Autodetachment spectroscopy of the metastable HD^- and D_2^- anions

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The autodetachment spectrum of decaying HD^- and D_2^- has been recorded with a velocity-map imaging spectrometer. Coincident detection of the neutral molecule permits the selection of events occuring in the center of the spectrometer. The discrete features of the spectrum are assigned to transitions between well identified ro-vibrational levels. Energies, lifetimes and branching ratios predicted with the non-local potential framework developed for associative detachment satisfactorily reproduce the experimental results.

The existence of long-lived molecular hydrogen anions was established by accelerator mass spectrometry [1]. Their metastability is a result of fast molecular rotation causing the potential energy minimum of the H_2^- ground state to lie outside the H_2 potential well. A single ro-vibrational level of D_2^- was found to decay significantly by tunneling to D + D^- [2]. All other levels decay via autodetachment, with lifetimes ranging from fs to ms [3, 4]. Coulomb explosion imaging of H_2^- and D_2^- has provided the average internuclear distance distribution [5, 6].

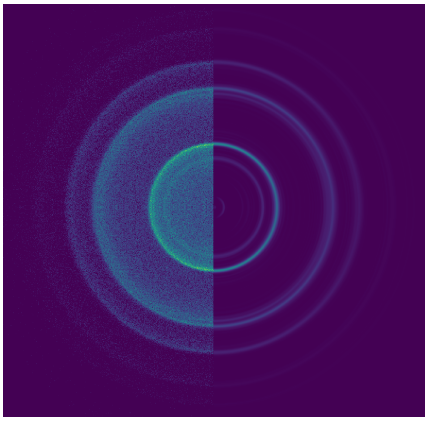

Figure 1: Velocity map of ejected electrons from D_2^-. Left: histogram of hit positions. Right: map after Abel inversion.

In the present study, we directly record the ejected electron energy spectrum by means of a velocity-map imaging (VMI) spectrometer (see Fig. 1). The coincident detection of the electron and its parent molecule permits to locate the emission point, hence to

retrieve regular momentum images. A rich spectrum emerges, that allows for a unique determination of the initial and final ro-vibrational levels. The detachment occurs in the exponential tail of the vibrational wavefunctions, with branching among all accessible vibrational levels of the neutral molecule.

The energies and lifetimes of the relevant HD^- and D_2^- levels were computed earlier in the nonlocal potential scattering framework [4]. This work has been expanded here to take explicitly into account the change in angular momentum imposed by parity conservation. The vibrational branching ratios are satisfactorily reproduced by analyzing the partial cross sections for the associative detachment reaction $H(D) + D^- \rightarrow HD(D_2) + e^-$.

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