Photo-Nernst current in graphene

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Photocurrent measurements provide a powerful means of studying the spatially resolved optoelectronic and electrical properties of a material or device6–7. Generally speaking there are two classes of mechanism for photocurrent generation: those involving separation of electrons and holes, and thermoelectric effects driven by electron temperature gradients. Here we introduce a new member in the latter class: the photo-Nernst effect. In graphene devices in a perpendicular magnetic field we observe photocurrent generated uniformly along the free edges, with opposite sign at opposite edges. The signal is antisymmetric in field, shows a peak versus gate voltage at the neutrality point flanked by wings of opposite sign at low fields, and exhibits quantum oscillations at higher fields. These features are all explained by the Nernst effect8–10 associated with laser-induced electron heating8–11,14. This ‘photo-Nernst’ current provides a simple and clear demonstration of the Shockley–Ramo nature of long-range photocurrent generation in a gapless material6.

In a semiconductor, electrons and holes generated by photons can live long enough to be spatially separated by an electric field and diffuse to the contacts. This is the basis of photovoltaic cell operation. In contrast, in a gapless material such as graphene the full electron distribution rapidly thermalizes, eliminating the distinction between electrons and holes. Nevertheless, photocurrent is readily produced when light is focused on inhomogeneous regions or junctions in graphene devices5,7,11–13. Detailed measurements of the dependence on gate voltage and time delay have shown that it is primarily of a thermoelectric nature6,11,13,17,19, and that the heating of the electrons is sometimes enhanced by slow energy transfer to the lattice due to the large optical phonon energy and high electron velocity11,12–16. However, the means by which a thermoelectric current near the laser spot results in photocurrent in the contacts, which may be located some distance away, has received much less attention.

Unlike in a semiconductor, in graphene one cannot talk about diffusion of majority carriers to the contacts. Rather, in such a gapless material the localized current density source \(j_{\text{loc}}\) produces a global photocurrent \(I_{\text{ph}}\) by setting up an electric field that drives ambipolar carriers outside the excitation region and into the contacts, as recently discussed by Song and Levitov6. These authors derived an expression giving \(I_{\text{ph}}\) as an integral over \(j_{\text{loc}}\), analogous to the Shockley–Ramo theorem that gives the current generated between two conducting plates when a charge moves in the insulating space between them27–28. Our measurements on graphene devices in a magnetic field demonstrate the existence of the photo-Nernst effect which produces a photocurrent according to this theorem. In the presence of a perpendicular field \(B\) a transverse current proportional to \(B\) tends to circulate around the laser spot, that is, perpendicular to the electron temperature gradient in the graphene generated by the laser. When the laser is near a free edge, the integral of this chiral \(j_{\text{loc}}\) points along the edge, producing a photocurrent which is independent of distance from the contacts. The agreement of the observed dependence on gate voltage and magnetic field with conventional thermoelectric measurements on graphene Hall bars6–10 firmly establishes the photo-Nernst origin of the response.

We studied two-terminal monolayer devices of graphene on SiO\(_2\) selected to have relatively low disorder (see Methods). Results from one device are shown throughout, but consistent results were obtained on several devices. An optical image of the device is shown in Fig. 1a. A gate voltage \(V_g\) is applied to the conducting Si substrate, and all measurements are made at a stage temperature of 40 K. Figure 1b shows the conductance at \(B = 0\) as a function of gate voltage \(\Delta V_g\) measured relative to the conductance minimum (neutrality point), which in this device was at \(V_g = 1.3\) V. The field-effect mobility is roughly \(2 \times 10^5\) cm\(^2\) V\(^{-1}\) s\(^{-1}\). To measure photocurrent, a laser (wavelength 632 nm, spot size \(\sim 1\) µm) is focused on the surface and the short-circuit current \(I_{\text{ph}}\) is detected, in the direction indicated. The laser power was kept at 40 µW, after determining that \(I_{\text{ph}}\) is linear in power up to 60 µW (Supplementary Fig. 1). Scanning photocurrent microscopy (SPCM) was performed by measuring \(I_{\text{ph}}\) as a function of the laser spot position.

Figure 1c shows SPCM images taken at \(B = 0\) at a series of four gate voltages. As usual in higher-mobility graphene devices, photocurrent is generated mostly near the contacts and is larger at gate voltages near the neutrality point. Figure 1d,e show corresponding images at \(B = +0.3\) T and \(-0.3\) T. Substantial additional photocurrent is now seen when the laser is near the graphene free edges yet far from the contacts. The polarity of this field-induced photocurrent is opposite at the upper and lower edges and reverses when the field is reversed. In Fig. 2a,b we plot the \(B\)-symmetric and antisymmetric parts of \(I_{\text{ph}}\), respectively, at \(\Delta V_g = -5\) V. The symmetric part is very similar to the \(B = 0\) photocurrent, whereas the antisymmetric part consists of two stripes, of opposite polarity, parallel to the edges and having almost uniform intensity along each edge.

All of these observations can be understood by starting with the assumption that there is an increase in the electron temperature \(T(x,y)\) in and around the laser spot, which generates a thermoelectric current. Because the electron distribution thermalizes in \(\sim 100\) fs, \(T\) should be well defined on a scale of \(\sim 100\) nm. In similar zero-field measurements it has been shown to rise a few degrees above the lattice temperature6. At zero magnetic field and a given gate voltage, the thermoelectric current is parallel and proportional to \(\nabla T\), and by mirror symmetry this produces no net photocurrent when the spot is near an edge but far from the contacts. However, in a normal magnetic field \(B = Bz\) there exists a thermoelectric current density component \(j_y\) that is odd in \(B\) and directed normal to \(\nabla T\), as indicated in Fig. 2c. When the spot is near an edge this current density tends to point along the edge. When the spot is near the opposite edge the current density points in the opposite direction. Because of its long-range Shockley–Ramo

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nature, the resulting photocurrent is independent of the location of the spot along the edge and reflects the directionality of $j_n$. The gate-voltage dependence is produced by the dependence of the thermoelectric coefficients on carrier density.

We can model the results quantitatively by applying Song and Levitov’s theorem to the extrinsic thermoelectric current $j_{th} = -\hat{\alpha} \nabla T$, with $\hat{\alpha}$ the 2D thermoelectric tensor. Use of this local relationship is justified as the momentum relaxation length is less than $\sim 1 \mu m$. The total current density $j = j_{th} + j_e$, which obeys the continuity equation $\nabla \cdot j = 0$, also includes a part $j_e$ (not parallel to $\nabla T$ if $\hat{\alpha}$ is non-diagonal) driven by the electrochemical potential gradient that builds up. The resulting photocurrent is then given by $I_{ph} = \int j_e \cdot \nabla \psi \, dx \, dy$. Here the auxiliary scalar field $\psi(x, y)$ is such that $- (\rho')^{-1} \nabla \psi$ is the current distribution created when a unit bias is applied between the contacts at $V = 0$, and $\rho'$ is the 2D resistivity tensor.

For the particular case of a rectangular strip of isotropic 2D material, with contacts at $x = 0$ and $L$ and free edges at $y = 0$ and $W$, far from the contacts one finds $\nabla \psi = (\beta / L) (\hat{\mathbf{x}} + \hat{\mathbf{r}})$, where $\rho' = (\rho_{xx} / \rho_{xy})$ and the coefficient $\beta < 1$ depends on contact details. The photocurrent is then

$$I_{ph} = -\beta \frac{L}{L} \int \left[ (\alpha_{xx} + r \alpha_{xy}) \frac{\partial T}{\partial y} + (\alpha_{yy} + r \alpha_{yx}) \frac{\partial T}{\partial x} \right] \, dx \, dy$$

The term involving $(\partial T / \partial x)$, which is even in $B$, gives zero on integration if the temperature rise at the contacts is negligible (or if it is the same at both ends). The other term is odd in $B$ and is proportional to the average temperature difference $\Delta T_{av}$ between the two free edges:

$$-\beta \left( \alpha_{xy} + r \alpha_{yx} \right) \frac{1}{L} \int_0^L dx \int_{-W/2}^{W/2} dy \frac{\partial T}{\partial y} = -\beta \left( \alpha_{xy} + r \alpha_{yx} \right) \Delta T_{av}.$$  

From the definition of the Nernst coefficient $N$ as the ratio of the transverse electric field to the longitudinal temperature gradient times the magnetic field at zero charge current, it immediately follows that $- (\alpha_{xy} + r \alpha_{yx}) = NB / \rho_{xx}$ for isotropic material. Hence, for a rectangular device, when the laser spot is far from the contacts,

$$I_{ph} = \beta NB \rho_{xx}^{-1} \Delta T_{av}$$

We conclude that the photocurrent is proportional to the Nernst coefficient divided by the longitudinal resistivity. This is likely to remain approximately true for less regularly shaped graphene.

It can be seen from equation (1) that this ‘photo-Nernst’ current will be odd in $B$, largest when the laser is near an edge (thus maximizing $\Delta T_{av}$), opposite in polarity at opposite edges, and independent of position along the edge when the laser is far from the contacts. All these facts are consistent with our observations.

To be more quantitative, we performed a calculation of $I_{ph}$ for the rectangular strip model. We calculated $\Delta T_{av}$ as a function of laser spot position by solving the heat equation given parameters reported in the literature for graphene on SiO$_2$, of similar mobility (see Supplementary Information). With these parameters, heat flow to the substrate (by optical phonon coupling) dominates lateral heat flow in the graphene, unlike in very high mobility devices. In this limit the temperature rises only close to the region where power is absorbed, explaining why photocurrent is produced mostly at distances roughly within the size of the laser spot from the contacts or edges; and $\Delta T_{av}$ is not very sensitive to the thermal conductivity of the graphene. A spatial pattern very similar to that in Fig. 2b was obtained with just two fitting parameters, the laser spot size and an overall multiplicative factor, as shown in Fig. 2d. A comparison of the calculated transverse profile with the measured photocurrent along the dotted green line in Fig. 2b is shown in Fig. 2e.

To provide more intuition for the long-range photocurrent generation process, we show in Fig. 2f,g calculated patterns of the $B$-antisymmetric component of the total current density $j$ that can readily be computed in this simple geometry (see Supplementary Information). When the spot is near the top edge (f), the Nernst effect drives current predominately to the right (taking $N$ to be negative). Continuity, and the fact that $j_h$ cannot flow in a closed loop, causes the current flow lines to continue into the contacts. When the laser spot is in the centre of the strip (g), the net photocurrent is zero, although the Nernst effect produces a circulating current within the graphene. The largest photocurrent occurs slightly inside the edge because the temperature rise is slightly larger when more of the laser spot is on the graphene.

We now consider the dependence of the photo-Nernst current on $V_g$ and $B$. The Nernst effect in graphene Hall bars has been measured before by conventional means, and considered theoretically. If the Mott relation applies then $N$ can be written in terms of the

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**Figure 1** | Scanning photocurrent microscopy (SPCM) of a two-terminal monolayer graphene device in a perpendicular magnetic field. a, Optical image of the device and indication of the experimental set-up. The graphene free edges are marked by dashed lines. b, Conductance at 40 K and $B = 0$ versus gate voltage $\Delta V_g = V_g - V_{g0}$ measured relative to the charge neutrality point, $V_{g0} = +1.3 \, V$. c-e, SPCM images at $B = 0$ (c), $+0.3 \, T$ (d) and $-0.3 \, T$ (e), for the indicated values of $\Delta V_g$. The contact edges are indicated by thin white lines. Scale bar, $2 \, \mu m$. 

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The Mott relation assumes purely elastic, independent-electron transport and a slowly varying density of states. It should not apply when $\epsilon_F$ is within several $kT$ of the Dirac point in perfect graphene. However, the behaviour of transport properties near the neutrality point in real devices is obscured by density inhomogeneities, and deviations from equation (2) may be hard to detect.

Figure 3 shows how the photocurrent depends on gate voltage at a moderate magnetic field, $B = -0.3$ T. Its magnitude and polarity change with $V_g$, whereas the maxima remain at the same spatial positions near the edges, as can be seen in Fig. 3a. The top axis here shows the 2D carrier density, defined by $n = (C/\epsilon) (V_g - V_0^\phi)$, taking $(C/\epsilon) = (\epsilon_F/\epsilon_0) e d_ox = 7.2 \times 10^{10}$ cm$^{-2}$ V$^{-1}$ for oxide thickness $d_ox = 300$ nm and $\epsilon_F = 3.9$. $n$ is negative for holes, and loses meaning near the neutrality point owing to density inhomogeneities. Figure 3b shows the dependence on $V_g$ at a fixed laser position (corresponding to the white dashed line in Fig. 3a). The photocurrent exhibits a peak at the neutrality point, changes sign for electron or hole density above $\sim 10^{11}$ cm$^{-2}$, and decays slowly at higher densities. Both the peak at the neutrality point and the opposite-sign tails at higher density are consistent with previous Nernst measurements$^{5-10}$ and are qualitatively explained by the energy derivative of $\rho xx$, in equation (2). If the width of the peak, $\Delta n \sim 10^{10}$ cm$^{-2}$, is set by density inhomogeneities, it should be similar to the width of the resistance peak at the neutrality point, shown in Fig. 3c. The two are indeed fairly similar. We note that the slight difference in the gate-voltage position of the peaks on the upper and lower edges apparent in Fig. 3a can be explained by a density variation across the sample.

At higher magnetic field quantum oscillations develop$^7$ as a function of $V_g$, as illustrated in Fig. 4a. The variation of the peak photocurrent with $V_g$ at a series of magnetic fields is shown in Fig. 4b. For the data at 3 T the filling factor $\nu = nh/eB$ is shown on the top axis. Again, the results resemble previous Nernst measurements$^5-10$ and are qualitatively explained by equation (2), with the oscillations reflecting the energy derivative of the Shubnikov–de Haas oscillations in $\rho xx$. The oscillations are thus large on a small background, in contrast with the much weaker oscillations in the resistance, which is shown in Fig. 4c. We note, however, that one needs to measure $\rho xx$ employing a multiterminal device to determine $N$ accurately using equation (1).

We finish by pointing out that scanning photo-Nernst microscopy could be used as a powerful new probe of materials and devices. For graphene the signal is large and easily detected at low fields (for example, 50 mT, Supplementary Fig. 2), and at room temperature (Supplementary Fig. 3). It can be measured on a simple two-terminal sample, where scanning can reveal spatial variations of properties. It can be separated from other photocurrent contributions by extracting the $B$-antisymmetric signal. Extended to lower temperatures and higher magnetic fields, the technique should yield new insights into the integer and fractional quantum Hall regimes. In those regimes, edge states and correlations may violate the conditions of locality or elastic
Figure 4 | Quantum oscillations in the photo-Nernst effect. a, Variation with carrier density of the photocurrent along a line across the graphene strip at 3 T. The top axis shows the corresponding Landau level filling factor. b, Maximum photocurrent versus carrier density at a series of magnetic fields. c, Two-terminal resistance measured under the same conditions as in b.

single-particle transport. Finally, as the photo-Nernst signal shows much stronger quantum oscillations than the resistivity, it can be used to investigate the quasiparticle properties of materials with short scattering times, as has been done with conventional Nernst measurements on cuprates14 and bismuth35.

**Methods**

Methods and any associated references are available in the online version of the paper.

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Author contributions

X.X. conceived the experiment; G.A. built the experimental set-up; H.C. performed the experiments, assisted by G.A.; H.C., Z.F. and J.R. fabricated the devices; H.C., X.X., D.H.C. and Z.F. analysed the results; Z.F. and D.H.C. did the modelling; and H.C., D.H.C. and X.X. wrote the paper with comments from all authors.

Additional information

Supplementary information is available in the online version of this paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to D.H.C. or X.X.

Competing financial interests

The authors declare no competing financial interests.
Methods
Monolayer graphene flakes were prepared by exfoliation onto substrates consisting of 300 nm-thick thermally oxidized SiO$_2$ on highly p-doped silicon, which served as a back gate. Metal (Cr/Au) contacts were patterned by standard e-beam lithography and metallization in an e-beam evaporator. The two-terminal resistance was measured with an a.c. 100 nA current excitation at 20 Hz. For photocurrent measurements, the laser was mechanically chopped at 2 kHz and the current in phase with the chopper was detected.