Dirac imprints on the \( g \)-factor anisotropy in graphene

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Dirac electrons in graphene are to lowest order spin 1/2 particles, owing to the orbital symmetries at the Fermi level. However, anisotropic corrections in the \( g \)-factor appear due to the intricate spin-valley-orbit coupling of chiral electrons. We resolve experimentally the \( g \)-factor along the three orthogonal directions in a large-scale graphene sample. We employ a Hall bar structure with an external magnetic field of arbitrary direction, and extract the effective \( g \)-tensor via resistively-detected electron spin resonance. We employ a theoretical perturbative approach to identify the intrinsic and extrinsic spin orbit coupling and obtain a fundamental parameter inherent to the atomic structure of \( ^{12} \text{C} \), commonly used in ab-initio models.

One of the great triumphs of the Dirac relativistic theory for the electron was the prediction of the \( g \)-factor with the value \( g_0 \simeq 2 \) [1]. As a major departure from previous quantum theories, Dirac’s equation describes indeed spin 1/2 particles with 4-component spinors or bispinors, allowing to introduce the concepts of chirality and helicity. Chirality is an inherent property of the particle, whereas helicity depends on its momentum: namely, it is positive (negative) when the momentum aligns (anti)parallel to the spin. In the massless limit, both qualities are related [2]: positive chirality corresponds to positive helicity and vice versa.

The linear dispersion at the Fermi level of graphene is cognate with the Dirac cones of the massless relativistic particles [3], motivating extensive research towards the parallelism with relativistic quantum mechanics in a solid-state material [4–9]. The inherent chirality of the Dirac carriers leads to a topologically non-trivial band structure [10–12]. Although the carriers are, to lowest order, spin 1/2 particles, their chirality induces a coupling of spin, valley and orbital degrees of freedom [13]. Here, we address this particular coupling that appears as a measurable \( g \)-factor anisotropy.

From a theoretical perspective, the Zeeman Hamiltonian that describes the interaction with an applied field is given by the sum of the contributions of the orbital and spin angular momentum, \( \hat{L} \) and \( \hat{S} \), respectively [14],

\[
\hat{H}_Z = \mu_B \hat{B}(\hat{\mathbf{L}} + g_0 \hat{S}),
\]

with \( g_0 \) representing the pure \( g \)-factor, \( \mu_B \) the Bohr magneton and \( \hat{B} \) an external magnetic field. On the other hand, the effective spin model, commonly employed experimentally to describe the Zeeman energy, includes an effective \( g \) tensor and fictitious spin operators [15–17],

\[
\hat{H}_{\text{eff}} = \mu_B \hat{B} \hat{g} \hat{S}.
\]

\( \hat{g} \) must be constructed such that the energies obtained with the effective spin Hamiltonian capture the corrections due to the internal molecular orbital angular momentum. In electron-spin resonance (ESR) experiments, this internal structure modifies the strength of an external field necessary to meet the resonant condition [18]:

\[
hv = \mu_B \langle \hat{B}(\hat{\mathbf{L}} + g_0 \hat{S}) \rangle = \mu_B \langle \hat{B} \hat{g} \hat{S} \rangle, \tag{1}
\]

where \( \hat{g} \) is a tensor that contains the effective (or experimental) \( g \)-factors measured with the field along the corresponding directions and \( \langle \cdot \rangle \) indicates the expectation value. Since the \( g \)-tensor is diagonal along the crystallographic directions, the angular dependence for the general rhombic symmetry can be expressed in terms of \( g_{xx} \), \( g_{yy} \) and \( g_{zz} \) [16]:

\[
g(\theta, \varphi) = \sqrt{g_{zz}^2 \cos^2 \theta + g_{yy}^2 \sin^2 \theta \sin^2 \varphi + g_{xx}^2 \sin^2 \varphi \cos^2 \varphi}, \tag{2}
\]

for an arbitrary magnetic field with axial and azimuthal angles \( \theta, \varphi \). In this letter we resolve experimentally the effective \( g \)-factor along the three main directions in a mesoscopic graphene sample, \( g_{\alpha\alpha} = g_0 + \Delta g_{\alpha\alpha} \), whereas the corresponding theoretical correction is evaluated perturbatively via the expectation value of the angular momentum, \( \Delta g_{\alpha\alpha} = \langle \hat{L}_\alpha \rangle, \alpha = x, y, z \). We employ a microscopic perturbative model to obtain \( \Delta g_{\alpha\alpha} \) in terms of atomic parameters [13]. We then compare these theoretical values to our experimentally obtained \( g \)-factors and extract the atomic spin-orbit coupling (SOC) corrections.

It is commonly accepted that near the Dirac points (DPs) the eigenstates are described by \( \pi \)-orbitals near the Fermi edge [19]. The conduction and valence bands, to lowest order, are linear in momentum, with the corresponding chiral states given in terms of the main \( (p_z\text{-orbital}) \) contribution at sublattices \( A \) and \( B \) [19, 20]:

\[
|\varphi_{\pm}^{(0)}\rangle \simeq c_A |p_z^A\rangle + c_B |p_z^B\rangle, \quad \frac{c_B}{c_A} = \pm e^{i\varphi_{q\tau}}, \quad \varphi_q = \arctan \frac{q_y}{q_x}, \tag{3}
\]
where the sign $\pm$ labels the conduction band (CB) and valence band (VB), respectively. These DPs are the celebrated $K$ and $K'$ points, which are assigned the valley index, $\tau = 1$ and $-1$, respectively, and $\vec{q} = (q_x, q_y)$ is the small vector off the nearest DP. The chirality-preserving Kane-Mele intrinsic SOC term [10], $\hat{H}_{\text{KM}} = \tau \hbar \tilde{s}_z \sigma_z$, with $\tilde{s}_z$, $\sigma_z$ being the Pauli matrices representing the electron spin and sublattice-spin, respectively, leads to the spin Hall effect and a measurable intrinsic SOC gap $\Delta_{12} = 2\lambda_1$ [12, 21, 22]. As we will show, the intrinsic SOC leads as well to chiral spin-valley orbit coupling and additional corrections to the measured $g$-factor. To lowest order, the quasiparticle eigenenergies are given by:

$$\varepsilon_{\pm} = \pm \sqrt{(\hbar v_F q)^2 + \lambda_1^2}.$$ 

with $v_F$ being the Fermi velocity. The axial symmetry of the $p_z$-orbitals involves $(L_\sigma) = 0$, and hence, the $g$-factor at lowest order is that of free electrons. Dominant corrections to the $g$-factor are due to (i) band hybridization, (ii) atomic SOC, (iii) Bychkov-Rashba effect and (iv) structural SOC, which we consider in the following.

As pointed out by McClure et al. [23], the $\tau$-bands are $p_z$-orbitals hybridized with $d_{xz}$- and $d_{yz}$-orbitals of the nearest neighbor (NN). Owing to the large energy difference, the $p_z$-contribution is dominant near the Fermi energy. We obtain perturbatively the $d$-band contribution, [24–26]

$$|\varphi_d^{(1)}\rangle = \frac{3i\tau V_{pd\tau}}{2\varepsilon_{pd\tau}} \left[ c_A |2\tau\rangle^B + c_B |2 - \tau\rangle^A \right],$$

where $\varepsilon_{pd\tau}$ is the energetic difference between the $d$- and $p$-orbitals and $V_{pd\tau}$ is the relevant $p-d$ coupling. Here, we have expressed the $d$-orbitals in the angular momentum representation, $|l, m_l\rangle$, with $\sqrt{2l+1} |d_{xz}\rangle \pm i |d_{yz}\rangle$. It is worth noting in Eq. (5) that $m_l$ relates to the valley index, $\tau$, commonly termed as *valley-orbit* coupling. This is connected to the chirality of the Dirac electrons: in one sublattice, the $p_z$-electrons couple to those of $d$-orbital with $m_l = 1$ ($m_l = -1$) in valley $K$ ($K'$), and the converse occurs for the other sublattice [13]. As we will see below, this has important consequences for the $g$-factor corrections (see diagram of Fig. 1).

On the other hand, the $\sigma$-band is constituted of $s$ and $p_{x,y}$ of the NN [27]. The $\pi$-bands described by Eqs.(3) and (5) can mix with the $\sigma$-bands either intrinsically via atomic spin-orbit interaction or extrinsically, via structural SOC or Bychkov-Rashba effect. The latter emerges as the horizontal mirror symmetry breaks and is linear in (uniaxial) electric field [25, 28–30], leading to an atomic dipole moment, commonly termed as Stark effect. Microscopically, the induced dipole results in a non-zero intraatomic coupling between the $p_z$- and $s$-orbitals. The structural SOC is related to a horizontal plane mirror asymmetry (PIA) [31–34] originated by ripples, defects or adsorbates, coupling $p_z$ and $p_{x,y}$-orbitals.

The $\sigma$-band mixing near the Fermi energy is expected to be smaller than the $d$-band contribution, since the atomic SOC parameter and the Stark parameter, $\lambda_z = eE|\hat{z}|\langle\hat{p}_z\rangle$ are small compared to the $p-d$ coupling, $\lambda_{soc}^d, \lambda_{soc}^s, \lambda_z \ll V_{pd\tau}$. We thus consider the $\sigma$-band mixing perturbatively, $\hat{V} = \hat{V}_{soc} + \hat{V}_{\text{PIA}} + \hat{V}_{\text{ERP}}$, with:

$$\hat{V} = i\epsilon_{\tau\alpha\beta\gamma} \lambda_{soc}^\alpha \langle \varphi^{(0)}_\tau | p_\beta | \varphi^{(0)}_{\tau'} \rangle + \lambda_{soc}^s | p_\tau \rangle | p_{\tau'} \rangle \rangle + \lambda_z | s^\tau \rangle | s_{\tau'} \rangle \rangle + \hbar c$$

where we have included the atomic $p$-orbital coupling $\lambda_{soc}^p$ and structural SOC in $\lambda_z$ and the Einstein summation convention is assumed. The projection over the orbital eigenstates yielding finite angular momentum contributions are [13],

$$\hat{P} |\varphi_{\tau}^{(1)}\rangle = (\tau \alpha^\tau c_A - is_{\tau} \sigma_z c_{\beta\tau}^\beta |1 - \tau\rangle^A - (\tau \alpha^\tau c_B - is_{\tau} \sigma_z c_{\alpha\tau}^\alpha |1\rangle^B),$$

where $\hat{P} = |11\rangle\langle11| + |1\rangle\langle1| |1\rangle - |1\rangle\langle1| |1\rangle$ and $\alpha^\tau$, $\alpha^\sigma$ are the $\sigma$-band intrinsic and extrinsic SOC coefficients:

$$\alpha^\tau = \sqrt{2}\lambda_{soc}^p \left( \frac{\sin^2 \gamma}{\varepsilon_\tau^+} + \frac{\cos^2 \gamma}{\varepsilon_\tau^-} \right), \quad \alpha^\sigma = \alpha_{\text{BR}} + \alpha_{\text{PIA}},$$

with $\tan \gamma = 3\sqrt{2} \lambda_{soc}^p / 2|\varepsilon_{\tau}^+|$, $V_{soc}$ being the $\sigma$-coupling of the $p$- and $s$-orbitals and $\varepsilon_{\tau}^\pm = \varepsilon_{\tau} \pm \sqrt{\varepsilon_{\tau}^2 + 2(3\lambda_{soc}^p)^2}/2$. Finally, $\alpha_{\text{BR}}$ and $\alpha_{\text{PIA}}$ accounts for the Bychkov-Rashba and the SL asymmetric SOC, respectively.

Eqs.(5) and (6) yield three different second order contributions for $\langle \hat{L}_z \rangle$, that is, $\Delta g_{zz} = \sum_i \langle \varphi_i^{(1)} | \hat{L}_z | \varphi_i^{(1)} \rangle$, giving:

$$\Delta g_{zz} \simeq \tau \rho^0_{\tau} \left( |\lambda_{soc}^s \rangle \langle \lambda_{soc}^s | - \langle \alpha^\tau \rangle^2 - \langle \alpha^\sigma \rangle^2 \right),$$

where we have defined $\rho^0_{\tau} = \langle \varphi^{(0)}_\tau | \sigma_z | \varphi^{(0)}_{\tau} \rangle = |c_A|^2 - |c_B|^2$, and we have used the result of Konschuh et al. [25], $\lambda_I = 9V_{pd\tau}^2 \lambda_{soc}^d / (2\varepsilon_{pd\tau}^2)$, with $\lambda_{soc}^d$ being the atomic SOC for the $d$-orbitals. We note that all three terms are proportional to $\tau (\sigma_z)$, due to the valley-orbit coupling, and the first term is dominant, as we will see.

![FIG. 1. Illustration of the spin-valley-orbit coupling of Dirac carriers in the top valence band. Spin ‘down’ carriers couple to counter-clockwise ($m_l = 1$) rotating orbitals, whereas spin ‘up’ carriers couple to clockwise ($m_l = -1$) rotating orbitals.](image)

Fig.1 illustrates the underlying nature of the spin-valley-orbit coupling for the lowest bands given in Eq.(4).
The highest populated state is characterized by $\tau \sigma_z \sigma_z = -1$. For sublattice $B$ (blue, $\sigma_z = -1$), the state has spin ‘up’ in the $K$-valley, and it couples to an anti-clockwise rotating $d_-$-orbital, whereas the spin ‘down’ in the $K'$-valley couples to the clockwise rotating $d_+$-orbital. The converse occurs for sublattice $A$ (magenta, $\sigma_z = 1$), where the spin ‘up’ (‘down’) is in the $K'$- ($K$-) valley, but it couples to the $m_l = -1$ ($m_l = 1$) $d$-orbital.

Hence, in the presence of spin-valley-orbit coupling, the Dirac carrier’s spin direction opposes that of the $m_l$ quantum number of the coupled $d$-orbital, reducing the effective $g$-factor at leading order.

We now consider the in-plane corrections, $\langle L_x \rangle$ and $\langle L_y \rangle$, with $\Delta g_{xx} = 2R\langle \varphi_{\pm}^{(0)} | \hat{L}_x | \varphi_{\sigma}^{(1)} \rangle$. We choose the $\hat{x}$ axis to be parallel to a zig-zag direction. The theoretical model assumes a well-defined crystalline zig-zag direction, which can be generalized as the transport direction in the polycrystalline, continuum limit. Using Eq.(6) and $\sqrt{2} \hat{L}_x | 1, \pm \tau \rangle^a = | p_+^a \rangle$, we obtain first order corrections:

$$\Delta g_{xx} = \pm 2\sqrt{2} (\alpha_{\pm}^a \tau \sigma_z^0 + \alpha_{\pm}^0 \sigma_z \tau \sin \varphi_{\pm}),$$

$$\Delta g_{yy} = \mp 2\sqrt{2} \alpha_{\pm}^0 \sigma_z \cos \varphi_{\pm}. \quad (8)$$

The intrinsic contribution results in a (dominant) negative correction for the highest populated band, whereas the sign of the extrinsic one depends on the electric field direction. In a single-particle theoretical picture, all corrections would vanish, as $\langle \tau \sigma_z \rangle$ averages out to zero. However, under real experimental conditions and in a macroscopic graphene sample with a spin imbalance $n_1 - n_\parallel \neq 0$, the problem becomes many-body and corrections to the $g$-factor emerge.

![FIG. 2. Schematic setup of our ESR measurements. The external magnetic field $|\vec{B}| \lesssim 1$ T can freely rotate, while a Hertzian loop antenna induces an AC field. A constant current flows along the $x$ direction, and the longitudinal voltage $V_{xx}$ is measured. The graphene layer rests on 300nm SiO$_2$ on top of a p-Si substrate that is grounded. The blow-up on the right-hand side illustrates the granular nature of the CVD graphene. The polycrystallinity retains zig-zag directions (red bold lines) parallel to the transport current.](image)

![FIG. 3. (b) Resonance frequency as a function of field strength $|\vec{B}|$ at constant $\theta$ in the $x-z$ plane. The data are taken at $T = 1.4$K, with a microwave radiation power of 21 dBm. The $g$-factor is given by the slope of the linear fit, $g_{xx} = 1.87 \pm 0.01$.](image)

The graphene that was synthesized by chemical vapor deposition is described by Lyon et al. [35]. We employed low temperature (1.4K) resistively-detected electron-spin resonance (RD-ESR) [12, 36, 37], a spin-selective probing technique that couples carriers of opposite spin by microwave excitation, and detects the response resistively. The large dimension of our device ensures the continuum limit with a well-defined bulk gap and chirality for the charge carriers [10], and the polycrystalline nature of the sample retains the theorized zig-zag directions parallel to the transport directions, as illustrated in Fig.2. Polycrystallinity also induces disorder, which broadens the resonant signal and facilitates the resistive detection.

The microwave excitation field is generated by a Hertzian loop antenna adjacent to the sample [see Fig. 2]. A constant low frequency current $I = 1$ mA is passed through the sample along $x$-direction, which we can relate to the propagating zig-zag direction [blow-up in Fig. 2], while a standard lock-in technique probes the resulting longitudinal resistance, $R_{xx} = V_{xx}/I$, as a function of the magnetic field $\vec{B}$. The magnetic field vector $\vec{B}$ can freely rotate with respect to the sample plane. Hence, we use spherical coordinates to denote the orientation of $\vec{B}$, with $\theta$ as the out-of-plane angle and $\varphi$ for in-plane rotations. All measurement are performed without the application of a gate voltage (substrate grounded), corresponding to an intrinsic density of $n \approx 6 \times 10^{11}$ cm$^{-2}$.

Whenever the microwave frequency $\nu$ matches the resonant condition of Eq.(1), the increased band population reduces $R_{xx,\nu}$. We can resolve these resonances as peaks in the microwave-induced differential resistance, $\Delta R_{xx}(\nu) = R_{xx,\nu,\text{dark}} - R_{xx,\nu}$.

Fig. 3 shows the electron spin resonances for different values of $|\vec{B}|$ under constant angle $\theta$ in the $x-z$ plane.
FIG. 4. Angular dependence of the g-factor: (a) g-factor for a rotation of \( \theta \) in the \( y-z \) plane and (b) for a rotation of \( \theta \) in the \( x-y \) plane and (c) for a rotation of \( \phi \) in the \( x-y \) plane. Along the in-plane, we extract \( \Delta g_{xx} = -0.19 \pm 0.01 \) and \( \Delta g_{yy} = 0.03 \pm 0.01 \), whereas \( \Delta g_{zz} = -0.05 \pm 0.01 \).

Each data point is the result of a Gaussian fit to the resonance curves \( \Delta R_{zz}(\nu) \) (not shown). The data points follow a linear dispersion that reflects the magnetic field dependence of the Zeeman energy. Its slope thus represents the effective g-factor of the Dirac electrons for the magnetic field under \( \theta \).

The procedure is repeated for \( \vec{B} \) pointing in different directions, i.e. for various \( \theta \) and \( \phi \), allowing us to resolve the anisotropic g-factor as defined in Eq.(2). Fig.4 is an angle-resolved study of the effective g-factor within the planes marked schematically inside each graph. In Fig.4(a) we explore the \( y-z \) plane, where a sinusoidal dependence of the g-factor on the axial angle \( \theta \) is apparent. When the external field is oriented perpendicular to the sample plane (\( \theta = 0 \)), we obtain \( g_{zz} = 1.95 \pm 0.02 \)[12], whereas for \( \theta = 90^\circ \), we obtain \( g_{yy} = 2.03 \pm 0.02 \). Fig.4(b) shows a rotation of \( \theta \) in the \( y = 0 \) plane. Here, the effective g-factor is smallest when \( \vec{B} \) is collinear to the current direction and becomes \( g_{xx} = 1.81 \pm 0.02 \). The in-plane variation of \( \phi \) at fixed \( \theta = \pi/2 \) is shown in Fig.4(c) for completeness. We attribute the asymmetry to the current-induced Rashba effect [38] and to the polycrystalline structure of the graphene, where the current direction is defined only locally, as illustrated in Fig. 2. The largest correction is thus obtained for \( \Delta g_{xx} = -0.19 \pm 0.01 \), consistent with the first-order intrinsic SOC, and the smallest correction is \( \Delta g_{yy} = 0.03 \pm 0.01 \), consistent with a small extrinsic SOC in the absence of gating. Finally, \( \Delta g_{zz} = -0.05 \pm 0.01 \), corresponding to a second order correction. Table I summarizes the experimentally extracted elements of the g-tensor and the g-factor anisotropy.

We can extract atomic SOC parameters that lead to the observed g-factor corrections. For the in-plane corrections, using (8) with \( \varphi_q = 0 \), we obtain \( \alpha^d_{\varphi} \simeq (0.01 \pm 0.03) \). We also obtain \( \alpha^d_\psi \simeq (0.067 \pm 0.003) \), giving an upper limit for \( \lambda_{soc}^p < 0.05 \mu \text{eV} \), consistent with the theoretical value [25–27, 29, 30, 39, 40].

For the axial correction, using \( \lambda_f \simeq 21 \mu \text{eV} \)[12] in Eq. (7) we obtain SOC parameter for \( d \)-orbitals, \( \lambda_{soc}^d \simeq (0.31 \pm 0.09) \) meV, which compares quite well with the DFT obtained value of 0.8 meV [25]. As pointed out by Konschuh et al., unlike in the \( \lambda_{soc}^p \) case, there is no possible fitting of the energy spectrum to obtain numerically this value, since the needed high-energy states in the conduction bands cannot be identified. We stress that this value is intrinsic: it is the atomic spin-orbit states not only for the special case of graphene, but for all \(^{12}\)C atoms.

In summary, we experimentally resolved the g-factor anisotropy in graphene using an angle-dependent ESR method. The chiral nature of the Dirac electrons in graphene entails corrections to the g-factor that originate from a peculiar spin-valley orbit coupling. Along the transport direction, we observe a negative first order correction, owing to the intrinsic, chiral SOC with the propagating \( p_x \)-orbital. We extract an intrinsic coupling of \( \alpha^d_\psi \simeq 0.067 \pm 0.003 \). Along the \( y \)-direction, the sign and magnitude of the g-factor correction reflects a extrinsic SOC, consistent with the absence of inversion symmetry. We extract an extrinsic coupling of \( \alpha^d_{\varphi} \simeq 0.001 \pm 0.003 \). In combination with the axial correction, we were able to extract intrinsic SOC parameter \( \lambda_{soc}^d = 0.31 \pm 0.09 \) meV.

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| \( g_x \) | \( g_{x-y} \) | \( g_y \) | \( g_{y-z} \) | \( g_z \) | \( g_{z-x} \) | Error |
|---|---|---|---|---|---|---|
| 1.81 | 1.91 | 2.03 | 1.99 | 1.95 | 1.87 | 0.01 |
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