The high-magnetic-field properties of single-crystalline epitaxial graphite.

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Graphite is an allotrope of carbon, composed by a Bernal stacking of graphene layers. In this material, the layer-to-layer coupling is made by Van-der-Waals forces, which result in highly anisotropic electronic properties.

Unfortunately, graphite is not a single crystal. Instead, this material is conventionally found in its highly-oriented form, obtained either synthetically, as a byproduct of industrial processes, or found in nature. The absence of well-defined crystallographic in-plane directions in bulk crystals imposes challenges to the understanding of graphite's properties, which are otherwise absent in systems with similar properties - such as Bi and Sb.

Perhaps one of the most curious properties of graphite regards its resistance in the presence of strong magnetic fields. It is conventionally observed that, in low temperatures, the resistance of the material increases almost-linearly with magnetic fields, reaching a maximum for magnetic fields around 15 T. Above this field, a markedly reduction of the sample resistance is observed with the increase of B, before the emergence of a high resistance state (HRS) is triggered by magnetic fields around 35 T.

Although the origin of the region of negative magnetoresistance ($dR/dT<0$) remains to be clarified, the triggering of the high-resistance state is thought to be associated to the induction of an out-of-plane charge density wave in the material, associated with a 3D – to – 1D dimensional crossover when the degeneracy of the lowest Landau subbands is adequately lifted. In this context, no in-plane anisotropy should manifest as a consequence of the HRS, as this would be an exclusively out-of-plane phenomenon.

As graphite is not a single crystal, however, attempts to probe such an (an)isotropy invariably run into the issue of bulk graphite not possessing well-defined in-plane crystallographic axes. In principle, such an issue could be addressed by considering small graphite samples, to ensure that measurements addressed a single grain of the material. However, such an approach compounds to the problem at hand, as samples of reduced size pertain to the realm of "few-layered-graphene" - often treated as a different entity than the bulk compound.

In this work, we address this issue by probing the in-plane isotropy of bulk graphite crystals grown by diffusion through single-crystalline Ni foils. Graphite obtained with this technique has been shown to present long-range crystalline order, permitting the measurement of electrical transport properties along in-plane high symmetry directions. Our results show the presence of in-plane anisotropy for magnetic fields above 15 T, indicating a surprising emergence of planar ordering during the out-of-plane electronic transition in the material. We also report an anomalous non-saturating magnetoresistance in the compound, hinting that subtleties in the crystal structure leading to new properties in this monoatomic compound.