Negative magnetoresistance in Weyl semimetals NbAs and NbP: Intrinsic chiral anomaly and extrinsic effects

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Chiral anomaly-induced negative magnetoresistance (NMR) has been widely used as critical transport evidence for the existence of Weyl fermions in topological semimetals. In this mini-review, we discuss the general observation of NMR phenomena in non-centrosymmetric NbP and NbAs. We show that NMR can arise from the intrinsic chiral anomaly of Weyl fermions and/or extrinsic effects, such as the superimposition of Hall signals; field-dependent inhomogeneous current flow in the bulk, i.e., current jetting; and weak localization (WL) of coexistent trivial carriers. The WL-controlled NMR is heavily dependent on sample quality and is characterized by a pronounced crossover from positive to negative MR growth at elevated temperatures, resulting from the competition between the phase coherence time and the spin-orbital scattering constant of the bulk trivial pockets. Thus, the correlation between the NMR and the chiral anomaly need to be scrutinized without the support of complimentary techniques. Because of the lifting of spin degeneracy, the spin orientations of Weyl fermions are either parallel or antiparallel to the momentum, which is a unique physical property known as helicity. The conservation of helicity provides strong protection for the transport of Weyl fermions, which can only be effectively scattered by magnetic impurities. Chemical doping with magnetic and non-magnetic impurities is thus more convincing than the NMR method for detecting the existence of Weyl fermions.

Keywords Weyl semimetals, chiral anomaly, negative magnetoresistance, extrinsic effects

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1 Introduction

Quasi-particle excitations of massless Dirac fermions in solids were first proposed in graphene by Wallace in 1947 [1], and the idea of spin non-degenerated Weyl fermions was published even earlier by Weyl in 1929 [2]. However, the experimental exploration of relativistic fermions in solids remained a niche field until the discov-
ery of anomalous quantum Hall effects of Dirac fermions in graphene [3, 4]. The birth of topological insulators [5, 6] has triggered intensive competition in the search for new topological semimetals (TSMs), such as Dirac semimetals (DSMs), Dirac nodal-line semimetals, and Weyl semimetals (WSMs) [7–13]. These materials not only host massless fermions, which are the condensed-matter-physics realizations of the long-sought relativistic fermions in high-energy physics, but also exhibit extraordinary physical properties for potential device applications. In general, three-dimensional (3D) DSMs with four-degenerate Dirac nodes near the Fermi level can lift the spin degeneracy of energy bands and transform into 3D Weyl semimetals, by breaking either time-reversal symmetry (TRS) or inversion symmetry (IS) [14]. The latter has been theoretically predicted [11, 12] and experimentally confirmed [15, 16] in the non-magnetic, IS-broken TaAs family, which is widely considered as a major breakthrough in TSMs. Unlike their magnetic counterpart of pyrochlore iridates [10], WSM states in the TaAs family can be directly detected by angle-resolved photoemission spectroscopy (ARPES), as evident by the existence of Weyl node pairs and linearly dispersed WSM bands [15–18]. WSMs also host symmetry-protected topological surface states. The bulk Weyl nodes with opposite chirality of $\chi = +1$ and $\chi = -1$ are the source and drain points of Berry flux in the momentum space, respectively. The projections of these paired singularities on any surface must be connected by open Fermi arcs [10, 11], which have also been visualized by ARPES. The transport signatures of WSM states [19–22] have been reported simultaneously with the spectroscopy results. However, WSMs share some common features with DSMs [9, 23] in the transport measurements [24]. For example, NbP shows one of the highest records of extremely large magnetoresistance (XMR), which is quasi-linear and non-saturating up to 30 Tesla [21, 22]. Similar XMR was also reported in DSM of Cd$_3$As$_2$ [9, 23]. By measuring Shubnikov–de Haas (SdH) oscillations, a nontrivial Berry’s phase of $\pi$ is expected for Weyl fermions, but such a quantum geometrical phase is general for all quasi-particles associated with the massless linear spectrum [25]. In the ultra-quantum regime of strong magnetic field and very low temperature, which was first considered by Nielsen and Ninomiya, two Weyl nodes with opposite chirality can exchange particles when the Fermi level lies within the zeroth Landau level. This effect, which is well known as the Adler–Bell–Jackiw anomaly or the chiral anomaly [26], manifests as negative magnetoresistance (NMR) when the external field is collinear with the applied electric field ($B//E$), as illustrated in Fig. 1(a).

However, anomalous NMR phenomena have been widely observed in the transport measurements of DSMs, such as Cd$_3$As$_2$ [9], Na$_3$Bi [27], Bi$_{1-x}$Sb$_x$ [28], and ZrTe$_5$ [29], along with the transport reports of chiral anomaly in the TaAs-family WSMs [20, 22, 30, 31]. This raises the question of whether such NMR arises from the intrinsic chiral anomaly or extrinsic factors. One attempt to reconcile the discrepancy between the theoretical prediction and experimental observations was made by Kim et al. who proposed that DSMs may transform into WSMs because of the lifting of spin degeneracy by the external magnetic field [28], as illustrated in Fig. 1(b). A similar idea has also been adopted by Hirschberger...
et al. to explain the possible chiral transport in half-Heusler GdPtBi, which is a zero-gap semiconductor with quadratic bands accidentally touching at the \( \Gamma \) point [32].

In real systems, the intrinsic Fermi levels of TaAs-family WSMs are not located exactly at the energy of the chiral nodes. The resulting WSM Fermi surfaces are ellipsoids enclosing the chiral nodes. In such a semiclassical regime, NMR can also arise from the non-zero Berry’s curvature, which is satisfied by both DSMs and WSMs [33–35]. The resulting NMR is expected to be quadratic in the weak-field limit [34]. In contrast, the experimental observations of NMR in DSMs and WSMs often extend to the high-field regime, and non-saturating behavior has been reported by different groups [30, 32]. This unusual field dependence, which can surprisingly persist above 100 K, strongly suggests that the current flow in these measured samples is inhomogeneous in the collinear configuration of \( B \) and \( E \), a geometrical artifact known as current jetting [36, 37]. Using spot welding to make point-like contacts, dos Reis et al. showed that current jetting may play a dominant role in the NMR of the TaAs family [38]. The authors claimed that with \( B/E \), the field-dependent resistivity anisotropy \( \rho_{zz}/\rho_{xx} \), which is the ratio of the resistivity (\( \rho \)) values perpendicular and parallel to the field direction, can explain all the NMR characteristics reported in the literature [38]. Conversely, Zhang et al. suggested that such field-dependent inhomogeneous current distribution can be effectively suppressed by using long and thin bar-shaped samples with electrodes fully crossing the sample width; thus, the intrinsic chiral anomaly-induced NMR can be probed in the low-field regime [39]. However, analytical modeling of the intrinsic chiral anomaly NMR becomes formidable when the specific defect structures of individual samples must be considered. Transmission electron microscope (TEM) has recently revealed that defects are predominantly high-density stacking faults in TaAs, but a mixture of stacking faults, vacancies, and anti-sites in TaP [38].

In this mini-review, we overview our recent studies on the anomalous MR in NbAs and NbP, with a focus on the physical origins of the NMR phenomena with \( B \) parallel to \( E \). We elucidate the critical roles of the weak antilocalization (WAL) and weak localization (WL) of bulk trivial carriers in NbAs and NbP, which dominate the anomalous MR in low-quality crystals and at elevated temperatures above 50 K. Unlike massless Weyl fermions, which always produce a steep positive MR in low fields when a Berry’s phase of \( \pi \) is accumulated for time-reserved scattering paths, the quantum correction of the trivial pockets is not only field-dependent but also temperature-dependent. Consequently, the trivial-pocket controlled MR is characterized by pronounced crossover behavior from low-field positive growth to NMR, as a result of the competition between the WAL and WL effects. Surprisingly, such crossover behavior is also distinctive above 50 K in high-quality NbP, which has unprecedented charge-carrier mobility of \( 10^7 \ \text{cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1} \), one order of magnitude higher than the other TaAs-family members. This suggests the importance of understanding the defect structures in these binaries. A systematic understanding of NMR and the chiral anomaly will require the preparation of high-quality thin films of the TaAs family to eliminate the inhomogeneous current distribution and to achieve gate-tunable chiral anomaly. For transport experiments, a comparative chemical-doping study of magnetic and non-magnetic impurities provides a more reliable way than NMR of detecting the existence of WSM states. The helicity protection of Weyl fermions in IS-broken WSM can only be invalidated by magnetic impurities, as indicated by our recently published results.

## Discussion

### 2.1 General NMR characteristics: XMR effect and WAL

We first briefly discuss the band structures of NbP and NbAs, which are noticeably different from the prototype TaAs. In NbP, there exist eight large trivial hole pockets, forming four pairs of inner and outer Fermi surfaces along the \( S-Z \) symmetry line [21, 22]. This trivial hole population is compensated by four n-type Weyl fermion pockets in the \( k_z = 0 \) plane near the high-symmetry points \( \Sigma \), with each such WSM FS enclosing a trivial electron pocket [22]. In NbAs, compared with NbP, the band top of the trivial hole pockets is closer to the Fermi energy, which effectively reduces the charge-carrier concentration in the binary (Fig. 2). This change mainly comes from the hybridization of the As-4p and Nb-4d orbitals, while the spin-orbital coupling (SOC) magnitude in both systems is comparable with the same transition metal Nb cations.

Due to the broken IS, the energy bands of NbP and NbAs are spin-lifted. Thus, all the charge carriers in these two compounds are spin-polarized, similar to the case in noncentrosymmetric WTe\(_2\) [41]. For WSM pockets enclosing chiral nodes, the spin orientation is either parallel or anti-parallel to the momentum, i.e., helicity, which is rooted in the massless Hamiltonian of WSMs: \( \hat{H} = i\hbar v_F \mathbf{\sigma} \cdot \mathbf{K} \). Helicity provides extremely strong protection for the transport of Weyl fermions against the scattering of non-magnetic defects and leads to a spectacular charge-carrier mobility of \( 1 \times 10^7 \ \text{cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1} \) at 1.5 K in NbP with a moderate residual-resistivity ratio (RRR) of \( \sim 100 \) [22]. For NbAs, we did not observe comparable mobility, which is about \( \sim 3 \times 10^5 \ \text{cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1} \).
The energy band structures of NbP (a) and NbAs (b). The change from P-3p to As-4p orbitals effectively reduces the trivial hole-pocket size and the overall charge carrier density.

for high-quality crystals with RRR of ~ 100. Distinctively, the anomalous MR of the NbAs samples with $B//E$ shows a steep MR upturn in low magnetic field, which is generally observed for TaAs [39] and TaP [30] at the helium temperature but nearly indistinguishable in NbP [22].

One possible contribution of this low-field MR hump is the XMR effect induced by a small misaligned angle between $B$ and $E$. In our experiments, we adopted a different angle-rotation configuration from that in the literature, as shown in Fig. 3, in which $E$ is along the $b$-axis and $B$ is rotated in the $a$-$b$ plane. The zero degree is defined as the minimum point of the averaged MR versus angle curve for $B = \pm 3$ T, respectively, measured by a rotation step of $0.5^\circ$. As shown in Fig. 3, at the presumptive $0^\circ$, the anomalous MR in NbAs is characterized by a low-field resistance hump, followed by the intermediate-field NMR growth. Above 3 T, the MR curve resumes slow positive growth, an indication of the good angle alignment by our method. Above 0.2 T, the dwindling of the XMR contribution to the anomalous NMR in NbAs is evident when $\theta$ changes from $-4.2^\circ$ to $-0.2^\circ$. The nominal $0^\circ$ MR curve surprisingly shows a weaker field dependence in NMR growth, which deviates from

It is intriguing that the steep MR upturn is also sample quality dependent. Figure 4 shows the angle-dependent MR curves in the vicinity of $0^\circ$ for a high-quality sample with RRR exceeding 130 (NbAs-S12), in contrast to RRR $\approx 8$ for the sample shown in Fig. 3 (NbAs-S4). Despite that the overall MR characteristics are highly sensitive to the rotation angle ($\theta$), the low-field parts are nearly identical between $\pm 0.2$ T. Above 0.2 T, the dwindling of the XMR contribution to the anomalous NMR in NbAs is evident when $\theta$ changes from $-4.2^\circ$ to $-0.2^\circ$. The nominal $0^\circ$ MR curve surprisingly shows a weaker field dependence in NMR growth, which deviates from

Fig. 3 (a) Schematic of quantum interference induced by time reversed scattering paths. (b) Angle-dependent anomalous MR in low-quality NbAs-S4. The pink dashed line is calculated from the transverse MR at $90^\circ$, by assuming a large $B$ and $E$ misalignment of $5^\circ$. Inset: Sample geometry: Single crystals of NbAs and NbP are polished into long and thin bar-shaped slabs with current contacts fully covering both long ends. The voltage leads are not point contacts on the top surface of the slab, but crossing the whole side surface along the $c$-axis to minimize the current jetting effect. Note that $E$ and $B$ are in the same plane during the angle rotation, and $a$- and $b$-axis are equivalent due to the lattice symmetry.

Note that the experimental low-field positive MR grows faster than the parabolic-like curve, which demonstrates that the low-field MR cannot be simply explained by the coupling of the XMR effect.
1/B2 and extends to substantially higher fields compared with the negative angles. Further increase in θ gradually weakens the field dependence of NMR, which becomes non-saturating at 1.3°, 1.8°, and 2.8°. At angles larger than 3.3°, NMR is completely absent, and positive MR growth dominates above 0.2 T.

Although these experimental results can be interpreted in various ways, as proposed in the literature [30, 38, 39], the θ-independent growth suggests that the low-field MR is not dominated by the XMR effect or current jetting, both are sensitive to the alignment of B and E. With the presence of massless Weyl fermions and spin-polarized trivial carriers, quantum interference-induced MR correction is expected. Such a quantum effect is illustrated in Fig. 3(a), which shows a closed scattering trajectory (the solid line) and its time-reversed path (the dotted line). For Weyl fermions, backscattering along these two time-reversed paths is equivalent to a closed circle movement in the momentum space, which leads to a Berry’s phase of π [42]. Consequently, destructive quantum interference, i.e., WAL, is typical for WSM pockets at low temperatures. In contrast, the conservation of the spin-momentum geometry of trivial carriers, which is not well-defined quantum numbers, is determined by the nature of individual scattering events. The resulting quantum correction is a pronounced competition between WAL and WL, which is determined by the spin-orbital scattering time (τSO) and the phase decoherence time (τϕ), respectively. When non-magnetic impurity scattering is dominant, i.e., a very small τϕ, the quantum interference is always in-phase and constructive, producing WL correction to MR [43]. On the other hand, if τϕ ≫ τSO, the quantum correction is antiphase, and WAL dominates. In the intermediate regime of τϕ ≈ τSO, there can be a field-dependent transition from WAL to WL [44].

In both the τϕ ≫ τSO and τSO ≫ τϕ regimes, the quantum correction to the conductivity with collinear B and E can be formulated by [45]

\[
\Delta\sigma(B) \equiv \frac{-e^2}{2\pi^2}\ln\left(1 + \beta \frac{e^2d^2}{\hbar B_F^2}B^2\right),
\]

in which \(\hbar\) is the Planck’s constant, \(e\) is the electron charge, \(d\) is the sample thickness, \(B_F\) is the critical field required to dephase the quantum interference, and \(\beta\) is a fitting parameter. The difference between WAL and WL lies in the coefficient \(\alpha\), which is 1/2 for the former and −1 for the latter. For practical purposes, the low-field WAL correction can be simplified as \(\sigma_2 + \alpha\sqrt{B}\) [28, 46]. By assuming a parabolic chiral magnetic conductivity of \(C_WB^2\) [28, 34], we fit the experimental anomalous MR using an empirical model:

\[
\rho(B) = \frac{1}{\sigma_{\text{WL}}} \cdot (1 + C_WB^2) + A_1B^2 + A_2B,
\]

where \(\sigma_0\) is the zero-field conductivity, \(A_1B^2\) is the XMR contribution due to θ misalignment, and \(A_2B\) is a linear correction to the resistivity, which includes the contributions of large Hall signals in NbAs or current jetting. The validity of Eq. (2) is justified by the good alignment between B and E, which ensures that the XMR contribution is negligible under weak fields. Indeed, the model captures all the essential features of the anomalous MR, including the low-field MR hump below 0.2 T, followed by the parabolic NMR, and the positive XMR growth above 2 T, as shown in Fig. 5 for the −4.2° data in Fig. 4. However, for 0° and a positive θ, Eq. (2) does not fit the experimental results.

**Fig. 4** Angle-dependent anomalous MR in high-quality NbAs-S12, in the vicinity of \(B//E\). The definition of the nominal 0° is explained in the main texts. Distinctively, the steep low-field MR hump is independent of angle rotations.

**Fig. 5** Fitting of the anomalous MR curve at −4.2° for NbAs-S12, using the Eq. (2) in the main texts. The low-field MR follows the \(B^2\) model, while the intermediate-field NMR growth is inversely proportional to \(B^2\). Note that a linear correction from Hall or current-jetting is superimposed on the anomalous MR characteristics.
not fit the NMR growth, which deviates from $1/(C_W B^2)$ significantly.

For WSM associated WAL, it is also expected to be robust at elevated temperatures, although electron-phonon and intravalley electron-electron interactions will weaken the helicity-protection mechanism. Figure 6 shows the $T$-dependent MR at the nominal 0° for NbAs-S12. It is clear that NMR has stronger $T$ dependence than the WAL-dominated low-field MR cusp, which is nearly identical below 20 K. At 50 K, NMR disappears completely, while low-field WAL becomes broader in field dependence and is followed by parabolic-like XMR above 2 T. Above 50 K, the anomalous MR is dominated by the XMR effect, which becomes saturating under strong field, as in the case of conventional semimetals [47, 48].

2.2 Non-saturating NMR: Contribution of WL from trivial pockets

Noticeably, sample quality plays a critical role in the $T$-dependent anomalous MR in NbAs. In Fig. 7, we show the $T$-dependent MR curves for NbAs-S4, which has an RRR ($\sim 8$) one order of magnitude lower than that of NbAs-S12. The low-field positive MR cusp in this sample is largely extended to 0.5 T and shows significant $T$ dependence below 20 K. Unlike the high-quality NbAs-S12, there are broad transitions from positive to negative MR in NbAs-S4. At 50 K, the NMR in NbAs-S4 becomes non-saturating up to 5 T. Further increase in $T$ to 100 K, the low-field positive MR becomes parabolic-like, suggesting its origin in the XMR effect. However, above 3 T, negative MR growth gradually takes over. Li et al. proposed that this unusual crossover feature is correlated to a pronounced field-dependent competition between the WAL and WL of trivial bulk pockets [49]. It is noteworthy that the coexistence of nontrivial and trivial pockets is not only reported for the MPn$_2$ family (M = Nb and Ta; Pn = As and Sb) [49–52], but rather general for topological SMs with strong SOC.

In the regime of $\tau_{SO} \sim \tau_\phi$ [44], such trivial pocket-contributed broad crossover with $B//E$ can be modeled by assuming two independent scattering processes for WAL and WL:

$$\frac{\Delta R}{R_0} = \alpha \left[ \frac{1}{2} \ln(1 + bB^2L_\phi^2) - \frac{3}{2} \ln(1 + bB^2L_{SO}^2) \right],$$

(3)

where $L_{SO} \propto \tau_{SO}$ and $L_\phi \propto \tau_\phi$ are the spin-orbital scattering length and the phase coherent length, respectively. WAL is dominant in low fields when $\tau_\phi > \tau_{SO}$. However, WL gradually takes over in high fields due to a larger coefficient of $3/2$ than the $1/2$ for WAL. More importantly, $\tau_{SO}$ is an intrinsic parameter of SOC in the bulk and is thus weakly dependent on $T$ changes, whereas $\tau_\phi$ is highly sensitive to $T$. Such distinct $T$ dependence between $\tau_{SO}$ and $\tau_\phi$ leads to pronounced $T$-dependent
crossover characteristics. In Fig. 7, we show the data fitting for 50 and 100 K using Eq. (3). At 100 K, the model provides a satisfying description of the field dependence. In contrast, the fitting for the 50 K data is less successful, which implies the contributions of the chiral anomaly or other extrinsic effects to the NMR.

It should be emphasized here that Eq. (3) is a variation of the Dugaev–Khmelnitskii model [53], which in principle requires $d \ll l$. Here, $l$ is the mean free path of trivial carriers, which is several orders of magnitude smaller than the typical sample thickness ($d$) of 200 μm in our experiments. The applicability of Eq. (3) to thick NbAs samples may be correlated to the formation of unique stacking-fault defects in the $a$–$b$ plane, which essentially produces quasi-two dimensional structure of $I4_1/m$ lattice sandwiched by hexagonal lattice of $P6_m2$ [40]. In Fig. 8, we show the typical angle-dispersive X-ray diffraction patterns of a low-quality NbAs single crystal, which clearly shows stacking fault-related streaks. For NbP, highly stoichiometric single crystals can be synthesized, as manifested by the unprecedented charge-carrier mobility of $1 \times 10^7$ cm$^2$·V$^{-1}$·s$^{-1}$ [22]. However, we have also observed broad WAL-WL crossover in NbP above 50 K [22], which implies that stacking-fault defects may be universal for the TaAs family.

2.3 Probing Weyl fermions by magnetic impurities

Because the helicity protection of Weyl fermions in the TaAs class relies on intact TRS, comparative chemical doping of non-magnetic and magnetic impurities in this family provides an indispensable way in probing the existence of Weyl fermions. In our previous study [22], we have shown that a minimal amount of chromium ($\sim 1\%$) in NbP degrades the charge-carrier mobility by more than two orders of magnitude, while three times higher concentration of non-magnetic zinc yields comparable mobility to pristine NbP (see Fig. 9). Compared with the $B//E$ NMR method, which may be completely dominated by extrinsic effects from multiple sources instead of the chiral anomaly, chemical-doping experiments provide effective transport evidence of the existence of WSM states. Equally importantly, the interaction of magnetic impurities with Weyl fermions at low temperatures may lead to novel Kondo physics of massless fermions [54], which has not been experimentally explored.

3 Conclusion

The discovery of WSM states in the IS-broken TaAs class has been a major breakthrough in condensed matter physics. The experimental observations of NMR have been extensively used as a hallmark of WSM states in TaAs and the other three binaries. However, the recent experiments have raised the question of whether NMR can be used as unambiguous transport evidence for the existence of Weyl fermions. Here, we have discussed the general observation of anomalous MR in NbP and NbAs in the configuration of collinear $B$ and $E$, which is a prerequisite for the chiral anomaly. We have elucidated that the low-field steep positive MR is WAL correlated, but not necessary to be associated with WSM states, whereas the following intermediate-field NMR may arise from the intrinsic chiral anomaly and/or various extrinsic effects. In particular, we highlight the WL contribution...
to NMR from trivial pockets in NbAs and NbP, which coexist with WSM states. The trivial pocket-controlled WL is sensitive to crystal quality, and low-RRR samples exhibit more pronounced extrinsic NMR, normally characterized by broad positive-to-negative MR transition behavior and weaker field dependence than the theoretical parabolic magnetoconductance of the chiral anomaly. At elevated temperatures, the WL-contributed NMR becomes predominant and shows non-saturating behavior in field growth, which is expected to be general for topological SMs with strong SOC and coexisting nontrivial and trivial pockets.

Chemical doping of non-magnetic and magnetic impurities provides a more reliable way to probe the existence of WSM states than other methods by detecting the helicity protection mechanism of Weyl fermions. Note that such a method is also valid for other relativistic fermions with spin-momentum locking, such as helical Dirac fermions in Rashiba SMs and TIs. A complete understanding of chiral anomaly induced NMR would be feasible by preparing the TaAs class in thin films using methods such as chemical vapor deposition and pulse laser deposition. With thin-film samples, it will be possible to introduce gate tunability to study Fermi energy-dependent chiral anomaly in this family, which would allow the ultra quantum-limit chiral magnetoconductance to be studied for the first time.

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