Inhomogeneous gas model for electron mobility in high density neon gas

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Abstract

Experimental studies of electron mobilities in Neon as a function of the gas density have persistently shown mobilities up to an order of magnitude smaller than expected and predicted. A previously ignored mechanism (gas in–homogeneity which is negligible in the thermal mobilities for He and other gases) is found to reproduce the observed Neon mobilities accurately and simply at five temperatures with just one variable parameter. Recognizing that a gas is not a homogeneous medium, a variation in local density combined with the quantum multi–scattering theory, shifts the energy and cross section – which in turn changes the collision probability and finally the mobilities. A lower density where a momentum transfer interaction occurs moves the mobility strongly in the same direction as the anomalous experiments. By going backwards from the observed mobilities, the collision frequency at each temperature and density is made to reproduce the experimental data by looking for the local (as opposed to average) density at which the (rare) momentum transfer interactions occur. These density deviations give a picture of the size and behavior of the wave packets for electron motion which looks very much like the often discussed wave function collapse.

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I. INTRODUCTION

The study of the response of excess electrons in gases under the action of an externally applied electric field has attracted the attention of many researchers since very early times. On one hand, at quite low density, electron mobility measurements have been carried out to determine the electron-atom momentum-transfer scattering cross section in the framework of the traditional approach of the Boltzmann equation.

On the other hand, the transport properties of electrons in dense gases and liquids have also been and still are subject of extensive studies because of their potentiality in technologically relevant applications, such as high-energy particle detectors, as well as for the rich wealth of information they can provide on the basic physical mechanism of electron-atom interaction in disordered and condensed media.

The mobility of extra electrons can be used as a probe to investigate the nature, energetics and dynamics of electron states in a disordered medium and their evolution as a function of the gas density. In particular, the research is aimed at investigating the transition from classical single scattering in the completely dilute phase to multiple scattering and weak localization at low and moderate densities and finally leading to the formation of fully localized or extended states in the condensed phase.

Dense gases are the simplest realization of dense disordered systems and the problem of the electron motion in a dense environment of randomly located scatterers is a model problem for several phenomena in various areas of physics, such as the physics of doped semiconductors, non polar liquids, electrolytes in solution, non ideal plasmas.

The classical theory of scattering in the binary collisions approximation predicts that the electron mobility $\mu$ depends on the nature of the electron-atom interaction potential through the momentum transfer scattering cross section, and also on the gas temperature, on the applied electric field $E$, and that it is inversely proportional to the gas density $N$, so that the so called density–normalized mobility $\mu_0N$ at thermal energy (i.e., at zero electric field) does not depend on the density.

However, experiments have revealed anomalous density effects on the electron zero-field mobility. It is now well established that in certain gases $\mu_0N$ decreases with increasing $N$, thus showing a negative density effect, while in other gases the anomalous effect is positive, i.e., $\mu_0N$ increases with $N$. 
The amount of the density effect is a function of several factors, including gas density and temperature. Among the noble gases, $\mu_0N$ decreases by a factor $\approx 10$ in He at $T = 77.4$ K in the range up to $N \approx 60 \times 10^{26}$ m$^{-3}$, whereas it increases by a factor $\approx 30$ in Ar at $T = 152$ K in an extended density range going up to $N \approx 100 \times 10^{26}$ m$^{-3}$, or a little bit less at higher temperatures.

The sign of the density effect on the electron mobility has been found experimentally to depend on the sign of the electron-atom scattering length $A$, which distinguishes whether an atom attract or repels low energy electrons. It is observed that the effect has a sign in the opposite direction to $A$. The effect is positive for attractive gases such as Ar with $A < 0$ and negative for He and Ne with $A > 0$. The reasons for this are not immediately obvious and depend on the way that the quantum multiple scattering theory affects the mobility.

The multiple scattering theory (MS) of waves treats the environment in which a propagating electron wave is immersed as a Fermi’s infinite sea of atoms which acts as a potential well or barrier depending on whether the atoms attract or repel low energy electrons. At finite densities MS is a source of potential scattering, as distinct from the (relatively rare) momentum transfer interactions which change the electron’s total asymptotic energy.

The real part $\Delta$ of its effect raises or lowers the electron’s kinetic energy level accordingly. Its imaginary part $\Gamma$ acts rather as a quantum energy uncertainty or width which is associated with possible barrier tunneling.

The density effect in attractive gases is a straightforward application of multiple scattering theory. Starting with the classical equation for the mobility given by Huxley and Crompton and assuming the gas to be a homogeneous medium, one simply adds the shift to the electron energy.

When MS shifts the electron energy, this change is passed directly from energy to cross section to collision frequency – to electron energy distribution function – and finally to mobility. For Ar these changes predicted the observed density effects always in the direction found by experiment and with at least fair accuracy. However the effect for repulsive gases is not so simple and direct.

In the repulsive gases at the highest densities and low temperature, the electron-atom repulsion may be so effective as to give origin to the formation of electron states localized inside fluid dilations named bubbles, as it is well known in liquid He and Ne, dense He gas and dense Ne gas. No such an effect has been observed in gases with a positive...
density effect. The equation for repulsive gases like He and Ne at low and moderate densities

\[ \frac{\mu_0 N}{(\mu_0 N)_c} \]

FIG. 1: Experimental zero–field density–normalized mobility ratio \( \frac{\mu_0 N}{(\mu_0 N)_c} \) data in He gas at \( T = 77.4 \text{ K} \) (points) and the prediction of MS theory for a homogeneous gas (solid line).

(the subject of present interest) it is now commonly accepted that multiple scattering theory has the effect of defining a mobility edge such that electron states with large wavelength compared to their mean free path are effectively localized and do not propagate.

The mobility edge, as found by OMalley in 1980 and from a different point of view in 1992, is defined in Sec. II below with the corresponding equations for electron mobility. Alternative theories involving a mobility edge have also been advanced by other researchers. Ions shown in Sec. II below predicted fairly closely the negative density effect in He, H\(_2\), and CO\(_2\) and were expected to be valid for any repulsive gas. Figure 1 shows a typical case of the predicted and observed density effect in He at 77K. However, when the Neon data appeared, the relative values of its density–normalized mobilities for \( T = 46.5 \) and 47.9K were reduced by a full order of magnitude beyond what was predicted, and this disagreement persisted to room temperature. The low temperature situation is shown in Figure 2 where the observed mobilities relative to the classical values (\( N = 0 \)) are shown.

This strong disagreement shows that the theory was seriously insufficient for the special case of Neon and presents a serious challenge to theory if an unified picture is to be maintained for the physical process of electron scattering off noble gas atoms in a dense
FIG. 2: Experimental zero–field density–normalized mobility ratios \((\mu_0 N)/(\mu_0 N)_{cl}\) data in Ne gas at \(T \approx 46.5\), and \(T = 47.9\) K (points). Solid line: prediction of MS theory for a homogeneous gas.

In Section III we reexamine the generally overlooked assumption that a gas may be always treated as a homogeneous medium when electrons transfer momentum. What we have found is that the Neon experiments themselves tell us (through the collision probability \(\nu\)) that the assumption is not valid for Neon and that \(\nu\) actually favors momentum transfers occurring at densities less than the average.

II. EXISTING THEORY FOR THE THERMAL MOBILITY IN A GAS AS A HOMOGENEOUS MEDIUM

We start with an existing theory for drift and diffusion in a gas considered as homogeneous. Its basic component is the Boltzmann theory of Huxley and Crompton which treats the electrons as freely propagating classical particles with the energy balance determined by their momentum–transfer collisions, but with the collision frequency at the point of momentum transfer determined from the quantum mechanical cross section.

The effect of elevated density is first included through quantum multiple scattering theory, a generalization of the Fermi energy shift. The theory predicts a complex shift
in a free electron’s (kinetic) energy $\epsilon$ whose value is

$$\Delta \epsilon = \Delta + i \Gamma$$ \hspace{1cm} (1)$$

where

$$\Delta = 4\pi \text{Re} [f(0)] = -4\pi NA \hspace{1cm} (2)$$

and

$$\Gamma = \hbar N \sigma v \hspace{1cm} (3)$$

Quantities are generally in atomic units, (a.u.) ($m = e = h = 1$) with energy in Ry units (energy = $p^2 = v^2$). $f(0)$ is the forward electron-atom scattering amplitude, $A$ is the scattering length, $\sigma$ is its momentum transfer cross-section, and $v$ is the electron’s classical velocity ($v = p/m$).

For the present case of Neon, a repulsive gas (negative scattering length $A$) as is He, the problems created by shifting the lowest electron energy in the negative direction was solved in terms of a mobility edge $E_c$, and resulted in the following equation for the electron’s density-normalized mobility $\mu N$

$$\mu N = -\text{const} \int_0^{\infty} \left[ \frac{p}{\nu (p)} \right] \frac{dg}{d\epsilon} p^2 dp$$ \hspace{1cm} (4)

where

$$g(\epsilon) = \text{const} \int_0^{\epsilon} \left[ k_B T + \frac{C}{\nu^2 (p)} \right]^{-1} d\epsilon_0$$ \hspace{1cm} (5)

where $C = e^2 (M/m) E^2 / 3$. Eq. 5 defines the distribution function $g(\epsilon)$ for the electron’s energy of propagation $\epsilon = p^2$ in Rydberg units, where $p$ is the electrons momentum.

The initial $p$ in the integrand of Eq. 4 is the velocity (in a.u.), whose energy integral determines the drift velocity $v_D = (\mu N) \times (E/N)$. It effectively cancels the $p$ factor in the collision frequency $\nu$ (Eq. 3 below) so that the mobility in a repulsive gas depends effectively on $1/\sigma$ rather than on $1/(\sigma v)$.

In Eq. 4, the shifted energy of propagation is $p^2$ is defined by

$$p^2 = \epsilon - E_c \hspace{1cm} (6)$$

where $\epsilon$ is the un-shifted energy including states below the mobility edge $E_c$ which is defined as

$$E_c = p_c p \hspace{1cm} (7)$$
with

\[ p_c = 2N_{loc}\sigma(p_{av}^2) \] (8)

and \( p_{av} \) is the average momentum. The momentum or wave number \( p_c \) is the Kubo upper limit for diffusing electrons. Finally the very important collision frequency \( \nu \) in eq. 4, which directly determines the mobility or drift velocity, is

\[ \nu = N\sigma v = N\sigma(p^2)p \] (9)

where \( 1/N \) here is a measure of the volume over which the collision frequency (probability) is determined, and \( N_{loc} \) is the local density where the average momentum transfer occurs.

It is interesting that \( \bar{\hbar}\nu \) is equal to the mobility edge \( E_c \), and also to \( \Gamma \), the imaginary part of the multiple energy shift. \( \Gamma \) was originally identified as the mobility edge in 1980 with Eq. 4 predicting the electron mobilities closely in He, H\(_2\), N\(_2\) and CO\(_2\) with no free parameters.

III. TREATING THE GAS AS AN INHOMOGENEOUS MEDIUM (NEON)

The completely unexpected observations of Borghesani et al.\(^{17,24}\) in Neon have shown that the model in Sec. II (MS theory in a gas treated as homogeneous) is definitely overlooking something if it is to describe the special case of Neon as well.

In fact no gas is truly a homogeneous medium. A gas is rather a collection of individual atoms whose range for interacting with an electron wave is very small (a few atomic units). Therefore, in the region where a collision actually occurs, the local average density of atoms may be more or less than the global average density \( N \).

There was an earlier different but equally anomalous density effect observed by Schwarz\(^{27}\) in He. As the electric field strength was increased, the density effect rapidly increased from very negative up to zero and above. The effect of increasing the electric field is governed by the energy distribution function\(^{5}\), and the effect was finally understood and reproduced\(^{20}\) by recognizing first that the density need not be the same everywhere and second that the experiments were showing that the collisions which determines the energy distribution function throught the collision frequency have a strong preference for occurring in regions of less than average density.
For the present case of thermal Neon mobility, Borghesani and Santini\textsuperscript{17,24} noticed that adding a Fermi shift $-\Delta$ could move the mobility strongly in the direction of their anomalous experimental data, and also that the long-wavelength limit of the structure factor $S(0)$ needed to be included.

The theory in Sec. II already incorporates the full effect of the average density $N$. However, where the local density in Neon as an inhomogeneous medium is less than $N$, the local density $N_{\text{loc}}$ is reduced by

$$N_{\text{loc}} = N(1 - \delta)$$  \hspace{1cm} (10)

where $\delta$ is defined as $\delta = (N - N_{\text{loc}})/N$. This further shifts the energy $p^2$ of Eq. 3 by the Fermi shift $\Delta$ of Eq. 2 to

$$p^2 = \epsilon - E_c + \Delta(N_{\text{loc}} - N)$$  \hspace{1cm} (11)

If $N_{\text{loc}} < N$, i.e., if $\Delta N = -N\delta$ is negative ($\delta > 0$), then $\Delta$ for Ne or He in Eq. 11 increases the energy of propagation $p^2$ and $\sigma(p^2)$ and, by Eq. 9, increases the collision frequency $\nu$ (which determines both the distribution function and the mobility).

Thus, collisions are more probable at lower densities, and a larger $\nu$ also decreases the mobility $\mu$, moving it in the direction of the experiment, as was noted by Borghesani and Santini\textsuperscript{17,24}. (Conversely, where $\Delta N$ is positive the mobility would be moved away from experiment, but this is less probable and so the smaller densities should dominate.)

IV. PROCEDURE

Starting from the above pressure effect model of sections II and III, with the effects of multiple scattering, Kubo diffusion and the structure factor $S(0)$ given and the e–Ne cross sections determined\textsuperscript{28} as functions of energy it remains only to explore the effect of possible local density deficits $\delta$ of Eq. 10 on the mobilities to be calculated.

In particular we look to see how the $\Delta N$’s change the mobilities and whether a simple choice of $\delta$’s can match all the observed mobilities as a function of temperature and density. We have already shown how only negative values of the variation $\Delta N$ from average can move the mobility in the direction of experiment.
FIG. 3: Experimental zero–field density–normalized mobility ratios \( (\mu_0 N)/(\mu_0 N)_{cl} \) data in Ne gas at \( T = 47.9 \) K (solid squares) and \( T = 46.5 \) K (solid dots and open squares). Dotted line: prediction of the present one parameter MS theory for a inhomogeneous gas (dashed line). The agreement is quite good up to a density of \( \approx 70 \times 10^{26} \text{ m}^{-3} \).

V. RESULTS FOR THE MOBILITIES IN THE LOW \( E/N \) LIMITI

At each temperature (\( \approx 46.5 - 47.9, 77.4, 101.2, 196 \) and 294 K) for which mobilities have been measured and for all moderate densities, we found a single \( \delta \) which made the predicted mobilities match the observed very closely.

Figure 3 compares the observed pressure effect \( (\mu_0 N)/(\mu_0 N)_{cl} \) at 46.5–47.9 K to the present model with \( \delta = 9\% \). The fit may be seen to be good up to about \( N = 70 \times 10^{26} \) m\(^{-3}\), where additional high density mechanisms begin to be important.

Figure 4 shows the same comparison (in terms of the absolute values of \( \mu_0 N \)) at all 5 temperatures in the moderate density range. We note that the 101.2 K data were never published before.
FIG. 4: Absolute zero-field density normalized mobility $\mu_0N$ as a function of the gas density in neon gas for several temperatures ($T = 47.9, 77.4, 101.2, 196.0,$ and $293.0$ K, from top). The lines are the predictions of the present one parameter MS model for an inhomogeneous gas. The $101.2$ K data were never published before.

VI. DISCUSSION

A. Neon vs. Helium

Given the dramatic effect that inhomogeneities have on thermal mobilities in Neon, why is the same not true in He where the homogeneous gas model with no free parameters predicts the mobilities very well?

The answer is that the thermal mobility (Eq. 4) is determined by $1/\sigma$. In He the cross section is nearly constant, while in Neon it varies rapidly with energy especially at the lowest energies.

On the other hand, Schwarz found a dramatic effect for the electric field dependence of the non–thermal mobilities which was explained by a crude inhomogeneous gas model.

The reason that the electric field dependence in He is sensitive to density variations (unlike its mobility) is that, although the He cross section varies hardly at all, the contributions to the electron energy distribution are determined at small fields by $\nu^2 = (N\sigma p)^2$ by Eq. 5. So it is the significant shift in the momentum $p$ that causes Schwarz’ effect. (Unfortunately the
very small size of the Neon cross sections makes this electric field effect barely significant in Neon).

**B. Temperature dependence of the density defects $\delta$**

Figure 5 shows the values of our empirical $\delta$ as a function of temperature. They lie almost precisely on a straight line going from 9% at 47 K to 59% at 294 K and vanishing at T=0. As a result, the experiments at all 5 temperatures and moderate densities are all fitted closely by a $\delta(T)$ curve with an only single variable parameter

$$\delta = 0.202 T$$

(12)

The close agreement seems remarkable in view of the anomalous nature of the observations and their great distance from what was previously understood. What the agreement seems to be saying is the following.

According to the experiments and Eq. 12 the density deficit is proportional to $T$, which is itself proportional to the inverse square of the electron’s thermal wavelength $\lambda_T = h/\sqrt{2m_\text{e}k_B T}$. It follows that $\lambda_T^2$ goes to infinity at $T = 0$ K and so the density sampled over such a range can only be the average $N$. This means that $\delta$ must be zero there as Eq. 12 predicts. And, as $T$ increases the area $\lambda_T^2$ decreases. The smaller area allows progressively larger deviations from the average density $N$ also as Eq. 12 would predict.

**FIG. 5:** Fractional density deficit $\delta$ as a function of temperature. The solid line is the equation $\delta = 0.2T$. 
But what is the significance of the area $\lambda^2_T$? The interaction of an electron with an atom is known to be limited to a distance no greater than a wavelength. Thus, as the electron’s wavepacket advances forward, $\lambda^2_T$ may be interpreted as a measure of the largest area over which it can overlap with the e–atom interaction and sample the local density.

C. The quantum inferences

The theory used in this work starts with pure Boltzmann theory at very low density, but uses quantum cross sections for the important momentum-transfer collision frequency. It describes an electron moving classically in a well defined direction from one momentum/energy transfer to the next.

However, in light of quantum theory, we know that the electrons motion is physically described by a wave function spreading out broadly, with Feynman paths passing through every atom in its forward direction. The total wave may alternately be represented as a superposition of wave packets, each beginning at the electron’s starting point and ending on a possible interaction with an individual atom in its path. For each wave packet, there is a well defined momentum vector in between the starting point and any atom.

What the earlier close agreement of the classical Boltzmann theory with the low density experiments indicates is that, after a momentum/energy transfer with one atom, the electron moves forward starting from that one point. In other words all the remaining members of the original superposition become irrelevant as the electron must be continuing on from the point of momentum transfer as a new wave.

The narrowing of the original superposition plus the electron continuing from that point closely parallels Keller’s derivation of wave function collapse in which he proved, using conditional probability theory, that a direct observation results in a collapsed single component wave–function at the observation point ready to continue from there.

What these results add to Keller’s conclusion is that observation may be generalized to momentum transfer interaction. More detailed evidence of this collapse to something like a particle has been seen at higher densities, particularly in neon gas.

When the density $N$ was increased in electron mobility experiments, known density dependent mechanisms were recognized as influencing the outcome, the principal one being the multiple scattering of waves.
What this mechanism does first is to modify the electron wave–function’s kinetic energy, and therefore its collision frequency $\nu$ (Eq. 4) by the density dependent kinetic energy shift $\Delta$ (Eq. 3). ($-\Delta$ is the cumulative potential energy of the electron summed over all the atoms in the full wave propagating wave-function).

This potential scattering, resulting from multiple scattering, is in addition to the rare energy exchange at the actual momentum transfers. The latter is completely determined by $\nu$, as well as the energy balance, through $g(\epsilon)$, (Eq. 5).

D. The present Neon case

With Neon as the gas, the fact that its scattering cross section is close to zero and changing rapidly makes the collision frequency exceptionally sensitive to gas density where momentum transfers occur. This sensitivity enabled us, as discussed above, to explain and predict the anomalous density effect in Neon and also to discover that these transfers were occurring mostly at local densities less than the average one $N$ by an amount proportional to the temperature.

The fact that the present model’s densities at points of momentum transfer were found to be significantly different from the average $N$ implies that the individual wave packet at the interaction, even before moving forward, is more compact than the full wave packet during its propagation - making it look much more like a particle there.

This presently inferred shrinking or collapse of the electron’s extended wave packet at the point of momentum transfer is further evidence of the behavior of the wave function demonstrated by Keller about the point of observation.

Our finding from the experiments that the density defect $\delta$ is such that at most momentum transfers $N_{loc}$ can differ from the average $N$ by as much as 59 % accordingly shows that the wave function for an electron’s motion, where it interacts, is very much smaller than the free propagating wave function. It might understandably be called a particle – as in the photoelectric effect, where light waves become point–like photons when they exchange energy and momentum with electrons in a metal, or the way photons and electrons were described earlier when they were emitted or observed.

Where the present finding from Neon mobilities differs from Keller’s is first that it is the wave and wave packet for electron motion which shrinks and second that the collapse–like
behavior happens at every momentum/energy transfer whether or not it is observed directly.

VII. SUMMARY

A straight-forward extension of existing theoretical models, combining the inhomogeneous nature of a gas with multiple scattering theory, has been found to reproduce the anomalous and previously puzzling measurements of the pressure effect on electron mobilities in Neon gas at 5 temperatures and moderate densities both simply and accurately with only one variable parameter - the deviation $\delta$ (proportional to the temperature) of local from average gas density at actual momentum transfers.

The special window that these, and also previous, electron mobility experiments offer on fundamental quantum processes was also discussed.

Further experimental investigations of this kind of relation between collisions in a gas and electron (or atom wave) packets are strongly recommended.

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