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Photon pair generation from multiply excited states of N_2, NO and O_2 excited by single-photon absorption

Takeshi ODAGIRI[*1], Keishou FUNATSU[1], Takehiko TANABE[1],
Isao H. SUZUKI[2,3], Masashi KITAJIMA[1] and Noriyuki KOUCHI[1]
[1]Dept. of Chem., Tokyo Institute of Technology, Meguro-ku, Tokyo 152-8551, Japan
[2]KEK-PF, Tsukuba, Ibaraki 305-0801, Japan
[3]AIST, Tsukuba, Ibaraki 305-8568, Japan

Introduction

Molecular multiply excited states 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 dynamics is hence one of the important subjects in atomic and molecular science. We have recently developeded *(γ, 2γ) method* to observe the multiply excited states of molecules: cross sections for the generation of a pair of photons emitted by neutral fragments in photoexcitation of a molecule,

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

are measured as a function of incident photon energy [1, 2]. Such a cross section curve is free from ionization and thus the features originating from the multiply excited states are highlighted. In the present study, the (γ, 2γ) method has been applied to N_2, NO and O_2 and multiply excited states of these molecules have been investigated.

Experiment

The experiments were carried out by using a 3m-normal incidence monochromator at BL-20A of KEK-PF. The two photons in process (1) emitted parallel to the electric vector of the incident light were detected in coincidence by two photon detectors, each of which is composed of an optical window (MgF_2 or LiF) and a microchannel plate.

Results and discussion

Figure 1 shows the doubly differential cross sections for the generation of a pair of photons (process (1)) in the photoexcitation of N_2(a) [2], NO(b) and O_2(c) as a function of the incident photon energy. The cross section curves reveals, for the first time, multiply excited states of these molecules in the energy range where the ionization from innermost valence electrons takes place. It is remarkable that some of the multiply excited states lie above the double ionization thresholds [3] indicated by hatched vertical bars. Interestingly such multiply excited states dissociate into neutral fragments escaping from the electronic autoionization and make a contribution to photon pair generation comparable to those below the double ionization thresholds.

For NO and O_2 sharp dips are observed just above the double ionization thresholds. They would be attributed to increase of branching ratio for autoionization of the multiply excited states observed which competes with process (1). The mechanism of the autoionization seems to be the spectator Auger decay of the multiply excited states.

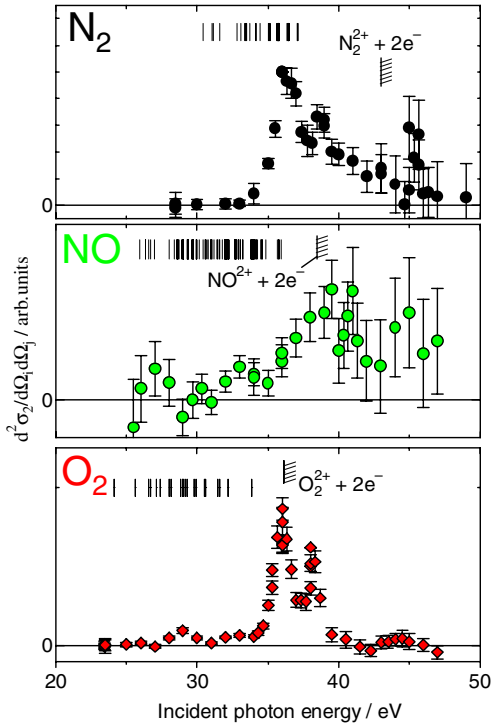

Figure 1 The cross sections for the generation of a pair of photons in the photoexcitation of N_2, NO and O_2 differential with respect to the solid angles for the photons. The hatched and short vertical bars indicate the double ionization thresholds [3] and dissociation limits of process (1), respectively.

References

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* joe@chem.titech.ac.jp