High-resolution, broad-bandwidth spectroscopy of molecular clusters by real-time wave packet imaging

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High-resolution spectroscopy of molecular complexes is one of the powerful tools to study dynamics and structures governed by intermolecular interactions. It is, however, difficult to measure pure rotational (microwave) and intermolecular vibrational (THz) spectra with a single setup, although rich information on intermolecular interaction can be deduced in these frequency regions. Recently, we have developed broad bandwidth (>3 THz), high-resolution (~50 MHz), spectroscopy for weakly-bound molecular clusters. Our method relies on high-precision rotational/vibrational wave packet imaging.[1,2] Rotational/vibrational wave packet of a molecular dimer is created upon broadband femtosecond pump, and subsequent dynamics is tracked by real-time Coulomb explosion imaging. Rotational/vibrational Raman spectrum can be obtained as a Fourier transform of a time trace of an observed wave packet movie (Figure 1).[3,4] We applied the method to Ar_2, $(N_2)_2$, $(CH_4)_2$, $(C_2H_4)_{2,3}$, and other important systems which have been difficult targets of typical microwave spectroscopy. We also succeeded in measuring rotationally-resolved vibrational spectra for the first time. Details on experimental setup and selected results will be presented.

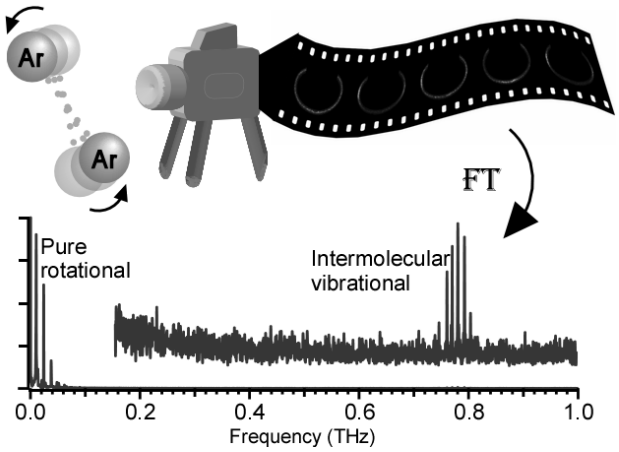

Fig. 1 Schematic of rotational and vibrational wave packet imaging spectroscopy of the argon dimer.

[1] Mizuse, K., Kitano, K., Hasegawa, H., Ohshima, Y. Quantum unidirectional rotation directly imaged with molecules. *Sci. Adv.* **2015**, 1, e1400185.
[2] Mizuse, K., Fujimoto, R., Ohshima, Y. Space-slice ion imaging: High slice resolution imaging in the polarization plane of arbitrarily polarized ionizing light. *Rev. Sci. Instrum.* **2019**, 90, 103107.
[3] Mizuse, K.; Sato, U.; Tobata, Y.; Ohshima, Y. Rotational spectroscopy of the argon dimer by time-resolved Coulomb explosion imaging of rotational wave packets. *Phys. Chem. Chem. Phys.* **2022,** *24*, 11014-11022.
[4] Mizuse, K.; Takano, K.; Kakizaki, E.; Tobata, Y.; Ohshima, Y. Time-Domain Rotational Raman Spectroscopy of the Ethylene Dimer and Trimer by Coulomb Explosion Imaging of Rotational Wave Packets. *J. Phys. Chem. A* **2023,** *127*, 4848-4855.