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Hole transport in the rare-gas solids Ne, Ar, Kr, and Xe

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This paper reports an investigation of the drift mobility of excess holes in solid Ne, Ar, Kr, and Xe. Thin-crystal specimens (50–500 μm thick) were grown from the liquid between parallel-plate electrodes in a chamber attached to a miniature cryostat after purification of the starting gas. As in previous work on the electron transport in rare-gas solids and liquids, an electron-beam technique was used to generate excess carriers near one of the electrodes. Holes were extracted by the applied field and their transit time was measured directly, leading to the drift mobility μ_h. Close to the triple points, μ_h values in the above crystals lie between 1×10^{-2} and 4×10^{-2} cm^2 V^{-1} sec^{-1}, several orders of magnitude lower than the corresponding electron mobilities. The form of the temperature dependence of μ_h changes progressively from Xe ($\mu_h \propto T^{-1.6}$) to an essentially activated mobility in Ar and Ne. The experimental results have been analyzed in terms of small-polaron theory, using both the adiabatic and nonadiabatic approximations. The theory can account for the different forms of the temperature dependence and possible ranges of values for the predominant phonon energy, the polaron binding energy and the transfer energy for holes have been deduced in each case. These quantitites, characterizing the hole hopping transport, vary systematically from Xe to Ne and their correlation is discussed in some detail.

I. INTRODUCTION

The transport of excess electrons in the rare-gas solids has been studied in some detail during recent years.[1–5] In solid Ne, Ar, Kr, and Xe the electrons move in a wide conduction band with mobilities approaching 4000 cm^2 V^{-1} sec^{-1}, remarkably high in comparison with other molecular solids. Values of the electron mobility μ_e near the triple points are given in Table I, which also includes similar data for the corresponding liquids. Although fairly detailed information on the properties of electrons is available, surprisingly little is known about the hole transport in these solids. The only available drift-mobility results are for solid Xe,[2,3,6] and they suggest that hole transport differs fundamentally from that of the electrons by the pronounced localization of the excess holes. It appeared therefore of some interest to carry out a systematic investigation of hole mobilities and their temperature dependence in the rare-gas solids. In this paper we report the results, attempt an interpretation of the data based on small-polaron theory, and finally discuss the parameters deduced from the theory.

II. EXPERIMENTAL METHOD

The experimental approach was essentially similar to that used in our previous work[3,6,7] and only a brief summary will be given here. The crystals were grown from the melt using ultrapure gases[8] as the starting material. These were passed through a carefully outgassed charcoal trap or a molecular sieve (Oxysorb) to decrease the oxygen content still further. The sample was grown between two conducting electrodes in the form of a thin platelet of thickness d. A detailed description of the specimen chamber used has been given in previously published work on the diatomic solids.[9] An important feature of the design is the flexible top electrode which is essential to avoid cracking of the sample at temperatures below the triple point. Although this arrangement minimized the effect, the temperature range that could be covered in each material was ultimately limited by crystal fracture due to thermal strain.

A fast electron pulse of high-energy electrons (45 keV) generated excess carriers near one surface of the specimen. The holes were extracted from the generation region by a synchronized field pulse of magnitude E applied to the crystal and their transit time t_t across the specimen was measured directly on a Tektronix 7704 oscilloscope. The drift velocity of the holes, $v = d/t_t$, was measured as a function of the applied field and was found to be proportional to E up to the highest fields used (50 kV cm^{-1}). From these data a value for the drift mobility, $\mu_h = v/E$, was deduced at each temperature.

In order to obtain a well-defined extraction signal it is necessary that the holes complete a transit of the specimen in a time that is short compared to their lifetime with respect to deep traps. In Xe and Ne, this was possible even with specimens a few hundreds of microns thick, but in similar Ar and Kr platelets no clear signals were observable due to lifetime limitation. However, by restricting crystal thicknesses to 100 μm or less, it became possible to obtain meaningful results for the hole mobility and its temperature dependence in Ne, Ar, Kr, and Xe.

TABLE I. Mobility of electrons and holes in the rare-gas solids and of the electrons in the liquids at temperatures close to the triple-point temperatures.

Material	Triple point (K)	Drift mobility ($cm^2 V^{-1} sec^{-1}$) Electrons (Solid)	 (Liquid)	 Holes (Solid)
Ne	25	600 a	1.6×10^{-3} a	1.05×10^{-2}
Ar	84	1000 b	475 b	2.3×10^{-2}
Kr	116	3600 b	1800 b	4×10^{-2}
Xe	161	4000 b	1900 b	1.8×10^{-2} c

[a] From Ref. 5.

[b] From Ref. 3. Where the mobilities differ slightly from those published previously, recent work in our laboratory has enabled more accurate determinations which are given above.

[c] From Ref. 6.

Polarization effects, similar to those observed in electron transport, were also found in the present experiments. They cause a gradual decrease in the amplitude of successive hole transit signals and are associated with the buildup of internal space charge, probably in the contact region. To keep these effects within manageable limits the electric field was applied in the form of a pulse, a few milliseconds long, which was triggered just before the excitation pulse. In addition, the repetition frequency was reduced to 2 Hz or less. Under these conditions the signals appeared to be independent of polarization effects, at least in Ne, Ar, and Xe.

Experiments on Kr crystals were affected far more severely by polarization effects and by lifetime limitations than the measurements on the other solids. It is possible that this difficulty arises from the fact that the commercially available Kr gas used for growing the specimens had a relatively large impurity content, at least a factor 10 higher than in the other starting gases.[8] Reasonably consistent data could only be obtained for Kr specimens with single-shot operation and the use of a fast-storage oscilloscope (Tektronix 7623) for recording the signals. As a result the mobility values are less certain than those for the other rare gas solids, as is evident from the scatter of the experimental points for different Kr specimens in Fig. 1.

The accuracy of our mobility data depends critically on the knowledge of the sample thickness d, since $\mu_h \propto d^2$. One disadvantage of working with flexible spacings of about 100 μm is that the experimental error in d, and therefore in μ_h, becomes relatively large. To determine d in each run, a number of independent measurements were made. First, the transit times of the positive ions in the liquid were measured as a function of field and a value of d was calculated from the predetermined ionic mobility. The latter had been measured in separate runs using rigid spacings of up to 500 μm and was therefore known fairly accurately ($\pm 10\%$). As a check on this we also measured the transit time of the electrons at high fields where the drift velocity saturates. Using the known saturation velocity[2,3] it was thus possible to determine a second value of d in the liquid phase. Unfortunately, the flexible electrodes generally tended to move slightly as the liquid solidified so that the determined value of d could only be relied upon to about ± 20 μm in the solid phase. We, therefore, measured the transit time of the electrons in the solid at high fields and calculated another value of d from the known saturation velocity in the solid.[2,3] With our very thin spacings, however, these transit times were rather short and the measurements were subject to a relatively large error. In view of these uncertainties in d we estimate that our absolute values of mobility could be in error by up to 50% in Ne, Ar, and Xe, and by up to a factor of 2 in Kr. The relative errors in a particular temperature run were probably less than 20% for the Kr data and 10% for the remainder.

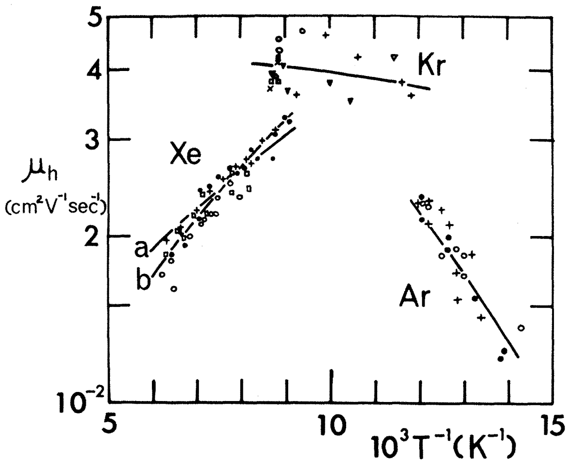

FIG. 1. Semilogarithmic plot of the hole drift mobility in solid Xe, Kr, and Ar as a function of reciprocal temperature. The points were measured in a number of experiments using samples ranging in thickness from 250 to 385 μm in Xe, from 60 to 130 μm in Kr and from 50 to 85 μm in Ar. The solid lines have been calculated from the nonadiabatic theory of Holstein (Ref. 17) using Eq. (5) and the following values of the parameters: Xe (curve a): $E_b = 20$ meV, $J = 1.5$ meV, $\hbar\omega_0 = 25$ meV, $\gamma = 0.8$, $a = 4.47$ Å; (curve b): $E_b = 5$ meV, $J = 0.76$ meV, $\hbar\omega_0 = 30$ meV, $\gamma = 0.17$, $a = 4.47$ Å. Kr: $E_b = 40$ meV, $J = 4$ meV, $\hbar\omega_0 = 15$ meV, $\gamma = 2.7$, $a = 4.12$ Å. Ar: $E_b = 70$ meV, $J = 12$ meV, $\hbar\omega_0 = 7$ meV, $\gamma = 10$, $a = 3.90$ Å.

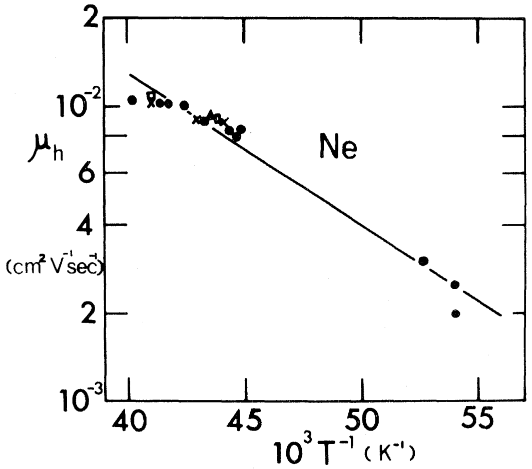

FIG. 2. Semilogarithmic plot of the hole drift mobility in solid Ne as a function of reciprocal temperature. The points are experimental data from a number of runs using samples from 180 to 470 μm in thickness. The solid line is the theoretical mobility calculated from the adiabatic theory (Ref. 19) using Eq. (4) and the following values of the parameters: $E_b - 2J = 24$ meV; $\hbar\omega_0 = 5$ meV and $a = 3.12$ Å.

III. EXPERIMENTAL RESULTS

The temperature dependence of the hole mobilities in Xe, Kr, and Ar is shown in Fig. 1 on a semilogarithmic plot of μ_h vs $10^3/T$. For clarity, the Ne results are presented separately in Fig. 2. Measurements at lower temperatures in Ne were difficult because of the relative ease with which the crystals cracked. The data in this range are therefore rather limited. All hole mobilities are of the order of 10^{-2} cm^2 V^{-1} sec^{-1}, four to five orders of magnitude lower than the corresponding electron mobilities. Values of μ_h near the triple point are listed in Table I and it can be seen that μ_h attains its maximum in Kr and decreases again in Ar and Ne.

The most interesting feature is, however, the progressive change in the form of the temperature dependence of μ_h as one goes from Xe to Ne. Our previous results for Xe, included in Fig. 1, led to a temperature dependence of the form $\mu_h \propto T^{-1.6}$, which is indicated by curve (b). In Kr there appears to be little or no temperature dependence of μ_h over the range investigated, whereas in Ar and Ne we observe essentially an activated hole transport. The interpretation and discussion of these results forms the subject of Sec. IV.

IV. DISCUSSION

Calculations of the band structure of the rare-gas solids indicate that the valence bands should be of the order of 1 eV wide.[10–13] On the basis of simple theoretical considerations one would expect that carriers in these bands should have mobilities greater than unity and that their transport should be describable by conventional band theory. The results presented in Sec. III clearly show that the hole transport in the rare-gas solids does not fit into this pattern. The theoretical treatment of mobilities of the order of 10^{-2} cm^2 V^{-1} sec^{-1} requires a fundamentally different approach which first of all takes account of the increased localization of the charge carrier, and secondly treats the carrier-phonon interaction not as a perturbation, but as the principal interaction.

The hole localization in the rare gas solids is a problem which has been discussed by Druger and Knox[14] and by Song.[15] In both papers it is suggested that localization occurs through the formation of a rare-gas R_2^+ molecular ion. One might envisage the hole self-trapped between two rare-gas atoms in a potential well, which is produced by the resulting lattice and electronic distortions. A center, isoelectronic to the R_2^+ molecular ion, is the well-known V_k center found in the alkali halides.[16] The close analogy suggests that the R_2^+ center should be stable and the detailed calculations confirm this.[14,15]

We believe that the small polaron theory provides a useful approach to the understanding of the hole transport. The localized hole will interact with the vibrational modes and occasionally tunnel to a neighboring atom, where it will reestablish an R_2^+ center. Probably the most detailed and applicable theory dealing with low mobility transport in molecular crystals is the small polaron theory developed by Holstein[17] and others.[18] In the following we shall first present the main results of this theory and then apply it to the hole data. Finally, we shall attempt to correlate the values of the parameters obtained from fitting the data to the theory and comment on their physical significance.

A. Small-polaron theory

The small polaron, i.e., the carrier self-trapped in its polarization field, moves from one site to another by a tunnelling or hopping process. The hopping mobility is given by

$$\mu = (ea^2/kT)P\,, \qquad (1)$$

where a denotes the spacing between the hopping sites and P is the hopping probability.

The transfer of the small polaron from one site to another is most likely to occur at an instant when the states associated with the conduction process coincide energetically on both sites. This means that phonon interaction must momentarily create equal distortions on both sites before a transfer can take place. We shall assume that the carrier interacts principally with phonons of energy $\hbar\omega_0$. In his theory Holstein[17] distinguishes two cases which depend on the relative magnitudes of the transfer integral J between the two hopping sites, the polaron binding energy E_b, the phonon energy $\hbar\omega_0$ and the temperature T.

In the classical limit, which requires that[19]

$$kT \gg \left(\frac{E_b}{2}\right)^{1/4}\left(\frac{2kT}{\pi}\right)^{1/4}\left(\frac{\hbar\omega_0}{\pi}\right)^{1/2}, \tag{2}$$

the so-called *adiabatic* approximation applies if[17]

$$J \gg \left(\frac{E_b}{2}\right)^{1/4}\left(\frac{2kT}{\pi}\right)^{1/4}\left(\frac{\hbar\omega_0}{\pi}\right)^{1/2}. \tag{3}$$

In the adiabatic case the calculation of P and Eq. (1) then lead to the expression[19]

$$\mu = \frac{ea^2}{kT}\frac{3\omega_0}{4\pi}\exp\left(\frac{-(E_b - 2J)}{2kT}\right). \tag{4}$$

In the nonadiabatic approximation on the other hand, the mobility is given by the expression[17]

$$\mu = \frac{ea^2}{kT}\frac{J^2}{\hbar^2\omega_0}\left(\frac{\pi}{\gamma\,\mathrm{csch}(\hbar\omega_0/2kT)}\right)^{1/2} \times \exp\left[-2\gamma\tanh\left(\frac{\hbar\omega_0}{4kT}\right)\right], \tag{5}$$

under the assumption that

$$2\gamma\,\mathrm{csch}(\hbar\omega_0/2kT) \gg 1. \tag{6}$$

$\gamma = E_b/\hbar\omega_0$ is a measure of the coupling between the charge carrier and the lattice. In the regime where $kT \gtrsim \frac{1}{3}\hbar\omega_0$ the applicability of the nonadiabatic approach is limited by a condition opposite to that in Eq. (3), namely,

$$J \lesssim \left(\frac{E_b}{2}\right)^{1/4}\left(\frac{2kT}{\pi}\right)^{1/4}\left(\frac{\hbar\omega_0}{\pi}\right)^{1/2}. \tag{7}$$

The essential difference between the adiabatic and nonadiabatic limits can be understood[18] by considering the probability of the hole tunnelling from one localized site to another during a time approximately equal to the "coincidence time." In the adiabatic case the wave function overlap described by J is sufficiently large so that the hole can tunnel from one site to another during one excited state of the system. In the nonadiabatic approximation on the other hand the chance of the hole tunnelling from one site to the other is small during the same time.

It is important to note that the equations in this section apply to a one-dimensional lattice. The extension to three dimensions would involve the coordination of the localization center, which is uncertain. It will become apparent in Sec. IV B that the fitting of the data to the theory leads to fairly extensive possible ranges of the parameters. In view of this it was felt that Eqs. (4) and (5) form a reasonable basis for the following analysis.

B. Analysis of experimental results

In the analysis a value for the hopping distance a is required. We have taken this to be the nearest neighbor distance in each of the crystals. If, as has been suggested, hopping takes place between R_2^+ centers then this should be appropriate.

First, let us consider the experimental results in relation to the nonadiabatic theory given by Eq. (5). A fit to the data can be obtained over a fairly wide range of values for each of the parameters involved in Eq. (5). Table II lists the range of E_b, J, $\hbar\omega_0$, and γ from which a set of values can be chosen to obtain agreement between the nonadiabatic theory and the experimental data. The particular choice of parameters leading to the solid lines in Fig. 1 are given in the figure caption. It is encouraging to see that the theory can account satisfactorily for the progressive changes in the temperature dependence of μ_h.

The case of Xe is of interest in this connection. To fit the data we require weak polaron binding and a small value of γ. Expanding the csch term in Eq. (5) gives, to first order, a preexponential term proportional to $T^{-1.5}$, which now dominates the temperature dependence. As pointed out before, this is in agreement with the earlier results, represented by curve (b) of Fig. 1. Curve (a) of Fig. 1 is another possible fit corresponding to a lager γ and illustrates the latitude inherent in the fitting procedure. However, it should be pointed out that the choice of parameters for curve (b) of Fig. 1 and also for some of the combinations of values in Table II for Xe, either do not satisfy the condition expressed by Eq. (6), or are only marginally in agreement with it. It is possible that in the weak coupling limit suggested by the Xe data, the more general expression given in Ref. 18 should be used in preference to Eq. (5). In view of the complications involved, this has not been attempted here. As pointed out by Emin,[20] the applicability of the nonadiabatic expression Eq. (5), may well extend beyond the original limits set by Holstein.[17] However, it should be stressed that the parameters in Table II for all the other solids satisfy Eq. (6), so that this complication does not arise.

The scatter in the experimental data for Kr makes a fit to Eq. (5) rather uncertain. We have

TABLE II. Values of the parameters obtained by fitting the hole mobility to the expression given by Eq. (5) for the nonadiabatic hopping theory of Holstein (Ref. 17).

Solid	a (Å)	E_b (meV)	J (meV)	$\hbar\omega_0$ (meV)	γ
Xe	4.47	1–50	0.5–2.5	10–50	0.1–1
Kr	4.12	~40	~4	~15	~3
Ar	3.90	60–90	8–18	5–9	~10
Ne	3.12	20–30	5–10	0.5–2.0	~30

therefore included in Table II simply the values of the parameters that lead to the solid curve in Fig. 1. These parameters and those for Xe satisfy the condition for the applicability of the nonadiabatic theory given by Eq. (7), but this is not the case for Ar and Ne. At best, the Ar parameters only marginally satisfy Eq. (7); in the case of Ne it is not possible to find a set of parameters which fit the results in Fig. 2 and at the same time satisfy Eq. (7).

This raises the question as to the possible applicability of the adiabatic theory given by Eq. (4). Table III summarizes the values of the parameters which will, in principle, lead to a fit of Eq. (4) to the experimental data. One difficulty is that E_b and J cannot now be determined separately. The small values of $\hbar\omega_0$ required in the case of Xe suggest that adiabatic hopping can be excluded as a likely interpretation. However, this does not apply to Ar and particularly to Ne, where Eq. (4) can lead to a satisfactory fit with acceptable parameters. The solid line in Fig. 2 for Ne was calculated from the adiabatic theory with the parameters shown in the caption.

The analysis of the experimental data thus leads to the conclusion that small-polaron theory can account for the hole transport in the rare-gas solids. It is however uncertain, particularly for Ar, whether the adiabatic or nonadiabatic approximation is the correct interpretation. A further complication is introduced by the possible occurrence of correlated small polaron hopping.[18,21] In the original theory it was assumed that the time characterising the relaxation of the lattice after a hop is short compared to the mean time between hopping events. Emin[21] pointed out that the opposite can occur and that in this case the value of E_b may be reduced significantly below that for a relaxed lattice. Using the conditions derived by Emin and a 10% dispersion in the phonon frequencies, one finds that correlated hopping cannot be excluded for the rare-gas solids. If it occurs, then the deduced E_b values will be too small.

C. Correlation of the deduced hopping parameters

It is generally found that the physical properties of the rare-gas solids vary in a systematic way as one goes through the family of these simple solids. If the same applies to the physical quantities involved in the hole transport, and we shall assume it does, then a correlation of the deduced parameters may help to resolve some of the uncertainties of the analysis.

In Fig. 3 we have plotted the nonadiabatic binding energies from Table II as a function of the reciprocal of the atomic mass number M. It should be emphasized that this independent variable has been chosen primarily as a convenient method of representation and it is not intended to imply any particular physical model. In fact a number of other physical quantities, such as the triple-point temperature, could have been used instead and still produced approximately linear plots.

TABLE III. Values of the parameters obtained by fitting the hole mobility to the expression given by Eq. (4) for the adiabatic hopping theory (Ref. 19).

Solid	a (Å)	$E_b - 2J$ (meV)	$\hbar\omega_0$ (meV)
Xe	4.47	$\ll 25$	< 0.8
Kr	4.12	$\lesssim 40$	$\lesssim 6$
Ar	3.90	60–80	20–100
Ne	3.12	20–25	8–25

The values for Xe, Kr, and Ar lie on a reasonably straight line, but the Ne value deviates markedly. This is not entirely unexpected, because the analysis of the results led to the tentative conclusion that the adiabatic rather than the nonadiabatic theory should be applicable to Ne. However, Fig. 3 offers the possibility of estimating E_b for Ne by extrapolation, assuming that the linear relation between E_b and M^{-1} remains valid. This gives $E_b = 150 \pm 30$ meV and, with the value of $E_b - 2J \simeq 24$ meV from Table III, we obtain $J = 63 \pm 15$ meV for Ne. This set of parameters would satisfy the condition given by Eq. (3) for the applicability of the adiabatic approach.

The data presented in Fig. 3 indicates, to a first approximation at least, that $E_b \propto M^{-1}$. Holstein's theory[17] expresses E_b in the form

$$E_b = A^2/2M\omega_0^2, \tag{8}$$

where A is a constant for a particular material and enters the theory by expressing the carrier-lattice interaction energy as

$$E(x) = -Ax. \tag{9}$$

Physically, $E(x)$ represents the energy of the localized hole on an isolated molecule. If the correlations of Figs. 3 and 4 are meaningful then the equation implies that $A \propto \omega_0$ and also $A \propto M$. The calculation of A is a difficult theoretical problem and no information seems to be available to check this prediction.

An attempt will now be made to correlate the electron transfer integrals with the hopping distances. In its simplest form J can be expressed by the relation[22]

$$J = I e^{-\alpha a}, \tag{10}$$

where I is a quantity of the order of the ionization energy of the atoms[23] and α is the decay parameter of the localized-hole wave function. Figure 4 is a semilogarithmic plot of J/I against a. The solid line has been calculated from Eq. (10), with $\alpha^{-1} = 0.5$ Å. Once again the nonadiabatic value for Ne does not fit into the correlation, whereas the previously estimated adiabatic value does.

The fit of the data with an approximately constant value of α may at first sight be surprising because of the relatively large variations in atomic size of the rare-gas atoms. However, it must be borne in mind that Eq. (10) is used here merely to represent the relatively weak tail of the wave function associated with the localized hole and in such a case the overlap is unlikely to be strongly dependent on the details of the center producing the localization.

Finally, it is of interest to consider the ranges of $\hbar\omega_0$ in Table II for Xe, Kr (and possibly Ar), and those in Table III for Ar and Ne. The lack of any systematic variation makes positive identification difficult. In most hopping models the phonon energy is identified with a predominant optical mode of vibration in the material, but this is not possible in the rare gas solids. An alternative interpretation is in terms of zone boundary acoustic phonons. Neutron studies[24–26] and other experiments[27–30] indicate that their energies are about 5, 6, 8, and 6 meV for Xe, Kr, Ar, and Ne, respectively. Although we can not exclude this interpretation it is perhaps more likely that the hole interacts primarily with the vibrational modes of its own R_2^+ center. Unfortunately, we know of no independent information on this point.

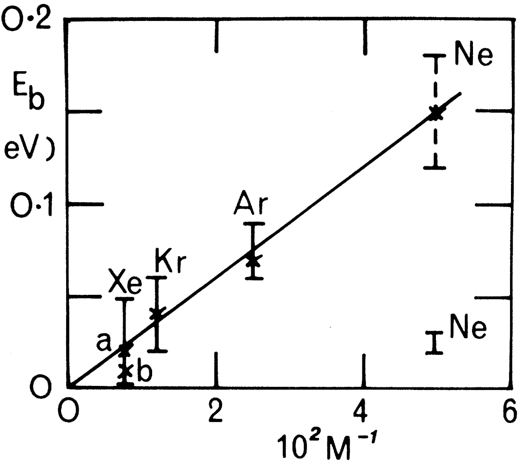

FIG. 3. Correlation of the polaron binding energy E_b with the reciprocal of the rare-gas mass number. The solid vertical lines are values taken from Table II, whereas the dashed line for Ne has been obtained by extrapolating the other data. The crosses denote the values of E_b used to calculate the theoretical lines in Figs. 1 and 2.

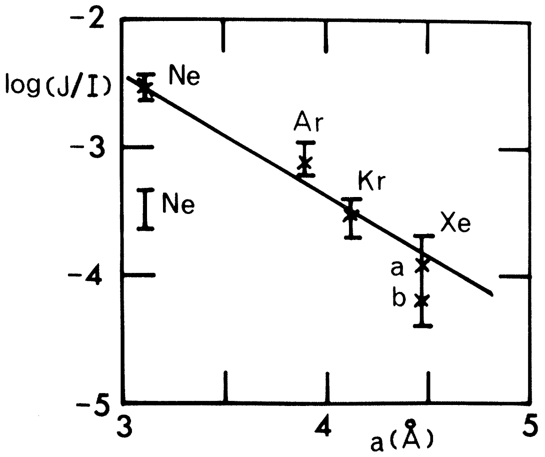

FIG. 4. Correlation of the transfer integral J, normalized to the ionization energy I of the gas, with the hopping distance. Values of J are from Table II, with the exception of the upper range of values for Ne which are deduced from Table III and Fig. 3 as described in the text. The crosses denote the values of J used to calculate the theoretical lines in Figs. 1 and 2. The solid line is calculated from Eq. (10) with $\alpha = 0.5$ Å. Values of I from Ref. 23.

V. CONCLUSIONS

(i) The hole mobilities in the rare-gas solids Ne, Ar, Kr, and Xe at temperatures close to their respective triple points, lie between 1×10^{-2} and 4×10^{-2} cm^2 V^{-1} sec^{-1}, several orders of magnitude lower than the electron mobilities in the same crystals (see Table I). It is likely that holes are localized by the formation of R_2^+ molecular ions.

(ii) The nature of the temperature dependence of μ_h varies progressively from Xe to Ne and the results can be accounted for by the small-polaron theory. The data for Xe and possibly Kr fit the nonadiabatic approximation, whereas the adiabatic theory provides the best interpretation of the Ne results.

(iii) The small-polaron parameters obtained from fitting the theory to the experimental results show the following trends on going from Ne to Xe: (a) the polaron binding energy E_b decreases, (b) the transfer integral for holes decreases almost exponentially as a function of the hopping distance, and (c) the phonon energy $\hbar\omega_0$ shows no systematic variation and may possibly be associated with the vibrational modes of the R_2^+ center.

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