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Field tuned critical fluctuations in $\text{YFe}_2\text{Al}_{10}$: Evidence from magnetization and ^{27}Al NMR and NQR investigations.

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We report magnetization, specific heat, and NMR investigations on $\text{YFe}_2\text{Al}_{10}$ over a wide range in temperature and magnetic field and zero field (NQR) measurements. Magnetic susceptibility, specific heat and spin-lattice relaxation rate divided by T ($1/T_1 T$) follow a weak power law ($\sim T^{-0.4}$) temperature dependence, which is a signature of critical fluctuations of Fe moments. The value of the Sommerfeld-Wilson ratio and linear relation between $1/T_1 T$ and χ suggest the existence of ferromagnetic correlations in this system. No magnetic ordering down to 50 mK in $C_p(T)$ and the unusual T and H scaling of the bulk and NMR data are associated with a magnetic instability which drives the system to quantum criticality. The magnetic properties of the system are tuned by field wherein ferromagnetic fluctuations are suppressed and a crossover from quantum critical to FL behavior is observed with increasing magnetic field.

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Critical fluctuations (CF) close to the absolute zero in temperature in materials as diverse as heavy-fermion systems¹⁻⁴, cuprates^{5,6}, and iron pnictides⁷ intimate the proximity to a quantum critical point (QCP). Whether the quantum critical point is underpinned by a magnetic phase transition or not depends on the location of the material on the generic phase diagram that connects a paramagnetic ground state with antiferromagnetic (AFM) order through an intervening QCP. The spatial dimensionality of the material has recently been demonstrated as an insightful approach in tuning the degree of fluctuations and the physics of quantum criticality⁸. Controlling a magnetic phase transition by means of a tuning parameter such as chemical composition, magnetic field (H) or pressure has proven to be a profitable route to QC^{4,9}. Non-Fermi-liquid (NFL) behavior of thermal properties is a hallmark of QC. The T and H scaling of magnetic systems are an insightful approach to unravel detail such as the universality of quantum phase transitions (QPT). For instance, unusual scaling in the resistivity $\rho(T) \sim T^n$ ($n < 2$), specific heat $C_p(T)/T \sim [-\ln T$ or $1 - \alpha T^{-n}$ ($n < 1$)] and magnetic susceptibility $\chi(T) \sim T^{-n}$ ($n < 1$) at low T are associated with QPT¹⁻⁴. A QPT typically causes a pile-up of entropy towards $T \rightarrow 0$. The NFL behavior in the local probe NMR leads to unusual power laws in spin-lattice relaxation rates (SLR) with $1/T_1 T \sim T^{-n}$ ($n < 2$) in the $T \rightarrow 0$ limit in contrast to simple local moment metals wherein $n=0$ is expected. From a theoretical point of departure, NFL behavior displaying these T scaling relations are native to instabilities in correlated electron systems, and spin fluctuations (SF) may also display similar NFL-like divergences. The concept of QC in $4f$ - and $3d$ -electron spin-density-wave systems in which an appropriate tuning parameter dissolves AFM order into a paramagnetic FL is by now well established^{1-4,10,11}. For

instance, in YbRh_2Si_2 , small fields suppress the AFM order and $1/T_1 T \sim T^{-0.5}$ is observed, which is due to a delicate interplay between FM and AFM spin fluctuations of local Yb^{3+} moments in the vicinity of a QCP^{12,13}. The significance of SF and their field dependence in itinerant FM like MnSi ¹⁴, $\text{Y}(\text{Co}_{1-x}\text{Al}_x)_2$ ¹⁵, ZrZn_2 ¹⁶, Ni_3AlC_x ¹⁷, $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$ ¹⁸, Sc_3In ¹⁹ and LaCoPO ^{20,21} have been explained by NMR in the framework of the self consistent renormalization (SCR) theory²². The notion of QC when the order parameter is FM on the other hand is controversial and currently a subject of debate. Recently, weak FM systems like UGe_2 ²³, MnSi ²⁴ and ZrZn_2 ²⁵ NbFe_2 ^{26,27} have been showing exciting features and it is observed that the magnetic field and/or pressure act as tuning parameters. Furthermore, unusual and weak power laws in $\chi(T)$ and C_p/T due to atomic disorder in a few itinerant and local moment systems are interpreted by quantum Griffith phase model²⁸⁻³⁷. The scaling behavior of observables especially in T and H offer a systematic approach towards establishing universality classes. In QC systems however our current understanding of the energy scaling that drives cooperative behavior offers very little commonality. The QC region of phase space appears to offer increasing layers of detail which complicates a consistent treatment of the QC region among different materials. The idea of universality still appears to be a distant target of our understanding of quantum criticality.

Ternary orthorhombic $4f$ -aluminides of $\text{RT}_2\text{Al}_{10}$ (R: Y, Yb, Ce; T: Fe, Ru, Os) type are currently attracting much interest because of a number of exotic properties such as unconventional structural and magnetic ordering in $\text{CeRu}_2\text{Al}_{10}$, metamagnetic transition as well as Kondo-insulating behavior in $\text{CeFe}_2\text{Al}_{10}$ ³⁸⁻⁴². In $\text{YFe}_2\text{Al}_{10}$, the most recently investigated member of this series, there is mounting evidence that this compound

is situated very close to a QCP of FM nature. The proximity to FM ordering is conveyed especially through an Arrott plot presentation of magnetization^{43,44}, but $\text{YFe}_2\text{Al}_{10}$ presents no unambiguous evidence of magnetic order above 50 mK.

In this communication, we present our investigations on magnetic susceptibility, specific heat, ^{27}Al NMR and NQR data in polycrystalline (PC) and single crystalline (SC) $\text{YFe}_2\text{Al}_{10}$. We have performed temperature and magnetic field dependent measurements in order to connect local magnetic dynamics through NMR and NQR with bulk thermal entropy and the consequences of FM correlations^{43,44}. Bulk data indicate that $\text{YFe}_2\text{Al}_{10}$ is in the vicinity of a magnetic instability^{43,44}. NMR investigations in the temperature range $1.8 \leq T \leq 300$ K and in the field range $0 \leq \mu_0 H \leq 7.27$ T on the PC sample have been carried out to provide better insights concerning the low energy spin excitations, and to contribute a deeper understanding of the effect of field on the correlated spin dynamics. Furthermore, we succeeded to perform ^{27}Al NQR measurements, which reflect the intrinsic magnetism of the system.

Details pertaining to the synthesis of PC and SC samples of $\text{YFe}_2\text{Al}_{10}$ are described elsewhere^{43,44}. The careful thermal and microprobe analysis inferred high quality and stoichiometric composition of the system without significant atomic disorder⁴⁵. We found no evidence for site exchange and only one Fe site is present in the host lattice⁴⁵. This poses a route towards understanding the QC in this system without complicating factors of atomic disorder whether accidental or whether induced as a consequence of doping when used to suppress FM order that is observed in other putative quantum critical systems. The magnetic susceptibility $\chi(T)$ is measured in different H in the T range $1.8 \leq T \leq 300$ K in PC samples using a Quantum Design (QD), MPMS SQUID magnetometer. As shown in Fig.1(a), the low field $\chi(T)$ increases upon lowering T , and $\chi(T)$ tends to saturate below 5 K in high magnetic fields. Above 50 K, $\chi(T)$ obeys a Curie-Weiss behavior (see Fig. 1b) and the effective moment ($\mu_{eff} = 0.52 \mu_B/\text{f.u}$) is rather small. The low value of saturation moment ($\mu_s \sim 0.02 \mu_B/\text{f.u}$) leads to $\mu_{eff}/\mu_s \approx 26$, which classifies $\text{YFe}_2\text{Al}_{10}$ being a weak itinerant FM in the Rhodes-Wolfrath plot^{46,47}. The enhancement of ac and dc magnetic susceptibility accompanied by $\chi(T) \sim T^{-0.5}$ divergence below 10 K in weak magnetic fields could be due to exchange enhanced $q=0$ excitations and suggesting the presence of spin correlations and spin fluctuations of Fe moments, which are suppressed with high magnetic fields. However, this power law in $\chi(T)$ is different from that observed in the SC, which can be associated with some sort of powder averaging in the PC sample⁴⁴. The magnetization isotherm at 2 K displays $H^{0.45}$ (as shown in Fig. 1c inset), which implying critical spin fluctuations. The M vs. H follows a mean field type M^2 vs H/M scaling (Fig. 1c), suggesting $T \rightarrow 0$ FM ordering. The nature of the Arrott plot is attributed to the zero-point spin fluctuations of

FIG. 1: (Color online). (a) The temperature dependence of $\chi(T)$ in different magnetic fields. The solid line displays the $T^{-0.5}$ behavior. (b) The T dependence of $1/\chi(T)$ with C-W fit (c) Arrott plot of the magnetization. The inset show M vs. H at 2 K with a fit to $H^{0.45}$.

Fe moment close to QCP, which are suppressed with H .

Specific heat on the PC sample measured in zero field down to 50 mK, using the dilution refrigerator and in various fields down to 0.35 K in the ^3He option of QD PPMS is shown in Fig. 4(a). The zero field specific heat coefficient $\gamma = C_p/T$ at low T is enhanced and follow a $T^{-0.35}$ behavior, while the C_p/T in 7 T is suppressed and independent of T at low temperature (see Fig. 4a). The low temperature C_p/T in 7 T is corrected for the nuclear Schottky contributions arising mainly from ^{27}Al ⁴⁸. The power law divergence of γ in zero field indicating quasi-2D ferromagnetic SF of Fe moments at $H=0$. It is noteworthy that in $\text{YFe}_2\text{Al}_{10}$, Fe atoms are stacked in 2D planes perpendicular to the b -direction of the orthorhombic crystal structure. Further to this point, we also draw attention to the fact that SF in $\text{YFe}_2\text{Al}_{10}$ reside largely within the ac -plane⁴⁴. The Sommerfeld coefficient, γ is enhanced (~ 14.5 mJ/mol Fe K²) compared to $\gamma \sim 3$ mJ/mol Ru K² in the isostructural $\text{YRu}_2\text{Al}_{10}$ ⁴⁴ indicating quasi-particle mass enhancement and suggests that the system is driven from a critical regime to a FL state with field. The value of the Sommerfeld-Wilson ratio $R_w = \pi^2 k_B^2 / \mu_0 \mu_{eff} (\chi/\gamma)$ (≈ 7 at 2 K) is relatively large implying the presence of FM correlations. These findings could be ascribed to the effect of SF in this weak itinerant ferromagnet wherein SF modulate the thermodynamical observables greatly²². The low T increase of C_p/T could be associated with the virtual scattering of electrons on the Fermi surface via $q=0$ excitations and CF of spins implying an enhancement of the effective mass of electrons^{4,9}. The exchange enhancement and mass enhancement are attributed to a FM instability in the close proximity of a QCP²². The specific heat in a

FIG. 2: (Color online). (a) Field sweep NMR PC and SC spectra ($H \perp b$) with simulation at 80 MHz. The inset shows the NQR spectra ($\pm 3/2 \longleftrightarrow \pm 5/2$) on PC sample at different T .

metal is related to the density of states, which seems to be modified around the QCP leading to unusual scaling laws in T and H which is substantiated in NMR and NQR data as discussed in the following sections²².

Shown in Fig. 2 are the field sweep NMR spectra of PC together with SC ($H \perp b$) samples at 80 MHz at 4.2 K. The presence of five inequivalent Al sites yields very complex NMR spectra. Each Al site sense different quadrupole couplings which makes the powder spectra rather broad. The low frequency spectra broaden inhomogeneously which originates from the broad distributions of H -independent quadrupole interactions admixed with the H -dependent anisotropic Zeeman interactions. The quadrupole Hamiltonian is given as: $H_Q = h\nu_q/6[(3I_z^2 - I^2) + \eta/2(I_x^2 - I_y^2)]$, where $\eta = (V_{xx} - V_{yy})/V_{zz}$ is the asymmetry parameter, $\nu_q = 3eQV_{zz}/20h$ is the NQR frequency, V_{zz} is the principal component of EFG tensor, which arises from the local charge distribution. The system presented here has five Al sites resulting in ten NQR lines since each Al site has two NQR lines viz., $\pm 1/2 \longleftrightarrow \pm 3/2$; $\pm 3/2 \longleftrightarrow \pm 5/2$ transitions. The SC NMR spectra exhibit more pronounced (first order) quadrupolar transitions with lines corresponding to the aforementioned pair of transitions. The central transition is rather broad for a SC but we interpret this as being due to strong correlations that are prevalent. To simplify the present spectra with five different Al sites (and five different quadrupole coupling constants, ν_q) we simulated NMR spectra with Al site corresponding to high frequency NMR line. This simulation yields $\nu_q = 1.6$ MHz which corresponds to the fraction of Al site having the highest quadrupolar interactions³⁸. For these Al sites, NQR lines are expected at $\nu_{Q1} = \nu_q$ ($\pm 3/2 \longleftrightarrow \pm 1/2$) and $\nu_{Q2} = 2\nu_q$ ($\pm 5/2 \longleftrightarrow \pm 3/2$). We therefore performed NQR at 3.2 MHz and we indeed ob-

served a relatively narrow NQR line (see Fig. 2. inset) which is in good agreement with our field sweep NMR results. Above 10 K, the NQR line develops singularities which are symptomatic of multiple quadrupolar transitions. Moreover, the well resolved NQR lines enable us to probe the low temperature magnetic fluctuations via SLR measurements in $H=0$. Because of the relatively weak T and H dependence of the line width and the absence of a sizable shift of the NMR spectra down to 1.8 K, spin lattice relaxation time measurements could reliably be performed over a wide T and H range of our investigation and the analysis of which is discussed in the following sections. Almost no shift in NMR and NQR spectra could be attributed to the symmetrical position of Fe with respect to Al nuclei and very weak hyperfine fields resulting in cancellation of transverse hyperfine fields via ²⁷Al. The line broadening at low T is associated with fluctuations of the Fe moments, and consequent strong magnetic correlations, which are in agreement with macroscopic data^{22,49}.

²⁷Al spin lattice relaxation time measurements have been performed by exciting the central transition (which is not affected by the quadrupole interactions) following saturation recovery method with suitable rf pulses in different magnetic fields. The recovery of longitudinal magnetization, $M(t)$, at a time delay t after the saturation pulse in all T and H ranges could consistently be fitted with a single component valid for $I=5/2$ nuclei $1-M(t)/M(\infty) = 0.0291e^{-t/T_1} + 0.178e^{-6t/T_1} + 0.794e^{-15t/T_1}$, where $M(\infty)$ is the equilibrium magnetization⁵⁰. The fit of $M(t)$ to the single component suggests the uniform distribution of Fe moments in the host lattice and the corresponding SLR results are shown in Fig.3. $1/T_1$ decreases with decreasing T and shows linear behavior with T in high magnetic fields, which is attributed to the metallic nature of the system due to the effect of non interacting conduction electrons. This leads to T -independent $1/T_1 T$ behavior in high H , and the system is in the FL state. A cross-over from Korringa to quantum critical behavior in $1/T_1 T$ is observed in weak magnetic field. As shown in Fig. 3(a), $1/T_1 T$ increases monotonically and follows $T^{-0.4}$ behavior below 100 K evidencing the persistence of CF of weak itinerant moments in weak magnetic field^{15,20}. This CF of Fe moments in weak magnetic fields is a consequence of the magnetic instability that is caused by hybridization between nearly filled d -electron shells and conduction electrons. On a first approach, this power law divergence in the spin lattice relaxation rate suggest a fragile interplay between FM and AFM components of spin fluctuations¹³.

Spin lattice relaxation rate in principle probes the q -averaged low energy spin excitations and it can be expressed as the wave-vector q summation of the imaginary part of the dynamic electron spin susceptibility

FIG. 3: (Color online). (a) The T dependence of $1/T_1 T$ in various magnetic fields and the solid line is a fit to $T^{-0.4}$ ($1/T_1 T$ values are scaled for better clarity) (b) The linear relation between $1/T_1 T$ and χ with T as an implicit parameter.

$\chi''(q, \omega_n)^{51}$:

$$\frac{1}{T_1 T} = \frac{2(27\gamma_n)^2}{N_A^2} k_B \sum_q |A_{hf}(q)|^2 \frac{\chi''(q, \omega_n)}{\omega_n}$$

where $A_{hf}(q)$ is the hyperfine form factor.

The SLR is linear with $\chi(T)$ in weak magnetic fields (Fig. 3b), which evidence the dominant role of FM spin fluctuations^{18,22,52–54}. It is worth to mention that $1/T_1 T \sim \chi(T)$ behavior represents the fact that $\chi(T)$ is the uniform spin susceptibility $\chi(q=0, \omega=0)$ and accounts for the dominant FM component of the static spin susceptibility⁵¹. $1/T_1 T$ reflects the dynamic SF which are not related to bulk effects, and the suppression of $1/T_1 T$ with field might be attributed to the fact that the relaxation mechanism is governed by the intrinsic magnetism of the system. The FM correlations develop between Fe moments, and the fluctuation of local hyperfine fields of electronic states with respect to ^{27}Al nuclei offers a channel for the relaxation mechanism at the FM wave vector $q=0$. Modes of SF's are localized in reciprocal space with small amplitude in $\text{YFe}_2\text{Al}_{10}$ leading to unusual power law in SLR, which infer that the system is in the vicinity of a QCP^{13,22}. NMR investigations on the SC are limited due to the poor signal to noise ratio in the available tiny SC and skin depth problems. Nonetheless, the values of T_1 in SC are found to be close to those in the PC and infer the absence of strong anisotropy in T_1 for both crystallographic directions $H \perp b$ and $H \parallel b$.

NQR spin lattice relaxation times are measured for the $\pm 3/2 \longleftrightarrow \pm 5/2$ transitions following the saturation recovery method for the $M(t)$. T_1 is determined by fitting the recovery of $M(t)^{50,55}$: $1 - M(t)/M(\infty) = 0.427e^{-3t/T_1} + 0.573e^{-10t/T_1}$. Zero field (NQR) $1/T_1 T$ is associated with intrinsic CF, the T

FIG. 4: (Color online). Temperature dependence of C_p/T and the solid line a fit to $T^{-0.35}$ (b) Field dependence of $1/T_1$ at 2 K with a fit as discussed in the text (c) $1/T_1 T$ vs T at 0.545 and 7.27 T.

dependence of which follow the same power law as in the case of low field $1/T_1 T$ data as shown in Fig. 3(a).

As mentioned previously, the close proximity of this system to a QCP could lead to unusual behavior of bulk and NMR data that are connected with magnetic interactions and collective excitations. Interestingly, the observation of same power law in $1/T_1 T$ and C_p/T is not unusual as these type of features also observed in a few systems^{10,30,31,56}. We rule out the possibility of Griffith phase in $\text{YFe}_2\text{Al}_{10}$, because of the absence of significant atomic disorder⁴⁵. It may be noted that the observed CF in χ , C_p/T and $1/T_1 T$ are suppressed with field suggesting the suppression of low lying spin excitations in the SF spectra. A magnetic field of 7 T is strong enough to dampen the fluctuations and diminishing the spin entropy towards a FL phase in $\text{YFe}_2\text{Al}_{10}$ ^{57–59}. The field dependence of the spin lattice relaxation rate at 2 K follows a dynamic scaling $1/T_1(H) = 1/T_1(0)[1 + (\mu_B H/k_B T)^2]^{-\xi}$, $\xi=0.74$ (see Fig. 4b). This scaling behavior is ascribed to the dominant role of the magnetic field in tuning the CF of spins in inducing the system to a FL phase^{60–63}. Here, the magnetic field accounts for the renormalization of collective excitations and suppresses the correlation effects in $\text{YFe}_2\text{Al}_{10}$, leading to suppression of spin fluctuations^{60–63}.

To summarize, the divergences observed in $\chi(T)$ and C_p/T of $\text{YFe}_2\text{Al}_{10}$ are consistently attributed to the quantum CF of Fe moments and support $T \rightarrow 0$ phase transition. Absence of magnetic ordering down to 50 mK and low T and H divergences in χ , C_p/T and $1/T_1 T$ can not be interpreted in terms of the standard theory predicted for disordered or NFL model. Bulk and NMR data are consistent with the persistence of FM correlations and infer the dominant role of $q=0$ components of

SF and CF in the spin excitation spectra in tiny magnetic fields. The observed static and dynamic magnetic properties are associated with a nearly continuous magnetic phase transition and the system seems to be a d -electron analogue of $4f$ -electron heavy fermions. The CF which are of FM nature in $\text{YFe}_2\text{Al}_{10}$ are suppressed and a cross over from a QC regime to a FL state is observed, and the magnetic field is concluded to play the significant role in driving such a phase transition. Further investigations concerning the effect of chemical pressure and SLR measurements at low temperature are highly desirable. Neutron scattering experiments are required to address the nature of spin fluctuation spectra along various q vectors.

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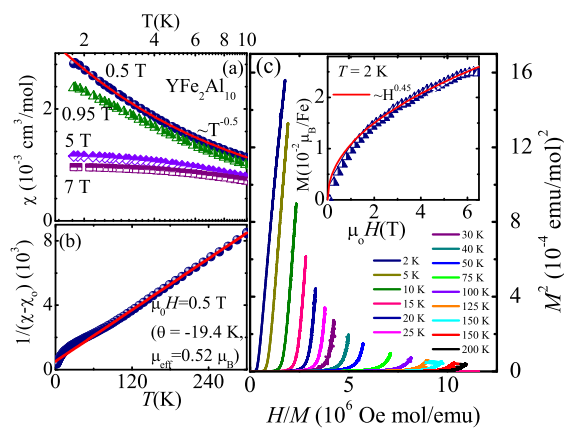


Figure 1

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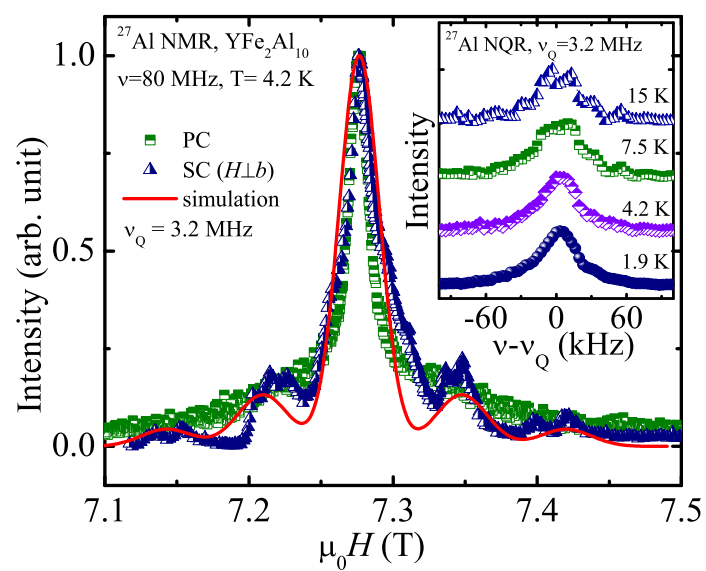


Figure 2

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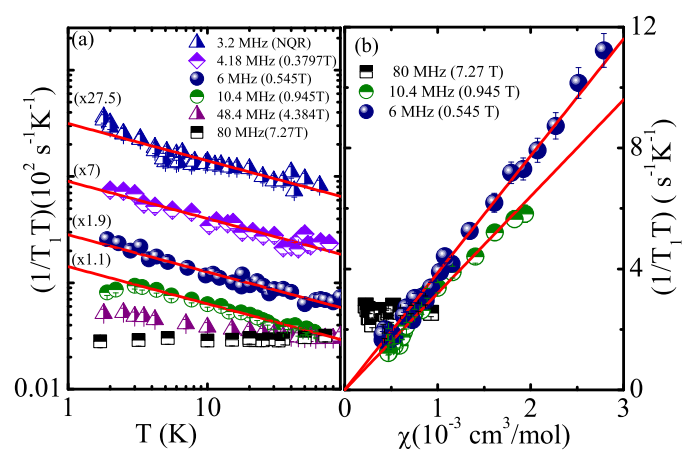


Figure 3

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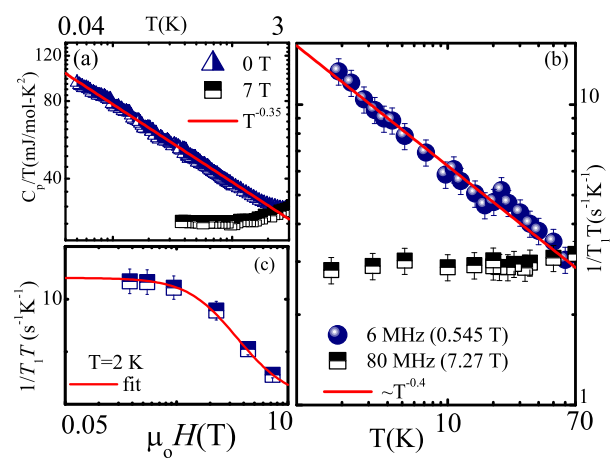


Figure 4

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