EXPERIMENTAL TESTS OF THE POST-COLLISION-INTERACTION MODEL FOR THE He^+-He EXCITATION FUNCTIONS*

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New data on the excitation of helium by low-energy helium ions have been obtained that provide strong support for the post-collision-interaction model proposed by Rosenthal and Foley in a companion paper. Certain details of the results that are not presently predicted by the theory are pointed out.

New experimental studies of low-energy He^+-He total excitation cross sections are presented that provide strong support for a post-collision-interaction model proposed by Rosenthal and Foley in a companion paper[1] (hereafter referred to as the RF model).

We previously reported the excitation of various neutral states of He in low-energy He^+-He collisions.[2] Two striking results of these observations were the large value of the cross section at low energies with thresholds slightly above the final-state energies, and the strong oscillatory dependence of the cross sections on projectile energy. These phenomena were interpreted in terms of the molecular-potential energy curves of the He_2^+ system. To understand the low-energy excitation it was noted that, in the diabatic approximation, the gerade ground term is repulsive and pseudocrosses all of the excited state terms.[3] These pseudocrossings occur at a value of internuclear separation R_I of about 1 a.u. In the course of collisions with small enough impact parameters, the internuclear separation decreases and passes through R_I. As the system passes through the pseudocrossing, excitation occurs by the mechanism discussed by Landau, Zener, and Stückelberg.[4] This accounts for the large cross section at low energies. Since the thresholds occur slightly above the final-state energies, they provide a direct measure of the energy location of the pseudocrossing.[5] In addition to other results discussed here, we report new precise values for these energies which can be compared with specific molecular models.

Previously we proposed that the oscillations were produced by an interference effect which arose from the double passage of the system through the crossing at R_I. However, it has now been shown (cf. RF) that any phase difference thus developed is strongly dependent on the impact parameter b; consequently, the integration over impact parameters required to obtain total cross sections averages out this interference effect. Such a phase difference might contribute to the oscillations reported in differential inelastic cross sections[6] but cannot explain the structure observed in the total cross-section data.

In the companion paper, Rosenthal and Foley show that in addition to the crossing between the ground-state and excited-state curves at R_I, other crossings between two or more excited states exist at relatively large values of internuclear separation, $R_0 \approx 20$ a.u. In this model the excitation mechanism is essentially identical with that previously considered and hence the large cross sections at low energies, and the threshold energies, are explained as before. However, the oscillations are now considered to arise when two excited states populated at R_I develop a phase difference between R_I and R_0 before mixing again at R_0. Since the resulting phase difference is weakly "b"-dependent, it manifests itself in the form of oscillatory structure in the total cross section. Rosenthal and Foley show that their model correctly predicts the oscillatory structure in the excitation functions of the 3^3S and 3^1S states, and, moreover, they predict that these oscillations should be in antiphase as is in fact observed.[2] New experiments whose results strongly substantiate the RF model are described in this paper.

Rosenthal and Foley find that in their model, outer crossings, which can efficiently mix the excited-state amplitudes, exist in the $n = 3$, 4, and higher states but not in the $n = 2$ states. Hence they predict oscillatory structure in the $n = 3$ and higher excitation functions but not in the $n = 2$ cross sections. In our earlier work we observed the $n = 3$, 4, and 5 levels and found strong oscillations. A new experimental study

has now been made of the 584-Å line (2^1P-1^1S) and 10 830-Å line (2^3P-2^3S) to test the predictions of the RF model. The apparatus was essentially the same as that used earlier except that in the case of the 10 830-Å line a liquid-nitrogen–cooled EMI model No. 9684B phototube was used to detect the radiation, and the line was isolated with an interference filter. Since no filter material exists for the 584-Å light, a McPherson model No. 220 uv spectrometer was used to isolate this line. The spectrometer viewed the bombarding chamber at an angle of 90° relative to the beam, and the light, after passing through the spectrometer, was detected with an EMI model No. 9603 particle multiplier. Although resonance trapping was initially considered to be a serious problem for the 584-Å line, simple estimates indicate that the transverse momentum transferred in the course of the collision is sufficiently large so that no trapping occurs. The resulting Doppler shift, at energies greater than 100 eV and for scattering angles greater than 1°, is larger than the thermal Doppler width of the line and little absorption occurs. The possibility of trapping is further reduced by the fact that the collision region is viewed over an angular region of at least ±3° and therefore the component of velocity in the direction of viewing is sufficiently large to insure a Doppler shift greater than the thermal Doppler width. The absence of trapping was tested experimentally by observing that the pressure dependence of the signal was linear and by comparing the signal for $^3He^+$-^4He and $^4He^+$-^4He collisions. Since the ^3He-^4He isotope shift is larger than the thermal Doppler width, any possible trapping would be seriously modified by changing isotopes; yet no difference in either signal strength or the form of the excitation function was observed, confirming the absence of trapping.

The $2P$ excitation functions are shown in Fig. 1(a). In accordance with the RF model and unlike the $n = 3$, 4, and 5 excitation functions, no large-scale oscillatory structure is observed. Some small-scale structure which does appear is discussed below. The fact, noted above, that there is no significant difference between the 584-Å excitation function for $^4He^+$-^4He and $^3He^+$-^4He collisions shows that nuclear symmetry is not important in these phenomena.

As mentioned previously, measurement of the excitation-function threshold gives an accurate value of the energy location of the inner pseudocrossing.[5] Careful measurements were made of

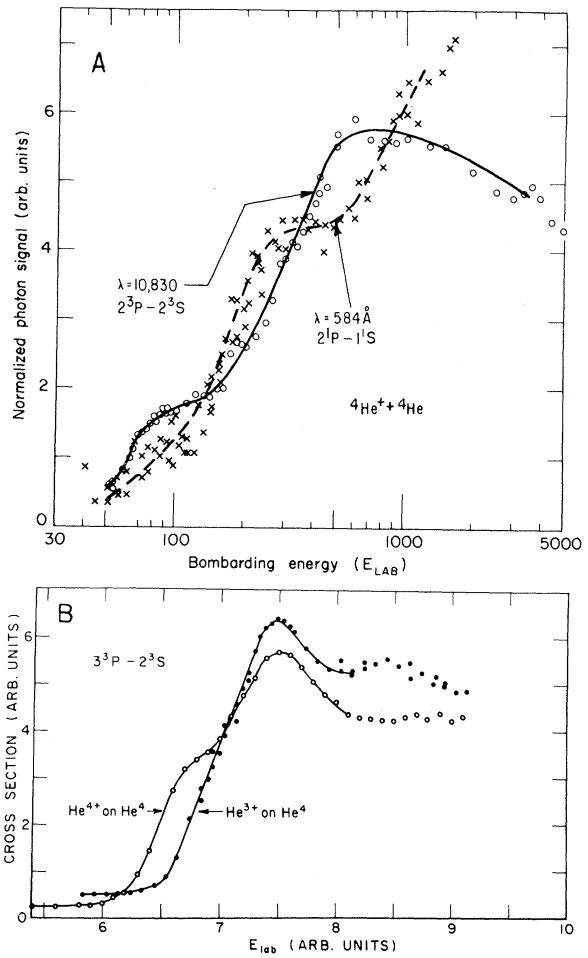

FIG. 1. (a) Cross sections for the excitation of the $2P$ states of He in He^+-He collisions. (b) Detailed threshold behavior of the 3888-Å excitation function for $^4He^+$-^4He and $^3He^+$-^4He collisions.

the signal near threshold for nine transitions. In extracting threshold values from these data, a linear threshold law was assumed in accordance with the observed steep nature of the onsets. In these experiments two phototubes were used simultaneously, one with a 3888-Å filter and the other with a filter chosen to isolate the line under study. In this way the thresholds could be obtained relative to the 3888-Å threshold. This technique was employed since there was evidence that the true beam energy differed by 1 or 2 eV from the applied potential. Such differences would be expected to arise from contact potentials, space-charge effects, and surface charging. Since these effects may vary with time and operating conditions, the comparison method was employed so as to obtain the best possible values for the difference in the thresholds. It is estimat-

ed that the differences were obtained to ±0.2 eV. The correction needed for obtaining the absolute value of the 3888-Å threshold was determined by measuring the $^3He^+$-^4He and $^4He^+$-^4He thresholds for this line and using the fact that the center-of-mass threshold for these two collision combinations should be identical. In this way the energy correction at the time these data were taken and the absolute value of the 3888-Å threshold were found to be -0.63 ± 1.56 and 60.50 ± 1.56 eV, respectively. The threshold values in the center-of-mass system and energies of the final states of the free atom are given in Table I.

The displacement of the thresholds above the final-state energies of the free atom indicates that the pseudocrossing at R_I occurs above the final-state energy. The observation of distinct thresholds for two excited states which cross at a large value of internuclear separation R_0 (e.g., 3^3S, 3^1S) allows us to make a qualitative statement concerning the transition probability at this outer crossing at low energies. If the transition probability at this crossing were large we would expect population of the upper state at R_0 even though the energy were so low that only the lower state was populated at the inner crossing. This would result in identical thresholds for both the states. The observation of distinct thresholds for these two states indicates that at low velocities the crossing at R_0 is essentially adiabatic. We are thus free to interpret the thresholds as a measure of the energy location of the inner crossings.

Table I. Thresholds for excitation of excited He I states.

State	Threshold relative to 3^3P (Lab. eV)[a]	Absolute threshold (c.m. eV)[b]	Difference between threshold and final-state energies (c.m. eV)[c]
2^3P	−7.30	26.60	5.64
3^3S	−2.57	28.97	6.26
3^1S	−1.85	29.33	6.42
3^1D	−0.40	30.05	6.98
3^3P	•••	30.25	7.25
3^3D	+0.10	30.30	7.23
3^1P	+1.80	31.15	8.07
4^3S	+1.87	31.18	7.59
5^3S	+2.70	31.60	7.63

[a]These energy differences are measured in the laboratory system and are believed to be reliable to ±0.2 eV.

[b]The energy of the 3^3P threshold is 60.50 ± 1.56 eV (lab system). The other values in this column (Col. 3) are obtained using the absolute 3^3P threshold and the values in Col. 2. Hence, the absolute values in Col. 3 are reliable to ±0.78 eV (c.m. system). However, the differences in the values in Col. 3 are reliable to ±0.10 (c.m.), as are those in Col. 2 from which they are derived.

[c]As in Col. 3, these values are reliable to ±0.78 eV (c.m.) and their differences are reliable to ±0.10 eV (c.m.).

We studied the velocity dependence of the oscillatory structure observed in a number of excitation functions by using ^3He and ^4He in various collisional combinations. For a given energy these different collisional combinations have different relative velocities. Since we believe that the oscillations are due to a phase-interference phenomenon, we scaled the cross sections according to phase-development time by plotting the isotopic data for excitation of a given state of He versus the relative velocity at infinite internuclear separation $v(\infty)$,

$$v(\infty)=\left[\frac{2E_L}{m_P}\right]^{1/2}=\left[\frac{2E_{\text{c.m.}}}{\mu}\right]^{1/2}, \quad (1)$$

where μ is the reduced mass of the system, E_L and $E_{\text{c.m.}}$ are the energies in the laboratory and center-of-mass frame, respectively, and m_P is the mass of the projectile particle.

When the cross sections are scaled according to Eq. (1), the high-energy peaks overlap, in accordance with the assumption of a phase-interference effect. The lack of overlap at low energies [Fig. 2(a)] is due to the difference between the true velocity and $v(\infty)$ in this energy domain. This difference arises from the effect of the interatomic potential $V(R)$ and the centrifugal barrier b^2/R^2. In considering b^2/R^2 we note that for excitation to occur b must be less than R_I. Yet in the RF model the region of relevant phase development varies between internuclear separations of R_I and R_0 which is of the order of $20R_I$. Therefore in this region the value of b^2/R^2 lies between 1 and 4×10^{-3}. In view of this we neglected the centrifugal barrier in scaling the oscillations versus velocity. The effective velocity in the region of phase development is then given by

$$V(R)=\{2/\mu[E_{\text{c.m.}}-\langle V(R)\rangle]\}^{1/2}. \quad (2)$$

Since in the model being considered the phase difference develops in the final state, we may take the average interatomic potential energy $\langle V(R)\rangle$ equal to the final-state energy E_f. In the case of the 4^3S level, the final-state energy is

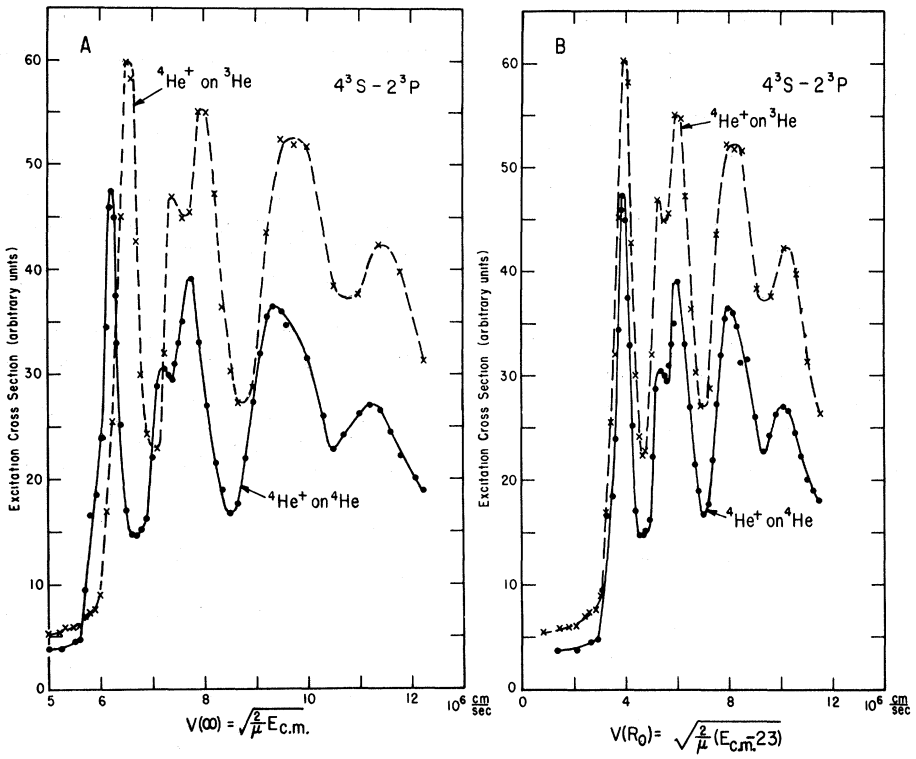

FIG. 2. (a) Excitation cross section for the 4713-Å He I line in $^4He^+$-^4He and $^4He^+$-^3He collisions plotted versus relative velocity at infinite internuclear separation $v(\infty)$. (b) Excitation cross section for the 4713-Å He I line in $^4He^+$-^4He and $^4He^+$-^3He collisions plotted versus relative velocity in the final state.

about 23 eV and this produces a substantial modification of the velocity at low bombarding energy. The isotopic data are plotted in Fig. 2(b) with the velocity given by Eq. (2) with $\langle V(R)\rangle = 23$ eV. In this plot the structure overlaps over the entire energy range in accordance with the RF model and the above discussion.

Two experimental results remain for which only tentative explanations exist. In carefully studying the threshold behavior of the various excitation functions, we find that additional structure appears in the 4713- and 3888-Å lines near threshold when the ^4He-^4He system is studied [Fig. 1(b)]. This structure does not appear for other collisional combinations. The difference in structure appearing in the isotopic data can be explained in terms of the phase-development time. As the energy decreases from the kilovolt range, and with it the velocity, more phase difference is developed and more oscillations appear. Near threshold the velocity in the 4-4 case reaches smaller values than that in the 4-3 case. Therefore, near threshold, the 4-4 system spends more time developing phase than the 4-3 system. These data indicate that enough additional time is spent for one more oscillation to appear. In the case of differential scattering experiments it is observed that the data, obtained in collisions with identical isotopes, i.e., ^3He-^3He and ^4He-^4He, exhibit more structure than in the case of the unsymmetric nuclear collisions ^3He-^4He.[7] This additional structure is attributed to the effects of nuclear symmetry. In the present case, in order to rule out this possibility, we performed an experiment in which we used ^3He ions on ^3He. The results obtained in this experiment did not show the additional structure seen in the ^4He-^4He case, and we conclude that nuclear symmetry is not responsible for the extra oscillation shown in Fig. 1(b).

One additional, unexplained experimental result involves the shoulders observed near threshold in the $2P$ excitation functions [Fig. 1(a)]. If one interprets these features as low-amplitude oscillations superimposed on a smooth excitation curve, then the oscillations in the 2^1P and 2^3P excitation functions are in apparent antiphase. This result seems to indicate that some small mixing occurs between these states although, as previously mentioned, these states are not coupled through an outer crossing. No theoretical interpretation of this result is as yet available.

However, we would like to suggest that this effect may result from a residual interference effect arising at the inner crossing.[2]

The results presented here have stimulated considerable efforts to understand the detailed interactions occurring in low-energy ion-atom collisions. The theory recently developed by Rosenthal and Foley has been remarkably successful in leading to an understanding of those results. Some unanswered questions remain, however, and further theoretical efforts will be needed to answer them. It is clear that the full elucidation of the detailed structure of the excitation functions will provide a powerful test of our understanding of the dynamics of the unbound molecular state formed during these inelastic collisions.

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VARIATIONAL CALCULATION OF THE PROPERTIES OF LIQUID He^3-He^4 MIXTURES AT 0°K

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The properties of liquid He^3-He^4 mixtures at 0°K are studied by a variational method using a Jastrow-Slater trial wave function and the Wu-Feenberg expansion of the energy expectation value: The system of mass-3 boson + mass-4 boson shows complete phase separation; the system of mass-3 fermion + mass-4 boson and the system of mass-3 boson + mass-4 fermion mixtures separate incompletely; and the system of mass-3 fermion + mass-4 fermion mixes completely. The limiting solubility of the mass-3 fermion in the mass-4 boson solvent is found to increase with pressure.

In this Letter we present the results of a variational calculation of the ground-state properties and phase equilibrium of liquid He^3-He^4 mixtures. The properties of such mixtures at zero and nonzero temperatures have already been investigated by Cohen and Van Leeuwen[1,2] (CVL). These authors, using a Fermi-Bose hard-sphere model and the Huang-Yang-Lee pseudopotential method up to first[1] and second[2] order in the hard-sphere diameter, were able to show that such a system has a phase diagram which is in excellent agreement with the experimentally observed He^3-He^4 phase diagram; in particular CVL predicted the existence of an incomplete phase separation at 0°K, which was later confirmed by experiment. The main purpose of our work is to show that this incomplete phase separation remains in a treatment which is not limited to low densities. This can be done by a variational calculation which has, however, the drawback of providing information only on the ground-state properties of the mixture. A variational calculation has already been made by Massey and Woo[3,4], who investigated only the properties of the dilute (less than 7%) solutions of He^3 in He^4 and did not attempt to describe the phase separation.

We assume that the system of n_4 He^4 (mass m_4) and n_3 He^3 (mass m_3) atoms, in a volume v (n_3