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Excitation of low excited states of Ne I and He I by He$^+$ + Ne collisions

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Emission cross sections for the resonance lines of Ne I (744 and 736 Å) and He I (584 Å) have been investigated during collisions of He$^+$ + Ne at energies from threshold to 9 keV. The threshold of excitation for the channel which dissociates into the upper state of the 736-Å line is 21.7 ± 2.0 eV, and the large modulation depth in the total cross section shows this channel to be strongly coupled at an internuclear separation of 5.3 Å to another excited channel which is presumed to be He*($1s2s$) + Ne$^+$($2p^5$). The onset of excitation of the 744-Å line occurs at 16.3 ± 0.5 eV, and the emission cross section exhibits oscillatory structure which is less pronounced than that of the 736-Å line. Excitation of the 584-Å line appears to occur through several channels.

I. INTRODUCTION

The growing interest in low-energy ion-atom collisions over the last several years has prompted many measurements of differential scattering cross sections and optical-emission cross sections. Much of the work has concentrated on interactions involving ion-atom pairs of various rare gases or alkali metals because both the ions and the atoms of these elements can be readily produced. Of these systems the $(\mathrm{He}+\mathrm{Ne})^+$ complex has been the object of some of the most extensive theoretical and experimental investigations.[1-12] Particular attention has been focused on explaining the interaction of the incoming channel, $\mathrm{He}^+(1s)+\mathrm{Ne}(2p^6)$, with excited channels that lead to $\mathrm{He}^+(1s)+\mathrm{Ne}^*(2p^53s)$. The elastic scattering shows a well-defined perturbation due to this interaction,[1,8] and direct excitations of the lowest excited states of neon are the most prominent features of the energy-loss spectrum.[2] *A priori* calculations have indicated that the primary excitations to these states are due to one or two isolated curve crossings[4,13] so that this system has been an attractive one for testing different theoretical approaches to the description of low-energy ion-atom collisions.

The inelastic differential cross section for producing $\mathrm{Ne}^*(2p^53s)$ displays certain interesting features which are characteristic of processes which proceed via mechanisms associated with curve crossings. There are several well developed Stueckelberg[14] oscillations which are slowly modulated in amplitude. The Stueckelberg oscillations arise from interferences between the elastic and inelastic channels. They depend upon the phases developed along the possible paths which the system may take between its two passages through the crossing point. At a fixed energy the phases depend upon the impact parameter and are, therefore, manifest in the differential cross sections; their effects are generally not apparent in the total cross section when it is plotted as a function of energy.[15]

The slow modulation of the Stueckelberg oscillations results from the coupling of two excited states as the quasimolecule dissociates.[15-17] Such interactions are usually localized in a region that is several a.u. outside the two primary crossings.[18] The frequency of the modulation depends upon the relative phase development between the primary crossing points of the two excited channels and this interaction region. Unlike the Stueckelberg oscillations, the effects of excited curve crossings also appear in the total cross sections. De Heer *et al.*[10] have observed such effects at collision energies from 0.3 to 10 keV in the emission cross section of the Ne I resonance line at 736 Å, the upper state of which arises from the $2p^53s$ configuration, and Hughes, Jones, and Tiernan[12] have seen indications of this mechanism in studies of the emission cross section for the neon lines at energies below 120 eV. Excitation of neon to states which lie above those of the $3s$ configuration have also been observed both by differential scattering and by observations of the spectra in the visible region.[11] The scattering data for excitation into these higher states are not so intense nor so well resolved as those for excitation of the $3s$ states, and the analysis of the results is by necessity restricted. The emission cross sections obtained by Tolk *et al.*,[11] both from direct excitation and from charge exchange into excited states, do oscillate as a function of the collision energy thus indicating that interactions between excited channels are quite prevalent during the dissociation.

In the present experiments we have measured the relative cross sections from threshold to 9 keV for the unresolved resonance lines of Ne I (736 Å, 744 Å) in order to obtain data which complement the studies of the differential scattering.

Measurements have also been made for the individual lines from threshold to 3 keV. The apparatus and experimental techniques are described elsewhere and are not reviewed here.[19] Particular attention has been paid to defining the oscillatory structure over the entire energy range of the unresolved data and to establishing the location of the secondary crossing. The relative emission cross section for the 584-Å line of He I which lies within our limits of detectability has also been obtained. There are no differential scattering experiments which relate to the excitation of the $2\,^1P$ state of helium, nor are there calculations of potential curves and of transition probabilities into channels which lead to this state. As a result, the features of this cross section are not so well understood as are those for the neon lines.

II. RESULTS

The initial investigations of the emission cross sections for the two neon lines were made using a 32-Å passband which left them unresolved but which provided sufficient intensity to detect small, closely spaced oscillations by using counting periods of 100 sec. These results are shown in Figs. 1 and 2. More than 5000 counts have been recorded at each point of the cross section above 34 eV so that one standard deviation of the statistical accuracy is less than 1.5% of the amplitude. Below this energy the uncertainty gradually increases as threshold is approached.

The most striking aspect of the emission cross sections shown in Figs. 1 and 2 is the appearance of a well-developed set of oscillations spanning the entire range. Three of the peaks, those at 64, 81, and 108 eV are quite apparent in the curve measured by Hughes, Jones, and Tiernan.[12] Oscillations at lower energies are not resolved in their data. The large modulation depth in the middle and high energy ranges indicates a strong mixing of excited states.

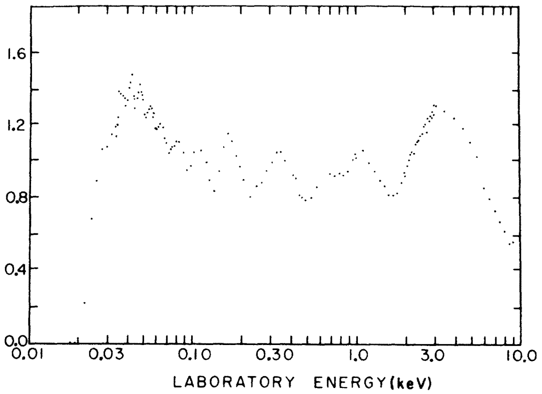

FIG. 2. Emission cross section from threshold to 9 keV for the $2p^53s(^1P, ^3P) \rightarrow 2p^6\,^1S$ doublet (736 Å, 744 Å) of Ne I.

After investigating the emission cross section for the unresolved doublet, measurements were made for the individual lines by using a bandwidth of 4 Å. These results are shown in Figs. 3 and 4. At impact energies greater than 80 eV the statistical uncertainty in each data point is less than 3% for the 736-Å line and less than 5% for the 744-Å line; fluctuations in the individual cross sections are well defined. But the ion current decreases rapidly from almost 1.0 μA at 80 eV to 0.1 μA at 30 eV, and the uncertainty in the total number of counts at each point becomes comparable to the amplitude of the real fluctuations in the low-energy region so that the details of the individual cross sections are not accurately determined.

The oscillations which are so evident in Fig. 2

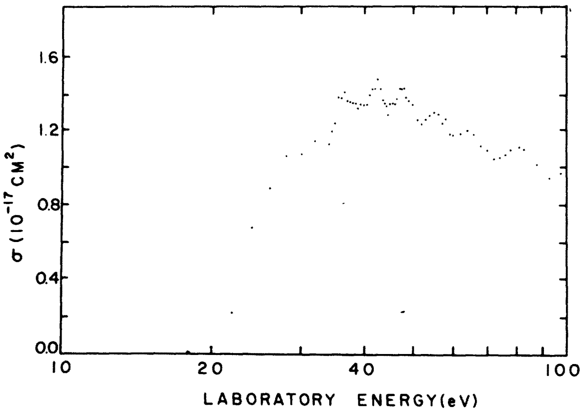

FIG. 1. Emission cross section from threshold to 100 eV for the $2p^53s(^1P, ^3P) \rightarrow 2p^6\,^1S_0$ doublet (736 Å) of Ne I.

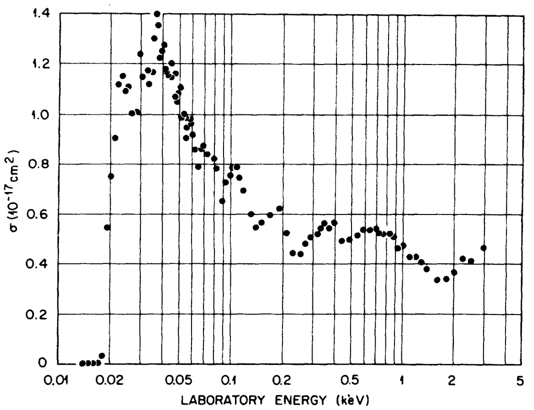

FIG. 3. Emission cross section for the resolved 744-Å line of Ne I.

exist in the cross sections for each line, but the modulation is much more pronounced for the 736-Å line. The four maxima above 80 eV occur at energies of 108, 160, 330 and 1050 eV for the 736-Å line, and at 105, 190, 370, and 700 eV for the 744-Å line. The primary excitations of the two lines seem to arise from crossings between the incoming channel and two different excited channels; the thresholds in Figs. 3 and 4 appear to be at about 18 and 26 eV, respectively, although the poor signal-to-noise ratio at low energies makes an accurate determination from these results uncertain by ± 2 eV.

Figure 5 is the sum of the cross sections shown in Figs. 3 and 4. The general features agree well with the measurements made by observing both lines together, except for the slower rise from 1.7 to 3.0 keV. The oscillations of the composite curve are mainly due to the pronounced variability in the cross section of the 736-Å line.

The apparatus used in the present experiments is not calibrated to yield absolute values for the cross sections; but according to De Heer *et al.*,[10] the apparent emission cross section for the neon doublet is 0.86×10^{-17} cm^2 at 6 keV when the target pressure in their apparatus is 10^{-3} Torr. Absolute emission cross sections for the resonance lines of the target gas are often difficult to specify because the number of photons that reach the detector is influenced by the trapping of the radiation. Such factors as pressure, Doppler shifts of the lines emitted from energetic atoms, and the geometry of the apparatus all affect the radiative transfer. Their studies of the dependence of the apparent cross sections upon pressure of the target indicate that it is linear below 10^{-3} Torr, however, so it would seem that the reliability as an absolute measurement is comparable to that which they specify for the optically thin lines, i.e., a factor of two. The absolute cross sections assigned to Figs. 1–5 have been obtained by normalizing our data to that of De Heer *et al.* at 6 keV.

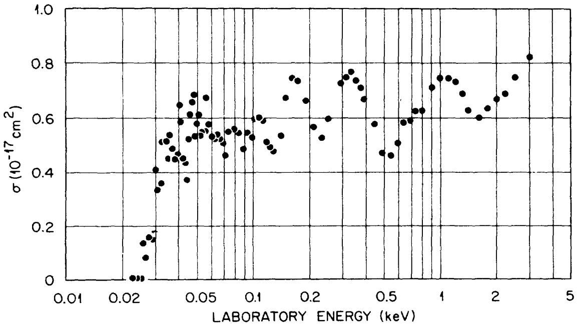

FIG. 4. Emission cross section for the resolved 736-Å line of Ne I.

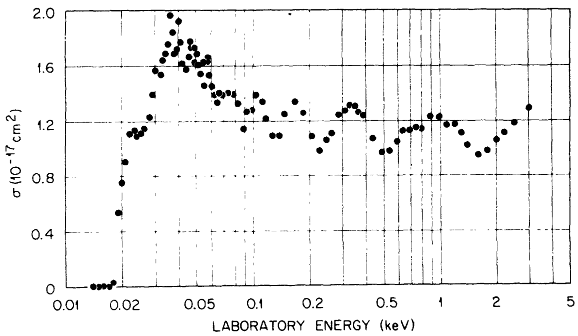

FIG. 5. Sum of the individual cross sections for the 736- and 744-Å lines of Ne I.

At an impact energy of 44 eV a quantum close-coupled calculation[20] utilizing the curves of Sidis and Lefebvre-Brion[4] gives a value of 1.86×10^{-17} cm^2 for transitions from the incoming channel B to the lowest excited channel C which dissociates directly into $\mathrm{He}^{+}(1s)+\mathrm{Ne}^{*}(2p^{5}3s)$. If it is assumed that the upper state of the 744-Å line, which has the lowest threshold of the two neon lines, is excited via the C channel, then from Fig. 3 we obtain a value of 1.17×10^{-17} cm^2 for comparison with the theoretical result. In view of the uncertainties that may exist in both the experimental and the computed values, the difference does not seem unreasonable.

Figure 6 shows the relative emission cross section for the 584-Å line of He I. There appear to be no oscillations which would indicate an interaction solely between two excited states over a large range of energies. Instead, the structure appears to indicate the effect of various primary-excitation modes becoming dominant at different energies. De Heer *et al.* do not state a cross section for this line, but it is possible to assign a

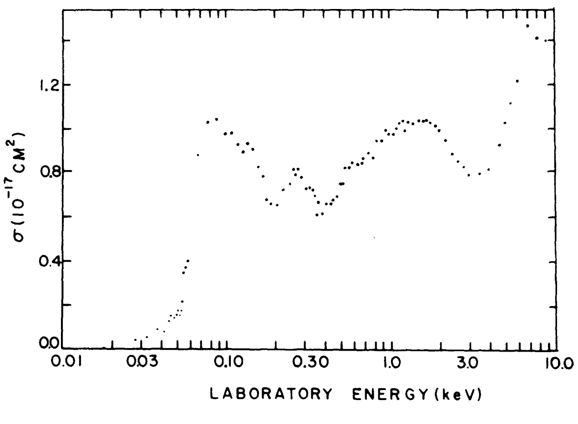

FIG. 6. Emission cross section from threshold to 9 keV for the $1s2p\,{}^1P \rightarrow 1s^2\,{}^1S$ transition (584 Å) of He I.

value by a somewhat circuitous method. Hughes, Jones, and Tiernan[12] do give absolute values for both the neon and the helium lines, but these appear to be too low in view of the consistency between the other measurements and the calculations already cited. However, if we assume that their relative calibration is accurate, the absolute value of the 584-Å line can be established by normalizing their neon data to the value given by De Heer *et al.*; this procedure has been used to assign absolute values to the measurements of the emission cross section of the 584-Å line.

Cascading appears to be of minor importance as a mechanism for populating the upper states of the spectral lines investigated in the present work. Spectral scans taken at a collision energy of 1 keV show that other vacuum ultraviolet lines which originate from higher states of both helium and neon have intensities that are less than 3.5% of the intensities of the first resonance lines. Also, the energy-loss spectra of Ref. 2 reveal that excitation to the $2p^53s$ configuration strongly dominates other direct excitations. As a consequence, it is estimated that the contributions of cascading to the emission cross sections of Figs. 1–6 are 7% at most, and that these curves closely represent the collisional excitation cross sections.

III. DISCUSSION

Several calculations have been performed to explain the results of inelastic scattering of the $\mathrm{He}^+ + \mathrm{Ne}$ system, but most of these have been for the purpose of understanding the features of the differential cross sections at certain selected energies. There are no theoretical computations of the total cross sections over a wide range of energies which would be directly applicable to the present results. Nevertheless, some qualitative insight can be obtained by examining our data with respect to the quasi-diabatic potential curves of Sidis and Lefebvre-Brion.[4] These curves indicate that the incoming channel which is designated as the B state $[\mathrm{He}^+(1s)+\mathrm{Ne}(2p^6)]$ is repulsive at all internuclear distances. At 0.99 Å it crosses the excited C and C' states, which upon dissociation correlate to $\mathrm{He}^+(1s)+\mathrm{Ne}^*(2p^53s)$ and $\mathrm{He}^*(1s2s)+\mathrm{Ne}^+(2p^5)$, respectively. The potentials of the C and C' channels have similar shapes, but they are separated by about 3.2 eV for internuclear distances less than 2.5 Å, the largest internuclear distance for which they are plotted in Ref. 4. At internuclear distances larger than 1.4 Å, the C and C' curves are dissociative. At 1.4 Å they turn over and become slightly attractive with a well depth of 1.5 eV at 0.95 Å. Inside 0.95 Å the curves become repulsive. Thus the B state crosses the C and C' states in a region inside a low potential barrier. At a smaller internuclear separation, about 0.66 Å, the incoming channel crosses a pair of states D and D' which are everywhere repulsive and dissociate into the same products as the C and C' states. Although the potentials of the Σ states that lead to the excited configuration $\mathrm{He}^+ + \mathrm{Ne}^*(2p^53s)$ have been computed, there are no calculations for the Π-state potentials and no theoretical studies of electron promotion through rotational coupling. Also, there are no calculated potentials for channels that produce excitation of the 584-Å line of He I.

Comparison of threshold energies provides one direct correlation that can be made between experimental results and theoretical potential curves. According to the curves of Sidis and Lefebvre-Brion, the energy required to produce the lowest excited state of the system (the C state) is 17.8 eV. The measurement of Baudon *et al.*[7] for this threshold agrees exactly with this value, but the result obtained by Coffey *et al.*[2] is about 4 eV lower. Our result for the threshold of excitation of the 744-Å line, which can be obtained with more confidence by using the data from the unresolved doublet (Fig. 1) than the data from the 744-Å line alone, is estimated to be 16.3 eV in the center-of-mass system, and thereby falls between the two values obtained from the measurements of the differential cross sections. It is possible that the experimental data for the emission cross section indicate a threshold which is higher than the minimum required to excite the C state if the probability of tunneling through the barrier is much smaller than the probability of a predissociation back to the B state. Such a discrepancy should be less than 1.5 eV if the calculated value of the well depth is correct.

Uncertainties in the energy of the incident ions usually limit the precision with which thresholds can be measured in the present apparatus. The mean energy of the beam is determined only within ± 0.5 eV. In addition the full width at half-maximum of our beam profile has been measured to be 3.6 eV, although this value undoubtedly overestimates the spread in the energies of the ions since the measurement was performed by employing only a single retarding grid. The low-energy oscillations seen in Fig. 1 would not be so well resolved if the width of the beam actually was as large as 3.6 eV. Previous investigations for excitation of the $3p^6\,{}^2S$ state of Ar^+ in $\mathrm{He}^+ + \mathrm{Ar}$ collisions have also indicated that the spread in energies within the ion beam is small enough that it does not influence the determination of the threshold significantly. The systematic error is, therefore, taken to be ± 0.5 eV.

The onset of emission of the 736-Å line appears to be at 21.7 ± 2 eV, with the large uncertainty being caused by the poor signal-to-noise ratio for the resolved lines at low impact energies. Of the curves investigated by Sidis and Lefebvre-Brion, only the *C* and *C′* channels which are calculated to have crossings at 17.8 and 21.0 eV above the asymptotic level of the *B* state can be excited at such a low energy. It would appear from the observed thresholds that the 744-Å line is excited via the *C* state and the 736-Å line via the *C′* state. But since the *C′* channel correlates to $\mathrm{He}^*(1s2s)+\mathrm{Ne}^+(2p^5)$, a secondary crossing is required to populate the upper state of the neon line. Sidis and Lefebvre-Brion have, in fact, pointed out that a crossing between the *C′* and *D* channels may be significant in exciting the $\mathrm{Ne}^*(2p^5 3s)$ configuration. Under the assumption that crossings of the *B* channel with the *C* and *C′* channels are responsible for the excitation of the two neon lines near threshold, we specify the energy at the crossing points as

$$E_{BC}=16.3\pm 0.5 \text{ eV} \tag{1}$$

and

$$E_{BC'}=21.7\pm 2.0 \text{ eV}. \tag{2}$$

At energies greater than 72 eV it should be possible to produce the neon lines by a primary excitation through the *D* state; however, there is no indication in Figs. 3 or 4 of the sudden onset of a second mode of strong excitation for either line.

The origins of oscillations in the total cross sections for inelastic processes have been discussed most thoroughly by Ankudinov, Bobashev, and Perel.[17] They consider two cases in which excited states can be mixed outside the primary crossing points: (i) the two excited channels mix at a point where they cross; (ii) the two excited channels approach one another, do not cross, but are strongly coupled in some well-localized region. The first mechanism was studied by Rosenthal[15] and the second by Demkov.[16] The phase of the oscillations is given by

$$\varphi=\frac{1}{\hbar v}\int_{R_1}^{R_3}\Delta V(R)\,dR+\Gamma\,. \tag{3}$$

For the case of term crossing

$$\Gamma_{tc}=\pi/4\,, \tag{4}$$

if the modulation depth is not small, and for the case of term approach

$$\Gamma_{ta}=(H_{22}-H_{11})/\hbar v\lambda\,. \tag{5}$$

Here, v is the average velocity of separation in the two channels, $\Delta V(R)$ is the difference in energy of the two channels as a function of internuclear distance, and R_1 and R_3 are the primary and secondary crossing points. For the case of term approach, $(H_{22}-H_{11})$ is the asymptotic energy difference of the two excited states and λ is a parameter such that the off-diagonal matrix elements are proportional to $\exp[-\lambda(R-R_3)]$. Equation (3) is valid if the phase difference between the two excited channels is independent of the impact parameter, in which case φ should be a linear function of $1/v$.

Figure 7 shows a plot of the reduced phase angle as a function of $1/v$ for the peaks of the unresolved data of Figs. 1 and 2. As noted in Sec. II, the oscillations at energies above 100 eV are due primarily to the 736-Å line, but at the lower energies the individual contributions cannot be determined. The points in the middle of the plot appear to lie on a straight line with a slope of 2.05×10^8 cm/sec; the points at either end deviate somewhat from this line. It is expected that the peaks at low velocities may not be uniformly spaced in $1/v$ because the phase development is not completely independent of the impact parameter. It is seen from Fig. 1 that the peak of the lowest oscillation is also rather ill-defined and its location may be slightly in error. The uncertainty in the slope of the line does not permit the initial phase to be determined accurately enough to differentiate which of the two coupling mechanisms is operating. Because the points at high and intermediate values of $1/v$ lie on the same straight line, it is presumed that all the oscillations shown in Figs. 1 and 2 reflect the coupling of the upper state of the 736-Å line to another excited state of the system. Although the relative phases of the four well-defined peaks in the cross

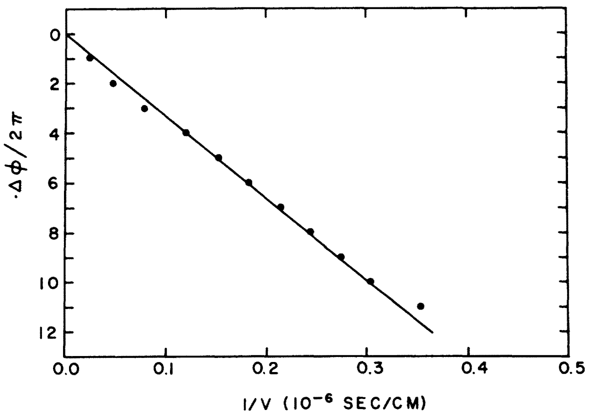

FIG. 7. Reduced phase of the oscillatory peaks of the Ne I excitation vs the reciprocal velocity at which the peaks occur.

section for the 744-Å line do not scale linearly as $1/v$, the slope of the average straight line through the points of a plot of $\varphi/2\pi$ vs $1/v$ is almost the same as the slope of the function in Fig. 7. It appears, at least at the intermediate and the low impact energies, that the upper states of both neon lines may exhibit oscillations which are due to couplings to the same charge-exchange state.

If the oscillations in the emission cross sections of the neon lines arise from secondary couplings of the C and D channels with the C' channel, the position at which the mixing takes place can be estimated. The channel which leads to the 736-Å line, $\mathrm{He}^+ + \mathrm{Ne}^*(3p^5[^2P_{1/2}]3s, J=1)$, is separated asymptotically from the channels $\mathrm{He}^*(2s\,^3S) + \mathrm{Ne}^+$ and $\mathrm{He}^*(2s\,^1S) + \mathrm{Ne}^+$ by $+0.5$ and -0.75 eV, respectively, but the computed differences are almost constant at -3.2 eV from the primary crossings out to 2.7 Å, the limit to which they are plotted in Ref. 4. By neglecting for the present the possibility that the term Γ_{ta} may have to be taken into account in computing the phase development, Eq. (3) and the slope of the function shown in Fig. 7 lead to

$$\int_{R_1}^{R_3} \Delta V(R)\, dR = 13.5 \text{ eV Å}, \tag{6}$$

for either the case of term approach or term crossing. By setting $\Delta V(R)$ equal to a constant 3.2 eV, and the location of the primary crossing R_1 equal to 0.99 Å, the location of the coupling region R_3 can be established as

$$R_3 \approx 5.2 \text{ Å}. \tag{7}$$

If the excited state mixing is due to the term approach mechanism, the neglect of Γ_{ta} must be justified. Ankudinov *et al.* show that Γ_{ta} should be ≤ 1 if the modulation depth is large. By assuming that the modulation around the peak at 160 eV is deep enough for this criterion to be applied, and that the maximum asymptotic separation of the interfering states is 0.75 eV, it is found that $\lambda^{-1} \leq 0.4$ Å and $(H_{22} - H_{11})/\lambda \leq 0.3$ eV Å. In view of the approximations which have been made to obtain $R_3 - R_1$, it is appropriate to neglect Γ_{ta}. It is interesting to note that the modulation which appears in the Stueckelberg oscillations of the inelastic differential cross section for exciting the $2p^5 3s$ configuration comes from an interaction which takes place at an internuclear separation of about 2.1 Å[20] and, hence, does not correspond to the interaction which appears predominately in the total cross section.

In contrast to the extensive effort which has been placed upon analyzing the direct excitation of the C states of the $\mathrm{He}^+ + \mathrm{Ne}$ system, very little work has been done either experimentally or theoretically on charge exchange into excited states of the projectile. The lack of calculated potential curves for both Σ and Π states which dissociate into $\mathrm{He}^*(2p)$ inhibits the interpretation of the structure in the emission cross section of the 584-Å line. Certain aspects of this structure are worth noting. The threshold occurs at an impact energy of 22 ± 2 eV ($E_{\mathrm{c.m.}} = 18.3 \pm 1.7$ eV), very close to the threshold for exciting the C or C' states. The cross section rises slowly until an impact energy of 54 eV ($E_{\mathrm{c.m.}} = 45$ eV) is reached; at this point there is a steep change in the slope which undoubtedly signifies the onset of a second mode of excitation. The threshold and qualitative behavior for this cross section in the present experiments agree quite well with the results of Hughes, Jones, and Tiernan.[12]

The excitation mode for $\mathrm{He}^*(2p)$, which appears to set in at 54 eV, must occur through a state which has a strong coupling to the incoming channel since the emission cross section for the helium line is comparable to that for the neon doublet at impact energies above 65 eV. The energy of onset in the center-of-mass system is 15 eV lower than the calculated threshold for exciting the D state ($E_{\mathrm{c.m.}} = 60$ eV) and 27 eV above that for the C state ($E_{\mathrm{c.m.}} = 18$ eV). Both of these calculated values for the C and D states have been verified experimentally, and it would appear that the major mode of excitation of the 584-Å line comes from a primary crossing which is not included in the curves of Sidis and Lefebvre-Brion. If this excited channel has Σ symmetry, its inclusion in the group of states used for calculating the quasi-diabatic curves may be quite important.

The process which is responsible for the slowly rising portion of Fig. 6, from threshold to 54 eV, may well be a secondary excitation mode caused by a weak coupling of the C' channel to other states outside the primary crossing point. The lines investigated by Tolk *et al.* appear to have thresholds near 24 eV in the center-of-mass system, which may imply that they also are the result of secondary excitations.

ACKNOWLEDGMENTS

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[18] We will refer to curve crossings between the incoming channel and excited channels as primary crossings and excitations which proceed through such couplings as primary excitations. Excitations which proceed *via* coupling between two excited channels subsequent to the primary excitation will be called secondary excitations.

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