Mass Inversion at the Lifshitz Transition in Monolayer Graphene by High-Density, Diffusive, Flip-Chip Alkali Doping

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The physical properties of 2D materials can be tuned by doping to extreme charge carrier density, enabling the investigation of phenomena such as superconductivity, charge density waves, and Lifshitz transitions. For example, it is estimated that an electron density of $3.7 - 5.1 \times 10^{14}$ cm^{-2} is required to induce a Lifshitz transition in monolayer graphene [1,2]. Achieving such charge density has been limited to chemical doping in ultra-high vacuum (UHV) conditions and observation by angle resolved photoemission spectroscopy (ARPES).

We report here [3] an integrated flip-chip method to dope graphene by alkali vapour in the diffusive regime, suitable for charge transport measurements at ultra-high charge carrier density. We introduce a liquid cesium droplet source into a sealed cavity filled with inert gas (argon) to dope a monolayer graphene Hall bar on a quartz substrate by the process of cesium atom diffusion, adsorption and ionization (Fig. a). Doping is monitored by operando ac Hall measurement of longitudinal R_{xx} and transverse R_{xy} resistance (Fig. b), with an optical image of a representative graphene Hall bar shown in Fig. c (scale bar = 100 μm). The measured R_{xx} and R_{xy} during Cs exposure can be used to determine the time dependent electron density n and electron mobility μ.

Upon sealing, the flip-chip assembly containing doped graphene is stable in ambient conditions, enabling sample characterization by various means, including non-resonant Raman scattering measurement through the transparent quartz window. Charge transport versus temperature and magnetic field in was performed, including Hall measurements (Fig. d). The Lifshitz transition is observed via the inversion of cyclotron effective mass, corresponding to sign inversion of the Hall coefficient R_H (Fig. d). Employing a third-nearest-neighbour tight binding (3NNTB) calculation, we estimate the Fermi energies and Fermi surfaces corresponding to the charge densities observed by high field magnetotransport (Fig. e). At the Lifshitz transition, the Fermi surface transforms abruptly from that of electron pockets around the K and K' points, to a hole pocket around Γ. Further experimental observations will be discussed.

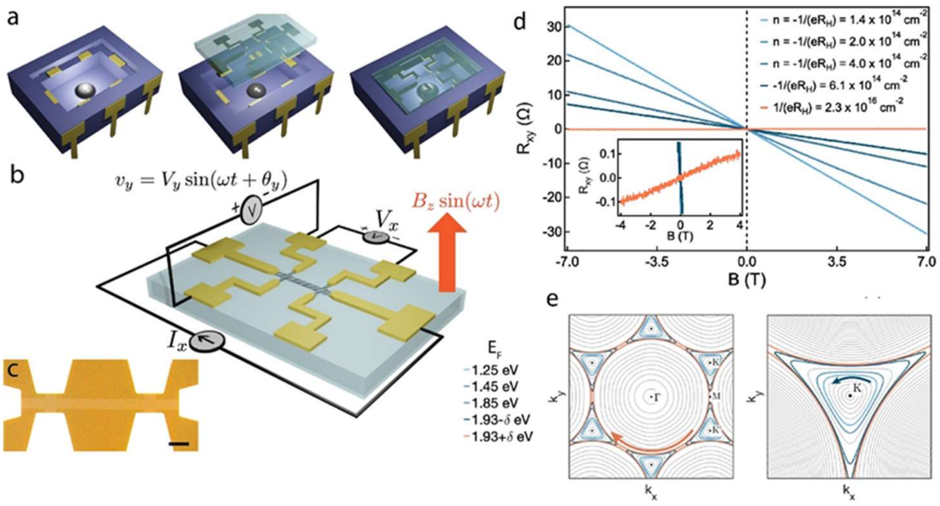

Fig.1. a) Schematic of flip-chip doping. b) Operando Hall setup. c) Optical micrograph of graphene Hall bar. d) High field Hall measurement. e) 3NNTB iso-energy surfaces.

In summary, our findings show that chemical doping, hitherto restricted to ultra-high vacuum conditions, can be applied in a diffusive regime at ambient pressure in an inert gas environment. We anticipate that flip-chip doping can be applied to a variety of 2D materials and dopant species.

References

[1] P. Rosenzweig et al., Physical Review Letters 125, 176403 (2020). [2] A. Zaarour et al., Physical Review Research 5, 013099 (2023). [3] A.M. Aygar et al., ACS Nano 18, 9092 (2024).