Surface Electronic Structure of K_3C_60

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C60 ions have an important role in the understanding of high-temperature superconductivity. C60-based salts with alkali metals (A) with stoichiometry A3C60 are well known to superconduct, whereas the stoichiometrically-neighboring compounds are usually insulating. K3C60 serves as a testing ground for investigations of their electronic structure, presumably because of the ease with which high-quality films can reproducibly be prepared [1]. Thus, while the experimental photoemission (PES) results have converged recently in terms of the spectral shapes, fundamental controversies remain to be elucidated. The disagreement between the theoretical electronic density-of-states (DOS) and the PES data is stark: Theory indicates narrow molecular-like bands [2], whereas experiment shows much broader bands with subtle structures. Recent photoemission spectroscopy results led to speculation that the empirical rule of three electrons per fulleride [3] might be relaxed in fewer dimensions [4],such as surfaces and grain boundaries [5]. EELS combined with PES results have been used to claim that the electronic structure of the surface layer is, for the most stable surface, exactly the same as in the bulk [6,7]. We present photoelectron spectra of K3C60, and detect a strong angle-dependence. This allows the unambiguous identification of bulk and surface components in the data, and explains the anomalous lineshapes universally observed for this system. We show that rigid band effects, strong electron-electron correlations, and a K-deficient surface layer explain the observations in PES of this system. We find that the states near the Fermi level are located predominantly in the bulk of the sample. There is also strong evidence of an insulating surface layer, which we ascribe to intermolecular electronic correlations. Thus the parameters derived from many bulk studies of the electronic structure of the alkali fullerides (see, e.g., [8,9]) are found to provide a good description of our results, which greatly simplify the picture of the surfaces of these superconductors.

[1] D.M.Poirier,Appl.Phys.Lett. 64 ,1356 (1994).

[2] S.Satpathy et al.,Phys.Rev.B 46 ,1773 (1992).

[3] T.Yildirim et al.,Phys.Rev.B 54 ,11981 (1996).

[4] R.Hesper et al.,Phys.Rev.Lett. 85 ,1970 (2000).

[5] R.Hesper et al.,Phys.Rev.B 62 ,16046 (2000).

[6] A.Goldoni et al.,Phys.Rev.B 58 ,11023 (1998).

[7] A.Goldoni et al.,J.Chem.Phys. 113 ,8266 (2000).

[8] J. E. Han, et al., Phys. Rev. Lett. 85, 1276 (1999)

[9] V. Brouet et al.,Phys. Rev. Lett. 86, 4680 (2000)