CeNiAsO: an antiferromagnetic dense Kondo lattice

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Abstract
A cerium-containing pnictide, CeNiAsO, crystallized in the ZrCuSiAs-type structure, has been investigated by measuring transport and magnetic properties, as well as specific heat. We found that CeNiAsO is an antiferromagnetic dense Kondo lattice metallic compound with Kondo scale \( T_K \sim 15 \text{ K} \) and shows an enhanced Sommerfeld coefficient of \( \gamma_0 \sim 203 \text{ mJ mol K}^{-2} \). While no superconductivity can be observed down to 30 mK, Ce ions exhibit two successive antiferromagnetic (AFM) transitions. We propose that the magnetic moment of the Ce ion could align in the G-type AFM order below the first transition at \( T_{N1} = 9.3 \text{ K} \), and it might be modified into the C-type AFM order below a lower transition at \( T_{N2} = 7.3 \text{ K} \). Our results indicate that the 3d–4f interlayer Kondo interactions play an important role in Ni-based Ce-containing pnictides.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
The discovery of the iron-based arsenide superconductor LaFeAsO\(_{1-x}\)F\(_x\) [1] has triggered enormous enthusiasm in searching for new high transition temperature \( (T_c) \) superconductors in Fe- or Ni-based pnictides [2–6]. \( T_c \) has been increased to above 50 K when La is replaced by other rare earth elements in the so-called 1111-type family [7, 8]. The parent compound of the 1111-type pnictides, LnTPnO (Ln = rare earths, T = transition metals, Pn = As, P), is the prototype of the ZrCuSiAs structure, where the conducting TPh layer is sandwiched by the insulating LnO layer. So far, superconductivity (SC) can be induced in the Fe(Ni)As-based pnictides by many different ways of chemical doping or by applying high pressure.

Among all of these LnTPnO, the members Ln = Ce show extremely interesting electronic properties. CeFeAsO, as a prototype parent compound of an Fe-based superconductor, is an itinerant bad metal. A spin-density-wave(SDW)-like antiferromagnetic (AFM) transition of d electrons was observed around \( \sim 140 \text{ K} \) and the AFM transition of the Ce\(^{3+}\) local moments was observed below 4 K [9]. SC has been induced by electron doping (i.e. F-for-O doping) when the SDW long range order of the Fe 3d electrons is suppressed [3]. On the other hand, CeFePO is a paramagnetic (PM) heavy-fermion (HF) system which is near the ferromagnetic (FM) instability [10, 11]. In comparison, CeOsPO shows the Ce\(^{3+}\) AFM order at 4.5 K while CeRuPO is a rare example of the FM Kondo lattice with \( T_c = 15 \text{ K} \) [12]. For the case of \( T = \text{Co} \), both CeCoPO and CeCoAsO show the FM-correlated Co 3d magnetism [13–15].

It is noted that the nickel arsenide LaNiAsO is a low \( T_c \) superconductor and \( T_c \) can be enhanced upon F-doping [5, 6]. Interestingly, the partial substitution of Fe by Ni in LaFeAsO, which introduces two more 3d electrons by each Ni\(^{2+}\) dopant, can induce SC in a very narrow doping regime [16]. Moreover, the normal state resistivity shows an upturn behavior at low temperatures, suggesting a possible Kondo-effect-induced semiconductor behavior. However, there are very few reports on the physical properties of 1111-type NiAs-
based pnictides other than LaNiAsO. The early report did not observe SC above 2 K and suggested a suspicious FM order in CeNiAsO [17], although its structure and the valence of the Ce ion has been studied [18].

In this paper, we report the systematic investigation of the physical properties of CeNiAsO. No evidence of SC is observed for temperatures down to 30 mK. While there is no local magnetic moment on Ni ions, two successive antiferromagnetic transitions related to Ce ions are observed. We propose that the Ce moments could align in ‘G’-type AFM order, first at $T_{N1} = 9.3$ K and then transform into ‘C’-type at a lower temperature $T_{N2} = 7.3$ K. The Sommerfeld coefficient is about 203 mJ mol$^{-1}$ K$^{-2}$, indicating that CeNiAsO is an enhanced correlated Kondo lattice compound. All these results imply a strong hybridization between 4f and 3d electrons and highlight the important role of the 3d–4f interlayer Kondo physics in nickel-based cerium-containing pnictides.

2. Experimental details

A polycrystalline CeNiAsO sample of high purity was synthesized by solid state reaction. Ce, Ni, As and CeO$_2$ of high purity ($\geq$99.95%) were used as starting materials. Firstly, CeAs was presynthesized by reacting Ce discs and As powders at 1320 K for 2 h. NiAs was presynthesized by reacting Ni and As powders at 970 K for 20 h. Secondly, powders of CeAs, CeO$_2$, Ni and NiAs were weighted according to the stoichiometric ratio, thoroughly ground and pressed into a pellet under a pressure of 600 MPa in an argon-filled glove box. The pellet was packed into an alumina crucible and sealed into an evacuated quartz tube, which was then slowly heated to 1450 K and kept at that temperature for 40 h. For comparison, a polycrystalline sample of LaNiAsO was also synthesized, in a similar process, where La$_2$O$_3$ was used as the oxygen source.

Powder x-ray diffraction (XRD) was performed at room temperature using a D/Max-rA diffractometer with Cu Kα radiation and a graphite monochromator. Lattice parameters were derived by Rietveld refinement using the program Rietan 2000 [19]. X-ray photoelectron spectrum (XPS) analysis was carried out by using a VG ESCALAB MARK II device with an Mg Kα ($h\nu = 1253.6$ eV) non-monochromatized source operating in constant analysis energy (CAE, 50 eV) mode, and data were collected in steps of 0.2 eV. Before taking the XPS data, the CeNiAsO sample was pre-polished by the argon beam in a vacuum to make a newly polished sample surface. Electrical resistivity was measured with a standard four-probe method in a Quantum Design physical property measurement system (PPMS-9, for $T \geq 2$ K) and a dilution refrigerator (for $T < 2$ K), while the Hall coefficient was measured by scanning the field from $-5$ to 5 T. Thermopower was measured by a steady-state technique and a pair of differential type E thermocouples was used to measure the temperature gradient. Specific heat was measured by the heat pulse relaxation method in PPMS-9. The dc magnetization measurement was carried out in a Quantum Design magnetic property measurement system (MPMS-5) in zero-field-cooling (ZFC) and field-cooling (FC) protocols under a magnetic field $H$ of 1000 Oe.

3. Results and discussion

Figure 1 shows Rietveld refinement of XRD patterns of CeNiAsO. All the XRD peaks can be well indexed to the tetragonal ZrCuSiAs-type structure with $P4/nmm$ (no. 129) space group, and no obvious impurity trace can be found. The quality factor ($\chi^2$) of this refinement reaches to as low as 0.88, guaranteeing the goodness of sample quality. The refined structural parameters are listed in table 1 with a comparison to CeFeAsO, from which we can find that CeNiAsO has a slightly longer $a$ axis but a much shorter $c$ axis compared to its iron-based counterpart. Because arsenic is drawn closer to the Ni plane, the thickness of the NiAs layer, $d_{\text{NiAs}}$, is much smaller than $d_{\text{FeAs}}$, while the angle of As–T–As ($T = \text{Fe or Ni}$, see the inset of figure 1), is enlarged.

Figure 2 shows temperature dependence of dc magnetic susceptibility of CeNiAsO measured under $H = 1000$ Oe. The data above 150 K exhibit good Curie–Weiss behavior, and can be well fitted to $\chi = \chi_0 + C/(T - \theta_W)$, with

**Table 1. Comparison among CeFeAsO, CeNiAsO and LaNiAsO.**

|                  | CeFeAsO | CeNiAsO | LaNiAsO |
|------------------|---------|---------|---------|
| $a$ (Å)          | 4.0002  | 4.0767  | 4.1231  |
| $c$ (Å)          | 8.6412  | 8.1015  | 8.1885  |
| $z_{\text{as}}$ | 0.1411  | 0.1465  | 0.1470  |
| $d_{\text{As}}$ (Å) | 0.6547  | 0.6434  | 0.6368  |
| $\theta_{\text{As–T–As}}$ (°) | 112.5 | 120.6 | 123.0 |
| $R_{\text{wp}}$ | 11.96%  | 8.11%   | 7.52%   |
| $R_{\chi}$ | 8.16%   | 5.73%   | —       |
| $\chi'$ | 1.12    | 0.88    | 2.02    |
| $\rho$ (300 K) ($\mu$Ω m) | 305.8 | 3.9    | 3.1     |
| $\rho_{\text{R}}$ (300 K) ($10^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$) | $<0$ | 0.68 | $-1.0$ |
| $S$ (300 K) ($\mu$V K$^{-1}$) | $-7.2$ | 2.9 | 0.62 |
| $Y_0$ (mJ mol$^{-1}$ K$^{-2}$) | $\sim59$ | $\sim203$ | $\sim5$ |
The XPS spectrum of CeNiAsO is shown in figure 3. After in this compound, we thus carried out the XPS experiment. The fitting reports that the valence of Ce is the presence of valence state Ce$^{4+}$, which has no 4f electron and local moment. To investigate the valence of Ce in this compound, we thus carried out the XPS experiment. The fitting reports that the temperature-independent term and $\theta_W$ the Weiss temperature. The fitting reports that $\theta_W = -31.8$ K. The obtained effective moment $\mu_{\text{eff}} = 2.24$ $\mu_B$ is slightly smaller than that of a free Ce$^{3+}$ ion, 2.54 $\mu_B$. For $T < 100$ K, we observed a change in the slope of the temperature dependence of inverse susceptibility (right axis of figure 2) and the fitting parameters are $\mu_{\text{eff}} = 2.16$ $\mu_B$, $\theta_W = -25.6$ K. The change in the slope should be ascribed to the crystal electric field (CEF) effect. It has been demonstrated both theoretically and experimentally that Ni ions do not exhibit magnetic ordering in the nickel-based pnictides. Therefore, the Curie–Weiss analysis suggests that the observed effective moments in CeNiAsO should come from Ce ions. When cooled to low temperature, a peak in susceptibility is observed around 9.3 K, followed by an upturn when further cooled. Since the Weiss temperature derived from the Curie–Weiss analysis is negative, it is reasonable to conclude that Ce moments are ordered antiferromagnetically below 9.3 K. The AFM ordering of Ce ions is compatible with the linear field dependence of magnetization (data not shown here), as well as the small magnitude of susceptibility in comparison with CeFeAsO.

One possible interpretation to the noticeable deviation of the obtained effective moment from that of the free Ce$^{3+}$ ion is the presence of valence state Ce$^{4+}$ which has no 4f electron and local moment. To investigate the valence of Ce in this compound, we thus carried out the XPS experiment. The XPS spectrum of CeNiAsO is shown in figure 3. After subtracting the background, the XPS spectrum can be well fitted by a combination of ten Gaussian peaks, i.e. Ce(III) = $v^0 + v' + u^0 + u'$ and Ce(IV) = $v + v'' + v''' + u + u'' + u'''$, with $u$ and $v$ corresponding to the spin–orbit-split $3d_{5/2}$ and $3d_{3/2}$ core holes, respectively. The positions of these peaks are taken from [24], where the XPS spectrum of CePO$_4$ (Ce$^{3+}$) and CeO$_2$ (Ce$^{4+}$) were presented. We found that Ce$^{4+}$ is dominant in CeNiAsO, but there is a small trace of Ce$^{4+}$ which should account for the small peak around 916 eV [25]. An early report on the Ce 3d XPS and Ce L$_3$-edge XANES analysis of CeNiAsO and CeFeAsO by Blanchard et al suggested that the valence of Ce should be close to 3+ [18]. In other Ce-containing compounds with strong Kondo interaction, for example, in CePd$_2$Al$_3$ [26], it is common that Ce has a valence very close to 3+. Therefore, although the valence effect may play some role in the reduction of the effective magnetic moment of Ce, a more likely interpretation to this reduction could mainly come from the CEF effect, which will be discussed hereinafter.

The transport properties of CeNiAsO are presented in figure 4. The resistivity exhibits several prominent features. Firstly, the magnitude of resistivity at room temperature ($T < 20$ K), with two arrows signifying the two magnetic transitions at 9.3 and 7.3 K. Lower inset: temperature-dependent resistivity of LaNiAsO and CeNiAsO, and Hall coefficient of CeNiAsO. Upper inset: the difference of resistivity between CeNiAsO and LaNiAsO, $\rho_{\text{Ce}} - \rho_{\text{La}}$, as well as the thermopower $S$. Office of CeNiAsO and CeFeAsO by Blanchard et al
The good metallicity of CeNiAsO can also be confirmed by the Hall coefficient $R_H$ measurement, as shown in the lower inset. $R_H$ remains as a nearly temperature-independent constant above 150 K and converges to 0.68 × $10^{-4}$ cm$^3$ C$^{-1}$. A rough estimate from the single-band model leads to a carrier density of $\sim 10^{24}$ cm$^{-3}$, which is an upper limit of charge carrier density in a multi-band system, about three orders of magnitude larger than those of LnFeAsO [27–29]. Secondly, in contrast to LaNiAsO, the resistivity in CeNiAsO shows a hump around 100 K. This could be ascribed to the f-electron contribution via Kondo scattering between the localized 4f and the conduction 3d electrons. We subtract the resistivity of LaNiAsO from CeNiAsO and the difference is shown in the upper inset. A broad maximum at 92 K becomes obvious. A similar broad peak centered at 50 K. This broad peak should be attributed to the Schottky anomaly caused by the excitations between CeNiAsO and the difference is shown in the upper inset. A broad peak at 92 K becomes obvious. A similar broad peak centered at 50 K. This broad peak should be attributed to the Schottky anomaly caused by the excitations between 

Figure 5. Evidence for the two magnetic transitions below 10 K, from (a) specific heat; (b) derivative susceptibility $(\Delta T/\Delta T)$ and derivative resistivity $(\Delta T/\Delta T)$ and (c) magnetoresistivity $(\rho_{\text{Hall}} - \rho_0)$ and the Hall coefficient $(R_0)$. The inset of (a) displays the Schottky anomaly fit of $C$ & $T$. The inset of (b) shows $\gamma_0$ derived from extrapolating the $(C - C_{\text{Sch}})/T^2$ plot to the zero limit.

Table 1). The good metallicity of CeNiAsO can also be confirmed by the Hall coefficient $R_H$ measurement, as shown in the lower inset. $R_H$ remains as a nearly temperature-independent constant above 150 K and converges to 0.68 × $10^{-4}$ cm$^3$ C$^{-1}$. A rough estimate from the single-band model leads to a carrier density of $\sim 10^{24}$ cm$^{-3}$, which is an upper limit of charge carrier density in a multi-band system, about three orders of magnitude larger than those of LnFeAsO [27–29]. Secondly, in contrast to LaNiAsO, the resistivity in CeNiAsO shows a hump around 100 K. This could be ascribed to the f-electron contribution via Kondo scattering between the localized 4f and the conduction 3d electrons. We subtract the resistivity of LaNiAsO from CeNiAsO and the difference is shown in the upper inset. A broad maximum at 92 K becomes obvious. A similar broad peak centered at 50 K. This broad peak should be attributed to the Schottky anomaly caused by the excitations between CeNiAsO and the difference is shown in the upper inset. A broad maximum at 92 K becomes obvious. A similar broad peak centered at 50 K. This broad peak should be attributed to the Schottky anomaly caused by the excitations between CeNiAsO and the difference is shown in the upper inset. A broad maximum at 92 K becomes obvious. A similar broad peak centered at 50 K. This broad peak should be attributed to the Schottky anomaly caused by the excitations between
linear dependence in the \((C - C_{\text{ref}})/T - T^2\) plot (see the inset of figure 5(b)) for the temperatures below 30 K. The extrapolated Sommerfeld coefficient \(\gamma_0 \sim 203 \text{ mJ mol}^{-1} \text{ K}^{-2}\), which is more than 40 times that of LaNiAsO (see table 1), manifesting the correlated effect contributed from the Ce 4f electrons. Although the phonon contribution and the CEF effect is hard to remove completely, our analysis should provide a good estimation of the Sommerfeld coefficient of CeNiAsO. The linear fit also produces that \(\beta = 0.2511 \text{ mJ mol}^{-1} \text{ K}^{-4}\) and Debye temperature \(\Theta_D = 314 \text{ K}\), which indicates the above analysis is quite self-consistent. For \(T < 10 \text{ K}\), two \(\lambda\)-shaped peaks are observed on the \(C/T\) curve, implying two different phase transitions. This can be further confirmed by the derivative of susceptibility \((T \cdot d\chi/dT)\), the derivative of resistivity \((d\rho/dT)\), the magnetoresistivity \((\rho_S T - \rho_0)\) and the Hall coefficient \((R_H)\) shown in figures 5(b) and (c). The excellent agreement among the \(C/T\), \(d\rho/dT\) and \(T \cdot d\chi/dT\) leads to a definition of two characteristic temperatures, i.e. \(T_{N1} = 9.3 \text{ K}\) and \(T_{N2} = 7.3 \text{ K}\). Actually, a similar analysis has been widely applied in other nickel-containing compounds, e.g. CeNiGe\(_x\) \([33]\) and LnNi\(_2\)B\(_2\)C \([34]\). Note that the magnetic entropy gain derived by integrating \(C/T\) over temperature reaches \(70\%\) of \(R\ln 2\) at \(10 \text{ K}\), and recovers the full doublet ground state \((R\ln 2)\) at \(30 \text{ K}\). All these indicate that both specific heat peaks are due to Ce 4f-electron-related magnetic transitions, though the ordered moment is partially screened by the Kondo coupling in the ground state doublet. The Kondo scale is estimated to be \(T_K \sim 15 \text{ K}\) by the entropy. A ratio of \(T_K/T_{N1} \sim 1\) can also be obtained by judging from the specific heat jump \(\Delta (C^d - C_{\text{Sch}})\) at \(T_{N1} \sim 35 \text{ K}\). Thus it is reasonable that the Kondo scale \(T_K \sim 15 \text{ K}\). We propose that the transition at \(9.3 \text{ K}\) originated from the Ce-AFM transition based on the magnetization measurement. We also notice that the similar phenomenon was also found in other Ce-based Kondo lattice compounds such as CeCu\(_2\)(Si\(_1-x\),Ge\(_x\))\(_2\), where the two peaks were explained by two incipient instabilities in the magnetic structure of pure CeCu\(_2\)Ge\(_2\) \([36, 37]\): the first one at a higher temperature, related to a reorientation of the moments, and the second one at lower temperature, related to a lock-in of the propagation vector.

Now we turn to discuss the possible magnetic configurations of CeNiAsO. First of all, we should emphasize that all the observed magnetism should arise from the Ce moments. The negative value of the Weiss temperature \(\theta_0\) indicates that Ce moments are AFM-correlated. The most simplified model involves the intralayer and interlayer magnetic interactions between the nearest-neighbor Ce moments, denoted by \(J_{\text{intra}} = J_0 + J_{\text{intra}}^{\text{RKKY}}\) and \(J_{\text{inter}} = J_{\text{inter}}^{\text{RKKY}}\), respectively, where \(J_0\) is the superexchange interaction via O\(^2\)- and As\(^3\)-anions in the absence of d-f coupling \([38]\), while \(J_{\text{intra}}^{\text{RKKY}}\) and \(J_{\text{inter}}^{\text{RKKY}}\) are the Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions mediated by conduction electrons via d-f coupling. Generally, \(J_0\) is always negative (\(J_0 < 0\)) and favors the AFM-ordered ground state \([38]\), while \(J_{\text{intra}}^{\text{RKKY}}\) and \(J_{\text{inter}}^{\text{RKKY}}\) are functions of Kondo coupling \((J_K)\) and density of state at the Fermi energy \((N_F)\) \([39]\). We can assume \(J_{\text{inter}}^{\text{RKKY}} \sim J_{\text{intra}}^{\text{RKKY}} = J_{\text{RKKY}}\), since they have almost the same \(k_F\) and the same nearest-neighbor distances\(^4\). \(J_{\text{RKKY}}\) may be either negative (antiferromagnetic) or positive (ferromagnetic) depending on \(J_K N_F\), which may account for either a G-type or C-type magnetic order, respectively, when cooled down (see the lower inset of figure 1). Actually, the changes in the Hall coefficient and thermopower at low temperatures indeed imply a temperature-induced change, either quantitatively or qualitatively, in either the density or relaxation time of different types of charge carriers. Thus it is plausible that a sign change occurs in \(J_{\text{RKKY}}\) when the temperature is lowered down to 7–9 K, resulting in the magnetic transition from the G-type to C-type magnetic configuration within the Ce-AFM phase. This scenario provides a possible explanation of the two magnetic phase transitions, and is consistent with the drop in the magnetic susceptibility \((\chi(T))\) in the G-type state and then an upturn as it enters the C-type state at \(T < T_{N2}\). Further experimental studies on the single-crystalline sample of CeNiAsO and neutron diffraction measurement are necessary to testify to this scenario.

Finally, it is interesting to compare CeNiAsO with its neighbors, CeFeAsO and CeCoAsO. Previous first-principles calculation on LaTaO \([21]\) suggested that Fe and Co ions show SDW-like and FM instabilities at low temperatures, respectively, while Ni ions display nonmagnetic behavior. For CeTaO\(_3\), Fe ions in CeFeAsO indeed show SDW-like AFM instability and Co ions in CeCoAsO are FM-ordered \([14, 15]\). Meanwhile neither local magnetic moment nor long range magnetic order of Ni ions is observed in CeNiAsO. However, this does not mean that the Kondo coupling between Ce moment and conduction carriers in the NiAs layer is weaker than that in the FeAs layer. The situation could be totally the opposite. We expect the Ce 4f-level is much closer to the conduction band in CeNiAsO than those in CeFeAsO and CeCoAsO. Thus an enhanced hybridization between 4f and 3d electrons is possible in CeNiAsO due to the smaller c-axis lattice constant (see table 1), as shown by the moderately large Sommerfeld coefficient and the absence of SC.

4 We argue that \(J_{\text{RKKY}}\) depends on the Ce–Fe–Ce distance. Thus interlayer and intralayer RKKY interactions have almost the same Ce–Fe–Ce distances.
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