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Exchange Forces Between Li^+ and He, and the Mobility of Li^+ in He*

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A calculation of the exchange force between Li^+ and He has been made with the use of simple hydrogenic state functions to represent the charge distributions of the ion and atom. The force so obtained was added to that arising from the polarizability and the van der Waals interaction between Li^+ and He. The result was then used to calculate the mobility of Li^+ in He gas. The theoretical value thus found, 19.4 cm²/sec. volt at a temperature of 18°C, is somewhat lower than the experimental one of 25.8 cm²/sec. volt, reported by Hoselitz.

INTRODUCTION

THE theory of the mobility of positive ions in gases has been developed first by Langevin[1] and later, with more rigor, by Chapman[2] and Enskog[3] independently. It has been applied by Hassé and Cook,[4] and, considering the fact that they used hard sphere models with long range polarizability forces and took little or no account of charge exchange, the results were highly satisfactory. With the development of quantum mechanics, it became possible both to calculate the correct force law and to take proper account of charge exchange. This was done by Massey and Mohr[5] but with very unsatisfactory results. Their value of 12 cm²/sec. volt for the theoretical value of the mobility of He^+ in He is in greater disagreement with the experimental value of 21.4 cm²/sec. volt quoted by Tyndall and Powell[6] than were earlier calculations. Massey and Mohr recalculated the force law by the method of Hylleraas[7] and found little difference from their original one calculated by the method of Majorana[8] and Pauling.[9] The present author has checked their interaction with that more recently suggested by Weinbaum[10] and found satisfactory agreement, so that the discrepancy cannot be ascribed to the force law.

The reason for this difference between theory and experiment is still unknown. Mott[11] has suggested that Massey and Mohr may have overestimated the effect of the exchange forces by their assumption that the mean period of electron exchange is small compared to the time of passage of the ion past the atom. This is equivalent to an overestimate of the range of the interaction and would lead to a value too low for the mobility. If this is the cause of the discrepancy, the calculation of the mobility for a similar structure in which the exchange effect is small should give better results. The most suitable molecular-ion combination is that of Li^+ in helium gas. Here the exchange forces are small except at very short distances. Electron exchange is impossible because of the large difference in the ionization energies of Li and He. Hence, we have chosen the above ion molecule combination for investigation.

Before calculating the mobility of Li^+ in He we must know the exchange forces between the ion and the atom. Since this is a problem of some importance in itself, this work will be considered first.

THE EXCHANGE FORCES BETWEEN Li^+ AND He

We have selected for this calculation simple hydrogenic state functions to represent the charge distribution of the ion and atom. Since the Hartree functions were not available for Li^+, we chose those screening constants for the atom and ion that minimized the atomic energy. The procedure followed for the greater part of this section

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* Part of a dissertation presented to the faculty of the Graduate School of Yale University in candidacy for the degree of Doctor of Philosophy.

[1] P. Langevin, Ann. Chim. Phys. **5**, 245 (1905).

[2] S. Chapman, Phil. Trans. **A216**, 279 (1916); **A217**, 115 (1917).

[3] D. Enskog, Inaug. Diss. (Uppsala, 1917).

[4] H. R. Hassé and W. R. Cook, Phil. Mag. **12**, 554 (1931).

[5] Massey and Mohr, Proc. Roy. Soc. **A144**, 188 (1934).

[6] A. M. Tyndall and C. F. Powell, Proc. Roy. Soc. **A134**, 125 (1931).

[7] E. A. Hylleraas, Zeits. f. Physik **71**, 739 (1931).

[8] E. Majorana, Nuovo Cimento **8**, 22 (1931).

[9] L. Pauling, J. Chem. Phys. **1**, 56 (1933).

[10] S. Weinbaum, J. Chem. Phys. **3**, 547 (1935).

[11] See A. M. Tyndall, *The Mobility of Positive Ions in Gases* (Cambridge University Press, 1938), p. 36.

was one outlined by Rosen.[12] The one-electron function used for Li^+ was

$$a=\left[\left(\frac{43}{16a_0}\right)^3\frac{1}{\pi}\right]^{\frac{1}{2}}\exp\left(-\frac{43}{16}r_a\right),$$

that for He

$$b=\left[\left(\frac{27}{16a_0}\right)^3\frac{1}{\pi}\right]^{\frac{1}{2}}\exp\left(-\frac{27}{16}r_b\right),$$

where a_0 is the radius of the first Bohr orbit, r is measured in a_0. The theory and some of the functions necessary for the evaluation of the integrals occurring in the expression for the energy are given by Rosen, whose notation will be followed here. For the long range repulsive forces, several approximations are valid. We may take

$$I^2K_2 \sim I^2/R,$$

$$IJ_3(a) \sim IJ_1(a),$$

$$IJ_3(b) \sim IJ_1(b).$$

The appearance of two screening constants in the integrals causes slightly more labor in evaluation of the integrals than in the case of one, but, with the exception of J_2, the exchange integral, little difficulty was encountered. J_2, on the other hand, turns out to be an infinite series and, when the difference in screening constants is unity as it is in this case, a large number of terms would be needed. To avoid this, the following approximation used previously by Gentile[13] was employed.

$$J_2=\int\int\frac{a_1b_1a_2b_2dv_1dv_2}{r_{12}} \sim \int\int a_1b_1a_2b_2dv_1dv_2\frac{\displaystyle\int\int\frac{a_1^*b_1^*a_2^*b_2^*dv_1dv_2}{r_{12}}}{\displaystyle\int\int a_1^*b_1^*a_2^*b_2^*dv_1dv_2}$$

TABLE I. Repulsion energy between Li^+ and He.

R in A	Δw in ergs
1.05	3.442×10^{-12}
1.58	1.508×10^{-13}
2.11	6.846×10^{-15}
2.64	3.109×10^{-16}
3.16	1.412×10^{-17}

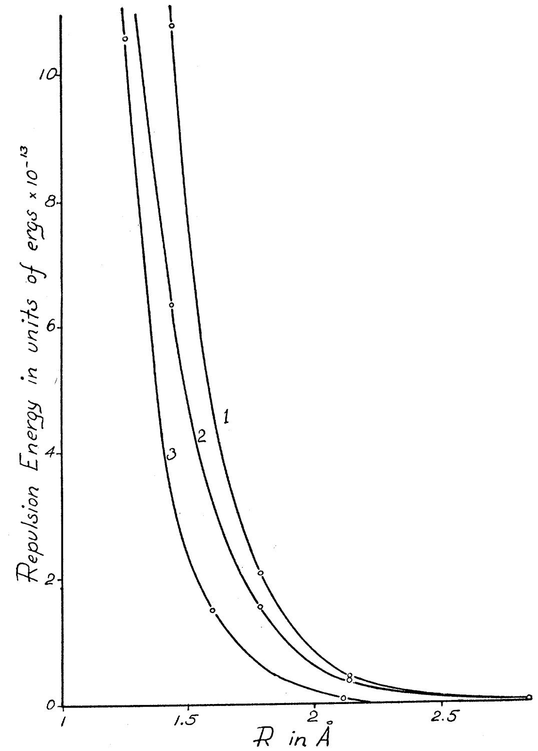

FIG. 1. (1) Repulsion energy between two He atoms as calculated by Slater. (2) Repulsion energy between two He atoms as calculated by Rosen. (3) Repulsion energy between $\mathrm{Li}^+-\mathrm{He}$.

where

$$a_1^*=b_1^*=\left[\left(\frac{43+27}{2\times16a_0}\right)^3\frac{1}{\pi}\right]^{\frac{1}{2}}\exp\left(-\frac{43+27}{2\times16}r_1\right).$$

Carrying out the above calculations, one obtains the values shown in Table I for Δw, the repulsion energy between $\mathrm{Li}^+-\mathrm{He}$. To the degree of accuracy of the calculations, Δw can be represented by

$$\Delta w = 1.61\times10^{-9}\,e^{-5.89R}\text{ ergs},\quad R\text{ in A.}$$

Figure 1 is a graph of this interaction compared with the repulsion energy between two helium atoms as calculated by both Rosen[12] and Slater.[14]

THE MOBILITY OF Li^+ IN He

The mobility K of ions in a given gas is proportional to the diffusion coefficient D_{12} of the ions in that gas. The expression for this is

$$K=D_{12}e/RT, \tag{1}$$

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[12] N. Rosen, Phys. Rev. **38**, 255 (1931).

[13] G. Gentile, Zeits. f. Physik **63**, 795 (1930).

[14] J. C. Slater, Phys. Rev. **34**, 1293 (1929).

e being the charge on the ion, R the gas constant, and T the absolute temperature of the gas. The theory necessary for the calculation of D_{12} is given by Chapman.[2]

$$D_{12}=\frac{1}{1-\epsilon_0}\frac{3}{4\pi\nu_0 JM}\frac{1}{\mathrm{K}'_{12}(0)}, \tag{2}$$

where $M=M_aM_b/(M_a+M_b)$ and M_a and M_b are the masses of the ion and gas molecule, respectively, $J\equiv 1/2RT$, ν_0 is the number of molecules per unit volume, ϵ_0 is a correction term that is usually small, and

$$\mathrm{K}'_{12}(0)=8\pi^{-\frac{1}{2}}\{JM\}^{5/2} \times\int_0^\infty \exp\left(-JMV^2\right)Q_D V^5 dV. \tag{3}$$

V is the velocity of the ion relative to the atom, and Q_D, the cross section for diffusion, is given by

$$Q_D=2\pi k^{-2}\sum_{n=0}^{\infty}\left\{(2n+1)\sin^2\eta_n -2(n+1)\left(\frac{\sin 2\eta_n \sin 2\eta_{n+1}}{4} +\sin^2\eta_n\sin^2\eta_{n+1}\right)\right\}, \tag{4}$$

where $k=2\pi MV/h$, and the η_n are the phase shifts introduced by the potential $\delta(r)$. The phases are defined in the following way. Let $G_n(r)$ be the bounded solution of

$$G_n''(r)+\left[k^2-U(r)-\frac{n(n+1)}{r^2}\right]G_n(r)=0, \tag{5}$$

with $U(r)=(8\pi^2M/h^2)\delta(r)$, then, for large r, $G_n(r)$ will have the asymptotic form

$$G_n(r)\sim\sin\left(kr-\tfrac{1}{2}n\pi+\eta_n\right). \tag{6}$$

η_n is thus defined. Approximate expressions for η_n have been derived for special cases. When the phases are small, i.e., less than $\pi/2$, and n large, the Born[15] approximation

$$\eta_n=-\frac{4\pi M}{h^2}\int_0^\infty \delta(r)\left[J_{n+\frac{1}{2}}(kr)\right]^2 r dr, \tag{7}$$

in which $J_{n+\frac{1}{2}}(kr)$ is the Bessel function of order $n+\frac{1}{2}$, is particularly useful. When the phases are large and n small, the approximation due to Jeffreys,[16]

$$\eta_n=\int_{r_0}^{r}\left[k^2-U(r)-\frac{n(n+1)}{r^2}\right]^{\frac{1}{2}}dr +\frac{(2n+1)\pi}{4}-kr, \tag{8}$$

r_0 being the largest zero of $[k^2-U(r)-(n(n+1)/r^2)]$, is valid. In the intermediate region where both approximations fail, the phases are usually obtained by plotting the phases as calculated by both methods against the order n and interpolating.

A difficulty arises when the above procedure is applied to the present problem. The phases are not continuous functions of the order n; several discontinuities may occur. In our case a discontinuity occurs in about the region where both the above approximations fail. This makes interpolation difficult and inexact. To avoid this difficulty a modification of the Jeffreys approximation has here been developed which enables one to pass, without interpolation, from the region where the phases are large into that where the Born approximation is valid.

Most of the error in the Jeffreys approximation arises in the neighborhood of the point $r=r_0$. Here the terms, assumed to be small, become infinite and, the phase not being known exactly, is taken to be $\pi/4$. A much better point to start the integration is at the first zero of G_n after r_0, which we will call r_1, i.e., $G_n(r_1)=0$. Here the phase is π and we have passed over the region where the neglected terms contribute anything appreciable. If this is done, the phase becomes

$$\eta_n=\int_{r_1}^{r}\left[k^2-U(r)-\frac{n(n+1)}{r^2}\right]^{\frac{1}{2}}dr +\frac{(n+2)\pi}{2}-kr. \tag{9}$$

r_1 is obtained by expressing $U(r)$ at r_0 in the form

$$U(r)=A+B/r^2.$$

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[15] See N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, 1933), p. 28.

[16] H. Jeffreys, Proc. Lond. Math. Soc. [II] **23**, Part 6; Phil. Mag. **33**, 451 (1942).

TABLE II. Comparison of the phases for $k=6\times10^8$ as calculated by different methods.

n	η_n (Jeffreys)	η_n (Modified Jeffreys)	η_n (Born)
23	0.86π	0.76π	0.62π
24	0.71π	0.64π	0.55π
25	0.60π	0.55π	0.49π
26	0.50π	0.48π	0.45π
27	0.44π	0.42π	0.39π
28	0.40π	0.37π	0.35π
29	0.36π	0.33π	0.32π
30	0.33π	0.30π	0.29π
31	0.29π	0.27π	0.26π
32	0.26π	0.23π	0.24π
33	0.24π	0.22π	0.22π

Then Eq. (5) takes the form

$$G_n''(r)+\left[k^2-A-\frac{n(n+1)+B}{r^2}\right]G_n(r)=0. \qquad (10)$$

This is Bessel's equation of order $p=[n(n+1)+B+\frac{1}{4}]^{\frac{1}{2}}$ in the variable $cr=r(k^2-A)^{\frac{1}{2}}$. The value of r_1 can be obtained from a table of first roots of Bessel's functions of order p. Table II shows the phases for $k=6\times10^8$ in the critical region. Here the Jeffreys approximation is usually too high while the Born approximation is too low. Our modified Jeffreys approximation seems both to divide the region and converge rapidly to the Born result which is known to be valid for large r.

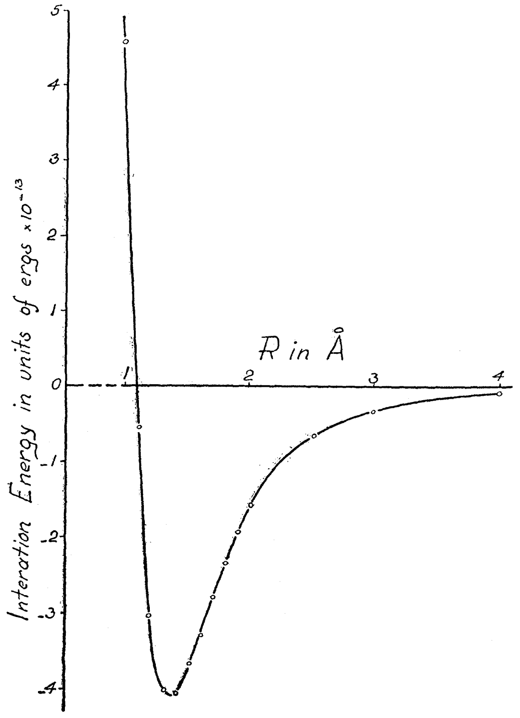

FIG. 2. Calculated interaction energies $\delta(r)$ of Li^+-He.

The calculation of the phases for collisions between Li^+ and He requires a knowledge of the potential $\delta(r)$ (or $U(r)$) in Eq. (5). This potential is an attraction at large distances of separation and turns to a repulsion only at small distances due to the effect of the exchange forces calculated in the previous section. Margenau[17] has shown that the attraction may be approximated by two terms; one varying as r^{-4}, the other as r^{-6}. These terms arise from the polarizability of both the ion and the atom. When the atom moves into the field of the ion, it acquires not only an induced dipole moment but also quadrupole moment and higher moments. The dipole moment gives rise to the term in r^{-4} while the quadrupole moment contributes to the term in r^{-6}. The remaining contribution to this term arises from the polarizability of the ion itself. This is analogous to the van der Waals interaction between neutral atoms and also varies as r^{-6}. The total interaction is given by

$$\delta(r)=v(r)-(\alpha/r^4)-(\beta/r^6), \qquad (11)$$

where $v(r)$ is the repulsion energy. From Margenau's paper we obtain the following values for these constants $\alpha=2.36\times10^{-44}$ erg cm^4; $\beta=1.84\times10^{-60}$ erg cm^6. The graph of this interaction is shown in Fig. 2.

To simplify the calculation, we have employed an approximation used by Massey and Mohr[5] and others. The phases start out large for small values of n and suddenly become small (at the discontinuity mentioned before), then diminish asymptotically to zero. In the region of large phase, $\sin\eta_n$ oscillates rapidly. Hence we permit ourselves to replace $\sin^2\eta_n$ and $\sin\eta_n$ by their average values $\frac{1}{2}$ and 0 in this region. Thus, if the first small phase occurs for $n=m$, the contribution to the collision cross section, $Q_D^{(1)}$, say, is

$$Q_D^{(1)}=\tfrac{1}{2}\pi k^{-2}m(m+1). \qquad (12)$$

The contribution for $n>m$, $Q_D^{(2)}$, is calculated and added to $Q_D^{(1)}$, to give the total collision cross section for diffusion. Thus

$$Q_D=Q_D^{(1)}+Q_D^{(2)}. \qquad (13)$$

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[17] H. Margenau, J. Phil. Sci. 8, 603 (1941).

For $n \geqq m$, η_n was calculated by the improved Jeffreys approximation up to that value of n for which this approximation agreed numerically with the Born approximation. For higher n, the Born expression was used. The cross section Q_D was calculated for $k\times 10^{-8} = 2, 4, 6, 8, 10$, and 20. The results plotted against $1/k$ are shown in Fig. 3 to a good approximation; Q_D can be expressed as

$$Q_D = 2\pi\times 10^{-16}\left(\frac{28.95}{k} - 0.68\right). \tag{14}$$

When this is put into expressions (3), (2), and (1), we obtain the following result for K in cm^2/volt sec.

$$K = \frac{19.7}{1.265 - 0.0146T^{\frac{1}{2}}}. \tag{15}$$

Table III shows the value of K for various temperatures as calculated by this formula and the experimental ones as given by Hoselitz.[18]

DISCUSSION OF RESULTS

It is seen that the calculated values of the mobility are lower than the experimental ones. At 18°C the error is about 30 percent. This discrepancy is greater than the errors introduced into the calculations. One source of error might be the approximation involved in setting $\sin^2\eta_n = \frac{1}{2}$ for large phases, hence, this was checked for several values of k. For $k = 4\times 10^8$, the approximation was found to yield too low a value, while for $k = 6\times 10^8$ it was exact. Since this error is likely to be greater at lower k values where there are fewer terms to average out the deviations in $\sin^2\eta_n$, the points for determining Q_D were taken in the region $k\times 10^{-8} = 6, 8, 10, 20$, as shown in Fig. 3. This is also the important region in determining the mobility for the temperature range considered. It seems unlikely that the error in Q_D as derived from our potential function is greater than 10 percent. It was also thought that the neglect of ϵ_0 in Eq. (2) might give rise to an appreciable error. This was checked and preliminary calculations indicate that it can be neglected. This is to be expected as the mobility is determined largely by the r^{-4} term in $\delta(r)$ and, classically, ϵ_0 is zero for this force law.

TABLE III. Experimental and theoretical values of K at various temperatures.

T	K_{exp}	K_{th}
20	20	16.4
78	21.8	17.4
90	22.2	17.5
195	23.9	18.6
291	25.8	19.4
389	27.8	20.1
483	29.2	20.9

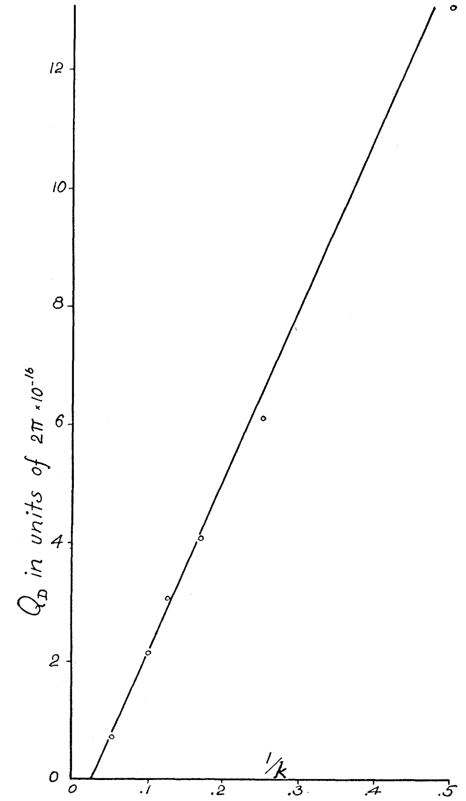

FIG. 3. Q_D plotted as a function of $1/k$.

It should also be pointed out that theoretically we are not justified in using the zero velocity approximation for the force law, whereas the correct expression contains velocity-dependent terms.[19] This approximation should cause little trouble in the present calculation, as the mobility is determined primarily by the polarizability term, and there is very little difference between the polarizability of He for radiofrequencies and for optical frequencies.

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[18] K. Hoselitz, Proc. Roy. Soc. **A177**, 200 (1940).

[19] See H. S. W. Massey and R. A. Smith, Proc. Roy. Soc. **A142**, 142 (1933).

It is very probable that the greatest error arises in the addition of the first-order exchange energy as calculated from wave functions satisfying the Pauli principle to that arising from the polarizability calculated by an entirely different method. In the region of the minimum in $\delta(r)$ both calculations break down, so that its depth can never be accurate. Calculations of this potential minimum from clustering data of Li^+ and He at low temperatures indicate indeed that our minimum may be several times too deep.[20] Should the exact calculation give rise to a saturation in the polarizability of He at a distance of separation of about 2.5A, good agreement between the theoretical and experimental mobility of Li^+ in He would result. It would seem very desirable, therefore, to check the addition of the polarizability term in a way similar to the check made on the van der Waals term by Margenau.[21]

While the present calculations are not in complete agreement with experimental determinations of Li^+ in He gas, the agreement is better than between the calculations of the mobility of He^+ in He and the accepted experimental value. This improvement might be taken to support Mott's suggestion that Massey and Mohr may have overestimated the effect of the exchange forces in their assumption concerning the mean period of electron exchange, which they took to be small compared to the time of passage of the ion past the atom.

Unfortunately the He^+-He problem is not so clear-cut. It is also possible that the experimental results may be confused by "clustering" of the ion. An examination of the exchange forces between He^+ and He shows that they have a considerable range; they become important at about 3A separation between the atom and the ion. The exchange energy between two He atoms becomes important at a shorter range, about 2A. Considering the fact that the internuclear separation between the two atoms in He_2^+ is only 1A, it is quite possible that the repulsion energy between the clustered ion and He, i.e., between He_2^+-He, would not be much greater than at 3A. In this case, the interaction between this "clustered" ion, He_2^+, and He would differ very little from the antisymmetrical case V_a, calculated by Massey and Mohr.[22] They calculated, using V_a alone, that the mobility of He^+-He should be 25 cm^2/volt sec. If this were corrected for an ion of mass 8 instead of 4, the value comes out around 22 cm^2/sec. volt. This agrees very well with the value 21.4 cm^2/sec. volt measured by Tyndall and Powell. If this conjecture proves correct, we have the anomalous situation of the "clustered" ion, He_2^+, having a higher mobility than the unclustered one, He^+.

An examination of the work of Tyndall and Powell[23] lends support to this hypothesis. These workers have made two different determinations of the mobility of He ions in He gas. The gas, in both cases, was of equal purity, but a different ion source was used in each case. From their first measurements using an α-particle source they obtained ions having the mobility of 13 cm^2/sec. volt. They express their disappointment in obtaining a value so much lower than the theoretical values existing at that time. They made another determination using another ion source, this time ions formed from a glow discharge in He, and were gratified in obtaining a mobility, which they attribute to He^+ in He gas, of 21.4 cm^2/sec. volt, which agreed favorably with the theoretical values at that time. Three years later, Massey and Mohr made their calculation of the mobility of He^+ in He and found that their value of 12 cm^2/sec. volt was in sharp disagreement with the accepted value.

The question as to whether the above disagreement is due to an anomalous behavior of the clustered He_2^+ ion or to an error in the calculations can be settled unambiguously only by an identification of the ions whose mobility is measured. However, some information can be gained from an examination of the type of ion sources used. From a consideration of these ion sources it would seem that an incorrect assignment of mobilities may have been made. Arnot and M'Ewen[24] have investigated the production of He_2^+ in He and find that the relative number

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[20] R. S. Munson and K. Hoselitz, Proc. Roy. Soc. **172**, 43 (1939).

[21] H. Margenau, Phys. Rev. **56**, 1000 (1939).

[22] See reference 5. Massey and Mohr calculated the mobility using the symmetrical V_s and antisymmetrical potential V_a alone to investigate the effect of charge exchange. This case corresponds to the latter.

[23] A. M. Tyndall and C. F. Powell, Proc. Roy. Soc. **A129**, 162 (1930).

[24] F. L. Arnot and M. B. M'Ewen, Proc. Roy. Soc. **A171**, 106 (1939).

of He_2^+ ions increases with pressure. At discharge pressures one would expect that many, if not all, the ions would be He_2^+. For their gas discharge source, Tyndall and Powell find only *one* ion present; one would expect, then, that this ion be He_2^+. They find this ion to have the high mobility of 21.4 cm²/sec. volt. While this is not a proof of the incorrectness of the identification of the mobility ions in He, this point should certainly be cleared up before any more theoretical work is done to attempt removal of the discrepancy.

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The author wishes to express his thanks to Professor H. Margenau of Yale University for suggesting this problem and for valuable suggestions and criticisms during the progress of the work and also to Professor Lars Onsager for valuable discussions and criticisms.

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The Photo-Conductance of Evaporated Bismuth Films

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The electrical conductance of Bi films evaporated on Pyrex in high vacuum was measured when the films were "dark" and when illuminated with 2537A radiation. A photo-conductance effect was observed for all films less than about 292 atom layers thick whether the Bi was deposited at room temperature or at liquid-air temperature. The photo-conductance effect disappears with increasing film thickness much more rapidly for films kept at liquid-air temperature than for those at room temperature. For films deposited discontinuously (that is, successive layers are allowed to age), the effect disappears at about 11 atom layers thickness for liquid air-temperature-deposited films and at about 172 atom layers thickness for room-temperature-deposited films. The experiments definitely point to the conclusion that *true* photo-conductance is not present in these films, that the films which exhibit photo-conductance are patch-like in structure; therefore, the observed photo-conductance is assigned to photoelectric emission between film patches.

IN reported[1] investigations of the effect of light on thin metallic films there is evident uncertainty about the existence of *true* photo-conductance in these films. *True* photo-conductance may be defined as the increased electrical conductance of a metallic substance caused by additional electrons (which have absorbed incident quanta) in the conduction band. True photo-conductance has been shown to be a property of the semimetallic element selenium and of compounds like zincblende, but its existence, or nonexistence, in pure metals has not been established. Photo-conductance in general is here defined as the electrical conductance of an illuminated substance; it has a wider meaning than *true* photo-conductance and is contrasted with dark conductance.

The present paper reports an investigation of the photo-conductance of Bi films, deposited on Pyrex by evaporation in high vacuum, aimed at determining whether true photo-conductance exists. The influence of temperature (both of deposition temperature and varying film temperature), of film thickness, and of film aging were systematically studied.

EXPERIMENTAL

All experiments were with Bi deposited on Pyrex by evaporation (at 1.11 atom layers min.$^{-1}$) in high vacuum. The experimental tube, outgassing procedure, and measurement of film thickness have been described.[2] Figure 1 with its legend describes the circuit used for measuring

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[1] B. Gudden and R. Pohl, Physik. Zeits. **23**, 417 (1922); R. S. Bartlett, Phys. Rev. **26**, 247 (1925); A. Etzrodt, Physik. Zeits. **35**, 433 (1935); R. Suhrmann and G. Barth, Zeits. f. Physik **103**, 133 (1936); T. Fukuroi, Sci. Pap. Inst. Phys. Chem. Research **32**, 187 (1937); Q. Majorana, Physik. Zeits. **38**, 663 (1937); T. C. Wilson, Phys. Rev. **55**, 316 (1939); A. H. Weber and D. F. O'Brien, Phys. Rev. **60**, 574 (1941).

[2] A. H. Weber and L. J. Eisele, Phys. Rev. **60**, 570 (1941).