Electron dynamics in condensed argon and xenon

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From accurate measurements of the drift velocity in condensed argon and xenon over a wide range of temperature (80–135 K for argon, 90–230 K for xenon) and of electric fields (5–1.5×10⁵ V cm⁻¹) the explicit form of the electron energy distribution function has been obtained so that the dynamics of electrons in these materials can be fully described. The description was based on the Landau "hot" electron theory. The mean electron energies and diffusion coefficients were calculated for different temperatures and electric fields. Values for the scattering cross sections for electrons in condensed argon and xenon at different temperatures were obtained.

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Condensed and highly compressed noble gases are constantly attracting the attention of investigators designing new apparatus for nuclear physics, as possible working materials for radiation detectors. Prediction of the possible characteristics of detectors, especially with electronic data collection, requires a knowledge of the behavior of charge carriers in these materials.

Measurement and theoretical calculations of such quantities as the drift velocity of electrons, their mean energy, diffusion coefficient, and the scattering cross section at different temperatures and electric field strengths are also important from the point of view of the physics of condensed matter.

However, experimental results are at present very scanty and contradictory even for argon, which is the gas most studied, while information about the dynamic characteristics of electrons is practically non-existent. Measurements of drift velocity were carried out either for a fixed value of electric field strength in the temperature range from the triple point T₃ to the critical temperature T₄ (Refs. 2–5), or on the other hand over a wide field range but at a fixed temperature, usually near T₃ (Refs. 6–10).

We have earlier, given preliminary results of studies of drift properties of condensed argon and xenon and, in particular, of an attempt to describe electron drift in argon in the range 70 to 110 K and 0.8 to 8 kV cm⁻¹ with the help of the Davydov distribution. In the present work we give the results of accurate measurements of electron drift velocities in condensed argon (80 to 135 K; 20 to 1.5×10⁵ V cm⁻¹) and xenon (80 to 230 K; 5 to 1.5×10⁵ V cm⁻¹) and also the results of calculations of the dynamic characteristics of electrons based on the experimental results obtained.

METHOD AND EXPERIMENTAL ERRORS

Measurements of drift velocity were carried out with apparatus similar to that described earlier. We used a modified Townsend pulsed discharge method, firstly, as the most direct time-of-flight method of measuring the drift velocity and, secondly, as the most direct method of measuring and controlling the purity of the condensed matter. In this method the electron component of the current or voltage pulse which arises in a two-electrode ionization chamber on irradiation from the lower electrode side by a short pulse of x rays is amplified and analyzed with, for example, an oscilloscope. It can be shown that for an amplifier time constant RC large enough an accurate value of the drift velocity is

\[ V = \frac{L}{t} \left[ 1 + \frac{\tau_m}{2} \left( \frac{1}{RC} \frac{1}{t} \left( 1 - \frac{L}{D} \right) \right) \right]. \]

where \( L \) is the drift distance, \( r \) the duration of the x-ray pulse, \( \tau_m = L_m/V \), \( L_m \) is the effective penetration depth of the x-ray beam into the material under investigation, defined as the tenfold attenuation layer, \( t \) is the rise time of the voltage pulse, measured by the intersection of the extrapolation of the linear part of the pulse rise front with the zero and maximum amplitude levels, and \( D \) is the mean electron path before capture, inversely proportional to the concentration of electronegative impurities (mainly oxygen). As can be seen from the above expression, neglect of the correction in the curving brackets can lead to a considerable error in measurements of the drift velocity. In our experiments RC = 400 μs, % = 2 to 50 μs, \( \tau_m < 0.3 \mu s, \quad L_m = 1 \text{ to } 0 \text{ cm, the x-ray pulse energy if } 50 \text{ MeV} \cdot \mu \text{s}^{-1}, \text{ the maximum energy of the x-ray quanta is } 23 \text{ keV, } L_m = 30 \text{ to } 80 \mu \text{m, and } D/L < 0.03. \) The overall error in the measurement is governed also by the following factors:

1. The uncertainty in the distance between the electrodes. If the geometrical dimensions are measured with an accuracy of 0.1 mm and \( L = 1 \text{ cm, the error in the drift velocity is } 1\% \).
2. The error in measuring \( t \) on the oscilloscope screen is not worse than 1.5%.
3. The error in calibrating the horizontal oscilloscope sweep is not more than 3%.
4. The uncertainty \( \Delta U = \pm 0.1, \pm 10, \text{ and } \pm 150 \text{ V in the value of the voltage applied to the chamber in the ranges } 0–1, 1–3, 3–15 \text{ kV, respectively.}\)
5. The error in temperature measurement. The temperature could be measured to an accuracy no worse than \( \pm 0.5 \text{ K and could be stabilized to no worse than } \pm 0.1 \text{ K by using a temperature control system similar to that described earlier.} \)

The production of cathode plasma \({ }^{(1)} \) in the form \( 10^{-10} \text{ to } 10^{-7} \) mbar with anode fields \( 5 \times 10^5 \text{ to } 3 \times 10^{-8} \text{ mbar and \( 5 \times 10^{-8} \text{ to } 1 \times 10^{-7} \text{ mbar with anode fields } 5 \times 10^5 \text{ to } 1 \times 10^{-7} \text{ mbar.}\)} \)

The measurement of the electrode gap between charge populated plates \( 0.5 \mu \text{s to } 1 \mu \text{s \( t \) is the rise time of the voltage pulse, measured by the intersection of the extrapolation of the linear part of the pulse rise front with the zero and maximum amplitude levels, and \( D \) is the mean electron path before capture, inversely proportional to the concentration of electronegative impurities (mainly oxygen). As can be seen from the above expression, neglect of the correction in the curving brackets can lead to a considerable error in measurements of the drift velocity. In our experiments RC = 400 μs, % = 2 to 50 μs, \( \tau_m < 0.3 \mu s, \quad L_m = 1 \text{ to } 0 \text{ cm, the x-ray pulse energy if } 50 \text{ MeV} \cdot \mu \text{s}^{-1}, \text{ the maximum energy of the x-ray quanta is } 23 \text{ keV, } L_m = 30 \text{ to } 80 \mu \text{m, and } D/L < 0.03. \) The overall error in the measurement is governed also by the following factors:

1. The uncertainty in the distance between the electrodes. If the geometrical dimensions are measured with an accuracy of 0.1 mm and \( L = 1 \text{ cm, the error in the drift velocity is } 1\% \).
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5. The error in temperature measurement. The temperature could be measured to an accuracy no worse than \( \pm 0.5 \text{ K and could be stabilized to no worse than } \pm 0.1 \text{ K by using a temperature control system similar to that described earlier.} \)
that described by Voronova et al. The temperature gradient over the chamber height did not exceed 1 K.

The positive space charge which is formed near the cathode by the extremely small ionic (hole) mobility \((-10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})\) and weakens the external field can introduce an appreciable error when working in weak fields. The drift velocity then obtained corresponds to some total field, the value of which is not known, while the mobility \(\mu = V/E\) is less than the true value. In the present work the x-ray pulse produced was 10^5 electron-ion pairs in the chamber. Under these conditions the current pulse observed in fields <100 V cm^-1 was rectangular at an x-ray pulse repetition frequency 0.1 to 1 Hz. Increasing the repetition frequency led to distortion of the current pulse shape (or to a nonlinear voltage pulse front), which indicated field distortions in the gap between the electrodes. For comparison, the radiation-pulse repetition frequency used by Miller et al. was 50 Hz and the charge produced by a pulse was not less than 2 x 10^4 electron-ion pairs.

The total error in a single measurement of the drift velocity at a fixed field value thus did not exceed 3.5%.

EXPERIMENTAL RESULTS

The results of this investigation are shown in Figs. 1 to 4 and in Table I, and the main conclusions are as follows.

1. The doubly-logarithmic plot of drift velocity vs electric field strength has some clearly defined linear sections (Figs. 1 and 2), so that in each section the drift velocity can be well approximated by the expression \(V = BE^\alpha\), where \(B\) and \(\alpha\) are constants for a given temperature. As the field strength increases, \(\alpha\) changes from 1 (\(V = \mu E\)) to 0 (\(V = V_0 = \text{const.}\)).

2. The linear section \(V = \mu E\) was reliably measured and contains 20 to 30 experimental points for each temperature. The mobility \(\mu\) was determined from the value of the drift velocity at the intersection of the extrapolation of the linear section \(V = \mu E\) on the doubly logarithmic plot with the ordinate axis at \(E = 1\) V cm^-1. As noted before, \(\alpha < 1\) the temperature dependence of \(\mu\) (Fig. 3) is very nonmonotonic; the maximum electron mobility (6500 cm^2 V^-1 s^-1) is reached in liquid xenon at 220 K. In solid xenon, \(\mu\) increases monotonically with decreasing temperature from 2900 cm^2 V^-1 s^-1 at \(T = 155\) to 6000 cm^2 V^-1 s^-1 at \(T = 80\) K.

3. The form of the electric field dependence of the electron drift velocity does not change with temperature, except for liquid xenon where there is a characteristic dip at \(E = 10^2\) to 10^3 V cm^-1 (Fig. 1) far from the triple point in the region of the maximum of the \(V(E)\) curve; this dip is absent for gaseous noble gases. The electron drift velocity in liquid argon evidently behaves in a similar way near 145 K.

4. In condensed xenon the drift velocity reaches saturation in fields above 2 to 4 kV cm^-1, and the satura-
FIG. 4. Temperature dependence of drift velocity at saturation: 0—argon, $V_d$; ——argon, $V_0$; ——argon, $V_d$. In solid argon $V_d = 1.8 \times 10^4$ cm$^2$ s$^{-1}$ at $T = 80$ K.

5. The electron drift velocity and mobility in argon and xenon crystals do not depend, within the limits of experimental error, on the rate of crystal growth (i.e., on the crystal quality) in the range from 1 to 10 mm s$^{-1}$.

ELECTRON SCATTERING CROSS SECTIONS IN CONDENSED NOBLE GASES

The behavior of electrons in matter in an external electric field is described by an electron distribution function in momentum, $f(p)$, or in energy, $F(E)$, that satisfies the Boltzmann equation, with its exact form depending on the type of scattering (elastic or inelastic). Calculation of the main parameters of the electron dynamic characteristics is not difficult if the form of the function and the values of the parameters entering into it are known. The transport coefficients, the drift velocity $V$, the mean energy $\overline{E}$, and the electron diffusion coefficient $D$ are given by the expressions

$$V = \frac{eE}{m} \left( \frac{2}{m^2} \right)^{1/2} \left( \frac{1}{\overline{E}} \right) \frac{\partial}{\partial z} F_v(z) dz,$$

$$\overline{E} = \int \frac{1}{\overline{E}} F_v(z) dz,$$

$$D = \int \frac{1}{\overline{E}} \frac{1}{2 \overline{E}} \frac{\partial}{\partial z} F_v(z) dz,$$

where $x = e/kT$, $m$, and $e$ are the electronic mass, charge and energy, $k$ is Boltzmann’s constant, $T$ is the absolute temperature, $E$ is the electric field strength, $\lambda(x)$ is the electron mean free path for momentum transfer, and $F_v(x)$ is the isotropic part of the distribution function.

In 1967 Lekner obtained, by solving the Boltzmann equation for elastic electron scattering, the distribution function in the following form

$$F_v(x) = \exp \left[ - \int \frac{1}{x} \frac{b(x)}{x} dz \right], \quad b(x) = \frac{dE}{d\theta} \frac{\lambda(x)}{\theta} \frac{\lambda(x)}{\theta}(kT)^2,$$

and extended this result to gases, liquids, and solids. In Lekner’s solution, which takes the structure factor $S(k)$ into account ($k$ is the electron wave vector), the mean free paths for energy transfer ($\lambda_\sigma$) and momentum transfer ($\lambda_\lambda$) are different in condensed matter and only become close in value in a gas or for very hot ($c \gg kT$) electrons, when $S(k) \approx 1$.

In a pure noble gas at not too strong fields, the electron dynamics is determined by elastic scattering, since interelectron scattering can be neglected at the electron densities ($\leq 10^{13}$ cm$^{-3}$) typical of drift measurements, so that the dynamic characteristics of an electron in condensed argon and xenon can be calculated from Eqs. (1) to (3). The main difficulty lies then in the evaluation of $\lambda_\sigma$ and $\lambda$. This problem can be solved either by quantum-mechanical calculations or by determining $\lambda_\sigma$ and $\lambda_\lambda$ directly from experimental data.

Methods of calculation of the mean free paths (scattering cross sections) of electrons in a weakly ionized gas are now fairly well developed (see, for example, Ref.19, especially for the case of only elastic scattering). The most accurate analysis method is based on successive approximations in which a power law depen-
The simple theory of elastic scattering of electrons in semiconductors leads to a differential cross section independent of electron energy.\(^\text{11}\) This is, evidently, also valid for the liquid state if the liquid is considered as a system with short-range order, having a corresponding phonon spectrum. Lekker's calculations,\(^\text{12}\) using an effective scattering potential, show that \(\lambda_0\) in liquid argon depends extremely weakly on electron energy. Similar results have been obtained\(^\text{10}\) for liquid argon, krypton, and xenon. In condensed noble gases, \(\lambda_0\) is thus independent of energy at a given temperature.

Since the Maxwell distribution function is valid for weak electric fields, such that \(V=\mu_0E\), while Lekker's theory gives \(\lambda_0=\lambda_0^0(0)\) for hot electrons, we have from Eq. (1)

\[
\lambda_0 = \frac{4}{\lambda_0^0} - \frac{3}{2} \left( \frac{nkT}{2} \right)^{\frac{3}{2}} \frac{\mu_0^2}{c^2} S(0),
\]

where \(S(0)=NkT \chi,\) where \(\chi\) is the isothermal compressibility.

For convenience, the masses and \(kT\) are expressed in eV in all expressions here and further on; \(c\) is the velocity of light.

The electron scattering cross section relative to energy transfer, \(\sigma_0\), calculated from Eq. (4) with the substitution of \(\lambda_0\) as measured by us, is that linear in argon and xenon a sharp minimum at temperatures correspond to the peak in the mobility (Fig. 5). As the temperature increases, \(\sigma_0\) tends to the value calculated for the gas at the critical point. On the left of \(T_0\), \(\sigma_0\) increases as the temperature decreases both in the liquid and in the crystal. Lekker\(^\text{13-20}\) explained the non-monotonic behavior of \(\sigma_0\) and \(\mu_0\), showing that the effective electron scattering length \(\lambda\), the sign of which depends on the nature of the scattering potential, is negative in a gas but positive in a liquid near the triple point. There must, therefore, exist a temperature region in the interval \(T_0\) to \(T_c\) where \(\sigma_0=4\pi\alpha^2\) has a minimum while \(\mu_0\) is a maximum. Unrestricted growth of the mobility is prevented by the scattering of the electrons by thermodynamic fluctuations of the electron-atom potential\(^\text{20}\) or by its tails, which do not completely overlap.\(^\text{21}\) A liquid is evidently a system with short-range order, i.e., a quasi-crystal at \(T<T_0\) and going over gradually at \(T>T_0\) into a system of individual scattering centers. The problem of the mobility peak is still not completely resolved and continues to attract interest. An attempt has been made,\(^\text{22}\) for example, to attribute this phenomenon to the existence of percolation channels, which are regions of high mobility that thread through the liquid at \(T=T_0\).

Having determined \(\lambda_0\), we pass to the calculation of \(\lambda_1\), which now depends on energy. Suppose that for some range of electron energy \(\lambda_1\) is approximated by the power series \(\lambda_1(x)=ax^n\). Substituting this expression in Eq. (3) and neglecting the thermal term in \(P(x)\), i.e., considering the field region where \(c \gg kT\), we obtain from Eq. (1)

\[
V=(4ak/3\lambda_0^0)^{\frac{c}{2}} (2-\frac{n}{2})^{\frac{n}{2}} \Gamma\left(\frac{3}{2-n}\right) \left\{ \frac{3}{2-n} \right\} \Gamma\left(\frac{3}{2(2-n)}\right),
\]

where \(\lambda_0=b(x)/x^n\), and \(\Gamma\) is the gamma-function.

If the experimental dependence of the drift velocity on the electric field, for a certain range of \(E\), is approximated by the expression \(V=BE_n\), then equating the latter with Eq. (7) we obtain the relations connecting the experimental values of \(B\) and \(a\) with the approximation parameters \(\lambda_1=ax^n\), in particular

\[
n=(2n-1)/(n+1).
\]

Since \(a=0\) in the saturation region, we have \(n=-1\) and the effect of saturation of the drift velocity in condensed gases is simply explained by the hyperbolic dependence of mean free path for momentum transfer on electron energy. The scattering cross section increases linearly with energy. For the gaseous state, when \(b=1\), \(n=1\) \(-3\). The drift velocity of electrons in a gas cannot, therefore, reach saturation in a noticeable range of electric field change, as confirmed by experimental results.\(^\text{6}\) This method of calculation, first applied to condensed noble gases, gives thus immediately good qualitative results, explaining in a simple way the saturation of the drift velocity.

In general, analysis methods based on a power-law approximation of the scattering cross section are not considered very suitable in the case of a strong dependence of cross section on electron energy, for example near a Ramsauer-Townsend minimum in heavy
monatomic gases such as argon, krypton and xenon. As can be seen from Fig. 6, good agreement is achieved only to the right of the Ramsauer minimum. However, if the ultimate aim of the calculations is the determination of not so much the scattering cross section as other features, such as the characteristic or mean energy, the agreement between calculation and experiment is appreciably better.

In condensed argon and xenon $\alpha$ ranges from 1 to -0.15, so that $n<0$ and the cross section is an increasing function of electron energy if $e\gg kT$. On the other hand, it was pointed out earlier that either the Ramsauer minimum is completely absent in condensed noble gases\(^{20}\) or that it exists, at least in liquid argon at low energies $\sim 2 \times 10^{-4}$ eV, but is shallow.\(^{24}\) At first sight, the analysis method considered cannot give information about the behavior of the cross section at such energies. However, energy considerations lead to the conclusion that the Ramsauer minimum does exist in the condensed phase. If we equate the electron energy loss in a single elastic scattering event

$$\frac{(2m/M)\varepsilon F_\alpha(x)}{e^\alpha}$$

to the gain in energy from the external electric field between collisions

$$\lambda e^\alpha \frac{V}{(2e/m)^{\alpha} F_\alpha(x)} e^\alpha d\varepsilon$$

and carry out an integration for the Maxwell distribution function, an expression can be obtained for the electric field strength $E_0$, for which the relation $V=\mu_0 E$ is preserved:

$$E_0=\frac{e}{\mu_0} \left[ \frac{2kT}{MS(0)} \right]^{\frac{1}{\alpha}}$$

(7)

The values of $E_0$ calculated from Eq. (7) are appreciably lower than the experimental values (Fig. 7). At the same time, thermodynamic equilibrium is not
determined, as shown above, from experimental results
for only a single transport coefficient (the drift velocity
in the present case), a comparison of the calculated and
experimental values of any other quantity, for example
the characteristic energy, also provides a valuable
check on the correctness of the analysis method and
on the calculated cross section.

MEAN ENERGY AND ELECTRON DIFFUSION
COEFFICIENT

After the electron diffusion function has been
determined and the connection between its parameters and
the experimental values of B and α established, there
is no difficulty in calculating the mean energy and
the diffusion coefficient for electrons. The results of the
calculations are shown in Fig. 8 and in Table II. Over
the temperature range studied, the mean electron energy
in the liquid increases monotonically with increasing
temperature in line with the reduction in σ and α.
In crystalline xenon the mean energy is practically
independent of temperature in the range 80 to 160 K.
At the liquid-crystal transition, α in argon increases while
in xenon it decreases slightly.

Only the characteristic energy D/μ in liquid argon
in the range of fields 2 to 10 kV cm\(^{-1}\) has been
determined experimentally.\(^\text{21}\) We used the Townsend
relation in the form

\[ D/\mu = \varepsilon \]

to compare \( \varepsilon \) and \( D/\mu \), where \( F \) is a dimensionless coefficient.
In weak fields, obviously, \( F = 1 \). If \( \varepsilon \gg kT \),
in general \( F \) depends on \( \varepsilon \). Calculation gives \( F = 0.62 \) in
liquid argon for \( E = 10^8 \) V cm\(^{-1}\), so that \( \varepsilon = 1.2 D/\mu \).
Taking this into account, the agreement between the
experimental values of \( \varepsilon \) and those calculated by us is very
good.

Finally, if we consider how the accuracy in the mea-

\[ T = (\frac{V_{\mu E}}{\varepsilon})^{\frac{1}{2}} \]

\[ D = (\frac{V_{\mu E}}{\varepsilon})^{\frac{1}{2}} \]

that every percent error in measuring \( \alpha, V, E, \mu, \) and
\( T \) introduces an error of several percent in \( \varepsilon \) and especially
in \( D \).

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