Observing Zero-Field Energy Gap in Graphene Grown on Sapphire Substrate

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Introducing an energy gap into graphene is a pivotal challenge for the developing graphene-based devices. An innovative approach involves breaking the sublattice symmetry, a technique successfully applied to graphene on a hexagonal boron nitride (hBN) substrate. In this study, we examine the energy gap in single-layer graphene, which was grown via chemical vapor deposition (CVD) on a sapphire substrate, using the resistively-detected electron spin resonance (RDESR) technique. We conducted RDESR measurements by recording the longitudinal resistance ($R_{xx,\nu}$) under microwave irradiation at a fixed frequency (ν) while sweeping the magnetic field (B) either perpendicular ($\perp B$) or parallel ($//B$) to the sample plane, within a 4 K cryostat.

Initially, the RDESR measurement was performed at $\nu = 27$ GHz. To extract the microwave-induced component, we subtracted the background resistance ($R_{xx,\mathrm{background}}$) from the $R_{xx,\nu}$, obtaining ΔR_{xx}. Figure 1 shows the ΔR_{xx} curves obtained with $\perp B$ (upper) and $//B$ (lower), both exhibiting distinct peaks near $B = \pm 1.0$ T, marked by blue arrowheads. Notably, the ΔR_{xx} curve for $//B$ displays shoulder features on both sides of the main peak, as indicated by red arrowheads.

To further investigate these peak and shoulder features, RDESR measurements under $//B$ were carried out at various frequencies. A gray-scale map of the derivative of ΔR_{xx} with respect to B ($d(\Delta R_{xx})/dB$) as functions of ν and B (Fig.2 (a)) reveals that the peak and shoulder features evolve along the three parallel lines; this is supported by Fig. 2(b), which plots the corresponding ν - B positions. A linear fit of the middle line (blue) yielded a slope of 27.9 ± 0.6 GHz/T and verifies that the extrapolated line crosses the origin. This indicates that the observed peak corresponds to the ESR signal, expected to show simple Zeeman gap in the B field, i.e., $h\nu = g\mu_B B$ (h: Planck's constant, μ_B: Bohr magneton), with a corresponding g-factor of 2.00 ± 0.05. Conversely, significant deviations from the origin at zero field by 4.6 ± 0.3 GHz (higher) and -5.0 ± 0.6 GHz (lower) were observed for higher and lower red lines, respectively. This deviation implies an energy gap $\Delta \sim 20$ μeV, indicating band splitting in graphene at zero field. Such splitting, observed in both CVD [1] and mechanically exfoliated [2] graphene on the hBN substrate, is attributed to sublattice splitting due to symmetry breaking. Our findings demonstrate the potential for the substrate-induced gaps due to symmetry breaking on sapphire substrates, suggesting the possibility of extending these observations beyond hBN substrates.

[1] U. R. Singh, et al, Phys. Rev. B. **102**, 245134 (2020), [2] C. Bray, et al, Phys. Rev. B. **106**, 245141 (2022).

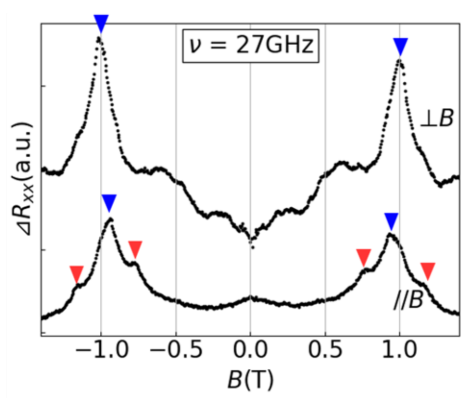

Fig.1. B-field dependence of ΔR_{xx} (= $R_{xx,\nu}$ - $R_{xx,\mathrm{background}}$) at $\nu = 27$ GHz. The B-field direction with respect to the sample plane is indicated in the figure. The blue (red) arrowheads indicate main peak (shoulder features).

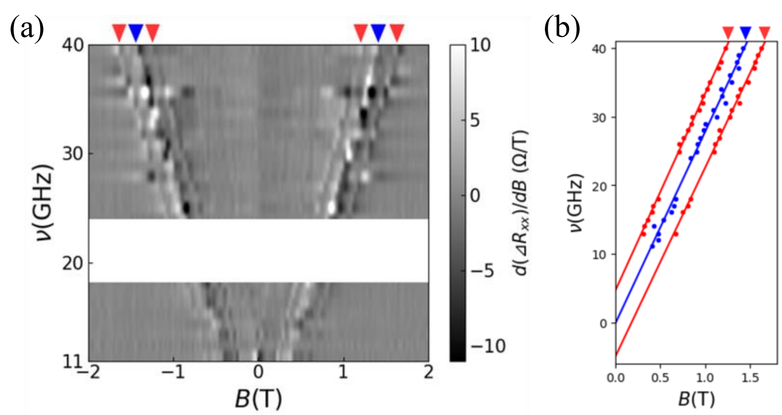

Fig.2. ν - B map of ΔR_{xx} signal under $//B$. (a) Gray-scale map of derivative amplitude: $d(\Delta R_{xx})/dB$. (b) Peak positions of ESR signal (blue dots) and satellite signal (red dots) along with liner fitting curves. The blue (red) arrowhead indicates main signal (shoulder features).