Characterization of sp^3 content in amorphous carbon films

Yuji Muraoka[a], Takanori Wakita[a] and Takayoshi Yokoya[a]

[a]*Research Institute for Interdisciplinary Science, Okayama University, 3-1-1 Tsushima-naka, Tsushima, Kita-ku, Okayama 700-8530, Japan*

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Q-carbon (*quenched* carbon) is a newly discovered amorphous phase of carbon [1], and shows extraordinary physical properties such as room-temperature ferromagnetism and high-temperature superconductivity by boron doping [2]. Despite much interest, the research of Q-carbon is limited because of the difficulty in preparation of Q-carbon using rapid nonequilibrium laser processing. In this study, we prepared Q-carbon by adjusting the sp^3 content in diamond-like carbon (DLC) films and the laser energy density of pulsed laser annealing (PLA).

The amorphous DLC films are fabricated on sapphire Al_2O_3(0001) substrates using a pulsed laser deposition technique with a KrF excimer laser (λ = 248 nm). The sp^3 content in the films varied between 20% and 42% by changing the laser energy density. To determine the sp^3 content in the films, X-ray photoemission spectroscopy (XPS) measurements were carried out on the beamline BL-5, at the Hiroshima Synchrotron Radiation Center in Hiroshima University with non-monochromatic Mg Kα X-ray source ($h\nu$ = 1253.6 eV). C 1*s* core-level spectra were measured under an ultrahigh vacuum of ~10^{-7} Pa using a VSW hemispherical analyzer. The total energy resolution was about 1.8 eV. The binding energy of the films was corrected using the peak position of molybdenum spectra. Before measurements, the films were annealed at 100 °C under ultrahigh vacuum (~10^{-6} Pa) for 1 h to clean the film surface. PLA was performed on the DLC films using the KrF excimer laser with energy densities between 0.5 and 1.2 J/cm^2 at ambient atmosphere and temperature.

Figure 1 shows the XPS spectra of the C 1*s* core-level for the as-grown DLC films. A peak at the binding energy of ~284 eV was observed in the spectra for all the films. The peak position of the films moved towards higher binding energy, with slight broadening, as the laser energy density increased. Results of the spectral fitting are also shown in Fig. 1. After subtraction of a Shirley background, the C 1*s* spectra of the films were fitted using a Gaussian-Lorentzian mixed function. Lorentzian linewidth was set to 0.095 eV [3]. In this fitting process, for simplicity, symmetric line shapes were assumed, and one component for each sp^3 and sp^2 bond was considered. Full-width-half-maximum (FWHM) of peaks was set to the same value in each film. The experimental results were well reproduced by the three components with peak positions of 283.9, 284.9, and 286.5 eV. The peaks of ~284 and ~285 eV corresponded to sp^2 carbon atoms and sp^3 C-C bond, respectively [4]. The feature at 286.5 eV was assigned to C-O contamination formed on the film surface due to air exposure [5]. Gaussian linewidth was estimated from FWHM of peaks to be 1.7, 1.5, 1.8 eV for the films with laser energy densities of 0.4, 0.8, and 1.3 J/cm^2, respectively.

The sp^3 content in the films was determined from the ratio of the corresponding sp^3 peak area over the sum of sp^3 and sp^2 peak areas to be 20%, 35% and 42% for the laser energy density of 0.4, 0.8, and 1.3 J/cm^2, respectively. The sp^3 content in the films increased with increasing the laser energy density. This indicated that the sp^3 content in the DLC films was successfully varied by changing the laser energy density during deposition. These results are qualitatively in good agreement with the result recently reported by Joshi [6].

Since DLC films with different sp^3 content were obtained, PLA was performed on the DLC films. Consequently, for the combination of 20% sp^3 content and laser density of 1.0 J/cm^2, as well as 42% sp^3 and 0.5 J/cm^2, the films showed characteristic features of Q-carbon: filamentary nanostructures, the presence of a T peak in the Raman spectrum, room-temperature ferromagnetic behavior (Fig. 2), and ~80% sp^3 content. The results indicate that Q-carbon can be obtained by using a proper combination of sp^3 content in DLC films and an appropriate PLA energy density [7].

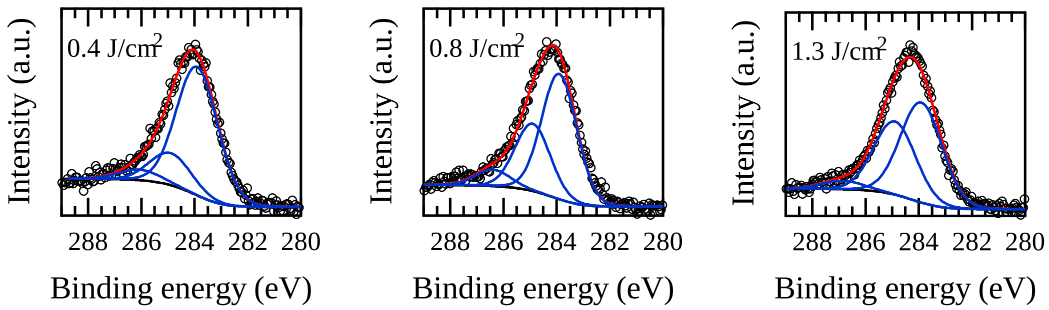

FIGURE 1. XPS spectra of C 1*s* core-level for the as-grown DLC films prepared with the laser energy density of (left) 0.4, (middle) 0.8, and (right) 1.3 J/cm^2. The results of spectral fitting are also shown. Open circles are experimental data, red lines are the fitting result, black lines are the Shirly background, blue lines are the component used for the fittings.

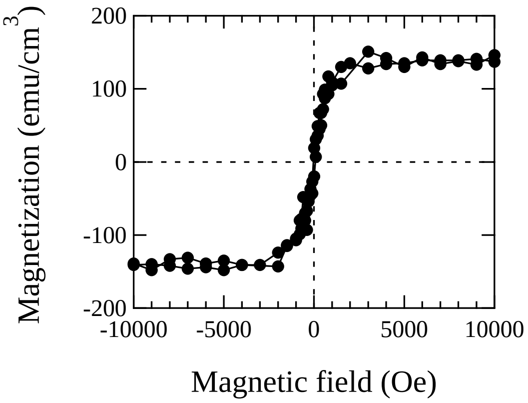

FIGURE 2. Field dependence of magnetization (*M-H* curve) for the 20-*sp*^{3} and 1.0-PLA film, measured at 300 K.

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