very good AgCl crystals in the wide range 2-21 eV has been made recently at the University of North Carolina.<sup>10</sup>

Reflectivity may be strongly influenced by electronic states due to surface imperfections having no measurable effects on the bulk crystal absorp-