THEORY OF SLOW ELECTRONS IN LIQUIDS

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The authors investigate the mobilities of electrons in polar liquids and indicate possible mechanisms for conductivity. They find the structure of the energy spectrum of electrons in a nonpolar liquid. They present qualitative considerations on the temperature dependence of the mobility for the latter case.

Much attention has recently been paid in the literature to the problem of slow electrons in liquids, particularly in connection with problems in radiation chemistry [1] and biology [2]. Papers are now appearing in which it is shown that solvated electrons may take part in kinetics. Particular interest naturally attaches to the case when the liquid is water. In the first part of this paper we shall therefore consider polar liquids. Here we shall not be trying to derive any new formulas, but shall limit ourselves to qualitative consideration of the theory of electron mobility in polar liquids. In the second part we shall consider nonpolar liquids. In contrast to the case of polar liquids, there is at present no satisfactory theory for nonpolar liquids: we shall therefore examine the latter in more detail.

The theory outlined below does not apply only to normal liquid dielectrics and semiconductors; the physical ideas and mathematical apparatus which are developed below can also be applied to solid organic semiconductors.

Mobilities of Electrons in Polar Liquids

In order to give a clearer exposition of the physical mechanism underlying electrical conductivity in liquids, we shall first give some qualitative results of the theory of electron mobility in polar crystals, obtained in the last five years. We shall then show how these results can be generalized to the case of liquids.

There is an extensive class of semiconductors whose properties cannot be represented by conventional zone theory. They are characterized by an activation-type dependence of mobility on temperature, $u \sim \exp(-E_a/kT)$. This phenomenon cannot be explained in terms of ordinary zone theory, since thermal motion of the crystal lattice increases the probability of scattering of the current carriers, thus reducing the mobility. Increase of mobility with temperature is found in substances with very small mobilities. For example, for Fe_2O_3 and NiO the mobility $u \approx 4 \cdot 10^{-3}$ cm^2/V · sec, and the free path of the current carriers (in conventional zone theory) is much smaller than the lattice constant. To explain the anomalous temperature dependence of the mobility, Ioffe [3] suggested that in these semiconductors the transport of charge should be regarded as "jump-over" of electrons from one nucleus to the next. In a polar crystal, "diffusion" transfer of electrons will require energy to repolarize the crystal lattice. Calculations [4-6] show that in this case the mobility will actually have an activation-type temperature dependence. A difficulty, however, is that the zone solutions of the problem depend on the translation symmetry of the crystal, which is unbroken in these semiconductors. Several authors [4-10] have attempted to settle the question of how the conductivity mechanism changes, i.e., how to make the transition from zonal (delocalized) electrons to localized electrons. These authors showed that at low enough temperature the mobility begins to increase exponentially: in a certain sense, this corresponds to the transition from the zonal mechanism to the jump-over mechanism.

The picture outlined above is valid only for the case when the electron interacts strongly only with the particular atom in which it finds itself at any given moment—in other words, when the exchange integral for a pair of neighboring atoms is sufficiently small. For this reason the current carriers in such systems are usually called small polarons.

In the case when the electron interacts weakly with each separate atom of the crystal, but interacts strongly with the polarization, the valid theory is that of the large-radius polaron, developed by Pekar [11].

At first sight, it might seem that the physical picture arising in a polar liquid is much more complex than that described above. However, we shall show that in many cases there is a complete analogy between a crystal and a liquid. Let us in fact consider all the possible cases which might arise in a polar liquid:

1. When there is little interaction between the electron and the atoms of the liquid. In this case the mobility of the electrons is due to the large-radius polaron mechanism. As shown by Pekar [11], the radius of a large polaron is considerably greater than the lattice period, and thus also greater than the mean distance between neighboring molecules of the liquid. We can therefore use a continuous model for the liquid, and we find no difference from the normal Pekar theory for a crystal. In fact this approach was already used in [12-13] for metal-ammonia solutions.

2. When the electron interacts strongly with the atoms of the liquid. In this case it is natural to use the small-radius polaron theory. However, the results of this theory can be applied directly only at low enough temperatures – i.e., when electrons are transferred by a "diffusion" mechanism (by means of jumping-over). In the next two sections we shall consider the zone mechanism of the small-radius polaron.

According to [4, 6], the mobility of an electron is related to the diffusion coefficient by the Einstein relation $u = eD/kT$. Here the diffusion coefficient is given by $D = a^2/\tau$, where a is the mean distance between molecules of the liquid and τ^{-1} is the probability of electron jump-over per unit time.

$$\tau^{-1} = \varkappa \cdot \omega_0 / 2\pi \cdot \exp\left(-E_s / 4kT\right). \tag{1}$$

Here ω_0 is the frequency of the liquid's optical vibrations with which the electron is interacting (e.g., for water the vibration frequency is $\sim 10^{11} \cdot \mathrm{sec}^{-1}$). For adiabatic transitions, the transmission coefficient $\varkappa$ is equal to unity, while for nonadiabatic transitions

$$\varkappa = |V|^2 \cdot \left(\frac{4\pi^3}{\hbar^2 \omega_0^2 kTE_s}\right)^{1/2}, \tag{2}$$

where V is the volume integral and E_s the energy of "repolarization" of the liquid due to electron jump-over.

3. Zonal polaron of small radius. If the temperature is such that the mobility of the polaron is due to a zone mechanism, then generally speaking we cannot apply the results found for a crystal. However, we can consider the case when the correlation length between molecules of the liquid (i.e., the short-range order distance l) is greater than the free-path length of the zonal polaron λ. In this particular case, we can expect that the electron mobility will be quite accurately given by the same formula as for a crystal.

4. Zonal polaron of small radius; $\lambda > l$. This case is more complex, and there is as yet no theory which can give an expression for the electron mobility. When $\lambda > l$ we can physically expect that the zonal polaron will be scattered by inhomogeneities in the liquid. This leads to the result that the effective free-path length of the polaron will be determined by the correlation length l.

The ideas advanced in sections 3 and 4 above are purely qualitative, and the corresponding mechanisms await strict theoretical treatment.

The Energy Spectrum Structure for Electrons in Nonpolar Liquids

In the last five to seven years there have been a number of theoretical papers [14-21] on the structure of the energy spectrum of electrons in various unordered systems. These investigations have followed two paths: some authors have studied solids (metals, semiconductors, alloys, etc.) containing impurity centers in unordered arrangement [19, 21]; other authors have studied liquids [15-18]. These investigators have calculated the density of electron energy levels, averaged by means of the distribution function of the random variates R_i – i.e., the coordinates of the impurity atoms or the coordinates of the atoms in the liquid.

It is easy to see that this approach corresponds in fact to the zero adiabatic approximation when the kinetic energy of the atoms in the liquid is negligible. As it is liquids which we are interested in, we shall not further consider the trivial case of a solid with impurities. The zero adiabatic approximation will lead to specially important limitations if we consider various kinetic effects in which electrons take part. For example, we do not have to consider processes involving nonadiabatic jump-over of an electron from one energy level to another. Furthermore, in the case of an adiabatic transition we cannot apply the Landau-Zener theory, because the kinetic energy of the atoms figures explicitly in it. A simple analysis shows that even in the zero adiabatic approximation there arises an additional limitation due to the fact that the distribution function $\Phi(R_i)$ should not depend on the electron state. Physically this means that, although the electron spectrum depends markedly on the coordinates of the atoms in the liquid, we can neglect the reverse action of the electron on the spatial distribution of the atoms.

In this paper we attempt to study the qualitative aspect of the energy spectrum structure of an electron in a nonpolar liquid and to devise an adequate mathematical approach. We shall therefore base our model on a number of assumptions. We shall consider a one-dimensional model of a liquid [22] with an extra electron which interacts strongly only with one atom of the liquid (the approximation of strongly bound electrons). The Hamiltonian of this system, in its double quantized form, is

$$H = \sum_k \varepsilon_0 a_k^+ a_k + \sum_k J_{k,k+1}(a_k^+ a_{k+1} + a_{k+1}^+ a_k), \tag{3}$$

where k is the number of the atom ($k = 0, \pm 1, \ldots, \pm N$), ε_0 is the energy of the electron in an isolated atom of the liquid, $J_{k,k+1}$ is the exchange integral for neighboring atoms k and k + 1, and a_k^+ and a_k are as usual the operators for formation and annihilation of an electron from the k-th atom. In this Hamiltonian, the $J_{k,k+1}$ are functions of the $R_{k+1} - R_k$ and will be regarded as random. Formula (3) can also be written in the momentum form, for which we make a unitary transformation by means of the matrix

$$S_{pk} = (2N+1)^{-1/2} e^{ipk}$$

$$H = \sum_p \varepsilon_0 a_p^+ a_p + \sum_{p,p'} I_{p,p'} a_p^+ a_{p'}. \tag{4}$$

Here

$$\varepsilon_0(p) = \varepsilon_0 + 2\langle J\rangle \cos p; \tag{5}$$

$$I_{pp'} = \frac{1}{2N+1} \sum_k I_{k,k+1} e^{i(p-p')k}(e^{ip} + e^{-ip'}), \tag{6}$$

while $I_{k,k+1} = J_{k,k+1} - \langle J\rangle$ is the deviation of the exchange integral from its mean. From the cyclicity conditions we find the possible values $p = 2\pi n/(2N+1)$, where $n = 0, \pm 1, \ldots, \pm N$.

To determine the electron spectrum we can use Green's function. In the one-electron approximation Green's function can conveniently be written as

$$G_{k,k'}(t-t') = -i\theta(t-t')\langle [a_k(t);\ a_{k'}^+(t')]\rangle_0, \tag{7}$$

where

$$\theta(t) = \begin{cases} 1, & t > 0 \\ 0, & t < 0 \end{cases}$$

while $\langle \ldots\ldots \rangle_0$ indicates quantum-mechanical averaging in vacuum.

In matrix form, the equation for Green's function is

$$G = G^0 + G^0 I G, \tag{8}$$

where $G^0_{p,p'} = \delta_{pp'}/[E - \varepsilon_0(p)]$. From (8) we can get a formula for $\langle G\rangle$ (here the pointed brackets indicate averaging over random R_i):

$$\langle G\rangle = \left\langle \frac{1}{1-G^0 I}\right\rangle \cdot G^0 \quad \text{or} \quad \frac{1}{\left\langle \frac{1}{1-G^0 I}\right\rangle}\langle G\rangle = G^0 .$$

Twice applying the formula for a geometrical progression to this last expression, we get

$$\langle G\rangle = G^0 + G^0 M \langle G\rangle , \tag{9}$$

where the effective potential or mass operator M is found from the formula:

$$M = (G^0)^{-1}\sum_{s=1}^{\infty}(-1)^{s+1}\left\{\sum_{r=1}^{\infty}\langle (G^0 I)^r\rangle\right\}^s . \tag{10}$$

In view of the diagonality of $< G >$ in the momentum form, from (9) we get directly that

$$\langle G\rangle = \frac{1}{E-\varepsilon_0(p)-M(p,E)} . \tag{11}$$

If the mean Green function has a simple pole at the point $E = \varepsilon(p) - i\gamma(p)$, which is a solution of

$$E(p) = \varepsilon_0(p) + M(p,E) , \tag{12}$$

then we can show that if $\gamma(p) \ll \varepsilon(p)$, the density of levels

$$D(E) = \left| \frac{1}{\pi}\operatorname{Im}\operatorname{Sp}\langle G(E)\rangle \right| \tag{13}$$

is related to $\varepsilon(p)$ by the equation

$$D(E) \cong dp/d\varepsilon(p) . \tag{14}$$

In this sense, we shall call $\varepsilon(p)$ the mean spectrum. In addition, we can show that $\gamma(p)$ has the meaning of the reciprocal of the decay time of state $\varepsilon(p)$ (on the usual condition of the theory of perturbations, $\gamma(p) \cdot t \ll 1$).

To calculate M(p, E) we have to make some kind of assumption about the distribution function $\Phi(R_i)$. In what follows we shall assume that correlation exists only between two neighboring atoms, i.e., that all the exchange integrals are noncorrelated. As regards the pair-wise correlative function $\Phi(R)$, we shall for the present assume only that it is bell-shaped with some characteristic width determined by l. Assuming also that $J(R) \sim e^{-R/R_0}$, we easily verify that

$$\frac{\langle I\rangle^n}{\langle J\rangle^n} \sim \left(\frac{l}{R_0}\right)^n = \mu^n ; \qquad n = 2k . \tag{15}$$

We shall now examine the two limiting cases when $1 \ll R_0$ and when $1 \gg R_0$. In the first case, in particular when $n = 2$, we have $< I^2 > \ll < J >^2$, and therefore for brevity we shall call the assumption $1 \ll R_0$ the low dispersion approximation.

a) Low dispersion. We shall first of all attempt to develop the ordinary theory of perturbations, assuming that operator I is small and confining ourselves to the first nonzero term in the mass operator

$$M^{(2)}(p,E) = 2\langle I^2\rangle \left\{ \frac{1}{(E-\varepsilon_0)\sqrt{1-\frac{4\langle J\rangle^2}{(E-\varepsilon_0)^2}}} + \frac{1}{2\langle J\rangle}\left(\frac{1}{\sqrt{1-\frac{4\langle J\rangle^2}{(E-\varepsilon_0)^2}}} - 1 \right)\cos p \right\} . \tag{16}$$

This expression must be substituted into (12). We then get

$$\varepsilon(p) \approx \varepsilon_0(p) + \operatorname{Re} M^{(2)}(p, \varepsilon_0(p)) = \varepsilon_0 + 2\mu |\langle J\rangle| (1 - \mu^2/2) \cos p \tag{17}$$

$$\gamma(p) \approx |\operatorname{Im} M^{(2)}(p, \varepsilon_0(p))| = \mu^2 \left| \frac{\langle J\rangle}{\sin p} \right| (1 + \cos^2 p). \tag{18}$$

The criterion of applicability of ordinary perturbation theory is that the damping must be small by comparison with the perturbation energy, i.e.,

$$\gamma(p) \ll |\varepsilon(p) - \varepsilon(0)| \approx |\langle J\rangle| p^2 \quad \text{or} \quad p \gg \mu^{2/3}. \tag{19}$$

This means that the standard perturbation theory is valid only at sufficiently great distances from the edges of a zone [14]. To get a solution near the edge of a zone ($p \ll \mu^{2/3}$), we shall construct an improved theory of perturbations. Expressions (17) and (18) were derived by neglecting in M(p, E) the terms of higher order than $< I^2 >$. In addition, (12) was solved approximately, retaining only terms of order μ^2. An improvement is made by finding an exact asymptotic solution of (12) near the edge of a zone. After this, the expression found for E(p) will be substituted into (10), and it will be shown that the remaining term $M^{(2)}(p, E)$ in this energy range is actually greater than the discarded terms. Let us examine, for example, the upper edge of the zone, and make the change of variables $E = \varepsilon_0 - 2 < J > \cdot (1+u)$, where the new variable $0 < u \ll 1$. Some simple calculations lead to the following expression for the spectrum:

$$E(p) = \varepsilon_0 + 2\langle J\rangle \left[1 + 2^{-1/3} \cdot \mu^{2/3} - \frac{(p')^2}{3}\right]; \quad p' = \pi - p; \quad p' \ll \mu^{2/3}, \tag{20}$$

To estimate the subsequent terms in the expansion of the mass operator, we shall here use an intuitive but fairly rigorous method; a rigorous proof confirms the result which we obtain. It is easy to show that, if $u \to 0$ for $G^0_{k,k'}$, the following asymptotic approximation is valid:

$$|G^0_{k,k'}| \sim \frac{1}{2|\langle J\rangle| \cdot \sqrt{2u}}. \tag{21}$$

We can thus write (10) as

$$M = \sum_{n=2} A_n \cdot \frac{\langle I^n\rangle}{\langle J\rangle^{n-1}} \cdot u^{(1-n)/2} \sim \mu^{2/3} \cdot \langle J\rangle \sum_{n=2} A_n (\mu^{1/3})^n, \tag{22}$$

where the A_n are numerical coefficients. From this expression it is seen that near the edge of a zone the mass operator takes the form of a power series in the small parameter $\mu^{1/3}$, which verifies our assumption.

From (20) there follow a number of important physical results. First of all, it is clear that the presence of dispersion leads to broadening of the former zone by $(2\mu)^{2/3}$. In addition, the effective mass calculated near the new zone edge differs from the effective mass found by the zero approximation [i.e., from (5)] by the factor $^3/_2$

$$m^* = \frac{3}{2} m_0^*. \tag{23}$$

The presence of the factor $^3/_2$ in this formula is due to the nonanalytic relation between E(p) and the small parameter μ [$E(p) \sim \mu^{2/3}$].

The above result can be compared with the result of Edwards and Beeby [16] who gave a solution to exactly the same problem. Their basic assumption was that at low dispersions the spectrum for a specific realization of the coordinates R_i must be of the form

$$E(p, R_i) = \varepsilon_0 + 2\sum_k J(R_k - R_{k-1})\cos p(R_k - R_{k-1}). \tag{24}$$

The mean spectrum can be obtained from (24) by averaging the second term over interatomic distances, $d_k = R_k - R_{k-1}$. It is easy to see that this treatment does not lead to appreciable alteration of the spectrum near the edge of a zone. In fact, by (24), for small enough μ the spectrum is exactly the same as the former one [see (5)], while by (20), for any small but finite μ the spectrum must take a different form owing to the nonanalytical relation between E(p) and μ.

Dyson [18] solved the problem of determining the phonon spectrum in a unidimensional chain of atoms with random elasticity coefficients. Although our problem differs from his physically, there is a certain mathematical similarity between them. Dyson solved his problem with asymptotic accuracy for small dispersions, assuming that the elasticity constants K were distributed according to a Gaussian law. Owing to the contribution made by large values of K, the phonon spectrum was diffuse, i.e., had no sharp boundaries. This discrepancy with out results is due to the fact that our exchange integral varied within finite limits. This assumption was also made in [16] and is physically justified. In fact, if we allow J to take large values, we shall violate the approximation of strongly bound electrons, and the whole solution will be invalid.

b) High dispersion. This case is specially interesting, because it is the most practical case for the liquids which we are interested in.

As was done in deriving (15), we can show that, for $l \gg R_c$,

$$\frac{\langle J\rangle^n}{\langle J^n\rangle} \sim \left(\frac{R_0}{l}\right)^{n-1} = \left(\frac{1}{\mu}\right)^{n-1} \ll 1. \tag{25}$$

We can easily verify this, e.g., by noting that $J(R) \sim l^{-R/R_0}$ and

$$\Phi(R) = \begin{cases} 0 |R-a| > l, \\ \frac{1}{2l}, \ |R-a| < l, \end{cases}$$

where a is the mean distance between atoms of the liquid.

From (25) it follows that we can regard the averaging symbol $<\ldots>$ as effectively a small parameter. In fact,

$$\frac{\langle J^k\rangle \cdot \langle J^{n-k}\rangle}{\langle J^n\rangle} = \frac{\langle J\rangle^n}{\langle J^n\rangle} \cdot \frac{\langle J^{n-k}\rangle}{\langle J\rangle^{n-k}} \sim \mu^{-k} \ll 1. \tag{26}$$

It is now easy to carry out partial summation in the mass operator, leaving in each term proportional to J^n the oldest with respect to the parameter $\mu^{-1} \ll 1$. However, in (10) I must be replaced by J, and G^0 correspondingly replaced by $\tilde{G}^0 = 1/(E - \varepsilon_0)$. As a result, we get the following expression for the mass operator:

$$M(p, E) \approx 2(E-\varepsilon_0)\left[\sum_{r=1}\left\langle\left(\frac{J}{E-\varepsilon_0}\right)^{2r}\right\rangle + \sum_{r=0}\left\langle\left(\frac{J}{E-\varepsilon_0}\right)^{2r+1}\right\rangle\cos p\right]$$

$$= 2\tilde{G}^0\left\langle\frac{J^2}{1-(\tilde{G}^0 J)^2}\right\rangle + 2\left\langle\frac{J}{1-(\tilde{G}^0 J)^2}\right\rangle\cos p. \tag{27}$$

By substituting this value into (12) we can in principle find the electron spectrum. However, to accomplish this procedure in practice, we need to give a specific form to $\Phi(R)$, since E in (27) comes inside the sign $<\ldots>$. To get qualitative results, it is reasonable to use a rectangular distribution for $\Phi(R)$ as shown above. As a result, we get the following formula for the spectrum:

$$\left.\begin{aligned} E_{\text{в}}(p) &= \varepsilon_0 + |J_m| \left(1 \pm \exp\left[-\frac{R_0}{2l(1-\cos p)}\right]\right) \\ E_{\text{н}}(p) &= \varepsilon_0 - |J_m| \left(1 \pm \exp\left[-\frac{R_0}{2l(1+\cos p)}\right]\right) \end{aligned}\right\} \qquad (28)$$

where $E_B(p)$ and $E_H(p)$ refer to the upper and lower zones (see continuous curve in Fig. 1), while J_m denotes the maximum value of the exchange integral corresponding to the minimum interatomic distance $R_{min} = a - l$.

By (28), the distance between the centers of the lower and upper zones is equal to $2\,|J_m|$, while the width of each zone $E_B(p)$ or $E_H(p)$ is equal to $2|J(a)|$, i.e., to the exchange integral for the mean distance a. To clarify the physical meaning of this result, let us compare (28) with the spectrum for a periodic chain as shown in Fig. 2. This problem can be solved exactly: it turns out that the spectrum takes the following form (dashed curves in Fig. 1)

$$\left.\begin{aligned} E_{\text{в}}{}^0(p) &= \varepsilon_0 + \sqrt{(J_1+J_0)^2 - 4J_1\cdot J_0\cos^2 p} \\ E_{\text{н}}(p) &= \varepsilon_0 - \sqrt{(J_1+J_0)^2 - 4J_1\cdot J_0\cos^2 p} \end{aligned}\right\} \qquad (29)$$

Here J_1 corresponds to the exchange integral for particles 1 and 2, and J_0 to that for particles 2 and 3 (see Fig. 2). The distance between the centers of zones $E_B^0(p)$ and E_H^0 is equal to $2(J^2+J_0^2)^{1/2} \approx 2\,|J_1|$, since $|J_1| >> |J_0|$, while the breadth of each zone is $2|J_0|$. Comparison of (28) and (29) shows that the mean spectrum of an electron in a liquid at high dispersions receives its main contribution from configurations of atoms such that two atoms in the liquid are as close together as possible. This picture suggests that such atomic configurations will also be important in calculations of mobility; this in a sense corresponds to electron jump-over between neighboring atoms. It is easily seen that the temperature dependence of the mobility has a qualitative correspondence with the case of a small-radius polaron, discussed in the first paragraph. In fact, by (18) the dispersion increases with temperature at low temperatures, whereas τ_p decreases; consequently the mobility $u \sim e\tau_p/m^*$ also decreases. On the other hand, at high dispersions a rise in temperature should apparently lead to an increase in jump-over, i.e., a rise in mobility.

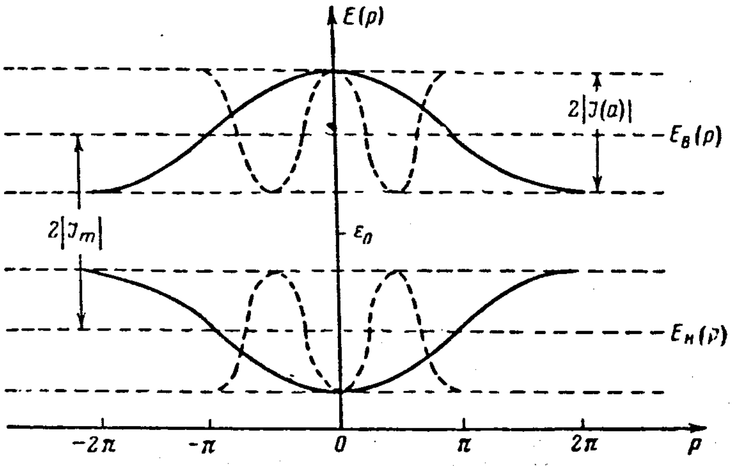

Fig. 1.

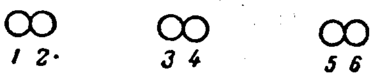

Fig. 2.

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