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LUMO photoemission lineshape in quasi-one-dimensional C_{60} chains

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The low-energy single-particle excitations of highly ordered C_{60} chains adsorbed on a vicinal copper substrate are investigated by angle-resolved photoemission spectroscopy. The interface state previously identified on $C_{60}/Cu(111)$ shows a one-dimensional dispersion on Cu(553). In contrast, no significant momentum dependence is detected for emission from the lowest unoccupied molecular orbital (LUMO). The LUMO displays similar phonon features as in $C_{60}/Cu(111)$ but it does not peak toward the Fermi level for all considered potassium dopings and its photoemission lineshape is broader than in any other monolayer system investigated so far. This behavior is not easily reconciled with existing theory and indicates that the one-dimensional character of the chains affects the electronic structure of the monolayer in an intricate way.

I. INTRODUCTION

Molecular nanostructures at surfaces have attracted attention in recent years due to their potential for applications in microelectronics. Long-range ordered molecular arrangements, from regular two dimensional (2D) to highly one dimensional (1D), can be prepared through self-assembly on templates with appropriate symmetry, surface patterns or chemical functions.[1,2]

In the prospective of possible applications it is fundamental to investigate the electronic structure of ordered molecular films, in particular the electronic states at the Fermi level which determine their transport properties. However, little experimental information is available so far. Band formation due to the overlap of molecular orbitals has been observed in a limited number of cases due to the difficulties to prepare single domain long-range ordered molecular arrangements and detect electronic dispersion in the presence of strong correlations, electron-lattice interactions and small Brillouin zones.

Dispersion of molecular valence bands, and in particular of the highest occupied molecular orbital (HOMO) typically lying at least 1 eV below the Fermi level has been observed in several systems.[3–7] The HOMO-derived peaks are usually rather broad and the peak center of gravity is often found to track the one-electron dispersion calculated within density-functional theory. However, recent work on C_{60} films indicates that an interpretation of dispersing HOMO peaks as coherent Bloch states is oversimplified. The large and temperature-dependent broadening of the C_{60} HOMO peak is more readily explained by multiphonon excitations whose maxima follow the one-electron dispersion in the frozen lattice.[5,6] At the Fermi level, electronic dispersion has been detected only for the LUMO-derived state in one monolayer (ML) of C_{60}/Ag doped with K to reach about three electrons/molecule.[8,9] In these 2D C_{60} arrangements, the bandwidth of the LUMO is renormalized compared to single-particle calculations and its spectral lineshape shows the characteristic features of coupling to intramolecular phonons.

C_{60} chains grown on stepped surfaces offer the opportunity to investigate the behavior of electrons confined in a 1D molecular structure.[10] We have shown previously[5] that the electronic coupling in the HOMO occurs predominantly along the chains with a hopping anisotropy $t_\parallel/t_\perp$ on the order of 10. In such quasi-one-dimensional (quasi-1D) molecular systems one might expect complex electronic phases arising from a competition of Luttinger physics with instabilities against spin-/charge-density wave formation and a tendency toward Mott-Hubbard insulating states.[11,12]

In this paper we present an angle-resolved photoemission study of the electronic states close to the Fermi level in C_{60} chains on Cu(553). Because of charge transfer from the metal substrate, the LUMO is partially occupied and contributes significantly to the density of states near the Fermi level. We find a peculiar lineshape which differs significantly from the typical LUMO signature in two-dimensional systems and discuss its origin in terms of many-body effects in C_{60} and one-dimensional character of the electronic structure.

II. EXPERIMENT

Photoemission experiments at low-temperature were performed with a Scienta SES 2002 spectrometer at Stanford University, using He $I\alpha$ radiation (21.22 eV) from a microwave-driven monochromatized discharge lamp (Gammadata VUV5000). Three differential pumping stages were used in order to reduce the He partial pressure in the measurement chamber to a level near the sensitivity limit of the ultrahigh vacuum ion gauges, sufficiently low to perform experiments with minimal surface degradation over several
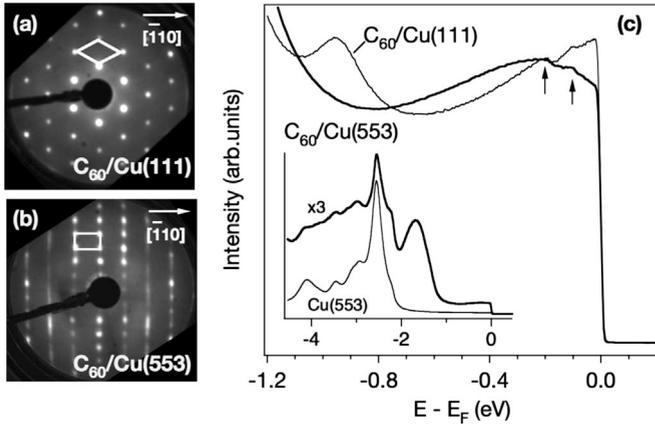
FIG. 1. (a) and (b) Low-energy electron diffraction (LEED) patterns of 1 ML $C_{60}/Cu(111)$ and $C_{60}/Cu(553)$ measured with 40 and 37 eV electron energy, respectively. The C_{60} chains on Cu(553) run along the $[\bar{1}10]$ direction of the copper substrate. (c) Low-temperature ($T = 10$ K) He $I\alpha$ angle-integrated photoemission spectra of one monolayer of C_{60} in a 1D and in a 2D molecular arrangement on Cu(553) and Cu(111), respectively. The arrows mark the positions of the satellites due to electron-phonon coupling. The inset shows angle-integrated spectra of 1 ML $C_{60}/Cu(553)$ and clean Cu(553) over a wider energy range.

hours, even on reactive samples such as doped C_{60} films. Energy and angular resolutions were set to 7.5 meV and $\pm 0.15°$ for all measurements. Additional data at various excitations energies and polarizations were taken at the Surface and Interface Spectroscopy (SIS) beamline at the Swiss Light Source using a Scienta SES 2002 analyzer.

Cu(553), the stepped surface used in this work, is a *B*-type vicinal of Cu(111) with $(11\bar{1})$ step facets. The nominal terrace width is 9.8 Å, which is close to the van der Waals diameter of the C_{60} molecules. The single crystal has been prepared by standard sputtering and annealing cycles. C_{60} powder (99.9%) was sublimated from a titanium crucible onto the surface held at 600 K. Potassium was evaporated from a well out-gassed SAES getter source onto the C_{60} monolayer held at room temperature.

III. RESULTS AND DISCUSSION

Before presenting the low-temperature photoemission data we briefly summarize the structure of one monolayer $C_{60}/Cu(553)$ and of the parent 2D system $C_{60}/Cu(111)$. In the latter the molecules form a regular hexagonal pattern and are adsorbed with the hexagon ring facing the substrate surface [Fig. 1(a)], an arrangement that is commonly observed upon adsorption on (111) metal surfaces.[13–15] On Cu(553), C_{60} molecules form regular chains aligned along the step direction of the substrate.[5,10] Compared to $C_{60}/Cu(111)$, the unit cell is larger, containing two C_{60} with different orientations. A different bond geometry to the copper atoms of the ascending step produces alternating chains of molecules adsorbed on the hexagon and the pentagon ring, which appear with different contrast in scanning tunneling microscopy (STM) images. The intermolecular distance along the chains

(10.2 Å) is the same as the nearest-neighbor distance in $C_{60}/Cu(111)$, while in the perpendicular direction it is expanded by 0.8 Å [Fig. 1(b)], giving rise to a pronounced 1D HOMO dispersion.[5]

In Fig. 1(c) we compare the low-temperature ($T = 10$ K) angle-integrated photoemission spectra of 1 ML C_{60} on Cu(553) and Cu(111). The photoemission spectrum of $C_{60}/Cu(111)$ shows the typical features of the partially occupied LUMO in a 2D arrangement. Upon adsorption on metal surfaces, C_{60} becomes negatively charged because of electron transfer from the metal to the molecule. Values between -0.8 and $-2.9 e/C_{60}$ have been reported for a complete C_{60} ML on Cu(111).[16–18] The high Fermi edge is indicative of the metallic character of the monolayer. At higher binding energies there are additional features, a peak at 0.1 eV and a peak-dip structure around 0.2 eV, which resemble those observed in gas phase C_{60}^-, where the shoulders were interpreted in terms of energy-loss processes due to electron-phonon coupling.[19] Similar spectra have been described for $C_{60}/Ag(100)$ (Ref. 20) and $C_{60}/Ag(111)$.[8] The peak at 1 eV binding energy is characteristic of C_{60} adsorption on copper and arises from hybridization of C_{60} with Cu sp orbitals resulting in a well-defined interface state.[18]

The LUMO spectral weight in the C_{60} chains on Cu(553) is remarkably different. It consists of a very broad hump, which extends from the Fermi level down to about 0.8 eV below E_F, with a width that is much higher than the noninteracting total bandwidth (0.3 eV) calculated within LDA for monolayer systems.[8] Maximum intensity occurs around 0.25 eV below E_F. Toward the Fermi level, the spectral weight decays gradually and only the satellites due to intramolecular phonon excitations appear at the same energies as in $C_{60}/Cu(111)$. Although a clear loss of intensity occurs toward E_F, the spectrum maintains a well-defined Fermi edge with major contribution from the C_{60} monolayer. Comparing spectra of $C_{60}/Cu(553)$ with those of the bare substrate [Fig. 1(c) inset], we estimate that less than 20% of the total intensity at E_F is due to emission from the substrate. Similar results have been obtained at different photon energies and polarizations of the incident light which exclude that this unusual C_{60} spectrum is determined by spurious matrix element effects.

The large width of the LUMO in the C_{60} chains resembles the LUMO peak width measured in bulk K_3C_{60} (Ref. 21) and is in contrast with the general trend in C_{60} monolayers on metals, where the photoemission spectra are narrower than in the bulk because of a combined reduction in the electron-plasmon coupling and of the Coulomb energy U in the proximity of the metal surface.[22,23] The line width in $C_{60}/Cu(553)$ seems consistent with the fact that electron correlations and many-body interactions are expected to be stronger in a one-dimensional system. Also the broad LUMO line width might be explained by the peculiar molecular arrangement of $C_{60}/Cu(553)$. Molecules along the two alternating chains have a different orientation relative to the substrate, a situation for which different charge transfer for the two chains can be expected.[24] This may lead to a shift in the LUMO energies between the two inequivalent chains, which contributes to the line width. Such an interpretation is supported by measurements with varying polarization of the in-
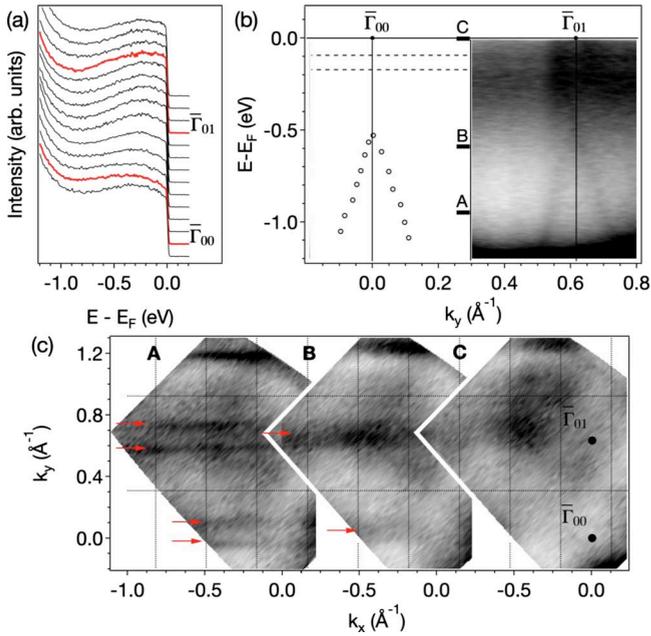
FIG. 2. (Color online) Angle-resolved photoemission data of C_{60} chains on Cu(553) measured at low temperature ($T=10$ K). (a) Set of high-resolution EDCs measured along the chains. (b) Dispersion plot illustrating the electronic structure of C_{60}/Cu(553) in the direction along the chains. The dashed lines highlight the satellites from the nondispersing intramolecular phonons; the black circles follow the dispersion of the *interface state* in the HOMO-LUMO gap. (c) Constant energy maps at (a) −0.95 eV, (b) −0.6 eV, and (c) E_F. The C_{60} Brillouin zones are indicated by a dashed grid. Maximum intensity corresponds to black. k_y is oriented along [$\bar{1}$10], the direction parallel to the chains. k_x is oriented along the perpendicular direction [$\bar{3}\bar{3}$10].

coming light and different photon energies which give changes in the photoemission intensity close to the Fermi level that are consistent with the presence of two LUMO components with different occupancy.

Angle-resolved photoemission data of C_{60} chains on Cu(553) are presented in Fig. 2. Systematic measurements at low temperature show no evidence of sharp quasiparticle peaks crossing the Fermi level at any point of the C_{60} Brillouin zone. Typical energy distribution curves (EDCs) are shown in Fig. 2(a). They were measured along the chains direction and show a broad hump and a loss of spectral weight toward E_F at all k points. For different emission angles there is a modulation of the photoemission intensity which does not follow the periodicity of the C_{60} reciprocal lattice. It is due to final-state scattering of the emitted photoelectrons (UV photoelectron diffraction)[25] which reflects the angular distribution of the molecular orbitals rather than the electronic band structure. The satellites due to phonon excitations show little dispersion as expected for C_{60} intramolecular vibrational modes.

The constant energy maps of Fig. 2(c) indicate that the momentum distribution of the lowest lying excitations is very different from the clear 1D dispersion of the HOMO and the interface state. We have shown in previous publications that the HOMO-derived peak has a very anisotropic
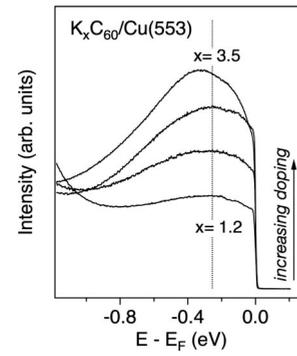
FIG. 3. K_xC_{60}/Cu(553) angle-integrated photoemission spectra measured for different doping levels of the LUMO ($T=10$ K). The LUMO occupancy x was calibrated with the procedure described in Ref. 30. We found that in the undoped monolayer $1.2e$/C_{60} are transferred from the substrate to the molecule.

dispersion (at least a factor of 10 difference between parallel and perpendicular directions), which is caused by the larger C_{60}-C_{60} distance perpendicular to the chains.[5] Moreover an interface state with one-dimensional dispersion is visible within the HOMO-LUMO gap. Figure 2(b) shows a dispersing state which has a minimum in binding energy around 0.5 eV at all $\bar{\Gamma}$ points and moves very rapidly toward the HOMO in two symmetric branches. It resembles the interface state found in C_{60}/Cu(111) (Ref. 18) although its intensity is much weaker and its energy position is shifted by about 0.5 eV toward the Fermi level. The constant energy maps of panel (c) illustrate the 1D character of the interface state in C_{60}/Cu(553). In map A there are pairs of straight lines running perpendicular to the steps. Moving toward the top of the interface band (map B) they merge into a single stripe crossing the center of the Brillouin zones. This defines an electronic state that disperses along the chains and is completely localized in the perpendicular direction. In contrast, the Fermi surface map (C) shows an aperiodic modulation of intensity and no clear contour lines.

The photoemission signature of the C_{60} LUMO is expected to change significantly upon intercalation with alkali atoms, which is the most effective way to dope electrons into the LUMO and modify the electronic properties of C_{60} compounds. At half-filling (K_3/C_{60}) 2D and three-dimensional (3D) systems are metallic and bulk materials show superconductivity; For an even number of electrons (K_4/C_{60}) bulk C_{60} behaves like a Mott-Jahn-Teller insulator.[26] STM studies of potassium-doped 2D monolayers have shown that the local density of states at the Fermi level is consistent with this picture.[27] In photoemission the two phases should show either a metallic spectrum with distinct quasiparticle peaks dispersing across E_F or a broad featureless Gaussian peak with negligible intensity at the Fermi level.[8,28,29] Figure 3 displays angle-integrated spectra of the C_{60} LUMO measured at different doping levels. As for the bare C_{60} chains, it reveals a behavior that is difficult to reconcile within the existing models. The LUMO peaks are very broad, dominated by a hump and a decrease in spectral weight toward the Fermi level. Upon increasing the doping, the LUMO peak integral increases linearly with the charge state of the molecule, while

the position of the maximum moves to higher binding energy only when the doping exceeds three electrons in the LUMO. Angle-resolved data for the K-doped LUMO are similar to those of the undoped chains and are not shown here.

The picture emerging from the above photoemission data indicates a very different behavior of the C_{60} chains compared to all monolayer systems investigated so far. This becomes particularly clear when comparing to the K_3C_{60} ML on Ag(111) which displays a dispersing quasiparticle peak and a nondispersing incoherent hump.[8] The unusual LUMO lineshape of the C_{60} chains is likely related to the quasi-one-dimensional character of the electronic band structure. In particular, it is tempting to interpret the loss of spectral weight toward E_F, still in the presence of a defined Fermi edge, as reminiscent of a Luttinger liquid, for which the weight of the quasiparticle and the interacting density of states are expected to vanish at the Fermi level. Similar photoemission lineshapes have been observed in bundles of single-wall carbon nanotubes[31] and in peapods,[32] where upon increasing the potassium doping a transition from a 1D Luttinger liquid to a 3D Fermi liquid has been postulated. For the C_{60} chains it is possible that a metallic Fermi edge remains because the 1D character of the LUMO is reduced due to hybridization with the substrate which may assist interchain hopping. Indeed, recent experiments aiming to investigate the charge-transfer dynamics along and perpendicular to the chains,[33] indicated an important role of the metal substrate in delocalizing the LUMO electrons across the steps.

An alternative way to describe the C_{60} spectral function starts from a Fermi-liquid picture and includes the phonon contribution to the spectral function. To this end we carried out model spectral function calculations for a coupled electron-phonon system. The idea was to test whether a highly anisotropic single-particle band coupled to high-energy phonons can produce a spectral function comparable to what is observed experimentally. Following Liechtenstein *et al.*,[34] we used a single-band model with coupling to local Einstein phonons on each site. Figure 4 displays the results of calculations for a half-filled band in a square lattice with increasing hopping anisotropy $t_\parallel / t_\perp$. We considered the coupling to two H_g phonons of the C_{60} molecules ($\omega_{ph}=0.195$ and 0.096 eV, respectively) and assumed the same electron-phonon coupling strength ($g=0.023$). For an isotropic dispersion ($t_\parallel / t_\perp = 1$), this simple model reproduces very well the prominent features of the angle-integrated LUMO spectrum in C_{60}/Cu(111), i.e., the peak-dip structures at about 0.2 and 0.1 eV binding energy and the maximum intensity just below E_F [Fig. 4(d)]. The calculated dispersion plot [inset of Fig. 4(d)] shows a renormalized sharp quasiparticle peak crossing the Fermi level and at higher binding energies an incoherent hump, which is consistent with the experimental results for the K_3C_{60} ML on Ag. Upon increasing the anisotropy between parallel and perpendicular hoppings, there is a transfer of spectral weight from the Fermi level toward higher binding energies. In a quasi-1D system [$t_\parallel / t_\perp = 10$, Fig. 4(f)], the maximum intensity in the angle-integrated spectral function is shifted to the bottom of the renormalized quasiparticle band. This resembles the angle-integrated spectrum of the chains and seemingly indicates that the different LUMO lineshape in C_{60}/Cu(111) and C_{60}/Cu(553) is due to the differ-
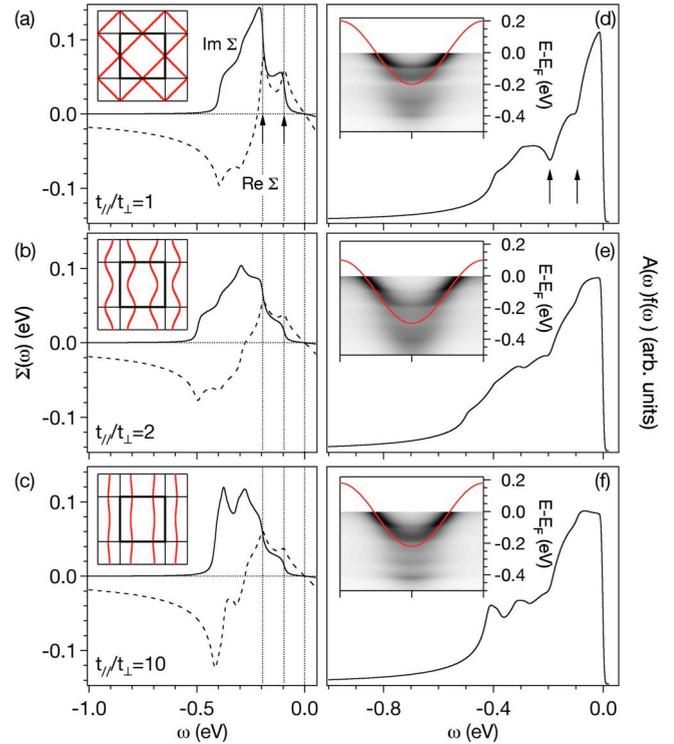
FIG. 4. (Color online) Spectral function calculations for a coupled electron-phonon system in a square lattice with different hopping anisotropies. (a)–(c) Electron self-energy calculated for a half-filled band coupled to two H_g phonons ($\omega_{ph}=0.195$ and 0.096 eV, respectively). The hopping anisotropy are as follows: (a) $t_\parallel / t_\perp = 1$, isotropic dispersion, (b) $t_\parallel / t_\perp = 2$, and (c) $t_\parallel / t_\perp = 10$, highly anisotropic dispersion similar to the C_{60} chains. The insets show the corresponding Fermi surfaces [gray (red) line]. In the calculations we included the fivefold degeneracy of these C_{60} vibrational modes. (d)–(f) Occupied part of the interacting DOS convoluted with a Gaussian ($\sigma=10$ meV) for the three different hopping anisotropies. The insets show calculated dispersion plots. A constant impurity scattering of 100 meV has been added for the plots of $A(k, \omega)$. The solid lines correspond to the noninteracting bands.

ent shape of the noninteracting density of states (DOS) in 2D and 1D systems. However, very poor agreement is found between the calculated dispersion plot and the k-resolved data for the C_{60} chains. The inset of Fig. 4(f) shows that also in the case of a highly anisotropic system we should expect dispersing quasiparticle peaks, which are not discernible in the experimental angle-resolved data (Fig. 2).

The lack of LUMO dispersion in the quasi-1D C_{60} chains may be due to a combination of effects that were not included in the model calculations. The presence of two different chains in the C_{60}/Cu(553) ML structure may complicate the electronic band structure and make it hard to discern dispersive peaks. However, this effect alone cannot explain the reduction in spectral weight toward the Fermi level and the complete lack of dispersion in the LUMO. We speculate that the properties of C_{60}/Cu(553) result from a combination of the structural complications, the effects of hybridization between the LUMO and Cu d and sp states and many-body interactions. In particular we cannot exclude that in this quasi-1D molecular system electron-phonon interaction

causes multiphonon excitations that remove weight from the dispersive quasiparticles and lead to a strong renormalization of the band dispersion.

IV. CONCLUSIONS

We presented an angle-resolved photoemission study of the low-energy electronic states of C_{60} chains on Cu(553). Both, the well-defined HOMO-derived peak and the interface state in the HOMO-LUMO gap show a strongly anisotropic dispersion indicating a quasi-1D character of the electronic system. Intriguingly, photoemission from the partially occupied LUMO almost completely lacks dispersion. Moreover, the LUMO has an unusual lineshape with a broad hump at high binding energy and a gradual loss of spectral weight toward the Fermi level. This behavior cannot be reconciled with common models of many-body interactions in bulk fullerides or in 1D electronic systems. Instead, the findings suggest that a good understanding of electron transport in molecular nanostructures on surfaces requires an integral approach including the combined effects of reduced dimensionality, many-body interactions and hybridization at the molecule-substrate interface.

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