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Superconductivity in $SrNi_2As_2$ single crystals

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The electrical resistivity $\rho(T)$ and heat capacity $C(T)$ on single crystals of $SrNi_2As_2$ and $EuNi_2As_2$ are reported. While there is no evidence for a structural transition in either compound, $SrNi_2As_2$ is found to be a bulk superconductor at $T_c=0.62$ K with a Sommerfeld coefficient of $\gamma=8.7$ mJ/mol K^2 and a small upper critical field $H_{c2}\sim 200$ Oe. No superconductivity was found in $EuNi_2As_2$ above 0.4 K, but anomalies in ρ and C reveal that magnetic order associated with the Eu^{2+} magnetic moments occurs at $T_m=14$ K.

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The $ThCr_2Si_2$ structure type is well known for accommodating a superconducting ground state, particularly in the heavy fermion community with superconductors such as $CeCu_2Si_2$ and URu_2Si_2.[1] Soon after the discovery of superconductivity in $LaFeAsO_{1-x}F_x$ at $T_c=26$ K with the structure type ZrCuSiAs,[2] it was realized that the related compounds (AFe_2As_2, with $A=$Ba, Sr, Ca, and Eu) in the $ThCr_2Si_2$ structure are also superconducting either with doping[3–7] or under pressure.[8–10] While systems with Fe_2As_2 planes have the highest T_c's to date, superconductivity has been found in both structure types with either Ni_2P_2 (Refs. 11 and 12) or Ni_2As_2 (Refs. 13–16) layers.

Here we report the observation of superconductivity in single crystals of $SrNi_2As_2$ at $T_c=0.62$ K, as determined by heat capacity, in the absence of a structural phase transition (below 400 K). Following our initial observation of superconductivity in $BaNi_2As_2$,[16] this represents the second superconducting system in the $ThCr_2Si_2$ structure with Ni_2As_2 layers. In addition, we report that our $EuNi_2As_2$ single crystals grown from Pb flux are not superconducting above 0.4 K.

Single crystals of $SrNi_2As_2$ and $EuNi_2As_2$ were grown in Pb flux in the ratio (Sr,Eu):Ni:As:Pb=1:2:2:20. The starting elements were placed in an alumina crucible and sealed under vacuum in a quartz ampoule. The ampoule was placed in a furnace and slowly heated to 1050 °C, as described in Ref. 16. The sample was then cooled slowly (5 °C hr^{-1}) to 600 °C, at which point the excess Pb flux was removed with the aid of a centrifuge. For $SrNi_2As_2$ the resulting platelike crystals were heavily embedded in a yet unidentified needle-like impurity phase. From single-crystal x-ray refinements, the platelike samples were confirmed to crystallize in the $ThCr_2Si_2$ tetragonal structure (space-group 139, $I4/mmm$). The refinement for $SrNi_2As_2$ $[R(I>2\sigma)=3.7\%]$ at 124 K yields lattice parameters $a=4.1374(8)$ Å and $c=10.188(4)$ Å and fully occupied (>98%) atomic positions Sr $2a(0,0,0)$, Ni $4d(0.5,0,0.25)$, and As $4e(0,0,z)$ with $z=0.3634(1)$ consistent with previous reports.[17–19] The refinement for $EuNi_2As_2$ $[R(I>2\sigma)=5.09\%]$ at 124 K gives lattice parameters $a=4.0964(6)$ Å and $c=10.029(3)$ Å and fully occupied (>98%) atomic positions Eu $2a(0,0,0)$, Ni $4d(0.5,0,0.25)$, and As $4e(0,0,z)$ with $z=0.3674(2)$ also consistent with previous reports.[20,21]

The in-plane electrical resistivity data for ANi_2As_2 ($A=$Ba,Sr,Eu) is shown in Fig. 1. All samples exhibit metallic behavior. $SrNi_2As_2$ is a relatively good metal with a RRR$[=\rho(300\text{ K})/\rho(4\text{ K})]$ of 11, and a residual resistivity of 7 $\mu\Omega$ cm. The resistivity of $EuNi_2As_2$ exhibits a kink at $T_m=14$ K associated with magnetic ordering of the Eu^{2+} moments, consistent with previous reports.[21] For $SrNi_2As_2$, there is no evidence of a structural transition below 400 K, in contrast to $BaNi_2As_2$, which has a clear first-order transition at $T_0=130$ K. The lack of a phase transition in $SrNi_2As_2$ is also provided by the heat-capacity data shown in the inset of Fig. 3.[22]

Figure 2 presents the low-temperature in-plane resistivity data for $SrNi_2As_2$ with fields applied parallel and perpendicular to the c axis. In zero field, a sharp superconducting transition is observed at $T_c=0.66$ K, defined as the midpoint of the resistive anomaly. With increasing magnetic field, the transition remains sharp and is quickly suppressed. The specific heat shown in Fig. 3 confirms the bulk nature of superconductivity in $SrNi_2As_2$. The zero resistance state coincides exactly with the onset of the specific-heat transition, from which we extract a superconducting transition temperature of $T_c=0.62$ K by an equal area construction. From a fit to the data from 0.7 to 3 K of $C/T=\gamma+\beta T^2+\delta T^4$, a Sommerfeld coefficient $\gamma=8.7$ mJ/mol K^2 is obtained. Using this value, the ratio of the specific-heat jump at T_c to the electronic specific heat is estimated to be $\Delta C/\gamma T_c\simeq 1.0$. From the

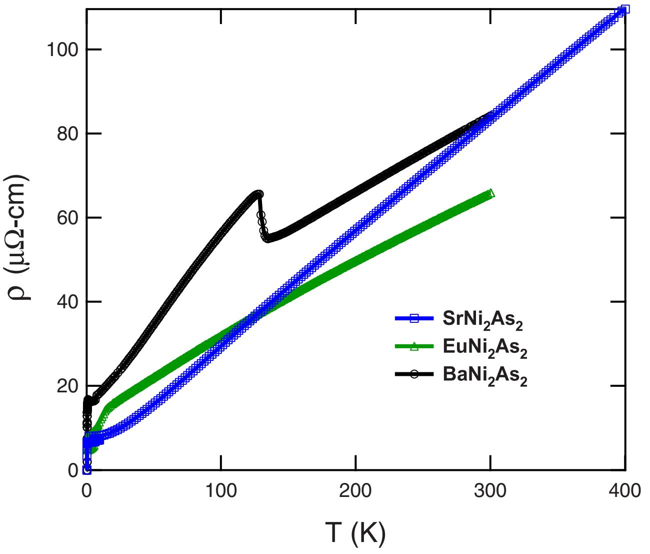

FIG. 1. (Color online) In-plane electrical resistivity $\rho(T)$ $(I\parallel ab)$ for selected ANi_2As_2 ($A=$Ba,Sr,Eu) compounds.

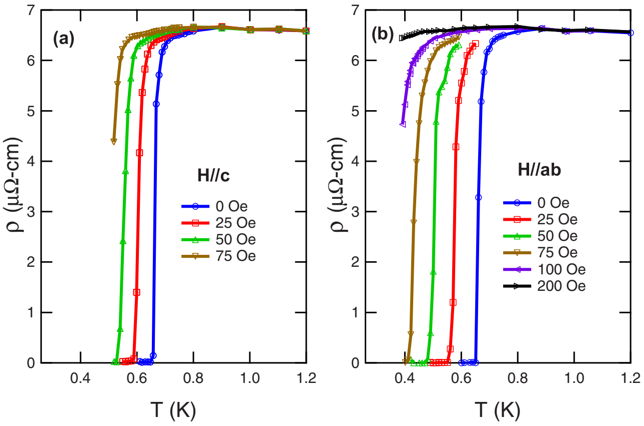

FIG. 2. (Color online) Electrical resistivity $\rho(T)$ of $SrNi_2As_2$ showing the superconducting transition for (a) $H\parallel c$ and (b) $H\parallel ab$. The current was maintained perpendicular to the magnetic field.

β coefficient=0.67 mJ/mol K^4, one obtains a Debye temperature $\Theta_D=244$ K.

The magnetic field-temperature H-T phase diagram of $SrNi_2As_2$ is shown in Fig. 4 determined from the $\rho(T)$ curves in Fig. 2, along with the data for $BaNi_2As_2$ for comparison.[16] For $SrNi_2As_2$, the zero-temperature orbital critical field[23] $H^*_{c2}(0)=-0.7T_c dH_{c2}/dT_c$ is determined to be 210 and 150 Oe for $H\parallel c$ and $H\parallel ab$, respectively. From this, the superconducting coherence length is estimated via $H^*_{c2}(0)=\Phi_0/2\pi\xi^2$,[24] yielding $\xi^{ab}=1477$ Å and $\xi^{c}=1250$ Å. Assuming $SrNi_2As_2$ is in the clean limit, values for the Fermi velocity $v_F^{ab}=7.1\times 10^6$ cm/s and $v_F^{c}=6.0\times 10^6$ cm/s are obtained from $\xi_0=0.18\hbar v_F/k_B T_c$. Surprisingly, while the absolute value of T_c in zero magnetic field is very similar for the two compounds, the anisotropy (H^{ab}_{c2}/H^{c}_{c2}) of the upper critical field, which is a factor of 2.1 in $BaNi_2As_2$, reverses sign $(H^{c}_{c2}/H^{ab}_{c2}=1.4)$ in $SrNi_2As_2$ and the overall magnitude of the upper critical field is nearly an order of magnitude smaller. The differences in the H_{c2} between the two compounds may be due to changes in electronic structure resulting from the structural transition in $BaNi_2As_2$ that is not present in $SrNi_2As_2$.

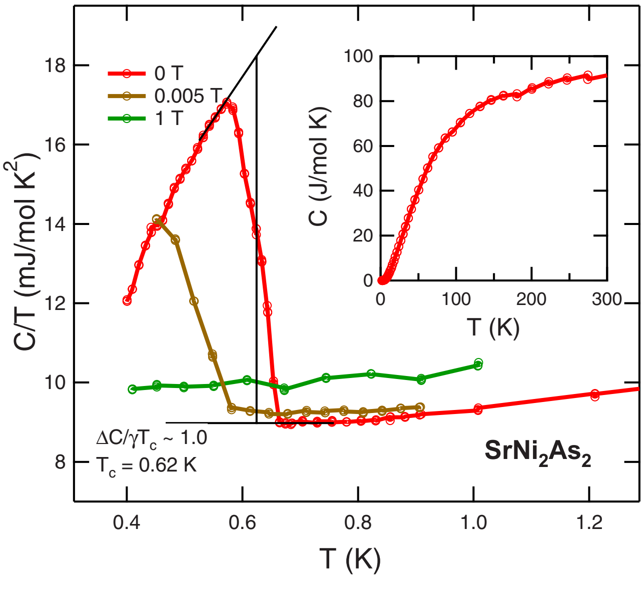

FIG. 3. (Color online) Low-temperature specific heat C versus temperature T of $SrNi_2As_2$ (Ref. 22) for various magnetic fields $(H\parallel c)$. The inset displays the high-temperature heat capacity with no indication of a first-order phase transition at higher temperatures.

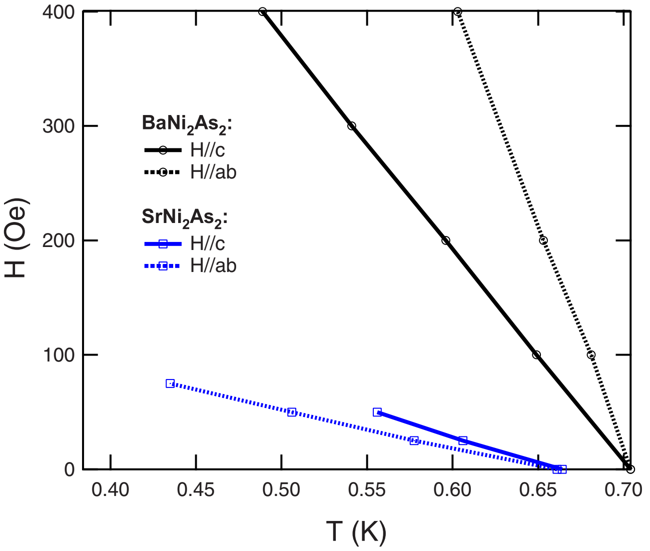

FIG. 4. (Color online) Magnetic field-temperature H-T phase diagram of $SrNi_2As_2$. The upper critical field H_{c2} for $H\parallel c$ and $H\parallel ab$ was determined by the resistive midpoint in Figs. 2(a) and 2(b). The data for $BaNi_2As_2$ from Ref. 16 is for the zero resistance state.

The specific heat, plotted as C/T, for $EuNi_2As_2$ is shown in Fig. 5 in magnetic fields up to 9 T $(H\parallel c)$. A sharp anomaly occurs at the magnetic ordering temperature $T_m=14$ K [consistent with the kink in $\rho(T)$, Fig. 1], as well as a broader hump at ~4 K. A magnetic field along the c axis modestly suppresses the transition, consistent with previous reports of antiferromagnetic ordering in polycrystalline samples.[21,25]

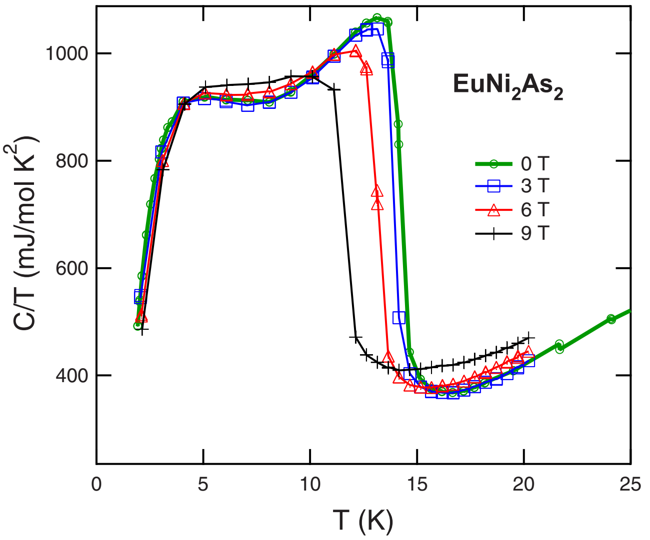

FIG. 5. (Color online) Heat-capacity data versus temperature for $EuNi_2As_2$ in zero and applied magnetic field (Ref. 22). The magnetic field was applied along the c axis.

There is no indication of superconductivity above 0.4 K in $EuNi_2As_2$.

It is interesting that superconductivity with very similar transition temperatures is found both in $BaNi_2As_2$ (Ref. 16) and in $SrNi_2As_2$ despite the differences in structural parameters caused by the smaller Sr^{2+} ions, as well as the presence of a first-order structural transition in $BaNi_2As_2$ that is possibly also magnetic. Recent theoretical work by Subedi *et al.*[26] indicate that the superconducting properties of the related Ni-analog LaNiPO may be explained within a conventional electron-phonon approach yielding a low value of $T_c = 2.6\text{K}$, consistent with experiment; the authors go on to suggest that the Fe-As superconductors may be in a separate class from their Ni-based counterparts. However, a scenario was put forth by Cvetkivoc and Tesanovic;[27] involving a multiband Fermi surface in layered FeAs superconductors to produce the large values of T_c may also be an appropriate description of these ANi_2As_2 superconductors as well. Further work is in progress to elucidate the nature of the superconductivity in these Ni-based materials and its relation to fine details of the electronic structure.

In conclusion, specific-heat and electrical resistivity measurements of $SrNi_2As_2$ single crystals reveal bulk superconductivity at 0.62 K, which shows no sign of a structural and/or magnetic anomaly below 400 K. Magnetic ordering associated with the Eu magnetic moments is observed in single crystalline $EuNi_2As_2$. No evidence for superconductivity is observed in this compound above 0.4 K.

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