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Effect of hydrostatic pressure on the superconducting properties of quasi-1D superconductor $K_2Cr_3As_3$

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Abstract

$K_2Cr_3As_3$ is a newly discovered quasi-1D superconductor with a $T_c = 6.1$ K and an upper critical field $\mu_0 H_{c2}(0) \approx 40$ T three times larger than the Pauli paramagnetic limit $\mu_0 H_p$ that is suggestive of a spin-triplet Cooper pairing. In this paper, we have investigated the effects of hydrostatic pressure on its T_c and $\mu_0 H_{c2}$ by measuring the ac magnetic susceptibility $\chi'(T)$ under magnetic fields at various hydrostatic pressures up to 7.5 GPa. The major findings include: (1) T_c is suppressed gradually to below 2 K at 7.5 GPa; (2) the estimated $\mu_0 H_{c2}(0)$ decreases dramatically to below $\mu_0 H_p$ above ~2 GPa and becomes slight lower than the orbital limiting field $\mu_0 H_{c2}^{orb}(0)$ estimated from the initial slope of upper critical field via $\mu_0 H_{c2}^{orb}(0) = -0.73 T_c \mathrm{d}H_{c2}/\mathrm{d}T_c|_{T_c}$ in the clean limit; (3) the estimated Maki parameter $\alpha = \sqrt{2} H_{c2}^{orb}(0)/H_p$ drops from 4 at ambient pressure to well below 1 at $P > 2$ GPa, suggesting the crossover from Pauli paramagnetic limiting to orbital limiting in the pair breaking process upon increasing pressure. These observations suggested that the application of hydrostatic pressure could drive $K_2Cr_3As_3$ away from the ferromagnetic instability and lead to a breakdown of the spin-triplet pairing channel. We have also made a side-by-side comparison and discussed the distinct effects of chemical and physical pressures on the superconducting properties of $K_2Cr_3As_3$.

Keywords: superconductivity, high pressure, $K_2Cr_3As_3$, upper critical field

(Some figures may appear in colour only in the online journal)

Introduction

Recently, much attention has been paid to the CrAs-based superconductors that arise as a new platform for studying the unconventional superconductivity. Soon after the discovery of pressure-induced superconductivity with a maximum $T_c \approx 2$ K in the binary compound CrAs [1, 2], a novel class of Cr-based materials, $A_2Cr_3As_3$, was synthesized and found to be superconducting at ambient pressure with $T_c = 6.1$ K for A = K [3–5], 4.8 K for A = Rb [5, 6], and 2.2 K for A = Cs [7], respectively. In contrast to the 3D helimagnetic CrAs, the crystal structure of $A_2Cr_3As_3$ is quasi-one-dimensional (Q1D) featured by $[(Cr_3As_3)^{2-}]_\infty$ double-walled subnanotubes with face-sharing $Cr_{6/2}$ ($As_{6/2}$) octahedral chains in the inner (outer) wall [3]. These Q1D novel superconductors were found to display intriguing physical properties at both the normal and superconducting states: the normal state above T_c is characterized by a linear-T resistivity in a wide

[5] These authors contributed equally to this work.

temperature range for the polycrystalline samples [3, 6], a large electronic specific-heat coefficient $\gamma_n = 70$–75 mJ mol^{-1} K^{-1} [2] signaling considerable electronic correlations [3], and a strong enhancement of spin fluctuations towards T_c with the nuclear spin relaxation rate $1/T_1$ following a characteristic power law predicted for the 1D Tomonaga-Luttinger liquid [8, 9]; in the superconducting state, on the other hand, the absence of Herbal–Slichter coherence peak in $1/T_1$ just below T_c [8, 10], the linear T-dependence of London penetration depth [11], and the $\sqrt{H}$-dependence of electronic specific heat coefficient [6] are suggestive of an unconventional superconductivity with line nodes in the superconducting gap [12, 13]. More intriguingly, the upper critical field $\mu_0H_{c2}(0) \approx 40$ T of $K_2Cr_3As_3$ at ambient pressure was found to be largely exceeding the Pauli limit $\mu_0H_p = 1.84\ T_c \approx 11$ T, implying a plausible spin-triplet Cooper pairing [3, 10]. According to the first-principles calculations [14], the electronic structures of $K_2Cr_3As_3$ near the Fermi level are composed of two Q1D Fermi surface sheets (α- and β-bands) and one 3D sheet (γ-band), all of which are essentially originated from the Cr-3d orbitals. Several theoretical investigations based on such an electronic band structures indeed found favorable solutions for the spin triplet paring driven by ferromagnetic fluctuations [14–19]. In light of these above experimental and theoretical results, it has been argued that the observed superconductivity in $A_2Cr_3As_3$ opens a new avenue for searching multiband triplet p-wave pairing driven by ferromagnetic fluctuations.

Although the majority of experimental and theoretical results on $A_2Cr_3As_3$ points to an unconventional, possible spin-triplet pairing superconductivity, consensus has not been reached yet regarding the mechanism of superconductivity in this novel class of Q1D materials. A high-field study on the anisotropic μ_0H_{c2} of $K_2Cr_3As_3$ by Balakirev *et al* [20] revealed a Pauli-limited behavior for $\mu_0H_{c2}^{||}(T)$ and the anisotropy reversal of $\mu_0H_{c2}^{||}(T) < \mu_0H_{c2}^{\perp}(T)$ below T_c; these observations were argued to be inconsistent with triplet superconductivity, but were proposed to be a form of singlet superconductivity with the electron spins locked onto the direction of Cr chains. In contrast, a recent study on the angular dependence of the μ_0H_{c2} on $K_2Cr_3As_3$ single crystal [21] evidenced a unique threefold modulation for the in-plane $\mu_0H_{c2}^{\perp}(\varphi)$, which has been interpreted as a signature for dominant triplet Cooper pairing with odd parity. In addition, based on the first-principles calculations, Subedi [22] proposed that the superconductivity and the above-mentioned peculiar properties in $K_2Cr_3As_3$ should be attributed to a strong, anisotropic electron–phonon coupling ($\lambda_{ep} = 3.0$) that involves in-plane motions of all three species of atoms. Experimentally, a polarized Raman scattering study has evidenced a temperature independent phonon mode with Fano line shape, which might provide a coupling medium between the magnetic fluctuations and the electronic structure [23]. Therefore, the exact mechanism of superconductivity in the Q1D $A_2Cr_3As_3$ remains an open issue.

High pressure has been known as a clean means to fine tune the structure and electronic properties of materials. Particularly, to investigate the effects of pressure on superconductors can not only provide some guidelines for further improving T_c, but also offer important information for understanding the mechanism of Cooper pairing. In general, the application of hydrostatic pressure has two major effects: on the one hand, pressure tends to broaden the bandwidth of conduction electron via shortening the bond lengths, which usually results in a reduced density of states at Fermi level and weakened electron correlations; on the other hand, pressure usually makes the materials harder and shifts the phonon spectrum toward higher energy. Thus, to study the effect of pressure can reveal the role of both the electron correlations and electron–phonon coupling.

The effects of pressure on the superconducting transition temperature T_c of $A_2Cr_3As_3$ have been investigated by Kong *et al* [4] up to 0.7 GPa for A = K and by Wang *et al* [24] up to 5 GPa for A = K and Rb. It was found that for both compounds T_c decreases gradually with pressure; T_c of $K_2Cr_3As_3$ decreases at a rate of -0.38 K GPa^{-1} for $P < 2$ GPa, while T_c of $Rb_2Cr_3As_3$ is suppressed at a higher rate of -0.814 K GPa^{-1}. Given the fact that T_c of $A_2Cr_3As_3$ increases progressively upon reducing the unit-cell volume from A = Cs (2.2 K) to Rb (4.8 K), and then to K (6.1 K), the observations of negative physical pressure effect on T_c are against the general expectations. Based on a high-pressure structural study on $K_2Cr_3As_3$, Wang *et al* [24] have correlated T_c of $A_2Cr_3As_3$ with the As–Cr–As bond angles and interchain distances, and further suggested a positive correlation between superconductivity with the degree of non-centrosymmetry in $A_2Cr_3As_3$. However, the effect of pressure on μ_0H_{c2}, which is the most intriguing property of $A_2Cr_3As_3$, has never been studied to date.

In order to shed more light on the mechanism of superconductivity in $A_2Cr_3As_3$, we have extended the previous high-pressure studies and investigated the effects of hydrostatic pressure on both T_c and μ_0H_{c2} of $K_2Cr_3As_3$ by measuring its dc and ac magnetic susceptibility under different external magnetic fields up to 7.5 GPa. Accompanying the gradually suppression of T_c under pressure, we observed a profound reduction of $\mu_0H_{c2}(0)$ from Pauli paramagnetic limiting to orbital limiting in the pairing breaking process. Our results indicated that $K_2Cr_3As_3$ is likely located near a magnetic instability with strong ferromagnetic fluctuations mediating the spin-triplet superconductivity; the application of hydrostatic pressure drives the system away from the ferromagnetic instability and leads to a breakdown of the spin-triplet pairing channel.

Experimental details

Needle-shaped $K_2Cr_3As_3$ single crystals were grown with the self-flux method as described elsewhere [3]. The crystals are very thin, soft, easy to peel off, and very sensitive to moisture and air, so we used a lump of randomly oriented crystals in our high-pressure measurements. The randomly oriented crystals were covered with Apiezon-N grease in an Ar-filled glove box and then inserted into the pressure cell. DC magnetization measurements under pressures up to 1.16 GPa were performed in a miniature BeCu piston cylinder cell fitted into the commercial magnetic property measurement system (MPMS-3, Quantum Design). The sample and a piece of lead (Pb) were loaded into a Teflon cell filled with Daphne 7373 as the pressure transmitting medium (PTM). The pressure values were determined from the shift of the superconducting transition temperature of Pb. AC magnetic susceptibility measurements with the mutual induction method were carried out by using the palm cubic anvil cell apparatus up to 7.5 GPa. We have applied GE varnish to fix the sample and coil assembly inside a Teflon capsule filled with Glycerol as the PTM. An excitation current of ~1 mA with a frequency of 317 Hz was applied to the primary coil and the output signal across two oppositely wound secondary coils was picked up with a Standford Research SR830 lock-in amplifier. Because it is hard to determine accurately the samples' mass and volume in the high-pressure measurements, we cannot estimate the superconducting volume fraction accurately. So we gave below the absolute values of the magnetization in the unit of emu and the output voltages in an arbitrary unit for the ac susceptibility. From the magnitude of the susceptibility drop $\Delta\chi'(T)$ at T_c, nevertheless, we can roughly examine the change of the superconducting volume as a function of pressure or magnetic field.

Results and discussions

We first studied the effect of pressure on the superconducting transition temperature T_c. Figure 1(a) shows the temperature dependence of dc magnetization $M(T)$ for $K_2Cr_3As_3$ and Pb at various pressures up to 1.16 GPa. The data were collected upon warming up under an external magnetic field of $H = 20$ Oe after zero field cooling from room temperature. As can be seen, the superconducting diamagnetic signal appears twice for Pb and $K_2Cr_3As_3$, respectively, and in both cases T_c decreases gradually with increasing pressure. The transition width of $K_2Cr_3As_3$ is not as sharp as that of Pb. T_c values were determined from the intersection of two straight lines above and below the transition, figure 1(b).The observed $T_c = 6.1$ K at zero pressure agrees well with the reported value [3], and shifts down gradually to 5.65 K at 1.16 GPa.

To further track down the evolution of $T_c(P)$ under higher pressures, we turn to the ac magnetic susceptibility $\chi'(T)$ measurements by using the palm cubic anvil cell apparatus. The $\chi'(T)$ data shown in figure 1(b) evidenced a continuous suppression of T_c upon further increasing pressure, and T_c reaches ~1.5 K at 7.5 GPa. As can be seen, the nearly parallel shift of $\chi'(T)$ curves illustrates that the superconducting transition width remains almost unchanged with increasing pressure, signaling a relatively good hydrostaticity in the investigated pressure range. Although the measured temperature range for superconducting transition becomes limited gradually with increasing pressure, the magnitude of $\chi'(T)$ drop converges to a similar value for all the investigated pressures as shown in figure 1(b). This indicates the superconducting volume keeps nearly the same under pressure. Figure 1(c) displays the pressure dependence of $T_c(P)$ obtained from the above measurements. As can be seen, our results are in excellent agreement

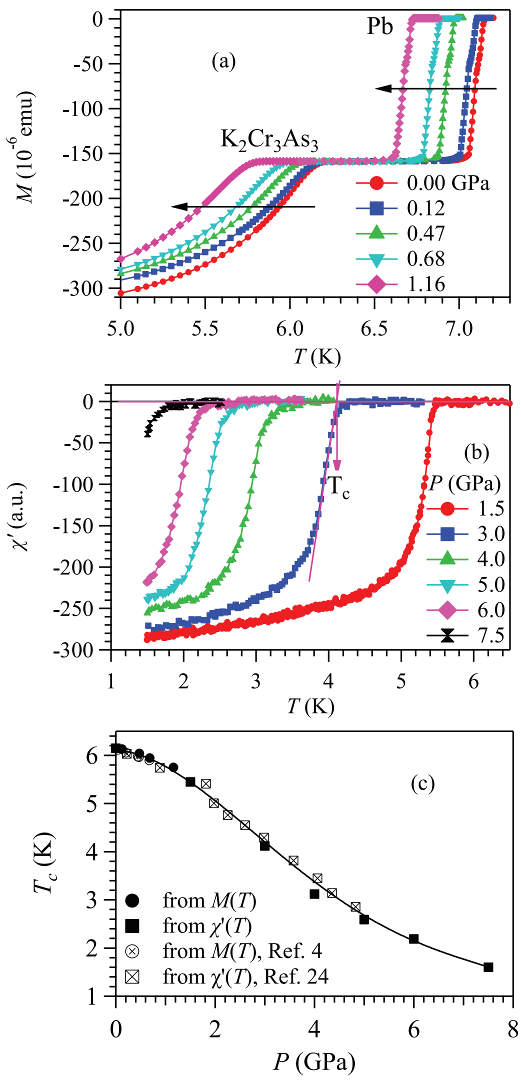

Figure 1. Temperature dependence of the (a) DC and (b) AC magnetic susceptibility of $K_2Cr_3As_3$ under various hydrostatic pressures up to 7.5 GPa. (c) Pressure dependence of the superconducting transition temperature T_c determined from the above measurements and the published data in [4, 24].

with those reported previously [4, 24]. But, our present study in an extended pressure range evidenced a reduced suppression rate of $T_c(P)$ above 5 GPa.

As mentioned above, another distinguished property of $K_2Cr_3As_3$ is the unusually large upper critical field $\mu_0H_{c2}(0) \approx 40$ T, which is three times larger than the Pauli limit $\mu_0H_p = 1.84$ T_c and thus implies a plausible spin-triplet Cooper pairing in this system. In order to gain more insight on the evolution of superconducting state with the reduction of T_c under pressure, we have also investigated the effect of hydrostatic pressure on $\mu_0H_{c2}(0)$ by measuring $\chi'(T)$ at different external dc magnetic fields under each pressure. Figure 2 shows some representative data at (a) 1.5 GPa, (b) 3.0 GPa, (c) 4.0 GPa, and (d) 6.0 GPa, respectively. For $P = 1.5$ GPa, the superconducting diamagnetic signal drops sharply at zero field, but the transition width becomes broaden and the superconducting volume is reduced gradually with increasing magnetic field as expected. Here, we define T_c as

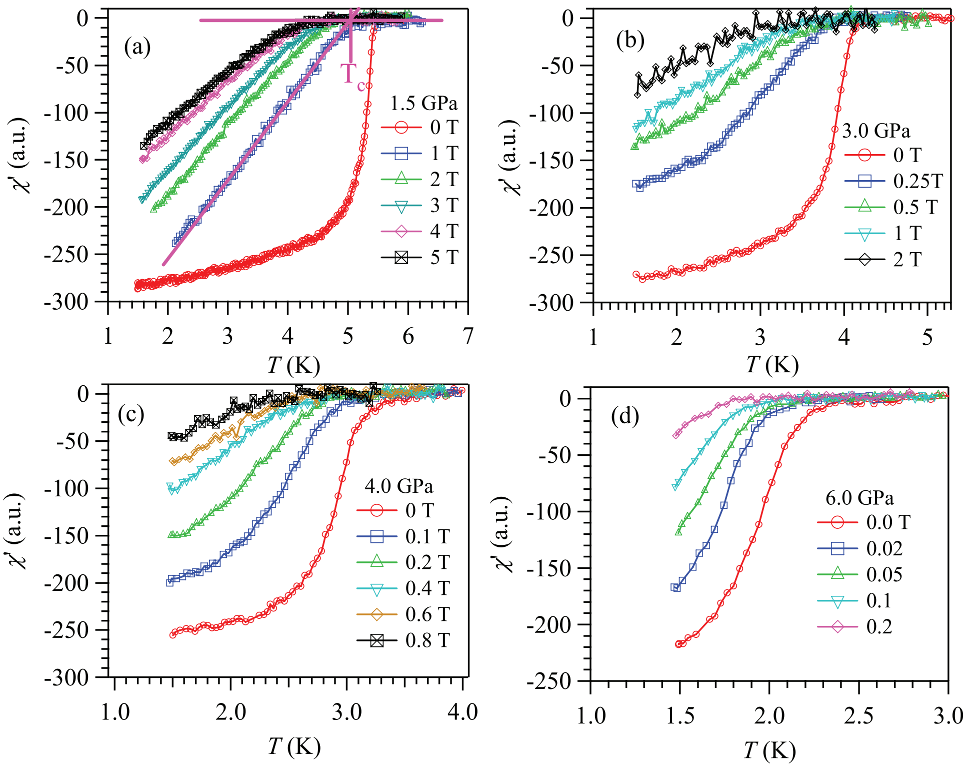

Figure 2. Temperature dependence of the ac susceptibility $\chi'(T)$ at different external magnetic fields under various hydrostatic pressures: (a) 1.5 GPa, (b) 3.0 GPa, (c) 4.0 GPa, and (d) 6.0 GPa.

the intersection for two straight lines below and above the transition as shown in figure 2(a). It is clear that T_c is suppressed progressively by magnetic fields, and reaches ~4 K at 5 T. Accompanying the suppression of T_c with increasing pressure, the external magnetic field needed to eliminate the superconducting diamagnetic signals also decreases quickly. As seen in figures 2(b)–(d), the superconducting signal becomes nearly diminished at about 2 T, 0.8 T, and 0.2 T for $P = 3$, 4, and 6 GPa, respectively.

Temperature dependences of upper critical field, $\mu_0H_{c2}(T_c)$, for $K_2Cr_3As_3$ under different pressures obtained from the above measurements are plotted in figure 3(b), together with those of $A_2Cr_3As_3$ (A = K [3, 4], Rb [5, 6], and Cs [7]) at ambient pressure for comparison. Although a pronounced anisotropy of $\mu_0H_{c2}(T)$ has been found for the single crystal samples, it is instructive to compare the results of randomly oriented crystals in our measurements with those of polycrystalline samples in figure 3. As can be seen, $\mu_0H_{c2}(T)$ curves of polycrystalline $A_2Cr_3As_3$ (A = K, Rb, and Cs) at ambient pressure are nearly parallel to each other despite of the large variation of T_c, suggesting that they share similar pairing mechanism. In striking contrast, the slope of $\mu_0H_{c2}(T)$ for $K_2Cr_3As_3$ decreases significantly with pressure accompanying the gradual suppression of T_c. It is noteworthy that T_c of $K_2Cr_3As_3$ at 6 GPa is very close to that of $Cs_2Cr_3As_3$ at ambient pressure, but the upper critical field is about an order of magnitude lower. These observations indicated that the superconducting pairing mechanism might undergo a dramatic change under pressure for $K_2Cr_3As_3$.

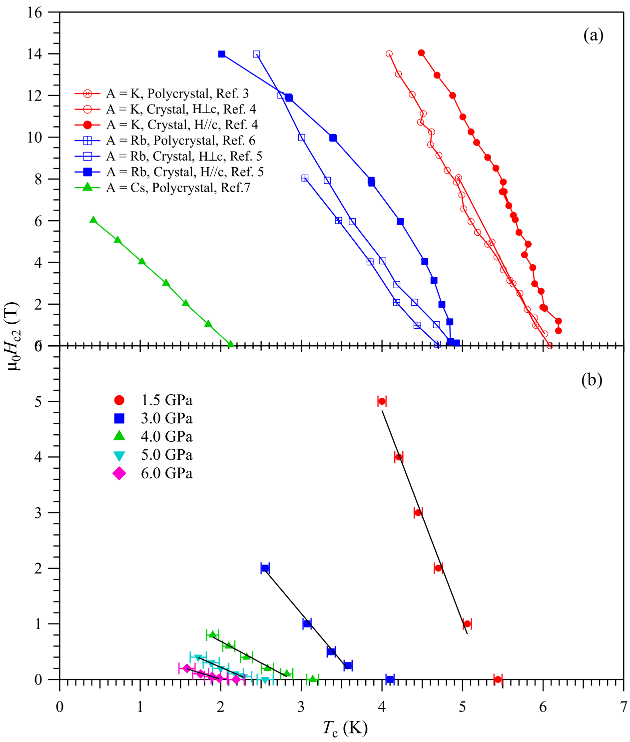

Figure 3. Upper critical filed μ_0H_{c2} of (a) $A_2Cr_3As_3$ (A = K [3, 4], Rb [5, 6], Cs [7]) at ambient pressure and (b) that of $K_2Cr_3As_3$ under various hydrostatic pressures up to 6 GPa. The solid lines in (b) represent a line fitting to the $\mu_0H_{c2}(T)$ data to extract the initial slope.

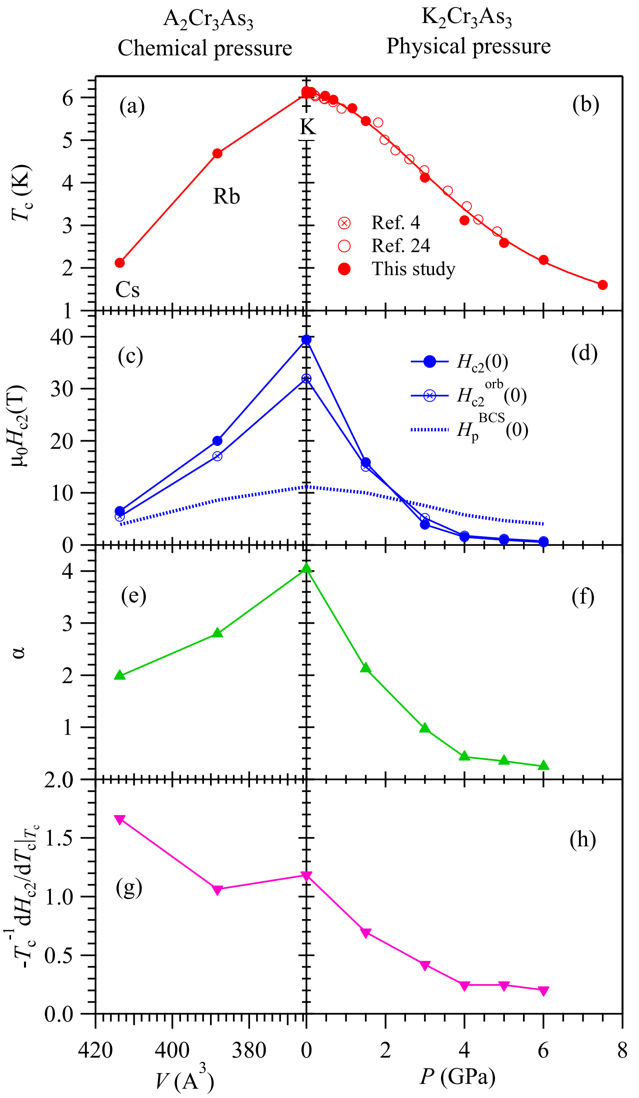

Figure 4. Comparison on the effects of chemical pressure versus physical pressure on the superconducting transition temperature T_c, upper critical field μ_0H_{c2}, Maki parameter α, and $-T_c^{-1}\mathrm{d}H_{c2}/\mathrm{d}T_c|_{T_c}$ of $A_2Cr_3As_3$ (A = K, Rb, Cs).

To quantify these changes under pressure, we have estimated the zero-temperature upper critical field $\mu_0H_{c2}(0)$ for each pressure by fitting the $\mu_0H_{c2}(T)$ with the empirical Ginzburg–Landau equation, *viz.* $\mu_0H_{c2}(T) = \mu_0H_{c2}(0)(1 - t^2)/(1 + t^2)$, where t is the reduced temperature T/T_c. Such a formula has been used to estimate $\mu_0H_{c2}(0)$ of $A_2Cr_3As_3$ polycrystalline samples at ambient pressure, and seems to be applicable for the randomly oriented crystals. It should be noted that although the presence of μ_0H_{c2} anisotropy and the limited temperature/field range for the present data should introduce a large error bar for our estimations of $\mu_0H_{c2}(0)$, its pressure dependence is not altered and does not influence our discussions below. The obtained $\mu_0H_{c2}(0)$ as a function of pressure for $K_2Cr_3As_3$ is displayed in figure 4(d), which also includes the Pauli paramagnetic limited $\mu_0H_p^{BCS}(0)$ and the orbital limited $\mu_0H_{c2}^{orb}(0)$ for comparison. The former was obtained as $\mu_0H_p^{BCS}(0) = 1.84\ T_c$ for a conventional BCS superconductor, while the latter was estimated from the initial slope of $\mu_0H_{c2}(T)$ according to $\mu_0H_{c2}^{orb}(0) = -0.73T_c|\mathrm{d}H_{c2}/\mathrm{d}T|_{T_c}$ in the clean limit of Werthamer–Helfand–Hohenberg model without spin–orbit coupling. As seen in figure 4(d), $\mu_0H_p^{BCS}(0)$ decreases gradually as $T_c(P)$, whereas $\mu_0H_{c2}^{orb}(0)$ exhibit a much faster drop with increasing pressure following the trend of $\mu_0H_{c2}(0)$. The most important finding of this present study is that $\mu_0H_{c2}(0)$ of $K_2Cr_3As_3$ changes from largely exceeding the Pauli paramagnetic limit for $P < 2\,\mathrm{GPa}$ to below the Pauli limit for $P > 2\,\mathrm{GPa}$. As seen in figure 4(d), at ambient pressure $\mu_0H_{c2}(0)$ is not only largely exceeding $\mu_0H_p^{BCS}(0)$ but also larger than $\mu_0H_{c2}^{orb}(0)$, implying that both orbital and spin-paramagnetic effects play a less important role for pair breaking in the multiband superconductor $K_2Cr_3As_3$. As mentioned above, this has been taken as an important indication for spin-triple pairing in $K_2Cr_3As_3$. The relationship of $\mu_0H_{c2}(0) > \mu_0H_{c2}^{orb}(0) > \mu_0H_p^{BCS}(0)$ still holds for $P = 1.5\,\mathrm{GPa}$, but their differences become much smaller. Similar situation was observed for the series of $A_2Cr_3As_3$ (A = K, Rb, Cs) with increasing the A-cation size, figure 4(c). But the relationship reverses completely for $P > 2\,\mathrm{GPa}$ in $K_2Cr_3As_3$, i.e. $\mu_0H_{c2}(0) \leqslant \mu_0H_{c2}^{orb}(0) < \mu_0H_p^{BCS}(0)$, implying that spin-paramagnetic effect becomes the major pair breaking mechanism under higher pressures. Since the Pauli paramagnetic limit is related to the Zeeman splitting for spin singlet pairs, the observed significant change of $\mu_0H_{c2}(0)$ thus points to a plausible crossover of superconducting pairing mechanism from exotic spin triplet to conventional spin singlet with increasing pressure.

This argument is further elaborated by the evolution of Maki parameter α defined as $\alpha = \sqrt{2}\frac{H_{c2}^{orb}(0)}{H_p(0)}$, which describes the relative importance of the orbital and spin-paramagnetic effects on μ_0H_{c2} for real materials. In general, $\alpha \ll 1$ since α is known to be the order of $\Delta(0)/E_F$, where the $\Delta(0)$ is the superconducting gap and E_F the Fermi energy. However, in real materials with a heavy electron mass or multiple small Fermi pockets, E_F can become quite small to result in $\alpha \geqslant 1$. In the case of $K_2Cr_3As_3$, α takes a large value of 4 at ambient pressure, signaling a pronounced mass enhancement as found from the bulk specific heat measurements [3]. With increasing pressure, α decreases quickly and becomes much smaller than 1 for $P > 2\,\mathrm{GPa}$. The reduced effective mass for $K_2Cr_3As_3$ under pressure can also be evidenced in figure 4(h), in which $-T_c^{-1}\mathrm{d}H_{c2}/\mathrm{d}T_c|_{T_c} \propto m^*$ decreases by an order of magnitude with pressure. These above observations confirmed a significant reduction of electron correlations under pressure. It has been proposed that the significant electron correlations in $K_2Cr_3As_3$ make it near a ferromagnetic instability [14], which can result in exotic spin-triplet superconductivity. The plausible pressure-induced crossover to spin-singlet pairing can be rationalized by considering the fact that the application of high pressure can weaken the electron correlations so as to drive the system far away from the ferromagnetic instability.

Based on the above results, we can comment briefly on the seemingly contradicted effects of physical versus chemical pressure on T_c. As seen in figure 4, T_c increases progressively in the series of $A_2Cr_3As_3$ (A = Cs, Rb, K) with reducing the

A-cation size or the unit-cell volume, but the application of physical pressure on $K_2Cr_3As_3$ that reduces further the volume does not enhance T_c. Although the difference becomes smaller with increasing the A-cation size, the relationship of $\mu_0 H_{c2}(0) > \mu_0 H_{c2}^{orb}(0) > \mu_0 H_p^{BCS}(0)$ still holds for the series of $A_2Cr_3As_3$ (A = K, Rb, Cs), in contrast to the reversal under physical pressure. In addition, the Maki parameter α remains larger than 1, and more importantly, the effective mass reflected by $-T_c^{-1}dH_{c2}/dT_c|_{T_c}$ does not vary significantly in comparison with the evolution under physical pressure. These side-by-side comparisons in figure 4 highlight a distinct effect of physical pressure with respect to the chemical pressure, which urges us to look into the details of the structural factors.

As pointed previously [5, 6], the replacement of A cations in $A_2Cr_3As_3$ (A = K, Rb, Cs) mainly changes the interchain distances but has negligible influence on the $[Cr_3As_3]_\infty$ chains that are believed to responsible for the observed superconductivity. This means that the effect of chemical pressure on the crystal structure is highly anisotropic. For instance, the lattice constant a and c of $Rb_2Cr_3As_3$ are, respectively, 3.2% and 0.5% larger than those of $K_2Cr_3As_3$. Correspondingly, the change of Cr–Cr bond distances is less than 0.6%, whereas the A–As bonds change 3–4% between $K_2Cr_3As_3$ and $Rb_2Cr_3As_3$ [5]. The reduced T_c with increasing the A-cation size should be attributed to the weakened interchain coupling [25]. Nevertheless, the nearly intact $[Cr_3As_3]_\infty$ chains maintains the similar pairing mechanism and strong electron correlations as manifested by the nearly parallel curves of $\mu_0 H_{c2}(T_c)$ in figure 3. In contrast, the application of physical pressure, especially the hydrostatic pressure in the present study, can have a profound effect on the $[Cr_3As_3]_\infty$ chains. Although the compressibility along the a and c axis are found to be anisotropic, the $[Cr_3As_3]_\infty$ chains are inevitably shortened progressively under pressure [24]. As shown in [24], the Cr–Cr distances decrease with pressure in a slope of about -0.014 Å GPa^{-1}. In such a case, the electron correlations are expected to be weakened substantially due to the bandwidth broadening with shorter Cr–Cr distances. This is consistent with the dramatic drop of $-T_c^{-1}dH_{c2}/dT_c|_{T_c}$ shown in figure 4(h). At the same time, a reduced density of states at Fermi energy due to a broaden bandwidth also drives the system away from the magnetic instability, which can rationalize the plausible crossover of superconducting pairing mechanism from spin triple to singlet. Indeed, first-principles calculations by Wu *et al* have found that the magnetism can be suppressed quickly by reducing the c axis under pressure [26].

As mentioned above, the observed unusually large $\mu_0 H_{c2}$ (~3–4 times of $\mu_0 H_p$) at ambient pressure has been regarded as a key sign for the unconventional spin-triplet superconductivity associated with the ferromagnetic correlations in $K_2Cr_3As_3$ [3, 10]. Later studies on the anisotropic $\mu_0 H_{c2}(T)$ provided either supportive or disapproved evidences for such an exotic mechanism [20, 21]. To the best of our knowledge, our present study is the *first* to investigate the pressure dependence of $\mu_0 H_{c2}$. Although the present study cannot settle down these contradicting results, our results on the evolution of $\mu_0 H_{c2}$ as a function of pressure provide new pieces of information; i.e. the significant reduction of $\mu_0 H_{c2}$, in particular, its evolution from far above the Pauli limit $\mu_0 H_p$ to well below $\mu_0 H_p$ with increasing pressure signals the possible breakdown of the spin-triplet pairing in $K_2Cr_3As_3$. In addition, our results illustrate that the relative strength of the spin-triplet and singlet pairing can be tuned via modifying the electronic/magnetic correlations with pressure for these novel Q1D superconductors.

Conclusions

In summary, we have elucidated the effects of hydrostatic pressure on the superconducting transition temperature T_c and the upper critical field $\mu_0 H_{c2}$ of the newly discovered quasi-1D superconductor $K_2Cr_3As_3$. We found that the application of hydrostatic pressure can reduce progressively T_c to below 2 K under 7.5 GPa. More interestingly, $\mu_0 H_{c2}(0)$ was found to decrease dramatically from largely exceeding the Pauli paramagnetic limit $\mu_0 H_p$ to well below it at ~2 GPa, signaling a plausible crossover of exotic spin-triplet to spin-singlet Cooper paring. Since $K_2Cr_3As_3$ is located near a magnetic instability with strong ferromagnetic fluctuations mediating the spin-triplet superconductivity, the application of hydrostatic pressure can drive the system away from the magnetic instability and lead to a breakdown of the spin-triplet pairing channel. We have also discussed the distinct effects of chemical and physical pressures on the superconducting properties of $K_2Cr_3As_3$.

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References

[1] Wu W, Cheng J-G, Matsubayashi K, Kong P P, Lin F K, Jin C Q, Wang N L, Uwatoko Y and Luo J L 2014 *Nat. Commun.* **5** 5508
[2] Kotegawa H, Nakahara S, Tou H and Sugawara H 2014 *J. Phys. Soc. Japan* **83** 093702
[3] Bao J-K *et al* 2015 *Phys. Rev.* X **5** 011013
[4] Kong T, Bud'ko S L and Canfield P C 2015 *Phys. Rev.* B **91** 020507
[5] Wang X F, Roncaioli C, Eckberg C, Kim H, Yong J, Nakajima Y, Saha S R, Zavalij P Y and Paglione J 2015 *Phys. Rev.* B **92** 020508
[6] Tang Z-T *et al* 2015 *Phys. Rev.* B **91** 020506
[7] Tang Z-T *et al* 2015 *Sci. China Mater.* **58** 16
[8] Zhi H Z, Imai T, Ning F L, Bao J-K and Cao G-H 2015 *Phys. Rev. Lett.* **114** 147004

[9] Watson M D *et al* 2017 *Phys. Rev. Lett.* **118** 097002
[10] Yang J, Tang Z T, Cao G H and Zheng G Q 2015 *Phys. Rev. Lett.* **115** 147002
[11] Pang G M *et al* 2015 *Phys. Rev.* B **91** 220502
[12] Adroja D T *et al* 2015 *Phys. Rev.* B **92** 134505
[13] Adroja D T *et al* 2017 *J. Phys. Soc. Japan* **86** 044710
[14] Jiang H, Cao G H and Cao C 2015 *Sci. Rep.* **5** 16054
[15] Wu X X, Yang F, Le C C, Fan H and Hu J P 2015 *Phys. Rev.* B **92** 104511
[16] Zhou Y, Cao C and Zhang F C 2015 *Sci. Bull.* **62** 208
[17] Zhong H T, Feng X-Y, Chen H and Dai J H 2015 *Phys. Rev. Lett.* **115** 227001
[18] Zhang L-D, Wu X X, Fan H, Yang F and Hu J P 2016 *Europhys. Lett.* **113** 37003
[19] Miao J J, Zhang F C and Zhou Y 2016 *Phys. Rev.* B **94** 205129
[20] Balakirev F F, Kong T, Jaime M, McDonald R D, Mielke C H, Gurevich A, Canfield P C and Bud'ko S L 2015 *Phys. Rev.* B **91** 220505
[21] Zuo H-K *et al* 2017 *Phys. Rev.* B **95** 014502
[22] Subedi A 2015 *Phys. Rev.* B **92** 174501
[23] Zhang W-L, Li H, Xia D, Liu H W, Shi Y-G, Luo J L, Hu J P, Richard P and Ding H 2015 *Phys. Rev.* B **92** 060502
[24] Wang Z *et al* 2016 *Sci. Rep.* **6** 37878
[25] Zhi H Z, Lee D, Imai T, Tang Z T, Liu Y and Cao G H 2016 *Phys. Rev.* B **93** 174508
[26] Wu X X, Le C C, Yuang J, Fan H and Hu J P 2015 *Chin. Phys. Lett.* **32** 057401