Field-induced tricritical phenomenon and magnetic structures in magnetic Weyl semimetal candidate NdAlGe

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Abstract
Non-centrosymmetric NdAlGe is considered to be a candidate for magnetic Weyl semimetal in which the Weyl nodes can be moved by magnetization. Clarification of the magnetic structures and couplings in this system is thus crucial to understand its magnetic topological properties. In this work, we conduct a systematical study of magnetic properties and critical behaviors of single-crystal NdAlGe. Angle-dependent magnetization exhibits strong magnetic anisotropy along the c-axis and absolute isotropy in the ab-plane. The study of critical behavior with $H \parallel c$ gives critical exponents $\beta = 0.236(2)$, $\gamma = 0.920(1)$, and $\delta = 4.966(1)$ at critical temperature $T_c = 5.2(2)$ K. Under the framework of the universality principle, $M(T, H)$ curves are scaled into universality curves using these critical exponents, demonstrating reliability and self-consistency of the obtained exponents. The critical exponents of NdAlGe are close to the theoretical prediction of a tricritical mean-field model, indicating a field-induced tricritical behavior. Based on the scaling analysis, a $H - T$ phase diagram for NdAlGe with $H \parallel c$ is constructed, revealing a ground state with an up-up-down spin configuration. The phase diagram unveils multiple phases including up-up-down domains, up-up-down ordering state, polarized ferromagnetic (PFM), and paramagnetic (PM) phases, with a tricritical point (TCP) located at the intersection $[T_{TCP} = 5.27(1)$ K, $H_{TCP} = 30.1(3)$ kOe] of up-up-down, PFM, and PM phases. The multiple phases and magnetic structures imply a delicate competition and balance between variable interactions and couplings, laying a solid foundation for unveiling topological properties and critical phenomena in this system.

1. Introduction
The interplay and correlation between geometry, charge, and spin at the quantum level result in topological properties that appear as exotic phenomena such as high carrier mobility [1, 2], extremely large positive transverse magnetoresistance [2–4], negative magnetoresistance [5], transport response and chiral anomaly.
show that AFM topological states provide an exciting platform for exploring prominent physics and inversion and time-reversal symmetry are broken [11, 13, 17]. CeAlGe is also considered to be a magnetic type-II Weyl semimetal without AHE due to an antiferromagnetic (AFM) ground state [18]. Recent studies topological phases due to both space-inversion symmetry and time-reversal symmetry broken [14]. PrAlGe, polarized ferromagnetic (PFM), and paramagnetic (PM) ordering phases. Moreover, a field-induced experimental report on its polycrystalline sample [21], and no study on a single crystal so far. Meanwhile, it phases is discovered.

The tricritical phenomenon with a tricritical point (TCP) at the intersection of up-up-down, PFM, and PM ordering phases. Moreover, a field-induced phenomenon with a tricritical point (TCP) at the intersection of up-up-down, PFM, and PM phases is discovered.

2. Experimental methods

Single crystals of NdAlGe were grown via self-flux technique method. High-purity powders of Nd (99.9%), Al (99.999%), and Ge (99.99%) were mixed with a mole ratio of 1 : 1 : 10 under an argon atmosphere, and sealed into a quartz tube. The sealed quartz tube was heated to 1150 °C followed by a slow cooling to 700 °C. Finally, NdAlGe single crystals were obtained by removing the extra Al flux with centrifugation method.

The chemical compositions were checked by the energy dispersive x-ray spectrometer as shown in the supplementary material [22], which give proportions of Nd:Al:Ge = 1 : 0.984 : 1.002. The crystal structure and orientations were determined by the x-ray diffraction (XRD), which was performed on a Rigaku-TTR3 x-ray diffractometer with high intensity graphite monochromatized Cu Kα radiation. The magnetization measurements including the angle-, temperature-, and field-dependent magnetization, were carried out by a Quantum Design vibrating sample magnetometer (SQUID-VSM). The sample was heated to room temperature and held for 2 min, then cooled to the target temperature under zero field to yield the initial isothermal magnetization. A no-overshoot mode was adopted to ensure a precise magnetic field before data acquisition, and the magnetic field was relaxed with an oscillation model. The magnetization under high magnetic field up to 30 T was performed on a water-cooling resistive magnet.

3. Results and discussion

Figure 1(a) presents the crystal structure of NdAlGe, which has a noncentrosymmetric tetragonal structure that belongs to the I41md space group [21]. The XRD patterns of single-crystal NdAlGe are shown in figure 1(b), while the surface morphology of the as-grown single crystal is depicted in the right inset of figure 1(b). The diffraction peaks in the XRD pattern can be indexed by (00l) Miller indices, indicating that the surface of the single crystal is the ab-plane and the c-axis is perpendicular to the surface. The lattice parameter \( c = 14.592 \, \text{Å} \) is calculated from the XRD pattern, which is very close to that of the polycrystalline sample [14.621(4) Å] [21]. The XRD rocking curve shown in the left inset of figure 1(b) has a single peak with a full-width-at-half-maximum \( \Delta \theta = 0.034^\circ \), indicating a high quality of the single crystal used.
Field-induced magnetic transitions. The two steps occur at temperature-dependent reciprocal of the susceptibility curves, indicating a first-order phase transition for $M_c$ increases monotonously as the field increases without noticeable transition. For $\Theta \perp c$, $M_c$ curves increase as the temperature decreases in the high temperature region. However, the FC and ZFC curves consist of a $\lambda$-shape at low temperature, implying an FM-like domain behavior with Curie temperature $T_C \sim 5.2(2)$ K. Furthermore, a thermal hysteresis appears between cooling and warming curves, indicating a first-order phase transition for $H \parallel c$. The inset of figure 2(a) plots the temperature-dependent reciprocal of the susceptibility $[\chi^{-1}(T)]$ for $H \perp c$ and $H \parallel c$, both of which exhibit linear behaviors in the high temperature range. The fitting of $\chi^{-1}(T)$ by the Curie–Weiss law $\chi(T) = C/(T - \Theta_{CW})$ gives $\Theta_{CW} = -6.66(4)$ K for $H \perp c$ and $\Theta_{CW} = 21.76(3)$ K for $H \parallel c$. The negative $\Theta_{CW}$ for $H \perp c$ indicates an AFM coupling whereas the positive one for $H \parallel c$ suggests an FM coupling.

Figure 2(b) depicts the field dependence of isothermal magnetization $[M(H)]$ at various temperatures with $H \perp c$ and $H \parallel c$ up to 30 T, with an inset magnifying $M(H)$ at $T = 1.8$ K in the low field region. For $H \perp c$, $M$ increases monotonously as the field increases without noticeable transition. For $H \parallel c$, on the other hand, $M(H)$ curves exhibit saturation behaviors after two abrupt magnetic steps, corresponding to two field-induced magnetic transitions. The two steps occur at $\sim 1.3$ kOe and $\sim 21$ kOe respectively, as shown in the inset of figure 2(b). The magnetization after the second step approaches the saturation magnetization $M_S$, whereas the value at the first step is $1/3M_S$ exactly [22]. Finally, both $M(H)$ curves for $H \perp c$ and $H \parallel c$ reach the same value up to a high field of $\sim 30$ T.

The magnetic behaviors described above reveal significant anisotropy in single-crystal NdAlGe. Angle-dependent magnetization $[M(\varphi)]$ at specific magnetic fields is performed to unveil the evolution of anisotropic magnetism. Figure 3 depicts a three-dimensional (3D) plot of angle-dependent magnetization $M(\varphi)$ at $T = 2$ K for single-crystal NdAlGe under selected magnetic fields ($H = 0.05$ T, $0.1$ T, $2$ T, and $3$ T). The in-plane $M(\varphi)$ curves are measured with the field rotated within the $ab$-plane and are projected onto the $xy$-plane. The out-of-plane $M(\varphi)$ curves are performed with the field rotated from the $ab$-plane to the $c$-axis and shadowed onto the $yz$-plane. As can be seen, regardless of the direction of applied field, all in-plane $M(\varphi)$ curves display circular shapes, indicating that magnetism within the $ab$-plane is absolutely isotropic. However, the out-of-plane $M(\varphi)$ exhibits dumbbell shapes with the longitudinal axis parallel to the $c$-axis, implying strong magnetization along the $c$-axis.

Owing to the strong anisotropic magnetization along the $c$-axis, the critical behavior of NdAlGe should be investigated to uncover the magnetic interactions and couplings. Figure 4(a) depicts the field-dependent initial magnetization $[IM(H)]$ for $H \parallel c$ with a temperature range from 2 K to 15 K, where all $IM(H)$ curves below $T_C$ exhibit two steps but no transitions above $T_C$. As the temperature rises, the steps become weaker. Figure 4(b) gives the Arrott plot of $M^2$ vs $H/M$ in the high field region based on the mean-field model. According to the theoretical prediction of the Landau mean-field model, the Arrott plot should be made up of a series of parallel straight lines [23]. Unfortunately, the Arrott plot of NdAlGe does not consist of a series of parallel lines, which implies that the mean-field model is invalid for NdAlGe.
Figure 2. (a) Temperature dependence of magnetization $[M(T)]$ using ZFC and FC sequences under $H = 200 \text{ Oe}$ with $H \perp c$ and $H \parallel c$ [inset shows the $\chi^{-1}(T)$ with fitted lines]; (b) field-dependent magnetization $[M(H)]$ at various temperature up to $30 \text{ T}$ [inset magnifies $M(H)$ at $T = 1.8 \text{ K}$ in low field region].

Figure 3. 3D-plot of angle-dependent magnetization $[M(\phi)]$ at $T = 2 \text{ K}$ under various field for single-crystal NdAlGe: in-plane $M(\phi)$ (xy-plane) with $H$ rotated within the ab-plane; out-of-plane $M(\phi)$ (xz-plane) with $H$ rotated from the ab-plane to the c-axis.

In general, $IM(H)$ curves should satisfy the Arrott–Noakes equation of state, which is written as [23]:

$$\frac{H}{M}^{1/\gamma} = \frac{T - T_C}{T_C} + \left(\frac{M}{M_1}\right)^{1/\beta},$$

(1)

where $M_1$ is a constant. According to the Arrott–Noakes equation of state, $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ curves should form a series of parallel straight lines, consisting of the modified Arrott plot (MAP). Considering the 3D nature of the crystal structure, three dimensional theoretical models are used to construct the MAP. Figures 5(a)–(d) plot the MAPs for single-crystal NdAlGe with $H \parallel c$ based on theoretical critical exponents of 3D-Heisenberg model ($\beta = 0.365, \gamma = 1.386$), 3D-XY model ($\beta = 0.346, \gamma = 1.316$), 3D-Ising model.
Figure 4. (a) Field dependence of initial magnetization $|M(H)|$ with $H \parallel c$; (b) Arrott plot of $M^2$ vs $H/M$.

Figure 5. MAPs under different theoretical models: (a) 3D-Heisenberg; (b) 3D-XY; (c) 3D-Ising; and (d) tricritical mean-field. ($\beta = 0.325, \gamma = 1.240$), and tricritical mean-field model ($\beta = 0.25, \gamma = 1.0$). All MAPs exhibit a bunch of quasi-straight lines in the high field region, however, some of which are not parallel to each other. The normalized slope defined as $NS = S(T)/S(T_C)$ are utilized to judge which model is the best one [where $S$ is the slope of a single line of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$]. Figure 6(a) plots the $NS$ of various theoretical models, with the one closest to ‘1’ being the best solution for NdAlGe [24]. According to $NS$, the tricritical mean-field model is the one that deviates the least from ‘1’ of the models considered. Nevertheless, the critical exponents must be precisely fitted.
The scaling hypothesis can also be used to verify the critical exponents. Defining the renormalized

\[
\log(1 - h / M_0) \equiv \epsilon, \quad \epsilon < 0, \quad T > T_C
\]

and reciprocal of initial susceptibility \(\chi_0^{-1}(T)\)

\[
\chi_0^{-1}(T) = (h_0 / M_0) \epsilon^\gamma, \quad \epsilon > 0, \quad T > T_C
\]

where \(M_0\) is spontaneous magnetization, \(\chi_0\) is initial susceptibility, \(h_0 / M_0\) and \(D\) are critical amplitudes. The reduced temperature \(\epsilon = (T - T_C) / T_C\) is used to scale the temperature. The parameters are critical exponents, where \(\beta\) is associated with \(M_0\), \(\gamma\) is responding to \(\chi_0\), and \(\delta\) is correlated with \(T_C\). These critical exponents can uncover spin interactions and couplings, which can be extracted by an iteration method [24]. Figure 6(b) shows the temperature dependence of spontaneous magnetization \([M_0(T)]\) and reciprocal initial susceptibility \([\chi_0^{-1}(T)]\). According to equations (2) and (3), it is obtained that \(\beta = 0.236(2)\) with \(T_C = 5.24(2)\) and \(\gamma = 0.920(1)\) with \(T_C = 5.26(4)\). Meanwhile, \(1 / \delta\) can be acquired from the slope of \(\log(M) vs \log(H)\) in the high field region \((H > H_s)\) using equation (4). The inset of figure 6(b) plots \(IM(H)\) on log–log scale at \(T_C = 5.2\) K, giving \(\delta = 4.966(1)\).

The self-consistency and reliability of the critical exponents should be examined. The Widom scaling law unifies these independently obtained critical exponents as [27, 28]:

\[
\delta = 1 + \frac{\gamma}{\beta}.
\]

It is calculated out that \(\delta = 4.895(4)\) approaching closely the experimentally obtained value \(\delta = 4.966(1)\). The scaling hypothesis can also be used to verify the critical exponents. Defining the renormalized magnetization \(m \equiv \epsilon^{-2}M(H, \epsilon)\) and the renormalized field \(h \equiv H e^{-(\beta + \gamma)}\), the scaling equation in the asymptotic critical region is as follows [26]:

\[
m = f_+(h),
\]

where \(f_+\) are regular functions with \(f_+\) for \(T > T_C\) and \(f_-\) for \(T < T_C\). With the critical exponents, \(MH\) curves are scaled into \(m(h)\), which should form two independent universal branches for \(T > T_C\) and \(T < T_C\) respectively [26]. Figure 6(c) depicts \(m vs h\) in the high field region \((H > H_s)\), where \(m(h)\) curves fulfill the universality principle to collapse onto two independent branches above and below \(T_C\) respectively. The conformity of the Widom scaling law and scaling equation demonstrates that the obtained critical exponents are self-consistent and reliable.

Table 1 lists the critical exponents of NdAlGe, PrAlGe, and those of theoretical models for comparison. The critical exponents of NdAlGe are mostly close to those of a tricritical mean-field model, which is in sharp contrast to the iso-structural PrAlGe of a 2D-Ising-like model [29]. A tricritical phenomenon is known to occur in a system with multiple phases when three phases converge at one point on the phase diagram. For a system with multiple phases, it means that there exist various interactions and coupling.

![Figure 6](image-url)
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Table 1. Critical exponents of NdAlGe, PrAlGe, and different theoretical models.

| Composition | Technique          | Ref.   | $T_C$  | $\beta$       | $\gamma$       | $\delta$       |
|-------------|--------------------|--------|--------|---------------|---------------|---------------|
| NdAlGe      | Experiment         | This work | 5.2    | 0.236(2)      | 0.920(1)      | 4.966(1)      |
| PrAlGe      | Experiment         | [29]    | 16     | 0.136(6)      | 1.801(7)      | 14.184(3)     |
| Tricritical mean-field | Theory          | [36]    | —      | 0.25          | 1.0           | 5.0           |
| 2D-Ising    | Theory             | [37]    | —      | 0.125         | 1.75          | 15            |
| 3D-Ising    | Theory             | [38]    | —      | 0.325         | 1.24          | 4.8           |
| 3D-XY       | Theory             | [38]    | 0.346  | 1.316         | 4.8           |
| 3D-Heisenberg | Theory           | [38]    | —      | 0.365         | 1.386         | 4.8           |
| Mean-field  | Theory             | [38]    | —      | 0.5           | 1.0           | 3.0           |

In addition, due to the delicate competition and balance, the properties can be easily modulated by external devices, such as pressure, field, doping, and other controlling parameters [30, 31]. In the case of NdAlGe, field-induced transitions and multiple phases are suggested to be the origin of the tricritical phenomenon. As a result, constructing a phase diagram is essential to understand the multiple phases of NdAlGe.

A scaling method can be used to generate a $H-T$ phase diagram for the field-induced phase transition [30, 31]. Divergence on the phase boundary is caused by the change in magnetic interactions induced by the phase transition, resulting in turning points on MAP [31–33]. In the high field region, the MAP of $M^1/H$ vs $(H/M)^{1/\gamma}$ shows a series of quasi straight lines, as seen in figure 5(d). Instead, the magnified MAP of $M^1/H$ vs $(H/M)^{1/\gamma}$ in the low field region exhibits more complex behaviors due to the field-induced phase transitions, as displayed on log–log scale in figure 6(d). Several turning points can be found in the MAP in figure 6(d), which are corresponding to the field-induced phase transitions. Extracting the turning points, a detailed $H-T$ phase diagram can be constructed.

The detailed phase diagram around $T_C$ for single-crystal NdAlGe with $H \parallel c$ is shown in figure 7(a), which distinguishes various phases. For more clear clarification of the magnetic structures and transitions, we draw the sketches only for the spin structures, as shown in figure 7(b). The spins do not represent the real space in the crystal, but only correspond to the spin evolution. The magnetic properties of NdAlGe differ significantly from those of PrAlGe with an Ising-like FM along the $c$-axis [15, 29] or CeAlGe with an AFM within the $ab$-plane [16, 34]. According to a recent study, the iso-structure NdAlSi exhibits an magnetic ground state in which the moments of Nd form an up-up-down configuration [35]. In the case of NdAlGe, structure analysis reveals that Nd ions can result in two different spin orientations [21]. Based on the bifurcation of FC and ZFC $M(T)$ curves, it is concluded that NdAlGe has an up-up-down ground state similar to NdAlSi [35]. Furthermore, the steps of $M(H)$ curves firmly support the up-up-down spin configuration. The saturation magnetization is $M_S \approx 2.681(8) \mu_B/(f.u.)$ corresponding to fully polarized up-up-down spin configuration [22]. It is noted that the first magnetic step on $M(H)$ is 0.892(2) $\mu_B/(f.u.)$, which is just 1/$3M_s$ consistent with the up-down configuration. However, NdAlGe exhibits a unique up-up-down domain behavior in low temperature range when $H \parallel c$. Under zero field, a multi-domain state will be formed to reduce the magnetostatic energy. Similarly, degenerate configuration of down-down-up domains can be formed which have equal energy with the up-up-down variants, which is depicted as phase I in figure 7(b). Three typical domains are marked as ‘1’, ‘2’, and ‘3’ in figure 7(b). Within a single domain, NdAlGe exhibits an up-down or a down-down-up state. However, the domains are random along the $c$-axis or its reverse direction, causing offsetting of the macroscopic magnetization under zero field. As the field increases with $H \parallel c$, the up-up-down multi-domains evolve into an up-up-down ordering state that persists from $\sim 1.3$ kOe to $\sim 15$ kOe, as shown as phase II in figure 7(b). As the field further increases, the up-up-down phase is gradually polarized into the PFM state, with the intermediate state of a canted up-up-down phase. The magnetic configuration of the canted up-up-down phase ranges from $\sim 15$ kOe to $\sim 29$ kOe, which is depicted as phase III in figure 7(b). Finally, when $H$ exceeds $\sim 29$ kOe, the magnetic moments are fully polarized into FM, as denoted as phase IV in figure 7(b). It is noted that $M(T)$ curves exhibit a hysteresis, suggesting that the magnetic transition from up-up-down ordering to PM phase is of a first-order type, as symbolled by the solid curve in figure 7(a). In general, a TCP appears at the terminal point of the first-order phase transition boundary line. In the $H$ $\rightarrow$ $T$ phase diagram of single-crystal NdAlGe with $H \parallel c$, it is found that the TCP is located at the intersection of up-up-down, PFM, and PM phases [$T_{TCP} = 5.27(1)$ K, $H_{TCP} = 30.1(3)$ kOe]. On the hand, when $H \perp c$, the multi-domain state is gradually polarized into PFM until to 30 T, as shown in figure 2(b). When $H \perp c$, the spins are polarized to PFM along an absolutely different route compared with $H \parallel c$. Two small inflections are noticed at $H \sim 1.1$ T and 8.4 T (see the supplementary material [22]). The inflection at the lower field may be due to the change of multi-domain to single-domain states, and the other one at the higher field may be owing to the polarization of up-up-down to up-up-up states. However, there is no abrupt magnetic transition for $H \perp c$. 
Figure 7. (a) $H$–$T$ phase diagram of single-crystal NdAlGe with $H \parallel c$ (the uud is short for up-up-down state); (b) variable magnetic configurations under different fields (the spins in magenta are for spin-up, and those in green are for spin-down; three typical domains are marks as '1', '2', and '3').

The field-induced multiple phases in single-crystal NdAlGe indicate complex interactions and couplings in this system. It is demonstrated that magnetic structures and interactions are essential for the formation and evolution of various topological nontrivial phases. Recently, the Fermi pockets and Weyl nodes correlated with the magnetic structures are studied by a combination of neutron diffraction, quantum oscillation measurements and density functional theory in isostructural NdAlSi compound [35]. Incommensurate magnetism has been testified to be mediated by Weyl fermions in NdAlSi, periodicity of which is linked to the nesting vector between two topologically nontrivial Fermi pockets [35]. For different magnetic structures, the position of Weyl nodes can be changed or gap is slightly opened. Generally, the calculated electronic band structure is performed at 0 K based on the magnetic ground state at zero field. In this case, it is not so sensitive to the variation of temperature below $T_C \sim 6$ K, at which Nd moments keep in the up-up-down spin configuration. On the other hand, in PrAlGe, it is also revealed that magnetic fields can be directly connected to Weyl nodes via the Pr magnetisation [15]. Furthermore, the topological magnetism has been discovered in the magnetic Weyl semimetal CeAlGe [39]. In RAlGe ($R$ = light rare earth) family, the formation of Weyl nodes is mainly protected by the non-centro-symmetric structure (space group $I41md$). Therefore, the family members of RAlGe share similar features of electronic band structures. Indeed, the Weyl cones have been experimentally observed by ARPES in nonmagnetic LaAlGe and FM PrAlGe compounds [13, 14]. Compared to the Ising-type FM state of PrAlGe, the moments of Nd$^{3+}$f-electrons in NdAlGe with different magnetic ground state (‘up-up-down’ spin configurations) will tune the positions of Weyl nodes more moderately. In particular, special attention should be paid to the TCP, which is expected to cause exotic topological phases and special critical phenomena.

4. Conclusion

In summary, the magnetism of single-crystal NdAlGe are thoroughly investigated. Angle-dependent magnetization exhibits strong magnetic anisotropy along the $c$-axis and absolute isotropy in the $ab$-plane. Furthermore, two filed-induced magnetic transitions are revealed when $H \parallel c$. The investigation of critical
phenomena for $H \parallel c$ indicates a field-induced tricritical behavior. Following that, a $H-T$ phase diagram for NdAlGe with $H \parallel c$ is constructed, where a ground state with an up-up-down spin configuration is determined. The $H-T$ phase diagram shows multiple phases, including up-up-down domains, up-up-down ordering, PFM, and PM ordering states, in which a TCP is revealed at the intersection of the up-up-down, PFM, and PM phases [$T_{TCP} = 5.27(1)$ K, $H_{TCP} = 30.1(3)$ kOe]. The recognition of the multiple phases and magnetic structure suggests delicate competition and balance between variable interactions and couplings, laying a solid foundation for understanding the topological properties and critical phenomena in this system.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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