Hydrogen ion desorption from amorphous carbon films induced by resonant core electron excitations

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Introduction

Carbon is a peculiar element which can form various structures in different dimensions and, moreover, with a wide variety of physical properties. Carbon materials are also known to transform with an unexpected ease to different structures by electronic excitations. Recently, some of the authors found that considerable structural changes are induced in tetrahedral amorphous carbon (ta-C) films by electron irradiation [1] and soft X-ray illumination exciting 1s core electrons in carbon atoms [2]. However, few studies have been done on the effects of soft X-ray illumination on carbon materials for technical reason of generally low light intensity in the C1s core-edge region due to carbon contamination in ordinary SOR beam lines. The present experiment aimed to get insight to the atomistic mechanism in the coreexcitation induced structural change in ta-C by studying the ionic desorption from the films under intense soft Xray illumination in the C1s core-excitation energy range.

Experimental

The ta-C film was deposited on a Si substrate by a filtered cathodic vacuum method. The soft X-ray experiments were conducted at BL-13C operated in the single-bunch mode using monochromatic soft X-rays of 10^{10} photons/s·cm² in intensity. The mass of desorbed ions was analyzed by a TOF spectrometer designed for coincidence spectroscopy [3]. Ion desorption spectra were measured as a function of the photon energy used for soft X-ray illumination, in one set of measurements for individual ions detected by TOF and in another for total ions collected by the TOF tube. All the experiments were conducted at room temperature under a ultra high vacuum (UHV) condition (< 3×10^7 Pa).

Results and Discussions

TOF spectra obtained when the sample was illuminated with photons in energies ranging near the C1s edge show the absence of C^+ (*m*=12) and C_2^+ (*m*=24) ions, which means that carbon atoms are not desorbed from the surface although atomic displacements leading to structural changes must be induced inside the film by the soft X-ray illumination.

The dominant ions in photo-stimulated desorption from the ta-C film were H^+ ions. In the core-excitation-induced

restructuring in ta-C films [2], the excitation spectrum exhibits a characteristic peak at 289 eV which might be attributed to a resonant excitation of C1s electrons to antibonding C-H σ^* state. A working hypothesis, therefore, was that the restructuring is triggered by rupture of C-H bonds. The blue curve in Fig. 1 shows the efficiency spectrum for desorption of ions dominated by H⁺. The circular marks indicate the desorption efficiency spectrum of H⁺ ions and the broken curve the X-ray absorption spectrum (XAS) for comparison. The desorption efficiency spectrum exhibited a sharp resonant structure near the C1s core absorption edge, but the peak position is lower by 3eV than the characteristic peak in the excitation spectrum for the photo-induced restructuring.

Thus, the absence of carbon ions in the desorbing species and the spectral discrepancy in the excitation efficiency disfavour the C-H bond rupture hypothesis.



Fig. 1 H⁺ ion desorption efficiency spectra measured by TOF experiments (marks), total ion counting (blue) and XAS (dotted black) for comparison.

References

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