Quantum critical behavior in transport properties of *i*-AuAlYb

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Point in phase diagram where second order phase transition temperature is suppressed to zero temperature is called quantum critical point (QCP). Physical properties of the system near QCP are governed by quantum fluctuations that become important even at higher temperatures indicating critical point occurrence. Recently, quantum critical phenomena was observed for the first time in quasiperiodic crystal in magnetic susceptibility and specific heat. [1] Icosahedral i-Au-Al-Yb is intermediate valence compound [2] with majority of Yb ions in trivalent high spin state (J=7/2) compared to divalent (J=0) Yb ions. Although approximant phase with very similar local structure and composition shows intermediate valence character but with larger asymmetry among amount of divalent and trivalent ions it does not show quantum criticality at low temperatures. We have measured electrical resistivity and thermopower down to 1.5 K and investigated magnetic field influence on thermoelectric transport properties. It turned out that electrical resistivity anomalous low-temperature behavior below 50 K that tend to vanish in magnetic field. Thermopower is positive at high temperatures but at 12 K changes sign and near 4 K has minimum. [3] It seems that this minimum and change of sign is somehow connected to quantum criticality since it disappears in magnetic field. We propose some kind of magnetic ordering of high spin Yb ions as ground state of i-Au-Al-Yb.

[1] K. Deguchi, *et al.* Nat. Mater. **11** (2012) 1013.
[2] T. Watanuki, *et al.* Phys Rev B **86** (2012) 094201.
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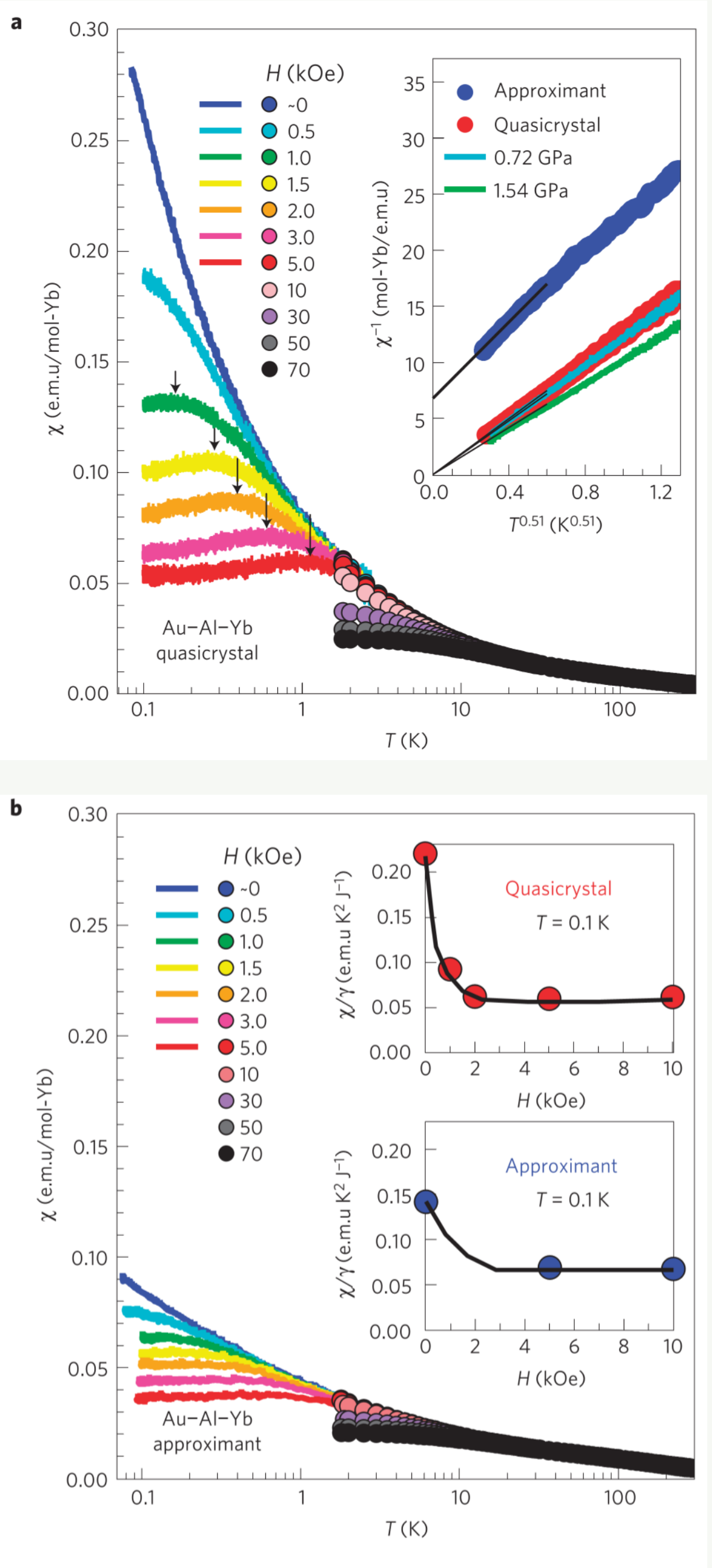
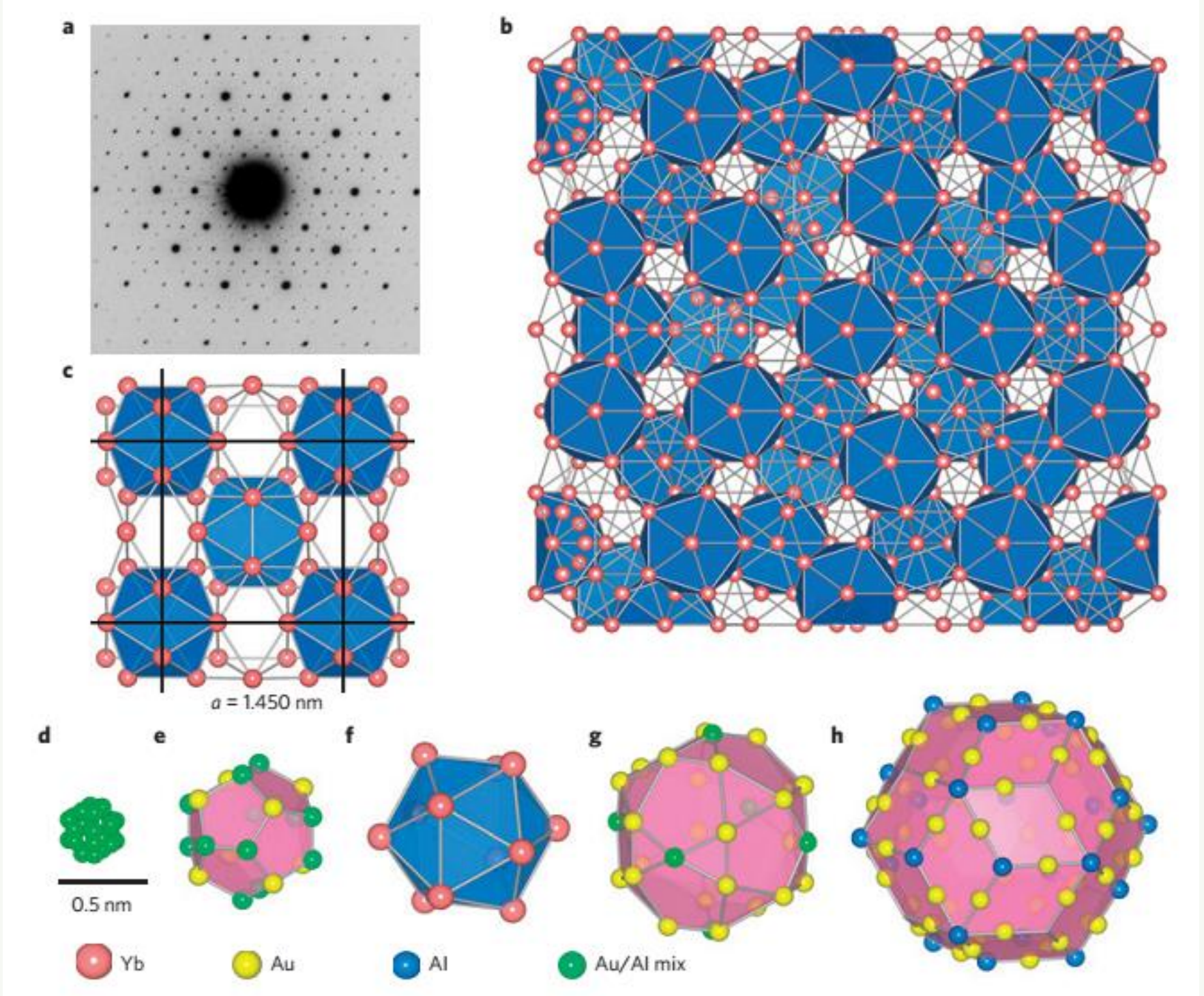
Point in phase diagram where second order phase transition temperature is suppressed to zero temperature is called quantum critical point (QCP). Physical properties of the system near QCP are governed by quantum fluctuations that become important even at higher temperatures indicating critical point occurrence. Recently, quantum critical phenomena was observed for the first time in quasiperiodic crystal in magnetic susceptibility and specific heat. [1] Icosahedral *i*-Au-Al-Yb is intermediate valence compound [2] with majority of Yb ions in trivalent high spin state (J=7/2) compared to divalent (J=0) Yb ions. Although approximant phase with very similar local structure and composition shows intermediate valence character but with larger asymmetry among amount of divalent and trivalent ions it does not show quantum criticality at low temperatures. Recently we have measured electrical resistivity and thermopower down to 1.5 K and investigated stabilization mechanisms.[3]

Here we present magnetic field influence on thermoelectric transport properties. It turned out that anomalous low-temperature behavior of electrical resistivity below 50 K tend to vanish in magnetic field. Thermopower is positive at high temperatures but at 12 K changes sign and near 4 K has minimum. It seems that this minimum and change of sign is somehow connected to quantum criticality since it disappears in magnetic field. We propose some kind of magnetic ordering of high spin Yb ions as ground state of *i*-Au-Al-Yb.

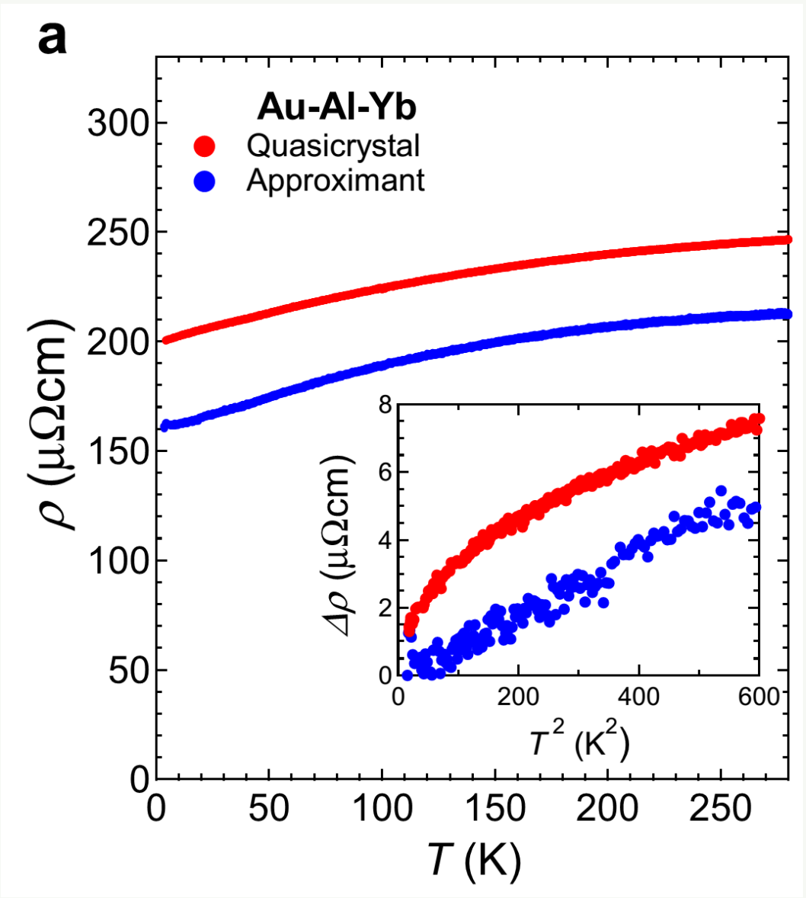
From literature



Quantum critical behavior seen from magnetic susceptibility *i*-$Au_{51}Al_{34}Yb_{15}$. Interestingly, approximant phase $Au_{51}Al_{35}Yb_{14}$ with very close composition and similar local atomic arrangement doesn't show critical behavior. Inset shows robustness of quantum criticality to hydrostatic pressure.



a) SAED of the Au–Al–Yb quasicrystal. **b**) Atomic arrangement of Yb atoms in the isostructural $Cd_{5.7}Yb$ quasicrystal. The icosahedral aggregate is highlighted. **c**) Yb arrangement in the Au–Al–Yb approximant in the projection along the [001] direction. **d) – h)** Concentric shell structures of Tsai-type cluster in the Au–Al–Yb approximant. Each vertex of the first cluster presented in d is occupied by Au/Al mixed atoms with an occupancy 1/6.

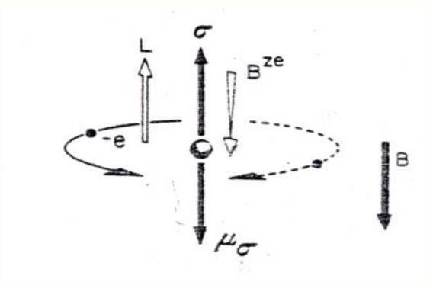


In electrical resistivity, approximant phase shows normal T^2 behavior at low temperatures, while i-AuAlYb has critical behavior. At high temperatures there is no difference.

Deguchi et al., Nature Materials ***11*** *(2012) 1013.*

Hall effect

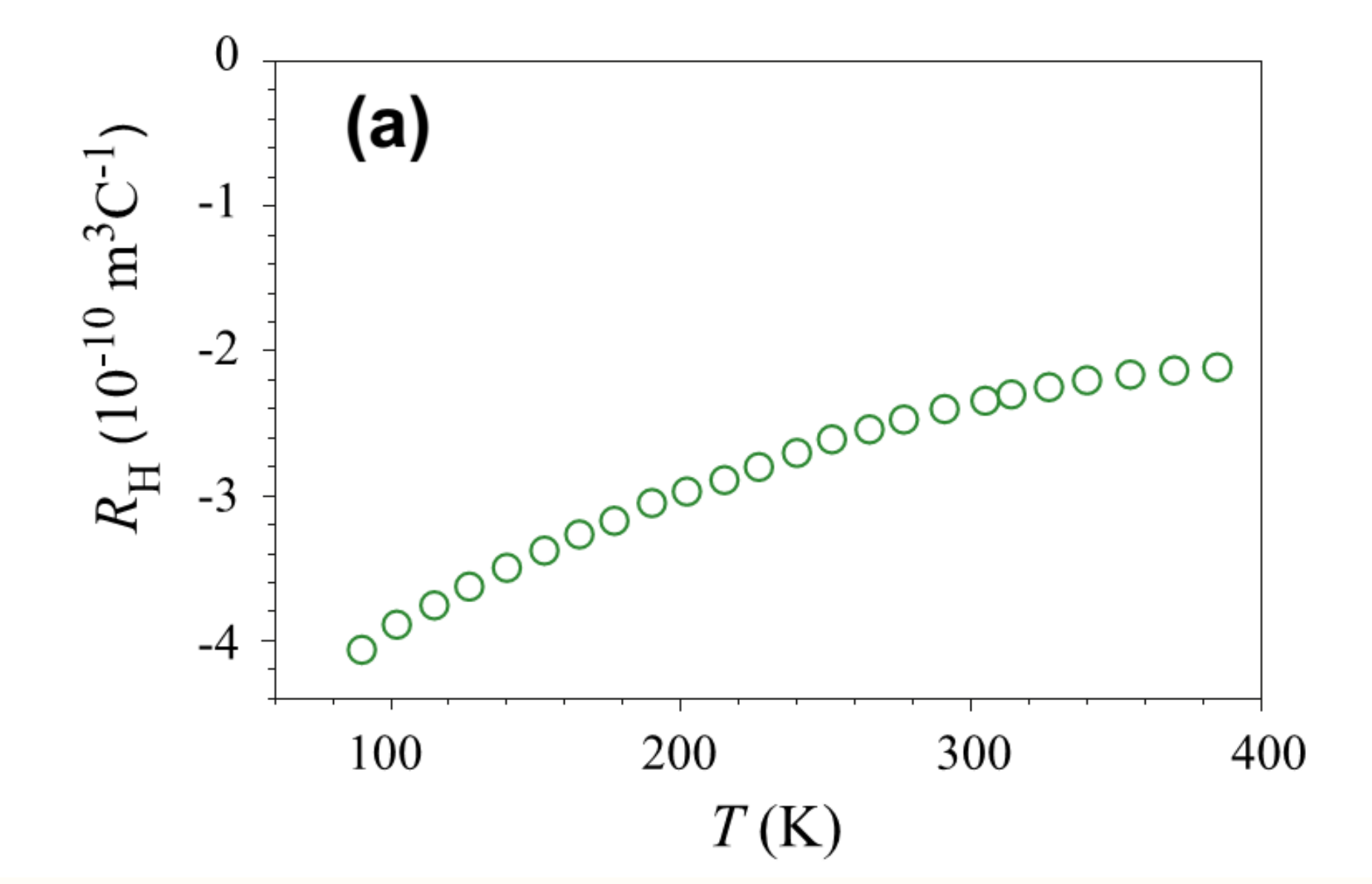
In some paramagnetic materials, in addition to Lorentz force, there is additional asymmetric scattering of electrons due to spin-orbit coupling [4].



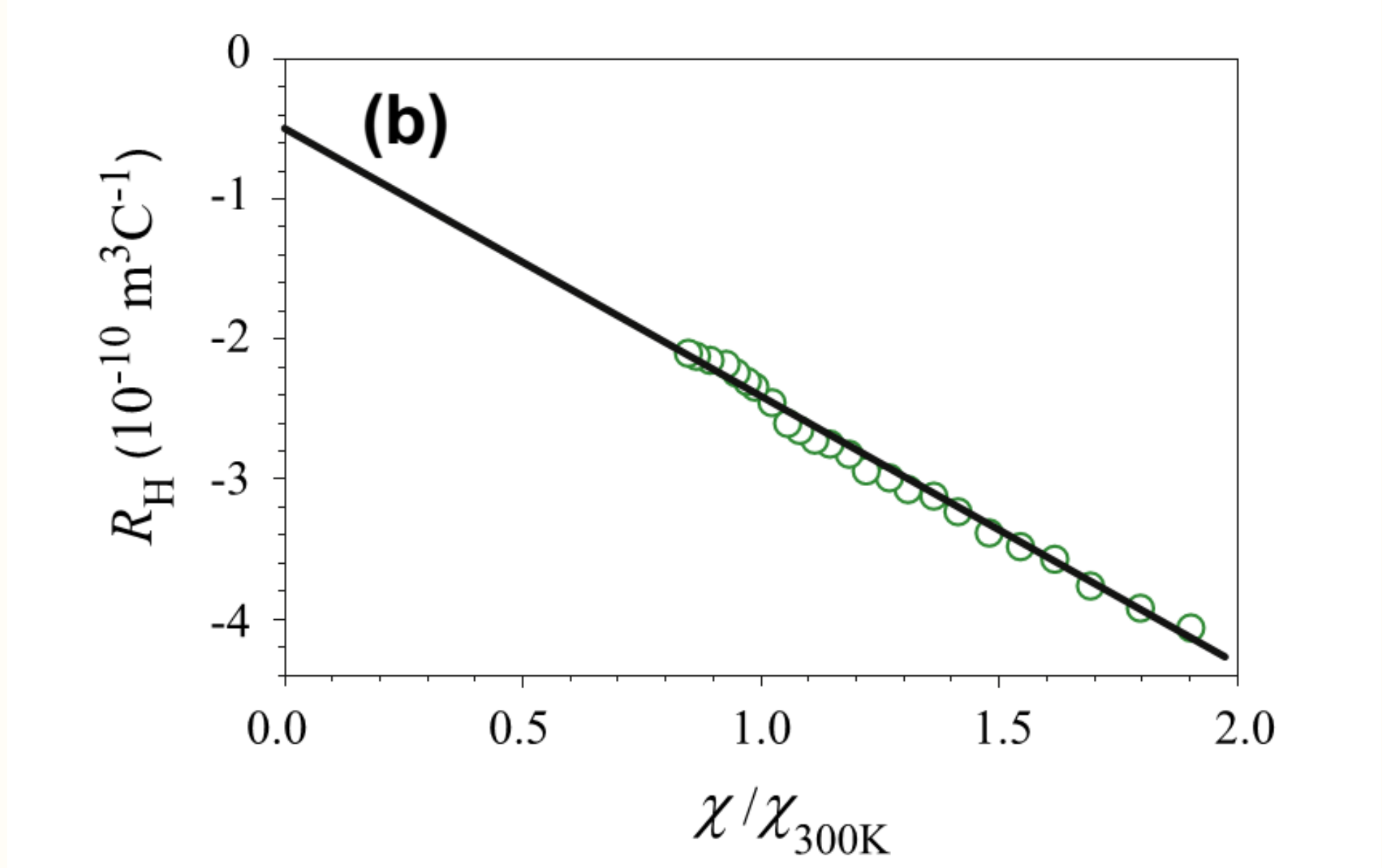
Full line trajectory has lower energy than dashed one leading to anomalous Hall effect.

In presence of anomalous Hall effect Hall resistivity has form:

$$\rho_H = R_0 B + R_S \mu_0 M$$

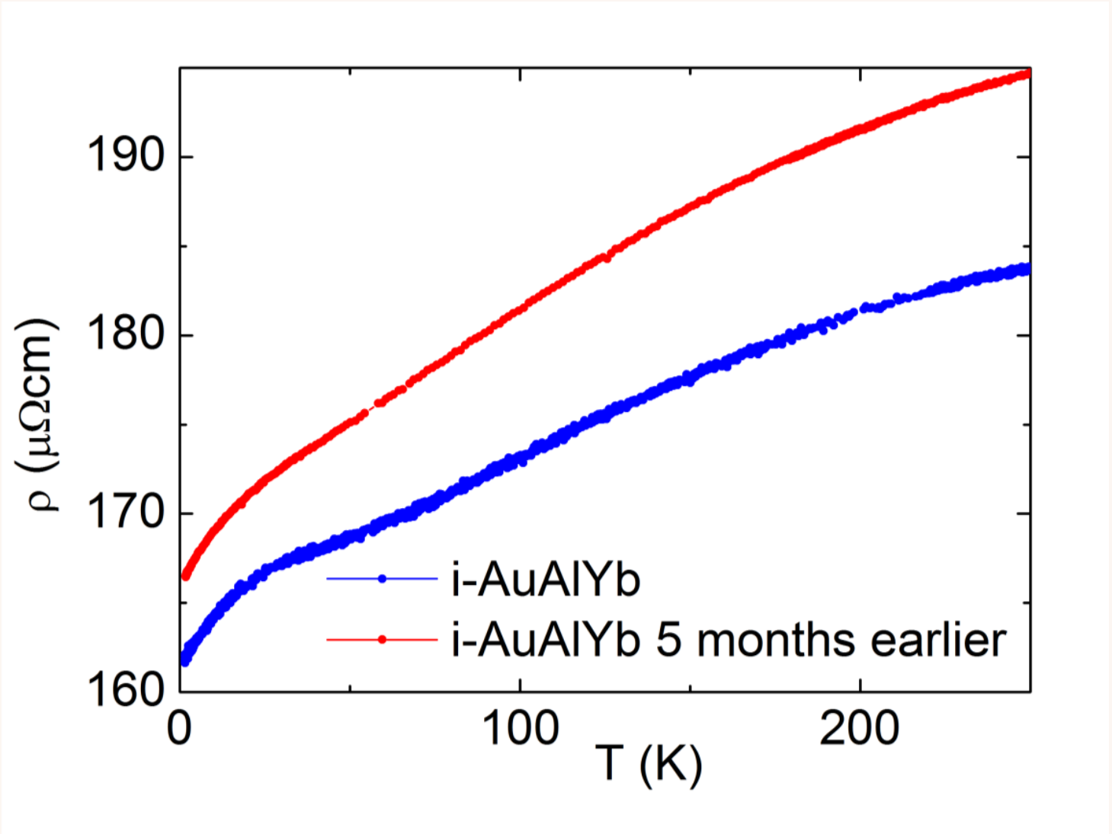


Temperature dependence of Hall effect $R_H(T)$ of the *i*-AuAlYb quasicrystal, due to anomalous contribution follows magnetic susceptibility temperature behavior.

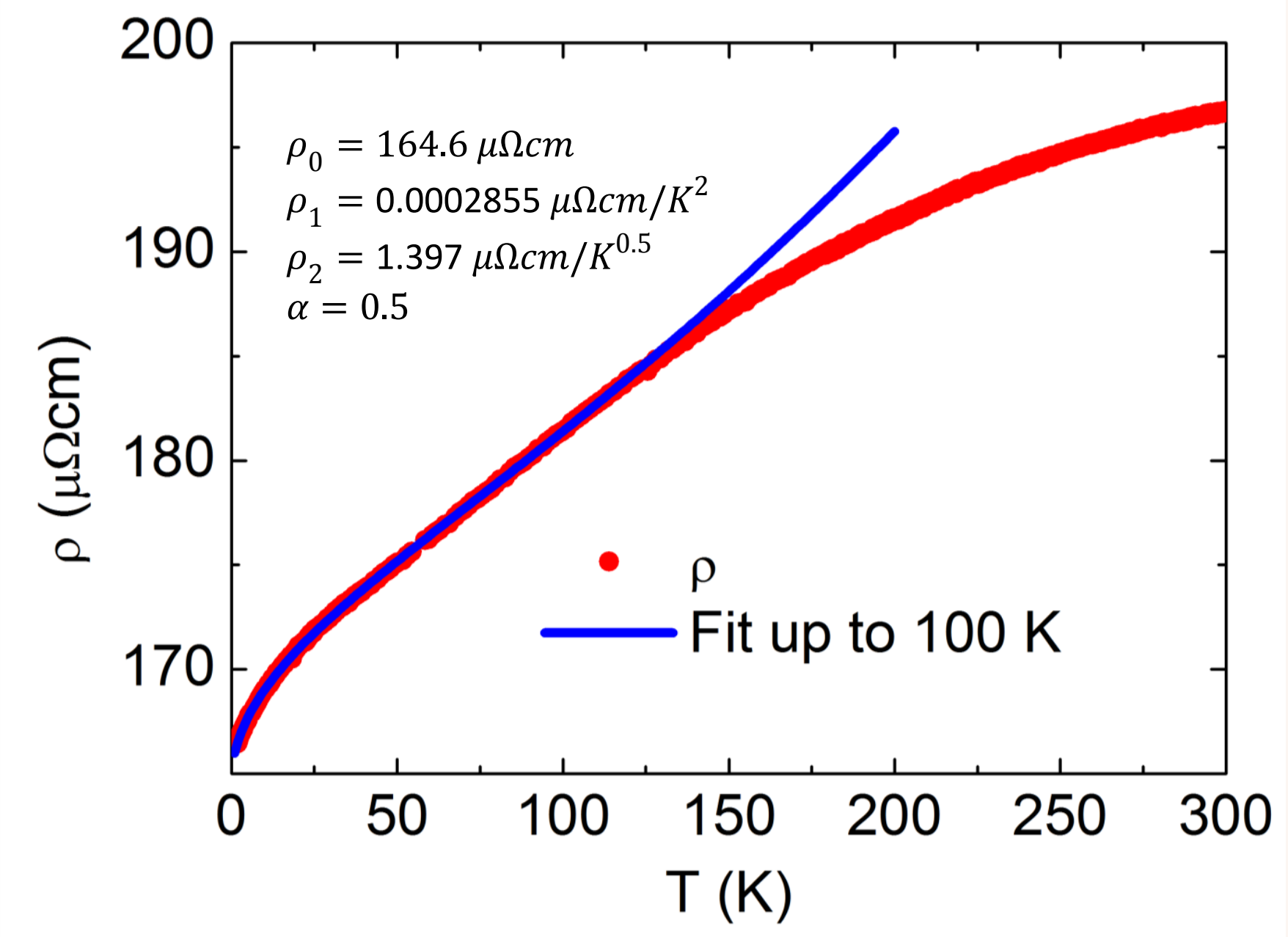


b) R_H as a function of the normalized magnetic susceptibility χ/χ_{300K}. Solid line is the fit with equation $R_H = R_0 + \chi R_s$. R_0 represents normal Hall coefficient and R_s is anomalous Hall coefficient.

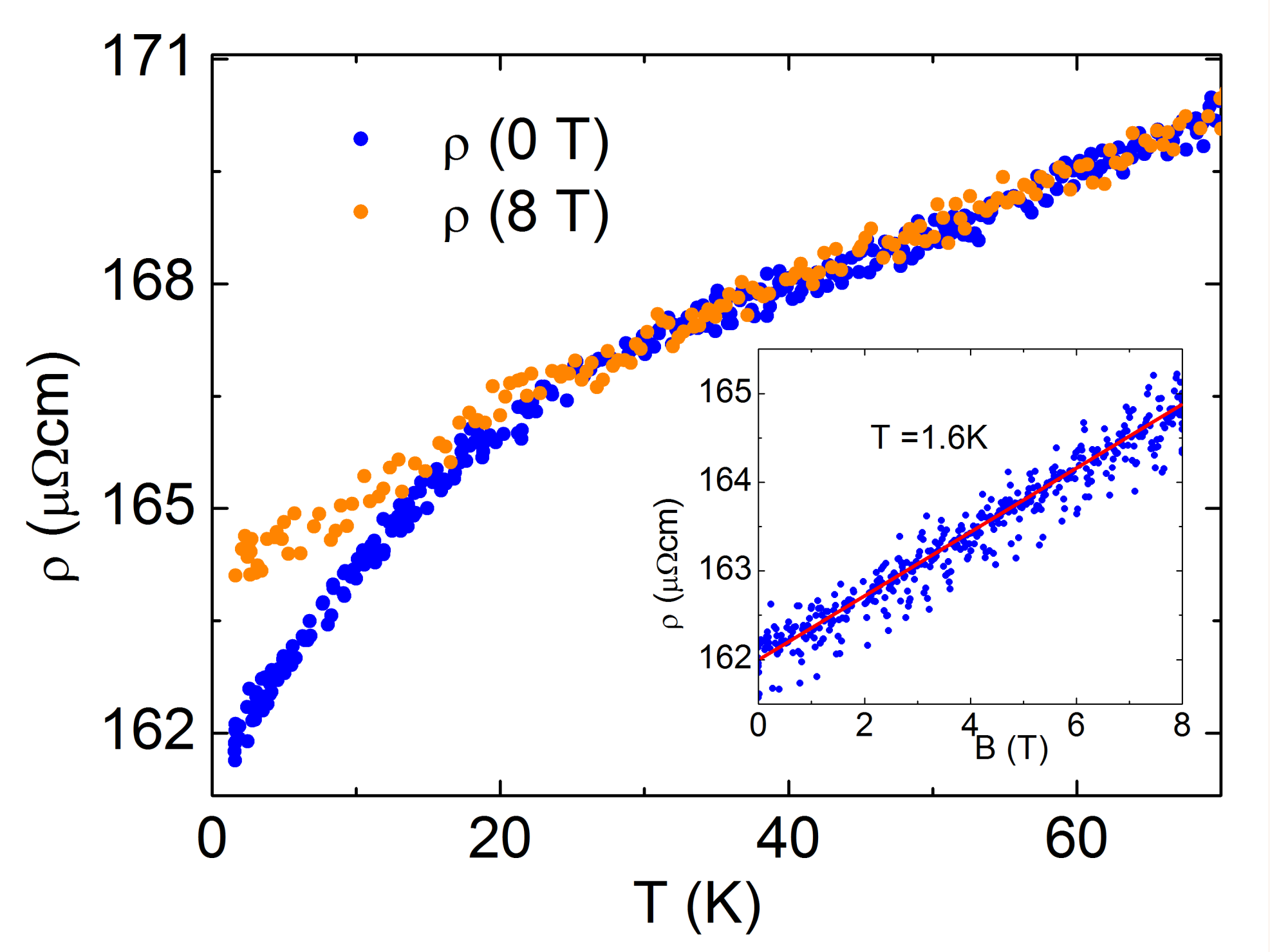
Electrical resistivity



Evidence of aging in electrical resistivity – smaller temperature coefficient -> more obvious evidence of quantum criticality in electrical resistivity
Metastable phase



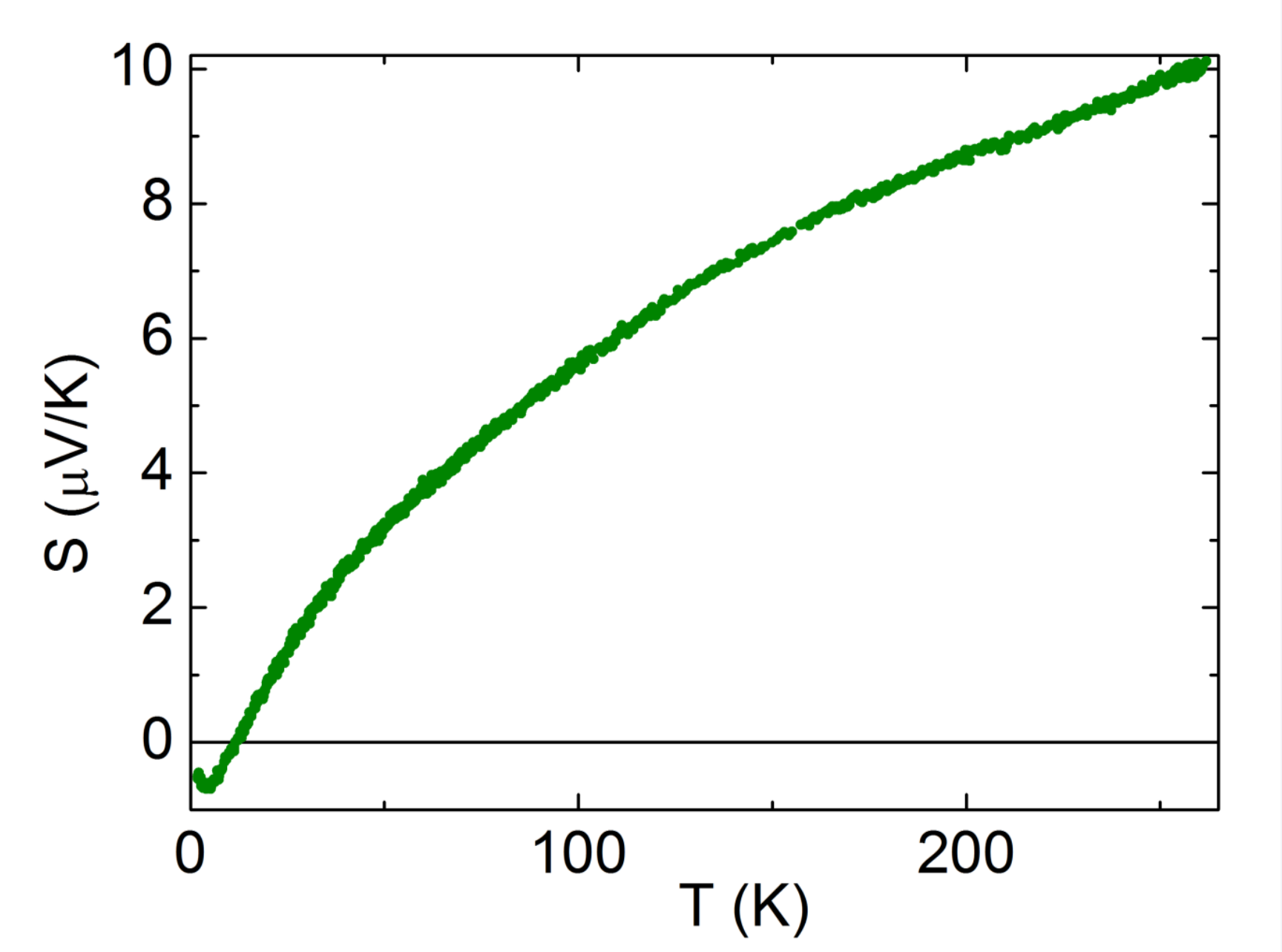
Electrical resistivity below 100 K was successfully fitted to $\rho_0 + \rho_1 T^2 + \rho_2 T^\alpha$ where ρ_0 represents residual resistivity, T^2 term is represents Fermi liquid behavior seen in approximant phase and whose presence can be perceived between 100 and 50 K before criticality overcome. $\rho_2 T^\alpha$ term represents critical behavior with exponent $\alpha = 0.5$ suggesting $\sqrt{T}$ temperature dependence of electrical resistivity in vicinity of quantum critical state.



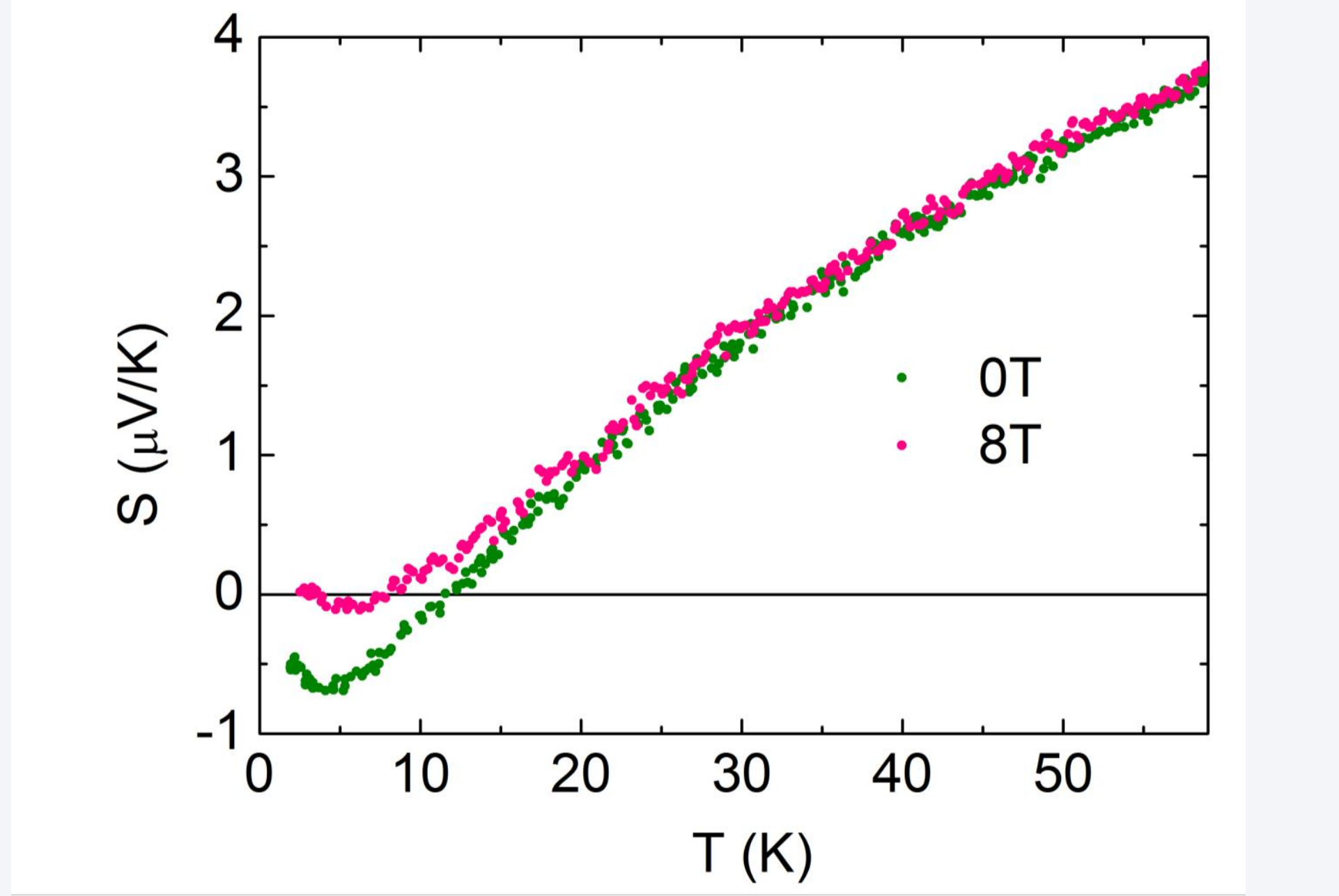
Suppression of $\sqrt{T}$ in electrical resistivity with magnetic field is clearly observed.
Inset shows nearly linear magnetic field dependence of electrical resistivity at 1.6 K. On the other hand Deguchi [1] reported $\sqrt{B}$ behavior at 0.1 K. Discrepancy can be vindicated with higher temperature in our measurement where quantum effects are smaller and can be hidden with the scattering of experimental results.

Rate of electrical resistivity change with magnetic field at 1.6 K is close to 0.35 $\mu\Omega cm/T$.

Thermopower



Critical behavior in thermopower is manifested through change of sign below 12 K, and at 5 K it has minimum.
In magnetic field however, this local minimum is suppressed, and thermopower reach zero value near 10 K, and doesn't change much at lower temperatures.



For thermopower measurements we used two chromel constantan thermocouples in electrical contact with sample. Such configuration allowed us simultaneously measurement of electrical resistivity and thermopower.
Thermopower of chromel thermocouple wire in magnetic field was corrected according to Chiang [5]

Summary: Change of sign of thermopower could indicate highly asymmetric density of states at Fermi level.
For better understanding of dynamics of the electronic system, Hall effect measurements at lower temperatures would be very helpful. Suppression of electrical resistivity at temperatures below 40 K, is probably caused by the increase of the correlation length between magnetic Yb ions. However, neutron scattering data are necessary to prove it.

References:

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[3] S. Jazbec, *et al*. J. Alloys Compd. **586** (2014) 343.
[4] C. M. Hurd, The Hall Effect in Metals and Alloys, Plenum press, 1972, New York
[5] C. K. Chiang, Rev. Sci. Instrum. **45** (1974) 985.