Anisotropic magnetotransport and extremely large magnetoresistance in NbAs$_2$ single crystals

G. Peramaiyan$^1$, Raman Sankar$^{1,2}$, I. Panneer Muthuselvam$^{1,2,3}$ & Wei-Li Lee$^1$

We report the extremely large magnetoresistance and anisotropic magnetoresistance in a non-magnetic semimetallic NbAs$_2$ single crystal. Unsaturated transverse XMR with quadratic field dependence has been observed to be $-3 \times 10^5$ % at 2 K and 15 T. Up to 12.5 K, clear Shubnikov de Haas (SdH) quantum oscillations were observed from which two distinct Fermi pockets were identified. The corresponding quantum electronic parameters such as effective cyclotron mass and Dingle temperature were obtained using Lifshitz-Kosevich formula. From the field dependent Hall resistivity at 2 K, carrier concentrations $n_e (n_h) = 6.7691 \times 10^{25}$ m$^{-3}$ and mobilities $\mu_e (\mu_h) = 5.6676 \times (7.6947 \times 10^7)$ m$^2$ V$^{-1}$ s$^{-1}$ for electrons (e) and holes (h) were extracted using semiclassical two-band model fitting. We observed large anisotropic magnetoresistance about 84%, 75%, and 12% at 0.75 T and 6 K for three different orientations $\gamma$, $\theta$ and $\phi$, respectively, similar to that in several topological semimetallic systems. Magnetic properties of NbAs$_2$ are similar to the case of graphite, without any phase transition in the temperature range from 5 K to 300 K.

Exploration of novel states of quantum matter with exotic physical phenomena is one of the new frontiers in condensed matter physics. Unusual transport properties such as large magnetoresistance (MR) not only provide signatures of unique states of matter but also play a vital role in device applications such as magnetic field sensors, random access memories, hard drives, spintronic devices, etc$^{1-3}$. The unsaturated large magnetoresistance with quadratic field ($H^2$) dependence, transverse and longitudinal linear magnetoresistance in nonmagnetic semimetals are unusual phenomena, and its origin is under debate in condensed matter physics. In some semimetals such as NbSb$_2$$^4$, LaSb$_5$ and LaBi$_6$, the origin of large unsaturated MR with $H^2$ is attributed to the electron-hole compensation. On the other hand, the electron-hole compensation with $H^2$ of MR and linear MR at intense high fields are observed in the topological semimetals such as a Dirac semimetal, ZrSiS$_7$ and a Weyl semimetal TaAs$_8$, but its origin is different from those aforementioned materials$^8$. Among the family of nonmagnetic semimetallic systems, NbAs$_2$ crystallizes in monoclinic with inversion center (C12/m1)$^{10}$, and is demonstrated to exhibit large MR with $H^2$ dependence$^{10-13}$. First principle calculations revealed that the NbAs$_2$ system possesses four types of Fermi surfaces$^{11}$. It is reported that the NbAs$_2$ single crystal shows unsaturated large transverse MR about 8000 at 9 T and 1.8 K, 8800 at 9 T and 2 K$^{12}$, 1000 at 14 T and 2.5 K$^{13}$ and ultra-high mobility of the order of $10^4$−$10^6$ cm$^2$ V$^{-1}$ s$^{-1}$, and its origin is attributed to electron-hole compensation. The field induced XMR with metal-insulator-like cross-over behavior followed by a resistivity plateau has been observed in a nonmagnetic semimetallic system NbAs$_2$,$^{12,13}$ where nontrivial Berry phase$^{11}$ and negative longitudinal MR$^{13}$ have also been observed. However, the detailed angle dependent magnetoresistance study will help to understand the anisotropic properties of NbAs$_2$, which has not been fully studied. In this work, we report a systematic study of anisotropic magnetoresistance (AMR) in NbAs$_2$ crystal. Large AMR in NbAs$_2$ may be linked to the non-trivial Berry phase of topological systems. High magnetic field transport measurement in the $I-H$ geometry shows the large unsaturated parabolic MR. The results of fitting with a semiclassical two-band model reveal electron-hole compensation with temperature dependent mobility in NbAs$_2$.

$^1$Institute of Physics, Academia Sinica, Taipei, 10617, Taiwan. $^2$Center for Condensed Matter Sciences, National Taiwan University, Taipei, 10617, Taiwan. $^3$Department of Materials Science, Central University of Tamil Nadu, Neelakudi, Thiruvurur, 610005, Tamil Nadu, India. Correspondence and requests for materials should be addressed to R.S. (email: sankarndf@gmail.com)
Results and Discussion

NbAs$_2$ crystallizes in a monoclinic system with the centrosymmetric space group of C12/m1. It belongs to a larger family of transition metal dipnictides MPn$_2$ (M = V, Nb, Ta, Cr, Mo, and W, Pn = P, As and Sb), which is found to crystallize in OsGe$_2$ structure type. In the NbAs$_2$ crystal structure (as shown in Fig. 1(a) and its inset), each Nb (Nb1) atom is bounded by six As (As1, As2) atoms and two As atoms lie outside the rectangular faces. Figure 1(b) shows the Rietveld refinement of the X-ray powder diffraction results (Bruker D8) using Cu-K$_\alpha$ radiation for the pulverized crystalline sample of NbAs$_2$. The inset of Fig. 2(a) shows the as-grown single crystals of NbAs$_2$. The refined lattice parameters, $a = 9.3560$ (2) Å, $b = 3.3828$ (1) Å, $c = 7.7966$ (2) Å, and $\beta = 119.440$ (15)°, are in good agreement with those reported in the literature.$^{10}$

Figure 2(a) shows the plot of resistivity as a function of magnetic field at various temperatures measured in the $I \perp H$ configuration. The data obtained by sweeping the magnetic field from 15 T to $-15$ T were then symmetrized using $\rho(H) = (\rho(H) + \rho(-H))/2$. NbAs$_2$ exhibits quite large magnetoresistance (MR) at low temperatures with a strong Shubnikov de Haas (SdH) quantum oscillation and quadratic field dependence as shown in Fig. 2(b). The MR percentage calculated from $[(\rho(H) - \rho(0))/\rho(0)] \times 100\%$, reaches 303,200% at 2 K without any signature of saturation in a field of 15 T. It is observed that the MR of NbAs$_2$ is very sensitive with respect to sample quality, since sample-1 shows MR about 115,200% at 2 K and 9 T with a residual resistivity ratio (RRR = $\rho_{300K}/\rho_{3K}$) of 107.04 (Fig. 1(c)), whereas the MR of sample 2 shows 170,800% with a RRR of about 110.49 as shown in Fig. S1. The RRR of NbAs$_2$ crystals attest to the good metallicity and quality of the grown crystals, which is comparable to that reported for the Weyl semimetal NbP (RRR = 115)$^9$, higher than those of TaAs (RRR = 49)$^9$ and NbAs (RRR = 72)$^{14}$, but lower than less than that previously reported for NbAs$_2$ (RRR = 222, and 317)$^{11,12}$ crystals. The unsaturated MR behavior of NbAs$_2$ is similar to the semimetals WTe$_2$$^{15}$ and NbSb$_2$. The large and unsaturated MR of NbAs$_2$ is higher than that for the semimetals NbSb$_2$ (MR = 1.3 $\times$ 10$^5$% at 2 K and 9 T)$^4$, LaBi (MR = 0.38 $\times$ 10$^5$% at 2 K and 14 T)$^8$, the Dirac semimetals ZrSiS$^7$ (MR = 1.4 $\times$ 10$^5$% at 2 K and 9 T) and Cd$_3$As$_2$ (MR = 1.6 $\times$ 10$^5$% at 2.5 K and 15 T)$^{16}$. It is comparable with that recently reported for NbAs$_2$ (8 $\times$ 10$^5$% at 9 T at

Figure 1. (a) Crystal structure of NbAs$_2$. The inset shows the triangular prism of NbAs$_2$ where a Nb (Nb1) atom is bounded by six As (As1, As2) atoms and two As atoms lie outside the rectangular faces. (b) Powder X-ray diffraction (XRD) pattern and Rietveld refinement results of for pulverized NbAs$_2$ single crystals. The inset shows the as-grown single crystal of NbAs$_2$. (c) shows the temperature dependence of resistivity with metallic profile, RRR about 107.04.
1.8 K\textsuperscript{11}, $1 \times 10^{5}$% at 14 T at 2.5 K\textsuperscript{13}, the topological semimetal LaSb ($9 \times 10^{5}$% at 9 T and 2 K)\textsuperscript{17} and the Weyl semimetal candidates NbP ($8.5 \times 10^{5}$% at 9 T at 1.85 K)\textsuperscript{9} and NbAs (MR = $2.3 \times 10^{5}$% at 9 T and 2 K)\textsuperscript{14}. In order to analyse the SdH quantum oscillations, the second order polynomial smoothed background was subtracted from the field dependent resistivity, $\rho(H)$. Figure 2(c) shows the total oscillatory pattern $\Delta \rho_{xx}$ as a function of inverse magnetic field ($1/\mu_0 H$) at various temperatures for the $I \perp H$ geometry. (d) FFT spectrum of quantum oscillations showing two distinct peaks at $F_\alpha = 266$ T and $F_\beta = 32$ T as well as their harmonics for various temperatures. (e) Temperature dependence of oscillation amplitude at fixed magnetic field for the observed Fermi pocket. Peak A&B represent the positions of resistivity oscillatory amplitudes. The solid line is the L-K fitting, used to extract the effective cyclotron mass and Dingle temperature. (f) It shows the log $|\Delta \rho_{xx}|$ vs $T$ plot at 3 K, and linear fitting yields the Dingle temperature.

Figure 2. (a) Field dependence of resistivity $\rho(H, T)$ of sample-1 along $I \perp H$ at various temperatures showing clear SdH quantum oscillations. The insets show the measurement geometry (left) and six-probe geometry for the simultaneous measurement of $\rho_{xx}$ and $\rho_{xy}$ (right). (b) It shows the quadratic field dependence of magnetoresistance $MR = \{\rho(H) - \rho(0)\}/\rho(0)$ of sample $-1$. (c) Total resistivity oscillatory patterns $\Delta \rho_{xx}$ as a function of inverse magnetic field ($1/\mu_0 H$) at various temperatures for the $I \perp H$ geometry. (d) FFT spectrum of quantum oscillations showing two distinct peaks at $F_\alpha = 266$ T and $F_\beta = 32$ T as well as their harmonics for various temperatures. (e) Temperature dependence of oscillation amplitude at fixed magnetic field for the observed Fermi pocket. Peak A&B represent the positions of resistivity oscillatory amplitudes. The solid line is the L-K fitting, used to extract the effective cyclotron mass and Dingle temperature. (f) It shows the log $|\Delta \rho_{xx}|$ vs $T$ plot at 3 K, and linear fitting yields the Dingle temperature.
estimate the cyclotron effective mass \( (m^*) \) and Dingle temperature \( (T_D) \) as shown in Fig. S2(b). From the Onsager relation \( F = (\phi_F/2\pi^2)A_F \), where the \( A_F \) is the extremal Fermi surface cross-sectional area perpendicular to the field, \( F \) is the frequency of the oscillation, and \( \phi_F \) is the magnetic flux quantum. The Fermi surface cross sections are calculated to be \( 25.3 \times 10^{-3} \) Å⁻² and \( 3.04 \times 10^{-3} \) Å⁻² for 266 T and 32 T, respectively. The total oscillatory pattern \( (\Delta \rho) \) can be expressed based on the Lifshitz-Kosevich (L-K) formalism \( ^{18} \):

\[
\Delta \rho(T, B) = \exp[-|X(T_D, B)|] \frac{X(T, B)}{\sinh[X(T, B)]} \Delta \rho^0
\]

where \( \Delta \rho^0 \) is the oscillatory component without damping, and \( X(T, B) = 2n^2k_BTm^*/\hbar^2. \) Here, \( m^* \) refers to the effective cyclotron mass, and \( T_D \) is the Dingle temperature. The temperature dependence of \( \Delta \rho \) is fitted well with the L-K formula as shown in Fig. 2(e). The fitting results yield the effective cyclotron mass \( m^* = 0.323 \pm 0.0009m_p \) where \( m_p \) is the electron rest mass. Figure 2(f) shows the fitting results of the respective \( \Delta \rho \) for various inverse fields \( (1/\mu_BH) \) at a fixed temperature of 3 K, which yields the Dingle temperature \( T_D^0 = 2.810 \pm 0.004 \) K. From the Dingle temperature, the single particle scattering rate is calculated to be \( \tau = 2\pi\hbar/\Delta \rho = 4.35 \times 10^{-13} \) s. The obtained results of NbAs₂ are consistent with previous studies.\(^{11-13} \)

The anisotropic magnetoresistance (AMR) is measured along three different field orientations of \( \gamma, \theta \) and \( \phi \) at different field strengths from 0.1 T to 0.75 T as shown in Fig. 3(a,c,e). The inset of Fig. 3(b) shows the AMR measurement geometry for the \( \gamma, \theta \) and \( \phi \) orientations. The AMR effect for the \( \gamma \) orientation is presented in Fig. 3(a) as a polar plot, which illustrates the two-fold symmetry with a variation of period \( \pi \). In this configuration, the magnitude of AMR reaches a maximum about 75% at \( \gamma = 10^\circ \) and minimum about 32% at \( \gamma = 100^\circ \) for the field of 0.75 T and temperature of 6 K, and a similar trend continues in the opposite way up to 180°. In the \( \phi \) orientation, the magnitude of AMR shows a minimum about 12% at \( \phi = 0^\circ \) when the current is parallel to \( H \), and maximum about 29.5% at \( \phi = 90^\circ \) when the current is perpendicular to \( H \). The magnitudes of AMR are observed to be 75% and 12% for \( \perp H (\theta = 0^\circ) \) and \( \parallel H (\theta = 90^\circ) \), respectively, in the \( \theta \) orientation. From the field dependent AMR measurements in three different orientations, it is clear that the AMR is positive, and its maximum always appears when the current is perpendicular to \( H \). In order to analyse the power law dependence of MR, a double-logarithmic value between \( H \) and MR was taken for \( \gamma, \theta \) and \( \phi \) orientations as shown in Fig. 3(b,d,f). The linear fitting of these plots yield the different slopes \( (m \) values) at various angles. The slope \( (m) \) varies from 1.23 at \( 0^\circ \) to 1.632 at 90° for \( \gamma \) orientation. In the \( \theta \) orientation, the slopes values are found to be 1.260 at \( \theta = 0^\circ \) (\( \perp H \)) and 1.201 \( \theta = 90^\circ \) \( \parallel H \)). For in-plane orientation \( (\phi) \), \( m \) varies from 1.175 at \( \phi = 0^\circ \) to 1.743 at \( \phi = 90^\circ \). It is noteworthy that the power law dependence of MR is close to 1 particularly at \( \phi = 0^\circ \) and \( \theta = 90^\circ \) with \( \parallel H \) orientation. We also remark that the behavior of AMR with two-fold symmetry for \( \gamma \) and \( \theta \) orientations remains the same regardless of the magnetic field strength up to 0.75 T, whereas, for \( \phi \) orientation, the two-fold symmetry in AMR gradually faded away in low field regime. The response of the charge carriers to the rotating magnetic field of magnitude about 0.75 T for three different orientations is studied as a function of temperature as shown in Fig. 4(a-c). The variation of AMR, \( \Delta \rho = \left[ \rho_{\text{peak}} - \rho_{\text{valley}} \right]/\rho_{\text{valley}} \) with respect to temperature is presented in Fig. 4(d) for three different orientations. From the temperature dependence of magnetoresistance, AMR increases with decreasing temperature, and it is almost saturated at low temperatures. The two-fold symmetry is well pronounced at low temperature (6 K), and it is sustained up to a measured temperature of 150 K.

Since the MR value of conventional metals is usually small in magnitude and saturated at high fields, and the consequences of unsaturated XMR and ultrahigh mobility in nonmagnetic topological semimetals such as Cd₃As₂, TaS₃, etc., is related to Dirac and Weyl fermions (topological surface states and linear band dispersion), the fact that NbAs₂ exhibits unsaturated XMR is extremely important. In order to identify the intrinsic magnetic property of NbAs₂, magnetization measurements as a function magnetic field and temperature were carried as shown in Fig. 5(a,b). The linear field dependence of magnetization in NbAs₂ is similar to that observed for graphite.\(^{19,20} \) Even though a sudden rise of magnetization below 25 K due to small amount of magnetic impurities, there is no significant effect in the AMR behavior of NbAs₂.

In ferromagnetic metals, AMR typically shows the maximum resistivity when the current is parallel to magnetic field due to spin-orbit scattering, and minimum resistivity when the current is perpendicular to magnetic field.\(^{21,22} \) Since the NbAs₂ belongs to nonmagnetic material category, the physical origin of the AMR effect in the present system is thus different from that in magnetic materials.

According to the semiclassical two-band model,\(^{23,24} \) the total conductivity tensor is expressed in the complex form of

\[
\hat{\sigma} = \frac{\sigma_{\parallel}}{1 + i\gamma H} + \frac{\sigma_{\perp}}{1 - i\gamma H}
\]

where the \( n(p) \) and \( \mu_{\parallel}\mu_{\perp} \) are electron (hole) concentration and electron (hole) mobility, respectively; \( e \) is the electron charge and \( H \) is the magnetic field. The total conductivity is then expressed as

\[
\hat{\sigma} = \left[ \frac{n\mu_{\parallel}}{1 + \mu_{\parallel}^2H^2} + i\frac{\mu_{\parallel}p}{1 + \mu_{\parallel}^2H^2} \right] + \left[ \frac{n\mu_{\perp}}{1 + \mu_{\perp}^2H^2} + i\frac{\mu_{\perp}p}{1 + \mu_{\perp}^2H^2} \right]
\]

In equation (2), the Re and Im \( \hat{\sigma} \) equal to \( \sigma_{\parallel}\mu_{\parallel} \) and \( \sigma_{\perp}\mu_{\perp} \), respectively where \( \sigma_{\parallel} = \frac{\rho_{\parallel}}{\rho_{\parallel} + \rho_{\perp}} \) and \( \sigma_{\perp} = \frac{\rho_{\perp}}{\rho_{\parallel} + \rho_{\perp}} \) and \( \rho_{\parallel} \) and \( \rho_{\perp} \) are Hall and transverse resistivities, respectively. The magnetoresistance (MR) then follows
From equation (2) \(n, p, \mu_n, \) and \(\mu_p\) can be obtained by fitting the \(\sigma_{xy}(H)\) data. For a perfect compensated system \((n = p)\), MR follows a quadratic field dependence which is shown in Fig. 2(b). Figure 6(a) shows the Hall resistivity as a function of magnetic field with nonlinear behavior at low temperatures. The expanded view of\(\rho_{xy}\) below 1 T of \(H\) at 2 K is shown in the inset of Fig. 6(a). It is clear that the sign of \(\rho_{xy}\) changes from positive \((\rho_{xy} > 0 )\) to negative at high fields with nonlinear band, suggesting the multiband effect in the NbAs$_2$ system. At 200 K, \(\rho_{xy}\) shows positive value below 8 T of magnetic field as shown in Fig. S1(c) which suggests that holes dominate over electrons in the transport properties. By fitting the \(\sigma_{xy}\) data as shown in Fig. 6(b), carrier concentrations of \(n = 6.7691 \times 10^{25} \text{m}^{-3}\) and \(p = 6.4352 \times 10^{25} \text{m}^{-3}\), and mobilities of \(\mu_p = 7.6947 \text{m}^2 \text{V}^{-1} \text{s}^{-1}\) and \(\mu_n = 7.6947 \text{m}^2 \text{V}^{-1} \text{s}^{-1}\).
$\mu_n = 5.6676 \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 2 K are extracted, as shown in Fig. 6(c). Figure 6(d) shows the ratio of $n$ to $p$ as the function of temperatures, suggesting that these two carriers are almost compensated in the NbAs$_2$ system.

The magnetic field and temperature dependent transport measurements revealed the highly compensated electron and hole pockets, which may be responsible for the observed XMR. Recently, the AMR effect is observed in several nonmagnetic materials such as ZrSiS$_2$, LaBi$_2$, WTe$_2$, etc. For example, the AMR effect with the combination of two and four-fold symmetry and unsaturated MR with electron-hole compensation as well as open orbital of Fermi surface had been reported in a Dirac semimetal ZrSiS$_2$. The large AMR may be regarded as the most prominent signature in transport for the non-zero Berry curvatures in topological systems. The transport features of NbAs$_2$ we observed turn out to be similar to the Dirac semimetal ZrSiS$_2$ and WTe$_2$. Further

Figure 4. (a–c) show the normalized plots of temperature dependence of AMR measured at fixed magnetic field 0.75 T for $\gamma$ (a), $\theta$ (b) and $\phi$ (c) orientations, respectively showing absence of phase transition up to the measured temperature range. (d) It shows the comparative plots of temperature dependence of magnetoresistance for three different orientations.

Figure 5. (a,b) show the $M$-$T$ for 1000 Oe and $M$-$H$ curves for 5 K and 100 K revealing the diamagnetic behavior in NbAs$_2$. 

μ$_n$ = 5.6676 m$^2$ V$^{-1}$ s$^{-1}$ at 2 K are extracted, as shown in Fig. 6(c). Figure 6(d) shows the ratio of $n$ to $p$ as the function of temperatures, suggesting that these two carriers are almost compensated in the NbAs$_2$ system.
theoretical calculations and band structure characterizations are keenly required to reveal the possible nontrivial band topology in NbAs₂.

In summary, the single crystals of NbAs₂ were grown using the chemical vapour transport method. We observed extremely large, unsaturated and anisotropic MR in NbAs₂. Transverse magnetoresistance of NbAs₂ reaches a large value of about 303,200% at 2 K and 15 T, and MR follows a quadratic field dependence, which is in accord with the electron-hole compensation with the n/p ratio of about 1.05 determined from semiclassical two-band model fittings. From the SdH quantum oscillations, two distinct Fermi pockets were identified, and its effective electron mass and Dingle temperature were extracted from the L-K fitting. Interestingly, apparent two-fold symmetry and large magnitude in AMR are observed for three different field orientations, and power law dependence of MR is close to 1 for $I\parallel H$ orientation. The origin of such large AMR effect in a non-magnetic semimetal NbAs₂ may be related to the presence of non-trivial Berry curvature in NbAs₂, where the magnetic contribution to the AMR effect has been excluded based on magnetization measurements.

Experimental Section
Sample preparation. Two step chemical vapor transport processes were used to synthesize and grow single crystals of NbAs₂. A quartz ampoule with a length of 30–40 cm was used for the synthesis and growth. At first, stoichiometric amounts of 5 N purity precursors of Nb and As in a molar ratio of 1:2 were sealed in an evacuated quartz ampoule. The vacuum-sealed quartz ampoule containing the binary mixture was treated at 950 °C for two days and then cooled to room temperature, yielding polycrystalline NbAs₂. Secondly, the polycrystalline powder of NbAs₂ was mixed with I₂ in a weight ratio of 100:1 and vacuum-sealed in a two-zone tube furnace having a thermal gradient of about 950-850 °C within ~40 cm. The resulting NbAs₂ single crystals have shiny surfaces with well-defined crystal facets as shown in the inset of Fig. 1(b).

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Author Contributions
R.S. and W.L.L. designed the study; R.S. synthesized and growth the sample; G.P., W.L.L. and I.P.M. performed transport measurements; all of the authors discussed the results and interpretations; G.P. wrote the manuscript, R.S. and W.L.L. revised the manuscript.

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