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The electronic structure of the conduction band of K_3C_{60} studied by photoemission and electron energy-loss spectroscopy

T. R. Cummins[a], J. F. Armbruster[a], M. S. Golden[b], M. Knupfer[b], H. A. Romberg[a], M. Sing[a] and J. Fink[b]
[a]*Kernforschungszentrum Karlsruhe, Institut für Nukleare Festkörperphysik, Postfach 3640, D-76021 Karlsruhe, Germany*
[b]*Institut für Festkörperforschung, IFW Dresden eV, Postfach 16, D-01171 Dresden, Germany*

Temperature-dependent photoemission spectra of the conduction band of superconducting K_3C_{60} confirm an earlier observation of a continuous transfer of spectral weight from the Fermi level (E_f) to higher binding energies with increasing temperature. This suggests the occurrence of a metal to non-metal transition at elevated temperatures. Electron energy-loss spectroscopy (EELS) in transmission measurements of the conduction band plasmon of K_3C_{60} show negligible dispersion as a function of the momentum transfer ($\boldsymbol{q}$), thus deviating from the behaviour expected for a simple metal and from some recent theoretical predictions.

1. Introduction

Previous photoemission spectroscopy (PES) and EELS studies of the A_3C_{60} fullerides (A=K, Rb) have provided valuable information on the normal state electronic structure of these new high temperature superconductors.[1] Here we further examine two points: the temperature dependence of the t_{1u}-derived conduction band (CB) spectral weight observed in PES and the $\boldsymbol{q}$-dependence of the charge carrier excitation as seen by EELS in transmission.

2. Experimental

For PES, films of global stoichiometry $K_{2.2}C_{60}$ (~150Å thickness) were prepared *in-situ* on freshly evaporated gold substrates. The film composition was chosen to avoid possible premature formation of K_4C_{60}.[2] Photoemission experiments were conducted using He I radiation (21.22eV) with a total energy resolution of 60meV. The spectrometer, and the preparation of the free-standing films (thickness ~1000Å) for EELS in transmission experiments are described elsewhere.[3] The energy resolution was set to 90meV and the momentum resolution to 0.04Å^{-1}.

3. Results and Discussion

The photoemission profiles of $K_{2.2}C_{60}$, within ~2eV of E_F, across the temperature range 15-425K are displayed in Fig. 1 . In this energy region the spectral weight is due exclusively to K_3C_{60} as a consequence of phase separation in the K_xC_{60} system, and the negligible spectral contribution of α-C_{60} in this energy window.[1,4] As has been observed in previous studies,[5] the width of the CB-derived spectral weight is ~1.3eV which is significantly greater than predicted from LDA band structure calculations.[6] The anomalous width and corresponding low density of states (DOS) at E_F arises because the ejected photoelectron couples to molecular phonon modes of the C_{60} balls and the collective excitation of the charge carriers, thus giving rise to satellites at ~0.25 and ~0.6eV binding energies (BE), which are seen in the low temperature spectra of Fig. 1. On increasing the temperature there occurs a spectral weight transfer from states at E_F to higher BE. Such a transfer of spectral weight has been previously observed in PES recorded at 425K of both Rb_3C_{60} and K_3C_{60}. The new results for K_3C_{60} in Fig. 1 demonstrate that the spectral weight transfer occurs continuously as a function of temperature. The observed shift of spectral weight cannot be due solely to a changing Fermi-Dirac distribution as a function of temperature and, since the changes are completely recyclable, they are not a result of temperature-dependent phase transitions. An alternative suggestion for the spectral weight transfer is of a temperature-dependent metal to non-metal (MN) transition. A MN transition could be driven by Anderson localisation induced by increased disorder at higher temperatures. No direct evidence for the non-metallicity of the A_3C_{60} phases at high temperatures exist, although in recent studies by Stepniak *al.*[7] an increase was observed in the resistance of a $Rb_{2.8}C_{60}$ thin film for temperatures

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between ~330 and 350K. More measurements of physical properties as a function of temperature are greatly desired to resolve this question.

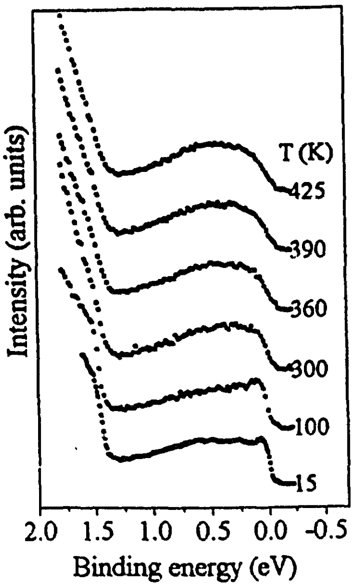

Figure 1. PES spectra of the conduction band of K_3C_{60} in the temperature range 15-425K.

Additional insight on the electronic structure of the conduction band of K_3C_{60} can be gained by EELS measurements of the valence band excitations. Here we report the loss function between 0 and 2eV as a function of q which is shown in Fig. 2. At low q, the features observed at ~0.6eV and ~1.2eV correspond to the charge carrier plasmon and interband transitions between the partially filled t_{1u}-derived band and the t_{1g}-derived bands, respectively. In keeping with earlier data on C_{60}/C_{70} mixtures,[3] for increasing q up to 0.5Å^{-1}, there is negligible dispersion of the plasmon energy, although the intensity of both features decreases. For a simple free electron gas the conduction band plasmon energy increases quadratically with q, resulting in a shift of ~340meV for changes of q to 0.4Å^{-1}. This behaviour is not seen in the q dependent data of K_3C_{60} in Fig. 2. Recently some theoretical work[8] have predicted an unusual dependence of the plasmon energy as a function of q. In calculations based on the random-phase-approximation (RPA) a prediction is made for a negative plasmon dispersion law in K_3C_{60}. For q changes up to 0.5Å^{-1} shown in Fig. 2, the magnitude of the predicted negative dispersion is ~150meV. The data presented in Fig. 2 demonstrate that this negative dispersion law appears not to hold.

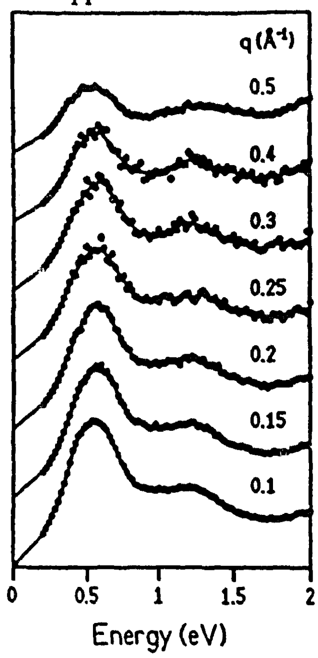

Figure 2. q-dependent EELS measurements of the loss function of K_3C_{60}.

4. Conclusions

The t_{1u}-derived spectral weight in photoemission of A_3C_{60} displays an anomalous temperature dependence with a spectral weight transfer to higher BE at high temperatures which suggests a temperature-dependent metal to non-metal transition, perhaps induced by disorder. Charge carrier plasmon dispersion in the q-dependent loss function of K_3C_{60} is less than 20meV for q values up to 0.5Å^{-1}, in contrast with that expected for a simple metal. This behaviour does not support a recent theoretical prediction of a negative plasmon dispersion law.

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