Surface phase transitions in fullerene-based systems

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In this talk we give some examples of the capabilities of high-resolution photoemission, in the study of surface specific thermal induced phenomena and phase transitions in fullerene-based systems.

First, we investigated the normal state temperature dependence of the core level and valence band photoemission spectra of the superconductor K_3C_{60}(111), revealing the existence of a phase transition at 200 K and the presence of several chemically shifted components in the C 1s core level. The merohedral disorder and the inhomogeneity of the conduction electron distribution at the molecular level were considered as possible explanations [2, 3]. Moreover, by exploiting the surface sensitivity of reflection Electron Energy Loss measurements, the temperature dependence of the surface resistivity from 120 K up to 600 K was obtained and compared with bulk sensitive investigations. We demonstrated that the normal state electronic and transport properties of the top molecular layer of K_3C_{60} are similar to the corresponding properties measured with bulk sensitive techniques [4].

We also measured the temperature evolution of the full width at half maximum of the C 1s core level photoemission spectra from 30 K up to 480 K in undoped C_{60}(111) films, with high overall energy resolution (~60 meV) and high temperature accuracy (±0.1 K). We obtained details on the disordering transition of the surface molecules, reporting the experimental evidence of a two-steps mechanism for the rotational disordering of fullerene molecules at the (111) surface of the solid. The rotational disordering of bulk C_{60} crystals is well known to take place at 260 K [5], while the surface counterpart has been observed at ~ 230 K [6]. Here we show that the rotational degrees of freedom of one molecule, out of the four inequivalent C_{60} molecules of the (2x2) surface unit cell in the low temperature ordered phase, melt about 110 K before the surface phase transition, in agreement with recent theoretical predictions [7].

Finally we investigated the interaction of a single C_{60} layer chemisorbed on Ag(100) and its temperature and alkali metals doping dependence. Surface metallic phases are

formed at any Na doping level, following a rigid band filling behaviour, until the conduction band is completely filled with 6 electrons/molecule. These observations, that contrast with the bulk phases behaviour, can be explained with an efficient screening of the Hubbard energy U in the C_{60} bands due to the close proximity to a metallic surface and highlight the important role of electron correlation in the electronic properties of these systems. Valence band angle-integrated photoemission shows the reversible opening of a gap at the Fermi level as a function of temperature. In the undoped system the gap reaches a maximum value of 10 meV at T ~ 70 K [8]. The gap opens up to a doping level of 4 electrons/molecule [9]. This is the first evidence of an electronic phase transition in C_{60} monolayers. The above observations can have important implications on the ongoing debate about surface superconductivity in C_{60}-based bulk materials [10-12].

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