Magnetic quantum ratchet effect in graphene

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A periodically driven system with spatial asymmetry can exhibit a directed motion facilitated by thermal or quantum fluctuations1, This so-called ratchet effect has fascinating ramifications in engineering and natural sciences. Graphene is nominally a symmetric system. Driven by a periodic electric field, no directed electric current should flow. However, if the graphene has lost its spatial symmetry due to its substrate or adatoms, an electronic ratchet motion can arise. We report an experimental demonstration of such an electronic ratchet in graphene layers, proving the underlying spatial asymmetry. The orbital asymmetry of the Dirac fermions is induced by an in-plane magnetic field, whereas the periodic driving comes from terahertz radiation. The resulting magnetic quantum ratchet transforms the a.c. power into a d.c. current, extracting from terahertz radiation. The resulting magnetic quantum ratchet effect2 has fascinating ramifications.

By exciting single-layer graphene samples with the alternating electric field of a terahertz laser22–24, we observe a d.c. current in the presence of an in-plane magnetic field. (For experimental details see Fig. 2, right inset, and Supplementary Section S3.) The current scales with the square of the electric field amplitude, increases linearly with the magnetic field, and reverses its sign with a change in the direction of the magnetic field, which is a clear signature of the magnetic ratchet effect. Figure 2 shows the temperature dependence of the current density j, measured in epitaxial graphene on SiC (refs 25–27; for sample descriptions see Supplementary Sections S1 and S3) for the experimental geometry of the model shown in Fig. 1 (that is, E(t)∥x and B∥y). The current magnitude remains almost constant up to ~100 K and decreases above this temperature. To prove that the signal stems from graphene and not, for example, from the substrate, in the control experiments we removed the graphene layer from one of the samples, and in another scratched the graphene layers, thereby isolating them from the contacts. In both cases we observed that the signal disappeared. Strikingly, we observe that the sign of the slope j,B is consistently opposite for two groups of epitaxial graphene samples (Fig. 2, left inset) that differ only in the manner of surface treatment. The surfaces of samples A and B, which exhibit a positive slope, were encapsulated in a thin polymer film (see ref. 29 and Supplementary Section S1), but the surface of sample C, which has a negative slope, remained unprotected and vulnerable to uncontrolled contamination from the ambient atmosphere. The observed difference in the current direction for covered and uncovered graphene samples demonstrates that the sign of the structure inversion asymmetry (SIA) is consistently opposite for these groups of samples. The magnetic quantum ratchet effect was also observed for graphene sample D, which was grown by chemical vapour deposition (CVD); the field dependence of the current for this sample is also shown in Fig. 2, left inset. The current strength in this sample is a factor of ~7 and ~20 times smaller than the currents in samples A and C, respectively. As in epitaxial graphene samples, the current decreases with increasing temperature, and at T > 100 K it could no longer be detected.

The origin of the ratchet effect suggests that the directed particle flow depends on the orientation of the alternating force with respect to the direction of built-in spatial asymmetry. In magnetic quantum ratchets, where the asymmetry stems from the magnetic field, the relevant parameter is the angle β between the a.c. electric field E(t) and the magnetic field B. The corresponding dependence j,B,
The experiments discussed so far were carried out for an alternating electric field aligned along a certain direction, a common geometry for electronic ratchet problems. However, ratchet transport can also be induced by a force rotating in space. By exciting the graphene samples with a clockwise or anticlockwise rotating in-plane electric field \( E(t) \), we also detect a d.c. current. The current measured along the \( x \)-axis, that is, perpendicular to the magnetic field, turns out to be insensitive to the rotational direction of \( E(t) \) and, as expected, just gives the value of \( j_x \). In contrast, the current along the magnetic field \( j_y \) is of the opposite sign for the clockwise and anticlockwise rotating fields (Fig. 4). This effect can be referred to as the circular magnetic quantum ratchet effect.

We now present a microscopic theory, supported by first-principles calculations, of the observed magnetic quantum ratchet effect and show that it yields a nice agreement with the experiment. The effect originates from the interplay of the a.c. electric field and the asymmetric electron scattering in the in-plane magnetic field previously considered for inversion channels in silicon and semiconductor quantum wells\(^{30-32} \). The electric current density is calculated by using the general expression

\[
j = 4e \sum_p \nu f(p, t) \tag{2}
\]

where \( e \) is the electron charge, the factor 4 accounts for the spin and valley degeneracy in graphene, \( p \) is the momentum, \( v \) is the velocity, \( \nu \) is the kinetic energy, and \( f(p, t) \) is the carrier distribution function. The distribution function can be found from the Boltzmann equation

\[
\frac{\partial f(p, t)}{\partial t} + eE(t) \cdot \frac{\partial f(p, t)}{\partial p} = S[t(f(p, t)] \tag{3}
\]

where \( E(t) = E_{\text{Exp}}(-i\omega t) + E^\ast \text{Exp}(i\omega t) \) is the a.c. electric field oscillating at angular frequency \( \omega \), and \( S[t(f(p, t)] \) is the collision integral. For elastic scattering, it has the form

\[
S[t(f(p, t))] = \frac{2\pi}{h} \sum_p \langle [V_{pp'}] f(p', t) - f(p, t) \rangle \delta(e - e') \tag{4}
\]

where the angular brackets denote impurity ensemble averaging and \( V_{pp'} \) is the matrix element of electron scattering between the initial and final states with momenta \( p \) and \( p' \), respectively.

The ratchet currents stem from the asymmetry of electron scattering caused by \( \sigma-\pi \) hybridization around the Dirac points in the in-plane magnetic field. Formally, for magnetic field \( B_y \), it is described by the matrix element

\[
V_{pp'} = V_{\sigma\sigma} - B_y(p_x + p'_x) z_{\sigma\sigma} e \frac{e}{m_0 c} V_{\sigma\sigma} \tag{5}
\]

where \( z_{\sigma\sigma} \) is the coordinate matrix element between the \( \pi \) and \( \sigma \)-band states, \( e \) is the energy distance between the two bands, \( V_{\sigma\sigma} \) and \( V_{\pi\pi} \) are the intraband and interband matrix elements of scattering at zero magnetic field, \( m_0 \) is the free electron mass and \( c \) is the speed of light.

We solve Boltzmann equation (3) by expanding the distribution functions in a series of powers of the electric field. The first-order correction to the equilibrium distribution function oscillates at the radiation field frequency and does not contribute to the d.c. electric current density is calculated by using the general expression

\[
j = 4e \sum_p \nu f(p, t) \tag{2}
\]

Figure 1 | Dirac electrons drive a ratchet. The ratchet wheel (analogue of electric current) turns as the a.c. electric field \( E(t) \) from the terahertz radiation drives the electrons in graphene. The ratchet-and-pawl mechanism is induced by the static magnetic field \( B \) and the spatial asymmetry of graphene induced by the hydrogen adatoms (blue spheres). The resulting spatial distribution of the electron density is shown in red. If the electrons, at any time, are driven to the right by the electric field, their orbitals are shifted upwards due to a quantum analogue of the Lorentz force (left panel). Consequently, their mobility decreases; that is, friction increases. Half a period later, when electrons are caused to flow to the left, their orbitals are shifted downwards and the mobility increases (right panel).

Figure 2 | Temperature dependence of current density \( j_x \) measured in sample A. The a.c. electric field is aligned along the \( x \)-axis and the magnetic field along the \( y \)-axis. Data are obtained for an electric field amplitude of \( \sim 10 \, \text{kV cm}^{-1} \) and a static magnetic field \( B_y = \pm 7 \, \text{T} \). Left inset: magnetic field dependence of \( j_x(B_y) \). Right inset: sample D at 4.2 K. Note that curves for samples A, B and D in the inset are obtained at \( T = 150 \, \text{K} \) and data for sample D at 4.2 K. Note that curves for samples A, B and D in the inset are obtained at \( T = 150 \, \text{K} \) and data for sample D at 4.2 K.
current. The d.c. current is determined by the second order in $E$ corrections. Such a calculation yields

$$j_i = M_i(\langle |E|^2 \rangle - < |E|^2 > |B_y| + M_i |E|^2 B_y)$$

$$j_f = M_f(E_x E'_x + E_y E'_y)B_y + M_f i(E_x E'_y - E_y E'_x)B_y$$

(6)

The general form of parameters $M_i$, $M_f$ and $M_g$ is given in Supplementary Section S4. For the particular case of scattering by short-range defects and degenerate statistics, relevant to our highly doped samples, the parameters assume the form

$$M_i = \frac{12\alpha^2 c z_{ss} \langle V_{\sigma\sigma} V_{\sigma\sigma} \rangle}{\pi n_F e \sigma \langle V_{\sigma\sigma}^2 \rangle} \frac{\tau^2 E_F}{1 + (\omega \tau)^2}$$

(7)

$$M_f = -\frac{4\alpha^2 c z_{ss} \langle V_{\sigma\sigma} V_{\sigma\sigma} \rangle}{\pi n_F e \sigma \langle V_{\sigma\sigma}^2 \rangle} \frac{(1 - \omega^2 \tau^2/2)^2 E_F}{1 + (\omega \tau)^2}$$

(8)

$$M_g = \frac{6\alpha^2 c z_{ss} \langle V_{\sigma\sigma} V_{\sigma\sigma} \rangle}{\pi n_F e \sigma \langle V_{\sigma\sigma}^2 \rangle} \frac{\omega^2 E_F}{1 + (\omega \tau)^2}$$

(9)

where $\alpha = e^2/\hbar c$ is the fine-structure constant, $\tau$ is the momentum relaxation time and $E_F$ is the Fermi energy.

Equations (6) to (9) describe all major features of the observed magnetic quantum ratchet effect in graphene. First, in agreement with the experiment, all current components are proportional to the magnetic field and the square of the electric field amplitude. The equations yield that the dependence of the current (which flows in the direction perpendicular to the magnetic field) on the orientation of $E$ has the form $j_i = (M_i \cos 2\beta + M_f) |E|^2 B_y$. Exactly such a behaviour is observed experimentally (Fig. 3a, inset). Moreover, for a rotating electric field, the current along the external magnetic field, $j_p$, is sensitive to the direction of rotation. Indeed, the last term on the right-hand side in the second line of equation (6) is proportional to the factor $i(E_x E'_y - E_y E'_x)$, which depends on the phase shift between $E_x$ and $E_y$ components and is equal to $|E|^2$ for the clockwise or anticlockwise rotating electric field in our experimental geometry. This contribution has been observed experimentally and describes the data shown in Fig. 4. The circular current, which is sensitive to the direction of a.c. field rotation, reaches a maximum at $\omega \approx 1/\tau$, which is achieved in the terahertz range for our graphene structures, and vanishes at smaller or higher frequencies (see equation (9)). As for the linear ratchet effect, it can also be excited, for example by microwave radiation.

The symmetry arguments forbid any d.c. current induced by an in-plane a.c. field in centrosymmetric systems. Therefore, no ratchet effect is expected in graphene with equivalent ‘up’ and ‘down’ surfaces, for example, in free-standing graphene. It would be observable in such layers only for non-equal numbers of adatoms on the opposite sides of the graphene sheet. The theory presented above demonstrates that the ratchet current emerges if the SIA of the graphene layer is broken by the environment (adatoms or substrate) and is proportional to $(V_{\sigma\sigma} V_{\sigma\sigma})/(V_{\sigma\sigma}^2)$, which is the measure of SIA. We believe that the coupling of the current direction to the sign of the SIA is responsible for the observed consistent difference in the sign of the current for two groups of samples having almost identical transport parameters but different post-growth treatment of the graphene surface. Note that the sign of the current is independent of charge carrier type. Thus, the magnetic quantum ratchet effect provides non-invasive experimental access to the inversion asymmetry in graphene.

Finally, we compare the current magnitude $j_f$ measured in sample A with that given by equations (6) and (7). To estimate the current density at low temperatures we use the momentum relaxation time $\tau = 4.6 \times 10^{-14}$ s and the Fermi energy $E_F \approx 150$ meV, both extracted from Hall measurements (Fig. 3b). The matrix element $z_{ss} \approx 0.15$ Å and the energy $e \sigma \approx 10$ eV were calculated using the tight-binding method. Orbital mixing due to adatoms is given by $(V_{\sigma\sigma} V_{\sigma\sigma})/(V_{\sigma\sigma}^2) \approx 1$, as shown in Supplementary Section S6 for hydrogen adatoms. The reason for the strong mixing is the hybridization of the $p_z$ orbitals with the orbitals of...
the adatoms (x for hydrogen), and subsequent mixing with the $\sigma$ orbitals. The above values give $j \approx 1 \mu\text{A cm}^{-1}$ at a magnetic field of $B = 7$ T, an electric field amplitude of $2E = 10 \text{ kV cm}^{-1}$, and a frequency of $\omega = 2.1 \times 10^{13} \text{ rad s}^{-1}$. The current density $j_x$ measured for the same conditions, is $\approx 18 \mu\text{A cm}^{-1}$ (Fig. 3). The estimation for the current density at $T = 260$ K gives $j \approx 0.7 \mu\text{A cm}^{-1}$, rather than the $j \approx 2.6 \mu\text{A cm}^{-1}$ measured in the experiment. The magnitude of the signal detected in CVD graphene sample D at a low temperature is $j \approx 7 \mu\text{A cm}^{-1}$ (Fig. 3, inset). Our simplistic model gives a result that is within an order of magnitude of the experimental value. This is very satisfactory. We attribute the occurrence of current enhancement in epitaxial graphene on SiC to the substantial role of a buffer layer between the substrate and the actual graphene sheet, which is known to interact strongly with the graphene and may considerably increase SIA. The estimation shows (Supplementary Section S5) that the ratchet current in an asymmetric bilayer-like structure can be two orders of magnitude larger than that in single-layer graphene. It also indicates that even a small interaction between two layers should enhance the magnetic quantum ratchet effect. In principle, each specific adsorbate and substrate imprint its own specific quantitatively SIA signature on graphene, which could then be quantified by our magnetic ratchet mechanism.

To summarize, we have experimentally demonstrated and theoretically explained the magnetic quantum ratchet effect in graphene driven by an a.c. electric field. The magnitude of the observed d.c. current is a quantifying measure of the SIA of graphene, while the sheer existence of this ratchet current is proof that generic graphene flakes exhibit a macroscopic space inversion asymmetry. Moreover, it indicates that the effects of electron orbital coupling can appear and play an important role in the transport properties of the purest possible two-dimensional crystals. The ratchet current can be calibrated to measure the strength of the structure inversion asymmetry, which plays an important role in graphene ferromagnetism14 and spintronics35,36.

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Author contributions

S.D.G. and S.A.T. conceived the experiments. C.D., P.O., J.Ka., M.H., F.M. and S.D.G. designed the experimental set-up and performed the measurements. C.D., P.O., S.D.G. and S.A.T. analysed the data. R.V., S.A., S.K., J.Ko., P.M.A., M.W. and R.V. co-wrote the paper. All authors commented on the manuscript.

Additional information

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Competing financial interests

The authors declare no competing financial interests.

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