Magnetic Properties of the Au-Al-Yb Approximant under Hydrostatic Pressure

Shuya Matsukawa, Kazuhioko Deguchi, Keiichiro Imura, Tsutomu Ishimasa, and Noriaki K. Sato

1Department of Physics, Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan
2Division of Applied Physics, Graduate School of Engineering, Hokkaido University, Sapporo 060-8628, Japan
E-mail: matsukawa.shuya@mbox.nagoya-u.ac.jp

Abstract. We report on ac magnetic susceptibility measurements of the Au-Al-Yb approximant under pressure. It is known that the approximant shows unconventional quantum criticality at the critical pressure $P_c \approx 2 \text{ GPa}$, and further that the magnetic ordered state emerges at a low temperature ($T_s \approx 100 \text{ mK}$) for $P > P_c$. We find that the nonlinear susceptibility shows a peak around the temperature $T_s$ at which the linear susceptibility peaks. We also find that $T_s$ increases with frequency of the modulation field. From these results, we suggest that the magnetic ordered state of the approximant below $T_s$ is a spin-glass-like short-range ordered state.

1. Introduction

Quantum critical phenomena have attracted much interest in last decades. These are observed near the quantum critical point (QCP) of strongly correlated materials where the second-order transition temperature vanishes to zero. The low-temperature properties of these materials differ from those of ordinary Fermi systems. Recently, the unconventional quantum critical phenomenon was observed in the Au-Al-Yb icosahedral quasicrystal: As the temperature $T$ is decreased toward zero temperature, the physical properties diverge like $\chi \propto T^{-0.51}$ and $C/T \propto -\ln T$ (where $\chi$ is the magnetic susceptibility and $C$ is the specific heat), suggesting that the system is just at the QCP without tuning [1]. These critical indices are very unusual and similar to those of the heavy fermion crystals $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$ [2]-[5]. In addition, the quantum critical behavior of the quasicrystal is robust against the application of hydrostatic pressure: At 1.54 GPa, the divergence of the magnetic susceptibility with the same critical index ($\gamma \approx 0.5$) was observed [1]. In contrast, the Au-Al-Yb approximant does not show such the diverging behavior at ambient pressure: The magnetic susceptibility is described as $1/\chi \propto T^{0.5} + \text{constant}$ [6]. Here we note that the presence of the constant term means that $\chi$ does not diverge (and instead saturates toward this constant susceptibility) as $T \to 0$, implying that the system is away from the QCP. At the critical pressure $P_c \approx 2 \text{ GPa}$, however, the constant term vanishes to zero and $\chi$ diverges with the same critical index as the quasicrystal [6]. By comparing the critical behaviors with theories, the quantum criticality is likely ascribe to the critical valence fluctuation [6],[7]. At high pressures greater than $P_c$, the $\chi$ of the approximant shows a peak anomaly at a very low temperature $T_s$ ($\approx 100 \text{ mK}$) [6]. It is unclear whether the magnetic
ordered state below $T_g$ is an antiferromagnetically long-range ordered state or a spin-glass-like short-range ordered state. To elucidate the long-range or short-range ordered state, it is useful to measure the nonlinear susceptibility $\chi_2$ defined as follows,

$$\chi(H) = \frac{dM}{dH} = \chi_0 + \chi_2 H^2 + \cdots.$$  

Here, $M$ is a magnetization, and the $\chi_0$ and $\chi_2$ are the linear and nonlinear susceptibility, respectively. Figures 1(a) and (b) show the temperature dependence of $\chi_2$ around an antiferromagnetic ordering temperature $T_N$ (a) and a spin-glass freezing temperature $T_g$ (b) [8]. In the case of the antiferromagnetic order, $\chi_2$ shows no divergence and a sign change at $T_N$. In the case of the spin-glass transition, on the other hand, $\chi_2$ diverges at both sides of $T_g$. To determine the long-range or short-range ordering, it is also useful to measure the modulation frequency dependence of the transition temperature [9]. In the case of an antiferromagnetic order, $T_N$ is independent of frequency of the modulation field, while in the spin-glass order, $T_g$ increases with frequency. Here, we report the nonlinear magnetic susceptibility and the modulation-field frequency dependence of the transition temperature at high pressure.

![Figure 1](image.png)

**Figure 1.** Schematic illustration of the nonlinear susceptibility of an antiferromagnetic order (a) and a spin-glass-like short-range order (b) as a function of temperature. $T_N$ and $T_g$ indicate a Néel temperature and a freezing temperature, respectively.

### 2. Experimental details

The approximant crystal with the nominal composition of Au$_{49}$Al$_{36}$Yb$_{15}$ was prepared by arc-melting starting materials of 4N (99.99 % pure)-Au, 5N-Al, and 3N-Yb and subsequently annealing the obtained alloy-ingot in an evacuated quartz ampoule at 750 °C for 116 hours [10]. The ac magnetic susceptibility $\chi$ was measured using the conventional mutual inductance method. A modulation field (frequency in the range of 10.03 to 3003 Hz and amplitude of 0.1 Oe) was superimposed on a dc magnetic field $H$ supplied by a superconducting magnet. To calibrate the ac magnetic susceptibility, we measured the dc magnetization $M$ using a MPMS machine (Quantum Design) at pressures of up to 1.2 GPa, above which we extrapolated the dc magnetization data. Hydrostatic pressure was generated using a NiCrAl-BeCu piston cylinder cell for the measurement of the ac magnetic susceptibility, together with Daphne oil 7373 as a pressure-transmitting medium. The pressure at low temperature was determined from the superconducting transition temperature of indium [11] that was put into the pressure cell together with the sample.

### 3. Results and discussion

Figure 2 shows the temperature dependence of the ac magnetic susceptibility $\chi(T)$ at $P = 2.79$ GPa for $H = 0$. There appears an anomaly suggestive of a transition into either an
Figure 2. (Color online) Temperature dependence of the conventional susceptibility, $\chi (= \chi_0)$, (left axis) and the nonlinear susceptibility, $-\chi_2$, (right axis) of the $\text{Au}_{49}\text{Al}_{36}\text{Yb}_{15}$ approximant at $P = 2.79$ GPa.

antiferromagnetically long-range-ordered state or a spin-glass-like short-range-ordered state. The nonlinear susceptibility $\chi_2$ is evaluated as the initial slope of the $\chi$ vs $H^2$ curve. We assumed here that the $\chi$ is proportional to $H^2$ below 300 Oe, and estimated $\chi_2$ using the following equation,

$$\chi_2(T) = \frac{\chi(T, H) - \chi(T, 0)}{H^2}.$$  

Thus obtained results are also plotted in Fig. 2, together with $\chi_0$. Note that $-\chi_2$ forms a positive peak around $T_g$ at which the linear susceptibility peaks. This strongly suggests that the ordering is of short-range nature, which is compatible with the fact that there are only limited number of examples of the long-range ordered icosahedral-approximant crystal [12].

Figure 3 shows the temperature dependence of the ac magnetic susceptibility measured at several frequencies of the modulation field. We clearly observe a peak at low temperature although it is not obvious for 3003 Hz. The peak slightly but definitely shifts to higher temperatures as the frequency increases. This results are fit to the following equation (Vogel-Fulcher law),

$$f = f_0 \exp \left[ -\frac{W}{k_B(T_g - T_0)} \right].$$  

Here, $f_0$, $W$, and $k_B$ are the characteristic frequency, the activation energy, and the Boltzmann constant, respectively. $T_0$ is called the ‘ideal glass’ temperature although its physical meaning is unclear. From the results shown in Fig. 4, we obtain the following values: $f_0 = 10^{13}$ Hz, $W/k_B = 760 \pm 100$ mK, and $T_0 = 68 \pm 4$ mK. This finding gives a supporting evidence for the glass-like transition at $T_g$.

There are two possible origins of the quantum criticality in the approximant. One is due to the QCP of the glass transition. However, this possibility is presumably excluded because the uniform susceptibility will not diverge at $T_g = 0$. The second possibility is due to the critical valence fluctuation. As mentioned in the introduction part, this possibility remains most probable. For the quasicrystal, we have detected no magnetic ordering as observed in the
Figure 3. (Color online) Temperature dependence of the magnetic susceptibility of the Au-Al-Yb approximant at $P = 2.79$ GPa, measured at different applied frequencies $f = 10.03, 50.03, 100.3, 500.3, 1003$, and $3003$ Hz.

Figure 4. (Color online) The variation of the freezing temperature $T_g$ with $1/\ln(f_0/f)$, using $f_0 = 10^{13}$ Hz. The dash line is a guide to the eyes.

approximant. Therefore, the first possibility mentioned above is unambiguously excluded. The second possibility remains to be a possible origin.

4. Summary
We measured the ac magnetic susceptibility of the Au-Al-Yb approximant crystal down to about 80 mK under hydrostatic pressure. We found that the nonlinear magnetic susceptibility at 2.79 GPa shows a peak anomaly at $T_g \approx 100$ mK and $T_g$ increases with frequency of the modulation field, which results in the Vogel-Fulcher law. These results suggest that the magnetic ordered
state of the Au-Al-Yb approximant below $T_g$ is a spin-glass-like short-range ordered state.

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