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Failure of the Adiabatic Criterion; Structure and Coherence in the Low Energy Excitation of Helium Atoms by Helium Ions*

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Low-energy ion-atom and ion-molecule collisions have recently been demonstrated to be effective in producing optical excitations.(1) These observations contradict the well-known adiabatic criterion(2) since they show that excitations readily occur in the low-energy (adiabatic) region. In this paper we present further evidence for this conclusion in the important and simple case of the optical excitation of helium atoms by helium ions. It is clear that curve crossings of the type first considered by Landau and Zener play an essential role in these collisions.(3) In the case of collisions which involve only "S" states in the initial and final channel, we find that the excitation function depends in a strongly oscillatory manner on the bombarding energy of

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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

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

Here a is a characteristic length of the order of atomic dimensions. Accordingly the collision becomes adiabatic, and the cross section decreases exponentially with decreasing bombarding energy, 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 10-100 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 adiabatic criterion.

Figure 1 shows, as an example, the excitation function for one of the He-I multiplets originating from the 3^3P levels, when helium gas is bombarded with helium ions. The laboratory energy in these studies ranged from $\approx$20 eV to $\approx$5 keV; the high energy limit was chosen to overlap with the lower limit employed in the work of de Heer and coworkers.[5] It is immediately evident that the cross section does not decrease monotonically at energies 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 predications 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, pseudo-crossings 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 pseudo-crossings.

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 pseudo-crossing between the molecular levels occurs <u>above</u> 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 excited, 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. Most of the structure cannot be explained by cascading from neighboring states. It is important to note that in the case of the 3888-Å line (3^3P-2^3S) our results connect smoothly unto those of de Heer at 5 keV; we have used this point to normalize the absolute cross section. 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.

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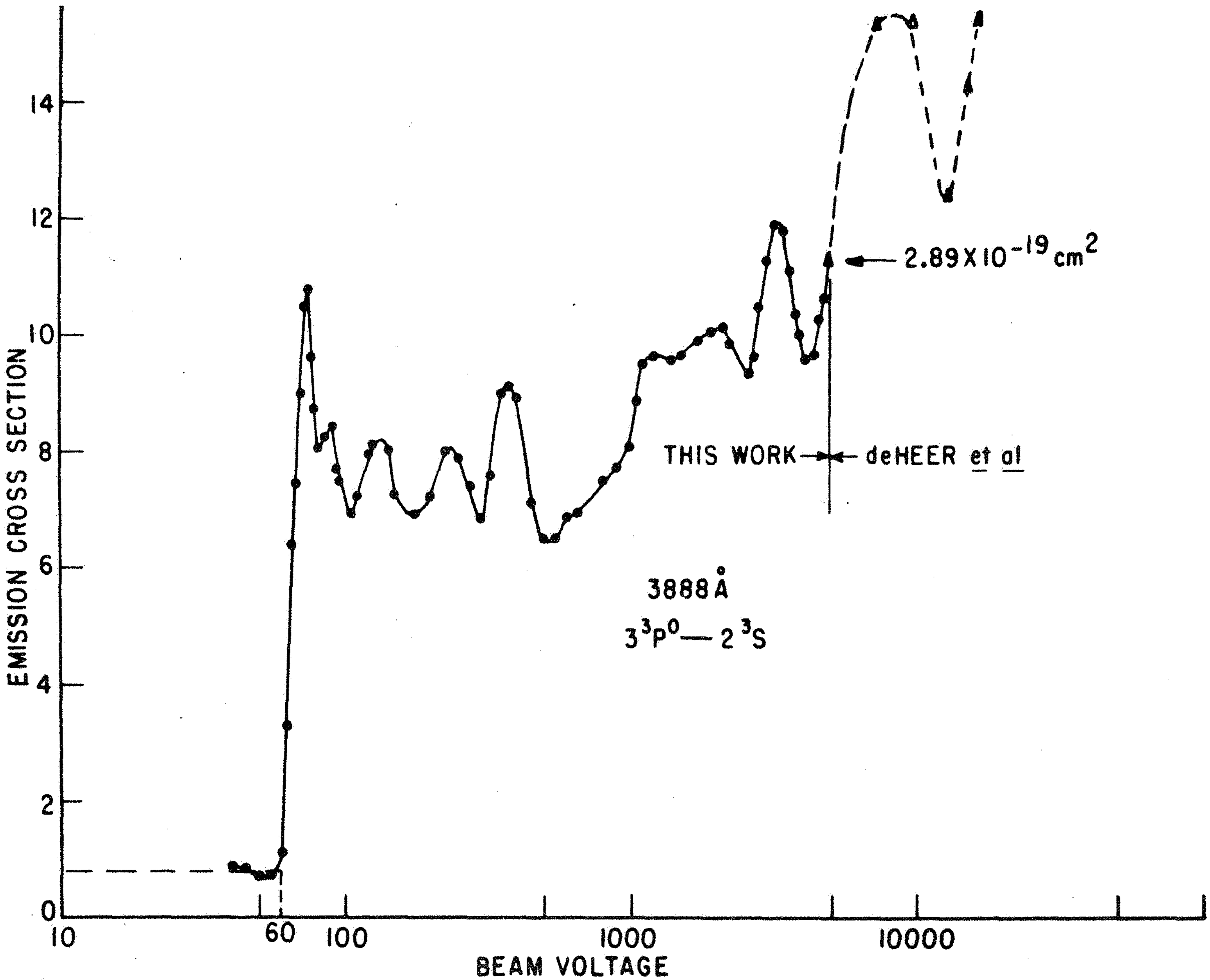
Figure Captions

Figure 1. Excitation function for the visible He-I multiplet at 3888 Å originating from the 3^3P state when He is bombarded by He^+ ions. The cross section is normalized at 5 keV to the absolute data of de Heer, Wolterbeek-Muller, and Geballe (reference 5). Their value of the cross section rises to $\approx 7.7 \times 10^{-19}$ cm^2 at 90 keV. No cascade corrections have been applied. The ordinate value below the marked threshold shows the background noise level.

Figure 2. Excitation function for the 4713-Å line of He (which orginates on the 4^3S state) with He^+ bombardment. Beam energy spread $\lesssim$ 1 eV.



14
12
10
8
6
4
2
0
EMISSION CROSS SECTION
10
60
100
1000
10000
BEAM VOLTAGE
2.89 X 10^-19 cm^2
THIS WORK
deHEER et al
3888 Å
3^3P^0 — 2^3S

FIG. 1

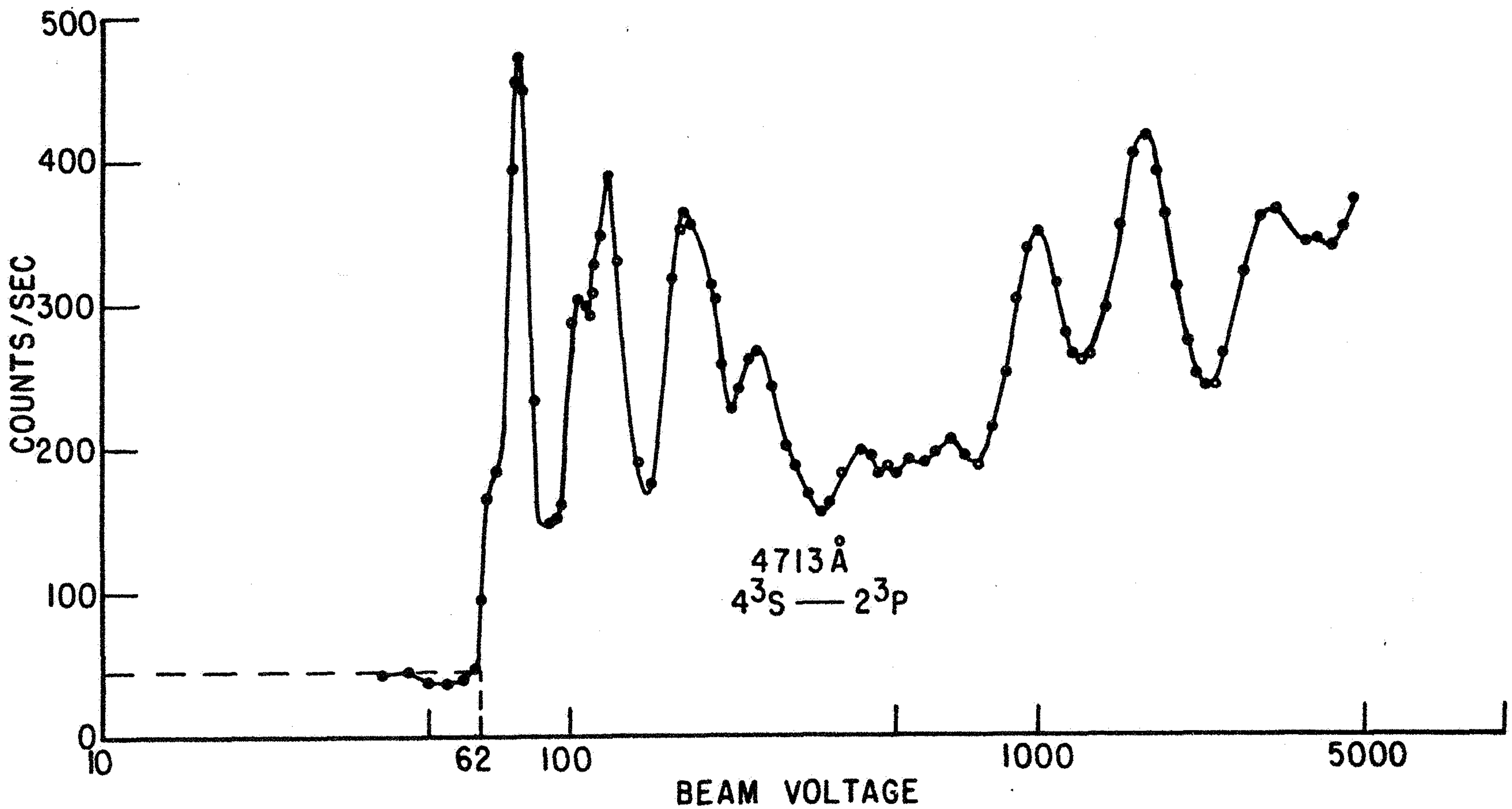

500
400
300
200
100
0
COUNTS/SEC
10
62
100
1000
5000
BEAM VOLTAGE
4713 Å
4³S — 2³P

FIG. 2