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ELECTRON MOBILITY TRANSITION IN A RANDOM SYSTEM OF HARD-CORE SCATTERERS*

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(Received 23 June 1969)

The mobility of an electron in an infinite system of randomly located identical hard-core scatterers is calculated to all orders in the density of scatterers. For fixed temperature a large drop in mobility is found at a characteristic density. The possible relevance of this result to the switching effect observed in amorphous semiconductors and the metal-insulator transition in a noncrystalline system is discussed.

The physics of systems displaying randomness in an essential characteristic is currently receiving considerable attention. The effect of randomness is presumed to be a common feature in numerous materials demonstrating metal-insulator transitions.[1] In theoretical investigations of electron transport, the introduction of a random potential at each site in the tight-binding model of a regular crystalline array leads to the Anderson transition.[2] Similarly, the presence of random fluctuations of charge density is responsible for the Mott transition when Coulombic interactions are involved.[3]

Transport in the presence of numerous identical scattering centers with randomness appearing in the spatial distribution has also been considered. Various authors have made quantum-transport calculations in the one-electron approximation. More recently, progress has been made on the problem of a system of interacting fermions.[4] All these theories consider a weak interaction and perform an expansion in powers of the strength of the interaction. The results are generally shown to be equivalent to those of the quantum Boltzmann equation in lowest order, with various corrections arising in succeeding orders of the expansion.

In this Letter we report the results of a mobility calculation in which a switching effect appears for an electron moving through a system of identical randomly distributed scattering centers. The Kubo[5] quantum-mechanical response formalism was used in the limits of weak uniform external field and s-wave (low-energy, spherically symmetric) scattering. An earlier paper[6] presented the lowest order work in which we derived the relaxation time τ characterizing the system and a mobility which showed a slight drop below the results of the semiclassical Langevin theory.[7] With the present extension of the work to all orders in the density two distinct changes are noted. A shift occurs in τ and, at constant temperature, the mobility shows a severe change of many orders of magnitude for a correspondingly small change in the density. In the language of Mott,[8] this is presumably caused by the appearance of localized electronic states.

For a weak external electric field in the z di-

rection, the electron mobility μ was shown by Kubo[5] to be given by

$$\mu = ieZ^{-1}\,\mathrm{Tr}\int_0^t e^{-itH}[z, e^{-\beta H}]e^{itH}p_z dt, \tag{1}$$

where p is the electron momentum operator, t is the length of time the system has been acted upon by the field, H is the Hamiltonian in the absence of the field, and $\hbar = m = 1$. Our specific model is described by

$$H = \tfrac{1}{2}p^2 + \sum_{j=1}^{N} V_j(\boldsymbol{r}_{ej}), \tag{2}$$

$$V(r) = 0, \quad r > a; \quad V(r) = \infty, \quad r \le a, \tag{3}$$

$$Z = \mathrm{Tr}\exp(-\beta H), \tag{4}$$

where a is the hard-core radius, N is the number of scatterers, and $\beta = 1/kT$. We consider a canonical ensemble of noninteracting electrons at temperature T. The assumption of random spatial distribution requires that μ be averaged over all possible configurations of the scatterers.

In our earlier work[6] we presented an expansion procedure tailored to the integrand of Eq. (1) which contains three exponentials, one of which is contained in a commutator. With the introduction of a leveling operator L and an ordering operator O it was possible to rewrite μ as

$$\mu = \beta e Z^{-1}\mathrm{Tr}\,p_z{}^2\exp(-\tfrac{1}{2}\beta p^2)\int_0^t dt\,\exp\{\langle \exp_{OL}(\sum_{j=1}^{N}[f_j(it) + f_j(\beta) + f_j(-it)]) - 1\rangle_c\}dt, \tag{5}$$

where $\langle\ \rangle_c$ denotes a cumulant average, and

$$f_j(x) = \exp[-\int_0^x \exp(\tfrac{1}{2}p^2 s)V_j \exp(-\tfrac{1}{2}p^2 s)ds\,] - 1. \tag{6}$$

The hard-core potential of Eq. (3) is replaced by an equivalent boundary value problem with the solution given by wave functions which represent an incident plane wave plus spherically symmetric outgoing scattered waves,[9]

$$\psi = (2\pi)^{-3/2}\left(e^{i\vec{k}\cdot\vec{r}} + \sum_{j=1}^{N}\frac{A_j e^{ikr_{ej}}}{kr_{ej}}\right), \tag{7}$$

where $\vec{k}$ is the set of quantum numbers for the electron energy. The coefficients A_j are evaluated by requiring ψ to vanish on the surface of each scatterer to order $N+1$ in the hard-core radius. The cumulant in the integrand of Eq. (5) is expanded as

$$\langle \exp\{\sum_{j=1}^{N}[f_j(it) + f_j(\beta) + f_j(-it)]\} - 1\rangle_c = \sum_n \frac{1}{n!}[\sum_{j=1}^{N}\langle f_j(it) + f_j(\beta) + f_j(-it)\rangle_c]^n. \tag{8}$$

When the thermodynamic limit ($N, V \to \infty$, N/V constant) is taken the cumulant expansion becomes equivalent to an expansion in the density ρ. Each order in ρ is further expanded in powers of ka. At this point the calculation bears similarity to the Gell-Mann and Brueckner expansion for the many-electron ground-state energy.[10] The diagrammatic representation will be seen to be in the form of single and double rings, and the integrals themselves resemble Coulomb-interaction terms. Only the leading (most divergent) terms in each order of ρ are retained and summation is taken before the integration. This analogy is mentioned only as an indication of the procedure employed and is not meant to imply any formal connection between the two works.[11]

The contributing terms correspond to linked diagrams proportional to $\rho^n a^{n+1}$. As an example, all the $n = 3$ terms generated by the multinomial expansion of the right-hand side of Eq. (8) are listed in Table I while Fig. 1 shows the diagrammatic representation of the nonvanishing terms which contribute to μ. The integrals corresponding to Figs. 1(a), 1(b), and 1(c) are [for general n and $f_j(it)$], respectively,

$$4\pi(-1)^n a^{n+1} n(n!)\int_0^\infty dk\,k^2 e^{-itk^2/2}\frac{\cos k(r_{e1} + r_{12} + \cdots r_{n-1,n} r_{ne'})}{r_{e1}r_{12}\cdots r_{n-1,n}r_{ne'}}, \tag{9a}$$

$$4\pi(-1)^n a^{n+1} n!\sum_{m=1}^{n-1}\int_0^\infty dk\,ke^{-itk^2/2}\frac{\sin k(r_{e1} + r_{12} + \cdots r_{m-1,m} + r_{me'} + r_{m,m+1}r_{m+1,m+2} + \cdots r_{nm})}{r_{e1}r_{12}\cdots r_{m-1,m}r_{me'}r_{m,m+1,m+2}\cdots r_{nm}}, \tag{9b}$$

$$4\pi(-1)^n a^n n!\int_0^\infty dk\,ke^{-itk^2/2}\frac{\sin k(r_{e1}r_{12} + \cdots r_{n-1,n}r_{ne'})}{r_{e1}r_{12}\cdots r_{n-1,n}r_{ne'}}. \tag{9c}$$

Table I. All $n=3$ terms from right-hand side of Eq. (8).

Argument of $\langle\rangle_c$	ρ dependence	Contribution to μ
$f_j(it)f_k(it)f_l(it)$ $f_j(\beta)f_k(\beta)f_l(\beta)$ $f_j(-it)f_k(-it)f_l(-it)$	ρ^3	$\sim\rho^3a^4$
$f_j^3(it)$ $f_j^2(it)f_j(\beta)$ $f_j^2(it)f_j(-it)$ $f_j^3(\beta)$ $f_j(it)f_j^2(\beta)$ $f_j^2(\beta)f_j(-it)$ $f_j^3(-it)$ $f_j(it)f_j^2(-it)$ $f_j(\beta)f_j^2(it)$		None (eliminated by leveling operator L)
$f_j^2(it)f_k(it)$ $f_j^2(it)f_k(\beta)$ $f_j^2(it)f_k(-it)$ $f_j(it)f_k^2(\beta)$ $f_j^2(\beta)f_k(\beta)$ $f_j^2(\beta)f_k(-it)$ $f_j(it)f_k^2(-it)$ $f_j(\beta)f_k^2(-it)$ $f_j(-it)f_k^2(-it)$		None (eliminated by leveling operator L)
$f_j(it)f_k(\beta)f_l(-it)$	ρ^3	None (separable thru order ρ^3a^4)
$f_j(it)f_j(\beta)f_k(-it)$ $f_j(it)f_k(\beta)f_j(-it)$ $f_j(it)f_k(\beta)f_k(-it)$	ρ^2	None (separable thru order ρ^2a^3)
$f_j(it)f_j(\beta)f_j(-it)$	ρ	None (separable thru order ρa^2)

One complication of the present calculation is the necessity of retaining all moments in the expansion of the nth-order cumulant, whereas in the more familiar applications of cumulant theory it is usually possible to neglect all except the nth moment as being of lower order in N.[12] Thus it is necessary to evaluate expression (9c) even though Fig. 1(c) is separable (i.e., singly connected and thus unlinked). Closed-form expressions were obtained for all summations. The time integral and trace of Eq. (5) were performed numerically, the trace being evaluated in the limit of infinite volume with the Gaussian factor $\exp(-\frac{1}{2}\beta p^2)$ providing a cutoff.

The resulting expression for μ is a function of

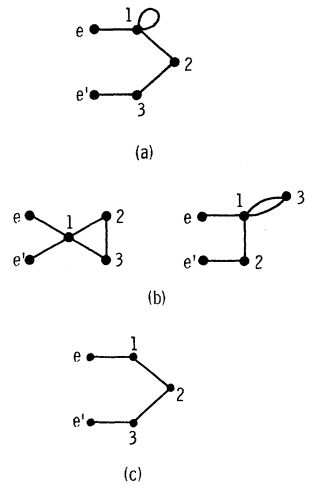

FIG. 1. Diagrams representing integrals contributing to μ for $n=3$.

β and the dimensionless parameter $R=2\pi\rho a\beta$. When $R\approx 2\pi$, μ undergoes a very severe change in response to small variations in R. Thus the switching effect is dependent upon the temperature as well as the density. Each expression entering the final result can be traced back to either one or the other of the two commutators in the linearized Liouville equation,

$$id\Delta P/dt=[H,\Delta P]+[\Delta H,P]. \tag{10}$$

The first commutator follows the motion in time of the perturbation of the total density matrix P under the influence of the equilibrium Hamiltonian and generates a relaxation time $\tau^{-1}=4\pi\rho a^2\beta^{1/2}\times(k^2+4\pi\rho a)^{1/2}$. This is in contrast to the first-order result[5] $\tau^{-1}=4\pi\rho a^2\beta^{1/2}k$. Thus one effect of including all higher terms is to shift k^2 by $4\pi\rho a$, which is exactly the shift in the free energy of an electron when it is introduced into a hard-core gas.[9] It is, however, the expression coming from $[\Delta H,P]$, the evolution of the equilibrium density matrix under the influence of the perturbing field, which causes the severe change in mobility. This leads to the conclusion that the basic mechanism responsible for mobility is no longer diffusion which is characterized solely by a relaxation time, but some other process. The fact that the sharp change is seen only when all orders of multiple scattering are considered, is independent of τ, and is in response to a randomly distributed set of scatterers is reminiscent of the circumstances governing the Mott and the Anderson transitions. The assumption is that the electron wave function ψ of Eq. (7) actually represents a series of spatially localized states

when the density is large enough. We have not yet been able to calculate the density of states in the neighborhood of the band edge ($\epsilon = 2\pi\rho a$). Such a calculation should show a tail representing the localized states developing on the single-scattering density of states[13] $[n(\epsilon) \sim (\epsilon - 2\pi\rho a)^{1/2}]$ and is contemplated for future work.

The above result for the mobility shows a reasonable correspondence with transport in helium vapor where, for thermal electrons, the electron-atom interaction can be characterized by a spherically symmetric hard-core repulsion.[14] The electron transport has been investigated by Levine and Sanders[15] using time-of-flight techniques at temperatures near 4°K and densities of the order of 10^{21} atoms/cm^3 with results as shown in Fig. 2. To our knowledge no physical process has been proposed which can adequately account for the features of the transition between high and low mobility.[16] Figure 2 also shows the result of the present theory which gives good qualitative agreement with the experiment in the transition region. This suggests that the electron's multiple interactions with randomly distributed scattering centers are sufficient to cause the observed decrease in μ with increasing density. The poor quantitative agreement is thought to arise from the lack of excluded volume between the scatterers (omission of the hard-core helium-helium interaction) which permits configurations contributing very strongly to the scattering. We hope to take account of this in future work since our present aim is to demonstrate the existence of a transition.

We conclude with speculation on the relationship of the above model to some currently observed phenomena involving disordered systems. Recall that our calculation was made in the limit of zero applied field. By use of the generalized Kubo formula the next order in the field can be computed. This should yield the analog of a coexistence curve, that is the field as a function of the temperature at which the transition takes place. We would expect that with a finite field the temperature at which the transition takes place would be lower, so that for a fixed density the transition temperature which we have found for zero field represents a critical temperature. This has been observed in the latest experiments on amorphous chalcogenide semiconductor films,[17] but the accuracy was not sufficient to give the shape of the coexistence curve in the neighborhood of the critical point. It appears that our simple model contains the basic features which

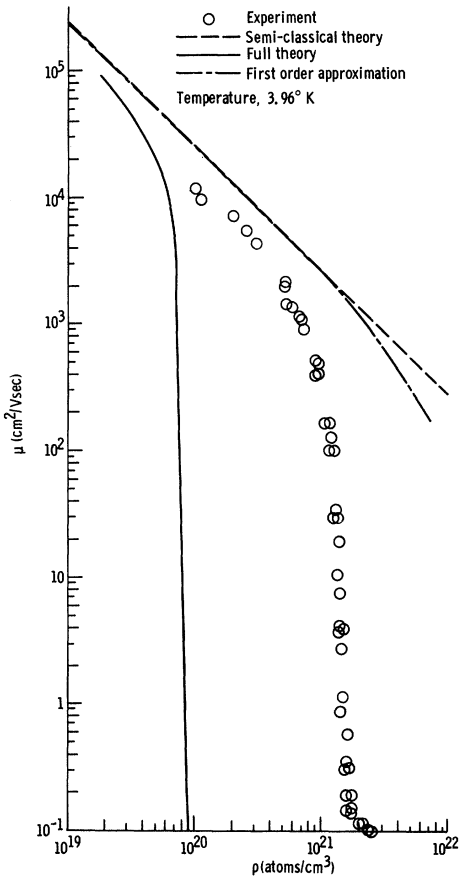

FIG. 2. Mobility versus number density at constant temperature for $a = 0.62$ Å.

are responsible for the effects outlined above. We hope to be able to demonstrate this quantitatively in the near future.

The authors are pleased to thank Professor Morrel Cohen for pointing out the possibility of regarding the present calculation as a simple model of a metal-insulator type of transition, and Professor Paul Zilsel for commenting on the effect of neglecting excluded volume between the scatterers.

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*Based on a dissertation submitted by Harold E. Neustadter to the Physics Department of Case Western Reserve University in partial fulfillment of the requirements for the degree of Doctor of Philosophy (unpublished).

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CRITICAL ATTENUATION OF SOUND BY SOFT MODES IN $SrTiO_3$

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(Received 7 July 1969)

Measurements of acoustic attenuation in $SrTiO_3$ reveal critical scattering behavior near the structural phase transition at $T_a \cong 105$ K. Power laws for the frequency and temperature dependences have been found. By these measurements the behavior of the soft mode frequencies could be studied in a temperature range one order of magnitude closer to T_a than by other methods so far.

It has recently been demonstrated[1-3] that the structural transition taking place in $SrTiO_3$ at $T_a = 105$ K has associated with it a soft optical mode at the R corner of the Brillouin zone for $T > T_a$. The mode in question represents a dynamic rotation of the oxygen octahedra. Below the transition temperature this dynamic rotation is superimposed on a net static rotation[4-7] φ giving rise to a tetragonal distortion and a splitting of the soft mode. The rotation angle φ is the order parameter of the low-temperature phase.

Although the experiments[1-3] have established significant lowering of the soft-mode frequencies $\epsilon_1(T)$ when approaching T_a, they have not revealed the very low frequencies that are to be expected in the range $T_a \pm 5$ K if the condition is truly second order.

This work was undertaken because one expected ultrasonic attenuation to be appreciably more sensitive to the temperature dependence of $\epsilon_1(T)$ in the region where these frequencies become really low: In the presence of coupling[6-9] between optical and acoustic phonons the expected high density of optical phonons when $\epsilon_1(T) \ll kT$ strongly suggests critical scattering of ultrasound at the transition.

In the present note we report the first observations of such critical scattering in $SrTiO_3$. The measurements were performed on a pure Verneuil-grown single crystal obtained from the National Lead Company. Acoustic propagation was chosen along a [100] direction. Double-ended operation with separate transducers for the generation and detection of ultrasonic waves was employed. The sample was mounted in a probe that was in thermal contact with liquid nitrogen through a copper rod. The temperature of the sample could be controlled by adjusting the temperature gradient in the rod. The runs were made using a chart-recording technique by which the ultrasonic signal was continuously monitored as a function of the thermocouple voltage repre-