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Absolute photoabsorption cross section of C_{60} in the extreme ultraviolet

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Abstract

The absolute photoabsorption cross section curve of C_{60} has been determined by means of mass spectrometry with the photon source of monochromatized synchrotron radiation of $h\nu = 24.5$–150 eV. Description has been made on a high-temperature source of gaseous fullerenes and an efficient time-of-flight mass spectrometer. The cross section was estimated by assuming an approximate expression of the number density of C_{60} in the ionization region. The resultant values were 762, 241, and 195 Mb at $h\nu = 24.5$, 90, and 110 eV, respectively, with about 10% errors. The cross section curve was then normalized at $h\nu = 25$ eV to the absolute photoabsorption cross section reported by Jaensch and Kamke [R. Jaensch, W. Kamke, Mol. Mater. 13 (2000) 143], the most reliable data so far available in the valence excitation region of C_{60}. Accordingly, the present cross section data were altered to 407, 144, and 114 Mb at $h\nu = 25$, 90, and 110 eV, respectively.

Keywords: Photoabsorption cross sections; Fullerenes; C_{60}; Extreme ultraviolet

1. Introduction

Relative photoabsorption cross sections of C_{60} have been reported by several groups. Most of the authors focused their attention to photon energies below 25 eV or those around the carbon 1s edge at ∼280 eV [1–3]. Furthermore, only a few experiments have been made on the absolute photoabsorption cross section, σ_{abs}, of C_{60}. Jaensch and Kamke have measured σ_{abs} at the photon energy below 25 eV from direct absorption of vacuum UV synchrotron radiation passing through a cell filled with the C_{60} vapor [4]. They pointed out that several especial cares should be taken in such measurements. First, the accessible photon energy regions are crucially restricted by the material of the windows of the cell. Good transmission of the windows should be secured up to the highest photon energy of interest, even at high ambient temperatures. Second, knowing the sample number density is prerequisite for calculating σ_{abs}, but the density is markedly unstable when C_{60} molecules are sublimated under conditions of high temperature. Owing to these difficulties, there has been no report dealing with σ_{abs} at photon energies above 25 eV, where only relative absorption cross sections were determined from the yields of positive ions produced from C_{60} [1,2,5].

In this paper, we attempt to obtain the σ_{abs} curve of C_{60} from 24.5 to 150 eV by means of photoionization mass spectrometry. For this purpose, several essential devices have been exploited in combination with a high-temperature source of gaseous fullerenes: a grazing-incidence monochromator, a conical nozzle, a quartz-oscillator thickness monitor and an efficient time-of-flight (TOF) mass spectrometer. The thickness monitor, the crux of our apparatus, was used for evaluating the C_{60} density in the ionization region.

2. Experiments

The detailed experimental setup for the measurement was described elsewhere [6]. Fig. 1 shows the side view of the apparatus. The powder of C_{60} was loaded in a copper sample holder and a molecular beam was discharged from a conical nozzle with a throat diameter of 0.5 mm and a diverging angle of 7.2°. The sample holder was heated up to approximately 743 K. Monochromatized synchrotron

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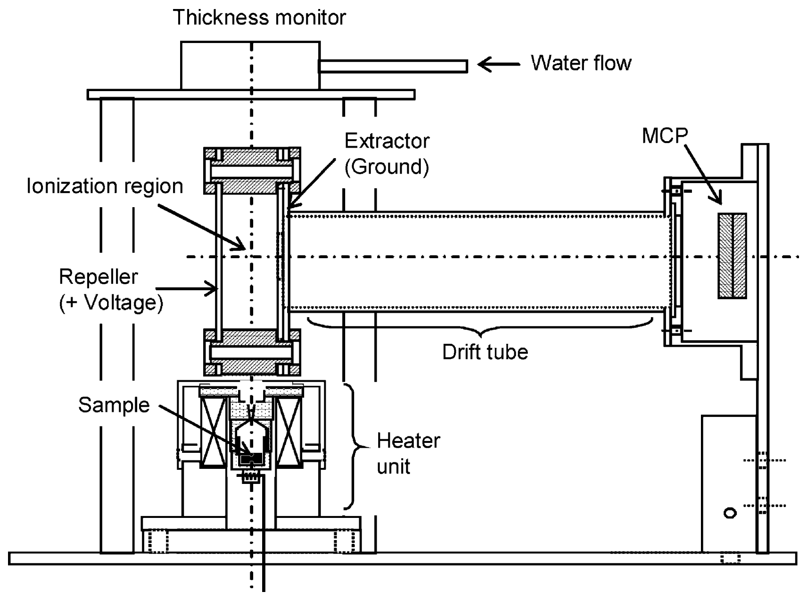

Fig. 1. Schematic diagram of the photoionization mass spectrometer for fullerenes [6].

radiation from a Dragon-type monochromator at the beamline 2B in UVSOR was focused onto the molecular beam and photoions of fullerenes were produced. The fullerene cations produced in the ionization region passed through a central hole (Ø 10 mm) of the grounded extractor electrode, as shown in Fig. 2, and mass-separated by a time-of-flight mass spectrometer. A pulsed voltage rising from the ground level to +100 V was applied to the ion repeller electrode as a start trigger for the TOF measurement. The duration and frequency of this pulse voltage were 5 μs and 5 kHz, respectively. Finally, the ions were detected with a microchannel plate (MCP) electron multiplier (Ø 14.5 mm; Hamamatsu, F4655). Either a time-to-amplitude converter (ORTEC, 567) or multichannel scaler (Laboratory Equipment, LN6500) was used to acquire mass spectra. The photon flux of the synchrotron radiation was estimated by measuring the photocurrent from a silicon photodiode (IRD, AXUV-100). A crystal-oscillator surface thickness monitor (Inficon, XTM/2) was employed to measure the flux of the molecular beam and evaluate fullerene density in the ionization region.

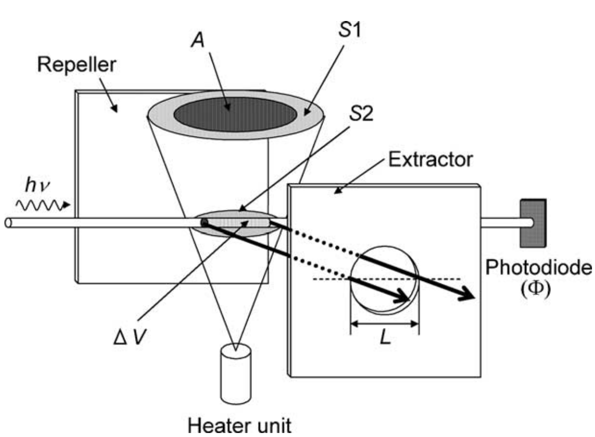

Fig. 2. Illustration indicating the quantities that are required for calculation of the absolute absorption cross section of C_{60}. L, reaction length; ΔV, ionization volume; Φ, photon flux; A, effective area of the thickness monitor; S_1, cross section of the molecular beam on the plane including the thickness monitor; S_2, cross section of the molecular beam on the plane including the ionization region.

It is possible that the detection efficiency of the MCP multiplier would depend on the charge of the impinging fullerene ions. We measured the ratio between the yield of Ar^+ and that of Ar^{2+} at $h\nu = 70$, 80, and 90 eV. Since the intensity ratio at every $h\nu$ was in good agreement with the reported value within experimental uncertainties [5], no correction has been made for the detection efficiency of the MCP. The ion detection efficiency of the mass spectrometer needs to be determined before the estimate of the total photoabsorption cross section of C_{60}. For this purpose, we measured the signal intensity of the ions produced by photoionization of Kr because its photoionization cross section has been well investigated. The Kr vapor was admitted to the vacuum chamber through a needle valve. The gas pressure of Kr was monitored with an ionization gauge, which had been calibrated beforehand by a capacitance manometer. The gas pressure was postulated to be almost uniform inside the vacuum chamber.

3. Results and discussion

Since the quantum yield for photoionization is nearly equal to unity in the present $h\nu$ range [7], the sum of the partial photofragmentation cross sections are considered to represent the total photoabsorption cross section. Singly and doubly charged ions of C_{60} have been observed in the TOF