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A pressure and magnetotransport study of binary quasicrystal $YbCd_{5.7}$

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Abstract

We have probed the quasicrystalline state in binary $YbCd_{5.7}$ by monitoring its electrical resistivity between 1.5 and 300 K in externally applied hydrostatic pressure up to 16 kbar and measuring its magnetoresistivity up to 12 T from 0.5 to 20 K. The thermal variation of the resistivity is practically unaffected by pressure, indicating the stability of the quasicrystalline state in this pressure regime. A positive magnetoresistance, $\Delta\rho/\rho$, of ~0.8% is observed at 0.65 K, which reduces to ~0.4% at 20 K in the maximum applied field of 12 T. Though the magnetoresistance of the sample investigated is about an order of magnitude larger than expected on the basis of the empirically derived relationship $\Delta\rho/\rho \sim \rho^{1.3}$ observed for a large number of quasicrystals, it is much below the unusually large anomalous magnetoresistance (20–200%) reported earlier in the literature. We believe that the intrinsic magnetoresistivity of $YbCd_{5.7}$ in different samples may be primarily masked by the presence of traces of free Cd which is known to have a giant magnetoresistivity at low temperatures.

The binary compound $YbCd_{5.7}$ was recently discovered to be a stable quasicrystal of P-type icosahedral symmetry (i-phase) [1]. Quasicrystals are materials that possess classically forbidden orientational point group symmetry but lack the translational invariance of the crystal lattice. It is believed that $YbCd_{5.7}$ should reflect the intrinsic properties of the quasicrystalline state due to its thermodynamical stability and binary nature, in contrast to the ternaries where the presence of parasitic phases, defects, etc may give rise to extrinsic effects. Indeed, the thermodynamic and various other physical properties of $YbCd_{5.7}$, such as magnetization, electrical and thermal transport, ultrahigh-resolution photoemission spectroscopy, etc have been reported in the literature [2–6]. Yb ions in $YbCd_{5.7}$ and in the cubic crystalline approximant $YbCd_6$ are in the divalent $4f^{14}$ state at room temperature and hence have a

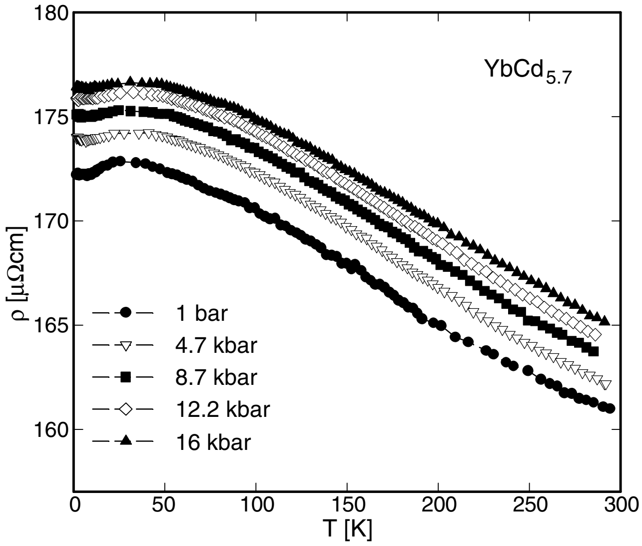

Figure 1. The temperature dependent electrical resistivity ρ of $YbCd_{5.7}$ for various values of applied hydrostatic pressure.

zero magnetic moment. It is believed that the divalent state of Yb ions is a necessary prerequisite for the formation of the quasicrystalline phase in $YbCd_{5.7}$. The thermal variation of the low-temperature heat capacity of $YbCd_{5.7}$ is satisfactorily described by the expression $C/T = \gamma + \beta T^2$, where γ and β are the coefficients of the electronic and lattice heat capacities, respectively. The Debye temperature θ_D (140–145 K) obtained from β is the lowest observed for the i-phases [3, 4]. The low Debye temperature indicates that the lattice of $YbCd_{5.7}$ is rather soft and it motivated us to study the effect of the external hydrostatic pressure on its quasicrystalline state. Additionally, the compression of the lattice may favour a shift towards the $4f^{13}$ state of the Yb ions, which has a smaller ionic radius than the divalent $4f^{14}$ state and possess a magnetic moment. Pressure induced changes of the Yb valence are well known in Yb compounds showing anomalies ascribed to valence fluctuations and the Kondo effect. In this work we have studied the effect of external pressure up to 16 kbar by monitoring the thermal variation of the electrical resistivity of $YbCd_{5.7}$. We have also investigated the electrical resistivity at various magnetic fields, and the magnetoresistance was determined at selected temperatures from 0.5 to 20 K up to 12 T.

Figure 1 shows the temperature dependent electrical resistivity ρ of $YbCd_{5.7}$ in the temperature range from 1.5 K to room temperature at selected pressures up to 16 kbar. The sample used in the present experiments is the same as that studied in [4], and the ambient pressure, room temperature resistivity was adjusted accordingly. The details of the sample preparation and characterization are given in [4]. $\rho(T)$ of $YbCd_{5.7}$ is characterized by a negative temperature coefficient for $T > 30$ K. Around this temperature, a shallow maximum develops, followed by a marginal drop of $\rho(T)$. Increasing pressure leaves these general features unchanged, but causes an increase of the absolute $\rho(T)$ value. The absence of any anomaly in the data recorded under pressure with respect to the behaviour at ambient conditions shows that the quasicrystalline state of $YbCd_{5.7}$, as well as the Yb valence and the band structure, are stable within the pressure range studied. These results may evidence a relatively high value of the bulk modulus, B, although the Debye temperature hints to a rather soft lattice. Moreover, the only minor changes of the resistivity observed by the application

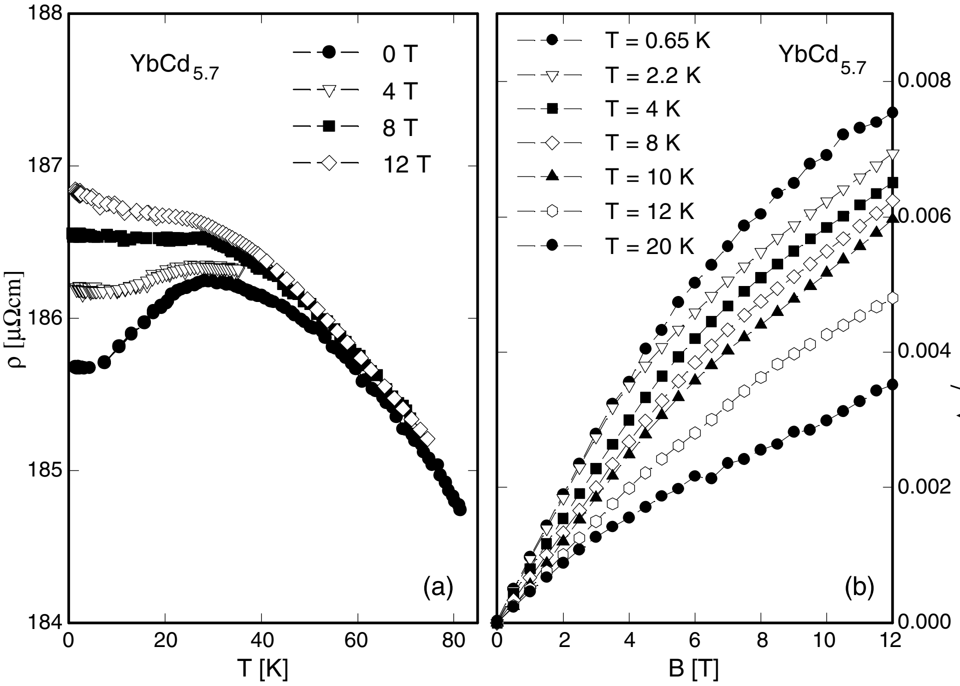

Figure 2. (a) The temperature dependent electrical resistivity ρ of $YbCd_{5.7}$ for various values of externally applied magnetic fields. (b) The isothermal magnetoresistance $\Delta\rho/\rho$ of $YbCd_{5.7}$ at various temperatures.

of hydrostatic pressure prove the absence of any strongly volume dependent energy scales determining the electronic properties of this quasicrystal. Since the 4f shell of divalent Yb is fully occupied it is energetically very stable, and that may lend additional stability to $YbCd_{5.7}$ against externally applied pressure.

Quasicrystals are characterized by unusual transport properties, which are often uncorrelated in this class of materials, and a proper quantitative understanding is still lacking. However, their magnetoresistivity (MR) is unique in the sense that most results can be quantitatively described by invoking the theory of quantum interference effects (QIE) in weakly disordered metals. A prerequisite for QIE is that the coherence length of the electron wave is much larger than the electron mean free path. The QIE manifest themselves as weak localization (WL) and electron–electron effects (EEI). A combination of WL and EEI has been successfully applied to account for the observed MR in a large number of quasicrystals whose low-temperature (~4 K) resistivity values differ by more than two orders of magnitude. Figure 2 shows the temperature dependent resistivity for various magnetic fields (a) and $\Delta\rho/\rho = [\rho(B) - \rho(0)]/\rho(0)$, for various temperatures up to 20 K (b). Here, $\rho(B)$ is the electrical resistivity at a magnetic field B and $\rho(0)$ is the zero field resistivity. The overall variation of $\Delta\rho/\rho$ is very small and decreases with increasing temperatures. The application of an external field suppresses the slight drop of $\rho(T)$ below about 30 K due to the positive MR, and as a consequence, $\rho(T)$ at high fields increases continuously with decreasing temperatures.

At the highest applied field of 12 T, $\Delta\rho/\rho$ is ~0.8 and 0.7% at 0.65 and 2.2 K, respectively. At 20 K, $\Delta\rho/\rho$ is below 0.4%. The observed change is roughly an order of magnitude larger than expected on the basis of the empirically derived correlation between the magnitudes of $\Delta\rho/\rho$ and ρ in ternary i-phases [7]. For ρ between $\sim 10^2$ and $\sim 10^5$ $\mu\Omega$ cm, it has been seen that $\Delta\rho/\rho \sim \rho^{1.3}$, where the value of ρ is taken at ~4 K and $\Delta\rho$ is the change of the resistivity at the highest field applied in that particular measurement, which in some experiments may be as high as few tens of tesla; see figure 5.9 in [7], which shows a plot of $\Delta\rho/\rho$ against ρ. An interpretation of this correlation has been given by invoking quantum interference effects (QIE) in weakly disordered metals, which predicts $\Delta\rho/\rho \sim \rho^{1.5}$. Based on the above-mentioned empirical relation, MR in our sample, in which $\rho(300\text{ K})/\rho(4.2\text{ K}) \approx 0.92$ and $\rho(4.2\text{ K}) \approx 173\ \mu\Omega$ cm, is expected to change by ~0.06%. Though we observe a larger MR, our values are one to two orders of magnitude less than the extraordinarily large MR reported in [3]—about 20% for i-$Cd_{84.6}Yb_{15.4}$ ($YbCd_{5.5}$) and 200% for i-$Cd_{84.1}Yb_{15.9}$ ($YbCd_{5.3}$) at 4.2 K and 9 T. The authors of that work noticed the obvious disparity between their observations and the empirical relation derived in the literature. Though the two compositions studied in [3] are slightly different in their Cd:Yb ratio than the one that we have studied here, their resistivity (560 and 240 $\mu\Omega$ cm, respectively, at 300 K and $\rho_{2\text{ K}} < \rho_{300\text{ K}}$) is much lower than what one would infer from the observed $\Delta\rho/\rho$ and the empirically established relation. To put this in some perspective, Ahlgren and co-workers observed a MR of ~100% in i-AlPdRe in which ρ is ~$10^5\ \mu\Omega$ cm at 4 K [8]. We believe that the significantly enhanced values of MR reported in [3] and the relatively modest enhancement seen by us are of extrinsic origin. A microscopic examination of our samples showed traces of semimetal Cd in both $YbCd_{5.7}$ and the crystalline approximant $YbCd_6$, studied earlier in [4]. Further corroboration is provided by the occurrence of a superconducting transition in $\rho(T)$ at zero field in both compounds around 0.6 K, which matches the superconducting transition temperature of 0.6 K of pure Cd. Although the amount of free Cd in $YbCd_{5.7}$ is found to be very small, a flimsy sheet or network of Cd on the surface of the sample may be responsible for the incidence of a full developed superconducting transition as observed experimentally. Thereby, the volume of the superconducting phase may be well below the standard numbers for percolation. The surface of both $YbCd_{5.7}$ and $YbCd_6$ tarnishes on exposure to air while the contacts for the electrical leads are made.

Cd is well known for its giant magnetoresistance, sensitively depending on the RRR value of a particular sample [9, 10]. MR values up to ~10^7% in pure Cd at 1.4 K in maximum applied field of ~2.5 T were reported in [9]. In view of the 'extraordinarily' large MR of Cd, even trace amounts of free Cd can completely dominate the intrinsic MR of a sample, if the latter is low. The MR in the crystalline approximant $YbCd_6$ is large [11]. Just to give a few numbers, $\Delta\rho/\rho$ of $YbCd_6$ at $T = 2$ K is 385% at a field of 12 T; even at 22 K this value, at the same field, is as large as 100%. This shows that the amount of free Cd in $YbCd_6$ is more than in $YbCd_{5.7}$. That is understandable as $YbCd_6$ is formed by a peritectic reaction while $YbCd_{5.7}$ melts congruently. Due to its peritectic formation, $YbCd_6$ was annealed at 640 °C to enhance the formation of single phase, while no annealing treatment was given to $YbCd_{5.7}$ in [4]. It may also be noted that in [3] MR has been measured on two compositions which are located away from $YbCd_{5.7}$, the MR increasing with decreasing Yb:Cd ratio. Further, the alloys were prepared there by melting the constituents, wrapped in a molybdenum foil, in a quartz tube. Since no additional details about the sample preparation are given, we presume the reaction has been carried at low temperatures at which Yb gradually dissolves in molten Cd. At high temperatures, both Yb and Cd, which are volatile, would sublimate appreciably out of the molybdenum foil. On the other hand, we have melted the samples in sealed tantalum crucibles by heating up to above the liquidus temperature (900–950 °C). This results in a better mixing of the constituents and improved quality of the samples in terms of the volume of the bulk single phase. Even a few per cent of free Cd in the samples studied in [3] can easily give rise to large MR observed there, as observed by us in $YbCd_6$. It is absolutely essential to have a sample which is completely free of Cd to obtain the intrinsic value of the MR. Unless this precondition is assured, a discussion on the possible variation of the MR with Yb:Cd ratio will be meaningless. Measuring the resistivity in zero field down to at least 0.5 K would be a good check-up on the sample quality in this regard. Keeping in mind the presence of trace amounts of Cd in our sample of $YbCd_{5.7}$, it is natural to conclude that the intrinsic MR of this quasicrystal is lower and presumably follows the correlation between the MR and zero-field resistivity.

To conclude, we find that $YbCd_{5.7}$ is structurally and electronically stable against externally applied pressure up to 16 kbar, despite having a relatively low Debye temperature. The low-temperature magnetoresistivity is about an order of magnitude larger than expected on the basis of the correlation $\Delta\rho/\rho \sim \rho^{1.3}$ empirically established in a large number of quasicrystals. However, the deviation from the established trend is most likely not intrinsic but due to the presence of trace amounts of free Cd in the sample.

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References

[1] Tsai A P, Guo J Q, Abe E, Takakura H and Sato T J 2000 *Nature* **408** 537

[2] Guo J Q, Takakura H, Abe E and Tsai A P 2000 *Phys. Rev.* B **62** R14605

[3] Tamura R, Murao Y, Takeuchi S, Tokiwa K, Watanabe T, Sato T J and Tsai A P 2001 *Japan. J. Appl. Phys.* **40** L912

[4] Dhar S K, Palenzona A, Manfrinetti P and Pattalwar S M 2002 *J. Phys.: Condens. Matter* **14** 517

[5] Tamura R, Murao Y, Takeuchi S, Kiss T, Yokoya T and Shin S 2002 *Phys. Rev.* B **65** 224207

[6] Tamura R, Araki T and Takeuchi S 2003 *Phys. Rev. Lett.* **90** 226401

[7] See, Rapp Ö 1999 *Physical Properties of Quasicrystals* ed Z M Stadnik (Berlin: Springer) p 127

[8] Ahlgren M, Rodmar M, Gignoux C, Berger C and Rapp Ö 1997 *Mater. Sci. Eng.* A **226–228** 981

[9] Katyal O P and Gerritsen A N 1969 *Phys. Rev.* **178** 1037

[10] Katyal O P and Gerritsen A N 1969 *Phys. Rev.* **185** 1017

[11] Bauer E, Lindbaum A, Michor H, Müller H, Dhar S K, Surjeet Singh, Manfrinetti P and Palenzona A 2003 communicated for publication