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Temperature dependence of Intrinsic Spin Orbit Coupling Gap in Graphene probed by Terahertz photoconductivity

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Abstract— Graphene is a quantum spin Hall insulator, with a nontrivial topological gap induced by the spin-orbit coupling. Such splitting is weak ($\sim 45\mu\text{eV}$) in the absence of external magnetic field. However, due to rather long spin-relaxation time, graphene is an attractive candidate for applications in quantum technologies. When it is encapsulated in hexagonal boron nitride, the coupling between graphene and the substrate compensates intrinsic spin-orbit coupling and decreases the nontrivial topological gap, which may lead to phase transition into a trivial band insulator state. In this work, we have measured experimentally the zero-field splittings in monolayer and bilayer graphene by the means of subterahertz photoconductivity-based electron spin resonance technique. The dependence in temperature of such splittings have been also studied in the 2-12K range. We observed a decrease of the spin splittings with increasing temperature. Such behavior might be understood from several physical mechanisms that could induce a temperature dependence of the spin-orbit coupling. These includes the difference in the expansion coefficients between the graphene and the boron nitride substrate or the metal contacts, the electron-phonon interactions, and the presence of a magnetic order at low temperature.

I. INTRODUCTION

THE intrinsic spin-orbit (ISO) coupling in graphene comes from carbon atom d orbitals which hybridize with the p_z ones. Indeed, the p_z orbitals themselves have no net orbital momentum along z , so the spin-orbit interaction is expected to be weak [1]. As an electron state at K valley and a hole state at K' valley are connected by sublattice symmetry, the ISO opens a gap of opposite sign at each Dirac points. The emergence of this gap therefore moves graphene from the family of Dirac semimetals to the one of quantum spin Hall insulators [2]. However, this topological gap is on the order of a few tens of μeV , and therefore difficult to observe. This sublattice symmetry in graphene can be broken, for instance, by the coupling of graphene to the boron nitride substrate (hBN), which in its turn induces staggered potentials and opens a bandgap. This staggered sub-lattice potential competes with the ISO potential, as the former leads to a trivial band insulator state, while the latter induces a topological insulator phase.

These phenomena of ISO coupling and spin splitting of sub-lattices have been addressed both experimentally and experimentally on graphene on hBN by the means of resistively detected electron spin resonance (ESR) [3-5]. Measuring the spin-flip transitions by ESR techniques allows one to determine accurately the different zero-field splittings (ZFS) in graphene. As consequence, it becomes possible to study this ISO coupling as a function of various physical parameters, such as temperature, hydrostatic pressure, or different orientations between graphene layers. In this work, we adapt a sub-THz magneto-photoconductivity technique to detect ESRs in monolayer and bilayer graphene embedded or not in hexagonal boron nitride (hBN). We measure several spin resonances, so that their energy evolution extrapolated to zero magnetic field provide estimation of ZFS, and the sub-lattice and ISO potentials, of similar values for monolayer and bilayer graphene, and there temperature dependence.

II. METHODS

Our experimental method is indeed similar to the “Electrically Detected ESR” (EDESER) technique. The particularity of our photoconductivity technique is based on the optimization of the electromagnetic coupling between the incident THz wave and a two-dimensional electron gas in a Ratchet device usually used as a THz sensor. This technique therefore represents an effective EDESER method capable of operating with a high sensitivity over a wide range of frequencies ranging from tens of GHz to THz. The experiments were carried using radiation frequencies between 45 GHz and 220 GHz (sweeping the magnetic field up to 9 T), and temperature range from 2.6 to 12 K. When ESR condition is satisfied, the signal measured at the THz Ratchet sensor, made from a graphene monolayer or bilayer, undergoes a variation that is extracted thanks to a lock-in detection scheme, resulting in a photoconductivity signal. An appropriate data analysis routine permits to increase the visibility of the ESR signal obtained, as show in Fig. 1.

III. RESULTS

We measure several spin resonances (Fig. 1) and, by extrapolating their energy evolution to zero magnetic field, we observe two zero-field splittings (ZFSs) attributed to the sub-lattice and ISO potentials γ' and β respectively, while α correspond to the ordinary Zeeman splitting. From measurements, we estimated the Landé-factor $g=2.02\pm 0.02$ for monolayer graphene sample, and $g=2.00\pm 0.02$. From β line slope, we estimated the ISO splitting value to be $\Delta_I=51.1\pm 6.5$ μeV for monolayer graphene, consistent with the value obtained in bilayer graphene of $\Delta_I=45.7\pm 4.1$ μeV . From γ' line slope, we estimated the staggered sub-lattice splitting value to be $\Delta_{\gamma'}=25.3\pm 6.1$ μeV for monolayer graphene, consistent with the value obtained in bilayer graphene of $\Delta_{\gamma'}=28.8\pm 3.7$ μeV .

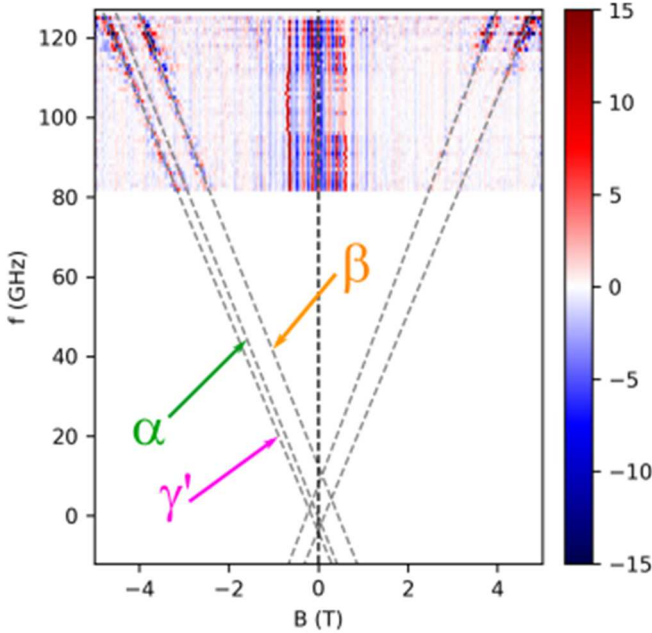


Fig. 1. Second derivative of photoconductivity signal measured in bilayer graphene as a function of the magnetic field at 2.6 K in the frequency range 82-125 GHz. The dashed lines are linear fits, allowing for the extrapolation of the experimental results to $B = 0$. The highest dashed line (β resonance) follows the linear evolution with a slope of -29.3 GHz/T ± 0.4 GHz/T and with an intercept at $B = 0$ of $+11\pm 1.0$ GHz (45.7 ± 8 μeV). The two other dashed lines corresponds to the α (resp. γ') resonance and have almost the same slope of -28.2 GHz/T ± 0.4 GHz/T (resp. -27.4 GHz/T ± 0.3 GHz/T) but their intercepts extrapolate to -1.1 ± 1.0 GHz (-4.5 ± 4.1 μeV) and -7.0 ± 0.9 GHz (-28.8 ± 3.7 μeV) at $B = 0$, respectively.

We also address experimentally the dependence in temperature of these electron spin resonances, from 2.6K to 12K. In that aim, the photoconductivity has been measured for a monolayer based Ratched THz sensor, at different temperatures and different magnetic fields for a fixed probe frequency of 60 GHz. It can be seen that the ISO ZFS increases from 43 μeV to 50 μeV between 12 K and 2.6 K, while the ZFS due to the sub-lattice potential increases from 26 μeV to 32 μeV in the same temperature range (Fig. 2).

IV. SUMMARY

Thanks to a new ultrasensitive ERS technique in the 50 GHz - 300 GHz range, based photoconductivity measurements, we

have investigated the influence of temperature on different spin-splittings in monolayer and bilayer graphene. By extrapolating these resonance frequencies at zero field, we extracted two ZFSs, attributed to ISO and sublattice potential, whose values are comparable to those in the literature. By varying the temperature of these two samples, we found that these two ZFS increase by about 20% when the temperature drops from 12 K to 2.6 K. The origin of this temperature dependence requires further theoretical and experimental investigations.

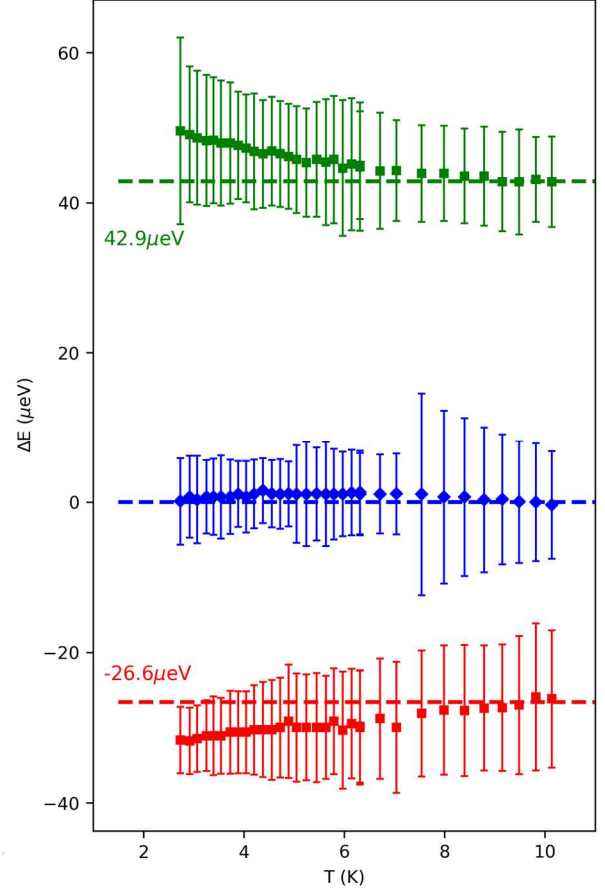


Fig. 2. Energies of the intercept at zero-field origin versus temperature for the three observed transitions: α (blue points), β (green points) and γ' (red points).. Dash line represents the converge values of energy at high temperature, corresponding to 42.9 μeV for ISO splitting and -26.6 μeV for the staggered sub-lattice splitting.

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