$(\gamma, 2\gamma)$ studies on multiply excited states of molecular nitrogen

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Introduction

The dynamics of electrons and nuclei in a multiply excited molecule is a subject of great interest because the independent electron model and Born-Oppenheimer appoximation would probably be much less available than for ground elecronic states and lower excited states. We mesuerd the relative values of the cross sections for the emission of the VUV fluorescences from excited nitrogen atoms produced in the photoexcitation of N₂ as a function of inicident photon enegry and revealed the doubly excited states in the range 22-23 eV [1]. However, the contribution from the dissociative direct-ionization leading to the emission of the atomic fluorescence dominates the fluorescence cross section curve in the energy range higher than approxmately 35 eV [2] and thus prevents the resonance peaks attributed to higher multiply excited states from being observed. Recently we have establised the $(\gamma, 2\gamma)$ method to overcome this difficulty: two fluorescence photons from neutral fragments are observed in coincidence to eliminate the contribution from ionization from ionization processes [3]. In the present study the $(\gamma, 2\gamma)$ experiments were carried out for molecular nitrogen and obtain cross sections for following process,

$$N_{2} + h\nu \to N_{2}^{**}$$

-> N* + N*'
-> N + N + hv' + hv'', (1)

differential with respect to solid angeles for the two photons, $d^2\sigma_2/d\Omega_i d\Omega_j$, to reveal higher doubly or mutiply excited states of N₂.

Experimental

Experiments were carried out at BL20A equipped with a 3-m normal incidence monochromator. A bandpass of the wavelength of the incident light was 0.167 nm: the energy width of 185 meV at 37 eV incident photon energy. Two VUV-photon detectors were placed opposite to each other and aligned on the line parallel with the electric vector of the linearly-polarized incident light. Each photon detector is composed of a microchannel plate and an MgF₂ window providing filter range of 115-150 nm. Pulses from the two detectors were fed into a standard delayed-coincidence circuit. True coincidence rates were normalized for the target number density and incident photon flux to obtain the relative values of the $d^2 \sigma_2 / d\Omega_i d\Omega_i$. The relative values of the singly differential cross sections for the emission of hv' or hv'', $d\sigma_1/d\Omega_1$, were also measured by using one of the photon detectors.

Results

In figure 1, the $d\sigma_l/d\Omega_i$ (a) and the $d^2\sigma_2/d\Omega_i d\Omega_j$ (b) are plotted as a function of incident photon energy. The $d\sigma_l/d\Omega_i$ curve (a) is dominated by the dissociative directionization with excitation leading to the emission of atomic fluorescences. On the other hand, the $d^2\sigma_2/d\Omega_i d\Omega_j$ curve (b) is attributed to the process (1), and thus it follows that unknown doubly or multiply excited states of N₂ above about 40 eV have been observed.

References

[1] M. Ukai et al., Phys. Rev. A 46, 7019 (1992)

- [2] A. Ehresmann et al., J. Phys. B 33, 473 (2000)
- [3] T. Odagiri et al., submitted to J. Phys. B

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Figure 1: The singly differential cross sections for the emission of the atomic fluorescences, $d\sigma_i/d\Omega_i$, in (a) and the doubly differential cross sections for the emission of the atomic fluorescences, $d^2\sigma_2/d\Omega_i d\Omega_j$, in (b) as a function of incident photon energy in the photoexcitation of N₂.