

Probing dissociative superexcited states of molecules by a detection of metastable atomic hydrogen in the 2s state

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1 Introduction

Doubly excited states of molecules are short-lived resonance states and often play an important role as intermediates in a wide range of reactions such as dissociative recombination, associative ionization, Penning ionization, and so forth. The formation and decay of them provide a unified view for understanding the various dynamic processes [1].

Such doubly excited states embedded in the electronic continua are well observed in cross sections free from ionization such as dissociation cross sections. Recently, we have developed a new method for measuring the cross sections for production of metastable atomic hydrogen in the 2s state, the H(2s) atom, formed in photoexcitation of a hydrogen containing molecule [2, 3]. In the present study, the method was applied to CH₄ and NH₃ for identifying the dissociative doubly excited states leading to the formation of the metastable atomic hydrogen.

2 Experiment

The measurements were carried out at BL-20A. The metastable hydrogen atom in the 2s state was detected by the combination of a localized electric field created by a stack of parallel plate electrodes and a detector of photons that is composed of a microchannel plate (MCP) coated with CsI and an MgF₂ window in front of it. The 2s state of atomic hydrogen mixes with the 2p state by the Stark effect due to the electric field and the atom decays to the ground state by emitting a Lyman- α photon, which was detected by the photon detector. Other details of the experiment were described elsewhere [2, 3].

3 Results and Discussion

Figure 1 shows the cross sections for the production of the H(2s) atom from photoexcited CH₄ (a) and NH₃ (b) as a function of the incident photon energy. By a fitting based on a semi-classical analysis, superexcited states of CH₄ and NH₃ were found in the cross sections as shown by the dotted curves in the figure. Among them, the three peaks at 29.7, 35.0 and 39.3eV in figure 1 (a) and the peak at 31.6eV in figure 1 (b) were attributed to the doubly excited states. The doubly excited states of CH₄ seem to be the same that were observed in the cross sections to form H(2p) atoms in the photoexcitation of CH₄ [4].

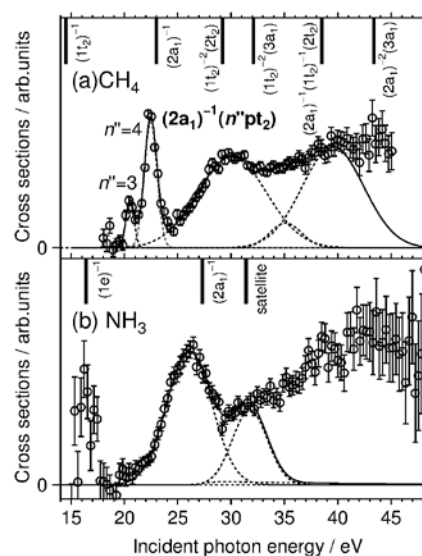


Figure 1. Cross sections for the production of the metastable hydrogen atom in the 2s state for (a) CH₄ and (b) NH₃ as a function of the incident photon energy. The vertical ionization potentials of CH₄ [5] and NH₃ [6] are displayed by thick vertical bars. The curves show the results of the fits by the semiclassical analysis.

References

- [1] Nakamura H, *Int. Rev. Phys. Chem.* **10** 123–88 (1991)
- [2] Odagiri T *et al.*, *Rev. Sci. Instrum.* **81** 603108 (2010)
- [3] Odagiri T *et al.*, *Phys. Rev. A* **84** 053401 (2011)
- [4] Fukuzawa H *et al.*, *J. Phys. B* **38** 565 (2005)
- [5] Potts A W *et al.*, *Proc. R. Soc. Lond. A* **326** 165 (1972)
- [6] Piancastelli M N *et al.*, *J. Chem. Phys.* **87** 1982 (1987); Banna M S *et al.*, *J. Chem. Phys.* **63** 4759 (1975)

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