

Supplementary Information:

Double imaging photoelectron photoion coincidence sheds new light on the dissociation of energy-selected CH_3Cl^+ ions

Xiaofeng Tang,^{1,2*} Xiaoxiao Lin,¹ Weijun Zhang,^{1,3} Gustavo A. Garcia² and Laurent Nahon²

1, Laboratory of Atmospheric Physico-Chemistry, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei, 230031 Anhui, China.

2, Synchrotron SOLEIL, L'Orme des Merisiers, St. Aubin BP 48, 91192 Gif sur Yvette, France.

3, School of Environmental Science and Optoelectronic Technology, University of Science and Technology of China, Hefei, 230026 Anhui, China.

* tangxf@aiofm.ac.cn

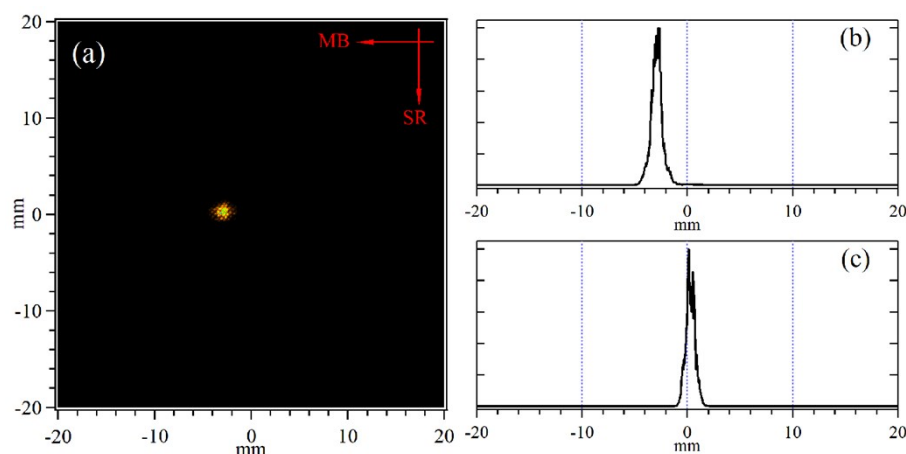


Figure S1. (a) CH_3Cl^+ ion image and its intensity projections along the directions of (b) x-axis (MB, molecular beam) and (c) y-axis (SR, synchrotron radiation). The polarization of the synchrotron light is linear and along the MB axis. The ion image is displaced along the horizontal axis due to the speed of the molecular beam. A translational temperature of the molecular beam is calculated to be ~ 25 K (Atomic and Molecular Beam Methods, Vol. 1, Oxford University Press, New York, 1988, pp. 14-54).

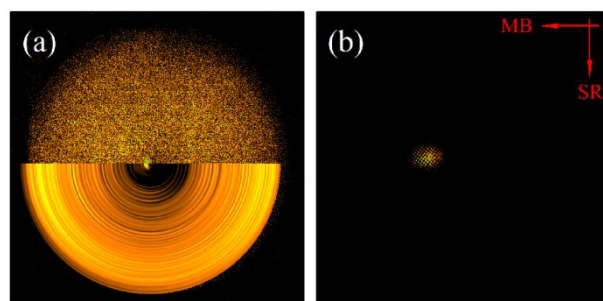


Figure S2. Mass-selected (a) electron image and (b) ion image corresponding to CH_2Cl^+ fragment ions recorded at $h\nu = 14.40$ eV. The upper half part of electron image represents the raw data and the lower corresponds to the result from the pBasex inversion algorithm (*Rev. Sci. Instrum.*, 2004, **75**(11), 4989-4996). The polarization of the synchrotron light is linear and along the MB axis. The ion image is displaced along the horizontal axis due to the speed of the molecular beam.

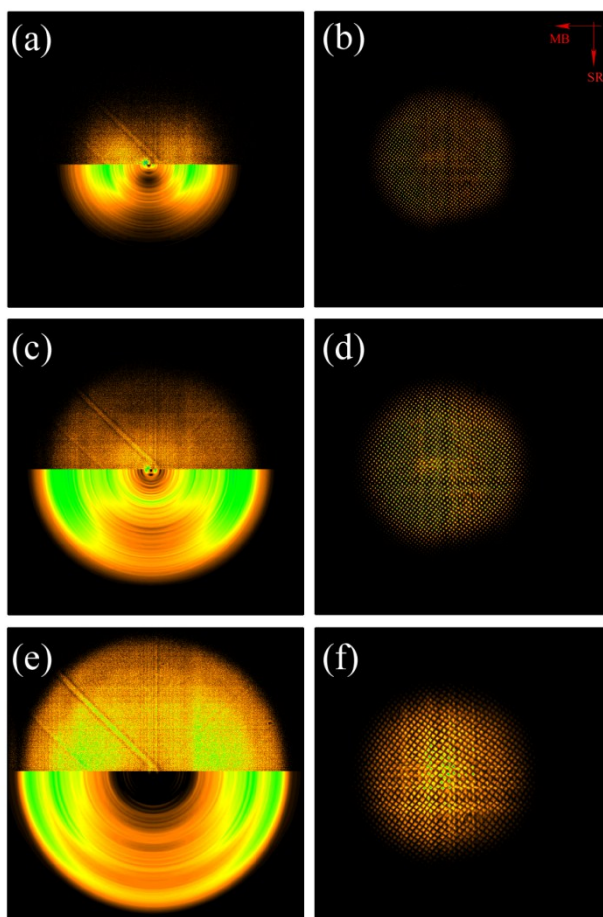


Figure S3. Mass-selected electron images (left) and ion images (right) corresponding to CH_3^+ fragment ions recorded at fixed photon energies of (a, b) 14.40 eV, (c, d) 15.40 eV and (e, f) 16.80 eV. The upper half electron image represents the raw data and the lower

corresponds to the result from the pBasex inversion algorithm (*Rev. Sci. Instrum.*, 2004, **75**(11), 4989-4996). The asymmetry parameters of electrons corresponding to the ionization into the A^2A_1 state have also been inferred from the electron images to be 0.9 ($h\nu=14.40$ eV), 0.7 ($h\nu=15.40$ eV) and 0.3 ($h\nu=16.80$ eV) showing a clear dependence with the photon energy, with a decreasing trend in reasonable agreement with previous calculated results (*J. Chem. Phys.* 2001, **115**, 4593).