Tomographic measurement of 3D ion velocity distributions from rotationally cold molecules using a kHz VMI spectrometer

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Synopsis: We measure a series of 2D velocity map images of I^+ fragments from laser-aligned iodobenzene and use a filtered back-projection algorithm to reconstruct the full non-cylindrical 3D ion velocity distribution. The use of tomography in VMI is not restricted to any particular symmetry of the velocity distribution.

The use of velocity map imaging [1] to measure 2D projections of 3D ion/electron momentum distributions has found increasingly wide application due to the ease of use and rapid data acquisition rates. VMI is most often used in experiments where cylindrical symmetry in the momentum distribution allows the use of Abel inversion to reconstruct the full 3D distribution. We overcome this constraint on the symmetry of the distribution by measuring a series of 2D projections along different directions in the plane of polarization (the laser polarization vector is rotated to do this), and reconstruct the 3D distribution using a filtered back-projection algorithm.

The method will be demonstrated for non-adiabatically aligned iodobenzene. The molecules are aligned with a linearly-polarized pump pulse, and a probe pulse polarized perpendicular to the alignment axis produces I^+ fragments. Tomographic reconstruction the 3D velocity distribution demonstrates the effect of probe selectivity on the measurement of alignment from 2D projections ($\langle\cos^2\theta_{2D}\rangle$), and makes it possible to measure the 3D value of $\langle\cos^2\theta\rangle$ from a slice of the 3D distribution (the probe applies no torque on the molecules and has no angle selectivity in the v_y=0 plane (Figure 1(a)).

The VMI spectrometer uses a fast phosphor, a CMOS camera and a multi-processor workstation to determine the position of each ion hit with sub-pixel resolution on a shot-to-shot basis at 1 kHz. The fast data acquisition rate allows the 3D measurement to be completed in a reasonable amount of time. A kHz supersonic valve provides a rotationally cold molecular target for laser-induced alignment experiments.

Tomographic reconstruction of 3D momentum distributions should be widely applicable because it can be used for a completely general velocity distribution. It also requires no modifications to a standard VMI spectrometer, only the ability to rotate the velocity distribution in space by simultaneously rotating all the laser polarizations in a collinear setup. The requirement that the VMI electric field doesn't influence the distribution is a potential limitation (for instance, the field might be strong enough to orient the molecules [2]).

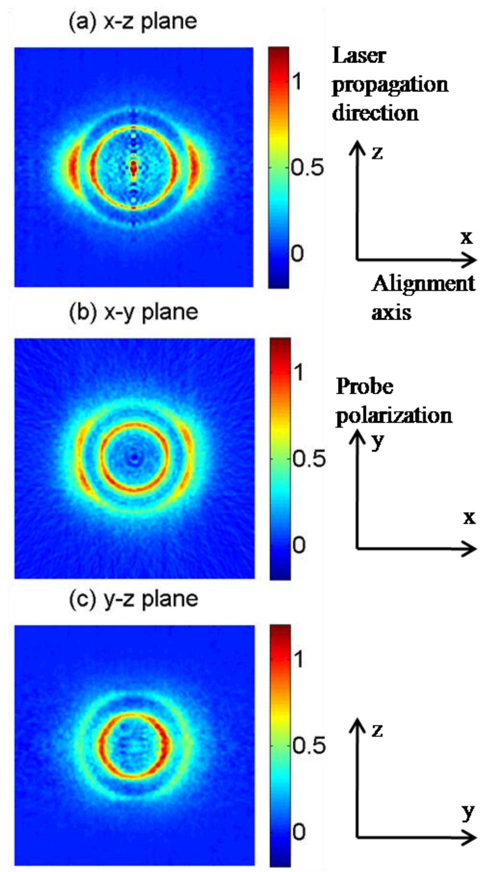

Figure 1: Three representative orthogonal slices of 3D I^+ ion velocity distribution.

References:

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2. L. Holmegaard *et al.*, PRL **102**, 023001-4 (2009).

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