Unusual Metallic Properties in Cubic FeGe at High Pressure

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During the last decade an increasing number of magnetically ordered metallic systems with anomalous low-temperature thermodynamic and transport properties have been reported. The majority of the anomalies occurred in rare-earth based compounds when their magnetic ordering temperature — of the order of several tens of Kelvin — is smoothly tuned to absolute zero by an appropriate control parameter like chemical composition or pressure. Close to such a quantum critical point (QCP) the thermodynamic and transport properties are dominated by quantum rather than thermal fluctuations. They lead to strong deviations in the temperature dependence of, e. g., the specific heat or the electrical resistivity, compared to that predicted by the Landau-Fermi-liquid (LFL) theory. Usually these non-Fermi-liquid (NFL) signatures fade away as the system is tuned away from the QCP. Common to all of these compounds is, however, that the metallic ground state, although exhibiting unusual properties, is conserved. Thus, the question arises whether or not it would be possible to observe a metal-to-insulator transition (MIT) as the QCP is reached. In this respect, pressure studies on the ferromagnetic metallic compound FeGe might provide further information.

Recent experiments on $FeSi_{1-x}Ge_x$ performed by Yeo and coworkers [1] showed a discontinuous transition from the Kondo insulating state in FeSi [2] to the metallic phase in FeGe [3] at a critical concentration $x_c \approx 0.25$. Upon further increasing the Ge-content the magnetic properties of FeGe were eventually established. The magnetic ordering in FeGe can be described as a long-range spiral with a period of about 700 Å, and moments perpendicular to the spiral propagation vector **k**. The spiral propagates along equivalent [100] directions (**k** is parallel [100]) just below $T_C = 280$ K at ambient pressure [4]. A change in the spiral direction to **k** parallel [111] occurs at a first-order transition at $T_1 \approx 210$ K [4]. Yeo and coworkers interpreted the MIT to be consistent with a reduction of the hybridization in conjunction with an increase of disorder at the Si-site. In order to elucidate disorder effects we studied the influence of a pure volume reduction on the transport properties of FeGe.

We performed electrical resistivity measurements on a single crystal of FeGe in a large temperature range (50 mK < T < 300 K) and pressures up to 23 GPa. Single crystals of the cubic modification of FeGe (non-centrosymmetric space group $P2_13$) were grown by chemical vapor transport using iodine as transport agent [5] in a homemade two zone furnace. The growth process is very slow and it took several months although the transport was made perpendicular to the tube axis. The resulting crystals had a volume of less than 1 mm^3 and the largest were examined thoroughly by various X-ray techniques to investigate orientation and quality and by electron-probe microanalysis to determine the composition. For the pressure experiment a parallelepiped piece was cut and polished to a final size of 690 × 105 × 23 μm^3. The current in the setup of the four-probe electrical resistivity measurement was applied perpendicular to the [111] direction and its density was in the range $4 < j < 80$ Acm^{-2}.

Figure 1 shows the temperature dependence of the magnetic contribution to the electrical resistivity,

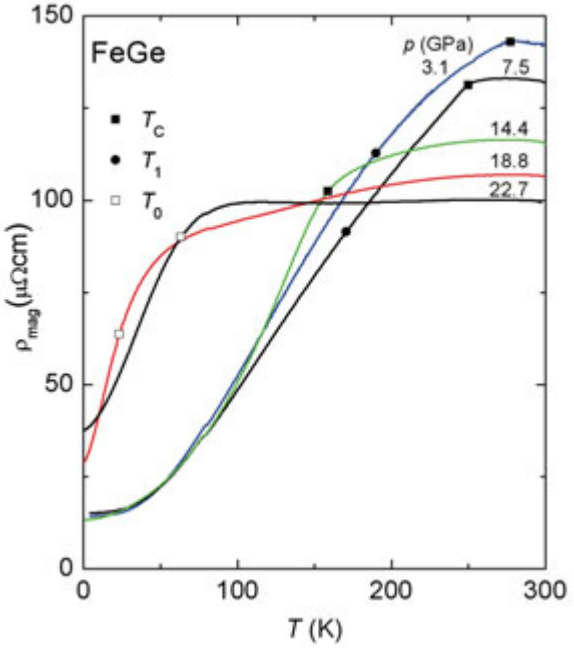

Fig. 1: Temperature dependence of the electrical resistivity, $\rho_{mag}(T)$, of FeGe at various pressures. Several anomalies in $\rho_{mag}(T)$ are attributed to the change of the magnetic structure at temperatures T_C, T_0, and T_1.

$\rho_{mag}(T)$. The kink in $\rho_{mag}(T)$ occurring at T_C is related to the onset of magnetic order. As pressure increases this kink broadens and the maximum slope of the first temperature derivative of $\rho_{mag}(T)$ has to be used to define T_C . Surprisingly, the position of the anomaly increases as pressure exceeds 19 GPa, and the anomaly is thus labeled T_0 in Fig. 1. The broad feature in $\rho_{mag}(T)$ visible below T_C is associated with the second transition temperature at T_1 and is unambigiously seen for $p < 12$ GPa. One important observation of this experiment is that, although the residual resistivity has doubled at 22.7 GPa, the ground state in FeGe remains metallic.

The dependence of these anomalies on pressure is depicted in Fig. 2. The long-range magnetic order seems to be suppressed at a critical pressure $p_c \approx 19$ GPa were $T_C \rightarrow 0$. Above p_c the (presumably short-range ordering) temperature T_0 increases. The monotonic decrease of T_C is in strong contrast to that found for $FeSi_{1-x}Ge_x$ [1] as $T_C(p)$ seems to be a second-order phase transition without any connection to an insulating state. To illustrate this difference we added the $T_C(x)$ data of $FeSi_{1-x}Ge_x$ in Fig. 2. For this purpose the $T_C(x)$ dependence was transformed into a $T_C(p)$ dependence using the measured $V(x)$ dependence [6] and a $p(V)$ Murnaghan equation-of-state (EOS) taking a linear change of the bulk modulus between FeSi (B_0 = 160 GPa [7]) and FeGe (B_0 = 130 GPa [8]) into account. For the pressure dependence of B_0, B', we used 4.71 for both systems. This comparison shows a stronger decrease of T_C in the alloy below a pressure of about 10 GPa. A good agreement with $T_C(p)$ in FeGe is seen at $p \approx 15$ GPa, corresponding to $x \approx 0.27$, which is close to the MIT. The effect of alloying and pure volume reduction, however, seems to influence the quantum fluctuations in such a way that an insulating state occurs in $FeSi_{1-x}Ge_x$ whereas the metallic state persists in FeGe [8].

In order to investigate how the crystal structure evolves at these extreme pressure and temperature conditions we performed X-ray powder-diffraction experiments. For the measurements at four different temperatures (82 K, 140 K, 230 K, 290 K) single crystals of cubic FeGe were ground into fine powder and the fraction with particle sizes below 20 µm was annealed in vacuum at 670 K for 24 h in order to remove residual stress. The resulting powder was loaded into a diamond anvil pressure cell using helium as pressure transmitting medium. Refinements

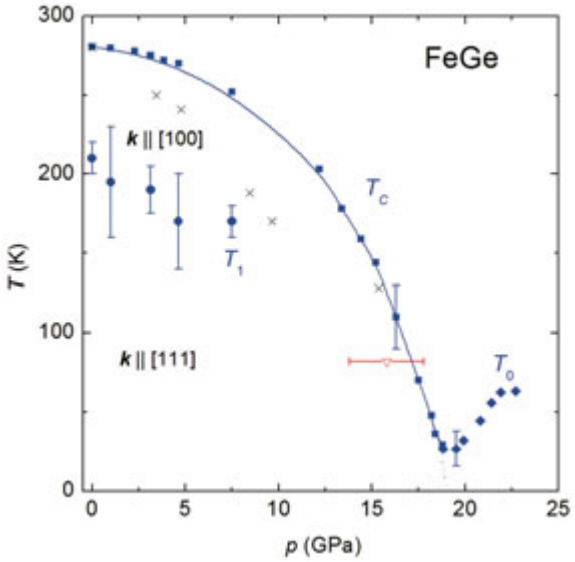

Fig. 2: (T, p) phase diagram of FeGe obtained from transport (bold symbols) and structural (open symbol) investigations. $T_C(x)$ data of $FeSi_{1-x}Ge_x$ (×) are included for comparison (taken from Ref. [1]). The long-range ordered phase is suppressed at a critical pressure $p_c \approx 19$ GPa. A presumably different kind of magnetic order (below T_0) seems to exist above p_c. The low-temperature and low-pressure phase (below T_1) seems to vanish at intermediate pressures.

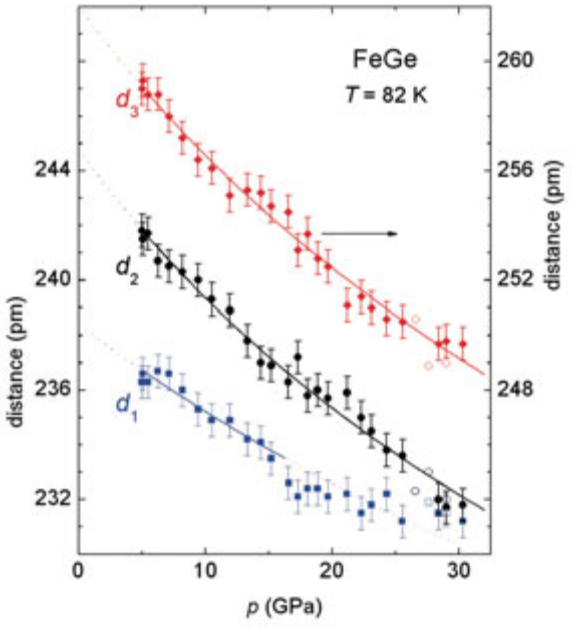

Fig. 3: Interatomic distances d(Fe–Ge) in cubic FeGe at 82 K. The shortest distance d_1 changes its low-pressure dependence at about 15.8 GPa (dashed line). The pressure of the discontinuity corresponds within experimental error to that of the change in the magnetic order (cf. $T_C(p)$ in Fig. 2).

of the crystal structure using powder diffraction data were performed by means of a least-squares procedure using the full profile method (program package WinCSD).

The interatomic distances d(Fe–Ge) as obtained by refinements of powder diffraction data recorded at 82 K are shown in Fig. 3. The pressure dependence can be described with a Murnaghan equation of state (lines in Fig. 3). However, for the shortest distance, d_1, which is between Fe and Ge atoms along the [111] direction, a deviation from the extrapolated low-pressure behavior can be seen at about 15.8 GPa. This pressure agrees rather well with the transition into the paramagnetic state as deduced from the electrical resistivity data ($T_C(p)$ data in Fig. 2). Thus, $d_1(p)$ seems to be sensitive to the loss of long-range magnetic order. All distances at higher temperatures can be well described with Murnaghan equations of state.

These results show that the pure volume reduction in FeGe leads to a suppression of the long-range magnetic order. The metallic state above p_c, however, exhibits unusual transport properties probably caused by small short-ranged moments below a characteristic temperature T_0.

References

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