Coherence and symmetry breaking in dissociative electron attachment to molecular hydrogen

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Synopsis Momentum images of H^- ions from the 14 eV dissociative electron attachment process in H_2 show strong forward – backward asymmetry while those of D^- from D_2 show much weaker asymmetry along with changes in direction of the asymmetry with electron energy. We explain these observations as due to the break of the inversion symmetry resulting from coherent excitation of odd and even parity resonances and subsequent interference between the two dissociating paths. The smaller asymmetry in D_2 is explained as due to losses from autodetachment and the reversal of the asymmetry as due to change in the relative phase.

Quantum coherence induced interference effects have been shown to break the inversion symmetry in the photoionization of homonuclear diatomic molecules [1, 2]. Such an asymmetry arises from the entanglement of the *gerade* and *ungerade* states and the subsequent loss of inversion symmetry. While these coherence induced effects have been identified in electron emission processes, such effects in electron attachment have not been observed till now. Our measurements using velocity slice imaging of H^- ions show strong asymmetry in the forward – backward direction for the 14 eV resonance in H_2, while this asymmetry is significantly less in D^- from D_2 for the same resonance.

The experiment consists of a velocity slice imaging spectrometer and comprises of a pulsed electron beam of 200 ns duration incident on an effusive jet of molecular hydrogen. The ions formed in the interaction region are extracted into a time of flight region after a delay of 200 ns using a pulsed extraction field [3, 4]. The ions are detected using three 75 mm diameter microchannel plates in Z-stack geometry followed by a phosphor screen and a CCD camera. The central slice of the Newton sphere is captured by applying a 2 kV pulse of 100 ns duration to the detector bias. The slice images of H^- from H_2 and D^- from D_2 at 14.5 eV are given below. In these images the electron beam direction is from top to bottom.

We explain the results in terms of coherent excitation of two resonances of Σ^+_u and Σ^+_g symmetries and the ensuing quantum interference as they take two different paths to the same dissociation limit. The difference between H_2 and D_2 is explained in terms of the larger time for dissociation of D_2 and subsequent faster autodetachment loss one of the states as compared to the other one, resulting in rather weak interference.

Quantum dynamic calculations using model potential energy curves show interference oscillations in the forward-backward asymmetry as a function of electron energy. In the absence of potential energy curves for these resonances we used different sets of curves as a function of the average lifetime of the Σ^+_u state and the known lifetime of the Σ^+_g state available from electron scattering measurements [5]. The calculations indicate that the two curves should be fairly close to each other in order to explain the measured asymmetry in the two istopomers as a function of the electron energy.

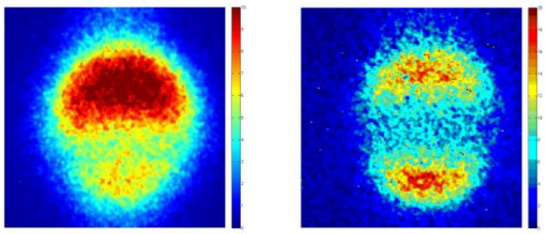

Figure 1. H^- from H_2 at 14.5 eV (left) D^- from D_2 at 14.5 eV (right).

References

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