1 Atomic and Molecular Science

1-1 Coincidence Studies of Core-Valence Double Photoionization

Double photoionization (DPI) of atoms and molecules has attracted special attention for a long time because this process is due entirely to electron correlation and, consequently, investigations of DPI are expected to reveal fundamental aspects of atomic and molecular physics. Until now, most DPI studies have concentrated on the removal of two valence electrons. Direct experimental observations of core-valence DPI have been limited to a few cases. In inner-shell photoelectron spectra, the formation of such states can be seen as the background enhancements lying at the converging limits of photoionization satellite states. However, investigations using conventional photoelectron spectroscopy offer neither direct insight into the doubly-ionized states nor the detailed DPI dynamics. By contrast, coincidence detection between the two photoelectrons emitted in DPI processes provides direct spectroscopic information on DPI processes themselves; however, a sophisticated coincidence method is required, because the DPI cross section is low compared with the main inner-shell ionization processes and, consequently, the events associated with these DPI processes are easily hidden behind ordinary inner-shell photoionization events. We have introduced a very efficient coincidence technique, the magnetic bottle time-of-flight electron-coincidence method, into the investigation of DPI associated with the removal of a core electron. The powerful capabilities of this coincidence method in electron coincidence observations have recently been described [1-3].

A multi-electron coincidence data-set for Ne was accumulated at an excess energy of 108.7 eV above the Ne 1s ionization threshold at 870.09 eV using ARNE1B. Figure 1(a) shows the photoelectron spectrum, where the energy range is adjusted to show the photoelectron peaks associated with the $1s^{-1}$ state and the satellite states. Figure. 1(b) displays a two-dimensional (2D) map showing the coincidences between the photoelectrons and slower electrons in the 0-30 eV energy range. Two intense diagonal stripes in the fast electron energy range of 30-60 eV are due to coincidence between two photoelectrons emitted through the formation of Ne^{2+} $1s^{-1}2p^{-1}(^{1,3}P)$ states by DPI. The available energy to form each Ne^{2+} state, given by (photon energy)−(binding energy of the Ne^{2+} state), is shared by the two photoelectrons emitted, and the corresponding coincidence yield thus results in the diagonal lines. The intensity distributions along the two diagonal stripes describe how

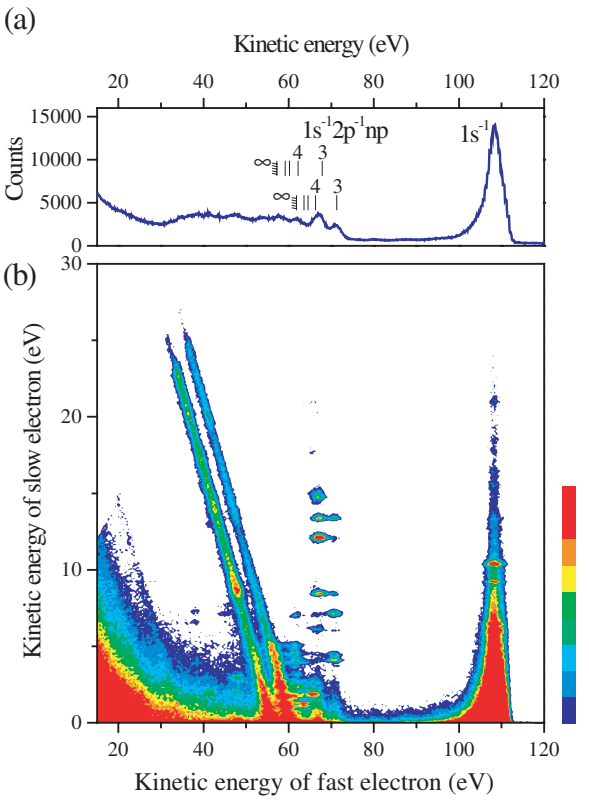

Figure 1
(a) Inner-shell photoelectron spectrum of Ne obtained at a photon energy of 978.8 eV. (b) Two-dimensional map of all coincidence pairs, represented as a function of the kinetic energies of fast and slow electrons. Coincidence intensities in the two dimensional map are plotted on a linear scale.

the two photoelectrons produced in the DPI processes share the available energy. Resonance structures corresponding to Ne^{+} $1s^{-1}2s^{-1}(^{1,3}S)$ ns are discernible on both distributions corresponding to the $1s^{-1}2p^{-1}$ $(^{1,3}P)$ DPI continua: interactions and interference of the Ne^{+} states with the DPI continua can be discussed in terms of the resonance features.

To reveal the Ne^{2+} states with a 1s core hole more clearly, the coincidence counts on the 2D map are integrated and projected on the diagonal axis X=Y in order to present the sum of the energies of fast and slow electrons for all coincident events. This projection is shown as a function of the total energy of the two electrons in Fig. 2. The high statistics resulting from the integration enables us to find, in addition to the $1s^{-1}2p^{-1}$ peaks, two weak peaks associated with the $1s^{-1}2s^{-1}$ states that are faintly visible on the 2D map. This is the first direct spectroscopy of the core-valence DPI states of Ne [4]. The intensity ratio of the ^{3}P and ^{1}P peaks differs significantly from the statistical value of $^{3}P/^{1}P = 3$. This observation manifests the breakdown of the sudden approximation near the DPI thresholds, which motivates us to advance our understanding of the DPI dynamics.

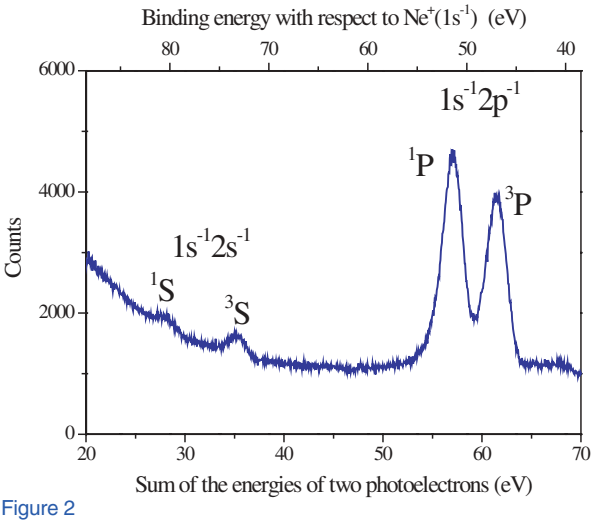

Figure 2
Histogram of the kinetic energy sum for two electrons detected in coincidence, as deduced from Fig. 1(b) by integrating the yields along the direction where for (fast electron energy) + (slow electron energy) = constant. The top abscissa gives the binding energies of the core-valence doubly ionized Ne^{2+} states with respect to the Ne^{+} $1s^{-1}$ state.

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1-2 Photon Pairs Produced from Multiply Excited Molecules in the Vacuum Ultraviolet Range

Molecular multiple excitation resonances in the extreme ultraviolet regime are highly correlated systems in which the electronic and nuclear motions strongly couple due to interactions with many electronic continua. Their formation and decay is hence one of the most interesting subjects today in atomic and molecular science. We have recently proposed a photon-photon coincidence experiment to observe the multiply excited states of molecules: cross sections for the generation of a pair of photons emitted by neutral fragments from the multiply excited molecules are measured as a function of incident photon energy. Such a cross section curve is free from ionization and thus the features originating from the multiply excited states are highlighted.

The cross sections for the generation of a pair of two Lyman-α photons in the photoexcitation of H_2 were measured at BL-20A for the first time, which are shown as a funtion of incident photon energy in Fig. 3 [1]. The spectral feature in Fig. 3 is attributed to the doubly ex-

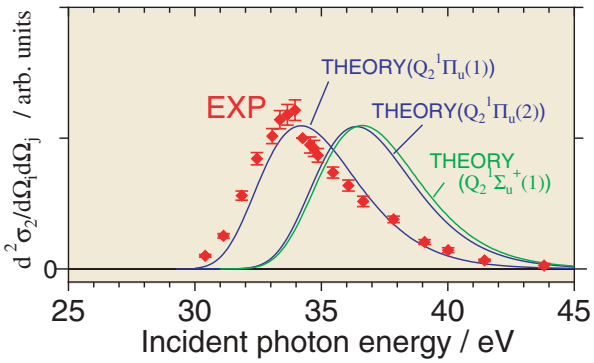

Figure 3
The cross sections for the generation of a pair of two Lyman-α photons in photoexcitation of H_2 as a function of incident photon energy. The calculated cross sections are shown as well (see text). The calculated curve for the Q_2 $^1\Pi_u(1)$ state is normalized to the experimental cross section around 34 eV and other calculated curves are normalized to the same height as this curve.

cited states of H_2. They are clearly Q_2 states built on the $(2p\pi_u)$ core leading to $H(2p)+H^+$. Taking into account also Wigner and Witmer correlation rules for connecting separated atoms and molecular states [2], a Q_2 $^1\Pi_u$ state of H_2 is expected to have important contribution to the cross section curve in Fig. 3. This has been confirmed by a theoretical calculation (solid curves in Fig. 3) based on the reflection approximation and semiclassical treatment for the dynamics: the experimental cross section curve agrees well with the calculated one for the lowest state in the Q_2 $^1\Pi_u$ series, i.e., the Q_2 $^1\Pi_u(1)$ state, of H_2. It has hence turned out that the decay dynamics of the Q_2 $^1\Pi_u(1)$ state is well described in the semiclassical manner.

Figure 4 shows the cross sections for the generation of a pair of two vuv photons in photoexcitation of N_2 [3]. The cross section curve reveals, for the first time, multiply excited states of N_2 in the energy range where the ionization from innermost valence electrons takes place: the four multiply excited states of N_2 were newly found. It is remarkable that the 45-eV peak is above the double ionization potential of N_2 (43.0 eV [4]). The precursor excited state of N_2 for the 45-eV peak thus turns out to be embedded in the double ionization continua. It is

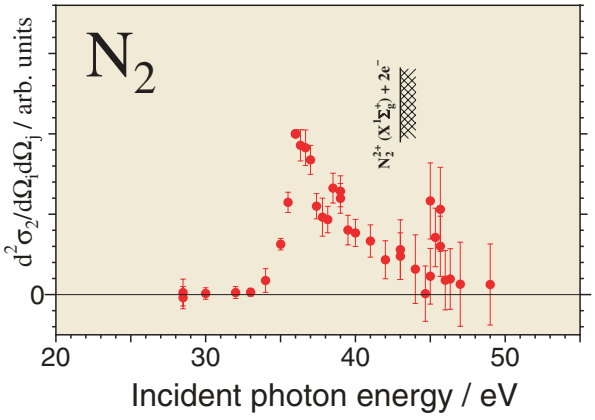

Figure 4
The cross sections for the generation of a pair of two vuv photons in photoexcitation of N_2 as a function of incident photon energy. The double ionization potential of N_2 is shown by the hatched bar.

Be+
convoluted Fano profile fitting

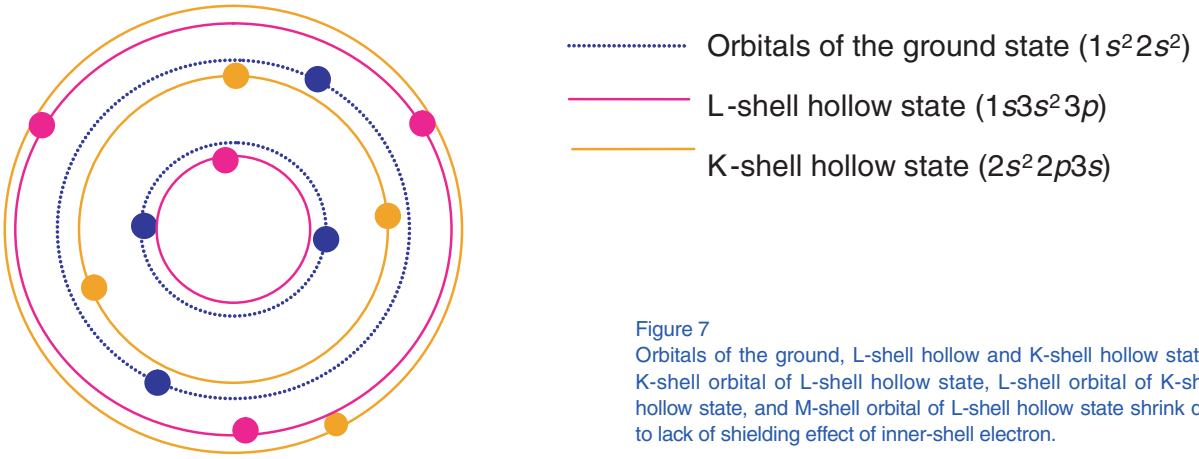

Figure 7
Orbitals of the ground, L-shell hollow and K-shell hollow states. K-shell orbital of L-shell hollow state, L-shell orbital of K-shell hollow state, and M-shell orbital of L-shell hollow state shrink due to lack of shielding effect of inner-shell electron.

Compared with the ground state, the orbitals of the hollow states shrink. Additionally, the transition probability from the ground state ($1s^2 2s^2$) to the $1s3s^2 3p$ state is nine times as large as that to the $2s^2 2p3s$ state. This implies that the transition probability for three-electron excitations is substantially larger than that for two-electron excitations. This is attributed to large overlaps between the 2*s*-electron wave function in the ground state and the 3*s*-electron wave function in the L-shell hollow state. Further experimental investigations are required to investigate the transition probabilities.

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