

MOLECULAR CHARGE TRANSFER COMPLEXES ON GRAPHENE

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Organic charge-transfer complexes (CTCs), especially those formed by the strong electron acceptor tetracyanoquinodimethane (TCNQ) and the strong electron donor tetrathiafulvalene (TTF), have been the topic of intense research for the last few years, both for their application in molecular electronics as well as for exotic effects such as 1D conductivity, charge density waves etc [1]. Recently, two-dimensional CTC of TTF-TCNQ self-assembled on Au(111) was seen to exhibit signatures of Kondo resonance [2]; however the molecular states were found to hybridise with the surface states of Au(111) [3] compromising the intrinsic properties of the CTC. Here, I will present low-temperature scanning tunneling microscopy (LT-STM) study of surface-confined CTC of TTF and fluorinated TCNQ (F_4 TCNQ), self-assembled on the surface of oxygen-intercalated epitaxial graphene on Ir(111) (G/Ir). G/Ir has emerged as a model surface to study the assembly and electronic property of molecules [4]; oxygen intercalation ensures further decoupling from the metallic bulk. Sequential deposition of the molecules on this surface lead to the formation of rotationally identical domains of CTC with alternating rows of TTF and F_4 TCNQ lying parallel to the surface. The adsorption geometry of the structure matches those predicted by calculations based on density functional theory (DFT). The frontier molecular orbitals of the molecular species in the CTC, as found from scanning tunneling spectroscopy (STS), are different from those in isolated islands of TTF and F_4 TCNQ on the same surface – this suggests charge transfer between them. Intriguingly, small-range tunneling spectra on the molecules show a large, asymmetric dip at zero bias. The shape and size of the dip remains unchanged in magnetic field up to 10 Tesla; it becomes weaker as temperature is increased from 2.2 K to 12 K – only the asymmetry remains for higher temperatures.

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