

Observation of a neutron spin resonance in the bilayered superconductor $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$

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Abstract

We report inelastic neutron scattering (INS) investigations on the bilayer Fe-based superconductor $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ above and below its superconducting transition temperature $T_c \approx 28.9$ K to investigate the presence of a neutron spin resonance. This compound crystallises in a body-centred tetragonal lattice containing asymmetric double layers of Fe_2As_2 separated by insulating CaF_2 layers and is known to be highly anisotropic. Our INS study clearly reveals the presence of a neutron spin resonance that exhibits higher intensity at lower momentum transfer (Q) at 5 K compared to 54 K, at an energy of 15 meV. The energy E_R of the observed spin resonance is broadly consistent with the relationship $E_R = 4.9k_B T_c$, but is slightly enhanced compared to the values observed in other Fe-based superconductors. We discuss the nature of the electron pairing symmetry by comparing the value of E_R with that deduced from the total superconducting gap value integrated over the Fermi surface.

Keywords: iron-based superconductors, neutron spin resonance, inelastic neutron scattering


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

1. Introduction

The discovery of iron-based superconductors in 2006 by Kamihara *et al* [1] has attracted considerable interest in condensed matter physics. Since then many families of iron-based superconductors have been reported [2–5]. Despite the

availability of many experimental results and various proposed theoretical models [6, 7] the origin of the superconductivity and the nature of electron pairing symmetry in iron-based superconductors are still under debate. It is widely believed that the interband interactions between the hole pockets at the zone centre (Γ) and the electron pockets at the zone edges (M) play an important role in the electron pairing and superconductivity in iron-based superconductors. The mechanism driving superconductivity in the iron-based materials is currently thought to involve spin fluctuations which mediate the electron

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pairing. These spin fluctuations, mainly arising from the excitations between electron and hole pockets, may also give rise to a neutron spin resonance centred around an energy (E_R) which scales linearly with the superconducting transition temperature T_c with the relationship $E_R \sim 4.9k_B T_c$ in iron-based superconductors [4, 8]. This has been tested using a variety of iron-based superconductors which fall into the well-known 1111-, 111-, 11-, 112- or 122-type families [4, 9, 10]. Furthermore, an inelastic neutron scattering study on $\text{CaKFe}_4\text{As}_4$ (1144-family) reveals the presence of three spin resonance modes and their intensity varies with the value of the momentum transfer (L) along the c -axis; these are hence called odd and even modes of the spin resonance [11]. A very similar relation between E_R and T_c has been observed in the cuprate high temperature superconductors (where the pairing symmetry is confirmed to be mainly d-wave pairing) with $E_R \sim 5.8k_B T_c$ [12–17]. Furthermore, heavy fermion superconductors, which have very low transition temperatures (below 3 K), also exhibit a neutron spin resonance [18, 19].

The spin resonance mode is argued to be a collective mode originating from singlet–triplet excitations of the Cooper pairs, and thus the spin resonance can be considered to be a spin-exciton bound state at a resonant energy (E_R) below the pair breaking energy (2Δ) [8, 17, 20]. Specifically in iron-based superconductors, the spin resonance mode was believed to be a key piece of evidence for the sign-reversal of the order parameter through Fermi surface nesting within the so-called weak coupling scenario [6, 21]. For this scenario, E_R should be within the total superconducting gap Δ_{tot} which is obtained by summing over the nesting pockets linked by momentum Q : $\Delta_{\text{tot}} = |\Delta_k| + |\Delta_k + Q|$. Alternatively, a strong coupling approach, in which the pairing of electrons comes from short-range magnetic interactions, can give s^\pm -pairing and a spin resonance, while the gap function is completely different from the Fermi surface nesting picture [22, 23]. Although it is generally accepted that the resonance is a signature of unconventional superconductivity [24], there is no consensus on its microscopic origin. Moreover, the spin resonance in iron-based superconductors has also been proposed to be a collective excitation with a magnon-like dispersion, which was later confirmed by neutron experiments [25–28].

Very recently, a totally new structural family was synthesized with general formula $\text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2$, where $A = \text{K}, \text{Rb}, \text{Cs}$. They thus have what we can call a ‘12442 formula’ and are superconductors with values of T_{cs} up to 33 K [29, 30]. The $\text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2$ family of materials crystallise in a body-centred tetragonal lattice containing asymmetric double layers of Fe_2As_2 separated by insulating CaF_2 layers. The alkali metal cations A^+ are sandwiched between the Fe_2As_2 layers (see figure 1). Note that the alkali-metal-containing ‘122’ block is nominally hole doped with 0.5 holes/Fe, while the ‘1111’ block is nominally undoped. Consequently, the 12442-type compounds are all hole self-doped at a level of 0.25 holes/Fe. This leads to superconductivity without the need for extrinsic doping; in the case of extrinsic electron doping through Co/Fe substitution, superconductivity gradually disappears, accompanied by a sign change in Hall coefficient [31].

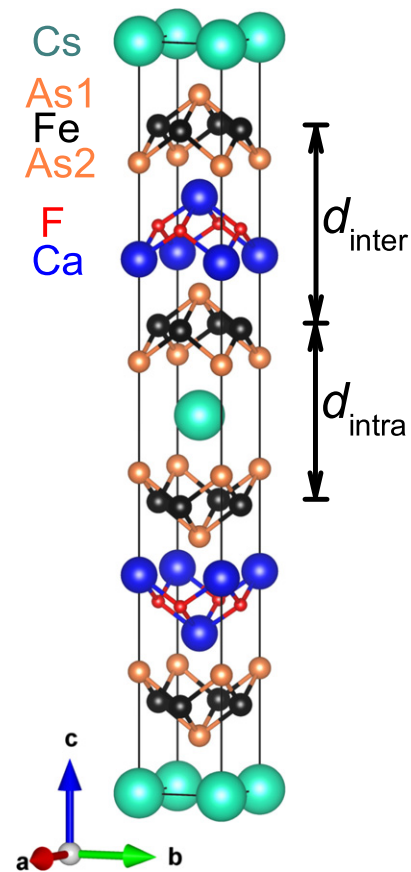


Figure 1. The crystal structure of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ with separate double Fe_2As_2 layers (after [30]). Adapted by permission from Springer Nature Customer Service Centre GmbH: Springer, Science China Materials [30].

By varying A , the structural parameters and atomic coordinates of almost all of the atoms vary [29, 30], and here we focus on $A = \text{Cs}$. It was found that the superconductivity is sensitive to these changes in the structural parameters, with T_c inversely correlating with the lattice parameters a and c . This contrasts sharply with other hole-doped Fe_2As_2 systems [32, 33] and so far the mechanism behind this variation is unknown. As the effective intra-layer coupling increases with a decrease in the lattice spacing, it is possible that T_c is either enhanced by inter-bilayer coupling or suppressed by intra-bilayer coupling, indicating that the superconducting properties may not be solely dependent on single FeAs layers [30]. First principles electronic structure calculations of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ reveal that the Fe-3d levels dominate the density of states at the Fermi level, and within these 3d states the contribution of e_g states is significant, demonstrating the multi-band nature of this superconductor [34]. The upper bound of the superconducting transition temperature, estimated using the electron–phonon coupling constant, is found to be 2.6 K. To produce the experimental value of transition temperature (28.9 K, see section 2), a 4–5 times increase in the electron–phonon constant is necessary, hinting that conventional electron–phonon coupling is not enough to explain the origin of superconductivity in this material [34].

This paper reports the discovery of a spin resonance mode in a powder sample of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ using inelastic

neutron scattering (INS). The compound contains the largest alkali atom in this family of superconductors and it has been characterised by muon-spin rotation which yielded a value for the in-plane penetration depth of $\lambda_{ab}(0) = 244(3)$ nm [35]. The temperature evolution of the penetration depth strongly suggested the presence of line nodes and is best modelled by a system consisting of both an *s*- and a *d*-wave gap [35] (with similar results for the Rb and K analogues [36, 37]), consistent with multiband behaviour. Recent single crystal measurements on $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ have highlighted the giant anisotropy of the upper critical field H_{c2} and the importance of two-dimensional fluctuations in this material [38].

2. Experimental details

A polycrystalline sample of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ was synthesized via the solid-state reaction and details are given in reference [29]. Our sample was found to have a sharp superconducting transition at 28.9 K, in agreement with the previous sample used in our μSR study [35]. The INS measurements were carried out using the high-neutron-flux MERLIN time-of-flight spectrometer at the ISIS facility [39]. The powder sample (mass ~ 4.1 g) was wrapped in a thin Al foil and mounted inside a thin-walled cylindrical Al can with diameter of 30 mm (and height of 40 mm), which was cooled down in a closed cycle refrigerator (CCR) with He-exchange gas around the sample. The measurements were performed with an incident neutron energy (E_i) 50 meV with chopper frequency of 350 Hz, which gave an elastic resolution of $\Delta E = 2.0$ meV. The measurements were carried out at 5 K (below T_c) and at 54 K (above T_c) with counting time of 22 hours per temperature point. The low-angle detectors on MERLIN are contaminated from direct neutron beam divergence and lead to a background which is weakly time-dependent, leading to a background which fails to subtract in difference plots. These detectors, below 7° scattering angles, were removed in our data analysis.

3. Results

Figures 2(a) and (b) shows 2D colour maps of the inelastic neutron scattering response, plotted as momentum transfer versus energy transfer, from the $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ powder sample measured at below T_c , 5 K and above T_c , 54 K. It is clearly seen that there is more scattering at low- Q near 15 meV in the 5 K data, than that of 50 K data (see figure 3 for 1D cuts). We attribute the excess scattering near 15 meV at 5 K compared with 54 K to the presence of the spin resonance mode below T_c . Note that the intensity of the spin resonance mode in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ seems weaker compare to that observed in $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ [9] and $\text{CaKFe}_4\text{As}_4$ [11, 40]. In order to see clearly the presence of the spin resonance and excess magnetic scattering intensity at 5 K compared with 54 K, we have taken the temperature difference of the two data sets, i.e. 5 K minus 54 K. Figure 4(a) shows the 2D colour map of the difference, 5 K–54 K, inelastic neutron scattering intensity, which clearly reveals the presence of magnetic scattering near 15 meV. The magnetic nature of the scattering can be deduced by noting

the fall in the intensity of this feature with increasing momentum transfer. The energy-integrated (between 13 and 17 meV) one-dimensional momentum (Q) cut showed a peak at $Q = 1.35(2) \text{ \AA}^{-1}$ [see figure 4(b)].

Figure 4(c) presents an energy cut through the temperature difference data (5 K minus 54 K) from low scattering angles, integrated over 7–20 degrees, that again reveals the presence of a clear sign of the spin resonance at ~ 15 meV. We note that the cut was made in scattering angle rather than Q to avoid the spurious scattering from the first few detector tubes at low scattering angles. It is clear from figure 4(c) that the integrated scattering intensity between 12 meV and 20 meV shows positive values. It is very difficult from our data to determine whether there is more than one resonance peak present in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$. Considering the double layer structure of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$, we would expect more than one resonance peak in the 12442-family due to multiple Fe *d*-bands contributing at the Fermi level. This has been supported through first-principle calculations on electronic structure and magnetic properties of $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ [41], which reveal a more complicated Fermi surface than other FeAs-based superconductors. Here there are ten bands crossing the Fermi level in the nonmagnetic state, resulting in six hole-like Fermi surface sheets along Γ – Z and four electron-like sheets along XP . The fixed spin moment calculations and the comparisons between total energies of different magnetic phases indicate that $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ has a strong tendency towards a stripe anti-ferromagnetic state. It has been found that the self-hole-doping suppresses the spin-density wave (SDW) state, inducing superconductivity in the parent compound $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ [41]. Furthermore, a comprehensive angle-resolved photoemission spectroscopy (ARPES) study on $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ shows divergent superconducting gaps with splitting electron bands on different sized Fermi pockets [42].

It is interesting to note that three resonance peaks were observed in the inelastic neutron scattering study on single crystals of $\text{CaKFe}_4\text{As}_4$ due to the very different superconducting gaps on the nesting pockets [11]. Furthermore, a powder inelastic neutron scattering study of $\text{CaKFe}_4\text{As}_4$ [40] also reveals a double peak type structure, near ~ 10 meV and ~ 15 meV, with magnetic intensity extending up to ~ 25 meV. The presence of multiple spin resonances has been also supported by a random phase approximation (RPA) calculation on $\text{CaKFe}_4\text{As}_4$ that reproduces the broad spectral features of the spin resonance anticipated as a result of multiple superconducting gaps on the different Fermi surface sheets in $\text{CaKFe}_4\text{As}_4$ [40]. It is observed for all the Fe-based compounds that the excess intensity due to the spin resonance mode disappears at T_c . A more detailed study of the temperature dependence of the spin resonance in our material would be an interesting subject for a future study which would be most informative if it could be carried out on single crystal samples.

We note that the resonance peak we observe at $Q = 1.35(2) \text{ \AA}^{-1}$ is larger than the wave vector from the Γ to the M point ($|\mathbf{Q}| = |(0.5, 0.5, 0)|_{\text{r.l.u.}} = 1.15 \text{ \AA}^{-1}$), which is usually the zone centre of magnetic fluctuations in 122 and 1144-type iron-based superconductors. However, if we consider an incommensurate resonance peak along the H direction, as

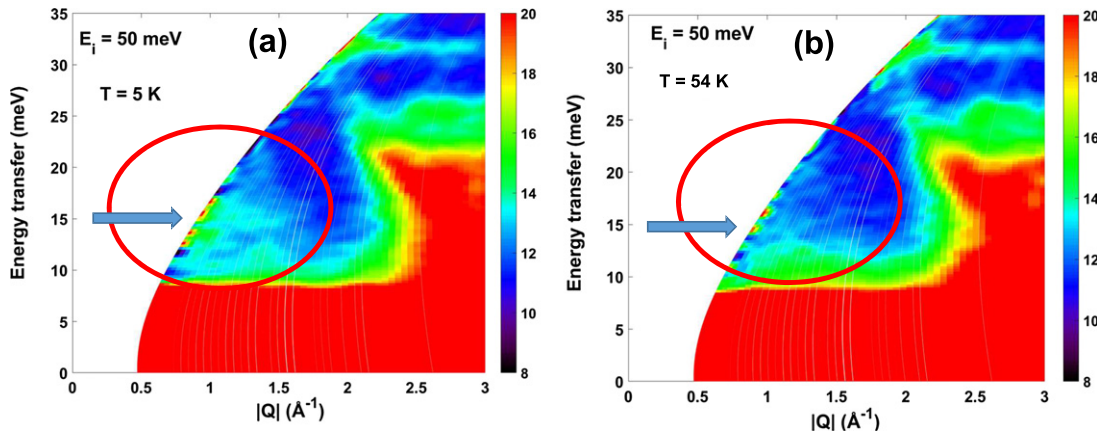


Figure 2. Inelastic scattering data at (a) 5 K and (b) 50 K. There is more scattering near 15 meV at 5 K than at 50 K, shown by arrow and circle, indicating the presence of a spin resonance. The incident neutron energy is 50 meV.

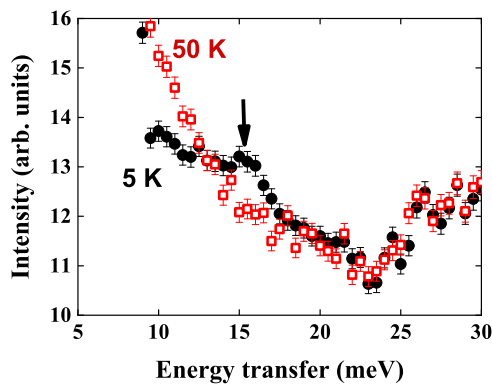


Figure 3. One-dimensional cuts taken from the data presented in figure 2 for scattering angles between 7 and 20 degrees, to avoid bad detectors at low angles. The presence of the spin resonance at ~ 15 meV is clear in the 5 K data. The data are from incident neutron energy of 50 meV.

is found in single crystal results on $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ [45], a reasonable explanation can be given. For example, assuming incommensurate resonance peaks at $\mathbf{Q} = (0.4, 0.4, 0)$ and $\mathbf{Q} = (0.6, 0.6, 0)$, respectively observed in a $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ single crystal, we also expect that the spin resonance in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ may originate from the same two Q positions. However, due to the limitation on the lowest detector angle of 7 degrees in the present data set, the lower- $Q = (0.4, 0.4, 0)$ (i.e. about 0.92 \AA^{-1}) side for the resonance peak is inaccessible and, we have mainly measured the higher- Q peak at $\mathbf{Q} = (0.6, 0.6, 0)$ (i.e. about 1.37 \AA^{-1}) of the resonance.

4. Discussion

The observation of a neutron spin resonance is usually taken as crucial evidence for spin-fluctuation mediated superconducting pairing in unconventional superconductors [4]. The spin resonance mode is argued to be a collective mode from singlet-triplet excitations of the Cooper pairs, so that the spin resonance can be considered as a spin-exciton bound state at resonant energy (E_R) below the pair breaking energy (2Δ) [8, 17, 20]. For s^\pm pairing symmetry the spin resonance

energy $E_R < 2\Delta(k, Q) = |\Delta_k| + |\Delta_{k+Q}|$, where the gap function is $\Delta_k = \Delta_0 \cos k_x \cos k_y$ or $\Delta_k = \Delta_0 (\cos k_x + \cos k_y)/2$. On the other hand for s^{++} pairing symmetry $E_R > 2\Delta(k, Q)$, so that a resonance-like hump with broad energy distribution can emerge above $2\Delta(k, Q)$ (maximum is $2\Delta_0$). The superconducting gaps estimated from the μSR for $s + d$ wave pairing in this compound are $\Delta^s = 7.5$ meV and $\Delta^d = 1.5$ meV, which would imply $E_R \geq 2\Delta(k, Q)$. These results may challenge the spin-exciton picture for the spin resonance under s^\pm pairing symmetry [43, 44], and probably suggest s^{++} pairing symmetry in the superconducting state of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$. Moreover, it is argued that the multiple superconducting gaps in $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ can be described by a gap function under the strong coupling picture [42], and its resonant energy also exceeds the measured maximum total gap. A similar situation in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ may also give a resonance mode beyond the spin-exciton picture [45]. Further investigation on the gap function in momentum space is highly desirable to clarify this issue.

In bilayer iron-based superconductors such as $\text{CaKFe}_4\text{As}_4$, two kinds of resonance modes with odd and even L -modulations are observed [11], much like the cases in cuprates [13–16]. However, this is may not the case in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$, even though it has a similar bilayer structure, and the resonance peak for the powder sample may be broadened due to average effect from L -modulations (figure 4(c)). Because both the intra-bilayer and inter-bilayer distances of $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ are large ($d_{\text{intra}} = 8.595 \text{ \AA}$, $d_{\text{inter}} = 7.6 \text{ \AA}$, see figure 1) [30], the magnetic interaction within the bilayer is likely decoupled, giving a degenerate spin resonance without any L -modulations. Indeed, a two-dimensional spin resonance was recently discovered in $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ [45], which is incommensurate around $\mathbf{Q} = (0.5, 0.5)$ in the $[H, K]$ plane. This is also consistent with the appearance of a spin resonance at $Q = 1.35 \text{ \AA}^{-1}$ in our results. As discussed above, the deviation of Q from $(0.5, 0.5)$ is more reasonable to explain by an in-plane incommensurability rather than via L -modulations, since the momentum transfer Q changes very little when increasing L within one Brillouin zone due to the large d -spacing along the c -axis ($c = 32.363 \text{ \AA}$).

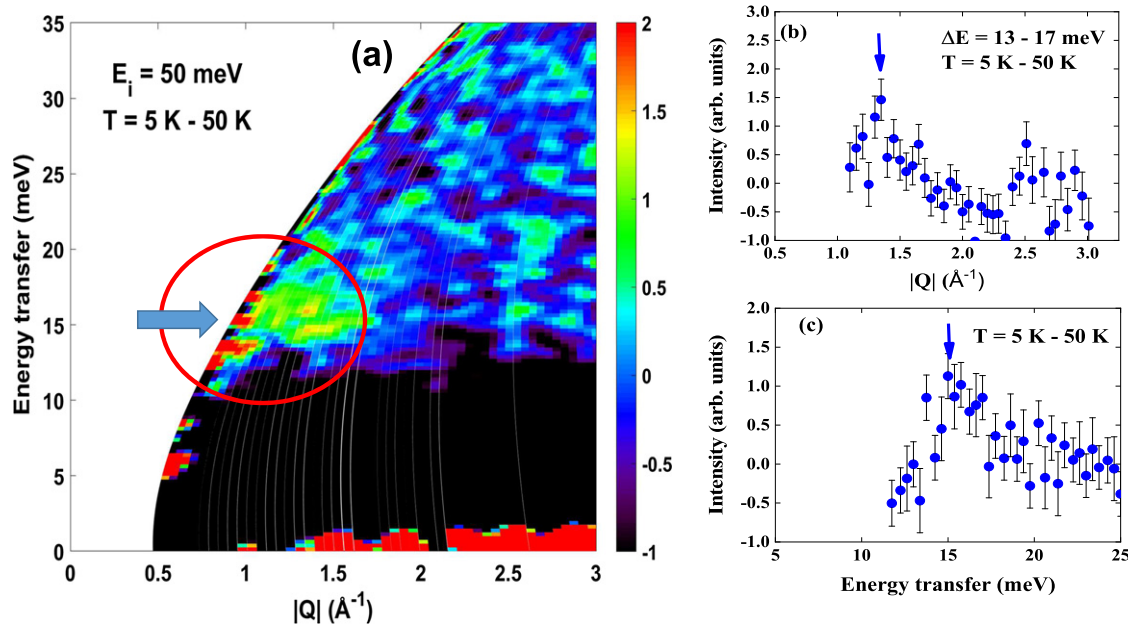


Figure 4. (a) The temperature difference colour map of the scattering intensity, between the 5 K and 50 K data is plotted as energy transfer vs momentum transfer. The arrow and circle indicate the presence of spin resonance and, (b) one-dimensional Q -cut taken from the data presented in (a) energy integrated between 13 and 17 meV. (c) One-dimensional energy-cut taken from the data presented in (a) for scattering angles between 7 and 20 degrees, to avoid bad detectors at low angles. The presence of the neutron spin resonance at ~ 15 meV is clear in the temperature difference data.

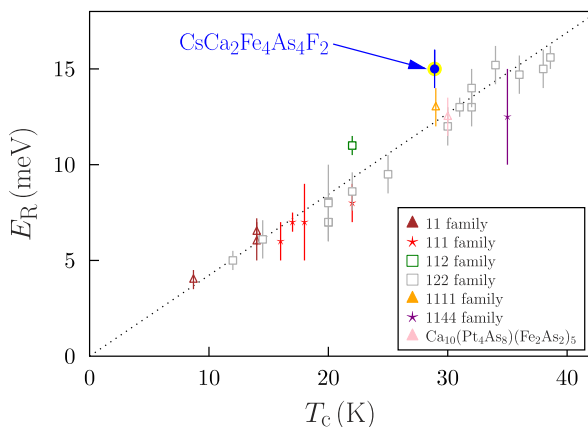


Figure 5. The neutron spin resonance energy E_R as a function of T_c for a collection of iron-based superconductors collated from [8, 46], together with the new data point from the present work. The straight line assumes the relationship $E_R = 4.9k_B T_c$. Adapted figure with permission from [8], Copyright (2018) by the American Physical Society.

As has been observed for many Fe-based superconductors the value of neutron spin resonance energy E_R is directly related to T_c . For $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ we have $E_R/(k_B T_c) = 6.02$, which is slightly higher than the value of 4.3–4.9 observed for other iron-based superconductors [46–50]. Recently the neutron spin resonance has also been investigated in $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ single crystals with $T_c = 33.5$ K [45], which reveals a spin resonance $E_R = 16$ meV. This give the ratio $E_R/(k_B T_c) = 5.5$ for $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ in broad agreement with what we have observed for $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$. The neutron spin resonance found in $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ can be plotted on a graph of E_R against T_c (see figure 5) and compared with

data obtained on a variety of other iron-based superconductors (collected in [11]). The new data point falls close to the correlation line $E_R = 4.9k_B T_c$, but with a small enhancement, possibly associated with the complex gap function in terms of a splitting on specific bands and strong bilayer couplings [42] (which would apply also to $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$). We note that the correlation found for d-wave pairing symmetry in the cuprates is $E_R = 5.8k_B T_c$, so that this enhancement may be connected with the s + d-pairing symmetry in this iron-based compound, which requires further studies on the momentum-resolved superconducting gaps. An alternative explanation arises from the highly two-dimensional nature of this superconductor. The resulting large anisotropy may be associated with weak coupling along the c -axis, which could reduce the bulk T_c value without similarly affecting E_R , thereby slightly enhancing the value of $E_R/k_B T_c$.

5. Conclusion

We have reported inelastic neutron scattering (INS) results on the bilayer Fe-based superconductor $\text{CsCa}_2\text{Fe}_4\text{As}_4\text{F}_4$ above and below its superconducting transition temperature $T_c \approx 28.9$ K to investigate the presence of the neutron spin resonance. Our INS study clearly reveals the presence of a neutron spin resonance, showing higher intensity at lower momentum transfer (Q) at 5 K compared to 54 K, at energy of 15 meV. The energy E_R of the observed spin resonance is broadly consistent with the relationship $E_R = 4.9k_B T_c$ that has been observed in other Fe-based superconductors, but our value is slightly enhanced, consistent with an effect observed in a $\text{KCa}_2\text{Fe}_4\text{As}_4\text{F}_2$ single crystal sample. This enhancement needs further investigation but we suggest that it may be an

effect associated either with the nature of the pairing found in this structural type or with the high two-dimensionality.

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