Gapless spin-excitations in the superconducting state of a quasi-one-dimensional spin-triplet superconductor

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Majorana zero modes form as intrinsic defects in an odd-orbital one-dimensional superconductor thus motivating the search for such materials in the pursuit of Majorana physics. Here, we present combined experimental results and first principles calculations which suggest that quasi-one-dimensional K$_2$Cr$_3$As$_3$ may be such a superconductor. Using inelastic neutron scattering we probe the dynamic spin-susceptibilities of K$_2$Cr$_3$As$_3$ and K$_2$Mo$_3$As$_3$ and show the presence of antiferromagnetic spin-fluctuations in both compounds. Below the superconducting transition, these fluctuations likely arise from nesting on one dimensional features of the Fermi surface. Considering these results we propose that while K$_2$Mo$_3$As$_3$ is a conventional superconductor, K$_2$Cr$_3$As$_3$ is likely a spin-triplet, and consequently, topological superconductor.

To realize scalable quantum computers, new phenomena on which to base the qubit are needed - ones robust and with intrinsic entangled properties such as exists in certain topological phases. Of the potential candidates, the Majorana zero mode (MZM) is one of the most promising due to its non-abelian anyon statistics which are suited for braiding while also potentially allowing for physical manipulation as is necessary for computation. However, generating and observing MZMs has proven challenging with several potential routes in active pursuit such as: at the interface of a topological insulator and a superconductor (SC), in a SC with a topological band structure, or in a SC whose pair operator is its own conjugate - a ‘spin-less’ or spin-triplet odd-orbital SC. Indeed, this last case (when restricted to one dimension) is related to the toy-model first proposed by Kitaev to generate physically separated MZM.

As a result, there is great interest in one-dimensional (1D) or quasi-1D (Q1D) systems which might exhibit spin-triplet SC (TSC) - especially those that exhibit both properties intrinsically. However, such materials are extraordinarily rare with few compounds showing either property and still fewer with both. Nonetheless, several candidate materials have been found (including the Bechgaard salts, purple bronze, and even possibly Sr$_2$RuO$_4$). More recently, the discovery of the Q1D potential TSC A$_n$H$_{2n}$TM$_4$As$_3$ (with A = Na,K,Rb or Cs, TM = Cr or Mo and n = 1 or 2) family has provided another route to realize these exotic physics.

The A$_n$H$_{2n}$TM$_4$As$_3$ materials exhibit numerous novel properties, several of which evince TSC. These materials crystallize with a Q1D structural motif of TM$_3$As$_3$ tubes which give rise to strongly Q1D features such as Luttinger-liquid physics, Q1D Fermi surfaces (FS) and highly anisotropic transport. Enticingly, their SC state appears to be unconventional with an unexpectedly high upper critical field, potential nodes in the SC gap, and a proximity to a quantum critical point with possible suggestions of TSC due to a spontaneous magnetization below the SC transition ($T_C$), an angular dependent upper critical field, ferromagnetic (FM) fluctuations, a $T_C$ suppressed by non-magnetic impurities and widespread findings of a leading TSC instability from theory.

However, the symmetry of the SC state, and thus the prospects for hosting MZM, remains disputed. While some studies support TSC, others have suggested a spin-singlet state. These investigations report anti-FM (AFM) instabilities, a proximity to a spin-glass state, a $s^\pm$ gap symmetry, and even claims of standard electron-phonon ($e$-$p$) coupling. Recently, it was proposed that the K$_2$TM$_3$As$_3$ family may straddle a boundary between an unconventional SC in K$_2$Cr$_3$As$_3$ ($T_C$$\sim$ 6 K) and a multi-gap conventional SC in K$_2$Mo$_3$As$_3$ ($T_C$$\sim$ 10 K), perhaps giving some guidance to understand the disparate reported features.

In this Letter, we assess the possibility of TSC in K$_2$Cr$_3$As$_3$ through a careful study of the dynamic spin susceptibilities of K$_2$Cr$_3$As$_3$ and K$_2$Mo$_3$As$_3$ using both experimental probes and first principles calculations. To start, inelastic neutron scattering (INS) experiments re-
veal spin-fluctuations (SF) in both compounds above $T_C$ which are consistent with an incipient AFM order. Below $T_C$, we find that for K$_2$Mo$_3$As$_3$ a resonanceless spin-gap opens while in K$_2$Cr$_3$As$_3$ no gap is observed implying a difference in the compounds’ SC states. Performing first principles calculations, we find that the AFM SF can be explained by FS nesting on one dimensional FSs. Consequently, we suggest that K$_2$Mo$_3$As$_3$ is an e-p SC whose low energy SF are suppressed due to the opening of SC gaps on all FSs. Contrastingly, the lack of a spin-gap in K$_2$Cr$_3$As$_3$ indicates that neither the AFM SF nor the associated FSs participate in SC leaving a single remaining FS which much be SC and is favorable to FM SF driven TSC. These conclusions support the scenario of FM driven TSC in K$_2$Cr$_3$As$_3$ and indicate its potential for hosting topological SC.

Large powder samples of K$_2$Cr$_3$As$_3$ and K$_2$Mo$_3$As$_3$ were synthesized using methods reported previously to obtain $\sim$ 10 g of materials per compound (see the supplemental materials (SM) for details) [20-27, 47]. Neutron powder diffraction (NPD) measurements were performed on the HB-2A diffractometer of Oak Ridge National Laboratory’s (ORNL) High Flux Isotope Reactor (HFIR) [51]. The resulting diffraction data were analyzed using the Rietveld method as implemented in the FullProf software Suite [52]. INS was performed on the HB-3 and CTAX triple axis spectrometers of HFIR using fixed analyzer energies of 14.7 and 5 meV respectively.

To calculate the electronic band structure, Density Functional Theory (DFT) calculations were performed using the generalized gradient approximation of Perdew, Burke and Ernzerhof (PBE) and the general potential linearized augmented plane-wave method as implemented in the WIEN2k code [55-57].

In fig. 1(a) we show the crystal structure of K$_2$Mo$_3$As$_3$ (space group P6$_m$2) which exhibits a unique Q1D structural motif comprised of two inequivalent, alternating, coaxial layers of Mo (and As) triangles. In fig. 1(b) we show a diffraction pattern of K$_2$Mo$_3$As$_3$ collected at 300 K together with a simulated pattern from our best-fit model (we note that we found no impurity phase in our diffraction data indicating the high quality of our sample). In the inset of fig. 1(b) we show a comparison of NPD patterns collected at 300 and 2 K demonstrating a lack of any significant changes which might be associated with the onset of magnetic order indicating that K$_2$Mo$_3$As$_3$ has no long-range magnetic order (see the SM for more discussion) [59].

Previously, K$_2$Cr$_3$As$_3$ was shown to have AFM SF arising from incipient k = (0, 0, $\frac{1}{2}$) type order. This was revealed as a column of scattering in the dynamic structure factor $S(q, \Delta E)$ (as probed via INS) which is proportional to the imaginary component of the spin-susceptibility [17, 57]. Such fluctuations and their temperature dependence offer significant insights to the SC state. Therefore, we now turn to similar experiments performed on K$_2$Mo$_3$As$_3$ and expand on our previous work on K$_2$Cr$_3$As$_3$ to compare their respective $S(q, \Delta E)$. Fig. 2(a) and (c) show the $S(q, \Delta E)$ of K$_2$Mo$_3$As$_3$ and K$_2$Cr$_3$As$_3$ collected at 20 and 10 K respectively (i.e. above either compound’s $T_C$). Here we focus on the low q and low $\Delta E$ region which is typically featureless at these temperatures for non-magnetic materials. However, for both materials a column of scattering is seen arising from $\sim$0.75 Å$^{-1}$. Such a signal is often indicative of incipient magnetic order caused by SF with a q characteristic of the incipient ordering vector [40-59, 52].

Qualitatively, the signal observed in K$_2$Mo$_3$As$_3$ is similar to that of K$_2$Cr$_3$As$_3$. Fitting the constant $\Delta E$ cuts of the two datasets with Gaussian functions, we find a slight shift in the position of the feature to lower q by $\sim$ 0.1 Å$^{-1}$ in K$_2$Mo$_3$As$_3$ compared to K$_2$Cr$_3$As$_3$ which is consistent with the larger c axis (see SM for details) [52]. Furthermore, the dispersion of the two signals is very similar (albeit they are both skewed due to the instrument’s resolution function). On the other hand, the fits reveal that the column in K$_2$Mo$_3$As$_3$ is broader in q by $\sim$ 20% and also is $\sim$ 30% weaker (though this is more difficult to reliably quantify between samples) which may indicate the fluctuations are shorter-ranged and a smaller fluctuating moment is present in K$_2$Mo$_3$As$_3$ - both of which have been suggested from prior DFT treatments [50]. Due to these considerations, we attribute the origin of this signal to similar causes as in K$_2$Cr$_3$As$_3$. 
FIG. 2. Inelastic neutron scattering spectrograms for K$_2$Mo$_3$As$_3$ at (a) 20 K and (b) 2 K and for K$_2$Cr$_3$As$_3$ at (c) 10 K and (d) 2 K. Intensity is in units of detector counts normalized to monitor counts. We note that the data showed in panel (c) includes data from ref. 47 but with additional counting statistics.

With the origin identified, we now turn to the temperature dependencies across $T_C$. Fig. 2(b) and (d) show the same region of $S(q, \Delta E)$ measured below $T_C$ at 2 K for both samples. Here a distinction between the two emerges. For K$_2$Cr$_3$As$_3$ the spectrograph looks qualitatively identical to the 20 K data set. In particular, no gap opens in the fluctuations despite the onset of SC. On the other hand, in K$_2$Mo$_3$As$_3$ (fig. 2(b)) there is a clear change in the column where the signal for $\Delta E < 7$ meV loses intensity. This observation is consistent with the opening of a SC gap which inhibits fluctuations below $2\Delta$ (i.e. the energy required to break a Cooper pair).

To characterize this feature more carefully, constant $q$ scans were taken at $q \sim 1.1 \, \text{Å}^{-1}$ above and below $T_C$ for both samples (fig. 3). For K$_2$Mo$_3$As$_3$ (fig. 3(a)), the gap becomes clear. While the 20 K data exhibit a constant increase in intensity below 5 meV (as the elastic line is approached), the 2 K data exhibit a qualitatively different behavior, dropping in intensity by $\sim 20\%$ below $\sim 5$ meV. Using the weak coupling Bardeen Cooper Schrieffer gap approximation (i.e. $\Delta(T = 0) = \frac{\Delta E}{2}k_B T_C$) we estimate $2\Delta$ as 6.2 meV which is consistent with our observed gap (a similar estimate is obtained using the empirical formula of $\omega_0 = 4.3k_B T_C$ with $\omega_0$ being the energy of the spin-gap in the SC state)[63]. In fig. 3(c) we show a difference curve between the 20 and 2 K data to remove background effects. Here, the gap is seen to open below $\sim 5$ meV and progressively widen to the lowest measured temperature of 2 K. We note that the shape of this curve may corroborate nodal or nodeless gap functions however, we do not believe our data are sufficient to allow such an analysis. We further associate this gap with $T_C$ by measuring the intensity at 1.05 Å$^{-1}$ and 3 meV as a function of temperature (fig. 3(b)) which shows the gap to close at $\sim 6$ K. This is a little below $T_C$ (10.4 K); however, the gap itself is a function of $T$ and so should become smaller than the certainty of our measurements before $T_C$ is exceeded.

Turning to K$_2$Cr$_3$As$_3$, we see discretely different behavior in the low energy spectrum (fig. 3(d)). Here, no obvious gap in the fluctuations is seen in the 2 K data.
If the gap size is estimated as before, $2\Delta \sim 3.7\text{ meV}$ and $\omega_0 \sim 2.2\text{ meV}$, both of which are within the limits of our energy resolution ($\sim 1\text{ meV}$). For comparison, in fig. 3(d) we plot an envelope showing the range equivalent to the percent change of the signal seen in $K_2Mo_3As_3$, demonstrating that, within our statistics, a similar decrease in intensity should be observable if present. Additional measurements were taken using a cold neutron triple-axis spectrometer to access lower energy transfers ($<1\text{ meV}$) and no gap was observed (see SM). Consequently, we take this observation to be a strong indication that no spin-gap opens in the SC state of $K_2Cr_3As_3$.

Such an observation has significant implications for the nature of SC in these systems as well as how it evolves between the two materials. That the SF in $K_2Cr_3As_3$ do not respond strongly to SC requires an explanation - naively SC should open a gap. Furthermore, though a spin-gap with an accompanying resonance has become a hallmark of unconventional SCs near magnetic order, here we see no evidence of a resonance above the gap in $K_2Mo_3As_3$ and no resonance or gap in $K_2Cr_3As_3$ undermining SF possible role in SC. To help interpret these observations we note that if the SF can be associated with specific features of the FSs then the presence (or absence) of a gap in those SF will directly correspond to the presence (or absence) of a gap on the associated FS. In a system such as $K_2Cr_3As_3$, where different FSs have different SC instabilities, such information can be key in determining the symmetry of the SC state.

To elucidate the origin of the SF, we consider the FSs of $K_2Mo_3As_3$ and (undistorted) $K_2Cr_3As_3$ (shown in fig. 4(a) and (b)) as determined by DFT calculations. Here we use undistorted $K_2Cr_3As_3$ due to ambiguity in the exact symmetry of the distorted structure. As reported, these two compounds have similar FSs, consisting of two Q1D $\alpha$ and $\beta$ sheets and one large 3D $\gamma$ sheet. Given the large sheet-like features of the FSs, nesting vectors have long been proposed as possible between both the upper and lower $\alpha$ and $\beta$ sheets as well as between the top and bottom surfaces of the $\gamma$ sheet any of which may lead to spin-or charge-density wave type orders with such a mechanism being proposed for the SF observed in $K_2Cr_3As_3$.

With the potential for FS nesting established, we next calculate the Fermi velocities ($v_F$) throughout the FSs of both compounds to predict the strength of electron correlations on the different surfaces (as shown in the color scale on fig. 4(a) and (b)). These calculations reveal two important features: for both compounds the large 3D $\gamma$ sheet has significantly lower $v_F$ indicating stronger correlated electron physics on this sheet. Additionally, $v_F$ is in general larger in $K_2Mo_3As_3$ suggesting weaker electron correlations. These results suggest both that magnetic interactions should be stronger on the $\gamma$ sheet and stronger in $K_2Cr_3As_3$ than in $K_2Mo_3As_3$ in general.

With the Fermiology indicating potential nesting, we turn to calculations of the imaginary component of the Lindhard susceptibility (which corresponds to the dynamic spin susceptibility) projected along $k_z$ for $K_2Mo_3As_3$ (fig. 4(c)) (for this purpose we used a very dense grid of $k$-points and the constant matrix element approximation). Peaks in the Lindhard susceptibility have previously been reported in $A_2Cr_3As_3$ and $ACr_3As_3$ and attributed to SF with a nesting vector along $k_z$ consistent with intra-band scattering. Here, we most prominently see a large broad peak near the zone center which corresponds to the FM SF observed in $K_2Cr_3As_3$. However, at larger $k_z$, near the zone boundary, we see a second feature at $k_z \sim 0.9$ which closely corresponds to the $k = (0, 0, \frac{1}{2})$ position for the AFM SF observed in INS. This peak is quite small which is consistent with it arising from nesting between the two high $v_F$ Q1D sheets.

These insights from first principles allow us to better interpret the experimental results. They show that both $K_2Mo_3As_3$ and $K_2Cr_3As_3$ have similar potential nesting vectors involving the 1D FSs consistent with the observed column of SF. That the AFM SF do not gap in $K_2Cr_3As_3$.
indicates that neither the AFM SF nor the 1D FSs participate in SC. This is expected as symmetry considerations for AFM SF mediated spin-singlet or spin-triplet SC disallow Cooper pairs between \( k_x \) and \(-k_x\) states. We can also eliminate AFM SF as a candidate mechanism in \( K_2Mo_3As_3 \) via the lack of a resonant-spin excitation in the SC state despite the observation of an SC gap. Turning to the upgapped 1QD FSs in \( K_2Cr_3As_3 \), this implies that SC must exist on the \( \gamma \) sheet. Given the strong FM SF present in \( K_2Cr_3As_3 \) which are known to gap below \( T_C \), this points to an interesting scenario. While AFM SF cannot pair \( k_x \) and \(-k_x\) states, FM SF do allow pairing of such states for a gap sign change as occurs in \( p_\pm \)-wave orbital symmetry. Furthermore, for FM SF the pairing potential is enhanced for low scattering vectors as found here on the \( \gamma \) surface which encompasses the zone center, consistent with the lower \(\nu_F\) found on this sheet. More generally, that both AFM and FM SF exist in \( K_2Cr_3As_3 \) but only the latter responds to SC is highly suggestive of TSC. Thus, our results are consistent with a \( p_\pm \)-wave TSC state in \( K_2Cr_3As_3 \) and encourage further work searching for MZMs, potentially pointing to a system which advantageously exhibits a \( p_\pm \)-wave state that avoids the singlet-triplet mixing, a highly 1QD crystal habit which may help with device design as well as in isolating such states, and predictions of other intrinsic topological band features.

In summary, we show that both \( K_2Cr_3As_3 \) and \( K_2Mo_3As_3 \) exhibit antiferromagnetic spin fluctuations which are consistent with an incipient \( k = (0,0,\frac{1}{2}) \) ordering vector. Comparing spectra collected above and below their respective \( T_{CS} \), we find that while \( K_2Mo_3As_3 \) exhibits a gap, \( K_2Cr_3As_3 \) exhibits no such gap. Furthermore, despite seeing a gap in \( K_2Mo_3As_3 \), we observe no evidence of a spin-resonance - the hallmark of spin-driven unconventional superconductivity. Using first principles calculations, we show that these two materials are susceptible to nesting across their Q1D Fermi surfaces consistent with a \( k = (0,0,\frac{1}{2}) \). As we observe no gap in the spin-fluctuations of \( K_2Cr_3As_3 \), we infer that these Fermi surfaces are not gapped by the superconducting state and that the remaining \( \gamma \) sheet, which should favor spin-triplet pairing, must host superconductivity. Furthermore, we rule out the antiferromagnetic and \( e-p \) coupling superconducting mechanisms in \( K_2Cr_3As_3 \), indicating that ferromagnetic fluctuation driven spin-triplet superconductivity is the likely mechanism. As \( K_2Cr_3As_3 \) is a Q1D material, its hosting spin-triplet superconductivity should have exciting implications for topological physics invoking aspects of Kitaev’s toy model for Majorana zero-modes.

Note: While this manuscript was in preparation another paper was published (ref. [52]) which came to similar conclusions via nuclear magnetic resonance measurements performed on a single crystal sample of \( K_2Cr_3As_3 \). These results and ours are quite complementary with the Knight shift providing a more direct measurement of the superconducting state and our measurements and analysis approaching the question of superconductivity in the \( A_2TM_3As_3 \) family more comprehensively.

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