The quantum limit is quite easy to achieve once the band crossing exists exactly at the Fermi level ($E_F$) in topological semimetals. In multilayered Dirac fermion systems, the density of Dirac fermions on the zeroth Landau levels (LLs) increases in proportion to the magnetic field, resulting in intriguing angle- and field-dependent interlayer tunneling conductivity near the quantum limit. BaGa$_2$ is an example of a multilayered Dirac semimetal with its quasi-2D Dirac cone located at $E_F$, providing a good platform to study its interlayer transport properties. In this paper, we report the negative interlayer magnetoresistance induced by the tunneling of Dirac fermions between the zeroth LLs of neighboring Ga layers in BaGa$_2$. When the field deviates from the c-axis, the interlayer resistivity $\rho_{zz}(\theta)$ increases and finally results in a peak with the applied field perpendicular to the c-axis. These unusual interlayer transport properties are observed together in the Dirac semimetal under ambient pressure and are well explained by the model of tunneling between Dirac fermions in the quantum limit.
The quasi-2D Dirac cone at K with the Dirac point located at $E_F$ is confirmed by the first-principles calculations, angle-dependent dHvA oscillations and angle-resolved photoemission spectroscopy (ARPES) measurements. The above-mentioned NIMR and peak in angle-dependent interlayer resistivity are both observed clearly in BaGa$_2$ for the first time and well explained by the model of Dirac fermion tunneling between the zeroth LLs.

Results

Materials characteristic and in-plane transport properties. BaGa$_2$ single crystals are grown by self-flux method (see the “Methods” section). The crystal structure consists of Ba planes and the Ga honeycomb-net planes which shows 2D characteristic. The (00l) lattice plane and lattice parameters are determined by the X-ray diffraction (XRD) measurements on BaGa$_2$ single crystal and powder as shown in Fig. 1b and c. Both the in-plane resistivity $\rho_{xx}$ and the interlayer resistivity $\rho_{zz}$ show metallic behavior (Fig. 1d). The ratio of $\rho_{zz}/\rho_{xx}$ is about 26 at 2.5 K, indicating a moderate electronic structure anisotropy. The measurements of Hall resistivity $\rho_{xy}$ reveal that the in-plane dominant carrier is hole-like with the carrier concentration and mobility estimated to be $n_h = 4.28 \times 10^{21}$ cm$^{-3}$ and $\mu_h = 3277$ cm$^2$ V$^{-1}$ s$^{-1}$ at 2 K. The MR of BaGa$_2$ reaches 400% and shows no sign of SdH oscillation (Fig. 1e).

dHvA quantum oscillations. dHvA oscillations at various temperatures with the magnetic field along [00l] direction is shown in Fig. 2a. Four fundamental frequencies $F_x = 36.9$ T, $F_y = 56.6$ T, $F_z = 356.0$ T, $F_{xy} = 1862.0$ T are confirmed to exist after fast Fourier transform (FFT) analysis of the oscillatory component $\Delta M$ as shown in Fig. 2b and c.

Fig. 1 Crystal structure and in-plane transport properties of BaGa$_2$. a Crystal structure and schematic of the interlayer tunneling of zeroth LL’s Dirac fermions in BaGa$_2$. The red and blue balls represent Ga and Ba atoms, respectively. b Single crystal X-ray diffraction pattern. Inset: the picture of BaGa$_2$ crystal. c Powder X-ray diffraction pattern with refined lattice parameters $a = 4.43$ Å and $c = 5.08$ Å (SG: P6/mmm). d Temperature dependence of in-plane $\rho_{xx}(T)$ and interlayer $\rho_{zz}(T)$. Inset: field dependence of Hall resistivity $\rho_{xy}$ at 2 K. e In-plane MR at different temperatures.
The dHvA oscillations can be described by the Lifshitz–Kosevich (LK) formula\(^\text{38}\):

\[
\Delta M \propto -B' R_T R_D \sin \left[ 2\pi \left( \frac{F}{B} - \frac{1}{2} + \beta + \delta \right) \right],
\]

where \(R_T = (\Lambda T\mu / B) / \sinh (\Lambda T\mu / B)\), \(R_D = \exp (-\Lambda T\mu / B)\) and \(\lambda = (2\pi^2 k_B T_m / \hbar\vee)\). \(\mu\) is the ratio of effective mass to free electron mass \(m_0\), and \(T_D\) is the Dingle temperature. \(\gamma = 0, \delta = 0\) for a 2D system, and \(\gamma = 1/2, \delta = \pi/8\) for a 3D system. \(\beta = \phi_B/2\pi\) and \(\phi_B\) is the Berry phase. The inset of Fig. 2c displays the temperature-dependent FFT amplitudes and fittings using the thermal factor \(R_T\) in LK formula. The obtained effective cyclotron masses are quite small, comparable with those in topological semimetals, due to the almost linear dispersion though they all originate from trivial bands lately determined by the magnetotransport analysis and first-principles calculations. In order to extract the Berry phase conveniently, we applied four-band LK formula to the almost linear dispersion though they all originate from quite small, comparable with those in topological semimetals, due directly. The dots being experimental results and the red line being the curve. All of the Berry phase of these four pockets greatly deviate from the non-trivial value \(\pi\) implying that these bands may be trivial. Detailed data is exhibited in Table 1. The angle-dependent dHvA oscillation measurements are applied to further reveal the Fermi surface of BaGa\(_2\), as exhibited in Fig. 3a. Figure 3b and c shows the FFT spectra of the corresponding dHvA oscillations with the B rotating from B/c to B/ab. The fundamental frequencies increase with the angle \(\theta\) and vanish at \(\theta = 90^\circ\), indicating the moderate electronic structure anisotropy of these trivial bands. If these trivial bands are highly 2D-like, these frequencies should vanish once the magnetic field rotates by a small angle, which is inconsistent with the experimental measurements that \(F_{xx}, F_{xy}\) and \(F_{yy}\) are still observable at \(\theta = 56^\circ\). Besides, the twofold symmetry of polar plot MR (Fig. 3d) with \(B\) always vertical to \(I\) at 2.5 K and the ratio of \(\rho_{xx}(14\,\text{T}, 0^\circ)/\rho_{xx}(14\,\text{T}, 90^\circ) = 5\) also indicate a moderate electronic structure anisotropy in BaGa\(_2\).

**First-principles calculations.** The band structure and Fermi surfaces of BaGa\(_2\) calculated with spin–orbit coupling (SOC) effect included are shown in Fig. 4. As exhibited in Fig. 4a, the Dirac cone locates exactly at K point (\(E_F\)) in the Brillouin zone (BZ), which is consistent with previous work\(^\text{37}\). In addition, there exist three trivial bands crossing the Fermi level with the corresponding FSs demonstrated in Fig. 4b–d. The hole-type FSs (Fig. 4b and c) are open-orbit FSs along the \(k_z\) direction exhibiting quasi-2D characteristic. Notably, different from the ideal cylindrical Fermi surface the lotus-root shape hole-type FS in Fig. 4b has two evident cross sections indicating the existence

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**Table 1 Parameters extracted from dHvA oscillations.**

| \(\alpha\) | \(F\) (T) | \(T_D\) (K) | \(m'/m_0\) | \(\tau_q\) (ps) | \(\mu_q\) (cm\(^2\) V\(^{-1}\) s\(^{-1}\)) | \(\phi_B\) | \(\psi_B\) | \(\nu_T\) (10\(^6\) ms\(^{-1}\)) | \(k_f\) (Å\(^{-1}\)) | \(E_F\) (eV) |
|---|---|---|---|---|---|---|---|---|---|---|
| 36.9 | 27.3 | 0.06 | 0.045 | 1334 | 1.39\(\pi\) | 0.66 | 0.033 | 0.144 |
| 56.6 | 8.0 | 0.08 | 0.152 | 3424 | 1.73\(\pi\) | 0.62 | 0.041 | 0.168 |
| 356.0 | 5.9 | 0.09 | 0.210 | 4107 | 0.75\(\pi\) | 1.37 | 0.104 | 0.937 |
| 1862.0 | 7.2 | 0.27 | 0.168 | 1179 | 1.37\(\pi\) | 1.02 | 0.238 | 1.597 |

\(F\) is frequency of the dHvA oscillations; \(T_D\) is Dingle temperature; \(m'/m_0\) is the ratio of effective mass to free electron mass; \(\tau_q\) is quantum relaxation time; \(\mu_q\) is quantum mobility; \(\phi_B\) represents Berry phase. \(\eta\), \(k_f\), and \(E_F\) represent Fermi velocity, Fermi vector, and Fermi energy, respectively.
of \( k_z \) dispersion, which is consistent with the angle-dependent dHvA oscillations (Fig. 3). According to the Onsager relation \( F = (\phi/2\pi^2) = (\hbar/2\pi e)A \), the frequency \( F \) is proportional to the extreme cross section \( A \) of FS normal to the magnetic field. The calculated dHvA frequencies from the hole-type Fermi surface Fig. 4b are 61.4 and 430.1 T corresponding to the observed frequencies \( F_{\alpha} \) and \( F_{\beta} \). The calculated frequency from the other hole-type Fermi surface Fig. 4c is 2273.4 T corresponding to the observed frequency \( F_{\gamma} \). The largest calculated frequency from the electron-type Fermi surface (Fig. 4d) is 4608.3 T which is too high to be observed at 2.5 K and 14 T. The observed frequency \( F_{\delta} \) may originate from the electron-type Fermi surface (Fig. 4d) as there is a branch along K–H according to the calculations. Thus, the trivial Berry phases obtained from the fitting are acceptable since they are all from the trivial bands. As for the Dirac band, no frequencies corresponding to the oscillation should be observed since it is located exactly at the Fermi energy (indicated by red circles in Fig. 5c) originating from surface states which is consistent with the first-principles calculations, as shown in Fig. 5d. Besides, the experimental dispersions which are away from Fermi energy show a mixture of calculated dispersions ranging from \( k_z = 0 \) and \( k_z = 0.5\pi \) (the dispersions at the \( k_z \) boundaries are colored black in Fig. 5d), suggesting a very strong \( k_x \)-broadening.

We zoom in the dispersion near \( E_F \) to further reveal the Dirac cone at the K point. The dispersion at \( k_z = 0 \) is shown in Fig. 5f. Due to the matrix element effect, the left branch is much weaker than the right branch. To enhance the visualization of the dispersion on both sides, we show in Fig. 5g the normalized spectra by integrating each energy distribution curve (EDC). In addition to the Dirac cone at the K point, there is an extra dispersion originating from the surface state, which is consistent with the calculations. The evolution of Dirac cone along the \( k_y \) direction is shown in Fig. 5f. Moving away from the K point, the dispersion shifts down in energy and becomes more parabolic, in agreement with a Dirac cone at the K point as shown in Fig. 5a. The \( k_z \)-dependence of the Dirac cone is shown in Fig. 5g. Similar behavior is also observed, with the dispersion shifting down in energy when moving away from K point (\( k_z = 2\pi^*/a \)). However, the \( k_z \) dispersion of the Dirac cone is much weaker than \( k_y \) dispersion and the Fermi velocity along \( k_z \) is almost one order of magnitude less than \( k_y \), suggesting that the Dirac cone is quasi-2D like centered at K point.

**Fig. 3** Angle-dependent dHvA oscillation and in-plane MR of BaGa2. a Angle-dependent dHvA oscillations at 1.8 K. b Corresponding FFT spectra with \( \theta \) varying from 0° to 90°. c FFT frequencies as a function of angle. Solid red line is the fitting curve with equation \( F = F_0/cos \theta \). Error bars are estimated from sample holders and de

**Fig. 4** Calculated band structure and FS topology of BaGa2. a Band structure of BaGa2 along high-symmetry paths of the Brillouin zone calculated with SOC. b and c Two hole-type Fermi surfaces and d one electron-type Fermi surface.

**ARCES measurement.** The electronic structure of BaGa2 is measured by ARCES and the quasi-2D Dirac cone at the K point is revealed. Figure 5a shows a schematic drawing of the 3D BZ and its projected surface BZ onto the (00l) surface measured. The FS is shown in Fig. 5b. The most obvious feature is a nearly circular pocket around the K point. Besides this pocket, there are several much weaker features around the F and \( k_z \) points where several bands cross the Fermi energy. This can be seen more clearly in the ARCES spectra along the high-symmetry directions as shown in Fig. 5c. The experimental spectra show an overall agreement with the calculated dispersions at different \( k_z \) values. There also exist a few extra band dispersions near the Fermi energy (indicated by red circles in Fig. 5c) originating from surface states which is consistent with the first-principles calculations, as shown in Fig. 5d. Besides, the experimental dispersions which are away from Fermi energy show a mixture of calculated dispersions ranging from \( k_z = 0 \) and \( k_z = 0.5\pi \) (the dispersions at the \( k_z \) boundaries are colored black in Fig. 5d), suggesting a very strong \( k_x \)-broadening.
Interlayer transport properties. BaGa₂ provides an ideal platform to study the unusual interlayer transport properties caused by the tunneling of Dirac fermions between the zeroth LLs since the Ga honeycomb-net layer contributes the quasi-2D Dirac cone and the Ba layer between adjacent two Ga honeycomb-net layers provides the barriers.

Figure 6b presents the angle-dependent interlayer resistivity $\rho_{zz}(B, \theta)$ at 2.5 K under different fields. It shows the interlayer resistivity peak at $\theta = 90^\circ$. $\theta$ is the angle between $B$ and $I$ as defined in Fig. 6a. Besides, the interlayer resistivity decreases with the increasing field, resulting in the NIMR at $\theta = 0^\circ$, as shown in Fig. 6b, d and e. The NIMR decreases quickly when temperature changes from 2.5 to 70 K. Both of these anomalous interlayer transport properties can be understood based on the tunneling of Dirac fermions between the zeroth LLs. According to previous works, the interlayer Dirac fermion tunneling conductivity $\sigma_{ll}^{(0)}(B, \theta)$ can be described as

$$\sigma_{ll}^{(0)}(B, \theta) = A|B\cos\theta|\exp\left[-\frac{1}{2}\frac{e^2}{\hbar c}B\sin\theta\right]$$

where $A$ is a field-independent parameter, $\theta$ is the angle between $B$ and $I$, and $c$ is the distance of the adjacent two Ga layers. When the field is applied parallel to $I$ along $c$-axis ($\theta = 0^\circ$), the tunneling conductivity is simplified as $\sigma_{ll}^{(0)}(B, 0^\circ) = A|B|$. In this case, the tunneling conductivity increases proportional to the magnetic field, leading to the NIMR as exhibited in Fig. 6d. When $\theta$ increases from $0^\circ$ to $90^\circ$, the interlayer tunneling conductivity decreases gradually because of the suppression of 2D LL quantization, resulting in the interlayer resistivity peak as shown in Fig. 6b.

In an ideal 2D Dirac system, the interlayer transport is only contributed by the tunneling of Dirac fermions between the zeroth LLs (tunneling channel) and the resistivity exhibits the equivalent peak values at $\theta = 90^\circ$ under different fields. However, as shown in Fig. 6b, the interlayer resistivity peak increases with the increasing field. The inconsistency is reasonable since there also exist trivial bands contributing the positive interlayer MR except for the Dirac band. As discussed in the angle-dependent dHvA oscillations, the results from first-principles calculations, the ratio of $\rho_{xx}(14T, 0^\circ)/\rho_{xx}(14T, 90^\circ) \approx 5$, and $\rho_{xx}/\rho_{zz} \approx 26$, the electronic anisotropy of these trivial bands are analyzed as moderate. Based on these discussion, we assume the trivial bands contribute to the interlayer transport through a momentum relaxation mechanism (i.e. coherent band transport) and describe it with Drude model. Considering both the tunneling mechanism and momentum relaxation mechanism, the total interlayer resistivity is described as

$$\rho_{zz}(B, \theta) = \frac{1}{\sigma_{xx}(B, \theta)} = \frac{1}{\sigma_{ll}^{(0)}(B, \theta) + \sigma_{xx}(B, \theta) + B_0}$$

where $\sigma_{xx}(B, \theta)$ is the conductivity from the coherent trivial bands. As shown in the Supplementary Table 1, all of the trivial bands do not reach the quantum limit, so the conductivity $\sigma_{xx}(B, \theta)$ is taken as $\sigma_0/(1 + k \cdot B_{xy})$ for low field approximation ($\sigma_0$ is the Drude conductivity, $k$ is a constant, $B_{xy} = |B|\sin\theta$). $B_0$ is a fitting parameter. At $\theta = 90^\circ$, the tunneling channel is suppressed $\sigma_{ll}^{(0)}(B, 90^\circ) = 0$ and the interlayer resistivity mainly originates from the trivial bands. In such case, the interlayer resistivity is described as $\rho_{zz}(B, 90^\circ) = 1/\sigma_{xx}(B, 90^\circ) = 1/(\sigma_{ll}^{(0)}(B, 90^\circ) + B_0)$. As shown in Fig. 6e the fitting curve matches well with the experimental data.
from trivial bands to interlayer transport can be described by Drude model. The total interlayer transport properties can be considered as the result of the competition between the tunneling mechanism and momentum relaxation mechanism. When θ is close to 0°, the trivial band-induced positive MR is relatively small because the field-induced scattering is very weak (absence of Lorentz force). However, the density of states (DOS) at Dirac point increases dramatically with the increasing field since the Dirac cone is located exactly at $E_F$. In this case, the tunneling mechanism dominates the interlayer transport, resulting in the NIMR as shown in Fig. 6b, d and e. The angle-dependent and field-dependent unusual interlayer resistivity can be well fitted by Eq. (3), as shown in Fig. 6c and e. When θ increases from 0° to 90°, the tunneling of Dirac fermions between the zeroth LLs is suppressed while the trivial band induced positive MR increases due to the enhancement of the Lorentz force-induced scattering. It is consistent with the observed interlayer MR, where the NIMR vanishes gradually and positive MR increases with θ varying from 0° to 90°. These resistivity curves are well fitted by Eq. (3) as shown in Fig. 6c and e. These anomalous interlayer transport properties induced by the tunneling of Dirac fermions between the zeroth LLs have also been observed in (BEDT-TTF)$_2$I$_3$, where the Dirac cone only exists under pressure$^{34,35}$. YbMnBi$_2$ is another quasi-2D topological material that exhibits the same anomalous interlayer transport properties$^{36}$, in which the NIMR was not observed possibly due to the large positive MR induced by other bands overwhelming the signature of NIMR. To our knowledge, BaGa$_2$ is the only material that exhibits both NIMR and interlayer resistivity peak at ambient pressure.

**Discussions**

To further understand the observed NIMR in BaGa$_2$, we discuss on the possibly different origins of the negative magnetoresistance (NMR) below. BaGa$_2$ is a non-magnetic material, spin flip may not be a suitable reason for the NMR in BaGa$_2$.$^{41}$ Secondly, the NMR has also been observed in topologically trivial materials at high field such as graphite, where the NMR is induced by the ellipsoidal Fermi surfaces approaching the quantum limit$^{42}$. As displayed in the Supplementary Table 1, all of the trivial bands in BaGa$_2$ are not under the quantum limit with the field of 14 T. In addition, the NIMR in BaGa$_2$ appears once a quite small field is applied. Thus, the scenario of trivial bands under quantum limit does not apply in BaGa$_2$. Thirdly, the NMR induced by chiral anomaly has been widely observed in 3D Dirac and Weyl semimetals$^{5,9,12,21,22}$, which usually has the form of $\Delta_{\sigma z} \propto B^2$ or $B$ depending on the Fermi energy$^{43}$. However, the Dirac cone in BaGa$_2$ opens a small energy gap (~10 meV) when SOC is considered and it will not evolve into Weyl cones when time reversal symmetry is broken (applying the magnetic field). So the chiral anomaly is absent in BaGa$_2$ when $I//B$. Besides, the in-plane NMR is not observed when the field is parallel to the current in BaGa$_2$ (Supplementary Fig. 1). Thus the scenario of chiral anomaly induced NMR does not apply either. Fourthly, current jetting can often cause the NMR$^{44}$, which has been explained in detail in ref. 45. Specifically, it is caused by the inhomogeneous spatial distribution of the current in the sample$^{22,45}$. In order to eliminate the current jetting effect, the four-probe AC transport method is applied in the interlayer transport measurement and the current probe with silver paste almost contact fully on the samples’ upper and lower surfaces (Supplementary Fig. 2) to make the current flow cross the sample homogeneously. Besides, the NIMR in BaGa$_2$ decreases quickly with the increasing temperature, inconsistent with the current jetting effect$^{44}$. Finally, the NIMR can arise in high-purity layered metal PdCoO$_2$ with the residual resistivity $\rho_{xx}$ ranging from about 10 to 40 nΩ cm in most single crystals$^{46}$. As for BaGa$_2$, the residual resistivity $\rho_{xx}$ and $\rho_{zz}$ are about 440 nΩ cm and 11.44 µΩ cm which are larger than that of PdCoO$_2$ at least one order of magnitude. So the condition $4\pi >
The crystal growth and magnetotransport measurements. The single crystal BaGa$_2$ was grown by self-flux method. The ingredient ratio with BaGa$_2$ $\approx$ 41:58:3 was put into the alumina crucible and sealed into a quartz ampoule. Then the ampoule was heated to 950 °C and kept for 50 h to make sure the materials have melted and was grown by self-flux method as implemented in the VASP package. The kinetic energy cutoff of the plane-wave basis was set to be 400 eV. A 20 × 20 × 18 k-point mesh was utilized for the BZ sampling and the Fermi surface was broadened smaller than 0.01 eV/Å. Once the equilibrium structures were obtained, the electric transport and magnetic properties were studied by using the maximally localized Wannier functions (MLWF) method. The scanning type-exchange-correlation potential was adopted. The kinetic energy cutoff of the plane-wave basis was set to be 400 eV. A 20 × 20 × 18 k-point mesh was utilized for the BZ sampling and the Fermi surface was broadened smaller than 0.01 eV/Å. Once the equilibrium structures were obtained, the electric transport and magnetic properties were studied by using the maximally localized Wannier functions (MLWF) method. To study the surface states, a 1 × 1 two-dimensional supercell with an BaGa$_2$ slab containing 59 atoms and a 20 Å vacuum was employed to simulate the BaGa$_2$ (001) surface.

Data availability

The authors declare that the data supporting the findings of this study are available within the article and its Supplementary Information. Extra data are available from the corresponding authors upon reasonable request.

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Author contributions

T.-L.X. coordinated the project and designed the experiments. S.X. and Y.-Y.W. synthesized the single crystals of BaGa$_2$, with the assistance of Q.-H.Y., L.-L.S. and Y.S. C.B. and S.Z. performed the ARPES measurements. P.-J.G., K.L. and Z.-Y.L. performed ab initio calculations. S.X. performed the magneto-transport measurements with the assistance of Y.-W. S.X., C.B. and P.-J.G. plotted the figures and analyzed the experimental data. S.X., Y.-W., and T.-L.X. wrote the paper. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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