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(γ, 2γ) studies on N_2 revealing multiply excited states embedded in the double ionization continuum

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Introduction

Molecular multiply excited 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 of great importance in collision physics. However, such resonances in inner valence range have been hardly observed by ordinary photoabsorption or photoionization spectroscopies because direct ionization processes dominate the spectra. Recently we have developed *(γ, 2γ) method* for investigating the formation and decay of multiply excited resonances: two fluorescence photons from fragment atoms, hν' and hν" in photodissociation process (1), are detected in coincidence to eliminate direct ionization and observe spectral features due to multiply excited states [1-3].

$AB+h\nu \rightarrow AB^{**} \rightarrow A^*+B^* \rightarrow A+h\nu'+B+h\nu''$ (1)

In the present study the (γ, 2γ) method has been applied to molecular nitrogen.

Experimental

The measurements were carried out at BL-20A. The linearly polarized synchrotron radiation from the 3-m normal incidence monochromator was introduced in a gas cell filled with N_2 with the pressure of approximately 0.13 Pa. Two VUV-photons emitted parallel to the electric vector of the incident light and opposite to each other were detected by two photon detectors, each of which is composed of an MgF_2 window and a microchannel plate providing a filter range of 115-150 nm. The coincidence counts were normalized for gas pressure, flux of the incident photons, detection efficiencies and geometrical factor to obtain cross sections for the emission of two photons (process (1)) in N_2 differential with respect to solid angles for the photons, $d^2\sigma/d\Omega_i d\Omega_j$ [3].

Results

Figure 1 shows the doubly differential cross sections for the emission of two photons of N fluorescence as a function of the incident photon energy. The short vertical lines in the range 30.42-37.15eV in the figure indicate the dissociation limits of N*+N*' which give two fluorescence photons detectable by the present photon detectors.

The doubly differential cross section as a function of the incident photon energy rises above approximately 33 eV and shows peak structures at 36, 39, and 45 eV and a broad structure around 40-44 eV. Photoelectron spectra in this energy range reveal the $(2\sigma_g)^{-1}$ N_2^+ states and the correlation states associated with them [4,5]. Hence the 36, 39 and 40-44 eV peaks can be attributed to multiply excited states of N_2 built on these ionic states [3]. It is remarkable that the 45 eV peak is above the double ionization potential of N_2 (43.0 eV [6]). The precursor excited state of N_2 for the 45 eV peak thus turns out to be embedded in the single and double ionization continua. It is interesting that such a highly excited state of N_2 dissociates into neutral fragments escaping from the electronic autoionization and makes a contribution comparable to those below the double ionization potential.

References

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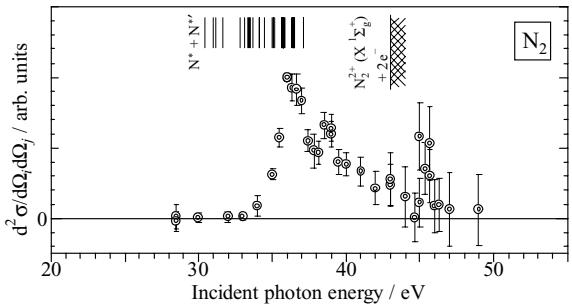

Figure 1. Doubly differential cross sections for the emission of two photons of N fluorescence as a function of the incident photon energy. The vertical line with crossed hatch indicates the double ionization potential of N_2 [6]. The short vertical lines indicate the dissociation limits of N*+N*' (see text).

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