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GROSS FAILURE OF THE CONVENTIONAL ADIABATIC CRITERION; STRUCTURE AND COHERENCE IN THE LOW-ENERGY EXCITATION OF HELIUM ATOMS BY HELIUM IONS*

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Recent work on low-energy ion-atom and ion-molecule collisions indicates that these processes can be highly effective in producing optical excitation,[1] in contradiction to the well-known "adiabatic criterion."[2] We here present further evidence for this conclusion in the important case of excitation of helium atoms by He^+ ions. These observations can be discussed in terms of molecular-potential curve crossings of the type first considered by Landau and Zener.[3] This simple model, however, does little justice to the wealth of observed effects. In particular, in the case of collisions which involve only S states in the initial and final channels, we find that the excitation function depends in a strongly oscillatory manner on the bombarding energy of the incident ion. These observations indicate the importance of treating the collision as a coherent process in which the relative phase of different components of the wave function is maintained.

It has been customary to discuss ion-atom and atom-atom collisions in terms of the adiabatic criterion.[2] According to this principle, the cross section for a collision which changes the internal (electronic) energy of the colliding partners by an amount ΔE is a maximum when the collision velocity is given by

$$v_m \cong \Delta E a/\hbar. \quad (1)$$

Here a is a characteristic length, of the order of atomic dimensions. The collision becomes adiabatic with decreasing bombarding energy, and the cross section decreases exponentially, becoming very small as the velocity is lowered below v_m. Many examples in the literature give interpretations of maxima in excitation cross sections in the 10- to 100-keV range by using the adiabatic criterion.[4] We have studied some of these published cases at lower energy and find that the observed high-energy maximum is only one of many such maxima, and that the interpretation in terms of the adiabatic criterion is incorrect as usually applied. The energy defect ΔE is often evaluated from the known energy levels of the isolated particles, and no allowance is made for the change in the electronic structure during the collision. In the case of the helium lines of interest here, this energy defect is about 22 eV, and the corresponding laboratory energy for maximum cross section is about 100 keV according to the conventional use of the adiabatic criterion.

Figure 1 shows the excitation functions for all of the He I visible lines originating from the $n = 3$ triplet and singlet levels, when helium gas is bombarded with helium ions. The laboratory energy in these studies ranged from ≈20 eV to ≈5 keV; the high-energy limit was chosen to overlap with the lower limit employed in the work of de Heer and co-workers.[5] It is immediately evident that the cross section does not decrease monotonically at energies

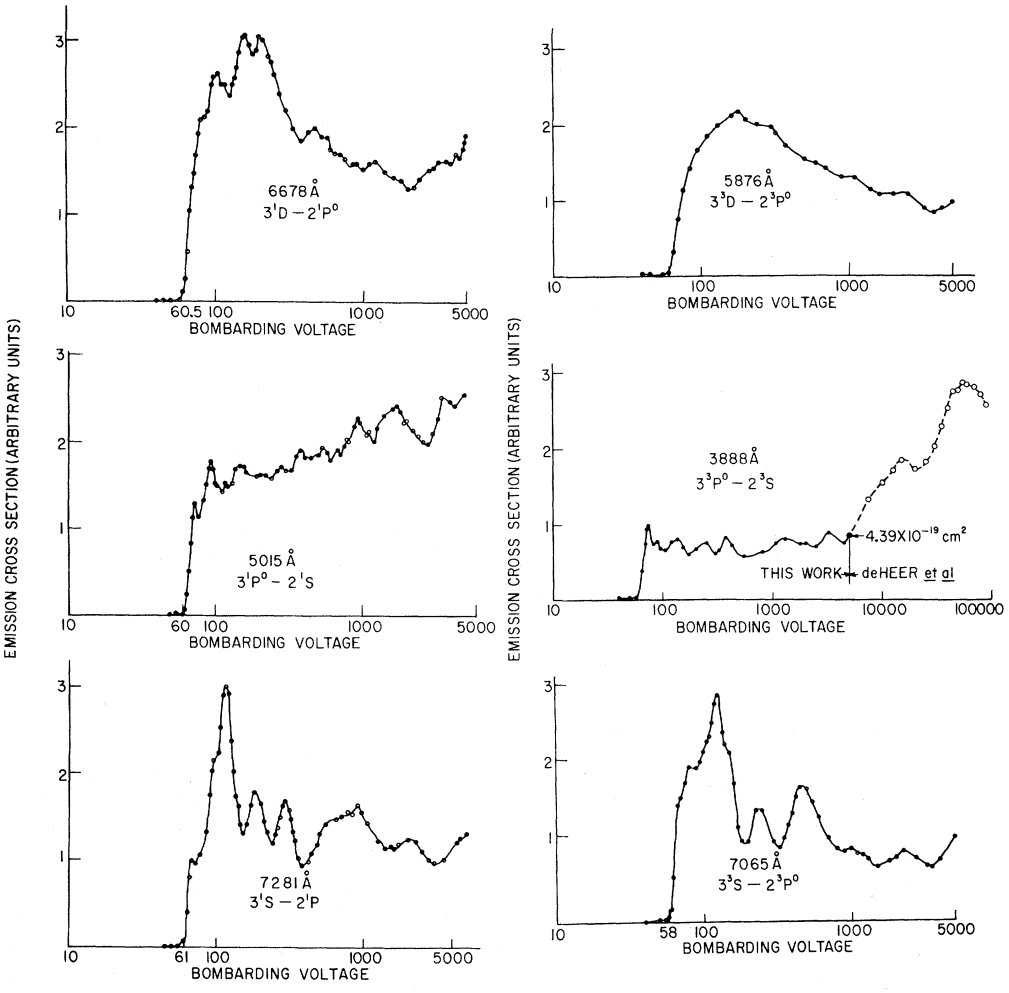

FIG. 1. Excitation functions for all of the visible He I lines originating from the $n=3$ levels when He is bombarded by He^+ ions. The cross section for the 3^3P state is normalized at 5 keV to the absolute data of de Heer, Wolterbeek-Muller, and Geballe (Ref. 5). Their value of the cross section rises to $\approx 7.7\times10^{-19}$ cm^2 at 90 keV. No cascade corrections have been applied.

below several kilovolts, and, in fact, in many cases the cross section attains a large fraction of its maximum value within a few electron volts of threshold. These observations are grossly inconsistent with the predictions based on the adiabatic criterion. This discrepancy certainly arises because the levels of the He_2^+ molecule formed during the collision are greatly modified from those of the free particles. In particular, pseudocrossings are known to occur between the initial state, $(1\sigma_u)^2(1\sigma_g)^2\Sigma_g$, and the final states leading to all of the excited states of the atom.[6] In this case low-energy excitations are to be expected since the energy defect is greatly reduced at the pseudocrossings.

It is noteworthy that the apparent thresholds for the various levels occur at somewhat higher energies than that required by simple energy conservation. This indicates that the relevant pseudocrossing between the molecular levels occurs above the energy corresponding to the final state of the isolated atoms. In particular, the crossings for the $n=3$ and $n=4$ levels both appear to be $\approx$7 eV higher than the levels of the free atom, in qualitative agreement with Michels' calculations.[7]

Several of the excitation functions show an extraordinarily rich and complex oscillatory behavior in the energy range studied. The structure in the excitation function depends markedly on the particular state in He which is ex-

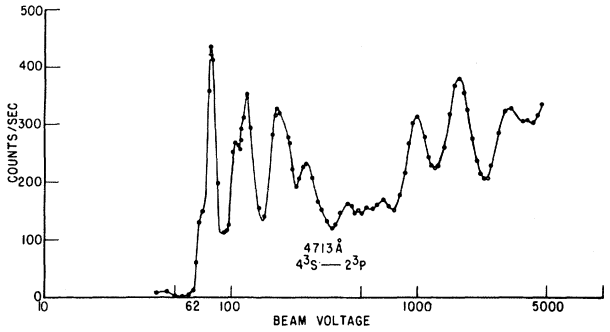

FIG. 2. Excitation function for the 4713-Å line of He (which originates on the 4^3S state) with He^+ bombardment. Beam-energy spread $\lesssim 1$ eV. (The excitation function is not normalized.)

cited, becoming less pronounced as we go from S to P to D states of the same principal quantum number. The 4^3S state (see Fig. 2) shows a bigger oscillatory effect than the $3S$ states. It is important to note that in the case of the 3888-Å line (3^3P-2^3S) our results connect smoothly onto those of de Heer at 5 keV; we have used this point to normalize the absolute cross section. In the cases studied, the gross structure could not be attributed to cascade contributions. Effects of low-energy electron contamination of the He^+ beam were ruled out by using an electrostatic lens system and a small transverse magnetic field in the interaction region.

It is suggested that the oscillatory behavior results from an interference effect between the incoming and outgoing passage through the crossing point. The S states can only be excited through a crossing of two Σ molecular states, the P states through Σ and Π states, and the D states through Σ, Π, and Δ. The greater multiplicity of channels for the P and D states may account for the apparent absence of structure in these cases.

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[4]The adiabatic criterion is stated very cautiously in Ref. 2. It has often been incautiously applied, assuming ΔE to be constant during a collision in order to obtain the energy for a maximum in the cross section. See, for recent examples, F. J. de Heer, Advances in Atomic and Molecular Physics (Academic Press, Inc., New York, 1966), Vol. 2, p. 349. The criterion has been extensively used to predict the location of maxima in the charge transfer cross section; see J. B. Hasted, Physics of Atomic Collisions (Butterworths Scientific Publications, Ltd., London, 1964), pp. 420-423.

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