Visualization of ultrafast molecular dynamics is one of the ultimate purposes in physical chemistry because it leads to deep understanding of dynamic properties of molecules. A time-resolved (pump-probe) charged particle imaging for gas-phase molecules is one of the powerful approaches for such a purpose. However, there have been several difficulties in existing imaging techniques. For example, a typical 2D imaging technique suffers from the impractical camera angle, and then it is quite difficult to measure an image in the laser polarization plane, although such an angle is often required to visualize the laser-induced molecular dynamics. 3D imaging was regarded as a versatile method; however, the spatial resolution and data count rate is much lower than those of 2D imaging. Because we have to take many "snapshots" at different pump-probe delay times to make a "molecular movie", low-throughput 3D imaging is not a desirable choice. An application of real-time imaging was therefore limited.

To overcome the above mentioned difficulties, we developed a new "space-slice" tomographic imaging technique. In our setup, 3D ion cloud (Newton sphere) is converted to a 2D tomogram using a mechanical slit and then imaged. By using 2D imaging detector, high spatial resolution and high count rate are achieved while the camera angle limitation was removed. The high count rate of the present method leads to a shorter data acquisition time, making time-resolved imaging study easier.

We applied the present imaging technique to the two important cases as shown below, and succeeded in visualizing the complicated quantum wave nature of the molecular motions. On the basis of high-resolution imaging data, new information on molecular structures and dynamics was also obtained.

(1) Rotational wavepacket of linear molecules

Laser-induced molecular rotation is a typical case in which we cannot utilize the existing imaging techniques. Detailed observation of such a rotational dynamics has never been reported although it is a good system to study quantum molecular motions and it is also used for the orientation control in diffraction studies. We combined our imaging technique with strong laser field-based rotational excitation. Rotational wavepacket of N_2 molecule was created by a train of strong laser pulses (pump process) and then probed with Coulomb explosion imaging. The observed N^{2+} ion distribution directly reflects the instantaneous angular distribution of molecular axis, i.e. squared rotational wave function. Owing to high-resolution of our imaging setup, we succeeded in observing highly complicated nodal structures, which govern the wavepacket dynamics.

(2) Real-time tracking of vibrational/rotational coherence in molecular clusters

Our imaging technique can be also used as a new tool for structural characterization. Nitrogen dimer $(N_2)_2$ is one of the important systems in atmospheric chemistry, however, because of the small permanent dipole, spectroscopic studies are limited. We used femtosecond impulsive Raman pump- Coulomb explosion probe imaging to investigate the structure and dynamics of $(N_2)_2$. This technique gives us instantaneous structural information of a molecular system investigated even when the system does not have a suitable dipole transition. Observed images show periodic rotational and vibrational motions, from which spectroscopic data can be deduced. The static and dynamic structure of $(N_2)_2$, which is governed by the weak intermolecular interaction, is discussed for the first time.