

Auger Decays from Highly-Excited Vibrational Sublevels in Core-Ionized N₂ Molecules

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1 Introduction

Photoelectron–Auger electron spectroscopy is a powerful method for disentangling complicated Auger decay paths following core photoionization of matter. With this method, the photoelectron and Auger electron emitted in each ionization event are detected in coincidence and their kinetic energies are recorded, which enables identification of the Auger decay paths starting from a specific core-hole state. In this report [1], we present an experimental study on Auger decays from highly excited vibrational sublevels ($v \geq 5$) of a core hole state by multielectron coincidence spectroscopy using a magnetic bottle time-of-flight (TOF) spectrometer. The aspects of Auger decay starting outside the Franck–Condon region of direct photoionization are studied.

2 Experiment

The experiment was performed at the BL-2B soft X-ray beamline. The combination of the hybrid-fill operation of the storage ring and the pulse selector apparatus [2] provided pulsed light at a repetition rate of 146 kHz. Multielectron coincidence spectroscopy was performed using the magnetic-bottle electron spectrometer [3]. The spectrometer was equipped with a strong permanent magnet (~ 0.7 T) and a 2.5 m-long flight tube, which enabled full solid angle detection and TOF measurements of the electrons emitted following the photoionization of N₂ molecules.

3 Results and Discussion

The double-excitation resonance lying above the K-shell ionization threshold was used to prepare highly excited vibrational sublevels of the core-hole state of the N₂ molecules. The autoionization decays from the doubly excited states lead to the production of vibrationally excited core-hole states [4] because of nuclear motion on the repulsive potential energy curves. These highly excited vibrational sublevels were hardly populated by direct photoionization of N 1s electrons because of the weak overlap between the vibrational wave functions in the ground and core-hole states. The multicoincidence dataset was

recorded at a photon energy of 415.7 eV, where the core-valence doubly excited states lie.

Figure 1 shows the kinetic energy correlations between the N 1s photoelectrons and the Auger electrons. The top panel shows a photoelectron spectrum obtained by projecting the coincidence counts onto the horizontal axis. The photoelectron peak shows a long tail on the lower-kinetic-energy side, which corresponds to vibrational progression.

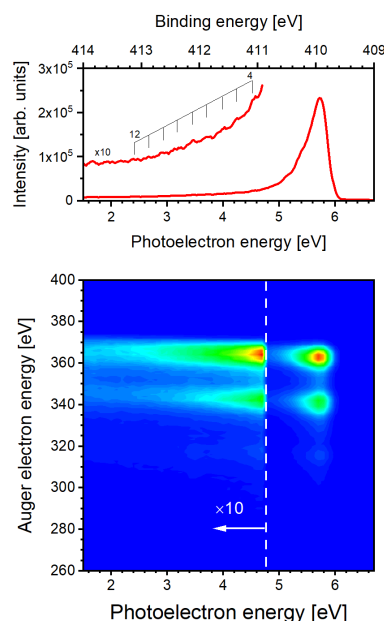


Fig. 1: Two-dimensional map of the kinetic energy correlation between N 1s photoelectrons and Auger electrons, measured at a photon energy of 415.7 eV.

We depict in Fig. 2 the coincidence Auger spectra derived from the two-dimensional map by selecting the photoelectron energy regions with binding-energy intervals of 409.6–411.0, 411.0–412.0, 412.0–413.0, and 413.0–414.0 eV, which correspond mainly to the vibrational sublevels $0 \leq v \leq 3$, $4 \leq v \leq 7$, $8 \leq v \leq 11$, and $v > 12$, respectively. The three band structures

in the spectra are assigned to the formation of doubly charged states with two holes in the valence orbitals.

A change in spectral shape is clearly observed in the coincidence Auger spectra in Fig. 2. The three band structures appear to become broader and less resolved as the binding energy of the core-hole state increases. These features can be essentially interpreted in terms of Franck–Condon overlap between the core-hole states and Auger final states. Most of the Auger final states have repulsive potential energy curves; therefore, the probability distribution for the vibrational wave function of the core-hole state is reflected in the energy space because of the reflection principle. The vibrational wave function for a higher vibrational level has a wider internuclear distance distribution. The spectral profile for the Auger decay is therefore expected to become broader for such higher vibrational levels of the core-hole state.

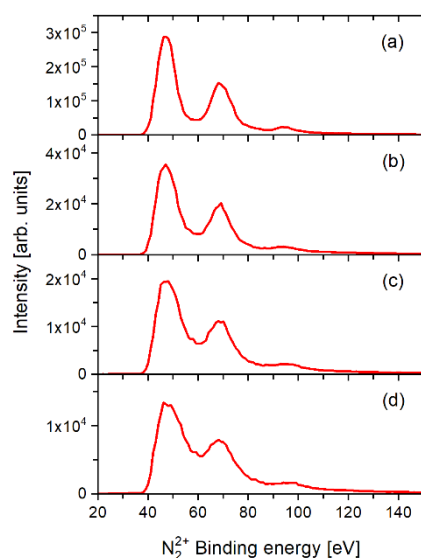


Fig. 2: Coincidence spectra for single Auger decays associated with binding-energy intervals of (a) 409.6–411.0 eV, (b) 411.0–412.0 eV, (c) 412.0–413.0 eV, and (d) 413.0–414.0 eV, respectively. The spectra are plotted as a function of the N_2^{2+} binding energy.

References

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