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3D-ion-momentum/high-resolution-electron coincidence measurements

G. Prümper[a,*], K. Ueda[a], U. Hergenhahn[a,d], A. De Fanis[b], Y. Tamenori[b], M. Kitajima[c], M. Hoshino[c,1], H. Tanaka[c]

[a] *IMRAM, Tohoku University, Sendai 980-8577, Japan*
[b] *JASRI, Sayo-gun, Hyogo 679-5198, Japan*
[c] *Department of Physics, Sophia University, Tokyo 102-8554, Japan*
[d] *Max-Planck-Institut für Plasmaphysik, 85748 Garching, Germany*

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Abstract

We have set up a coincidence apparatus consisting of a high-resolution electron spectrometer and a *k*-resolving ion spectrometer. As an example for measurements performed with this apparatus, we present ion mass spectra that are coincident with energy-resolved resonant Auger electrons emitted from F 1s excited SF_6 molecules and resonant Auger electron spectra that are coincident with mass-selected fragment ions, as a function of the detuning from the $F(1s^{-1})a_{1g}^*$ resonance in SF_6. Significant changes of these spectra due to detuning suggest that a substantial fraction of the excess photon energy goes into fragmentation rather than into electron kinetic energy.

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1. Introduction

Dissociation pathways from core-excited and ionized molecules have been investigated extensively in the last two decades. One of the most exciting findings in this field is that bond breaking of a core-excited polyatomic molecule often takes place near the excited atomic site. This site-specific fragmentation was first demonstrated by Eberhardt et al.: they found that only O ions are produced when the C 1s electron in the CO site in a $(CH_3)_2CO$ molecule is excited to the LUMO π^* [1]. In order to know a state-selected dissociation pathway, however, an energy-resolved Auger electron and ion coincidence technique is demanded [2–5]. The first demonstration of such experiments was performed by Eberhardt et al. for the N_2 molecule [2]. Later on Miron et al. successfully employed this technique to investigate site-specific fragmentation of the $ClCH_2Br$ molecule [6]. Ueda et al. [7], Morin et al. [8] and Kukk et al. [9] used this technique to investigate the influence of the nuclear motion in the core-excited state to the molecular dissociation.

In order to study the influence of nuclear motion in the core-excited state on the molecular dissociation in further detail, we have set up a system for coincidence measurements consisting of a high-resolution electron spectrometer and a *k*-resolving ion spectrometer. In the present work, fragmentation of the SF_6 molecule following F 1s $\rightarrow$ a_{1g}^* excitation is studied by the coincident measurement of the resonantly emitted electron and the fragment ion. A great variety of dissociation pathways are possible for this polyatomic molecule. With our apparatus we are able to disentangle the electron spectra with respect to the dissociation pathways and we can study the pattern of fragmentation for specific parts of the electron spectrum. The results recorded in coincidence with the atomic-like F(1s) Auger lines [10] are reported and discussed in detail elsewhere [11].

* Corresponding author.
[1] Present address: RIKEN, Wako, Saitama 351-0198, Japan.
E-mail address: pruemper@tagen.tohoku.ac.jp (G. Prümper).

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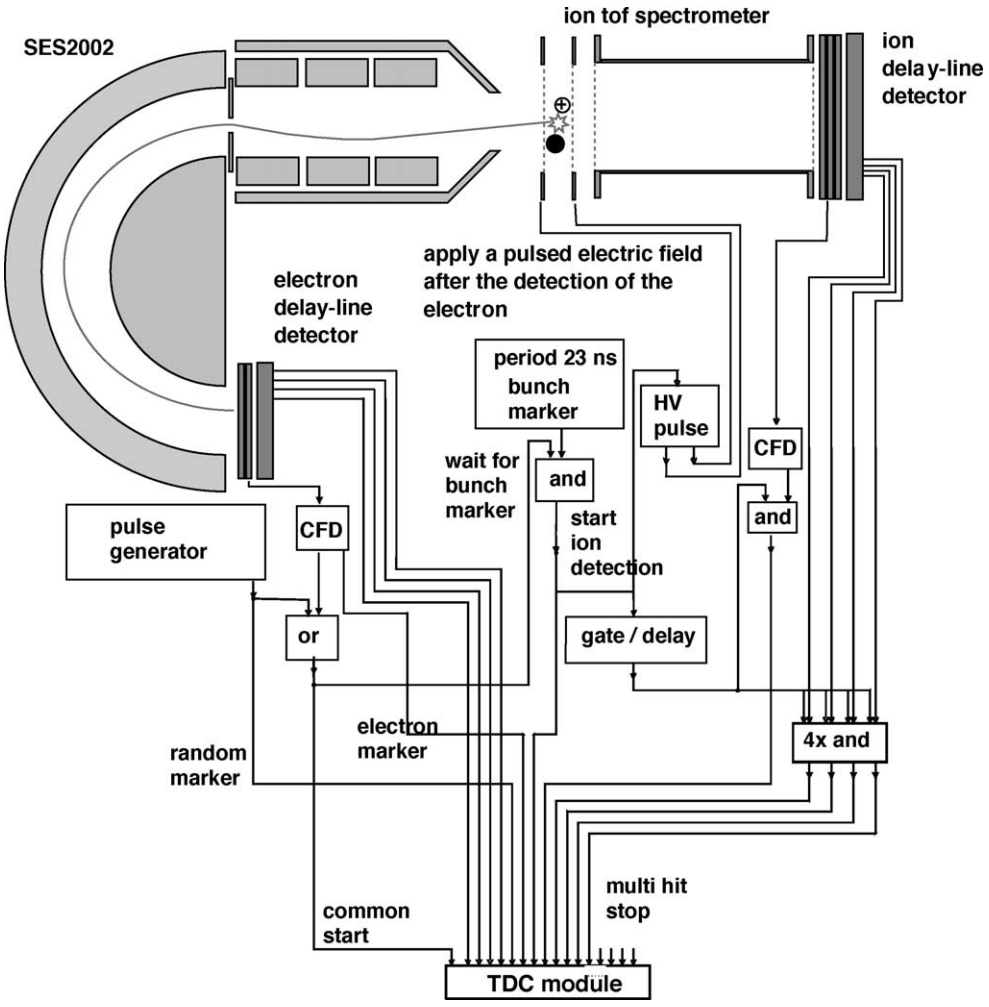

Fig. 1. A schematic diagram of the experimental setup including the data acquisition system. All signals from the two delay line detectors are feed into a 16-channel TDC system. See text for the details.

2. Experimental

Fig. 1 is a schematic of the setup including the data acquisition system. The electron and ion spectrometers are mounted inside a vacuum chamber. The sample gas (SF_6 in the present study) is introduced between the pusher and the extractor electrode of the ion spectrometer through a grounded copper needle. After ionization of the sample molecule (SF_6) some of the electrons pass the pusher electrode and enter the electron analyzer (Gammadata Scienta SES-2002). In order to make coincidence experiments possible, the standard CCD camera of the electron spectrometer was replaced by a delay line detector with an active diameter of 40 mm (Roentdek DLD40 [12]). During the coincidence experiment all voltages of the electron spectrometer were fixed. Four channels of a 16-channel time to digital converter (TDC) were used for the recording of the position information of the electron detection. The common start for the TDC comes from the electron detector.

Triggered by the electron detection, rectangular high-voltage pulses with opposite sign are applied to the pusher and extractor electrodes. The ion drift tube was held at a constant voltage. The electrodes of the ion spectrometer have 90% transmission copper grids (8 lines/mm) to produce homogenous electric fields between the different electrodes.

The timing, quality and stability of the high-voltage pulses is crucial for the measurement. In particular, the time between the trigger signal and the output of the HV pulse must not exceed some 100 ns, otherwise the fast ionic fragment might leave the region between the pusher and extractor electrode and thus is lost for the detection. The electronic insertion delay of the pulse generator [13] was less than 120 ns with rise/fall (10%/90%) times less than 15 ns. The ions are detected by another delay line detector with an active diameter of 80 mm (Roentdek DLD80 [12]). Four channels of the 16-channel TDC were used for the recording of the position information of the ion detection. The ion side of the detector needs electronic gates to mask the noise induced by the high-voltage-pulse generator (see Fig. 1).

The experiment was conducted on the c-branch of the high-resolution photochemistry beamline 27SU [14,15] at SPring-8, in Japan. The radiation source is a figure-8