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Single electron capture by He^+ in Mg and Pb vapors

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The single-electron capture cross section σ_{10} for He^+ ions on magnesium and lead vapors has been measured in the energy range 9–40 keV. The results are not in agreement with the conventional near-adiabatic criterion of Massey regarding the energy at which the cross sections σ_{10} are maxima. This is thought to be due to pseudocrossing of potential-energy curves. Cross sections for Mg are found to be larger than for Pb in spite of the fact that both targets have very similar ionization potentials for their outer electrons.

I. INTRODUCTION

Available experimental data on nonresonant ($\Delta E \neq 0$) electron transfer collisions between ions and atoms have been discussed mainly on the basis of Massey's criterion[1] and the Stueckelberg-Landau-Zener theory[2] (SLZ). Massey's rule predicts that the maximum cross section occurs at a relative velocity V_m given by

$$V_m \simeq a\Delta E/h, \tag{1}$$

where ΔE is the average energy defect of the reaction, h is Planck's constant, and a is a length of the order of atomic dimensions, representing a mean collision distance.

Hasted,[3] examining experimental data, found that Eq. (1) is a fairly good prediction for a large number of single-electron capture processes if a is given the value 7 Å and ΔE is replaced by ΔE_∞, the energy defect at infinite internuclear separation. Hasted and Lee[4] and Williams[5] have found that better agreement with experiment is obtained if the energy defect ΔE_∞ is corrected by the average polarization energy in the collision, although they have made the crude assumption that polarization forces dominate at internuclear distances where the electron clouds of the colliding systems partially overlap. These approximations are referred to as the conventional Massey criterion. In cases where there is an isolated crossing (in a diabatic sense) of the potential-energy curves of the initial and final states describing the interaction, it is known that the conventional Massey criterion does not hold[6] and the collisions can be treated by the two-state SLZ theory, which predicts a broad maximum in the cross section as a function of energy,

$$\sigma_{\max}^{\rm SLZ} = 0.452\pi R_X^2, \tag{2}$$

where R_X is the internuclear distance at the diabatic crossing.

Apart from the curve-crossing case, no general rule has been given for the dependence of the maximum value of the cross sections on characteristic parameters of the collision partners. It is found, however, that the maximum cross sections are larger, for a given collision pair, if the state of the products is such as to give a closer energy balance.

Earlier investigations on electron capture by helium ions have been limited mainly to collisions on noble gases, alkalis, and some molecules. It is believed that studies on targets of different electronic configurations can help to determine the conditions of applicability of the conventional Massey criterion and broaden our knowledge of the relevant factors influencing the shape and magnitude of the cross sections versus energy curves.

In this work, we report measurements on the single-electron capture cross section for He^+ ions on Mg and Pb vapors in the energy range 9–40 keV. These targets were chosen because their first two ionization potentials are very similar; hence the effect of other factors, like total angular momentum or spin multiplicity, could show up more easily. A preliminary account of this work was given in Ref. 7.

II. APPARATUS

A schematic drawing of the experimental apparatus is shown in Fig. 1. He^+ ions are produced by a radio-frequency ion source. They are extracted, focused, and accelerated electrostatically, and sorted by a double-focusing magnetic analyzer through a 20° deflection. After collimation, the beam enters the interaction region. A pair of deflecting plates can be used to sweep out the charge components of the beam. This allows us to determine the contribution to the measured signals from unwanted fast neutrals formed by charge-changing collisions in the residual vacuum of the apparatus before the interaction chamber, as will

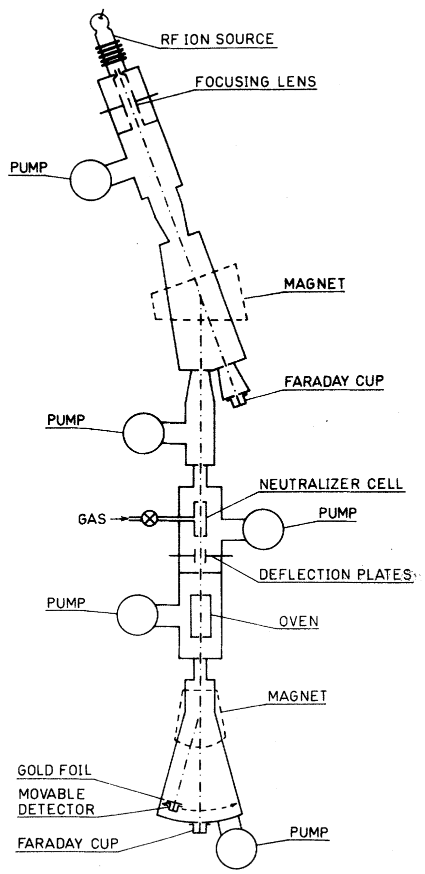

FIG. 1. Experimental equipment.

be shown later.

The interaction region is shown in Fig. 2. The stainless-steel tube A, 81 mm in length and 20 mm in internal diameter, contains the metal to be evaporated, whose purity was checked by spectroscopical analysis to be greater than 99.999% for Pb and 99.8% for Mg. The beam enters and leaves the cell through holes which are 1.75 and 2 mm in diameter, respectively. The holes present a sharp edge to the incoming beam so as to minimize scattering and/or charge-changing collisions on them.

The target cell is placed with a 0.1-mm clearance inside a quartz tube around which is wound a nichrome ribbon heater B. The stainless-steel cylinder D acts as a heat shield, and is closed at its ends by two plates that support the quartz tube. The alumina rods C passing through holes in these plates are fixed to the vacuum chamber, thus giving mechanical support to the oven.

The oven can be operated up to 800 °C with an input power of 100 W. Water flows through a copper tube F wound around the outside of the vacuum chamber. The temperature of the vapors is measured with a chromel P-alumel thermocouple E placed in a thin-walled stainless-steel finger extending inside the oven. The thermocouple was calibrated *in situ* against the melting point of lead at 600.7 °K, with an uncertainty of ±1 °K. The output of the thermocouple, whose cold junction was kept at 0 °C, was read with a digital voltmeter with 1-μV resolution.

The different beam components that emerge from the interaction region as a result of charge-changing collisions with the metal vapors are magnetically separated and collected in a movable secondary-emission-type detector with a thin (~ 200 $\mu g/cm^2$) gold foil mounted at its entrance.[8] Since this foil charge equilibrates each beam component, the detector response is charge independent.

Typical beam currents were 10^{-7} A at 40 keV and 10^{-10} A at 9 keV, while the neutral currents resulting from the interaction ranged from 10^{-9} to 10^{-13} A. These currents were measured with a Cary 401 vibrating-reed electrometer.

Vacuum is maintained in the equipment by means of diffusion pumps with their corresponding cold

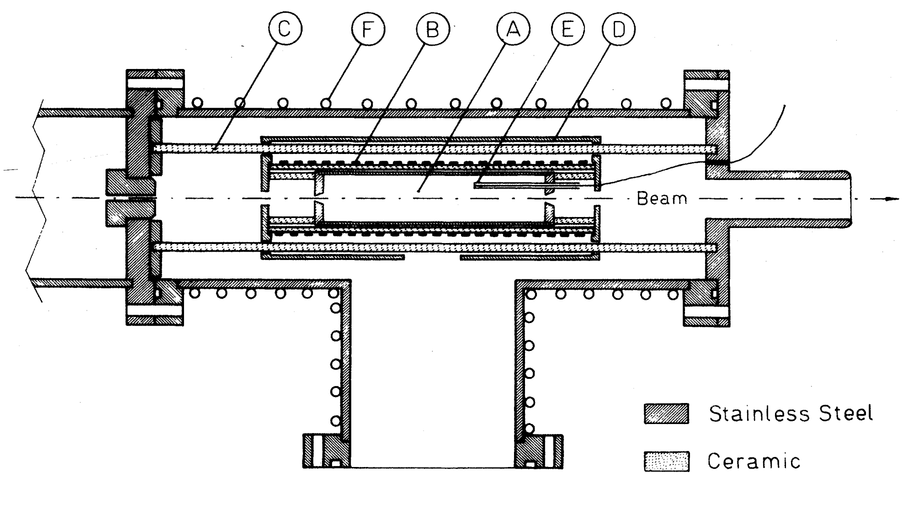

FIG. 2. Interaction region. A: oven; B: heater; C: supporting rods; D: radiation shield; E: thermocouple; F: water cooling.

traps at liquid-nitrogen temperature. The background pressures ranged from 4 to 9×10^{-7} Torr with the oven at room temperature. The pressure in the region outside the target cell was of the order of 5×10^{-6} Torr with the oven at 500 °C.

III. EXPERIMENTAL PROCEDURE

The dependence of the charge fractions F_j on target thickness Π can be described by the system of differential equations[9]

$$\frac{dF_j}{d\Pi} = -F_j \sum_{k\neq j} \sigma_{jk} + \sum_{i\neq j} F_i \sigma_{ij} \quad \text{with} \sum_j F_j = 1\,, \quad (3)$$

where σ_{if} are the cross sections for collisions which change the charge of the projectile from i to f. The fractions F_j are related to the beam currents I_j by

$$F_j = \frac{I_j}{\sum_k I_k} = \frac{I_j}{I_T}\,. \quad (4)$$

For an incident beam consisting only of single charged ions, Eq. (3) can be easily integrated, giving, in the limit of low-Π values, for the neutral component

$$F_0 = \sigma_{10}\Pi + b\Pi^2 + \ldots, \quad (5)$$

where b depends on sums of products of cross sections σ_{ij}. In this way, by observing the initial linear growth, the single-electron capture cross section σ_{10} can be determined.

A complication arises if the incident beam has a fraction G_0 of neutral atoms, and if there is background gas in the interaction cell, with thickness Π'. In this case, and considering $G_0 \ll 1$,[10]

$$F_0 = G_0 + \left(\sigma'_{10} - G_0 \sum_{j\neq 0} \sigma'_{0j}\right)\Pi' + \left(\sigma_{10} - G_0 \sum_{j\neq 0} \sigma_{0j}\right)\Pi + \ldots, \quad (6)$$

where the cross sections σ'_{ij} are for collisions with the background gas.

The unwanted contribution of G_0 may be subtracted with the help of the deflecting plates located before the oven in the following way. Let I_0^E be the neutral current measured with the deflection field on, I_0 that with this field off, and I_T the total current; then

$$I_0 = I_T G_0 + I_T\left(\sigma'_{10} - G_0 \sum_{j\neq 0} \sigma'_{0j}\right)\Pi' + I_T\left(\sigma_{10} - G_0 \sum_{j\neq 0} \sigma_{0j}\right)\Pi + \ldots, \quad (7)$$

$$I_0^E = I_T G_0 - I_T G_0 \sum_{j\neq 0} \sigma'_{0j}\Pi' - I_T G_0 \sum_{j\neq 0} \sigma_{0j}\Pi + \ldots, \quad (8)$$

and then

$$F_0^* = \frac{I_0 - I_0^E}{I_T} = \sigma'_{10}\Pi' + \sigma_{10}\Pi + \ldots. \quad (9)$$

Therefore, measuring I_0, I_0^E, and I_T, we can from Eq. (9) determine the cross section σ_{10} from the initial slope of $F_0^*(\Pi)$, the intercept at $\Pi = 0$ giving the contribution of collisions with the background gas.

The target thickness Π is given by the effective length of the target cell, the pressure of the vapor, and its temperature. The effective length was calculated from the geometrical length of the cell, a correction being made to account for the Knudsen effusion through the holes. This correction amounted to less than 3%. The vapor pressure was derived from its temperature with the help of data given by Hultgren *et al.*[11] for magnesium and by Kim and Cosgarea[12] for lead. The functional dependences $P(T)$ used were

$$\log_{10} P = 8.659 - 7516.87/T \quad (\text{for Mg}),$$
$$\log_{10} P = 11.6091 - (10186/T) - 1.1071 \log_{10} T \quad (\text{for Pb}), \quad (10)$$

where the pressure P is in Torr and the temperature T is in degrees kelvin. In order to use Eq. (9) to determine σ_{10}, it must be ascertained that the background contribution remains constant, i.e., that Π' does not vary with temperature. To achieve this, the oven was thoroughly outgassed at high temperatures during several hours before taking data. The fact that the neutral fractions measured have the functional dependence of Eq. (9), as shown in Fig. 3, indicates that Π' remained constant or varied by a negligible amount.

To avoid hysteresis effects[13] between data taken with increasing and decreasing temperatures that would result from nonequilibrium conditions, the oven was allowed to stabilize at the desired temperature for at least 20 min. No hysteresis phenomena were thus observed within experimental error, as shown in Fig. 3.

Each point in the $F_0(\Pi)$ curves results from at least four independent measurements (usually eight) of both the neutral and the charged fractions emergent from the oven, at a temperature which was kept constant to within 0.25 °K. The accelerator voltage was determined to within 0.4% from the current drained through a calibrated chain resistance. To obtain the value of the beam energy the extraction voltage applied to the ion source

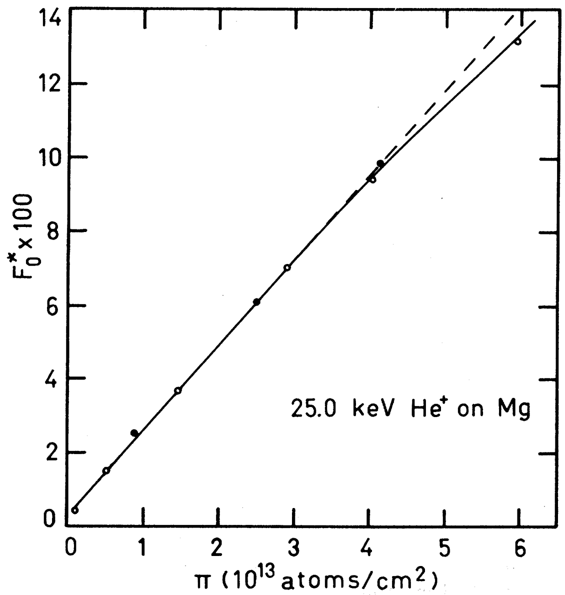

FIG. 3. F_0^* vs Π for 25-keV He^+ on Mg. The points ○ and ● indicate data taken with increasing and decreasing temperature, respectively.

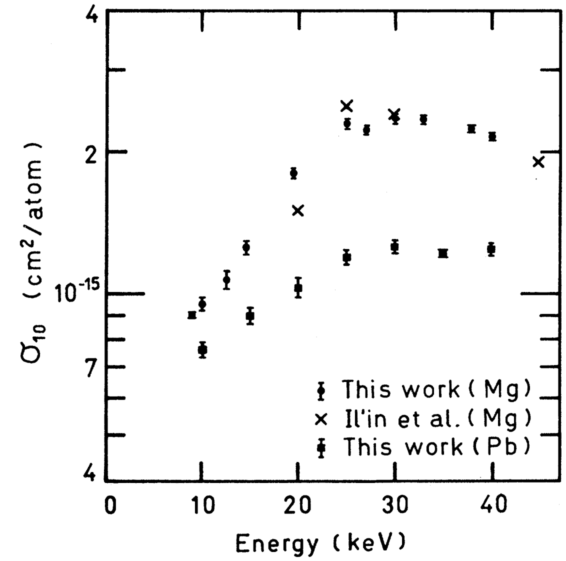

FIG. 4. Single-electron capture cross section σ_{10} for He^+ on Mg and Pb. Also shown are the results of Il'in *et al.* (Ref. 14) for Mg.

was added to the acceleration voltage. The plasma potential in the radio-frequency ion source for normal operating conditions was found to be negligible compared to the total voltage.

The values of the measured neutral fractions were not affected, within experimental errors, when changing the exit aperture of the target cell from 2 to 3 mm diam. This suggests that errors due to large-angle scattering are negligible.

TABLE I. Cross sections σ_{10} for He^+ on Mg and Pb in units of 10^{-16} cm²/atom. Not included are uncertainties in the vapor pressure data, 20% for Mg and 9% for Pb.

Energy (keV)	σ_{10} (Mg)	σ_{10} (Pb)
9.00	8.95 ± 0.26	...
10.00	9.47 ± 0.33	7.64 ± 0.30
12.50	10.70 ± 0.40	...
14.75	12.55 ± 0.43	...
15.00	...	8.99 ± 0.26
19.56	18.20 ± 0.33	...
20.00	...	10.30 ± 0.48
25.00	22.99 ± 0.33	11.93 ± 0.36
27.00	22.25 ± 0.14	...
30.00	23.48 ± 0.39	12.55 ± 0.39
33.00	23.57 ± 0.51	...
35.00	...	12.22 ± 0.07
38.00	22.34 ± 0.41	...
40.00	21.81 ± 0.20	12.39 ± 0.28

IV. RESULTS AND DISCUSSION

The measured values of σ_{10} for He^+ on magnesium and lead vapors are presented in Table I and plotted in Fig. 4, together with the results of Il'in *et al.*[14] for He on Mg, which are in good agreement with ours. Statistical errors, arising mainly from uncertainties in the initial slopes of the curves $F_0^*(\Pi)$, were always smaller than 5%, and on the average are 2.5%. Not included in Table I and the graphs are uncertainties in the vapor-pressure data which are quoted as 20% for Mg[11, 15] and 9% for Pb.[16] These uncertainties must be added to the statistical errors mentioned above in order to obtain the absolute accuracy of the experimental results.

Lockwood[17] has studied the electron capture process for He^+ in N_2. He finds that σ_{10} depends on the type of ion source used to produce He^+, which he attributes to different populations of metastable $He^+(2^2S)$. In our experiment, the electric fields used to extract, focus, and accelerate the beam, and the magnetic field in the magnetic analyzer, were such that only a fraction of 10^{-6} would not be Stark quenched to the short-lived ($\sim 10^{-10}$ sec) He^+ (2^2P) state. Thus, the influence of those states in our data should be negligible.

The energies E_m at which the cross sections reported in this work are maxima (~30 keV) do not agree with those resulting from the application of the conventional Massey criterion for capture into the ground (~175 keV) or first excited states (4–9 keV) of helium. It must be pointed out that an agreement could be found if we assume electron capture to highly excited states of He, but this would not explain the large measured values of the cross sections.

Diabatic crossings of the potential-energy curves at large internuclear distances, which occur if an electron is captured to levels with $n \geq 2$ of the He atom, may explain the results. These crossings occur because of the much larger polarizabilities of the excited states of He as compared to those of the ground-state target atoms, polarization forces being predominant at large internuclear distances. However, since many crossings are involved, and these may not be isolated, the SLZ formula (2) cannot be directly applied, for it is a two-state approximation to the problem. The reason for σ_{10} being larger on Mg than on Pb, in spite of the fact that both targets have very similar ionization potentials, is not known at present.

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