Multiple Auger-Electron Emissions from Core-Excited States in N_2

Decay dynamics of core-excited states in N_2 were studied in great detail by a multi-electron coincidence technique combined with the pulse property of the synchrotron light.

Resonant photoexcitation of a core-electron in atoms or molecules into an unoccupied valence or Rydberg orbital can take place just below its K-shell ionization threshold (IP_k), which gives pre-edge structures in XANES. The core-excited states decay mainly by Auger electron emissions for the case of atoms or molecules with low-Z elements, where the electron promoted in the initial photoexcitation can either participate or behave like a spectator, leading to a variety of decay routes as compared with the Auger processes of the core-ionized states formed above IP_k. Such multiple electron processes provide good opportunities for investigating electron correlation effects.

In the decay of such core-excited states, more than two Auger electrons may be emitted simultaneously (direct multiple Auger process) or sequentially (cascade multiple Auger process). The dynamics of these multiple Auger decays is more complicated and interesting for molecules than atoms since it additionally contains nuclear degrees of freedom as demonstrated in CO by Kaneyasu *et al.* [1]. However, the multiple Auger decays of molecular core-excited states remain little studied because coincidence measurements are required for a detailed understanding of the mechanisms. In the present study we measured the electron-energy correlations of the Auger electrons emitted in the double and triple Auger transitions of core-excited N_2.

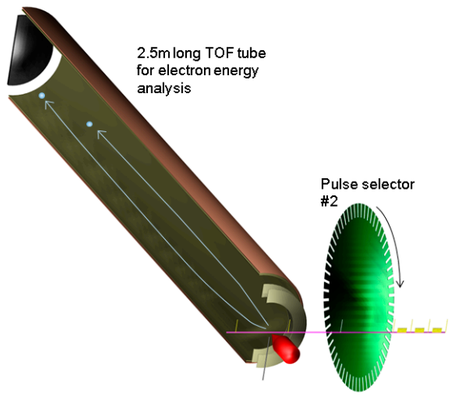

Figure 1: Schematic of the experiment. Isolated light pulses passed through the pulse selector #2 cross a beam of gas from a capillary. Near the interaction region a permanent magnet (red colored) is placed, forming an inhomogeneous magnetic field for the electron detection with extra-high efficiency.

A schematic of the experiment is shown in Fig. 1. The measurements were carried out under the hybrid-fill mode of the PF ring in which half of the storage ring was filled by a train of low-current bunches and an isolated high-current bunch was located at the center of the opposite half. The light pulses were chopped out by using the pulse selector #2 [2] to obtain isolated light pulses with the repetition rate of 146 kHz. The Auger electrons were collected by an inhomogeneous magnetic field, energy-analyzed by a 2.5-m long TOF tube and detected in coincidence by a time-to-digital converter. The collection efficiency of the electrons was nearly 100% for kinetic energies less than 400 eV, which is a great advantage for multi-electron coincidence measurements.

Figures 2(a) and 2(b) represent the energy correlations between the two Auger electrons in the double Auger processes after the core excitations at 401.08 eV ($1s \rightarrow \pi^*$) and 406.15 eV ($1s \rightarrow 3s\sigma$) in N_2. The kinetic energies of the fast electron KE_{fast} were converted to the binding energies of singly charged states by subtracting them from the photon energy, i.e. $h\nu - KE_{fast}$. The bottom and left panels represent projections on each axis. The features that are the most prominent in the energy correlations are attributed to electrons emitted in autoionizations of superexcited neutral N atoms which are produced in the cascade double Auger processes: $N_2^* \rightarrow N_2^+ + e^-(fast)$, followed by $N_2^+ \rightarrow N^* + N^+$, and then by $N^* \rightarrow N^+ + e^-(slow)$. It is noted that the molecular dissociations proceed before the electronic relaxations complete. The shaded areas in the left panels indicate the estimated fraction of the direct double Auger decays. By integrating the whole KE_{slow} range the fractions are estimated to be approximately 34 and 24% for the π^* valence and the $3s\sigma$ Rydberg excited states, respectively.

Careful analysis of numbers of *n*-electron coincidence events enables us to obtain branching ratios between single, double and triple Auger decays for each core-excited state, which are shown in Table 1. The ratios change dramatically between the normal ($h\nu > IP_K$) and resonant core-excitation ($h\nu < IP_K$) processes: probabilities of the multiple Auger processes are much greater for the resonant core-excitation. Moreover, the ratios are also different from each other among the resonant processes. The differences come from the existence of spectator Auger decays in the first-step ionization of the cascade multiple Auger processes for the resonant excitations. It is interesting that the weakly bound excited electrons can modulate the decay of the molecular states with a core-hole. We also succeeded in obtaining branching ratios between final ions for each of the multiple Auger decays (see details in [4]).

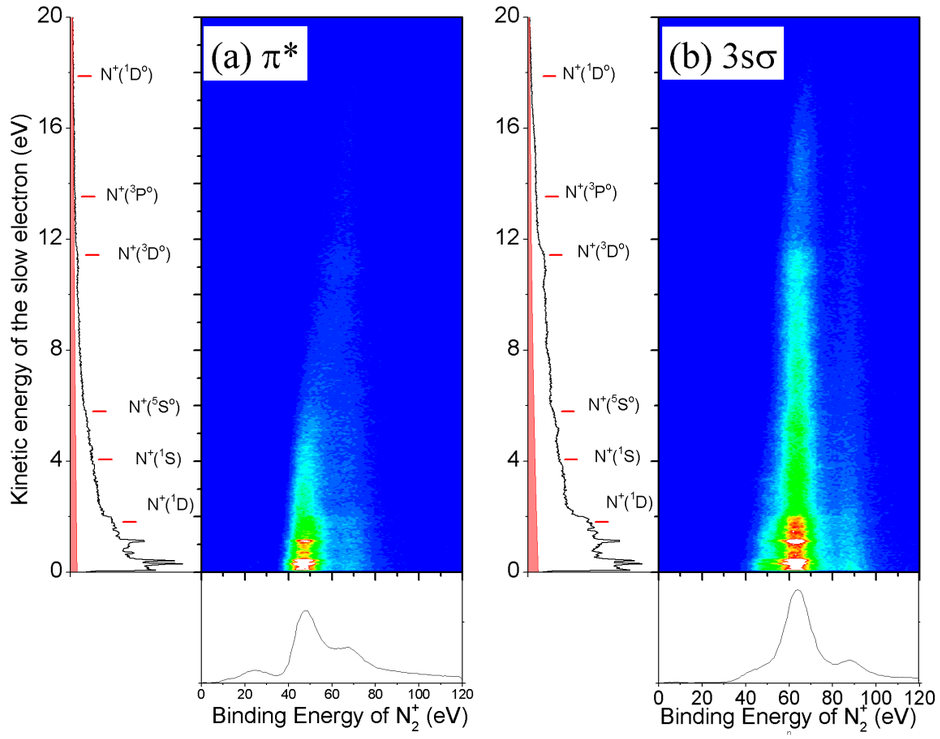

Figure 2: Electron-energy correlations in double Auger decays of the resonant core-excited states of N_2: (a) $1s \rightarrow \pi^*$, (b) $1s \rightarrow 3s\sigma$. Short bars in the left panels represent the energies of N^+.

Table 1: Branching ratios between single, double and triple Auger decays for the core-ionized [3] and core-excited states of N_2.

	π^*	$3s\sigma$	Normal [3]
$h\nu$ / eV	$IP_K - 8.86$	$IP_K - 3.84$	$> IP_K$
Single Auger	67 ± 4	46 ± 4	90.6
Double Auger	30 ± 3	48 ± 3	9.4
Triple Auger	3 ± 1	6 ± 1	–

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