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Threshold electron-electron coincidence study on dissociative photoionization of O_2 followed by fragment autoionization

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Introduction

There has been a growing interest in the mechanism of single photon double ionization of molecules. Direct double photoionization, where two electrons are simultaneously ejected into the double ionization continuum, is the basic process above adiabatic double ionization threshold. In addition, indirect double photoionization via intermediate singly-charged ion state occurs when the ion state subsequently autoionizes or produces an autoionizing fragment:

$AB + h\nu \rightarrow AB^+ + e^- \rightarrow AB^{2+} + 2e^-$ or

$AB + h\nu \rightarrow AB^+ + e^- \rightarrow A^* + B^+ + e^- \rightarrow A^+ + B^+ + 2e^-$.

By using photoelectron-photoelectron coincidence technique, Price and Eland found the latter process on 40.8 eV double ionization of the oxygen molecule [1]. Later, Bolognesi et al. investigated this indirect double ionization process in the photon energy range 32-42 eV using threshold photoelectron-photoelectron coincidence technique, and discussed on the indirect double ionization mechanism via intermediate formation of O_2^+ lying around 33 eV [2]. Despite these efforts, the full understanding of the double ionization mechanism of oxygen is still not obtained. In order to gain the entire picture of the double photoionization process of oxygen, we have measured hundreds of threshold photoelectron-photoelectron coincidence spectra in the photon energy range 46-50 eV and assembled the spectra in such a manner as that coincidence yields are plotted as functions of both photon energy and electron kinetic energy [3]. We have suggested the prominent indirect double ionization process with the resultant two-dimensional spectrum. in this report, we present a detailed interpretation of the two-dimensional spectrum to display the O_2 double photoionization process.

Experimental results

Figure 1 shows a two-dimensional plot of threshold electron-electron coincidence yields, which is newly obtained for a wider photon energy region. The two electrons ejected for each double photoionization event share the energy difference between incident photon energy and ionization energy for O_2^{2+}, and the kinetic energy of one of the electrons observed is restricted to essentially zero. The available energy increases linearly with the incident photon energy, and therefore formation of O_2^{2+} states results in diagonal lines on the two-dimensional plot. Besides the diagonal lines due to formation of O_2^{2+} states, we can see in the photon energy region 38-55 eV three or more vertical stripes running parallel to the photon energy axis. These are assignable to autoionization from O*[converging to $O^+(^2D)$] to $O^+(^4S)$ [4]. This shows that O_2^+ state(s) lying in the corresponding ionization energy region dissociates into fragment pairs, one of which is O*[converging to $O^+(^2D)$], in preference to molecular autoionization to O_2^{2+}. Here, counterpart ion fragments $O^+(^4S)$, $O^+(^2D)$ and $O^+(^2P)$ are energetically possible. In order to determine the counterpart ion fragments, we have performed threshold photoelectron-photoion coincidence spectroscopy at 42.4 eV and 46.4 eV. The photoion kinetic energy spectra measured in coincidence with threshold electrons (not shown) present the main contributor at 42.4 eV is O^+ fragments associated with dissociation to $O^* + O^+(^4S)$, while dissociation to $O^* + O^+(^2D)$ is more preferred at 46.4 eV to $O^* + O^+(^4S)$.

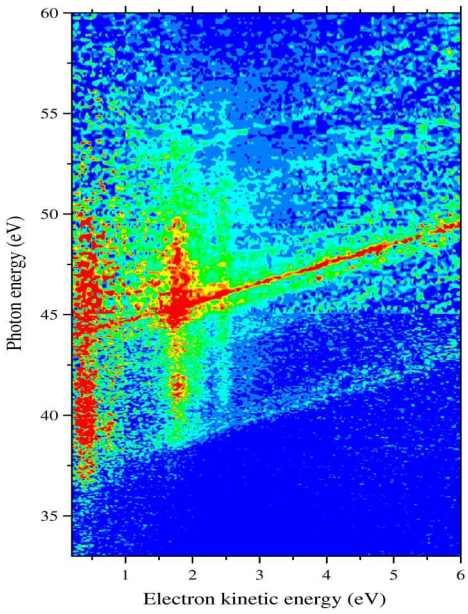

Figure 1. Two-dimensional plot of threshold electron-electron coincidence yields. Intensities are presented by the plots from blue to red on a linear scale.

References

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