J. Phys. B: Atom. Molec. Phys. 13 (1980) L199–L204. Printed in Great Britain

**LETTER TO THE EDITOR**

# Optical observations of molecular dissociation in thin foils

T J Gay† and H G Berry
Argonne National Laboratory, Argonne, Illinois 60439, USA

Received 15 January 1980

**Abstract.** We present measurements of the intensity and polarisation of light emitted from two excited states of neutral helium, after break-up of the $HeH^+$ molecule in thin foils. We vary the foil thickness to study their dependence on the internuclear separation of the He and proton at the final foil surface and postulate that the variations observed are sensitive to the molecular orbitals of the dissociating $HeH^+$ molecule. These vicinage effects provide the distance scale of the final state interaction at the foil surface.

The production and approach to equilibrium of outer-shell electronic excited states in fast ions having traversed thin foils are generally accepted to take place at the exit surface of the foil. We have previously shown that these ion–surface interactions are sensitive to the foil temperature and have suggested that this is due directly to changes in the secondary electron distribution as the ion leaves the surface (Gay and Berry 1979a).

In order to probe the distance parameter of these surface interactions, we have used the bombarding molecular ion $HeH^+$ to observe the influence of a nearby dissociated proton on the populations and alignment of neutral helium states. The proximity of the proton at the exit surface is varied by the simple procedure of using different carbon foil thicknesses.

We find that the total light yields of neutral helium transitions vary strongly, by factors of up to five, with the proximity of the exiting proton at beam energies between 25 and 200 keV $amu^{-1}$. In addition, the variations depend strongly on the excited states being observed. The optical polarisations or atomic alignments also vary with proton proximity, and these variations are also very different for different excited states.

We suggest that the variations between different excited states must be interpreted on the basis of long-range quasi-molecular curve crossings. The large scale variations in light intensity may be caused by inhibition of surface de-excitation by the nearby proton.

Ion beams of $He^+$ and $HeH^+$ were produced at beam energies of 50–200 keV at the University of Chicago's 250 kV electrostatic accelerator, and at beam energies of 450–1000 keV at the Argonne National Laboratory Dynamitron accelerator. A standard beam–foil target chamber and automated optical detection system (Gay and Berry 1979a) were used at both experimental facilities. Relative total light yields and polarisation of the 2p $^3P$–3d $^3D$ (5876 Å) and 2s $^3S$–3p $^3P$ (3889 Å) transitions in neutral helium were measured as a function of foil thickness for both $He^+$ and $HeH^+$ projectiles. For the polarisation measurements, the phase plate or polariser in the

† Also at the Department of Physics, University of Chicago, Chicago, Illinois 60637, USA.

optical train was stepped in equal angle increments for fixed beam charge collected at a shielded Faraday cup. The foil was moved by a stepping motor after each polarisation measurement to obtain a quantum beat curve. For the 3p $^3$P emission, the $J=0$–2 quantum beat was not resolved and the polarisation data presented here are also averaged over each beat wavelength of the $J=1$–2 beat. The polarisation of the 3d $^3$D emission is averaged over the $J=2$–3 and 2–1 quantum beat wavelengths, while the $J=3$–1 quantum beat was not resolved. The Fano–Macek alignment parameter, $A_0^{\mathrm{coll}}$, may be directly obtained from these averaged polarisations.

For the total light yield measurements, photons were measured for a fixed time at each foil thickness for a known incident beam current. This was measured with no foil in place immediately before and after each optical measurement. Typically, sets of three measurements were made to allow for beam fluctuations. We have measured the foil thickening rate for a typical beam current of 1 $\mu$A of $HeH^+$ at 650 keV to be 0·3 ng cm$^{-2}$ s$^{-1}$ at a vacuum of $8\times10^{-7}$ Torr. Since individual runs took about 20 minutes, foil thickening effects in these experiments are negligible.

The RMS separations of the H and He fragments at the exit surface of the carbon foil were calculated taking into account the relative energy losses, and the velocity and angular straggling due to multiple scattering which dominates at low energies and the

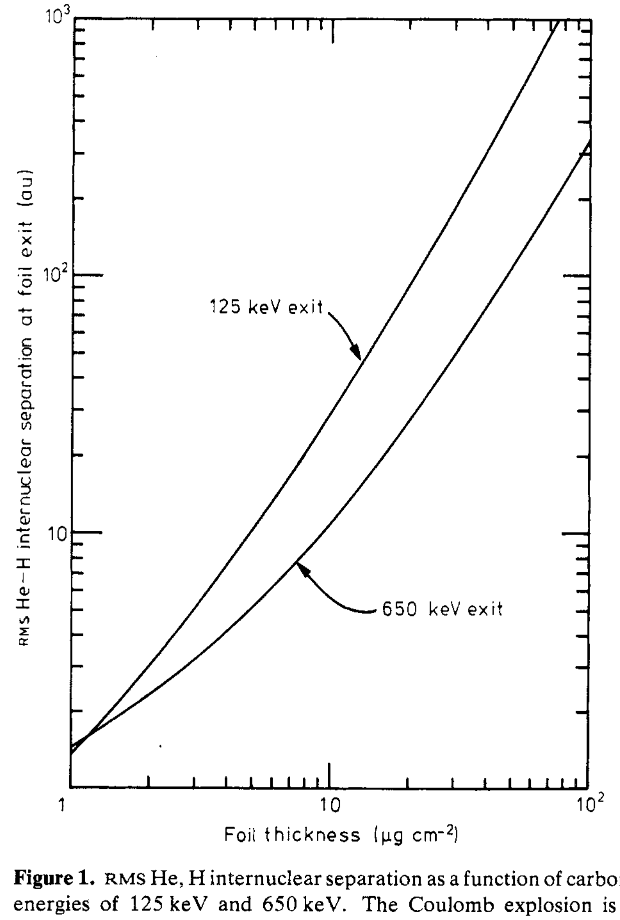


**Figure 1.** RMS He, H internuclear separation as a function of carbon foil thickness at He exit energies of 125 keV and 650 keV. The Coulomb explosion is dominated by multiple scattering at low energies.

Coulomb explosion (Gemmell *et al* 1975) which becomes increasingly important at higher energies. The results of our calculations for He exit energies of 125 keV and 650 keV are shown in figure 1. Dwell time in the foil, the parameter most often considered in molecular experiments of this type, is meaningful only when the Coulomb explosion completely dominates the other processes. This is not the case below 200 keV $amu^{-1}$. All optical measurements were compared for equal *exit energies* of the helium atoms, the incident beam energy being adjusted to take account of energy losses in the different thickness foils.

In figure 2, we show the total light yields from the 2p $^3$P–3d $^3$D and 2s $^3$S–3p $^3$P transitions for a He exit energy of 650 keV. Note that the 3p $^3$P population is unchanged by the proximity of the proton. This is to be contrasted with less detailed measurements we have made of eight other transitions in He I, He II and H I, all of which show population enhancement for thinner foils. Similar results are obtained for a He exit energy of 125 keV.

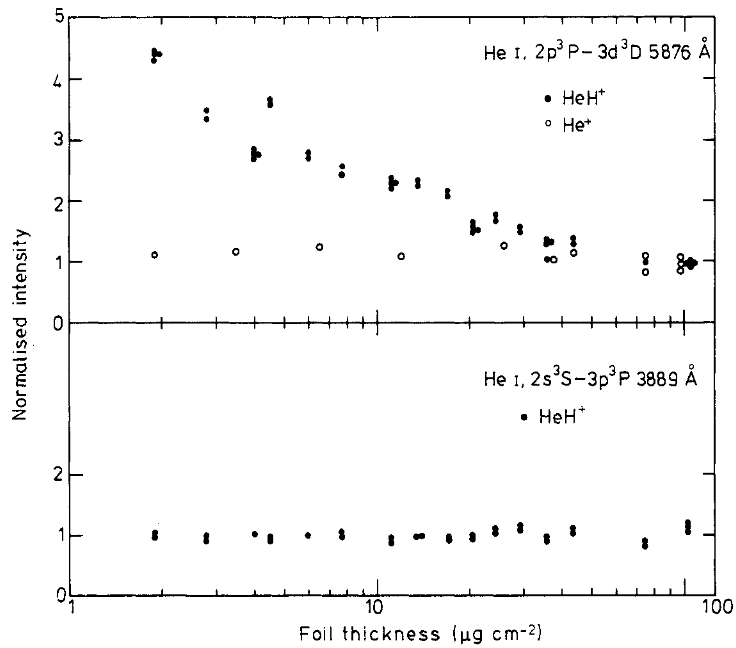


**Figure 2.** Relative total light yields plotted against foil thickness. Intensities normalised at 100 µg $cm^{-2}$. He exit energy 650 keV.

The variation of linear polarisation with foil thickness is shown in figures 3 and 4. Measurements taken for the 3p $^3$P state at 125, 500, 550 and 650 keV He exit energy all give a consistent value of 35 ± 10 au for the distance at which the $HeH^+$ values begin to equal those for $He^+$ projectiles. The results for the 3d $^3$D values at different energies do not scale so well. At 650 keV, the equilibrium distance is 150 ± 50 au. The equivalent value at 125 keV seems to be considerably less.

The total intensity and polarisation of the 2p $^3$P–3d $^3$D transition for $HeH^+$ projectiles begin to differ from their atomic values at roughly the same value of foil

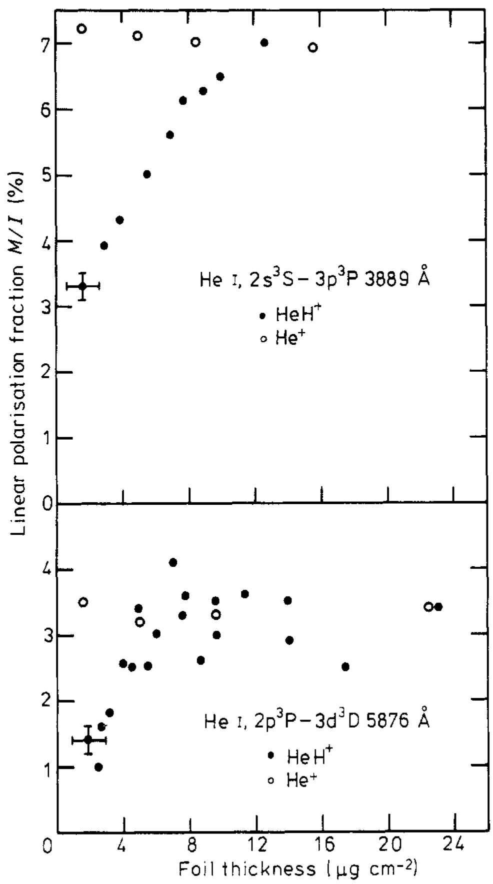


**Figure 3.** Linear polarisation fraction plotted against foil thickness. 125 keV He exit energy. Error bars shown are typical.

thickness. It is tempting, therefore, to draw the conclusion that the proton is isotropically exciting the $^3$D levels and that this is the specific polarisation reduction mechanism. This model fails, however, to explain the 3p $^3$P data.

The slow variations of $M/I$ with thickness for $He^+$ projectiles are due to the increasing temperatures produced in thicker foils for a given beam current density (Gay and Berry 1979a).

We suggest that the above results can be explained by a combination of two effects: first, a single He atom or ion leaves the surface in a high state of excitation and is then de-excited and/or neutralised by non-radiative resonance tunnelling and Auger de-excitation processes (for a brief review, see Hagstrum 1977). When the additional proton is close to the outgoing projectile, these de-excitation processes are inhibited and further excitation occurs as a result of the proton–helium post-surface interaction. The excited states of the projectile whose energies lie at or below the Fermi level of the foil electrons should be less strongly affected by the tunnelling and Auger processes.

Secondly, we suggest that the differences in detail of the light enhancement and the alignment for these excited states are due to quasi-molecular curve crossings. We have made similar, less detailed measurements of the intensity and polarisation of the transition from the 3p $^1$P state using $HeH^+$ projectiles. These results are qualitatively different from those for the 3p $^3$P state. These differences imply that formation of

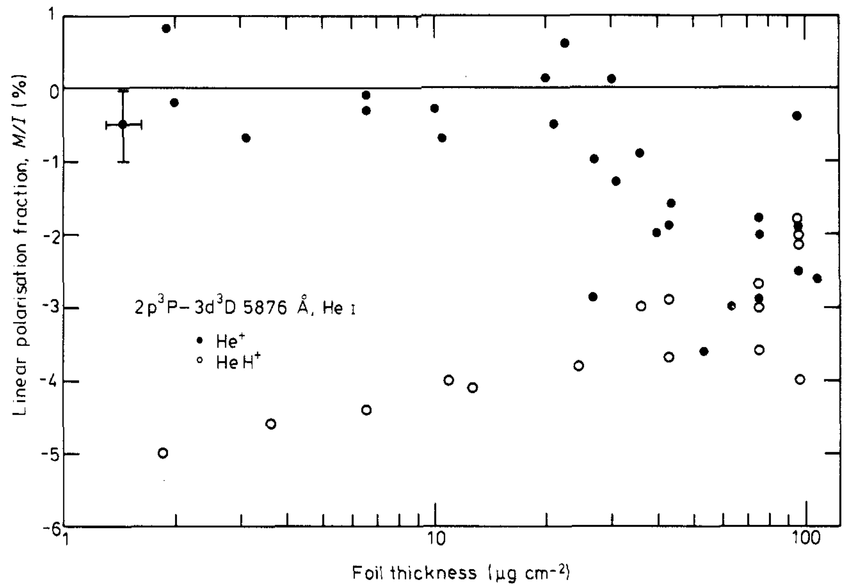


**Figure 4.** Linear polarisation fraction plotted against foil thickness. 2p $^3$P–3d $^3$D (5876 Å) transition. 650 keV $He^+$ exit energy.

quasi-molecular states at the surface has to be considered. Green (1976, 1978) has recently shown that level crossings for $n = 3$ singlet and triplet states of $HeH^+$ occur at quite different internuclear separations. Further analysis of the differences observed for these states requires detailed study of the appropriate $HeH^+$ potential curve crossings and matrix elements. This work is underway.

A principal conclusion from our results is that the surface interaction distance is less than 35 au for defining the production of these $n = 3$ He I states. This distance is about four times larger than the mean diameter of the excited atomic states.

Some initial results have been published previously by Gay and Berry (1979b); also, Schectman (1979) and Schectman and Ellis (1979) have made quantum beat observations of $H_\alpha$ using $H_2^+$ projectiles. K O Groeneveld *et al* (1980, private communication) have studied the quantum beats from decay of 3p $^3$P He I using $HeH^+$ projectiles. The results are in qualitative agreement.

We thank Professor J A Simpson for providing access to the University of Chicago LASR accelerator, and S Tejero for helpful technical assistance. This work was supported by the US Department of Energy, Division of Basic Energy Sciences.

## References

Gay T J and Berry H G 1979a *Phys. Rev.* A **19** 952–61

—— 1979b *Proc. Workshop on Physics with Fast Molecular Ion Beams* ed D S Gemmell ANL/PHY-79-3 pp 279–89

Gemmell D S, Remillieux J, Poizat J C, Gaillard M J, Holland R E and Vager Z 1975 *Phys. Rev. Lett.* **34** 1420–5

Green T 1976 *J. Chem. Phys.* **64** 3951–6

—— 1978 *J. Chem. Phys.* **69** 101–5

Hagstrum R D 1977 *Inelastic Ion–Surface Collisions* ed N H Tolk, J C Tully, W Heiland and C W White (New York: Academic) pp 1–25

Schectman R M 1979 *Bull. Am. Phys. Soc.* **24** 581

Schectman R M and Ellis D G 1979 *Proc. Workshop on Physics with Fast Molecular Ion Beams* ed D S Gemmell ANL/PHY-79-3 pp 269–78