Electron Detachment from Negative Halogen Ions in Collisions with Inert-Gas Atoms and Hydrogen Molecules

IU. F. BYDIN AND V. M. DUKEL'SKII

Leningrad Physico-Technical Institute, Academy of Sciences, USSR

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Measurements have been made of the effective cross sections for electron detachment in the negative ions F⁻, Cl⁻, Br⁻ and I⁻ in collisions with atomic He, Ne, A, Kr and Xe in the ion-energy region from 200 to 2,000 ev. The cross sections have also been measured for collisions between Cl⁻, Br⁻ and I⁻ ions and H_2 molecules. The following energy thresholds for electron detachment were found: Br⁻, He (150 ev); Br⁻, H_2 (160 ev); I⁻, H_2 (320 ev);   the threshold for I⁻, He (280 ev) was established accurately. The results are interpreted in terms of energy transitions in a system consisting of two slowly approaching atomic particles.

1. INTRODUCTION

IN collisions of negative ions with atoms or molecules of a gas the excess electron associated with the negative ion can be detached, making a transition to an unbound state. It has been shown experimentally[1-3] that for ion energies of about 1,000 ev the effective cross sections for this process (which in the following we shall call collision-stripping of negative ions) may reach values of the order of 10^{-15} to 10^{-16} cm^2.

Negative-ion stripping has not been studied adequately although it is of more than usual interest, being a particularly simple process and one which makes easily available for examination inelastic collisions between atomic particles. A study of negative-ion stripping may, in general, prove useful in establishing general features of collisions between slow-moving atomic particles and, in particular in clarifying the question of energy thresholds in the excitation and ionization processes which occur in such collisions. It was our purpose in the present work to study negative-ion stripping in a more detailed and systematic fashion than was done in Refs. 1 and 2, and as far as possible, to remove certain experimental deficiencies in this earlier work.

It was decided to investigate the stripping process in negative halogen ions since these form a homogeneous group with similar structure in the outer part of the electron shell (a closed group of p-electrons). Atoms of the inert gases were chosen as the target particles since these also have closed electron shells. We were also interested in extending the data on energy thresholds for the stripping effect. These thresholds are most accessible to measurement in collisions of heavy negative ions with light atoms[4]. Hence,
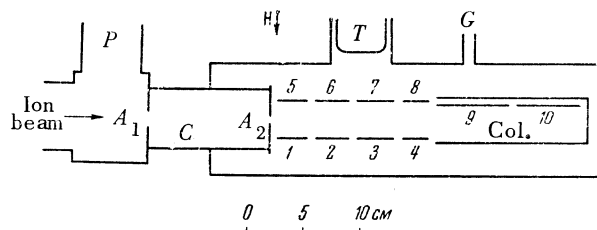

FIG. 1. Diagram of the collision chamber.

careful measurements of the ion-stripping effect were made for Cl⁻, Br⁻ and I⁻ in helium, and these were supplemented by studies in hydrogen.

The ion-stripping was studied for ion energies ranging from 200 to 2,000 ev; in this energy region the stripping effect is not complicated by the presence of the other inelastic processes which occur at higher ion energies[5].

2. EXPERIMENTAL ARRANGEMENT

To observe negative-ion stripping, use was made of a scheme in which the slow electrons produced in the passage of a beam of negative ions through a gas-filled chamber are detected. The experimental arrangement used in the present work is similar to that described in Ref. 1.

The source of negative halogen ions was a hot-cathode gaseous discharge in vapors of one of the salts KF, NaCl, NaBr or NaI. A homogeneous beam of halogen ions was extracted by a magnetic mass-analyzer with a revolving power of 40 and, following additional collimation, introduced into the collision chamber (Fig. 1). Here C is the collimator (the diameter of aperture A_1 is 4.0 mm, that of aperture A_2 is 2.0 mm). Electrons detached from negative ions in collisions with gas atoms were collected by two pair of semi-cylindri-

cal electrodes, 2 and 6 and 3 and 7, which surrounded the ion beam. The measuring electrodes were located between two pair of guard electrodes, 1 and 5 and 4 and 8. The collector Col for the primary ion beam was a deep cylinder designed to prevent the escape of secondary electrons. For this same reason there were two retarding electrodes, 9 and 10, inside the collector; these provided an electric field which was used to deflect secondary electrons. A large liquid-nitrogen trap T was used to freeze out condensable vapors in the collision chamber. The gas was admitted to the chamber through the tube G and was pumped out through the port at P. There was a pressure differential across the aperture A_2. With gas pressure in the collision chamber of a few times 10^{-4} mm Hg the pressure at the port P was approximately 100 times smaller.

The collision chamber was filled with so-called spectrally pure inert gases. Before being admitted to the chamber the gas passed through a coil which was cooled by liquid nitrogen (in the case of Xe it was cooled by solid CO_2 in acetone). The hydrogen was purified by passage through a heated palladium thimble. The gas purity was checked by mass-spectroscopic analysis. The only spurious line in the mass-spectrographic measurements was found to be one due to H_2O^+, the intensity of which was less than 1 percent of the intensity of the test gas. Aside from this no other contamination of the inert gas was found.

The current in the primary ion beam was measured by a dc electrometer amplifier connected to the collector Col. A second electrometer amplifier (type EMA-1) with a maximum usable sensitivity of 1×10^{-15} amp/div was used to measure the current due to electrons which appeared in the collision chamber as a result of ion stripping.

A particular effort was made to reduce the effect of the fringing magnetic field H of the mass-analyzer on the electron collection in the collision chamber. Because of the proximity of the ion source to the magnet of the mass-analyzer, the collision chamber and the ion source were well separated. In the region in which the electrons were detected the horizontal component of the fringing field of the magnet was the largest component; this component was perpendicular to the axis of the collision chamber. The other two components were less than five percent of this one. The measurement and guard electrodes were arranged in the collision chamber in such a way that the electric field between them was in approximately the same direction as the fringing field of the mass-analyzer. This configuration was chosen to reduce the possibility of displacing electrons, produced as a result of negative ion stripping, along the chamber axis.

3. METHOD OF MEASUREMENT

It was the purpose of the present measurements to obtain accurate data which would make possible a determination of the total effective cross section for negative-ion stripping in collisions with gas atoms. To realize this objective a strong electric field was set up between electrodes 1, 2, 3, 4 and 5, 6, 7, 8 to collect electrons which had been detached from negative ions along a definite path in the gas. Knowing the current i due to electrons produced in the gas along a path l, the current I in the negative ion beam, and the number of atoms n per cm^3 in the collision chamber, using the pressure and temperature of the gas it is possible to calculate the magnitude of the total cross section Q for the negative-ion stripping effect by the approximate formula

$$Q = i / Inl. \qquad (1)$$

The current i was measured by connecting the electrometer amplifier EMA-1 to one of the electrodes 2, 3, or 6, 7. In this case, the electrodes located in line with the measuring electrode were grounded and a negative potential was applied to all the opposite electrodes. A series of voltage-current curves which were taken indicated that a potential of -6 volts was sufficient in all cases to obtain the saturation current. The quantity I in Eq. (1) is the value of the current in the negative-ion beam on entrance into the measurement volume. To determine this quantity it is necessary to add the primary beam current at the side electrodes due to electrons produced by negative-ion stripping along the entire path from the entrance of the measurement chamber up to the collector.

The length l from which electrons are collected is taken to be the length of the measuring electrode. It was established in a series of special experiments that the ratio i/I was the same, within the limits of experimental accuracy, when any one of the four electrodes 2, 3, 6, 7 was used as a measuring electrode; this is an indication that edge effects were insignificant.

The gas pressure in the collision chamber was measured with an ionization gauge which was calibrated against a McLeod gauge.
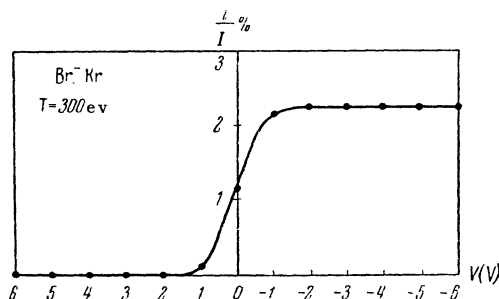
FIG. 2. Voltage-current curve. Br⁻ ions at 300 ev on krypton with $p = 2.6 \times 10^{-4}$ mm Hg.

The various spurious effects which can distort the results obtained with the scheme described here have been considered in Ref. 1. We have carried out a series of control experiments to evaluate these effects under the conditions of the present experiment. By passing a beam of negative ions through the collision chamber when it was pumped down to a pressure of 1×10^{-6} mm Hg it was found that the beam was not intercepted by any of the side electrodes even when a potential difference of ±6 volts was applied between opposite pairs of electrodes. It was also verified that secondary electrons from the edges of aperture A_2 and from the inner surface of the collector Col did not enter the measurement volume. When the collision chamber was filled with gas it was found that electrons produced as a result of negative-ion stripping within the collector did not enter the measurement volume whether or not a potential was applied to electrodes 9 and 10 inside the collector.

In all (negative ion and gas atom) pairs which were investigated, voltage-current curves $i(V)$ (where V is the potential of the electrode opposite the measuring electrode) were measured for the highest, lowest and one intermediate value of the ion energy, while V was varied from +6 volts to −6 volts. At low ion energies (<500 to 600 ev) the voltage-current curves were similar to that shown in Fig. 2 which pertains to a Br⁻ ion at an energy $T = 300$ ev and atomic Kr. In the region of the field which was used to accelerate electrons toward the measurement electrode, saturation current was achieved at $V \approx$ -2 to -3 volts. In the retarding field region the current to the measuring electrode practically vanished at $V = +2$ volts. The absence of a negative current in the left side of Fig. 2 is a consequence of the fact that the negative ions are scattered in the gas and do not reach the measurement electrode in any appreciable quantity; this situation may be explained by the
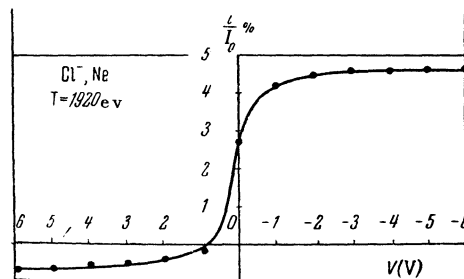

FIG. 3. Voltage-current curve. Cl⁻ ions at 1920 ev on neon with $p = 8.0 \times 10^{-4}$ mm Hg.

small probability for charge-conserving scattering of negative ions at large angles.

In a number of cases in which the ion energy was greater than 500 to 600 ev a small positive current was observed in the left side of the voltage-current curve. A typical case, in which this effect was particularly large, is the voltage-current curve shown in Fig. 3 which was obtained in collisions of Cl⁻ ions at energy $T = 1920$ ev with Ne atoms. The appearance of a positive current might be explained by the production of positive ions in the passage of negative ions through the gas or by the ejection of electrons from the surface of the measurement electrode, for example, as a result of the emission of radiation in the collision of the negative ions with the gas atoms. In this and similar cases, in determining the effective cross section for negative-ion stripping the current i was taken as the difference of the negative and the positive saturation currents. This was done in the present case when the positive current was not small compared with the negative current*.

The possibility that there might be errors in the present results because of the superposition, on the negative-ion stripping, of an exchange effect with atoms of the gas is obviously negligible because the inert gases, in which the bulk of the measurements were made, have zero electron affinity. The absence of charge exchange effects can be con-

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* To investigate the origin of this positive current, an experiment was performed in which an open grid was placed in front of the measuring electrode and slow positive ions were produced in the collision chamber (by charge exchange between Cl⁺ ions in Kr). The dependence of the positive current to the measurement electrode on the grid potential was noticeably different in this case from the dependence of the positive current observed in the ion-stripping case, Cl⁻ in Br. On the basis of these results it may be assumed that the origin of the positive current in the latter case is the ejection of electrons from the measurement electrode.
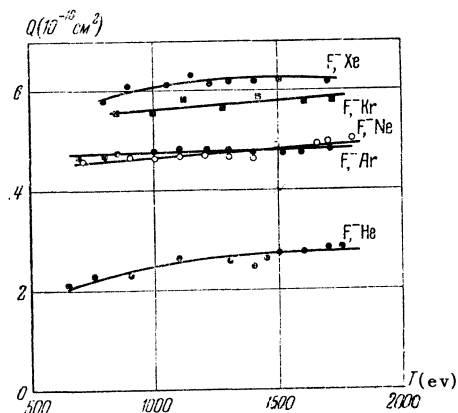
FIG. 4. Effective cross section for stripping of the negative ion F⁻ in inert gases as a function of the ion energy.

clusively demonstrated only in the hydrogen experiments. However, if the H_2 molecule has an electron affinity, the probability for charge exchange between negative halogen ions and molecular H_2 must still be small because of the great difference in the magnitude of the electron bonding energy.

In all cases in which the voltage-current characteristics were taken the dependence of the ratio i/I on the pressure of the gas p in the collision chamber was also investigated (up to a pressure of $\approx 1 \times 10^{-3}$ mm. Hg). The measurements which were used to determine the cross section Q were performed at the values of p which pertain to the linear part of the curve $(i/I)(p)$ in which the magnitude of i/I is less than five percent. Under these ex-
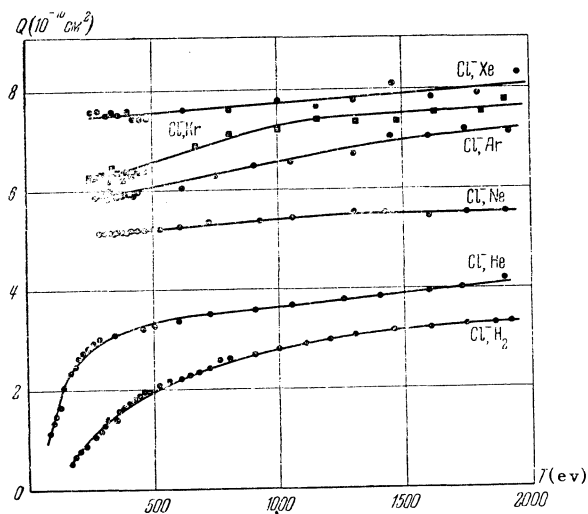
perimental conditions the current attributed to negative-ion stripping in the absence of a gas in the apparatus amounted to 0.1 to 0.2 percent of the current produced by the ion stripping in the gas being studied.

The reproducibility of the measured results was quite good: The average random error in the determination of Q was no greater than six percent. Systematic errors in the measurement of the gas pressure and the length from which electrons were collected in the collision chamber and the use of the approximate formula (1) may increase the total error in the determination of the absolute value of Q to 15 to 20 percent.
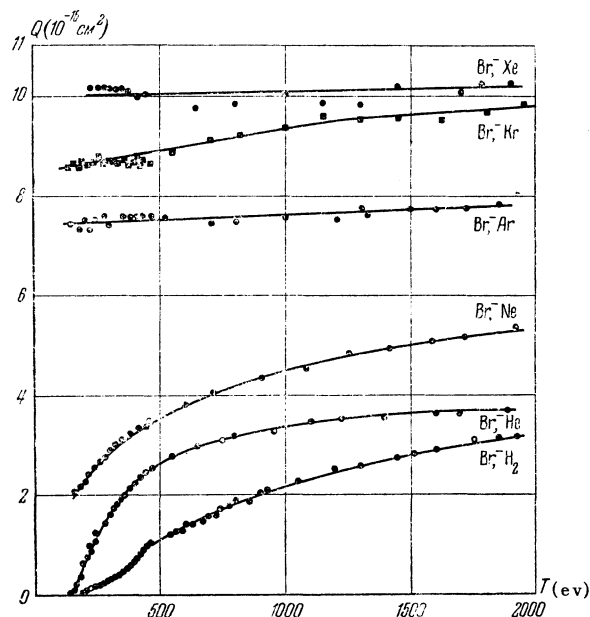

FIG. 6. Effective cross section for stripping of the negative ion Br⁻ in inert gases and hydrogen as a function of ion energy.

4. RESULTS

The measurements which were carried out in the present work have been used to determine the total effective cross sections Q for negative-ion stripping for all the halogens in all the inert gases in the ion-energy region from approximately 200 to 2,000 ev. The quantity Q was also determined for collisions of Cl⁻, Br⁻ and I⁻ uons with H_2 molecules. The results which were obtained are presented in Figs. 4-7 as curves giving the dependence of Q on the ion kinetic energy T.

We were also able to find the threshold for stripping (Fig. 6) in the case of the Br⁻ ion in

FIG. 5. Effective cross section for stripping of the negative ion Cl⁻ in inert gases and hydrogen as a function of ion energy.
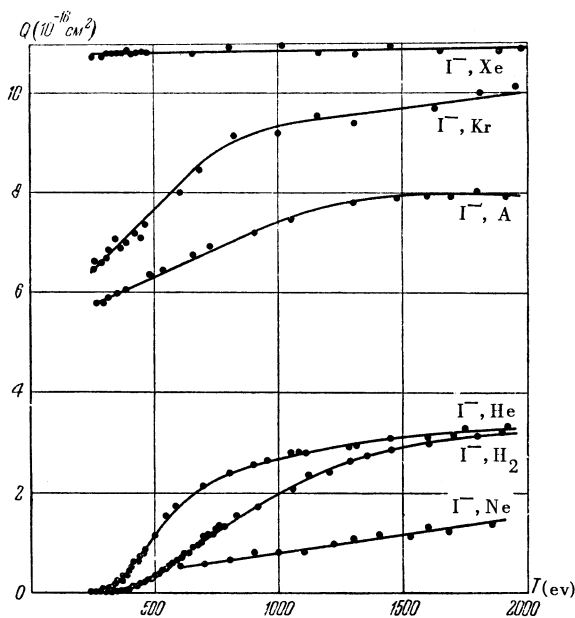
FIG. 7. Effective cross section for stripping of the negative ion I⁻ in inert gases and hydrogen as a function of ion energy.

He and H_2. We also verified the existence of a threshold for I⁻ ion stripping in He[1] and found a threshold for stripping of these ions in H_2 (Fig. 7). In those cases for which thresholds were found, the initial part of the curve $Q(T)$ was examined in detail. In Fig. 8 is shown, on an expanded scale, the function $Q(T)$ for stripping of I⁻ ions in He and H_2 for $T < 600$ ev. It is apparent from this figure that as the $Q(T)$ curves approach the threshold they are concave with respect to the ordinate axis, thus causing the threshold to be smeared out. We have determined the position of the threshold by square-law extrapolation of the initial part of the experimental curves. The uncertainty in the value of T because of the finite resolving power of the mass-analyzer is no greater
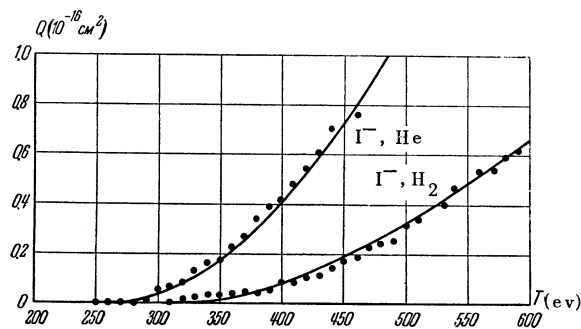

FIG. 8. Initial part of the curves $Q(T)$ for stripping of the negative ion I⁻ in He and H_2.

than ±2.5 percent. It follows from the conservation of energy and momentum that in the collision of a negative ion with a gas atom the stripping of the excess electrons can occur only with the consumption of kinetic energy W of the relative motion of the two particles $W = m_2 T / (m_1 + m_2)$, where m_1 is the mass of the ion and m_2 the mass of the gas atom. The energy threshold for stripping should occur at $W = W_0 = S$, where S is the binding energy of the excess electron in the negative ion. In the Table are shown the values T_0 and W_0 for threshold obtained from the present experimental curves and, for comparison, S, the electron bonding energy in the atoms Br and I. As can be seen from the Table, the observed thresholds lie above the thresholds determined from the conservation of energy and momentum. A similar result was obtained earlier in Ref. 4; there, however, an explanation for the origin of the experimentally observed threshold was proposed. It should be noted that the thresholds W_0 for ion stripping for Br⁻ and I⁻ in He lie higher than the thresholds for the

TABLE

Ion Atom	T_0, ev	W_0, ev	S, ev
Br⁻, He . . .	150	7.2	3.6 6
Br⁻, H_2	160	3.9	
I⁻, He	280	8.6	3.3 7
I⁻, H_2	320	5.0	

stripping of these same ions in H_2, whereas the thresholds determined by the conservation law should not depend on the gas. In the case of Br⁻, H_2, the observed threshold is only slightly higher than the threshold given by the conservation laws.

In the other (ion, atom) pairs, the thresholds could not be obtained in the present experiments, since these thresholds lie in the region of low ion energies. In the Cl⁻ in He case a rapid drop of the $Q(T)$ curve was observed in the region $T < 200$ ev (Fig. 5) indicating the proximity of the threshold.

Although the behavior of the curves $Q(T)$ and $Q(W)$ is not exactly the same in different (ion, atom) pairs, in the majority of the cases which were investigated these curves have common features. In Fig. 9 is presented the curve $Q(W)$ for the stripping of I⁻ in He which will be taken as typical. Close to the threshold the cross section increases rather rapidly with W, then the slope
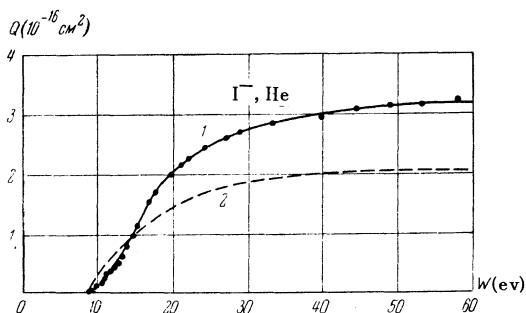
FIG. 9. The functions $Q(W)$ and $P(W)$ for stripping of the negative ion I^- in He. $1 - Q(W)$ determined experimentally; $2 - P(W)$ as calculated from Eq. (3).

falls off and the value of the cross section approaches a certain limiting value Q_m. In those cases in which the energy threshold for negative-ion stripping should be found at small ion energies (for example, collisions with Kr and Xe atoms) we were able to observe only the segment of the curve in which the value of Q was sensibly constant.

The present data reveal a correlation between the quantity Q_m and the atomic numbers of the ion and atom between which the collision takes place. For a given ion, for example Cl^- (Fig. 5), Q_m increases in going from He to Ne and then further from A, Kr and Xe. In collisions with the same atom, for example Xe, Q_m increases with increasing atomic number of the ion. A marked departure from this dependence was observed in the case of I^-, Ne for which the $Q(T)$ curve passes below the curve for I^- in the energy region 600 to 1,900 ev and does not saturate. This peculiarity was investigated further in supplementary measurements of the cross section for ion stripping of Sb^-
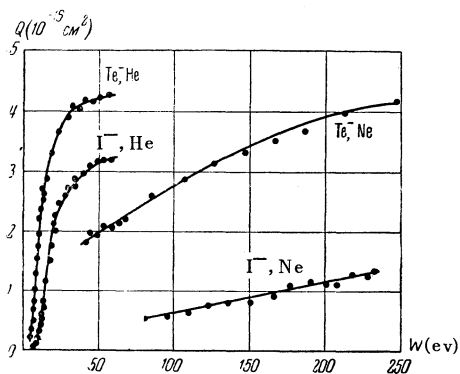

FIG. 10. Effective cross section for stripping of the negative ions I^- and Te^- in He and Ne as a function of the kinetic energy of the relative motion of the particles.

and Te^- (the mass of which is close to the mass of the I^- ion) in Ne and He. The same shape for the corresponding curves was found for these ions as for the I^- ion. In Fig. 10 are shown curves for $Q(W)$ in He and Ne for I^- and Te^- ions.

In a number of cases the present results can be compared with the data of Refs. 1-3. All the negative-ion stripping cross sections measured in the present work are smaller than the corresponding values given in Ref. 1. This difference may be explained by the more favorable experimental conditions in the present work as compared with Ref. 1. The general shape of the $Q(T)$ curves in the present work are similar to the curves presented in Refs. 2 and 3. The absolute value of Q, however, is considerably different from that given in Refs. 2 and 3. In the majority of cases the present values are smaller, and in only one pair, Cl^-,
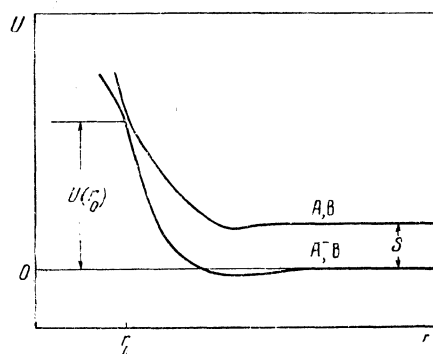

FIG. 11. Potential curves associated with the stripping of an electron from the negative ion A^- in slow collisions with the atom B.

Ne, are the values in good agreement with those given in Ref. 3.

5. DISCUSSION OF THE RESULTS

An approximate theoretical calculation (using the Born approximation) of the magnitude of the effective cross section for negative-ion stripping has been carried out in only one case, namely, the collision of H^- ions with He atoms[8]. For heavier atoms and ions a quantum-mechanical calculation of the cross sections would be prohibitively complicated. For cases in which the collision of an ion with a gas atom can be considered slow (the relative velocity of the approaching particles is smaller than the velocity of the electron in the negative ion), a qualitative description of the stripping process can be given in terms of a picture borrowed from the theory of diatomic molecules[1]. Two particles which approach and then

separate can be considered as a short-lived system
with a potential energy U which depends on the
distance r between the nuclei. Assume that the
curve $U_1(r)$ is associated with one system (a
negative ion A^- and atom B) and the curve $U_2(r)$
is associated with the other system (atom A and
atom B). At large r the curve $U_2(r)$ should lie
above the curve $U_1(r)$ by an amount equal to the
magnitude of S the electron bonding energy for
atom A (Fig. 11). At smaller r, U_1 and U_2 both
increase (because of the repulsive forces between
the particles) and if there is no interaction between
the systems (A^- and B) and (A, B and free elec-
tron), $U_1(r)$ and $U_2(r)$ can intersect at a certain
$r = r_0$. If there is an interaction no intersection
occurs; at the point r_0 the potential curves ap-
proach to within a minimum distance but become
separated again at smaller r. It is well known that
even for a finite value of $\Delta U = U_1 - U_2$ a transition
is possible from curve U_1 to curve U_2 at the ex-
pense of energy of the relative motion of the ap-
proaching particle. It has been shown theoreti-
cally[9-11] that the transition probability between
the potential curves is given by the expression

$$P = \exp \{ -4\pi (\Delta U)^2 / hv \tag{2}$$
$$\times (dU_1^0 / dr - dU_2^0 / dr) \},$$

where h is Planck's constant, v is the relative
velocity of the particles and dU_1^0 / dr and dU_2^0 / dr
are the values of the derivative dU/dr for each sys-
tem in the absence of any interaction.

If the curves U_1 and U_2 are such that U is a
strong function of r, then P will have a pronounced
maximum at $r = r_0$ and the transition probability
will be significant only at the critical distance
r_0. It is easy to show that in this case the transi-
tion probability P and the effective cross section
Q for the process associated with this transition
are related by the expression

$$Q = P(r_0, W) \pi r_0^2 (1 - U(r_0) / W), \tag{3}$$

where $U(r_0)$ is the energy of the relative particle
motion corresponding to the threshold for the process
in question. The validity of Eq. (3) in determining
the behavior of the probability for negative-ion
stripping P as a function of ion energy can be de-
termined from the experimental data on the function
$Q(W)$. This was done for stripping of I^- ions in
collision with He atoms [$U(r_0) = W_0$ is unknown
in this case] using the experimental curve given
in Fig. 9 and the function $P(W)$ given in this same

figure as curve 2.

If the potential curves $U_1(r)$ and $U_2(r)$ ap-
proach each other slowly, then the transition proba-
bility P should show only a small change over a
considerable range of r. Under these conditions the
notion of a "critical distance" has little signifi-
cance and the cross section Q should increase
comparatively slowly with W. The small values of
the cross sections and the difference in the form of
the $Q(W)$ curve from other cases in the stripping
of I^-, Te^- and Sb^- ions in neon may possibly be
explained on this basis.

It follows from Eq. (3) that for $W \gg U(r_0)$,
$Q = P(r_0 W) \pi r_0^2$. As has already been noted in
Sec. 4, in many (ion, atom) pairs virtually con-
stant values of the cross section Q were obtained
in a certain region of ion energy. This situation is
an indication that the probability for electron de-
tachment $P(r_0, W)$ is a weak function of W in these
regions.

The correlation which was observed between the
quantities Q_m and the atomic numbers of the ion
and atom (Sec. 4) may be reasonably explained by
the hypothesis that the value of r_0 is different for
different pairs and is connected with the "size"
of the colliding particles. It is well known that
the "radii" of atoms and negative ions increase
with increasing atomic number. This correlation
can, however, be disturbed by other factors which
influence the value of r_0; such factors might be
the polarizability of the interacting particles or
some peculiarity of the structure of the electron
shell. Thus, for example, for F^- ions the Q_m
in neon and A are almost identical (cf. Fig. 4).
It is impossible that for the pair F^-, Ne the value
of r_0 is unusually large because the configuration
of the electron shell of both particles is the same.

We may note the larger values of the cross sec-
tions Q_m for certain of the (ion, atom) pairs which
were investigated. For example, in Br^-, Xe, Q
$= 1.1 \times 10^{-15}$ cm^2 and I^-, A, $Q = 8 \times 10^{-16}$ cm^2.
These values are of the same order of magnitude
as the atomic cross sections determined from gas
kinetics. From this the conclusion may be drawn
that in the energy region for which Q reaches a
limiting value, the probability for negative-ion
stripping tends to unity as the impact distances
approach r_0.

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On the Theory of Atomic Semiconductors

A. G. SAMOILOVICH AND V. M. KONDRATENKO

Chernovitzkii State University

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On the basis of Vonsovskii's polar model of crystals several questions in the theory of absorption of light and the theory of photoconductivity in atomic semiconductors are examined, taking excitons into account. We consider terms of third order in the Hamiltonian, which determine the probability of different transitions among the elementary forms of excitation; this allows an examination of the kinetics of photoconductivity.

1. INTRODUCTION

THE polar model of crystals was proposed in 1934 by Shubin and Vonsovskii[1]. In this model the semiconductor in the normal state is considered as an ideal monocrystal, at the lattice sites of which are atoms with one valence s-electron. In the excited state sites can occur at which there are two electrons, and correspondingly, empty sites. The appearance of this type of excited state causes its electrical conductivity. Actually, on account of the translational symmetry, the states of the sites in which there are two electrons (or, respectively, none) can propagate through the crystal, which leads to the appearance of degenerate states. Since each state with definite sites occupied by two electrons, or with empty sites, is quasi-stationary, we obtain a whole band of energy levels. In this way the energy spectrum of an atomic semiconductor, from the point of view of the polar model, can even be continuous; but since the excitation of states with double and empty sites requires an expenditure of energy, the excitation of current states in the crystal requires a known activation energy, in spite of the existence of the continous spectrum. Thus, for

example, if one considers the exchange interaction between the electrons, the energy spectrum of the crystal has the form shown in Fig. 1, from which it can be seen that the conduction states occurring in the upper band, overlapping the lower one (due to the exchange interaction), require an activation energy for their excitation.
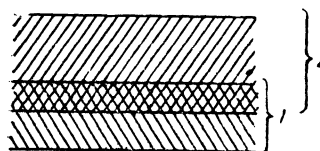

FIG. 1. *1* — Nonconduction band; *2* — Conduction band.

It is known[2] that an energy spectrum of this form (for weak excitation of the crystal) can be regarded as the energy spectrum of a system of non-interacting quasi-particles. In particular, the branch of the energy spectrum corresponding to the existence of conduction states in the crystal can be considered as the energy spectrum of a collection of positively charged (holes) and negatively charged (doublets) quasi-particles.