The electronic structure of K_6C_{60} studied by soft x-ray emission spectroscopy

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INTRODUCTION

The observation of superconductivity in alkali-metal doped fullerides (A_3C_{60}, A is alkali metal or mixture) has attracted much research attention [1,2]. The resulting electronic structure of doped C_{60} depends strongly on the character of the doping atom. There are strong chemical interactions and charge transfer processes, as typified by the alkali metal fullerides. Several phases of potassium doped C_{60} (K_xC_{60}) have been identified previously by x-ray measurements. For K_3C_{60} the structure is still fcc, like C_{60}, but with a slightly modified lattice parameter, K_4C_{60} has a bct structure and for K_6C_{60} the structure is bcc. Increasing the potassium concentration from zero the conductivity increases and reaches a maximum at x=3, where the conduction band is half filled. Further doping decreases the conductivity and K_xC_{60} becomes again semiconducting at x=6.

Soft x-ray emission spectra originate in transitions between valence states and core states, and thus information on the occupied valence states can be gained. In this paper, we report results from soft x-ray absorption and emission studies of the electronic structure of C_{60} and potassium doped C_{60}.

EXPERIMENT

The experiment was performed at beamline 7.0 of the Advanced Light Source (ALS) at Lawrence Berkeley Laboratory. The beamlines both use undulators and spherical-grating monochromators to provide intense, high-resolution soft x-ray radiation. The pure C_{60} film was made in situ by evaporation of C_{60} on clean Mo and Fe surfaces in the experiment chamber under vacuum [3]. Subsequent doping with potassium was done using a degassed getter source (SAES Getters, Italy). The approximate concentration of potassium in the C_{60} films was controlled by monitoring the exposure time of the C_{60} film to the potassium flux produced at a given getter current (5A). In one case, potassium was deposited onto a room temperature C_{60} film, while in another case, potassium was deposited onto a hot C_{60} film (at about 200•C). The diffusion and equivabra of potassium into the C_{60} film were completed by heating up the film to 200•C under 10^{-8} mbar of potassium vapour pressure.

The x-ray absorption spectra (XAS) were obtained using the total electron-yield-detection (TEY) method, done by measuring the sample current against photon energy of incoming monochromatized synchrotron radiation. The spectra were normalized to the photo-current from a clean gold mesh introduced in the synchrotron radiation beam in order to correct for intensity variations in the excitation beam.

The soft x-ray fluorescence was recorded in the polarization plane of and normal to the incident photon beam using a high-resolution grazing-incidence grating spectrometer with a two-dimensional detector [4]. The bandpass of the exciting photon beam was set to ~0.3 eV, both for emission and absorption measurements, and the spectrometer resolution was ~0.5 eV.

RESULTS AND DISCUSSION

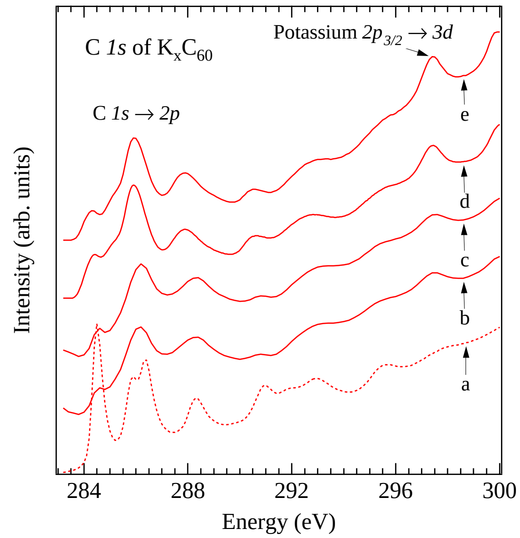

Figure 1

Figure 1 shows the soft x-ray absorption spectra for pure C_{60} and potassium doped C_{60} films obtained using total electron-yield detection. The lowest-energy peak in the C_{60} spectrum *a* is corresponds to a transition from the C *1s* to the LUMO. There are significant changes in the soft x-ray absorption spectrum upon deposition of potassium. The trend is in agreement with other XAS measurements, even though the saturation of K deposition was not reached in this case. The intensity contributed from the LUMO is reduced (see spectra *b*, *c*, *d* and *e* in figure 1) since the LUMO becomes occupied upon potassium doping, while the intensity of the K *2p* $\rightarrow$*3d* absorption (at about 297.4 eV) increases.

Figure 2 shows the C *K* emission of K_xC_{60} obtained at various excitation energies. The insert shows the x-ray absorption spectrum recorded for the same sample. In our previous investigations on pure C_{60} films, much larger shape variations were observed in C *K* emission of C_{60} with photon-excitation energy, and it was interpreted in terms of resonant inelastic x-ray scattering processes. Strong symmetry correlation including parity selection was verified. In contrast, here, the shape of the x-ray fluorescence spectra of K_xC_{60} film exhibits much less variations with excitation energy. This suggests that resonant inelastic x-ray scattering becomes less prominent due the formation of metallic system upon potassium doping. Nevertheless, the survival of some shape variations in the photon-excited emission of K_xC_{60} suggests a certain degree of molecular character remaining.

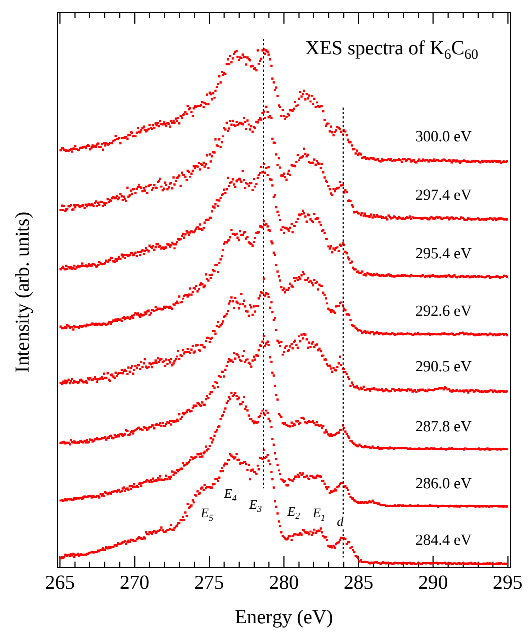

Figure 2

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