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Quenching of metastable $He^+(2s)$ ions in collision with noble gases

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A two-state theory is used to obtain the quenching cross sections of metastable helium ions in collision with noble gases (He, Ne, Ar, Kr, and Xe). The potentials used in the present calculation are based on information derived from the experimental scattering data by Smith *et al.* The results for the quenching cross sections are found to be in reasonable agreement with the recent data of Prior and Wang.

I. INTRODUCTION

The destruction of excited atoms in collision with gaseous targets has been the object of considerable interest in recent years. The total collisional destruction cross section results from the sum of three inelastic processes, namely, electron loss, electron capture, and collisional deexcitation. The dominant contribution to the cross section at low energies, however, comes from the collisional deexcitation process. The study of the deexcitation of hydrogen atoms in the metastable $2s$ state by low-energy collisions with atoms and molecules has been studied both experimentally[1-10] and theoretically.[11-14] The transition to the ground state could take place either directly or through an intermediate excited state. For low energies the major contribution to the cross section for the quenching process is most likely to come from the adjacent $2p$ state, i.e., via the processes

$$A(2s)+X \to A(2p)+X \to A(1s)+X+h\nu$$
$$\to A(1s)+X^*+\text{energy},$$

where A is the projectile (hydrogen atom or helium ion) and X is the target atom; X^* corresponds to the case in which the target atom is left in an excited state after the collision.

Byron and Gersten[11] used a pseudopotential approach to study the quenching of H($2s$) in collision with noble gases.

Slocomb *et al.*[13] studied the quenching of metastable hydrogen atoms in low-energy collisions with spherically symmetric collision partners in the adiabatic approximation. They used the Born-Oppenheimer (BO) approximation to simplify the treatment. Within this approximation one assumes two Σ potential curves arising from X and the $n=2$ state of hydrogen. The current understanding about the collisional process $H(2s)+X$ is that, for extremely large internuclear separations $(r>r_t)$ the hydrogen exists in either the pure $2s$ or pure $2p$ state. But during its passage near the target at some value of the internuclear distance (r_t) the character of these states changes, and they become s-p hybrids, i.e., $(2s+2p$ or $2s-2p)$. No transition occurs between the two Σ states for $r<r_t$. After passing through the transition region at r_t, the atom is still in the metastable $2s$ state, which is expressed as a linear combination of the BO states:

$$2s=[(2s+2p)/\sqrt{2}+(2s-2p)/\sqrt{2}]/\sqrt{2}\,.$$

Slocomb *et al.*[13] conclude that the $2s$-$2p$ transition proceeds in a manner identical to the symmetric charge transfer. In the present work we study the collisional quenching of $He^+(2s)$ ions in passing through noble gases. We compare our results for the quenching cross section with the recent experimental data of Prior and Wang.[15] Hartree atomic units are used throughout.

II. THEORY

In the two-state region of validity the scattering amplitudes for the elastic and inelastic processes are given, respectively by[16,17]

$$f_0=\frac{1}{2ik_0}\sum_l(2l+1)(e^{2i\eta_l^+}+e^{2i\eta_l^-}-2)P_l(\cos\theta) \quad \text{(1a)}$$

and

$$f_1=\frac{1}{2ik_0}\sum_l(2l+1)(e^{2i\eta_l^+}-e^{2i\eta_l^-})P_l(\cos\theta)\,, \quad \text{(1b)}$$

where $k_0=(2\mu E)^{1/2}/\hbar$ is the wave number, μ is the reduced mass of the colliding system, and E is the center-of-mass energy. $\eta_l^\pm$ are the elastic phase shifts for scattering on the two adiabatic potentials $V^\pm$. The corresponding elastic and inelastic differential cross sections are obtained as

$$I_0(\theta)=\frac{1}{4}\left|\frac{1}{k_0}\sum_l(2l+1)P_l(\cos\theta)\times[\sin\sigma_l\cos\delta_l+i(1-\cos\sigma_l\cos\delta_l)]\right|^2 \quad \text{(2a)}$$

and

$$I_1(\theta)=\frac{1}{4}\left|\frac{1}{k_0}\sum_l(2l+1)P_l(\cos\theta)(e^{i\sigma_l}\sin\delta_l)\right|^2, \qquad (2b)$$

where $\sigma_l=\eta_l^+ + \eta_l^-$ and $\delta_l=\eta_l^+ - \eta_l^-$.

The total cross sections are given by

$$Q_0=\frac{\pi}{k_0^2}\sum_l(2l+1)(\cos^2\delta_l+1-2\cos\sigma_l\cos\delta_l) \qquad (3)$$

and

$$Q_1=\frac{\pi}{k_0^2}\sum_l(2l+1)\sin^2\delta_l. \qquad (4)$$

In the calculation of the above cross sections we have evaluated the phase shifts in the JWKB approximation, which are given by

$$\eta_l^\pm=(l+\tfrac{1}{2})\tfrac{1}{2}\pi-k_0r_0+\int_{r_0}^{r_t}\{[F^\pm(r)]^{1/2}-k_0\}dr, \qquad (5a)$$

where

$$F^\pm(r)=k_0^2-(2\mu/\hbar^2)V^\pm(r)-[(l+\tfrac{1}{2})/r]^2; \qquad (5b)$$

r_0 is the outermost zero of the function $F^\pm$. In practice the value of r_t is so large that it can be taken as infinite for the purpose of the calculation of phase shifts in Eq. (5a). The lower limit of the integral of the phase shift in the JWKB approximation is taken to be the classical turning point r_0.[18]

III. POTENTIALS

The interaction potential between an ion and an atom consists of two parts, namely, a short-range repulsive potential and a long-range attractive potential. The repulsive potential arises when the ion and the atom come close enough for their charge clouds to overlap. It is usually expressed in a simple exponential form[19] given by

$$V_s=(b/r)e^{-r/\beta}, \qquad (6)$$

where b is a constant and β is a screening parameter. Several forms for the short-range repulsive potential have been suggested in the literature, such as the addition of more exponential terms in Eq. (6), the use of the Morse-type potential, the replacement of the exponential in Eq. (6) by a Thomas-Fermi function, etc.

The polarization potential in an ion-atom collision is given by[20]

$$V_p(r)=-\alpha e^2/2r^4, \qquad (7a)$$

where α is the polarizability of the target atom and e is the charge on the incident ion. In order to avoid the blowing up of this potential at the origin, a damping factor $f(r)$ is generally multiplied by the right-hand side of Eq. (7a). The function $f(r)$ given below is such that it becomes zero at the origin and is unity at infinity:

$$f(r)=1-e^{-r/d}\left(1+\frac{r}{d}+\frac{r^2}{2d^2}+\frac{r^3}{6d^3}\right), \qquad (7b)$$

where d is a constant.

In an attempt to derive information about the potentials from experimental measurements, Smith *et al.*[21] analyzed the data of Aberth and Lorents[22] and Fuls *et al.*[23] for the elastic scattering of He$^+$ ions on Ar and Ne at energies above 10 eV. Smith *et al.* found that the data can be fitted fairly well by a repulsive screened Coulomb potential of the form of Eq. (6). Considering the shell structure in the screening parameter, they obtained

$$V_{\rm rep}(r)=\frac{e^2}{r}\sum_i A_i e^{-r/C_i}, \qquad (8)$$

where the summation is over the various shells of the target atom. The coefficients C_i for Ar and Ne, based on the analysis of experimental data, are given in Table I of the work of Smith *et al.* In order to relate it to the theory, they took the coefficients C_i as equal to

$$C_i=a_0(I_{\rm H}/I_i)^{1/2}, \qquad (9)$$

where a_0 is Bohr's radius, $I_{\rm H}$ is the ionization potential of the hydrogen atom, and I_i is the ionization potential of the individual shells of the target. Good agreement was obtained between the values of C_i obtained from Eq. (9) and from the fits to the experimental data as gathered from Table 1 of Smith *et al.* In order to explain the deviation of the scattering data from the calculated values obtained by using the potential of Eq. (8), Smith *et al.* considered the polarization potential [Eq. (7a)] with the parameter d of Eq. (7b) taken equal to the screening parameters C_i of the outermost shell of the target.

In the present calculation for the quenching process we take the total potential to be

$$V^\pm=V_{\rm rep}\mp V_{\rm pol}. \qquad (10)$$

TABLE I. Constants involved in the repulsive potential.

Target	A_i	C_i
He	2	0.744
Ne	8	0.790
Ar	8	0.929
Kr	8	0.988
Xe	8	1.059

For $V_{\rm rep}$ and $V_{\rm pol}$ we take the forms as given above by Smith *et al.* [Eqs. (7)–(9)]. In our case, since the energy is very low (≈ 0.23 eV), we do not expect the shell structure of the target to play an important role. We therefore retain only the contribution from the outermost shell in Eq. (8). In the absence of any systematic information available on potentials for other targets (e.g., He, Kr, Xe),[24] we assume for them also a similar form as obtained for Ne and Ar by Smith *et al.*[21] We obtain C_t from Eq. (9) by using the respective ionization potentials of the various target noble gases. The coefficients A_t and C_t are given in Table I.

It may be emphasized here that the potentials reported by Smith *et al.* were derived from the scattering of ground-state ions from the noble gases, whereas in the present case we are considering the quenching of helium ions which are initially in the 2*s* state. We expect that this change in the initial state of the ion will not introduce a major change in the repulsive potential, because of the spherical symmetry of the states in both cases. Further, in our case, since the energy is low, the repulsive potential is not expected to play a substantial role as compared to the polarization potential [the leading contribution of which will still be given by Eq. (7)[25]].

Further, we note that Smith *et al.* have derived these potentials from the data at 10 eV and above. One might question whether these potentials can be used at large enough r. In our opinion there will not be any significant error in using these potentials, because the potential at large r is dominated by the polarization potential, which for ion-atom collisions is always expressed by Eq. (7).

IV. RESULTS AND DISCUSSION

Figures 1 and 2 give plots of $V^{\pm}$ potentials with the internuclear separation r. From Fig. 1 we note that at smaller internuclear separations the potential falls rapidly, and after that a gradual fall is observed. The dominant contribution to the potential for small values of r is from the screened Coulomb repulsive potential, and for larger values of r the main contribution comes from the polarization potential. In the whole range of r the potential V^+ remains repulsive in nature. From Fig. 2 it is seen that for smaller values of r the behavior of the V^- potential is repulsive and is almost the same as that of the V^+ potential. As r increases, the potential changes its behavior and be-

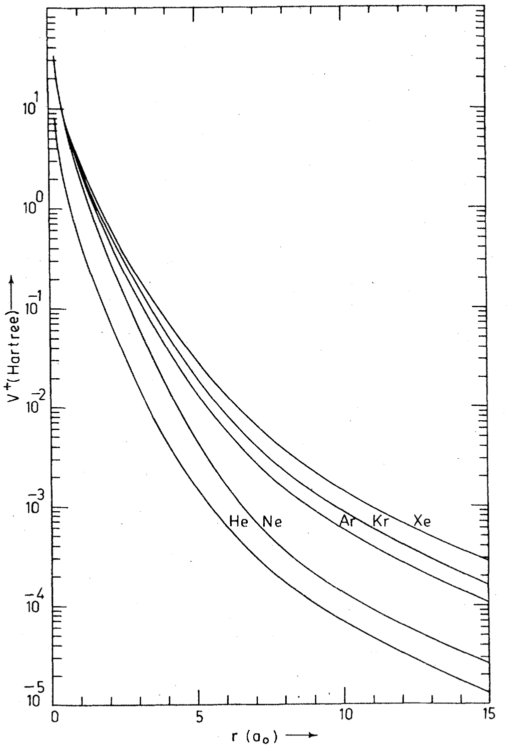

FIG. 1. Variation of potential V^+ with respect to internuclear separation r for He, Ne, Ar, Kr, and Xe.

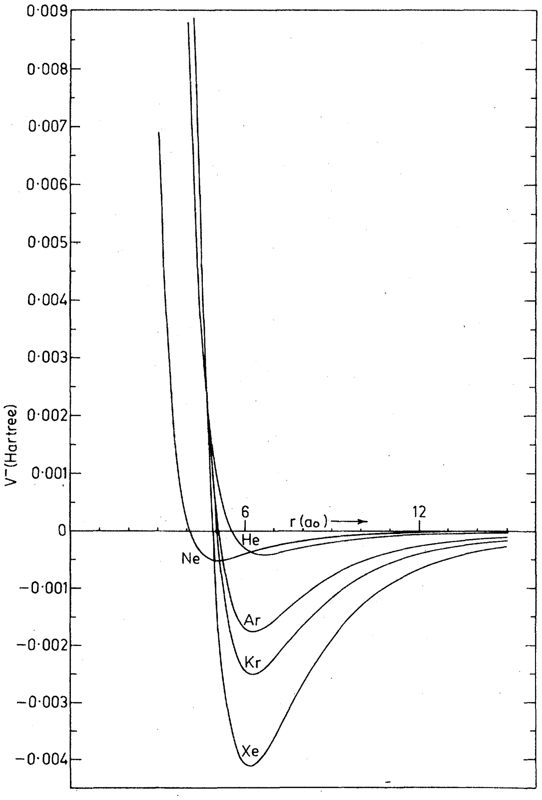

FIG. 2. Variation of potential V^- with respect to internuclear separation r for He, Ne, Ar, Kr, and Xe.

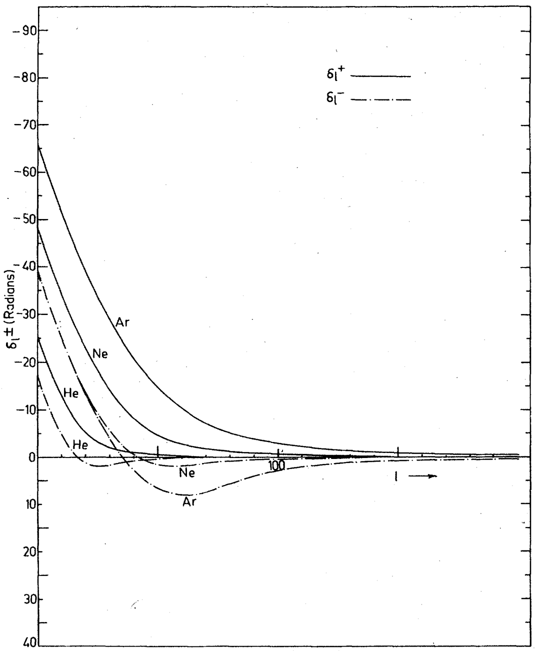

FIG. 3. Variation of phase shifts $\delta_l^{\pm}$ with respect to the partial waves l for He, Ne, and Ar. Solid curve, δ_l^+; dot-dashed curve, δ_l^-.

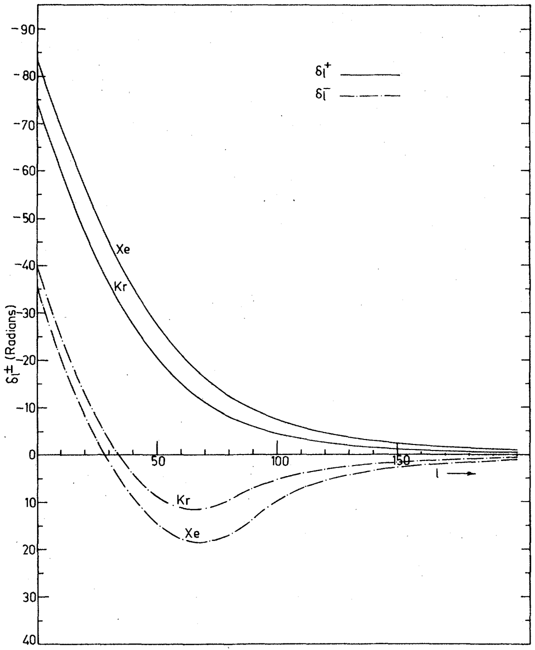

FIG. 4. Same as Fig. 3 but for Kr and Xe.

comes attractive in nature.

In comparison to the hydrogen metastable quenching, the interaction potential in the present case of helium metastable ion quenching is of larger range and varies as r^{-4} rather than r^{-7} at larger r. This is due to the presence of the positive charge on the helium ion.

Figures 3 and 4 show the variation of the phase shifts $\delta_l^{\pm}$ with respect to the partial waves l for the different target noble gases. From the figures it can be seen that for small values of l, δ_l^+ varies rapidly, and after that a slow variation is noted. For large l the phase shift δ_l^+ (corresponding to V^+ potential) becomes almost constant. The behavior of δ_l^- (corresponding to V^- potential) with respect to a lower value of l remains identical to δ_l^+, but, as l increases, the phase shift changes its sign and becomes constant for large values of l.

The above phase shifts are used in Eq. (4) to obtain the total cross sections for inelastic scattering. These are presented in Table II and compared with the effective cross sections reported by Prior and Wang (which are obtained by dividing the rates by the velocity). We have also included in this table the cross section obtained by using Eq. (11), which follows. It is noted from the table that our results for Q_1 are within a factor of 2 for all the target gases, in comparison with the data. However it may be mentioned that, in obtaining these cross sections from the rates, Prior and Wang estimate an average velocity

$$\langle v^{1/2}\rangle^2 = (3.3 \pm 1.7) \times 10^5 \text{ cm/sec},$$

in which there is approximately a 50% uncertainty.

TABLE II. Quenching cross sections for He$^+$(2s) in collision with He, Ne, Ar, Kr, and Xe (Å^2).

Target	Effective cross section (Prior and Wang[a])	Inelastic cross section Q_1 [Eq.(4)]	[Eq.(11)]
He	52.0	77.0	71.0
Ne	···	117.0	110.0
Ar	156.0	302.0	285.0
Kr	199.0	396.0	374.0
Xe	273.0	553.0	522.0

[a] Reference 15.

Dose *et al.*[3] have emphasized that at low collision energies the elastic scattering outside the detection angle of the experiment greatly enhances the cross section. The contribution of elastic scattering can be obtained by numerically integrating the differential elastic scattering cross section given by Eq. (2a). However, the elastic scattering will be important in beam attenuation measurements of the quenching cross section, where the elastically scattered ions miss the detector. In their experiment on the quenching of metastable helium ions, Prior and Wang examined the emission of light from the ions stored in the ion trap. Therefore, unless the large-angle elastic scattering actually removes ions from the trap in a time short compared to the 2-msec lifetime of the metastable ion, the elastic scattering will not be important. In view of this we ignore elastic scattering.

In Fig. 5 we plot our results for the ratio of the cross section Q/Q^{He} $(Q=Q_1)$ for various targets with respect to the square root of the polarizability. The results are compared with the corresponding ratio of the rates (R/R_{He}) obtained experimentally.[15] Prior and Wang have pointed out that the ratio R/R_{He} is equal to the ratio Q/Q^{He} provided the dependence of the cross section on relative velocity is the same and also that the relative velocity distribution is the same for both targets. They have shown that these conditions are nearly fulfilled in their experiment. From the figure we see that our results for the ratios are in reasonably good agreement with the data. For heavier target gases, however, there remains some disagreement.

A rough estimate of the quenching cross sections and their velocity dependence can be obtained if one uses the Massey-Mohr[26] approximation to obtain the phase shifts. Assuming $V^+ - V^- \approx \alpha/r^4$, the cross section can be shown to be given by[27]

$$Q_1 = \pi(\pi\alpha/4v)^{2/3}. \tag{11}$$

This gives a $v^{-2/3}$ dependence on velocity for the cross section. Prior and Wang[15] have predicted a $v^{-1/2}$ velocity dependence [Eq. (15) of their paper], which we feel can only be obtained by using a r^{-5} type of interaction potential.[25] The leading term of the interaction is, however, r^{-4}, which does not seem to have been considered by Prior and Wang in obtaining Eq. (15) (of their paper), although they do mention it in their Eq. (12).

Figure 6 shows our results [based on Eq. (4)] for the velocity dependence of the quenching cross section for $\mathrm{He}^+ + \mathrm{He}$ case. Also shown are the results obtained by using Eq. (11). It is seen that Eq. (11) provides a reasonable estimate of the cross section when compared to the accurate values [Eq. (4)].

We are unable to provide the velocity dependence of the cross sections [Eq. (4)] for other targets for lack of sufficient computer time. For heavier targets, at each velocity one needs to calculate a large number of phase shifts (η_l^+ and η_l^-) to get a reasonable estimate of the cross section. There seems to be no simple way to extract the velocity dependence from Eqs. (4) and (5). However, Eq.

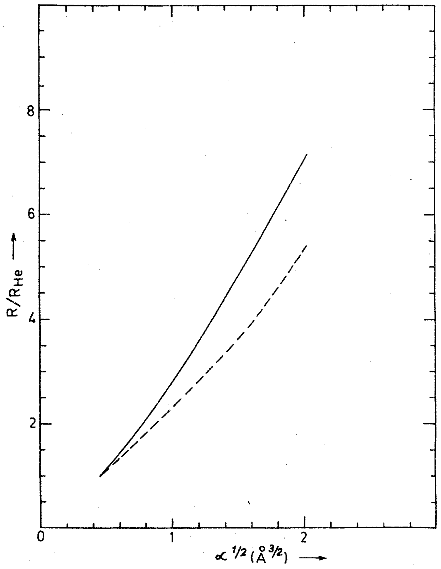

FIG. 5. Plot of ratio Q/Q^{He} vs square root of polarizability. Solid curve, present results for Q_1/Q_1^{He}; dashed curve, experimental results for R/R_{He} (Ref. 15).

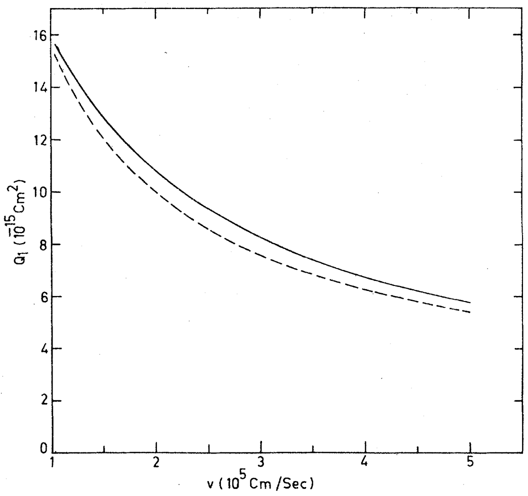

FIG. 6. Velocity dependence of quenching cross section for $\mathrm{He}^+ + \mathrm{He}$. Solid curve, present results [Eq.(4)]; dashed curve, present results [Eq. (11)].

(11) can be used to obtain a rough estimate.

It is concluded that the present approach provides a reasonable estimate of the cross section. We hope that refinements in the interaction potential will lead to a better agreement with the data of Prior and Wang, and shall investigate this in future work.

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