Semi-Annual Technical Report
August 1, 1971 to January 31, 1972

ARPA Order No. 1562
Profram Code No. OD10
Name of Grantee
University of North Carolina at Chapel Hill
Effective date of grant
January 1, 1971
Expiration Date
December 13, 1971
Amount of Grant
$31,745

Grant No.
DA-AROD-31-124-71-G52
Principal Investigators
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Phone no. 919-286-2285
Short Title: Electronic States in Disordered Materials

Sponsored by
Advanced Research Projects Agency
ARPA Order No. 1562

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2nd Technical Report

"Studies of Electron States in Structurally Disordered Materials Using Simple Liquids and Dense Vapors as Prototypes."

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Summary
During the period of August 1, 1971 to January 31, 1972, we have completed the calculations of the electronic mobility in dense helium gas based on the theory of Eggarter and Cohen. We have also formulated a simplified Monte Carlo method for calculating the transport of hot electrons in dense vapors. The calculation takes into account multiple scattering, image field, and energy lost due to momentum exchange and/or inelastic scattering. The method is used to analyze data of hot electron injection into argon, hydrogen, and nitrogen. Experimentally, we have now obtained preliminary data on electron drift in helium at 77°K. Above about $2.5 \times 10^{21}$ atoms/c.c., we observed a sharp drop (about three decades) in the mobility. We plan to investigate this mobility edge further by making more temperature and field dependence measurements. The general methodology and results of the works summarized above are given in the following two sections:
I. Mobility of Excess Electrons in Helium Gas

The theory of Eggarter and Cohen\(^1\) (E.C.) was used to calculate the mobility of excess electrons in dense helium gas as a function of gas density and temperature. The experimental low field mobility is reproduced over its five orders of magnitude range as a function of gas density. The recently measured temperature dependence (4-20°K) is also obtained. The low field Hall mobility is calculated. In order to derive the electric field dependence of the drift velocity the occupation probability \(F(E,\epsilon,\rho,T)\) in the E.C. theory have to be modified. The detailed description of this work is contained in the attached reprint from the Physical Review.

II. Monte Carlo Calculations of Hot Electron Currents in Dense Vapors

(A.) Momentum Exchange Scattering.---Until recently most electron injection experiments in systems where momentum exchange scattering between electron and scatterers dominates have been analyzed using the Young and Bradbury\(^2\) theory. Young and Bradbury pointed out that the current is limited by the probability of the injected electrons to be back scattered into the electrode. They considered the probability for back scattering to be the ratio \(\hat{n}(x)/4\pi\) where \(\hat{n}(x)\) is the return cone for a scattering event at a distance \(x\) from the injection electrode. They then calculated the current assuming that only the first scattering event was important and that there were no energy loss processes. They also neglected the image field of the electrons. We report here a simplified Monte Carlo method for calculating the hot electron currents including multiple scattering the image field and energy lost due to momentum...
scattering. The simplification in the present method is based on the following less drastic assumptions:

(a) The collision is S-wave.

(b) The direction of motion of the electron after each collision is independent of the previous collision.

(c) The trajectories of the electrons are linear, and their path length $s$ is distributed according to $e^{-s/\lambda_p}$ where $\lambda_p$ is the momentum exchange mean free path.

Assumption (a) can be easily corrected to include other partial waves; assumption (b) is equivalent to the molecular chaos assumption. In the presence of the image and driving fields, assumption (c) has to be corrected to take into account trajectories with turning points and the fact that most trajectories will be bent toward the direction of the field.

Since such calculations would require a tremendous amount of computer time, we correct for the effect in the following approximate way: Let us consider an electron located at a position $x$ beyond the potential maximum $x_M$. See Fig. 1. The fraction of electrons that will be scattered backward by the image and applied field will be proportional to the return cone:

$$\alpha(x) = 2\pi \left[ 1 - \left( \frac{V_M - V(x)}{E_0 - V(x)} \right)^{1/2} \right] \quad (x > x_M)$$

Thus one can define a reflection coefficient

$$W(x) = \frac{\alpha(x)}{4\pi}$$
and a transmission coefficient
\[ T(x) = 1 - W(x), \quad \text{for } x > x_M. \]

For \( x < x_M \), \( T \) and \( W \) simply exchange roles. In our model, we shall correct for assumption (c) approximately by weighting the forward and backward trajectories by \( T(x) \) and \( W(x) \) respectively.

Thus our simplified Monte Carlo calculation proceeds by introducing one electron at a time; the length of the trajectory is given by a distribution \( \exp(-s/\lambda p) \); the direction \( \theta \) with the \( x \)-axis is given by the distribution \( 1 - \cos \theta \) with the forward and backward direction weighted by \( T(x) \) and \( W(x) \). After each momentum exchange collision the electron loses approximately \( \frac{2m}{M} (1 - \cos \theta) \) of its kinetic energy of the molecule. Thus \( E_0 \) has to be reduced accordingly until the final point where the kinetic energy becomes less than the potential maximum \( x_M \). The electron is then considered transmitted if its final position is greater than \( x_M \) and returned otherwise. The result of a typical calculation is shown in Fig. 2.

(B). Inelastic Scattering. For systems like hydrogen and nitrogen the effect of inelastic scattering is also important. The dominant inelastic processes in these cases are the excitation of rotational and vibrational modes of the molecules. Assume that \( \sigma_i \) is the cross section for a given inelastic process. In our Monte Carlo calculation the number of inelastic processes compared with the number of elastic processes is distributed according to the ratio \( \sigma_i/\sigma_p \). The possibility of an inelastic process is also limited by a threshold for the excitation of a rotational or vibrational mode.

Aside from these modifications, the method for generating the series of events for the passage of an electron remains the same to that described in section A. A typical result for \( T = 0^\circ\text{K} \) is shown in Fig. 3.
We are now trying to extend our calculation to include finite temperature effects.

When the energy relaxation is very rapid, it might be more profitable to calculate the current by solving the continuity of current equation. One such model worked out by M. Silver and P. Smejtek is based on the two fluid model, $\rho_h$ describing the hot electron density and $\rho_t$ describing the thermalized electron density. Assuming only a single relaxation time $\tau$, the following hydrodynamic equation can be set up:

$$- \frac{d}{dx} J_h - \frac{\rho_h}{\tau} = 0$$

where

$$J_h = - D_h \frac{d\rho_h}{dx} + \nu_h (E - \frac{e}{4\epsilon x^2}) \rho_h$$

and

$$- \frac{d}{dx} J_t + \frac{\rho_h}{\tau} = 0$$

where

$$J_t = - D_t \frac{d\rho_t}{dx} + \nu_t (E - \frac{e}{4\epsilon x^2}) \rho_t.$$ 

The diffusion constant and mobility are $D_h$ and $\nu_h$ for the hot electrons and $D_t$ and $\nu_t$ for the thermalized electrons. The boundary conditions are assumed to be

$$\rho_t(0) = 0$$

and

$$J_0 = \rho(0)\langle -v_{0x} \rangle + \frac{1}{\tau} \int_0^\infty \rho_h(x) \, dx$$

where $J_0$ is the injected current and $\langle -v_{0x} \rangle$ is obtained by averaging the time of flight over all angles of hot carriers at a distance of one mean free path from the electrode in the presence of applied and image fields. The hydrodynamic equations can easily be solved in the free diffusion
approximation, i.e. retaining only the diffusion term in $J_h$. The solution is

$$
\frac{J}{J_0} = (1 + 3C \frac{x_0^2}{\lambda})^{-1} \int_0^L \frac{\exp\left(-\frac{\mu t}{D} E + \gamma\right)x + \frac{\beta}{x}\right)dx 
\int_0^L \frac{\exp\left(-\frac{\mu t}{D} E + \frac{\beta}{x}\right)dx
$$

where

$$
\beta = \frac{e^2}{4\pi kT}, \quad C = \left< -\nu_{0x} \right>/\nu_0,
$$

$$
\nu_0 = \sqrt{(3kT/n)}, \quad x_0 = \lambda \nu_0 \tau / 3, \quad \text{and} \quad \gamma = -\frac{\mu h E}{2D_h} + \left(\frac{\mu h E^2}{2D_h} + \frac{1}{x_0^2}\right)^{1/2}.
$$

The validity of the free diffusion approximation will be checked against the finite temperature Monte-Carlo calculations once they become available. Smejtek and Silver have also checked the validity of the free diffusion approximation by exact solution of the hydrodynamic equations using a computer. The results show that the accuracy is best when the electron's m.f.p. is long (> 20 Å) and when the electron's mean energy is large (> 1 eV).

Experiment

Experimentally we have been working on two problems: 1) energy distribution of electrons in thin films and 2) drift mobility measurements in very dense gases.

The former problem continues to give us difficulty. What is required is to grow a few micron thick film of argon on our tunnel cathodes. To date we have been able to grow films but they are not good enough to make meaningful measurements. The idea of the experiment is
to inject hot electrons into the film, let them thermalize, heat them up again with an electric field and then measure their energy as they emerge from the film into vacuum by a retarding grid method. While the current is dependent upon the presence of the film, it does not vary with film thickness in a consistent way.

We will try to improve our film growing technique by using higher thermal conductivity substrates and by using helium gas as a buffer. If we do not have any success during the next quarter we will drop this experiment and go to a more productive area.

The drift velocity apparatus is functioning very well and we have our first preliminary data on electron drift in He at 77°K. Above about \(2.5 \times 10^{21}\) atoms/cc we observed a short drop in mobility. The drop is about three decades from about \(10^2\) to about \(10^{-1}\). This drop is occurred at densities below where Harrison and Springett\(^3\) have made measurements at 18.1°K. At present we cannot account for the difference. Our data are consistent with self trapping in a fully formed cavity while their data are consistent with the pseudo-bubble analyzed in the paper by Hernandez\(^4\).

We plan to make further temperature and field dependent measurements to clarify this point.

References

Argon

Density (atoms/cm³)

× 3.7 x 10⁻²¹
× 1.15 x 10⁻²²
× 1.8 x 10⁻²³

Eq. 2

10⁻¹⁸
10⁻¹⁹
10⁻²⁰
10⁻²¹
10⁻²²

1/1.0
10⁻¹
10⁻²
10⁻³
\[ \sigma_0 = 3 \times 10^{-16} \text{ cm}^2 \]
\[ \sigma_i = 3.5 \times 10^{-18} \text{ cm}^2 \]

\[ \Delta \bar{E}_i = 0.25 \text{ eV} \]
The recent theoretical work of Eggarter and Cohen (EC) is used to calculate the drift velocity of electrons in dense helium gas, in the neighborhood of the mobility edge. The purpose of this paper is to show that, within the context of the EC theory, it is possible to account for the drift velocity of electrons over a substantial helium density range, as EC have already shown, and also over a substantial temperature range. Also the theory can be extended to account for the effects of non-negligible electric field strengths.

In their letter, EC calculated the zero-field mobility of electrons in helium gas with densities from $10^{18}$ atoms/cm$^3$ to $25 \times 10^{18}$ atoms/cm$^3$ at temperatures of 3.68, 3.90, and 4.19 °K. This range of density and temperature allowed comparison with the data of Levine and Sanders. Recently, Harrison and Springett have performed measurements over a substantially larger temperature range. Their data are fairly extensive in the region $4.2 \leq T \leq 18.1$ °K. It is possible to take the theory, without change, and calculate the zero-field mobility over this extended temperature range. It is found that the parameters appropriate to 4 °K are not quite appropriate at higher temperatures, but a fit can be found. The sensitivity of the theory to the parameters will be shown.

Further, using a theory of Cohen and Lekner, suitably modified for this problem, the manifestation of the influence of non-negligible electric fields on the observed drift velocity of electrons can be calculated. A comparison of the results with the field-dependent data of Levine and Sanders shows adequate agreement but emphasizes the fact that,
as pointed out by EC, an intermediate-scattering mechanism has been neglected.

**Formalism**

The electron drift velocity is to be calculated using

$$v_0(\rho, T) = \frac{\langle \mu_0(E, \rho, T) \rangle}{}$$

where $\rho$ is the average density at temperature $T$, $\delta$ is the applied electric field, $E$ is the energy, $n$ is the density of states, $\mu_0(E)$ is the zero-field drift mobility, and $F$ is the occupation probability.

$n(E)$ and $\mu_0(E)$

A brief review of the EC method follows for the density of states and energy-dependent drift mobility follows.

The density of states is obtained by summing over those in cubic cells of side $L(E) = c \lambda(E)/\rho$, where $\lambda(E)$ is the de Broglie wavelength of an electron of energy $E$ and $c$ is adjustable. The helium density within each cell is assumed constant which gives rise to a potential $V(N) = N^2/2m$; with $hr = \tan \left( \frac{k}{r} \right)$, $\lambda^2 = L^2/N$, and $a$ is the electron-helium scattering length (0.62 Å). This is the Wigner-Seitz potential due to $N$ atoms in the cell and leads to a cell density of states

$$n_p(E) = 2L^3 \left( \frac{2m}{\pi a^3} \right)^{3/2} \frac{(E - V(N))^{1/2}}{} \text{ for } E > V(N).$$

The probability of having $N$ atoms in a cell, $P(N, T)$, is taken as a Gaussian about $N = \rho L^3$ with $(\Delta N)^2 = kT \rho / \rho_0 L^3$ (thus taking into account the nonideal gas dependence of pressure with density). Adding over the cells in the total volume $\Omega$ we have

$$n(E) = (\Omega/L^3) \int P(N, T) n_p(E) dN.$$

At a given energy, the drift mobility is obtained from

$$\mu_0(E) = \frac{2e}{3m} \frac{\lambda(E, \rho, T)}{} \rho(E, \rho, T)$$

$$+ \left[ 1 - \rho(E, \rho, T) \right] \mu_0(E, \rho, T).$$

$\rho(E)$ is the probability that at this energy the electron is in an extended state, the electron velocity $v_0$ has to be appropriately averaged over the fluctuating potential; the mean free path accounts for scattering from single atoms and from clusters making up the density fluctuations and is taken to be

$$\frac{1}{\lambda} = \rho_0 + \frac{1}{\lambda_p},$$

where

$$\lambda_p = \frac{L C(E)}{1 - C(E)}.$$

and $C(E)$ is the allowed cell concentration at this energy. This mean free path lacks scattering due to clusters of atoms between one and $(\Delta N)_\text{max}$ and thus overestimates the zero-field mobility at relatively low densities (as EC pointed out). The lack will also show up quite clearly in the field dependence of the electron drift velocity. The term with $\mu_0$ in the energy-dependent drift mobility, accounts for the motion of trapped electrons. It is taken as a pseudobubble mobility defined as

$$\mu_b = \frac{e}{\delta \eta} \left( 1 + \frac{9n_0}{4nR(2nM_n + T)^{1/2}} \right),$$

with

$$R = c' \left( \frac{3 \Delta N(E)}{4n} \right)^{1/3},$$

$$\Delta N(E) = N - N_{\text{max}}(E), \quad V(N_{\text{max}}) = E,$$

$\eta$ is the viscosity, and $c'$ is adjustable. $F(E)$

The occupation probability $F(E, \rho, T)$ can be obtained from a modification of the work of Cohen and Lekner on the Boltzmann equation for electrons undergoing collisions with structureless, isotropic, and uncorrelated objects of mass $M$. Their result is

$$F = \exp \left( -\frac{1}{kT} \int \frac{N \times dx}{x + b(x)} \right),$$

with

$$b(x) = \frac{e^2 g^2}{3kT} \lambda_0(x) \left( \frac{M}{2m} \lambda_1(x) \right),$$

$$\Lambda_0^i = \rho_0, \quad \frac{2m}{M \Lambda_1} = \frac{2m}{M \rho_0},$$

Thus, they account for momentum- and energy-exchange mean free paths. For our purpose, modifications must be introduced to account for the two types of scattering mechanisms of electrons in extended states. The modifications are embodied in

$$\Lambda_0^i = \rho_0 + \lambda_1^i$$

and

$$\frac{2m}{M \Lambda_1} = \frac{2m}{M \rho_0} + \frac{2m}{M^*} \frac{1}{\lambda_1},$$

with

$$b(x) = \frac{e^2 g^2}{3kT} \lambda_0(x) \left( \frac{M}{2m} \Lambda_1(x) \right) \rho(x),$$

$\rho(x)$ takes into account that the scattering processes refer only to electrons in extended states. $M^*$ is the effective mass, in an energy-exchange sense, of the density fluctuations and will be discussed later along with the results.

As long as pseudobubbles do not contribute appreciably to the zero-field mobility, it seems reasonable to neglect their possible energy gain from
ELECTRON DRIFT IN GASEOUS He:

No. 1. Zero-field drift mobility vs density at 4.2, 7.3, 11.6, and 18.1 °K. The points are calculated with the indicated parameters and $c' = 0.4$. The solid lines are experimental results from Ref. 3 at the various temperatures.

The electric field on grounds of a large effective mass and short mean free paths. A possibility of field ionization will also be neglected for now. Clearly the above neglects are completely unwarranted once the trapped electrons dominate the zero-field mobility. It will be shown that experimental data are only available for the regime where the neglect of trapped electrons is appropriate. Along with a lack of experimental data is the fact that the present context is unsuitable for treating the electric-field-trapped-electron interaction. For processes such as field ionization the details of the electron-helium correlation are important but unavailable in the present context.

RESULTS AND DISCUSSION

Low Fields

For negligible electric fields the EC theory applies unchanged. Figure 1 shows the calculated zero-field drift mobility versus helium density at various temperatures compared with Harrison and Springett’s measurements. It should be noted that the parameter $c$ has to be varied with temperature. As an indication of the sensitivity of the theory, the calculations are presented for a best fit and for a 4.2 °K calculation with $c = 1.05$ (which gives a best fit at 18.1 °K) and an 18.1 °K calculation with $c = 1.4$ (best fit for 4.2 °K). It is also noteworthy that only when $\mu_D < 1$ cm$^2$ sec$^{-1}$ V$^{-1}$ there any appreciable contribution due to the motion of trapped electrons (their contribution is 10% at $\mu_D \approx 1$ and essentially 100% at $\mu_D \approx 10^{-1}$). Thus the calculation is insensitive to $c'$ for $\mu_D > 1$. The mobility drop is due to a decrease, with density, of the fraction of the electrons in extended states. The calculation overestimates at helium densities around $5 \times 10^{16}$ cm$^{-3}$, as EC noted, due to the neglect of scattering by clusters of atoms between 1 and $(\Delta N)_{max}$ which should be effective at energies slightly above $V(N)$. It is possible that the effect of this neglect is being partially absorbed into a temperature dependence of the parameter $c$ but this does not completely account for its temperature dependence.

As a consistency check, the low-field Hall mobility may be interesting. It is easily calculated from

$$\mu_H(\rho, T) = \frac{\int \mu_D(E) n(E) e^{-E/kT} dE}{\int \mu_D(E) n(E) e^{-E/kT} dE}$$

and the results shown in Fig. 2 for parameters which give best fits to the drift mobilities.

Non-Negligible Field Effects

Experimental data concerning the field dependence of the electron drift velocity have been given by Levine and Sanders. It is immediately clear that the qualitative dependence will be obtained correctly here. At low gas densities the majority of electrons are in the free-electron regime, the important mean free path is $(a\rho)^{-1}$; as the distribution gains

FIG. 2. Zero-field Hall mobility vs density at various temperatures. The calculations are at the same parameters that gave the best fits to Fig. 1.
energy from the electric field, the main effect is an increase of the random velocity of the electrons and thus a decrease in the slope of \( v_\parallel \) vs \( \delta \) as observed experimentally. The effect is similar to increasing the temperature. At higher gas densities the majority of electrons are in the trapped regime. Nevertheless, it is still the electrons in extended states (now only a small fraction of the total) which dominate the observable transport behavior. The important mean free path is limited by scattering from fluctuations. As the electrons are allowed to gain energy from the field, the first observable effect is an increase in the mean free path with the resultant increase in the slope of \( v_\parallel \) vs \( \delta \). It is only at yet higher field strengths that the random velocity increase dominates with the resultant decrease in the slope of \( v_\parallel \) vs \( \delta \). Both effects are experimentally observed. At gas densities where the trapped electrons dominate the transport, one must deal with the correlated electron-helium behavior in the presence of a non-negligible field. In the absence of experimental data relevant to this regime, and since the present context is unsuitable to undertake this calculation, it will not be pursued further at this time.

For a quantitative calculation of field-dependent effects, one must face the problem of the energy gain from the field by electrons which percolate through the potential fluctuations. As previously stated, we have transformed the problem into a parameter \( M^* \). By assumption, when an electron is attempting to percolate and collides with a potential fluctuation, it will give up on the average \( 2m/M^* \) of its energy. This problem is similar to that considered by Davis, Schmidt, and Minday in that the electron is giving up energy to internal structure of the cluster of atoms, and in this sense \( M^* \) should be small in comparison to that of the helium atom while still large compared to the electron mass.

Calculations of the drift velocity versus electric field are shown in Fig. 3 for various parameters and compared with experimental values. The mass ratio \( M_\text{He}/M^* \) is irrelevant when the important mean free path is \((\rho v)^{-1}\) (always for low gas density and at higher densities, for large fields). It is only for intermediate field strengths and zero-field mobilities below \(-10^3\text{cm}^2\text{sec}^{-1}\text{v}^{-1}\) that the mass ratio causes a measurable effect. The results clearly show that a mass ratio of 1 is too small while curve c indicates that a value of 81 is too large. The intermediate value of 36 seems roughly appropriate.

To clearly exhibit the field and mass parameter effects, Fig. 4 shows the number of electrons at a given energy versus energy for curve b. At zero field most of the electrons are trapped. At
300 v/cm and a mass ratio of 1 the entire distribution has been shifted to higher energies and broadened; however, when the mass ratio is increased to 36 the electrons whose mean free path is limited by fluctuation scattering are relatively unaffected by the field while those whose mean free path is limited by single-atom collisions are unaffected by the mass-ratio change.

We return to the comparison with experiment in Fig. 3. The agreement is as good as that for the low-field calculations except in the region where the $v_D$ vs $S$ slope starts to decrease where the theory once again overestimates. This discrepancy is evidence of an overestimate in mean free path since it is clearly unaffected by the mass ratio. The overestimate emphasizes the missing scattering mechanism with clusters of more than one atom which would make its presence felt at energies slightly above $V_W$. A final remark is that the factor $p(x)$, in the occupation probability, which limits the field effects to the extended-state electrons has a negligible effect on the calculations performed. When the percolation probability $p(x)$ is small, the mean free path is so small that the field effect would be negligible even if $p(x)$ were taken at its maximum value of 1. This fact provides further support for neglecting trapped electrons in the field-effect calculations, since they would have a smaller yet mean free path, compounded by a larger effective mass, than the electrons in extended states.

In summary, good agreement with experiment is obtained for calculations as a function of gas density, temperature, and external field strength.

ACKNOWLEDGMENTS

The aid of T. P. Eggarter and M. H. Cohen in providing full details of their calculation in the form of a computer program is gratefully acknowledged. Extensive discussions with M. Silver are also acknowledged.

*Work supported by the Advanced Research Projects Agency of the Department of Defense and monitored by the U. S. Army Research Office-Durham under Grant No. DA-AROD-31-124-71-G62.


5In Ref. 1 it is stated that $\lambda_p$ accounts for the scattering by clusters of ~$N$ atoms. This should be $\sim (\Delta N)_{ser}$ since it is the potential fluctuations which are responsible for the scattering.


8A first guess might be $M^a$ equal to the reduced mass of $(\Delta N)_{ser}$ helium atoms.