Excitons are spin integer particles that are predicted to condense into a coherent quantum state at sufficiently low temperature. Here by using photocurrent imaging we report experimental evidence of formation and efficient transport of non-equilibrium excitons in Bi$_{2-x}$Sb$_x$Se$_3$ nanoribbons. The photocurrent distributions are independent of electric field, indicating that photoexcited electrons and holes form excitons. Remarkably, these excitons can transport over hundreds of micrometers along the topological insulator (TI) nanoribbons before recombination at up to 40 K. The macroscopic transport distance, combined with short carrier lifetime obtained from transient photocurrent measurements, indicates an exciton diffusion coefficient at least 36 m$^2$ s$^{-1}$, which corresponds to a mobility of 6 × 10$^4$ m$^2$ V$^{-1}$ s$^{-1}$ at 7 K and is four order of magnitude higher than the value reported for free carriers in TIs. The observation of highly dissipationless exciton transport implies the formation of superfluid-like exciton condensate at the surface of TIs.
A variety of systems, including double quantum wells, microcavities, graphene, and transition metal dichalcogenides, have shown signatures of exciton condensation. Dirac materials such as graphene and topological insulators (TIs) with strong Coulomb attraction and vanishing effective mass emerge as a new promising platform for achieving exciton condensates potentially at room temperature. The gapless TI surface state is protected against backscattering and has a linear dispersion of the TI surface state results in further carrier recombination can be much slower, ranging from a few ps to over 400 ps. This long-lived population inversion allows electrons and holes in the transient state to form pairs. Photoexcited electrons and holes in TIs relax to the surface Dirac cones on sub-picosecond (ps) timescales, while further carrier recombination can be much slower, ranging from a few ps to over 400 ps. This long-lived population inversion allows electrons and holes in the transient state to form pairs (Fig. 1a). Because of the small effective mass, excitons in Dirac materials are expected to have long de Broglie wavelength and high transition temperatures. The figure of merit for exciton formation in materials is $\alpha = \frac{E_s}{E_K}$, where $E_s$ is the Coulomb energy and $E_K$ is the electron kinetic energy. The linear dispersion of the TI surface state results in $\sqrt{\alpha}$. The two-dimensional (2D) surface state of a three-dimensional (3D) TI, with a single non-degenerate Dirac cone, relative low $E_K$ (compared to graphene) and reduced $\epsilon$ at surface, has been theoretically identified as a promising candidate for realizing high-$T_c$ exciton condensations. In addition, the topological nature of the band structure may create exotic spin texture to the excitonic quantum state. The spin-momentum locking demands that the ground state of excitons must be a spin-triplet $p$-wave, which spontaneously breaks time reversal symmetry.

Previous experimental evidence of exciton condensation in gapped semiconductors has been obtained from spatially resolved photoluminescence (PL) measurements, where PL images exhibit macroscopically ordered patterns, or PL peak intensity sharply increases with reduced peak widths at lower temperature. More recently, exciton formation has been experimentally demonstrated in both graphene and TIs. Evidence of superfluidic excitons has also been obtained by quantum Hall drag in bilayer graphene. Photocurrent imaging is a powerful experimental technique that can be applied to visualize the transport of locally photoexcited charge carriers. Compared to spatially resolved PL, it does not require materials to have strong light emission and is hence ideal to study TIs. Previous photocurrent studies of TIs have largely been on degenerately $n$-doped TIs, where photoexcitation is weak with an external quantum efficiency (EQE) of <1% and photocurrent decays rapidly as the local photoexcitation is interrupted. The out-of-channel nanoribbon segment is electric field free and photoexcited carriers are expected to diffuse in this region. Photocurrent distribution in normal semiconductors free of external electric field is understood by the diffusion of minority free carriers. In this model, photocurrent decays exponentially with a characteristic length of $L_d = \sqrt{D_t}$, where $D$ is the diffusion coefficient and $t$ is the lifetime of minority carriers. Transient photocurrent measurements showed $t = 15 \pm 5$ ns in our samples (Supplementary Fig. 6). Limited by the bandwidth of electronics, this value should be treated as an upper limit of the actual lifetime. Taking $t = 20$ ns and $L_d = 0.9$ mm, we estimate a lower limit of mobility $\mu = \frac{eL_d^2}{\hbar k_B T} \approx 6 \times 10^4$ m$^2$/V-s at 7 K. This is 6 orders of magnitude higher than the field-effect mobility determined in our devices ($\mu = 0.037$ m$^2$/V-s), and 4 orders of magnitude higher than the highest reported mobility in 3D TIs ($\mu \sim 1$ m$^2$/V-s). Note that though the electron backscattering at the surface of a 3D TI is forbidden, scattering into other angles is possible, resulting in finite carrier mobility. Therefore, free carrier diffusion does not explain the observation.

Field-independent photocurrent distributions. To understand this unusual behaviour, we performed SPCM as a function of source–drain bias ($V_{SD}$) at 7 K and found that the in-channel photocurrent profiles remain largely independent of $V_{SD}$ (Fig. 2a, c). This is striking as $L_d$ in normal semiconductors is expected to strongly depend on electric field, as experimentally demonstrated previously. Free charge carriers move faster along the electric force, and slower against, leading to longer photocurrent decay near one contact and shorter near the other, as shown in Fig. 2d. Quantitatively, in this free carriers model, $L_d = \frac{2d\mu_0}{V_{SD}}$, where $L_{diff} = \sqrt{D_t}$, $L_{diff} = \mu_0E$ and the signs indicate $L_d$ measured at opposite electrodes. When $E$ increases above a threshold $E_c = \frac{\hbar k_B T}{e d}$, $L_d$ becomes drift dominated and strongly depends on $E$. As shown in Fig. 2b, we applied $E$ up to 20 times of $E_c$, but the measured $L_d$ values remained largely constant. At maximum applied field, the measured $L_d$ value is 20 times lower than that predicted from the free carrier model. This discrepancy from the free carrier model indicates the formation of excitons. The motion of these charge neutral particles is not affected by external electric field, resulting in $E$-independent $L_d$ (Fig. 2b, d). Note that the applied electric field here is still much lower than that...
needed to separate excitons as estimated in Supplementary Note 4. In addition, photoexcitation at low temperature mainly produce excitons, but with a small portion of free carriers presumably because of thermal activation, as evidenced by the small shift of the photocurrent baseline with $V_{\text{SD}}$ (Supplementary Fig. 8).

Effects of Sb doping, gate, excitation wavelength and intensity. Long $L_d$ is only observed in Sb-doped Bi$_2$Se$_3$ samples, in which the Fermi level ($E_F$) is close to the Dirac point evidenced by the ambipolar gate dependence (Fig. 3d) and angle-resolved photoemission spectroscopy (ARPES)\textsuperscript{39}. Micro-ARPES spectra of these nanostructures have demonstrated clear Dirac cones and indicated that the samples are slightly n-doped relative to the Dirac point, but with $E_F$ below the bulk conduction band (Supplementary Fig. 2). $L_d$ in samples with low Sb doping is shorter than that with more Sb (Supplementary Fig. 9). In pure Bi$_2$Se$_3$ that is degenerately n-doped, photocurrent with much lower magnitude is observed solely near the contacts (Fig. 3a). This explains why non-local photocurrent has not been reported in TIs, though photocurrent mapping in TIs has been studied in previous work\textsuperscript{26–28}. Consistent with the doping-dependent photocurrent, we also found that $L_d$ and IQE can be greatly modulated by gate voltage ($V_g$). Photocurrent first increases slightly at negative $V_g$ when $E_F$ is lowered closer to the Dirac point. But as $V_g$ becomes more negative and tunes the TI from n-type to p-type (Fig. 3d), both $L_d$ and IQE drop sharply (Fig. 4a, d).

The photocurrent distributions are also measured as a function of light polarization, light intensity and excitation wavelength. Both circularly and linearly polarized laser beams are applied, but the resulting photocurrent distributions are independent of the polarization due to the normal incidence of the laser used in this work\textsuperscript{28}. $L_d$ is found to decrease at higher laser intensity (Fig. 4b, e) but is independent of excitation wavelength in a wide range of 500–1700 nm (Fig. 4c, f). The latter rules out the possibility of surface plasmon polariton (SPP)\textsuperscript{40} since the SPP propagation length is expected to be wavelength dependent\textsuperscript{41,42}. The normal incidence configuration with light injection from free space also unlikely creates surface plasmon due to momentum mismatch. Surface plasmons are expected to exist at high electron density\textsuperscript{40}, but we only observed long photocurrent decay lengths in intrinsic samples. In addition, the wavelength independent $L_d$ confirms that the second Dirac cone 1.5 eV above the conduction band edge\textsuperscript{43} is not involved in the exciton formation.

Discussion

One possible way to understand the observed highly efficient carrier transport at low temperature is the formation of superfluid-like exciton condensate in TIs. Different from free carriers, which suffer from scattering, excitons are bosons and can condense into a coherent quantum state at low temperature. In this picture, the photoexcitation mainly generate charge carriers in the bulk of the TI material. Then, these photoexcited carriers undergo a fast relaxation process within a few picoseconds and relax to the surface states where recombination is much slower up to hundreds of picoseconds. The electrons and holes at the surface form excitons at sufficiently low temperature. The exciton formation opens many-body energy gaps at the surface states similar to the energy gap associated with Cooper pairs in superconductors\textsuperscript{10,11}. The excitons propagate across the TI surface ballistically over hundreds of micrometres. The flow of excitons does not generate an electrical current because excitons are charge neutral. However, as excitons reach the metal-TI contact, they are separated and create photocurrent. Bi$_2$Se$_3$ makes
Ohmic contact to metals but strong band bending of hundreds of meV at the junction facilitates efficient charge transfer (see band diagram and equivalent circuit model in Supplementary Fig. 3). The high IQE value at low temperature indicates that a large fraction of photoexcited carriers condense in the superfluid state.

This exciton condensate model is consistent with the sensitive dependence of photocurrent distributions on temperature, doping, gate and intensity. Both dependence of photocurrent distributions on temperature, doping and gate effects. The rapid drop of $L_d$ as $V_g$ becomes more negative (Fig. 4a, d) is likely because of faster carrier recombination caused by the mixing of surface states and bulk valence band in $\beta$-type TIs, as the Dirac point is close to the bulk valence band in $\beta$-type TIs. Furthermore, the strong dependence of $L_d$ on $E_g$ indicates that excitons are at the surface of TIs. While the carrier lifetime at the TI surface sensitively depends on $E_g$ and is over 400 ps in intrinsic samples, the lifetime in the bulk is always short in the order of picoseconds. As a result, excitons in the bulk are required to travel at a speed 2 orders of magnitude higher than the Fermi velocity in order to propagate across the 200-μm nanoribbon within this lifetime, which is highly unlikely. Finally, the strong light intensity dependence indicates that stronger screening at high intensity makes excitation formation more difficult. The theoretically estimated $T_c$ as a function of excitation power (Supplementary Note 4) is in good agreement with the experimental observation (Supplementary Figs. 10 and 11).

Different theories suggest that exciton condensates can be either an insulating or a superfluid state.

Experimental signatures of both excitonic insulator and excitonic superfluid have been reported. The observed highly dissipationless transport of photogenerated carriers in TIs provides strong evidence supporting superfluidity. As pointed out in reference, superfluidity in He or superconductors can be distinct from that in exciton condensate, where the former is via mass flow and the later is via energy flow. It is interesting to note that mass flow is not necessary for the observed long photocurrent decay. The energy flow from the photoexcitation point to electrical contact can also result in non-local photocurrent. Furthermore, Fig. 1e shows that $L_d$ decreases gradually when temperature is increased above $T_c$, indicating that the phase transition is not sharp. This can be understood by considering the
Fig. 3 Doping-dependent photocurrent profiles. a, b Photocurrent and optical images, where vertical yellow lines indicate the contacts. Colour scales are current in nanoampere. Laser power is 723 nW. c, d Gate-dependent conductance measured in the dark at 7 K. Insets: band diagrams showing $E_F$ positions. a, c are for pure Bi$_2$Se$_3$ where $E_F$ is close to the conduction band. Field-effect mobility and electron concentration are estimated to be $\mu = 329$ cm$^2$ V$^{-1}$ s$^{-1}$, $n = 3.25 \times 10^{18}$ cm$^{-3}$. The photocurrent is only observed when excitation is close to the contacts. b, d Sb doping lowers $E_F$ as evidenced by ambipolar conduction. $\mu = 371$ cm$^2$ V$^{-1}$ s$^{-1}$, $n = 9.3 \times 10^{16}$ cm$^{-3}$ for electrons. The scale bars correspond to 3 $\mu$m in a and 30 $\mu$m in b.

Fig. 4 Effects of wavelength, gate voltage and laser power on photocurrent decay lengths in Sb-doped Bi$_2$Se$_3$. The dashed lines indicate the contacts. The measurements are carried out at 7 K and zero source–drain and gate biases. a–c 2D photocurrent maps. d–f line cuts along the nanoribbons axis. a, d Gate voltage dependence. Inset, band diagrams showing $E_F$ position. Laser power is 166 nW. b, e Laser power dependence. c, f Wavelength dependence. Laser power from 77 to 280 nW was used for different wavelengths to maintain the same exciton injection rate. The scale bar denotes 30 $\mu$m.
Methods

**Nanoribbon growth and device fabrication.** The CVD growth was carried out in a Lindberg Blue M tube furnace, following similar procedures as in previous work. The system was first evacuated to a base pressure of 30 mTorr and Ar was then injected and a room pressure was maintained. For a typical growth, 116 mg of Bi$_2$Se$_3$ powder (99.999%, Alfa Aesar) was mixed with 20–35 mg of Pb powder (99.999%, Alfa Aesar) and placed in a small quartz tube at the centre of the tube furnace. Se pellets (250 mg) (99.999%, Johnson Matthey Inc.) were placed in another quartz tube upstream by a distance of 16 cm. A silicon substrate was placed 14 cm downstream from the centre of the furnace. Se pellets (250 mg) (99.999%, Johnson Matthey Inc.) were placed in another quartz tube upstream by a distance of 16 cm. A silicon substrate was placed 14 cm downstream from the centre of the furnace. The temperature at the centre of the furnace was 680 °C, the Ar flow rate was 150 sccm (standard cubic centimetres per minute) and the growth time was 5 h. After that, the furnace was cooled down to room temperature over ~3 h. The growth yields both nanoribbons and nanoflakes (Supplementary Fig. 1a). The as-grown nanoribbons were then transferred to 300 nm SiO$_2$ covered Si substrates, where yields both nanoribbons and nanoplates (Supplementary Fig. 1a). The as-grown nanoribbons were then transferred to 300 nm SiO$_2$ covered Si substrates, where yields both nanoribbons and nanoplates (Supplementary Fig. 1a). The as-grown nanoribbons were then transferred to 300 nm SiO$_2$ covered Si substrates, where yields both nanoribbons and nanoplates (Supplementary Fig. 1a).

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Author contributions

D.Y. and Y.H. designed the experiments. Y.H. synthesized Bi2Se3 nanoribbons and performed the measurements. R.X., L.M., H.C.T., J.F. and H.F. assisted the synthesis and measurements. A.R., I.V. and E.R. performed micro-ARPES measurements. R.W., O.E., B.W. and S.S. performed theoretical calculation. All the authors analysed the data. D.Y., Y.H., R.W., O.E. and I.V. co-wrote the paper.

Competing interests

The authors declare no competing interests.

Additional information

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