Fragmentation dynamics of nitrogen dimers: role of the neighbor and access to the 3D geometry

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Synopsis The fragmentation of molecular nitrogen dimers $(N_2)_2$ induced by collision with low energy 15 qkeV Ar^{9+} ions is studied by mean of the Coulomb explosion imaging technique. The effect of the environment on fragmentation dynamics within the dimer is investigated through the comparison of the KER spectra obtained for the 3-body channels $N_2^+ + N^{m+} + N^{n+}$ from $(N_2)_2$ dimers and those for the 2-body channels $N^{m+} + N^{n+}$ from isolated N_2 molecules [1]. In addition, a complete analysis of fragment momenta in 2-body, 3-body and 4-body fragmentation channels provides first elements for the 3D imaging of the $(N_2)_2$ dimer targets.

Ion collisions with weakly bound systems offer a unique tool to investigate the transition from gas phase isolated atoms/molecules to the condensed phase. Rare gas dimers are of particular interest as they offer a simple system consisting of two neighbor quasi-independent atoms where the electrons remain localized on each atom [2]. On the other hand, clusters of complex molecules have also been extensively studied, showing for example, that the surrounding molecules may act as a protective environment by dissipating the excitation energy among the degrees of freedom of the cluster [3].

Dimers formed by two diatomic molecules are small clusters of intermediate complexity for which the availability of detailed experimental data is still scarce. We consider here the dimer of the simple diatomic N_2 molecule. It constitutes a model system of polyatomic complex containing both covalent and van der Waals bonds. In the present work, $(N_2)_2^{q+}$ ionized dimers are produced in low energy collisions between highly charged ions and $(N_2)_2$ targets. A COLTRIMS (cold target recoil ion momentum spectrometer) set-up is used to measure in coincidence the time of flight and positions of the positively charged fragments resulting from the dimer dissociation. For all events corresponding to a fragmentation channel of interest, the complete momentum of each fragment is reconstructed in the center of mass, giving access to the KER (kinetic energy release) and to the angular correlations between fragments.

The KER spectra for the channels $N_2^+ + N^{m+} + N^{n+}$, with (m,n) = (1,1) or (1,2), were found very similar to those obtained for $N^{m+} + N^{n+}$ from the dissociation of isolated $N_2^{(m+n)+}$ cations (fig. 1). They show a simple energy shift of ~10eV, attributed to the deformation of potential energy curves in the presence of a N_2^+ cation, whereas the relative populations of the different dissociative states populated during the collision remain the same.

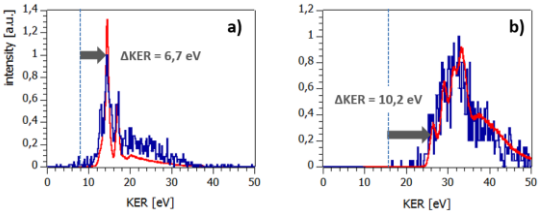

Figure 1. KER spectra for $N^+ + N^+$ (a) and $N^+ + N^{2+}$ (b) channels in the fragmentation of the dimer (blue) and of the isolated molecule (red). The red curves are shifted by respectively 6.7 eV and 10.2 eV.

A detailed analysis of the fragment momenta correlations in 2-, 3- and 4-body fragmentation channels was also performed to reconstruct the 3D geometry of the dimer target and provide stringent tests of theoretical predictions.

References

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