Tuning the Distance to a Possible Ferromagnetic Quantum Critical Point in $A_2Cr_3As_3$

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Although superconductivity in the vicinity of an antiferromagnetic (AFM) instability has been extensively explored in the last three decades or so, superconductivity in compounds with a background of ferromagnetic (FM) spin fluctuations is still rare. We report ^{75}As nuclear quadrupole resonance measurements on the $A_2Cr_3As_3$ family, which is the first group of Cr-based superconductors at ambient pressure, with A being alkali elements. From the temperature dependence of the spin-lattice relaxation rate ($1/T_1$), we find that by changing A in the order of $A = $ Na, $Na_{0.75}K_{0.25}$, K, and Rb, the system is tuned to approach a possible FM quantum critical point (QCP). This may be ascribed to the Cr2-As2-Cr2 bond angle that decreases towards 90°, which enhances the FM interaction via the Cr2-As2-Cr2 path. Upon moving away from the QCP, the superconducting transition temperature T_{sc} increases progressively up to 8.0 K in $Na_2Cr_3As_3$, which is in sharp contrast to the AFM case where T_{sc} usually shows a maximum around a QCP. The $1/T_1$ decreases rapidly below T_{sc} with no Hebel-Slichter peak, and ubiquitously follows a T^5 variation below a characteristic temperature $T^* \approx 0.6\ T_{sc}$, which indicates the existence of point nodes in the superconducting gap function commonly in the family. These results suggest that the $A_2Cr_3As_3$ family is a possible solid-state analog of superfluid ^{3}He.

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The interplay between magnetism and superconductivity is a key topic in condensed matter physics. In the past thirty years or so, a large amount of superconductors in proximity to an antiferromagnetic (AFM) ordered phase have been found. In heavy fermions [1,2], cuprates [3], and iron pnictides [4] superconductivity appears on the verge of antiferromagnetic instability, and the critical temperature T_{sc} usually takes a maximum at the AFM quantum critical point (QCP). However, superconductivity in the vicinity of a ferromagnetic ordered phase is still rare. UGe_2 is a ferromagnet, but becomes superconducting under high pressure when the Curie temperature T_C is reduced inside the ferromagnetic phase [5]. However, superconductivity has not been found when the ferromagnetic order is completely destroyed.

Ferromagnetic interactions can also promote quantum states other than superconductivity. For example, superfluidity in ^{3}He emerges in the background of ferromagnetic spin fluctuations. There are two phases, namely, the A phase and B phase, in ^{3}He [6–9]. The A phase is a p-wave ABM (Anderson-Brinkman-Morel) state with equal spin pairing ($\uparrow\uparrow$ and $\downarrow\downarrow$) [7,8]. The B phase is a p-wave BW (Balian-Werthamer) state with an additional component $1/\sqrt{2}(\uparrow\downarrow + \downarrow\uparrow)$ [9]. There are point nodes in the gap function in the ABM state but the gap is isotropic in the BW state [10]. It has been pointed out that the spin triplet pairing in ^{3}He is induced by FM spin fluctuations [8]. Notably, ^{3}He phases are topologically nontrivial, which has received new and intensive interest in the past few years [11]. The B phase of ^{3}He belongs to the so-called DIII topological class [12], and the A phase bares similarities to topological Weyl semimetals. Therefore, searching for a solid state analog of ^{3}He serves to bridge three large research areas: strong correlations, unconventional superconductivity, and topological quantum phenomena.

Recently, a 3d-electron system, chromium-based superconductors $A_2Cr_3As_3$ ($A = $ Na, K, Rb, Cs), has been discovered [13–16]. In this family, superconductivity emerges from a paramagnetic state. $K_2Cr_3As_3$ and $Rb_2Cr_3As_3$ have a $T_{sc} = 6.1$ and 4.8 K [13,14], respectively, and $Na_2Cr_3As_3$ has the highest $T_{sc} = 8.0$ K. Resistivity measurement suggests that $Cs_2Cr_3As_3$ superconducts below $T = 2.2$ K [15]. Although anomaly at $T = 2.2$ K was not found by nuclear quadrupole resonance (NQR) [17,18], magnetic susceptibility measurement does confirm bulk superconductivity below $T = 1.2$ K [19]. Density function theory (DFT) calculations show that there are three bands across the Fermi level, namely, two quasi one-dimensional (1D) bands α and β, and one three-dimensional (3D) band γ [20,21]. The γ band makes the main contribution to the density of states (DOS).

NQR, penetration depth, muon spin rotation (μSR), upper critical field H_{c2}, and specific heat measurements show signatures of unconventional superconductivity [22–29]. Theoretically, a spin-triplet superconducting state has been proposed [30–32]. In the normal state, ferromagnetic spin fluctuations have been found from the Knight shift and spin-lattice relaxation rate $(1/T_1)$ measurements in $Rb_2Cr_3As_3$ [23]. Neutron scattering measurements also suggest short-range magnetic order in $K_2Cr_3As_3$ [33]. However, how the spin fluctuations evolve with changing A is unknown. In addition, the gap symmetry is still controversial. For example, $1/T_1 \propto T^4$ was reported in $K_2Cr_3As_3$ [22], but $1/T_1 \propto T^5$ was found in $Rb_2Cr_3As_3$ [23]. The latter result suggests point nodes in the gap function [23]. On the other hand, line nodes were claimed by penetration depth and specific heat measurements [24,27].

In this work, we systematically study the normal and superconducting states of the family $A_2Cr_3As_3$ by NQR. We find that $1/T_1T$ in the normal state for all compounds can be fitted by Moriya's theory for 3D FM spin fluctuations [34]. Our results show that on going from A = Na to $Na_{0.75}K_{0.25}$, K, Rb, the system progressively approaches a possible FM QCP, which may be ascribed to the Cr2-As2-Cr2 bond angle that decreases toward 90° in the same order and enhances the FM interaction. In the superconducting state, $1/T_1$ for all compounds show no Hebel-Slichter peak, and follows a T^5 variation below a characteristic temperature $T^* \approx 0.6T_{sc}$, indicating a common formation of point nodes in the gap function. Our results indicate that $A_2Cr_3As_3$ shows some properties similar to superfluid ^{3}He.

Polycrystal sample of $Na_2Cr_3As_3$ was prepared by a low temperature ion-exchange method [16], and the others were prepared by solid state reaction method [13]. We crush the samples into powders or cut them into pieces to avoid skin depth problems in the NQR measurements. To protect the samples against air and water vapor, we seal the samples into an epoxy (stycast 1266) tube in an Ar-filled glove box. The T_1 was measured by using the saturation-recovery method, and obtained by a good fitting [23] of the nuclear magnetization to $1 - M(t)/M_0 = \exp(-3t/T_1)$, where $M(t)$ is the nuclear magnetization at time t after the single saturation pulse and M_0 is the nuclear magnetization at thermal equilibrium.

Figures 1(a) and 1(b) show the crystal structure of the system and the NQR spectra, respectively. Similar to $K_2Cr_3As_3$ [22] and $Rb_2Cr_3As_3$ [23], the NQR spectra of $(Na_{0.75}K_{0.25})_2Cr_3As_3$ and $Na_2Cr_3As_3$ also have two peaks originating from two inequivalent As sites. Since the temperature dependence of $1/T_1$ for two As sites is the same [22,23], we measured $1/T_1$ for A = Na, $Na_{0.75}K_{0.25}$, and K at the stronger peak.

As can been seen in Fig. 2, $1/T_1T$ is a constant above $T \sim 150$ K and increases with decreasing temperature

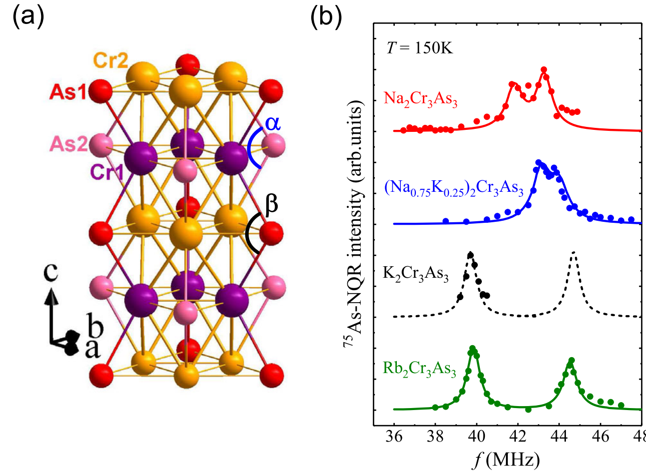

FIG. 1. (a) The structure of the $[Cr_3As_3]_\infty$ tube, the Cr2-As2-Cr2 bond angle (α), and the Cr1-As1-Cr1 bond angle (β). (b) ^{75}As NQR spectra of $A_2Cr_3As_3$ (A = Na, $Na_{0.75}K_{0.25}$, K, Rb) measured at $T = 150$ K. Solid lines are fitting results by two Lorentzian functions. The dashed curve represents data of Ref. [22].

down to T_{sc} for all samples. For a conventional noninteracting metal, $1/T_1T$ is a constant. Therefore, the results indicate electron correlations. Previous Knight shift measurements in $Rb_2Cr_3As_3$ found that spin susceptibility increases with decreasing temperature, which indicates that the electron correlation is ferromagnetic in character. DFT calculations also show that the interaction within each Cr sublattice is ferromagnetic [20,21,30]. Below we apply the 3D ferromagnetic spin fluctuations theory of Moriya to

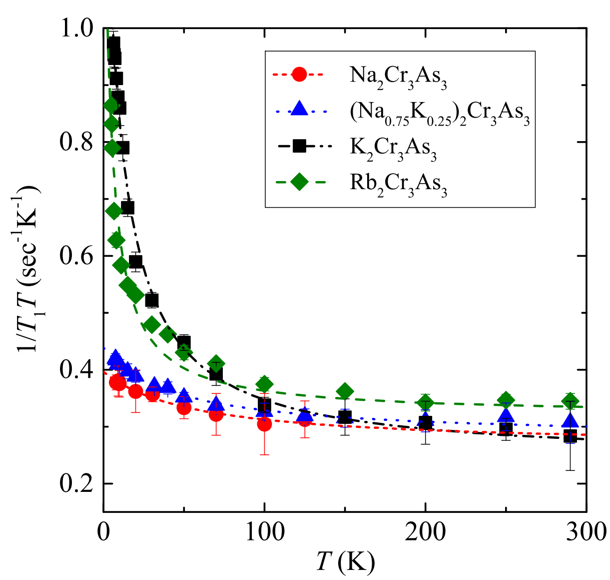

FIG. 2. ^{75}As nuclear spin-lattice relaxation rate $1/T_1$ divided by temperature. Data for $Rb_2Cr_3As_3$ are from Ref. [23]. The dashed curves on the normal state data are fittings to $1/T_1T = a + b/(T + \theta)$.

characterize the spin fluctuations [34]. The $1/T_1T$ can be expressed as $1/T_1T = (1/T_1T)_{SF} + (1/T_1T)_0$. The first part originates from spin fluctuations of 3d electrons, and the second part is due to noninteracting electrons. For 3D ferromagnetic spin fluctuations [34], $(1/T_1T)_{SF}$ follows a relation of $C/(T+\theta)$, with the parameter θ describing a distance to FM QCP. The obtained θ decreases in the order of $A =$ Na, $Na_{0.75}K_{0.25}$, K, and Rb. The parameter $\theta = 4 \pm 1.5$ K for $Rb_2Cr_3As_3$ and $\theta = 57 \pm 7$ K for $Na_2Cr_3As_3$.

Figure 3 shows the phase diagram of the family, where θ and T_{sc} are plotted as a function of the ionic radius of alkali element A. As the ionic radius increases, the parameter θ decreases, indicating that the system is tuned closer to a possible FM QCP. Concomitantly, T_{sc} decreases. Below we show that the transverse axis of Fig. 3 correlates with ferromagnetic interaction strength. The larger the ionic radius is, the stronger the ferromagnetic interaction is.

As shown in Fig. 4, analysis of the available data shows that the Cr2-As2-Cr2 bond angle α has a linear relationship with the ionic radius, while the Cr1-As1-Cr1 bond angle β is almost a constant. Notably, as the ionic radius increases, α decreases towards 90°. It was pointed out that the Cr-Cr coupling would be antiferromagnetic according to the Goodenough-Kanamori-Anderson rule [30]. However, since Cr is in an averaged valence of 2.3, double exchange through the Cr-As-Cr path is possible [30], which is ferromagnetic. At $\alpha = 90^\circ$, the As-4p_x and As-4p_y orbitals become degenerated with respect to Cr-3d orbitals, which will maximize the double exchange interaction between the two Cr2 along the c axis via the As-4p_x and As-4p_y orbitals. Therefore, on going from $A =$ Na to $Na_{0.75}K_{0.25}$, K, and Rb, an increase in the ferromagnetic interaction can be expected, which drives the system towards a FM QCP. In order to directly access such QCP, we propose to replace A with Ca, Sr, or Ba, thereby a long-range ordered phase can hopefully be obtained.

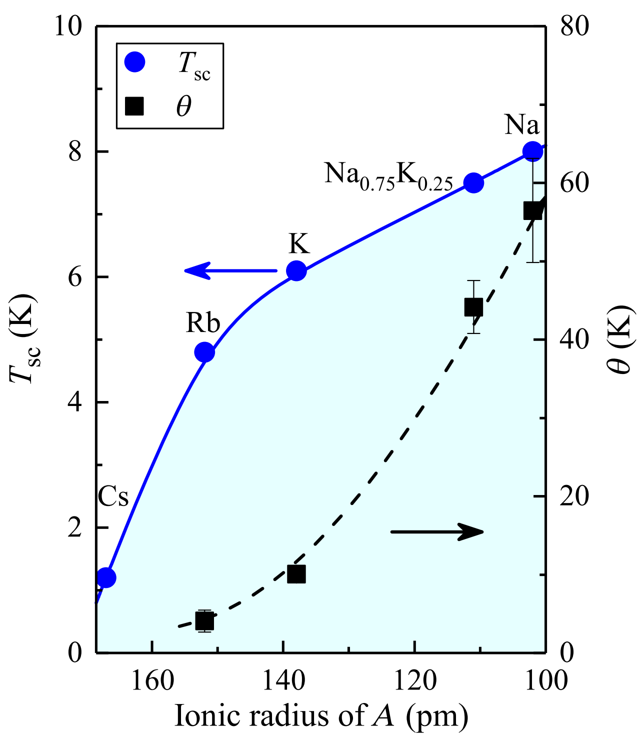

FIG. 3. Phase diagram of $A_2Cr_3As_3$ ($A =$ Na, $Na_{0.75}K_{0.25}$, K, Rb, Cs). The data of alkali ions with six-coordinations were taken from Ref. [35]. The filled squares are θ from the fitting of $1/T_1T$ data (see text).

In Fig. 3, one sees that T_{sc} decreases upon approaching the FM QCP. This is in sharp contrast to the AFM case where T_{sc} forms a peak around a QCP. Superconductivity near a FM QCP in the paramagnetic side was discussed by Fay and Appel in their seminal work [37], but to our knowledge, had not been confirmed thus far. In the AFM case, the pairing interaction is enhanced due to increased quantum fluctuations [38]. In the FM case, when it is approached from the paramagnetic side, increased quantum fluctuations also enhances pairing strength [37,38]. However, Fay and Appel found, based on a random phase approximation, that mass enhancement due to FM spin fluctuations will kill a spin-triplet superconducting state so that T_{sc} is zero right at ferromagnetic QCP [37]. Later on, Monthoux and Lonzarich [38] and Wang *et al.* [39] calculated T_{sc} in the strong-coupling limit. They pointed out that mass enhancement and a finite quasiparticle lifetime act as pair breaking, but found a nonzero T_{sc} at ferromagnetic QCP. In UGe_2, however, no T_{sc} was found in the paramagnetic side. In a related compound UCoGe [40], although superconductivity survives after ferromagnetic order is suppressed, the ferromagnetic-paramagnetic transition is a first-order phase transition [41] and thus cannot

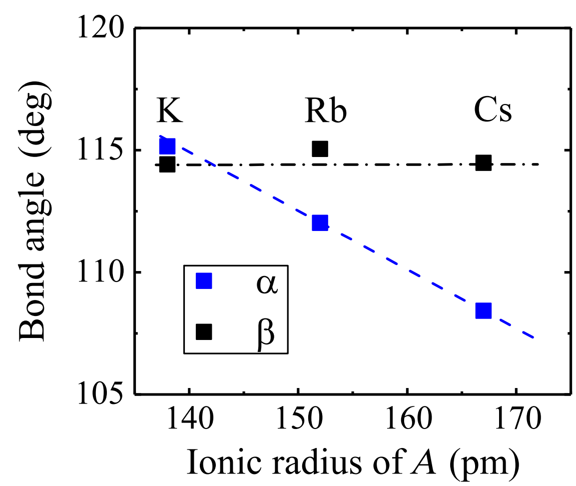

FIG. 4. Correlation between the ionic radius and the Cr2-As2-Cr2 bond angle α and Cr1-As1-Cr1 bond angle (β) for $Cs_2Cr_3As_3$, $Rb_2Cr_3As_3$, and $K_2Cr_3As_3$. The angles were calculated based on the available crystal-structure data in the literature [13,15,36].

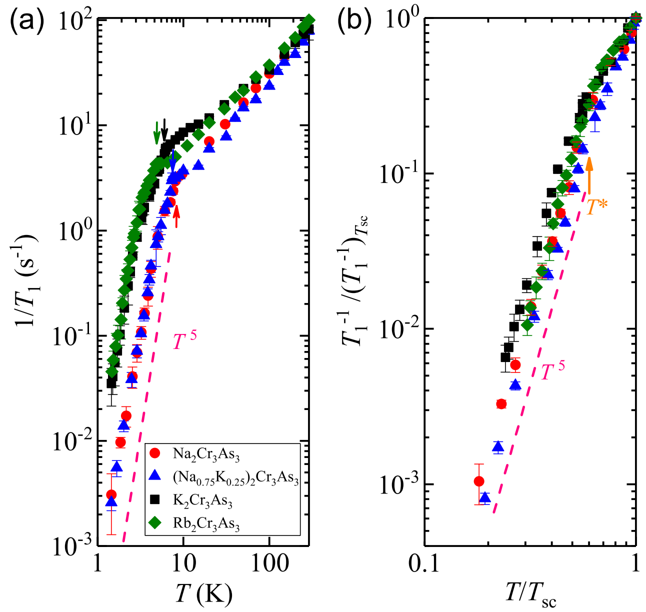

FIG. 5. (a) $1/T_1$ as a function of T for all samples. The arrows indicate T_{sc} of each compound. (b) $1/T_1$ normalized by its value at T_{sc}. The arrow indicates a characteristic temperature T^*, below which $1/T_1$ becomes proportional to T^5. The symbols are the same as (a).

be directly compared to theories. Our results experimentally demonstrate [42], for the first time, the evolution of T_{sc} in the paramagnetic side predicted by theories [37,38]. Looking forward, it would be interesting to experimentally probe the evolution of effective electron mass by London penetration depth at the zero-temperature limit [43], for example.

Next, we discuss the properties of the superconducting state. In Fig. 5(a) are shown the $1/T_1$ data for the four compounds. There is no coherence peak for all cases. In contrast to the previous report for a $K_2Cr_3As_3$ sample with a lower $T_{sc} = 5.7$ K than our case (6.1 K) [22,42], our data show that $K_2Cr_3As_3$ exhibits a temperature variation very similar to $Rb_2Cr_3As_3$. In Fig. 5(b), we show the data with reduced scales for the axes. As can be seen there, $1/T_1$ commonly shows a T^5 behavior below a characteristic $T^*/T_{sc} \approx 0.6$. This suggests that the gap symmetry is the same for all members of this family.

The $1/T_1$ in superconducting state can be expressed as

$$\frac{(T_1)_{T_{sc}}}{T_1} = \frac{2}{k_B T}\int N_s(E)^2\left(1+\frac{\Delta^2}{E^2}\right)f(E)[1-f(E)]dE, \tag{1}$$

where $N_s(E)$ is the DOS below T_{sc}, $f(E)$ is Fermi distribution function, Δ is the gap function. When there exist point nodes in the gap function, such as in the ABM model [7,8] with $\Delta = \Delta_0 \sin\theta e^{i\phi}$, $N_s(E) \propto E^2$. This results in $1/T_1 \propto T^5$ at low temperatures. Previously, Katayama *et al.* found such a T^5 variation in filled skutterudite superconductor $PrOs_4Sb_{12}$ under pressure [44]. Notably, $1/T_1$ does not deviate from T^5 in the lowest temperature even for $(Na_{0.75}K_{0.25})_2Cr_3As_3$, where partial substitution of K for Na would induce disorder. Generally, disorders or impurities cause finite DOS at the Fermi level in the case of a nodal gap, which will result in a deviation of $1/T_1$, which has indeed been observed in cuprates [45]. The current result therefore indicates that the superconducting state is not affected by the disorder out of $[Cr_3As_3]_\infty$ tube. Finally, the existence of T^* is unclear at the moment. A possible reason is multiple-band superconductivity, as seen in iron-based superconductors [46]. Another possibility is multiple phases arising from internal freedoms associated with spin-triplet pairing as seen in UPt_3 [47]. Clearly, more work is needed in this regard.

In summary, we have performed ^{75}As-NQR measurements on the $A_2Cr_3As_3$ family. we find that by changing A in the order of $A =$ Na, $Na_{0.75}K_{0.25}$, K, and Rb, the system is tuned to approach a possible ferromagnetic QCP. We propose that the Cr2-As2-Cr2 bond angle (α) that decreases towards 90° is responsible for the increase of ferromagnetic interaction. Upon approaching the QCP, the superconducting critical temperature T_{sc} decreases, which is in sharp contrast to the AFM case where T_{sc} usually forms a broad peak around a QCP. In the superconducting state, $1/T_1$ decreases with no Hebel-Slichter peak just below T_{sc}, and ubiquitously follows a T^5 variation at low temperatures, which indicates the existence of point nodes in the gap function commonly in the whole family. Our results indicate that the $A_2Cr_3As_3$ family is a possible solid-state analog of superfluid ^{3}He. Therefore, further investigations on this family promise to enrich the physics across multiple research areas of electron correlations, unconventional superconductivity, and topological quantum phenomena.

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SUPPLEMENTAL MATERIALS

for

Tuning the distance to a possible ferromagnetic quantum critical point in $A_2Cr_3As_3$

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1. Phase diagram of $A_2Cr_3As_3$ compared with UGe2 and theories

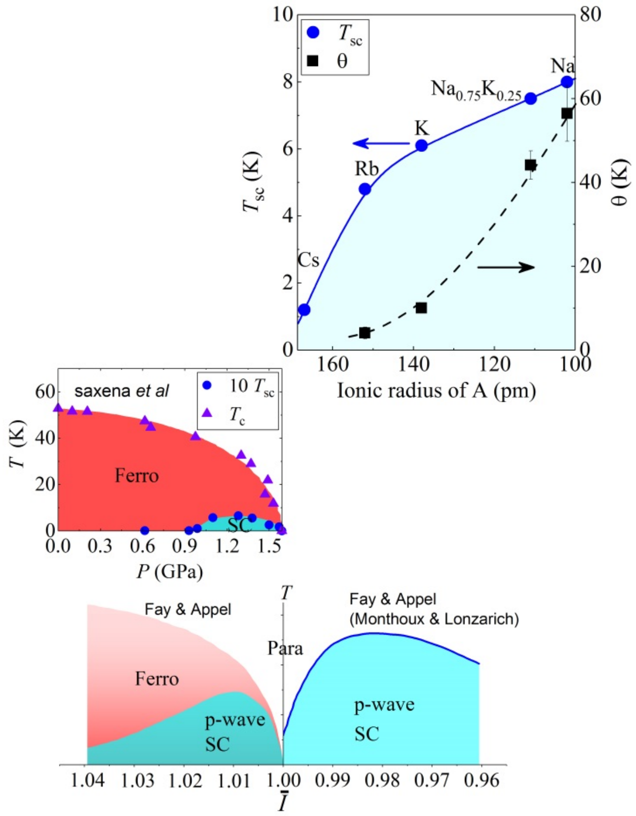

FIG.S1. (a) Phase diagram of $A_2Cr_3As_3$ (A = Na, $Na_{0.75}K_{0.25}$, K, Rb, Cs). (b) Phase diagram of UGe_2 under pressure [1]. "Ferro" denotes ferromagnetic ordered state. (c) Phase diagram proposed by Fay and Appel [2] and Monthoux and Lonzarich [3]. The horizontal axis is represented by the exchange-interaction parameter $\bar{I}$, which is 1 at FM QCP.

2. Lattice parameters of $A_2Cr_3As_3$

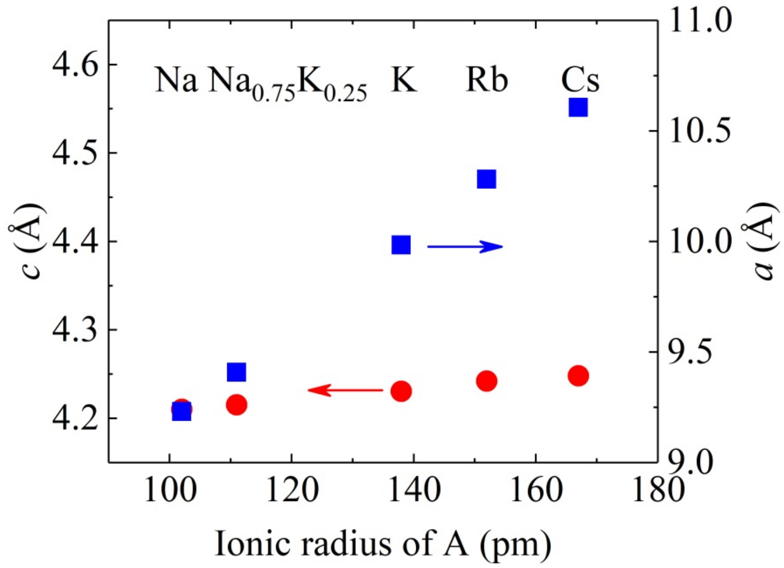

FIG.S2. Lattice parameters of $A_2Cr_3As_3$ plotted against ionic radius. The data for the lattice constants are taken from Ref.[4-7].

3. Comparison with previously reported $1/T_1$ of $K_2Cr_3As_3$

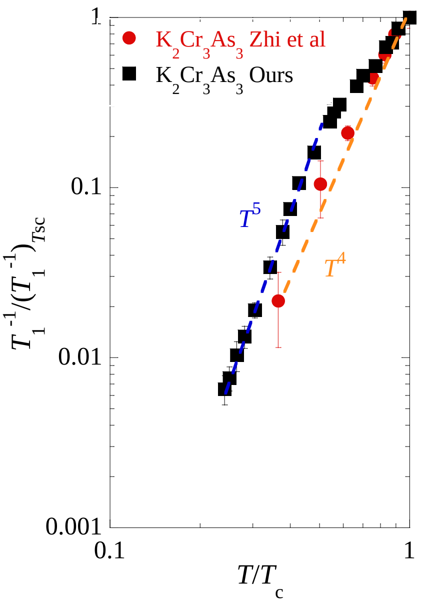

FIG.S3. Comparison of $1/T_1$ normalized by its value at T_{sc} with the data from ref. [8].

Previously, in a $K_2Cr_3As_3$ sample with a lower T_{sc}=5.7 K than our case (6.1 K), it was claimed that $1/T_1$ follows a T^4 variation [8]. We compare the reported data with ours in the above figure. It can be seen that the first three data points below T_{sc} agree well with ours, but the last three points disagree. The slower relaxation at low temperatures may be tied to the lower T_{sc} [8]. Namely, in-plane non-magnetic impurity or disorder brings about finite density of states at the Fermi level in nodal gap superconductors, which reduces T_{sc} and concomitantly results in $1/T_1T$=constant behavior at low temperatures [9]. As a result, in the intermediate temperature regime, $1/T_1$ exhibits a deviation from the otherwise T^5 variation.

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