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Comment on "Electronic structure of superconducting KC8 and nonsuperconducting LiC6 graphite intercalation compounds: evidence for a graphene-sheet-driven superconducting state". — Source link

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Comment on "Electronic Structure of Superconducting KC_8 and Nonsuperconducting LiC_6 Graphite Intercalation Compounds: Evidence for a Graphene-Sheet-Driven Superconducting State from electron-phonon interaction."

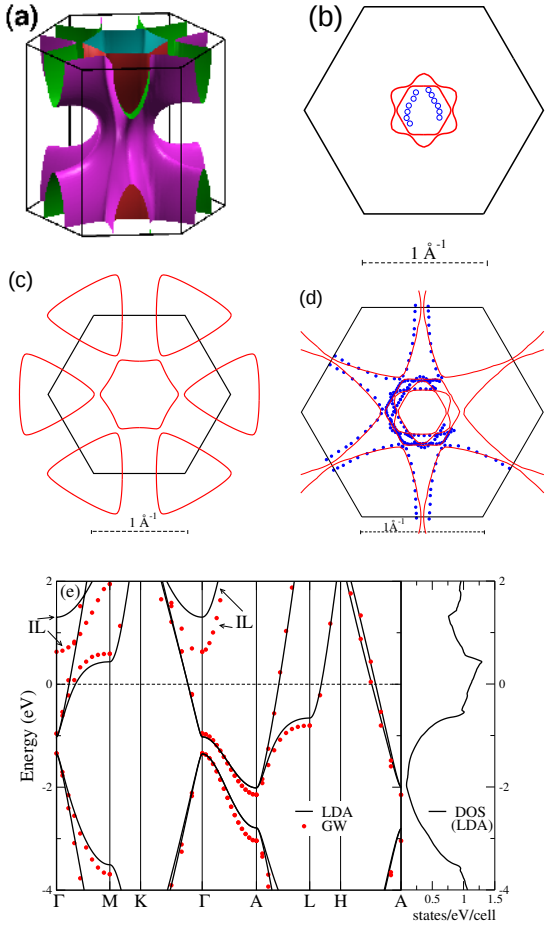

FIG. 1. (color online):(a) DFT Fermi surface of Bulk LiC_6 (Li stacking is AA) [2] and cuts over the $k_z = 0$ (b) and $k_z = \pi/c$ (c) planes within the local density approximation (LDA). In (b) empty blue dots are ARPES data from [1]. (d) DFT Fermi surface (red) of a Li-intercalated graphene bilayer (C_6LiC_6) compared with ARPES data (blue dots) from ref. [3]. (e) DFT and GW bulk LiC_6 electronic-structure. The Fermi level is at zero. IL labels the metal-interlayer state. In all plots we use the LiC_6 Brillouin zone where the graphene Dirac point is folded at Γ.

In [1] it is claimed that the charge transfer from the metal to the graphene layers is larger in KC_8 than in LiC_6. This relies on a two-dimensional analysis of photoemission (ARPES) data that, *a priori* and without experimental verification, totally neglects the electronic band dispersion perpendicular to the graphene planes. Pan *et al.* could have verified this assumption by performing ARPES measurements as a function of the photon energy, as it was done for the out-of-plane dispersion in graphite [4]. Here we demonstrate that, in LiC_6, the small interlayer distance results in a very high band-dispersion along the k_z direction, invalidating the analysis and the conclusions of [1].

The density-functional-theory (DFT) band-structure in Fig. 1 (e) is highly dispersive along k_z (ΓA direction) by more than 1.2 eV. This results in the three-dimensional Fermi surface shown in Fig. 1 (a),(b) and (c). Note, in particular, that the Fermi surface cuts in the $k_z = 0$ and $k_z = \pi/c$ planes are completely different, the $k_z = 0$ being closer to the experimental data in [1].

Correlation effects slightly increase the k_z dispersion in bulk LiC_6 as shown by our GW [5] calculation. Both GW and DFT find an empty interlayer metal state in LiC_6[6, 7] (complete charge-transfer from Li to graphene) and a partially occupied one in KC_8 (incomplete charge-transfer from K) [7, 8]. Thus the calculated charge-transfer is larger in LiC_6 than in KC_8, in disagreement with Ref. [1].

To validate the accuracy of DFT on Li intercalated graphite systems, we calculate[9] the Fermi surface of a Li-intercalated graphene bilayer, obtaining results in excellent agreement with ARPES data [3] (Fig. 1 (d)). The experimental Fermi surface splitting of the bilayer measures the hopping between graphene planes separated by Li atoms and, indirectly, the k_z dispersion in the bulk. Indeed, the hopping between graphene planes is a properties of the local geometry which is identical in bulk LiC_6 and in the C_6LiC_6 bilayer.

In conclusion, the three dimensional character of the Fermi surface of bulk LiC_6, neglected in the analysis of [1], invalidates the charge-transfer estimate of [1] and the subsequent claim of a graphene-sheet-driven superconducting state.

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