

Ultra-high surface conductivity in Weyl semimetal NbAs

Cheng Zhang^{1,2#}, Zhuoliang Ni^{1,2#}, Jinglei Zhang^{3#}, Xiang Yuan^{1,2#}, Yanwen Liu^{1,2}, Yichao Zou⁴, Zhiming Liao⁴, Yongping Du⁵, Awadhesh Narayan⁷, Hongming Zhang^{1,2}, Tiancheng Gu^{1,2}, Xuesong Zhu^{1,2}, Li Pi³, Stefano Sanvito⁶, Xiaodong Han^{8,9}, Jin Zou^{4,10}, Yi Shi^{2,11}, Xiangang Wan^{2,5}, Sergey Y. Savrasov¹², Faxian Xiu^{1,2,13*}

¹ State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China

² Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China

³ High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, China

⁴ Materials Engineering, The University of Queensland, Brisbane QLD 4072, Australia

⁵ National Laboratory of Solid State Microstructures, School of Physics, Nanjing University, Nanjing 210093, China

⁶ School of Physics and CRANN Institute, Trinity College, Dublin 2, Ireland

⁷ Materials Theory, ETH Zurich, Wolfgang-Pauli-Strasse 27, CH 8093 Zurich, Switzerland

⁸ Institute of Microstructure and Properties of Advanced Materials, Beijing University of Technology, Beijing 100124, China

⁹ Beijing Key Lab of Microstructure and Property of Advanced Material, Beijing University of Technology, Beijing 100124, China

¹⁰ Centre for Microscopy and Microanalysis, The University of Queensland, Brisbane QLD 4072, Australia

¹¹ School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, China

¹² Department of Physics, University of California, Davis, California 95616, USA

¹³ Institute for Nanoelectronic Devices and Quantum Computing, Fudan University, Shanghai 200433, China

These authors contributed equally to this work

* Correspondence and requests for materials should be addressed to F. X. (E-mail: Faxian@fudan.edu.cn)

Abstract

In two-dimensional (2D) systems high mobility is typically achieved in low-carrier-density semiconductors and semimetals. Increasing the carrier density in these systems strongly enhances the charged impurity and electron-electron scattering, which in turn degrades the mobility. Here, we discover that the surface state of Weyl semimetal NbAs can overcome such a limit and maintain a high mobility even in the presence of a high carrier density. In order to study its surface transport properties, we first develop a new approach to synthesize high-quality nanostructures of Weyl semimetal NbAs with tunable Fermi levels. Owing to their large surface-to-bulk ratio, the 2D surface state exhibits dominant quantum oscillations with multiple large Fermi surfaces that give rise to a high sheet carrier density, even though the bulk Fermi level locates near the Weyl nodes. Combined with the high mobility value, a record-high surface sheet conductance is achieved up to $5\sim 100$ S/ \square . This is far exceeding that of conventional 2D electron gas, quasi-2D metal films, and topological insulator surface states. Corroborated by theory, we attribute the origin of the ultra-high surface conductance to the disorder-tolerant nature of the Fermi arcs. Our results present the first transport evidence for the low-dissipation property of Fermi arcs in NbAs surface states and establish it as an excellent 2D metal with supreme conductivity for both fundamental studies and potential electronic applications.

Electric transport properties of low-dimensional systems have been one of the central topics of condensed matter physics in the last two decades.¹⁻³ Such intense interest is driven by both fundamental study and functional nanoscale device research. Significant advances have been made in low-carrier-density systems like carbon-based low-dimensional structures^{2,4} and quantum wells¹, in which high sample quality and tunability of electronic properties can be easily accessed. In contrast, much less progress has been made in the study of high-carrier-density systems owing to the lack of good low-dimensional metals. Conventional bulk metals suffer from surface roughness and defects when made into the nanoscale while most layered metallic materials are found to be unstable. Doping/Gating low-carrier-density systems into high-carrier-density metals^{1,2,5} is gradually being adopted as a common approach but additional charged impurity scattering is induced, hindering the study of intrinsic properties in low-dimensional metals.

Recently, Weyl semimetals have been discovered as a new group of topological materials.⁶⁻¹² In Weyl semimetals, conduction and valence bands touch at discrete points in the Brillouin zone, forming quasiparticles that can be described by the Weyl equation.^{6,7} The band touching points, or Weyl nodes, can be regarded as monopoles for Berry curvature. By breaking either time-reversal symmetry^{6,13,14} or inversion symmetry⁷, Weyl nodes with opposite chirality can be separated in momentum space. Gapless boundary states then emerge on the surface of Weyl semimetals, manifesting themselves as arc-like states connecting the surface projection of Weyl node pairs.⁶ Many exotic phenomena have been observed in Weyl semimetals such as chiral

anomaly¹⁵⁻¹⁸, Weyl orbits¹⁹⁻²¹, and anomalous Hall effect²²⁻²⁴. Apart from these physical aspects, the intriguing properties of Weyl fermions have also stimulated extensive studies towards the realization of functionalities, such as magnetization switching²⁵ and valley polarization^{26,27}. Apart from the bulk state, the surface Fermi arc is also predicted to host low-dissipation transport^{28,29} with exotic helical spin texture⁷, which may be a good candidate for a clean two-dimensional (2D) metal.

Despite the rapid progress in investigating the physical properties of bulk Weyl semimetals, the research of surface state transport and related device physics has been limited by the lack of an ideal material system with clean band structure and easy integration into nanoscale devices. One important reason is that most Weyl semimetals discovered so far involve heavy transition metals¹¹, which typically show low vapor pressure and are therefore difficult to be grown through conventional thin film synthesis techniques such as molecular beam epitaxy and pulsed laser deposition. Particularly, the widely-studied transition metal pnictide TaAs and its family suffer from the high evaporation temperature of Ta/Nb and the complicated phase diagram of Ta-As compounds. Top-down methods, such as focused ion beam etching, result in Ta/Nb residues owing to the significant difference of the surface binding energy.³⁰ In contrast, transport in layered materials WTe₂/MoTe₂³¹ and TaIrTe₄³² is dominated by trivial carriers rather than Weyl fermions although they can be exfoliated into micro-size. Hence, unlike Dirac semimetals Cd₃As₂ and ZrTe₅, the mesoscopic transport of Weyl semimetals remains largely unresolved.

Here we report the synthesis of NbAs nanostructures by chemical vapor deposition (CVD) technique and investigate the transport properties of the as-grown nanostructures. In order to overcome the high evaporation temperature problem mentioned above, we use metal chlorides (NbCl₅) to react with hydrogen as the source of Nb. As illustrated in Fig. 1a, NbAs is formed by heating NbCl₅ and As vapors to 800~850 °C along with H₂ gas as the reducing agent in a low-pressure CVD system (see Methods for details). A thin gold layer (~15 nm) is pre-coated on the substrate, acting as the catalyst. Fig. 1b is a typical scanning electron microscope (SEM) picture of the obtained NbAs nanobelts and nanowires. Fig. 1c shows a high-resolution transmission electron microscopy (TEM) image, revealing the good crystalline quality of the as-grown NbAs nanobelts. Fig. 1d is the corresponding selected area electron diffraction (SAED) pattern, consistent with the body-centered tetragonal structure of NbAs. By carefully examining the atomic image and the SAED pattern, we can determine the crystal orientation of the nanobelt. The long axis of the nanobelts is along the [100] direction and the out-of-plane direction is [001]. We also carried out energy dispersive spectroscopy (EDS) analysis, as shown in Fig. S1. These NbAs nanostructures present a good stoichiometric ratio with uniform distribution of Nb/As elements. Fig. 1e shows the Raman spectrum of the NbAs nanobelt. Similarly to NbAs bulk crystals, three peaks are resolved, corresponding to the B₁(1), B₁(2) and A₁ modes, respectively. The thickness of the nanobelts and the diameter of the nanowires are usually around several tens to a few hundred nanometers. In this study, we focus on the transport properties of NbAs nanobelts with a well-defined surface profile along the [001] crystal direction. Samples with thicknesses in the range of 100~300

nm are specifically chosen to enhance the surface state transport while preventing the bulk band structure to display quantum confinement effects.

Transport experiments were carried out in a series of NbAs nanobelts. Fig. 2a summarizes the temperature dependence of the resistivity of these nanobelts, along with another set of data from bulk NbAs for comparison. The inset of Fig. 2a shows a typical device fabricated using the as-grown nanobelts. All of them show a metallic behavior with a very small residue resistivity in the range of $0.03\sim 2 \mu\Omega \text{ cm}$ at low temperatures. Interestingly, the overall resistivity value is more than one order of magnitude smaller than that of the bulk counterpart. This is anomalous because the carriers experience more boundary scatterings in nanoscale samples. Normally this will result in a significant increase in resistivity owing to the decrease in mobility. Such behavior is particularly true when it comes to the high-mobility semimetal systems such as Weyl semimetals, where the original mean free path of carriers is very long up to micron range, exceeding the confined geometry. Fig. 2b presents a direct comparison of the resistivity in bulk crystals and nanostructures of three representative systems, namely NbAs, Cd_3As_2 and Bi. All three materials have a semimetallic band structure with a similar carrier density of $\sim 10^{18} \text{ cm}^{-3}$ and a high mobility over $10,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. In contrast to NbAs, the resistivity in Cd_3As_2 and Bi nanostructures is more than two orders of magnitude larger than that of their bulk crystals, as one would normally expect. With a similar thickness of around 100 nm, the sheet resistance of NbAs and Cd_3As_2 is of the order of $\sim 0.01 \Omega$ and $\sim 50 \Omega$, respectively.

In order to determine the origin of the anomalous decrease of resistivity in NbAs nanobelts, we measured the Hall effect and magnetoresistance (MR) as shown in Fig. 2c-e. The change of Hall coefficient from negative to positive suggests a crossover of the dominant carriers from *n*- to *p*- type. Here the Hall coefficient is estimated by performing a linear fit over the entire $\rho_{xy}\text{-}B$ range (Fig. 2e). These samples are obtained from slightly different growth conditions with the deposition temperature varying between $800 \square$ and $850 \square$. Higher deposition temperature tends to yield *p*-type carriers. In fact, the change of carrier type originates from the semimetallic band structure. In previous photoemission experiments, the Fermi level of NbAs was found to be close to the Weyl nodes.³³ Different growth conditions may shift the Fermi level, resulting in the change of the carrier type, as has been observed in TaAs bulk crystals³⁴. A similar transport behavior shown in Fig. 2c suggests that the Fermi level in our NbAs nanobelts is also near the Weyl nodes, a fact that enables an easy switching between the conduction and valence bands. Similar to bulk NbAs, the MR ratio in nanobelts, defined as $R(B)/R(0)-1$, is also very large, on the order of $10^3\sim 10^5$ (Fig. 2d). Fig. 2e plots the inverse of Hall coefficient $1/R_H$ and MR ratio of different samples. The MR ratio is found to achieve maximum value around the *n*-to-*p* transition. At the same time, quantum oscillations can be observed in both the MR and the Hall resistivity with the amplitude becoming stronger in *p*-type samples. Here, the Hall coefficient reveals a much higher carrier concentration in nanobelts than in bulk crystals of the TaAs family^{34,35}, e.g., $8.1\times 10^{15} \text{ cm}^{-2}$ for sample #1 and $\sim 10^{13} \text{ cm}^{-2}$ for bulk carriers for a thickness of 100 nm. According to previous transport

experiments^{36,37}, the Fermi surface of NbAs bulk crystals with (001) surface only shows two sets of oscillations with close frequency around 20 T from the two kinds of Weyl nodes. In contrast, here in sample #5 and #6, in addition to the low-frequency components, a series of high-frequency oscillations are observed (Fig. 2d). They correspond to the emergence of very large Fermi surfaces other than those originating from the Weyl cones. Furthermore, the beating pattern of the high-frequency oscillations indicates the coexistence of multiple orbits. Note that such fast oscillations have never been detected in bulk TaAs and NbAs even up to 90 T.^{37,38} It is well-known that in nanostructures, the surface-state component will be enhanced due to the large surface-to-bulk ratio. Thin films and nanobelts have been widely used in the study of the surface states in topological insulators (TI)^{39,40} and most recently in Dirac semimetals²¹. Therefore, the observed high-frequency oscillations and low resistivity could come from the surface states, which were overwhelmed and thus undetected in previous transport experiments based on bulk crystals.

In order to track the dimensionality of the additional large Fermi surface, we further carried out magneto-transport experiment in high magnetic field for two representative samples, #3 (*n*-type) and #6 (*p*-type). Fig. 3a shows the magnetoresistance of sample #3 with magnetic field tilting from 0° (out of plane) to 90° (in plane). High-frequency oscillations are observed above 10 T and disappear as the field is tilted towards the in-plane direction. The oscillations extracted from the magnetoresistance R_{xx} (0°) are shown in Fig. 3b. The fast oscillations are super-imposed to the slow ones. Figures 3c and 3d present the corresponding fast Fourier transform (FFT) spectra of the oscillation patterns and the angle dependence of the oscillation frequencies, respectively. In both samples, two low-frequency components, marked as F_{B1} and F_{B2} , are observed. Their values and anisotropic properties (Fig. S4) are consistent with previously-reported bulk Fermi surfaces of NbAs crystals^{36,37}. The frequency of F_{B1} and F_{B2} is smaller in sample #3, suggesting that the Fermi level is closer to the Weyl nodes. Four additional frequencies (F_{S1} - F_{S4}) are resolved in the FFT analysis of sample #6 while only two exist in sample #3. By tracking the angle dependence (Fig. 3d), we find that they follow a 2D behavior and disappear when approaching 90°, consistent with the property of surface states. Under the perpendicular magnetic field, the size of cyclotron orbits (F_{S1} - F_{S4}) is in the range of 0.029~0.040 Å⁻² (300~420 T). Compared to previous Fermi surfaces determined from photoemission experiments³³, such orbits may correspond to the Fermi arcs of Weyl nodes W2 close to the Brillouin zone center. However, these orbits form only a small part of the entire Fermi arc contour. The existence of the majority of the surface carriers is still not captured by the quantum oscillations. This point can be proved by making a comparison between the carrier density value calculated from the Hall effect and the quantum oscillations. Taking sample #3 as an example, the total area from two cyclotron orbits is 0.069 Å⁻². Considering a degeneracy of 2 from the mirror symmetry along k_x/k_y direction, it corresponds to a sheet carrier density of 3.5×10^{13} cm⁻², much smaller than the measured Hall carrier density (7.0×10^{14} cm⁻²).

Having established the surface state as the origin of high conductance in NbAs nanobelts, we now go back to re-evaluate the transport properties within the

framework of 2D systems. By comparing with bulk, we know that the sheet carrier density as well as the low conductivity in NbAs nanobelts originate mainly from the surface state. The corresponding sheet conductance (per surface) and carrier mobility at 2 K are in the range of $5\sim 100$ S/ \square and $1.5\sim 35\times 10^4$ cm²V⁻¹s⁻¹, respectively. Note that here we do not include the data of sample #5~6 since the one-carrier approximation may yield large errors in determining the carrier density in these two samples. A better idea of these values can be obtained by comparing them with other common 2D/quasi-2D conductors in a carrier density-mobility scaling plot as shown in Fig. 4. Here we include conventional 3D metals grown as ultra-thin films or nanostructure forms, intrinsic layered materials in the 2D limit, and quantum well systems. To the best of our knowledge, the NbAs surface state provides the highest sheet conductance among all these 2D systems, and it is over two orders of magnitude larger than typical 2D metals and previously-studied topological surface states (generally below 0.1 S/ \square). In comparison to 3D bulk materials, 2D systems tend to show lower mobility owing to various extrinsic and intrinsic effects such as surface roughness, boundary scattering and enhanced localization.¹ High-quality quantum well systems can overcome these mobility limits by confining carriers within two large potential barriers. However, the generated carrier density, typically around 10^{12} cm⁻², is usually low compared to metals, thus limiting the conductance. Although high-carrier-density 2D electron gases have been made using *d*-electron systems like SrTiO₃ ($10^{13}\sim 10^{15}$ cm⁻²)^{41,42}, the mobility is in turn significantly degraded owing to the large charged impurity scattering. Here, the Weyl semimetal surface states can overcome such a trade-off. Physically, we expect that the Fermi-arc surface state is capable of supporting strong surface currents because the corresponding wave functions extend well into the bulk near the Weyl points making them robust against disorder. Their scattering cross sections on impurities or phonons are generally similar to the case of 3D TIs, whose surface states consist of massless 2D Dirac fermions with helical orientation of spins at a circular Fermi surface. This spin-momentum locking in 3D TIs results in the suppression of 180 degrees backscattering from nonmagnetic impurities since states with opposite spin and momentum remain orthogonal. While there is no general spin-momentum locking mechanism for the Fermi-arc states of WSMs, backscattering of electrons from the arc associated with the top surface to the arc associated with the bottom surface is also forbidden due to zero spatial overlap between the corresponding electronic states.

There is, however, another important difference between a circular Fermi surface of a 3D TI and a disconnected Fermi arc of a WSM that originates in phase space availability for the transitions between scattered electrons in transport calculations³⁹. Under applied external field in certain direction α , the matrix element for the electron at state k to scatter to the state k' at the Fermi surface is weighted by the electron velocities pre-factor $(v_{k\alpha}-v_{k'\alpha})$ appearing as a generalization of a famous $(1-\cos\theta_{k-k'})$ for free electrons.^{43,44} For the disconnected arcs, the velocities of the electrons projected onto the direction of the current remain similar before and after scattering, which makes the surface resistivity tremendously reduced for the curved arc in general, and vanishingly small in the limit of a straight arc geometry. This conclusion is virtually

independent of the particular form of the scattering cross sections, but results from the fact that, in the limit of a straight arc, scattering only occurs between states with the same velocity along the direction of the current. Figures 4b-d summarize the scattering processes in a Fermi arc, in a TI surface state, and in a conventional Fermi pocket, respectively. The intra-arc scattering only slightly tilts the momentum direction without strongly affecting the transport lifetime in a Fermi arc as shown in Fig. 4b. In a TI surface state, although the momentum-reversal-type backscattering is strictly prohibited during the spin-independent scattering process, the oblique backward scattering (indicated by the green arrow in Fig. 4c) is still possible.⁴⁵ Finally, in a conventional Fermi pocket, scattering along all directions is allowed. Therefore, we can expect a long transport lifetime from Fermi arcs even in the presence of strong electron-electron scattering. For the real material NbAs studied here, there is a multiplicity of arcs that necessarily leads to inter-arc scattering of the electrons. However, this should not strongly affect the broadening of the Fermi arcs and their transport since the arcs are rather compact in k-space, and such scattering events are rare due to a requirement of a finite momentum transfer corresponding to wave vectors that separate the arcs.

Our recent theoretical work simulating the effect of surface vacancies on the Fermi arcs in an almost identical system, TaAs²⁹, revealed that the arc states acquire a very little broadening up to very high values of surface disorder. This is primarily attributed to a very small strength in hybridization of the surface states with the remaining electrons. This hybridization strength is at the heart of the impurity problem that appears when evaluating the disorder induced self-energies using a Coherent Potential Approximation, a self-consistent theory that we used to investigate the effect of quenched surface vacancies on the Fermi arcs electronic spectral functions. The resulting calculations of surface conductivity using Kubo-Greenwood formalism showed a great insensitivity to disorder, which is interpreted as driven primarily by the disorder-tolerant Fermi-arc electrons. Thus we can expect high carrier mobility from these Fermi arcs since they have a small effective mass and a low scattering rate. Indeed, the estimated carrier mobility (Fig. 4 and Fig. S2d) is comparable to that of the bulk Weyl fermions in TaAs family^{18,35,36}. In contrast, the large curvature of Fermi arcs^{9,10,33} gives rise to exceptional large density of states as we detected here, even when the Fermi level is close to the Weyl nodes. The 24 Weyl nodes in this material result in multiple sets of Fermi arcs widely distributed over the whole Brillouin zone. This unique feature actually helps to minimize the scattering between Fermi arcs and bulk electrons since the projection of bulk Fermi surface on surface state is small. The coexistence of high carrier density and high mobility in NbAs surface state results in large sheet conductance as observed in experiments. In contrast, Cd₃As₂ nanostructures show much smaller sheet conductance of $0.5\sim 10\times 10^{-2} \text{ S}/\square$ owing to the low carrier density (10^{12} cm^{-2}) from the short Fermi arcs.^{21,46}

In conclusion, we successfully synthesized high-quality NbAs nanostructures using a new CVD method and discovered ultra-high conductivity from the surface state. This growth method can be easily extended to other transition metal pnictides

and enables further studies towards the exotic transport properties of Fermi arcs. The record-high conductance of NbAs surface states may also facilitate Weyl-semimetal-based electronic applications such as thermoelectric conversion and supercapacitors.

Methods

Material growth and characterizations

The NbAs nanobelts were grown using NbCl_5 powders and As chunks as the precursor in a horizontal tube furnace with the mixture of argon (95%) and hydrogen (5%) as the carrier gas (Fig. 1a). The SiO_2/Si substrates were deposited with a thin gold layer (~15 nm) to achieve the vapor-liquid-solid growth process, and they were placed in the center of the furnace. During the growth, the temperature was first increased to 800~850 °C in 50 mins, held for 15 mins, and then decreased to 700 °C in 30 mins before naturally cooled down to room temperature. Higher growth temperature tends to achieve *p*-type samples with a larger mobility. The flow rate of the carrier gas was set to 50 sccm and the pressure was fixed at 200 Torr during the growth. After growth, the substrates appear to be silver-grey color by naked eyes. Both nanobelts and nanowires can be found on the substrates by the optical microscope. Increasing the initial gold coating thickness to 40 nm will only result in the growth of nanowires. The morphological, structural, and chemical characteristics of the as-grown NbAs nanostructures were investigated by SEM (JEOL 7800 and 7001) and TEM (FEI F20) equipped with EDS. The largest crystal plane of as-grown NbAs nanobelts is determined to be (001). The Raman spectrum was measured in a home-built Raman system with an excitation wavelength of 532.8 nm. Compared with previous work⁴⁷ on CVD growth of group Vb phosphides, the current method yields single crystals of nanostructures with higher crystalline quality.

Device fabrication and transport measurements

NbAs nanobelts with thickness in the range of 100~300 nm are chosen in this study. The devices used for transport experiments were fabricated by electron beam lithography technique and etched by standard buffered HF solution for ~5 s before depositing electrodes. Magnetotransport with low field regime (<9T) was carried out in a Physical Property Measurement System (Quantum design) with lock-in amplifier. The high field magnetotransport was carried out in High Magnetic Field Laboratory (CAS, China) and National High Magnetic Field Laboratory (USA).

Figure Captions

Figure 1| Growth and characterizations of NbAs nanostructures. (a) Illustration of the CVD growth process of NbAs nanostructures using a horizontal tube furnace. (b) The SEM image of the as-grown NbAs nanobelts and nanowires on SiO_2/Si substrate. The white scale bar is 20 μm . (c) The high-resolution TEM image of NbAs nanobelts. The scale bars in c corresponds to 2 nm. (d) The obtained SAED pattern of NbAs nanobelts with crystal orientation marked. (e) The Raman spectrum of the NbAs nanobelts, in which three phonon modes can be identified.

Figure 2| Transport results of a series of NbAs nanobelts. (a) The temperature dependence of the overall resistivity in NbAs nanobelts. In comparison, the resistivity of bulk NbAs is also displayed, which is over one order of magnitude larger. The inset is an optical image of a typical NbAs nanobelt device used to study the transport properties. The white scale bar is 20 μm . (b) The comparison of bulk and nanostructure resistivity in three representative semimetals, NbAs, Cd_3As_2 and Bi. The thickness of nanostructures is chosen to be around 100 nm. Only NbAs shows anomalous decrease of resistivity in nanostructure form. Part of the data (including NbAs bulk, Cd_3As_2 /Bi bulk and nanostructures) is taken from literature.^{21,36,46,48-51} (c-d) The Hall resistance (c) and relative MR (d) of a series of NbAs nanobelts. The samples are indexed in the order of n -type to p -type transition. (e) The extracted Hall coefficient $1/R_H$ and the MR ratio at 9 T. The MR ratio is found to achieve the maximum value near the n -to- p transition. The Hall coefficient is estimated using a linear fit to describe the overall trend.

Figure 3| Quantum oscillation analysis and the Fermi surface in NbAs nanobelts. (a) Angle dependence of MR in sample #3 at high magnetic field up to 31.5 T. (b) Extracted quantum oscillations in sample #3 and #6. Fast surface quantum oscillations are found to be superimposed on the slow bulk quantum oscillations. (c) The FFT spectra of the quantum oscillations with corresponding frequencies marked. $F_{B1}+F_{B2}$ corresponds to the harmonic frequency of two bulk oscillations F_{B1} and F_{B2} (d) The angle dependence of oscillations frequencies. The two low frequencies correspond to anisotropic 3D Fermi surfaces while the high frequencies are 2D.

Figure 4| The comparison of sheet conductance among various 2D systems and the illustration of scattering mechanisms. (a) Comparison of the carrier density-mobility scaling among various 2D systems. The surface state of NbAs nanobelts is found to host the largest sheet conductivity. Data of other materials are taken from literature with a typical thickness below 20 nm.^{5,21,39,41,42,46,52-63} The effective penetration depth of surface states in topological systems is also in this range. The mobility values of Cu and Pt thin films are estimated from the thin film sheet resistance by assuming the same carrier density as their bulk. The three dashed lines correspond to three sheet conductance values. (b-d) The scattering processes in the Fermi arc (b), TI surface state (c), and conventional Fermi pocket without spin texture (d), respectively. The red, blue and green arrows indicate the spin texture, the momentum directions, and the possible scattering vectors, respectively. The initial and final states of intra-arc scattering gives similar momentum directions. The spin-independent backscattering is strictly forbidden in TI surface states due to the helical spin texture.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

F.X. conceived the ideas and supervised the overall research. Z.N. synthesized NbAs nanostructures with help from C.Z., T.G., H.Z., and X.Z. C.Z. and Z.N. fabricated the devices. C.Z., X.Y., and Y.L. carried out the transport measurements assisted by J.Z. and L.P. X.Y., Y.Z., Z.L., X.H. and J.Z. performed the sample characterizations. C.Z. analysed the transport data. Y.D., X.W., A.N., S.S. and S.Y.S. provided the theoretical support. C.Z., S.Y.S. and F.X. wrote the paper with helps from all other co-authors.

Competing financial interests

The authors declare no competing financial interests.

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