

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

J. Luo,<sup>1</sup> J. Yang,<sup>1,\*</sup> R. Zhou,<sup>1</sup> Q. G. Mu,<sup>1,2</sup> T. Liu,<sup>1,2</sup> Zhi-an Ren,<sup>1</sup> C. J. Yi,<sup>1,2</sup> Y. G. Shi,<sup>1</sup> and Guo-qing Zheng<sup>1,3,†</sup>

<sup>1</sup>*Institute of Physics, Chinese Academy of Sciences, and Beijing National Laboratory for Condensed Matter Physics, Beijing 100190, China*

<sup>2</sup>*School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China*

<sup>3</sup>*Department of Physics, Okayama University, Okayama 700-8530, Japan*



(Received 12 March 2019; published 24 July 2019)

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  $^{75}\text{As}$  nuclear quadrupole resonance measurements on the  $A_2\text{Cr}_3\text{As}_3$  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  $A = \text{Na}, \text{Na}_{0.75}\text{K}_{0.25}, \text{K},$  and  $\text{Rb}$ , 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  $90^\circ$ , which enhances the FM interaction via the Cr2-As2-Cr2 path. Upon moving away from the QCP, the superconducting transition temperature  $T_{\text{sc}}$  increases progressively up to 8.0 K in  $\text{Na}_2\text{Cr}_3\text{As}_3$ , which is in sharp contrast to the AFM case where  $T_{\text{sc}}$  usually shows a maximum around a QCP. The  $1/T_1$  decreases rapidly below  $T_{\text{sc}}$  with no Hebel-Slichter peak, and ubiquitously follows a  $T^5$  variation below a characteristic temperature  $T^* \approx 0.6 T_{\text{sc}}$ , which indicates the existence of point nodes in the superconducting gap function commonly in the family. These results suggest that the  $A_2\text{Cr}_3\text{As}_3$  family is a possible solid-state analog of superfluid  $^3\text{He}$ .

DOI: 10.1103/PhysRevLett.123.047001

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  $T_{\text{sc}}$  usually takes a maximum at the AFM quantum critical point (QCP). However, superconductivity in the vicinity of a ferromagnetic ordered phase is still rare.  $\text{UGe}_2$  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  $^3\text{He}$  emerges in the background of ferromagnetic spin fluctuations. There are two phases, namely, the  $A$  phase and  $B$  phase, in  $^3\text{He}$  [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  $^3\text{He}$  is induced by FM spin fluctuations [8]. Notably,  $^3\text{He}$  phases are topologically nontrivial, which has received new and intensive interest in the past few years [11]. The  $B$  phase of  $^3\text{He}$  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  $^3\text{He}$  serves to bridge three large research areas: strong correlations, unconventional superconductivity, and topological quantum phenomena.

Recently, a  $3d$ -electron system, chromium-based superconductors  $A_2\text{Cr}_3\text{As}_3$  ( $A = \text{Na}, \text{K}, \text{Rb}, \text{Cs}$ ), has been discovered [13–16]. In this family, superconductivity emerges from a paramagnetic state.  $\text{K}_2\text{Cr}_3\text{As}_3$  and  $\text{Rb}_2\text{Cr}_3\text{As}_3$  have a  $T_{\text{sc}} = 6.1$  and 4.8 K [13,14], respectively, and  $\text{Na}_2\text{Cr}_3\text{As}_3$  has the highest  $T_{\text{sc}} = 8.0$  K. Resistivity measurement suggests that  $\text{Cs}_2\text{Cr}_3\text{As}_3$  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  $\alpha$  and  $\beta$ , and one three-dimensional (3D) band  $\gamma$  [20,21]. The  $\gamma$  band makes the main contribution to the density of states (DOS).

NQR, penetration depth, muon spin rotation ( $\mu$ SR), 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  $\text{Rb}_2\text{Cr}_3\text{As}_3$  [23]. Neutron scattering measurements also suggest short-range magnetic order in  $\text{K}_2\text{Cr}_3\text{As}_3$  [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  $\text{K}_2\text{Cr}_3\text{As}_3$  [22], but  $1/T_1 \propto T^5$  was found in  $\text{Rb}_2\text{Cr}_3\text{As}_3$  [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  $A_2\text{Cr}_3\text{As}_3$  by NQR. We find that  $1/T_1 T$  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  $A = \text{Na}$  to  $\text{Na}_{0.75}\text{K}_{0.25}$ ,  $\text{K}$ ,  $\text{Rb}$ , the system progressively approaches a possible FM QCP, which may be ascribed to the Cr2-As2-Cr2 bond angle that decreases toward  $90^\circ$  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  $T^* \approx 0.6T_{\text{sc}}$ , indicating a common formation of point nodes in the gap function. Our results indicate that  $A_2\text{Cr}_3\text{As}_3$  shows some properties similar to superfluid  $^3\text{He}$ .

Polycrystal sample of  $\text{Na}_2\text{Cr}_3\text{As}_3$  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  $\text{K}_2\text{Cr}_3\text{As}_3$  [22] and  $\text{Rb}_2\text{Cr}_3\text{As}_3$  [23], the NQR spectra of  $(\text{Na}_{0.75}\text{K}_{0.25})_2\text{Cr}_3\text{As}_3$  and  $\text{Na}_2\text{Cr}_3\text{As}_3$  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  $A = \text{Na}$ ,  $\text{Na}_{0.75}\text{K}_{0.25}$ , and  $\text{K}$  at the stronger peak.

As can be seen in Fig. 2,  $1/T_1 T$  is a constant above  $T \sim 150$  K and increases with decreasing temperature

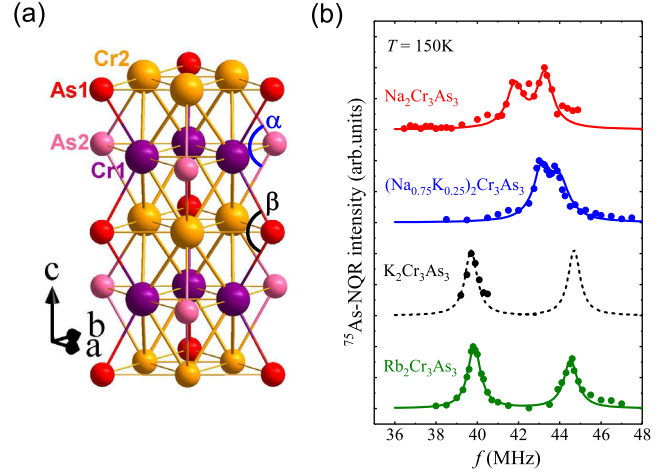


FIG. 1. (a) The structure of the  $[\text{Cr}_3\text{As}_3]_\infty$  tube, the Cr2-As2-Cr2 bond angle ( $\alpha$ ), and the Cr1-As1-Cr1 bond angle ( $\beta$ ). (b)  $^{75}\text{As}$  NQR spectra of  $A_2\text{Cr}_3\text{As}_3$  ( $A = \text{Na}$ ,  $\text{Na}_{0.75}\text{K}_{0.25}$ ,  $\text{K}$ ,  $\text{Rb}$ ) measured at  $T = 150$  K. Solid lines are fitting results by two Lorentzian functions. The dashed curve represents data of Ref. [22].

down to  $T_{\text{sc}}$  for all samples. For a conventional non-interacting metal,  $1/T_1 T$  is a constant. Therefore, the results indicate electron correlations. Previous Knight shift measurements in  $\text{Rb}_2\text{Cr}_3\text{As}_3$  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

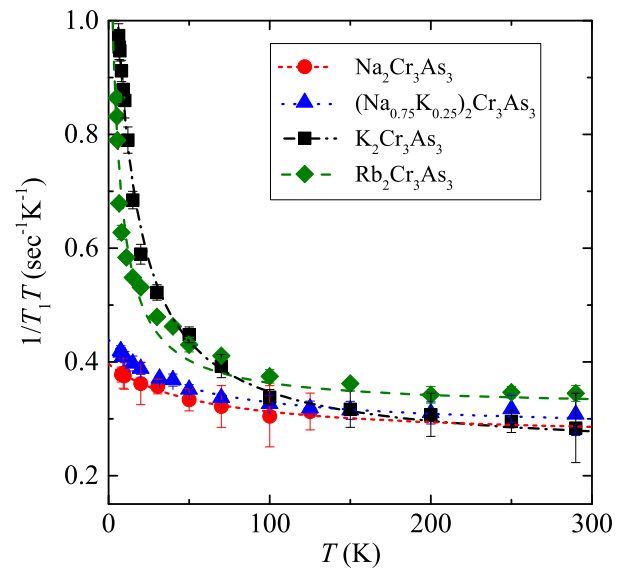


FIG. 2.  $^{75}\text{As}$  nuclear spin-lattice relaxation rate  $1/T_1$  divided by temperature. Data for  $\text{Rb}_2\text{Cr}_3\text{As}_3$  are from Ref. [23]. The dashed curves on the normal state data are fittings to  $1/T_1 T = a + b/(T + \theta)$ .

characterize the spin fluctuations [34]. The  $1/T_1T$  can be expressed as  $1/T_1T = (1/T_1T)_{\text{SF}} + (1/T_1T)_0$ . 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],  $(1/T_1T)_{\text{SF}}$  follows a relation of  $C/(T + \theta)$ , with the parameter  $\theta$  describing a distance to FM QCP. The obtained  $\theta$  decreases in the order of  $A = \text{Na}$ ,  $\text{Na}_{0.75}\text{K}_{0.25}$ ,  $\text{K}$ , and  $\text{Rb}$ . The parameter  $\theta = 4 \pm 1.5$  K for  $\text{Rb}_2\text{Cr}_3\text{As}_3$  and  $\theta = 57 \pm 7$  K for  $\text{Na}_2\text{Cr}_3\text{As}_3$ .

Figure 3 shows the phase diagram of the family, where  $\theta$  and  $T_{\text{sc}}$  are plotted as a function of the ionic radius of alkali element  $A$ . As the ionic radius increases, the parameter  $\theta$  decreases, indicating that the system is tuned closer to a possible FM QCP. Concomitantly,  $T_{\text{sc}}$  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  $\alpha$  has a linear relationship with the ionic radius, while the Cr1-As1-Cr1 bond angle  $\beta$  is almost a constant. Notably, as the ionic radius increases,  $\alpha$  decreases towards  $90^\circ$ . 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

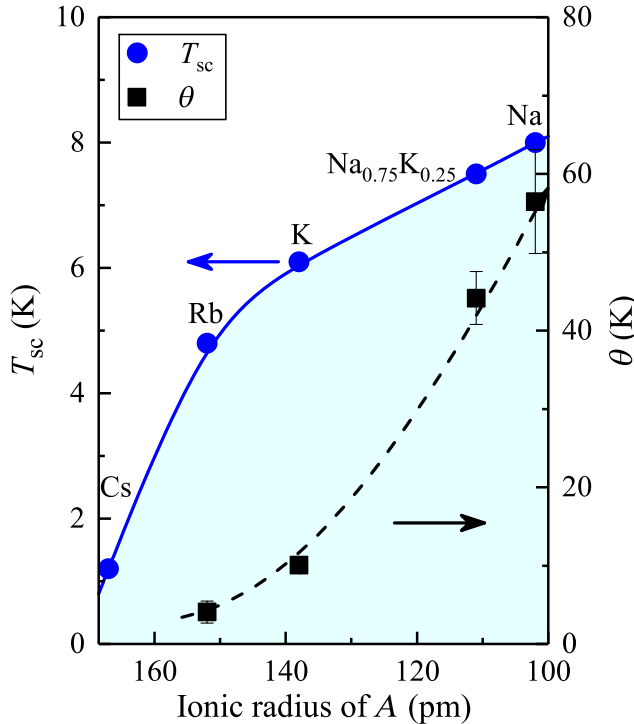


FIG. 3. Phase diagram of  $A_2\text{Cr}_3\text{As}_3$  ( $A = \text{Na}$ ,  $\text{Na}_{0.75}\text{K}_{0.25}$ ,  $\text{K}$ ,  $\text{Rb}$ ,  $\text{Cs}$ ). The data of alkali ions with six-coordinations were taken from Ref. [35]. The filled squares are  $\theta$  from the fitting of  $1/T_1T$  data (see text).

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  $A = \text{Na}$  to  $\text{Na}_{0.75}\text{K}_{0.25}$ ,  $\text{K}$ , and  $\text{Rb}$ , 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.

In Fig. 3, one sees that  $T_{\text{sc}}$  decreases upon approaching the FM QCP. This is in sharp contrast to the AFM case where  $T_{\text{sc}}$  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  $T_{\text{sc}}$  is zero right at ferromagnetic QCP [37]. Later on, Monthoux and Lonzarich [38] and Wang *et al.* [39] calculated  $T_{\text{sc}}$  in the strong-coupling limit. They pointed out that mass enhancement and a finite quasiparticle lifetime act as pair breaking, but found a nonzero  $T_{\text{sc}}$  at ferromagnetic QCP. In  $\text{UGe}_2$ , however, no  $T_{\text{sc}}$  was found in the paramagnetic side. In a related compound  $\text{UCoGe}$  [40], although superconductivity survives after ferromagnetic order is suppressed, the ferromagnetic-paramagnetic transition is a first-order phase transition [41] and thus cannot

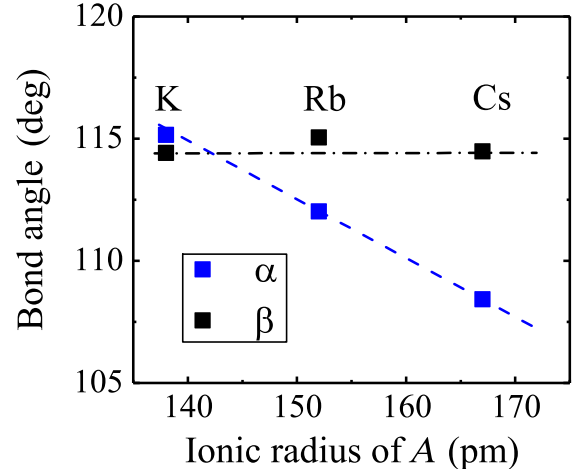


FIG. 4. Correlation between the ionic radius and the Cr2-As2-Cr2 bond angle  $\alpha$  and Cr1-As1-Cr1 bond angle ( $\beta$ ) for  $\text{Cs}_2\text{Cr}_3\text{As}_3$ ,  $\text{Rb}_2\text{Cr}_3\text{As}_3$ , and  $\text{K}_2\text{Cr}_3\text{As}_3$ . The angles were calculated based on the available crystal-structure data in the literature [13,15,36].

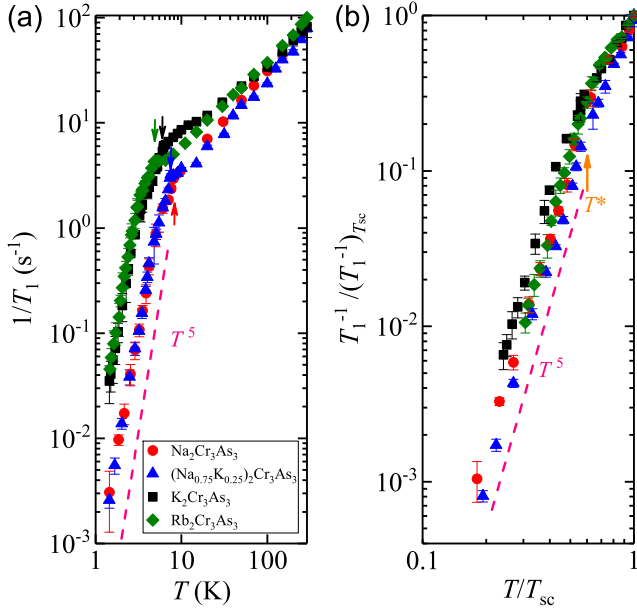


FIG. 5. (a)  $1/T_1$  as a function of  $T$  for all samples. The arrows indicate  $T_{sc}$  of each compound. (b)  $1/T_1$  normalized by its value at  $T_{sc}$ . The arrow indicates a characteristic temperature  $T^*$ , below which  $1/T_1$  becomes proportional to  $T^5$ . The symbols are the same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

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

where  $N_s(E)$  is the DOS below  $T_{sc}$ ,  $f(E)$  is Fermi distribution function,  $\Delta$  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 would induce 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 ( $\alpha$ ) that decreases towards  $90^\circ$  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  $^3He$ . Therefore, further investigations on this family promise to enrich the physics across multiple research areas of electron correlations, unconventional superconductivity, and topological quantum phenomena.

We thank G. H. Cao, Y. Haga, J. P. Hu, H. Kontani, K. Miyake, A. Sumiyama, Z. Q. Wang, H. M. Weng, G. M. Zhang, and Y. Zhou for helpful discussions, and C. G. Wang for technical assistance. This work was partially supported by NSFC Grants (No. 11674377, No. 11634015, and No. 11774399), MOST Grants (No. 2016YFA0300502, No. 2017YFA0302901, and No. 2017YFA0302904), as well as JSPS/MEXT Grants (No. JP15H05852 and No. JP19H00657). J. Y. also acknowledges support by the Youth Innovation Promotion Association of CAS.

\*yangjie@iphy.ac.cn

†gqzheng123@gmail.com

[1] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Magnetically mediated superconductivity in heavy fermion compounds, *Nature (London)* **394**, 39 (1998).

- [2] P. Coleman and A. J. Schofield, Quantum criticality, *Nature (London)* **433**, 226 (2005).
- [3] P. A. Lee, N. Nagaosa, and X. G. Wen, Doping a Mott insulator: Physics of high-temperature superconductivity, *Rev. Mod. Phys.* **78**, 17 (2006).
- [4] G. R. Stewart, Superconductivity in iron compounds, *Rev. Mod. Phys.* **83**, 1589 (2011).
- [5] S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Sheikin, D. Braithwaite, and J. Flouquet, Superconductivity on the border of itinerant-electron ferromagnetism in  $\text{UGe}_2$ , *Nature (London)* **406**, 587 (2000).
- [6] D. D. Osheroff, R. C. Richardson, and D. M. Lee, Evidence for a New Phase of Solid  $\text{He}^3$ , *Phys. Rev. Lett.* **28**, 885 (1972).
- [7] P. W. Anderson and P. Morel, Generalized Bardeen-Cooper-Schrieffer states and the proposed low-temperature phase of liquid  $^3\text{He}$ , *Phys. Rev.* **123**, 1911 (1961).
- [8] P. W. Anderson and W. F. Brinkman, Anisotropic Superfluidity in  $^3\text{He}$ : A Possible Interpretation of Its Stability as a Spin-Fluctuation Effect, *Phys. Rev. Lett.* **30**, 1108 (1973).
- [9] R. Balian and N. R. Werthamer, Superconductivity with Pairs in a Relative p Wave, *Phys. Rev.* **131**, 1553 (1963).
- [10] A. G. Leggett, A theoretical description of the new phases of liquid  $^3\text{He}$ , *Rev. Mod. Phys.* **47**, 331 (1975).
- [11] X.-L. Qi and S.-C. Zhang, Topological insulators and superconductors, *Rev. Mod. Phys.* **83**, 1057 (2011).
- [12] A. P. Schnyder, S. Ryu, A. Furusaki, and A. W. W. Ludwig, Classification of topological insulators and superconductors in three spatial dimensions, *Phys. Rev. B* **78**, 195125 (2008).
- [13] J. K. Bao, J. Y. Liu, C. W. Ma, Z. H. Meng, Z. T. Tang, Y. L. Sun, H. F. Zhai, H. Jiang, H. Bai, C. M. Feng, Z. A. Xu, and G. H. Cao, Superconductivity in Quasi-One-Dimensional  $\text{K}_2\text{Cr}_3\text{As}_3$  with Significant Electron Correlations, *Phys. Rev. X* **5**, 011013 (2015).
- [14] Z. T. Tang, J. K. Bao, Y. Liu, Y. L. Sun, A. Ablimit, H. F. Zhai, H. Jiang, C. M. Feng, Z. A. Xu, and G. H. Cao, Unconventional superconductivity in quasi-one-dimensional  $\text{Rb}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **91**, 020506(R) (2015).
- [15] Z. T. Tang, J. K. Bao, Z. Wang, H. Bai, H. Jiang, Y. Liu, H. F. Zhai, C. M. Feng, Z. A. Xu, and G. H. Cao, Superconductivity in quasi-one-dimensional  $\text{Cs}_2\text{Cr}_3\text{As}_3$  with large interchain distance, *Sci. China Mater.* **58**, 16 (2015).
- [16] Q. G. Mu, B. B. Ruan, B. J. Pan, T. Liu, J. Yu, K. Zhao, G. F. Chen, and Z. A. Ren, Ion-exchange synthesis and superconductivity at 8.6 K of  $\text{Na}_2\text{Cr}_3\text{As}_3$  with quasi-one-dimensional crystal structure, *Phys. Rev. Mater.* **2**, 034803 (2018).
- [17] H. Z. Zhi, D. Lee, T. Imai, Z. T. Tang, Y. Liu, and G. H. Cao,  $^{133}\text{Cs}$ , and  $^{75}\text{As}$  NMR investigation of the normal metallic state of quasi-one-dimensional  $\text{Cs}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **93**, 174508 (2016).
- [18] We have also performed NQR measurements on  $\text{Cs}_2\text{Cr}_3\text{As}_3$  down to  $T = 1.4$  K and obtained similar results as Ref. [17]. Namely, different from other compounds described in this Letter,  $1/T_1T$  for  $\text{Cs}_2\text{Cr}_3\text{As}_3$  shows a pseudogaplike behavior below  $T \sim 75$  K, whose origin is unknown.
- [19] A. Sumiyama, T. Nagasawa, A. Yamaguchi, J. Yang, Y. G. Shi, and G.-q. Zheng, Autumn Meeting of Japan Physical Society, Doshisha University, Program No. 10aPS-40 (Division 8), 2018.
- [20] H. Jiang, G. H. Cao, and C. Cao, Electronic structure of quasio-one-dimensional superconductor  $\text{K}_2\text{Cr}_3\text{As}_3$  from first-principles calculations, *Sci. Rep.* **5**, 16054 (2015).
- [21] X. X. Wu, C. C. Le, J. Yuan, H. Fan, and J. P. Hu, Magnetism in quasi-one-dimensional  $\text{A}_2\text{Cr}_3\text{As}_3$  ( $A = \text{K}, \text{Rb}$ ) superconductors, *Chin. Phys. Lett.* **32**, 057401 (2015).
- [22] H. Z. Zhi, T. Imai, F. L. Ning, J. K. Bao, and G. H. Cao, NMR Investigation of the Quasi-One-Dimensional Superconductor  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. Lett.* **114**, 147004 (2015).
- [23] J. Yang, Z. T. Tang, G. H. Cao, and G.-q. Zheng, Ferromagnetic Spin Fluctuation and Unconventional Superconductivity in  $\text{Rb}_2\text{Cr}_3\text{As}_3$  Revealed by  $^{75}\text{As}$  NMR and NQR, *Phys. Rev. Lett.* **115**, 147002 (2015).
- [24] G. M. Pang, M. Smidman, W. B. Jiang, J. K. Bao, Z. F. Weng, Y. F. Wang, L. Jiao, J. L. Zhang, G. H. Cao, and H. Q. Yuan, Evidence for nodal superconductivity in quasi-one-dimensional  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **91**, 220502(R) (2015).
- [25] D. T. Adroja, A. Bhattacharyya, M. Telling, Y. Feng, M. Smidman, B. Pan, J. Zhao, A. D. Hillier, F. L. Pratt, and A. M. Strydom, Superconducting ground state of quasi-one-dimensional  $\text{K}_2\text{Cr}_3\text{As}_3$  investigated using  $\mu\text{SR}$  measurements, *Phys. Rev. B* **92**, 134505 (2015).
- [26] D. Adroja, A. Bhattacharyya, M. Smidman, A. Hillier, Y. Feng, B. Pan, J. Zhao, M. R. Lees, A. Strydom, and P. K. Biswas, Nodal superconducting gap structure in the quasi-one-dimensional  $\text{K}_2\text{Cr}_3\text{As}_3$  investigated using  $\mu\text{SR}$  measurements, *J. Phys. Soc. Jpn.* **86**, 044710 (2017).
- [27] Y. T. Shao, X. X. Wu, L. Wang, Y. G. Shi, J. P. Hu, and J. L. Luo, Evidence of line nodes in superconducting gap function in  $\text{K}_2\text{Cr}_3\text{As}_3$  from specific-heat measurements, *Euro. Phys. Lett.* **123**, 57001 (2018).
- [28] F. F. Balakirev, T. Kong, M. Jaime, R. D. McDonald, C. H. Mielke, A. Gurevich, P. C. Canfield, and S. L. Bud'ko, Anisotropy reversal of the upper critical field at low temperatures and spin-locked superconductivity in  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **91**, 220505(R) (2015).
- [29] H. Zuo, J. K. Bao, Y. Liu, J. Wang, Z. Jin, Z. Xia, L. Li, Z. Xu, J. Kang, Z. Zhu, and G. H. Cao, Temperature and angular dependence of the upper critical field in  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **95**, 014502 (2017).
- [30] X. X. Wu, F. Yang, C. C. Le, H. Fan, and J. P. Hu, Triplet  $p_z$ -wave pairing in quasi-one-dimensional  $\text{A}_2\text{Cr}_3\text{As}_3$  superconductors ( $A = \text{K}, \text{Rb}, \text{Cs}$ ), *Phys. Rev. B* **92**, 104511 (2015).
- [31] H. T. Zhong, X. Y. Feng, H. Chen, and J. H. Dai, Formation of Molecular-Orbital Bands in a Twisted Hubbard Tube: Implications for Unconventional Superconductivity in  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. Lett.* **115**, 227001 (2015).
- [32] Y. Zhou, C. Cao, and F.-C. Zhang, Theory for superconductivity in alkali chromium arsenides  $\text{A}_2\text{Cr}_3\text{As}_3$  ( $A = \text{K}, \text{Rb}, \text{Cs}$ ), *Sci. Bull.* **62**, 208 (2017).
- [33] K. M. Taddei, Q. Zheng, A. S. Sefat, and C. de la Cruz, Coupling of structure to magnetic and superconducting orders in quasi-one-dimensional  $\text{K}_2\text{Cr}_3\text{As}_3$ , *Phys. Rev. B* **96**, 180506(R) (2017).
- [34] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).

- [35] R. D. Shannon, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, *Acta Crystallogr.* **A32**, 751 (1976).
- [36] G. H. Cao, J. K. Bao, Z. T. Tang, Y. Liu, and H. Jiang, Peculiar properties of  $\text{Cr}_3\text{As}_3$ -chain-based superconductors, *Philos. Mag.* **97**, 591 (2017).
- [37] D. Fay and J. Appel, Coexistence of p-state superconductivity and itinerant ferromagnetism, *Phys. Rev. B* **22**, 3173 (1980).
- [38] P. Monthoux and G. G. Lonzarich, Magnetically mediated superconductivity in quasi-two and three dimensions, *Phys. Rev. B* **63**, 054529 (2001).
- [39] Z. Wang, W. Mao, and K. Bedell, Superconductivity Near Itinerant Ferromagnetic Quantum Criticality, *Phys. Rev. Lett.* **87**, 257001 (2001).
- [40] N. T. Huy, A. Gasparini, D. E. de Nijs, Y. Huang, J. C. Klaasse, T. Gortenmulder, A. de Visser, A. Hamann, T. Gorklach, and H. V. Lohneysen, Superconductivity on the Border of Weak Itinerant Ferromagnetism in  $\text{UCoGe}$ , *Phys. Rev. Lett.* **99**, 067006 (2007).
- [41] M. Manago, S. Kitagawa, K. Ishida, K. Deguchi, N. K. Sato, and T. Yamamura, Enhancement of superconductivity by pressure-induced critical ferromagnetic fluctuations in  $\text{UCoGe}$ , *Phys. Rev. B* **99**, 020506(R) (2019).
- [42] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.123.047001> for the phase diagram compared to theories, lattice constants, and others.
- [43] C. G. Wang, Z. Li, J. Yang, L. Y. Xing, G. Y. Dai, X. C. Wang, C. Q. Jin, R. Zhou, and G.-q. Zheng, Electron Mass Enhancement near a Nematic Quantum Critical Point in  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ , *Phys. Rev. Lett.* **121**, 167004 (2018).
- [44] K. Katayama, S. Kawasaki, M. Nishiyama, H. Sugawara, D. Kikuchi, H. Sato, and G.-q. Zheng, Evidence for point nodes in the superconducting gap function in the filled skutterudite heavy-fermion compound  $\text{PrOs}_4\text{Sb}_{12}$ :  $^{123}\text{Sb}$ -NQR study under pressure, *J. Phys. Soc. Jpn.* **76**, 023701 (2007).
- [45] K. Asayama, Y. Kitaoka, G.-q. Zheng, and K. Ishida, NMR studies of high  $T_c$  superconductors, *Prog. Nucl. Magn. Reson. Spectrosc.* **28**, 221 (1996).
- [46] K. Matano, Z. A. Ren, X. L. Dong, L. L. Sun, Z. X. Zhao, and G.-q. Zheng, Spin-singlet superconductivity with multiple gaps in  $\text{PrO}_{0.89}\text{F}_{0.11}\text{FeAs}$ , *Euro. Phys. Lett.* **83**, 57001 (2008).
- [47] R. A. Fisher, S. Kim, B. F. Woodfield, N. E. Phillips, L. Taillefer, K. Hasselbach, J. Flouquet, A. L. Giorgi, and J. L. Smith, Specific Heat of  $\text{UPt}_3$ : Evidence for Unconventional Superconductivity, *Phys. Rev. Lett.* **62**, 1411 (1989).