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Semiempirical Calculations of the He* $(2^{3}S \text{ and } 2^{1}S)$ + Ar Ionization Total Cross Sections^{*}

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The ionization (associative and Penning) total cross sections for $He^*(2^3S) + Ar$ and $He^*(2^{1}S) + Ar$ have been calculated from 10^{-3} to 10^4 eV and compared with experimental data. The classical calculations used interaction potentials that agreed with *ab initio* theoretical calculations for the repulsive wall, possessed the correct long-range energy dependence, and were in agreement with glory-scattering experiments. The coupling width was an exponential with its free parameter determined by the thermal-energy ionization cross section for each system. The calculations were then extended over a wide range of energies. It was found that as the collision energy increased at the very low energies, the cross sections decreased until about 0.1 eV where they began slowly to rise. As the collision energy is further increased, the cross sections reach a maximum of about 15 Å^2 around 10 eV, and then decrease again at higher energies. The cross sections predicted by the high-energy results, 100-1500 eV, are compared with the data of Moseley, Peterson, Lorents, and Hollstein (preceding paper) and are found to agree within 20% and to have a similar energy dependence.

INTRODUCTION

The energy dependence of the ionization (associative and Penning) total cross sections for Ne* $(2p^{5}3s^{3}P)$ + Ar were first reported by Micha, Tang, and Muschlitz.¹ These cross sections stimulated interest to try to understand the unexpected energy dependence that was observed.^{1,2} In the energy range investigated, the measured cross sections were found first to decrease with increasing energy, then to pass through a minimum around 0.05 eV, and finally to rise gently at the higher energies ($E \gtrsim 0.05$ eV). In a previous paper,² this author was able to explain this phenomenon and theoretically reproduce these low-energy data by using available potential curves for the Ne* + Ar interaction and a realistic exponential form for the coupling to the continuum. Basically, this behavior occurs because of the exponentially increasing nature of the coupling width as the distance of closest approach decreases with increasing energy. Furthermore, these calculations led to the prediction that the cross sections would rise to a maximum of about 10 Å² at around 10 eV and then slowly decrease with increasing energy. There were, however, at that time no high-energy

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measurements on this system to test these predictions.

Work was therefore begun on the $\text{He}^{*}(2^{3}S) + \text{Ar}$ and $He^{*}(2^{1}S) + Ar$ systems since there exist several measurements at thermal energies³⁻⁸ and also recent measurements in the 100-1500-eV region by Moseley *et al.* (preceding paper).⁹ The energy dependences of the low-energy ionization cross sections, such as those measured by Tang, Marcus, and Muschlitz for the Ne*+noble-gas systems, ¹⁰ are not available for the He*(2s) + Ar systems, and hence the opportunity exists for theory to predict the energy dependence of the low-energy cross sections. The high-energy Ne*+Ar cross sections have also been measured⁹ and are compared with the previous theoretical prediction² in the preceding paper. Although the theory underestimates the cross sections somewhat, the agreement is satisfactory, particularly with regard to the energy dependence.

For the present $\operatorname{He}^{*}(2s) + \operatorname{Ar}$ studies, as in the Ne* + Ar study, the interaction potential was obtained from ab initio calculations and elastic-scattering information. The coupling width was again normalized to the thermal-energy cross section. The cross-section calculations were then extended over five orders of magnitude in energy and compared with the high-energy data to test the validity of the theoretical procedure used here and in the previous $Ne^* + Ar$ calculations. We find that at high energies the calculated cross sections are within 20% of the experimental results and also possess a similar energy dependence. At low energies, the computed cross sections exhibit the same type of energy dependence that was found in the Ne* + Ar system and has been more recently observed in the Ne* + Kr and Xe systems.¹⁰

It appears that there will be a common energy dependence to the cross sections for most of the Ne* $(2p \, {}^{5}3s \, {}^{3}P)$ + noble-gas and He* $(2 \, {}^{3}s \, {}^{a}nd \, 2 \, {}^{1}s)$ + noble-gas systems. That is, with increasing collision energy, the cross sections at low energies will first decrease to a minimum around 0.1 eV, then increase to a maximum of about 15 Å² at 10 eV, and finally decrease again at higher energies.

Since the experimental measurements are most conveniently made either at thermal energies $E \leq 0.1$ eV or at high energies $E \gtrsim 100$ eV the theoretical technique provides a means for interpolating the cross sections into the interesting energy region of 0.1 to 100 eV.

THEORY

The ionization total cross sections were calculated as a function of energy for the sum of the Penning ionization process

$$He^* + Ar \rightarrow He + Ar^* + e^-$$
(1)

and of the associative ionization process

$$He^* + Ar \rightarrow HeAr^* + e^-, \qquad (2)$$

where He^{*} is $He(2^{3}S)$ or $He(2^{1}S)$. The calculations were performed using the classical formulas found in a paper by Miller.¹¹ Here, the ionization cross section is

$$Q = 2\pi\alpha \int_0^\infty db \ b \ P(b) , \qquad (3)$$

where the ionization probability is given by

$$P(b) = 1 - \exp\left(-2\int \frac{dR\Gamma(R)}{\hbar v_b(R)}\right).$$
(4)

The radial velocity is calculated from

$$v_b(R) = v_0 \left[1 - V_0(R) / E - b^2 / R^2 \right]^{1/2}, \qquad (5)$$

where v_0 is the incident velocity and b is the impact parameter. In Eq. (3), α is the statistical weight factor that is unity for these systems. The interaction potential $V_0(R)$ is that of the metastable atom in the field of the neutral atom. The coupling to the continuum is provided by the width $\Gamma(R)$, which in this case we have assumed to be of exponential form. This form is similar to the shapes of $\Gamma(R)$ calculated by Miller, Slocomb, and Schaefer,¹² Bell,¹³ and Fujii *et al.*¹⁴ for the He* + H system and is similar to that deduced from He* + Na experimental data by Hotop and Niehaus.¹⁵ We have restricted the coupling width to the simple exponential form

$$\Gamma(R) = e^{-R/B} . \tag{6}$$

The functional dependence of the interatomic potential $V_0(R)$ was chosen so that it possessed a realistic shielded Coulomb form at small separations and the correct $-C_{ab}/R^6$ dependence at large separations. The long-range R^{-6} term is exponentially damped at small separations by using the shielding length of the outer Ar core. This removes unrealistic dominance of the attractive terms at small separations. For small interatomic distances, the potential parameters are chosen so that the potential reproduced the Thomas-Fermi-Dirac calculations by Abrahamson¹⁶ for Li + Ar. Because of the large range of collision energies studied here, it was found that the commonly used Lennard-Jones or Morse potentials were inadequate. The Lennard-Jones potential is much too repulsive at small separations while the Morse potential has too soft a repulsive core. We chose instead to use a form similar to that employed by Smith et al.¹⁷ for He⁺ + Ar scattering, employing the appropriate change in the long-range forces. The potential is parametrized by (all quantities are in atomic units)

 $V(R) = (2/R)(8e^{-R/C_M} + 8e^{-R/C_L} + 2e^{-R/C_K}) - (C_{ab}/R^6)$

(7)

$$\times \left[1 - e^{-X} \left(1 + X + \frac{1}{2}X^{2} + \frac{1}{6}X^{3} + \frac{1}{24}X^{4} + \frac{1}{120}X^{5}\right)\right],$$

where

$$X = R/C_M . (8)$$

For both the He* $(2^{3}S)$ and He* $(2^{1}S)$ + Ar scattering, we set $C_M = 0.98$, $C_L = 0.18$, and $C_K = 0.057$. C_L and C_K are the same screening lengths as given in Table I of Ref. 17. C_M was varied slightly from 0.93 so that the potential minimum for the $He^{(2^3S)}$ case would agree with the glory-scattering results of Rothe, Neynaber, and Trujillo.¹⁸ The long-range C_{ab} coefficients were obtained from the work of Bell, Dalgarno, and Kingston¹⁹ with C_{ab} for He*($2^{3}S$) + Ar being set at 226 a.u. and C_{ab} for $He^{*}(2^{1}S) + Ar$ being set at 325 a.u. The potentialwell parameters used in these calculations are given in Table I and the repulsive portion of the potential is displayed in Fig. 1. For the $He^{(2^3S)}$ + Ar glory scattering, the quantity derived from experiment is the product $\epsilon \times \sigma$, ϵ being the well depth and σ being the zero of the potential. The potential used for the $He^{*}(2^{3}S)$ scattering agrees to better than 1% with the value of $\epsilon \times \sigma$ obtained by



FIG. 1. Solid lines: the repulsive region of the potentials used in these calculations. The dashed line is from the Thomas-Fermi-Dirac calculations on Li +Ar by Abrahamson (Ref. 16).

Rothe et al.¹⁸

The value of the screening length used for the coupling width, Eq. (6), varies with the experimental thermal-energy cross section that is used for the normalization of the theoretical cross sections. The values of *B* will be given in the discussion of the next section.

RESULTS AND DISCUSSION

$He^{*}(2^{3}S) + Ar$

The results for the $\text{He}^*(2^{3}S) + \text{Ar}$ calculations are presented in Fig. 2. The width, Eq. (6), was normalized to the thermal-energy result of Sholette and Muschlitz.⁴ The exponential constant *B* in the width was found to be equal to $0.667a_0$. Several other experimental values^{3,5-8} for the thermal-energy cross section are shown in Fig. 2 along with the energy dependence of the low-energy cross sections measured by Jones and Robertson.⁶ All measurements are in reasonable agreement with one another.

By using an interaction potential obtained from elastic-scattering data¹⁸ and theoretical calculations¹⁶ and a width normalized to a thermal-energy cross section, the ionization total cross sections can be computed at several other energies. To test this theoretical extrapolation, we have compared the theoretical results with the high-energy cross sections of Moseley et al.⁹ Since the highenergy experimental results are four orders of magnitude in energy away from the thermal-energy normalization, this comparison provides a severe test of the theoretical procedure. The high-energy cross sections were measured by two separate methods that are completely described in the preceding paper.⁹ In brief review, however, some of the high-energy cross sections are optical measurements of the deactivation of $He^{*}(2^{3}S)$ by Ar and are depicted by the open circles in Fig. 2. The open squares, in turn, are the deactivation cross sections measured by detecting the slow ions produced when an unknown *mixture* of $He^{(2^3S)}$ and $\text{He}^{*}(2^{1}S)$ collides with Ar. Since the optical techniques show that the deactivation cross sections for $He^{(2^3S)}$ and $He^{(2^1S)}$ on Ar are almost the same and are similar to the cross sections measured by detecting the slow ions, the ionization cross sections for $\text{He}^{*}(2^{3}S) + \text{Ar}$ should be very close to those shown and provide a good test of the theoretical method. Furthermore, it can be argued that for deactivation of $He^{*}(2^{3}S)$, the only plausible mechanism is via ionization since excitation of the He* to the $2^{3}P$ state is not followed by an allowed transition to the ground state.

The theoretical cross sections are found to be approximately 15% below the high-energy experimental results. The energy dependence of the cross sections is also found to be in reasonable agreement. If the width were normalized to any of the other thermal-energy cross sections, there would be very little change at the higher energies [this will be seen in the next subsection on the $He^*(2^{1}S) + Ar$ cross sections]. Therefore, any difference between theory and experiment is probably caused by an inaccurate interaction potential or most likely by the simple form used for the width, Eq. (6). It does not seem appropriate, moreover, to vary the functional form of the width to reproduce the high-energy data because of the uncertainties in the interaction potential and the experimental data.

The theoretical low-energy cross sections have an energy dependence similar to that observed in the Ne* $({}^{3}P)$ + Ar, Kr, and Xe systems.^{1,10} We predict that the He* $(2{}^{3}S)$ + Ar, Kr, and Xe cross sections will also have an energy dependence similar to that shown in Fig. 2.

In light of the assumptions made in the theoretical calculations, it is estimated that the accuracy of the theoretical cross sections is $\pm 50\%$. By comparing the theoretical cross sections with the high-energy measurements, it is expected that the theoretical ones will be slightly below the true values at the higher energies.

$He^{*}(2^{1}S) + Ar$

The $\text{He}^*(2^1S)$ + Ar calculations proved to be a more difficult problem in that the thermal-energy cross sections used for the normalization of the width were not consistent with one another. Here,

Sholette and Muschlitz⁴ find a value for the ionization cross section of 7.6 Å²±7% and MacLennan⁵ finds 9.0 Å²±30%, while Schmeltekopf and Fehsenfeld⁷ find 16.4 Å²±30%. We have therefore used the two end values of the thermal-energy cross section to normalize the width and then have compared the results at high energies. Using the cross section of Schmeltekopf and Fehsenfeld,⁷ the value of *B* in the width is found to be 0.680 a_0 . From the data of Sholette and Muschlitz,⁴ we find *B* to be 0.629 a_0 .

The calculated cross sections for the two different normalizations are shown in Fig. 3. Even though the theoretical cross sections were normalized to thermal-energy results that differ by more than a factor of 2, at the high energies, 100-1000 eV, the two calculations differ by less than 25%. However, because of uncertainties in the interaction potential, the functional form for the width, and the high-energy experimental results, it is impossible to ascertain which thermal-energy result is most accurate.

The high-energy data displayed in Fig. 3 are taken from the preceding paper.⁹ The open circles are the deactivation cross sections for the He*($2^{1}S$) + Ar reaction as obtained by optical methods. The open squares are the deactivation cross sections obtained by detecting the slow ions produced when a mixture of He*($2^{1}S$) and He*($2^{3}S$) collides with Ar. To complicate this reaction further, it is probable that the deactivation cross sections do not represent only the ionization reaction. Unlike the He*($2^{3}S$) + Ar case, it is possible for the



FIG. 2. He* (2^3S) + Ar ionization total cross sections. The data at thermal energy are: MacLennan, Ref. 5, 9.0 Å² ±30%; Sholette and Muschlitz, Ref. 4, 7.6 Å² ±7%; Bolden, Hemsworth, Shaw, and Twiddy, Ref. 8, 7.0 Å² ±20%; Benton, Ferguson, Matsen, and Robertson, Ref. 3, 6.6 Å² ±20%; Schemeltekopf and Fehsenfeld, Ref. 7, 5.34 Å² ±30%; and the error bars are from Jones and Robertson, Ref. 6, who report 7.7 Å² ±9%. Their thermal-energy result is displaced slightly for clarity. The high-energy results are those of Moseley, Peterson, Lorents, and Hollstein, Ref. 9. The solid line is the theoretical calculation.

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FIG. 3. He*(2¹S) + Ar ionization total cross sections. The data at thermal energy are: Schmeltekopf and Fehsenfeld, Ref. 7, 16.4 Å² $\pm 30\%$; MacLennan, Ref. 5, 9.0 Å² $\pm 30\%$; and Sholette and Muschlitz, Ref. 4, who report 7.6 Å² $\pm 7\%$. The high-energy results are those of Moseley, Peterson, Lorents, and Hollstein, Ref. 9. The solid lines are the theoretical calculations.

He*(2¹S), on collision with Ar, to be deactivated by first being excited to the nearby 2¹P state and then transferring via an allowed transition to the ground state. Therefore, the experimental deactivation cross sections can only provide an upper limit to the ionization cross sections. It is therefore difficult to provide error limits for the theoretical cross sections since there is such a large discrepancy in the thermal measurements and since the high-energy measurements cannot be considered strictly ionization cross sections. We can only estimate, assuming that one or the other thermal-energy cross section is found to be correct, that the theoretical curve through it will be accurate to $\pm 50\%$.

CONCLUDING REMARKS

Precautionary statements should be made about the difficulties of determining the functionality of $\Gamma(R)$ from experimental data. These difficulties arise because of uncertainties in the elastic intermolecular potential and in the experimental data. For the systems studied here, the experimental cross sections measured at thermal energies differ appreciably among one another, and at the higher energies the experimental cross sections can only be considered an upper limit on the ionization total cross section. Therefore, the use of a simple exponential form for $\Gamma(R)$, Eq. (6), cannot be justified by the experimental data alone but must also be supported by theoretical calculations.¹²⁻¹⁴

The experimental data, however, can demand specific requirements on $\Gamma(R)$. The easiest way to understand the requirements on $\Gamma(R)$ is to utilize the small transition-probability formula for the ionization cross section given by Miller.¹¹ Here, the cross section is given by the simple integral

$$Q = \frac{4\pi}{\hbar v} \int_{R_0}^{\infty} dR R^2 \Gamma(R) [1 - V(R)/E]^{1/2} , \qquad (9)$$

where R_0 is the classical turning point for b = 0. Since it is reasonable to expect $\Gamma(R)$ to decrease with increasing R, the main contribution to the above integral comes from the region of R just outside R_0 . To the lowest order then, the cross section will be roughly proportional to the quantity $R_0^2\Gamma(R_0)/v$.

For the case at hand, we can now understand why the cross sections, with increasing collision energy, rise to a maximum at about 10 eV and then decrease again at the higher energies. The functional form of $\Gamma(R)$ is required to be such that the quantity $R_0^2 \Gamma(R_0) / v$ increases as the collision energy increases from thermal energy to about 10 eV and then decreases at the higher energies. If we review the energy dependence of the classical turning point R_0 , we find that in increasing the energy above thermal energy, R_0 decreases relatively rapidly so that with a realistic form for $\Gamma(R)$, such as an exponential, the quantity $R_0^2 \Gamma(R_0)/v$ will increase. At the higher energies, however, R_0 will remain relatively constant since we are now on the steeply repulsive wall of the potential. For these energies $R_0^2 \Gamma(R_0)/v$ can only decrease, and we can expect the same behavior from the cross sections.

Other functional forms for $\Gamma(R)$ can certainly meet the above requirments and explain the data. In fact, a better fit to the data could be made by using a two parameter form for $\Gamma(R)$ such as $\Gamma(R) = A e^{-R/B}$. However, this complication is unjustified since a better fit of the data would not imply a more accurate $\Gamma(R)$. An inversion to $\Gamma(R)$ has to be based on knowing accurately the elastic intermolecular potential and the experimental cross sections. Neither of these requirements exist here, so any $\Gamma(R)$ obtained certainly could not be considered unique. *Supported by the Office of Naval Research.

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Optical Excitation and Polarization in Low-Energy Collisions of $He^+ + H_2^{\dagger}$

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Optical studies of spectral lines produced in collisions of He⁺ + H₂ have been made at energies below 700 eV. Lines of atomic hydrogen and helium are the only features observed. Emission cross sections have been measured for Lyman- β , Balmer- α , Balmer- β , and for the following He I lines: 5876 Å ($3^{3}D \rightarrow 2^{3}P$), 6678 Å ($3^{1}D \rightarrow 2^{1}P$), 4471 Å ($4^{3}D \rightarrow 2^{3}P$). Emission cross sections are also listed at certain selected energies for all spectral lines which have been observed. Polarization measurements have been performed on the Balmer- α , the Balmer- β , and the visible helium lines. Only the Balmer- α line exhibits significant polarization, and the results have been interpreted as indicating that the excited hydrogen atoms dissociate primarily in a direction perpendicular to the ion beam.

I. INTRODUCTION

Stedeford and Hasted¹ have measured the total cross section for charge transfer in He⁺+H₂ collisions and have tentatively postulated that the products are helium atoms in the ground state and H_2^+ ions in the lowest antibonding state. This state of the molecular ion, $2\rho\sigma_u$, dissociates into a proton and a hydrogen atom in the ground state. However, Dunn, Geballe, and Pretzer² have concluded from the shape and magnitude of the excitation cross section for producing the Lyman- α line by these collisions that the charge transfer at energies below 2.5 keV takes place almost 50% of the time into states of H_2^+ which then dissociate into a proton and a hydrogen atom in the $2^{2}P$ state. Gusev et al.³ have shown that the Balmer- α and $-\beta$ lines are also produced rather strongly by He⁺+H₂ collisions. The implications of these optical observations are that most, if not all, of the charge exchange at low energies takes place into states which ultimately result in excited hydrogen atoms. In the present work we have extended the spectral observations in order to obtain a more complete picture of the excited atomic states which may result from collisions of the He^++H_2 system at energies below 700 eV. Both the Lyman and Balmer series as well as lines of He_I have been investigated. The emission cross sections for several lines have been measured from threshold to 700 eV, and values are tabulated at a few selected energies for all spectral features which have been observed.

The polarizations of the Balmer- α and $-\beta$ lines have also been measured as a function of the energy of He⁺ ions.⁴ The latter appears to be unpolarized, but the former exhibits a polarization which is highly dependent upon the ion energy, being a maximum of about 12% at 50 eV. Van Brunt and Zare⁵ have pointed out that both unequal populations of the magnetic substates and a nonisotropic angular distribution of the molecular dissociation axes are required in order for polarized light to be observed. Although the magnitude of the polarization can provide information about this angular distribution, the interpretation of the experimental results is ambiguous unless one knows which states