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Anisotropic effect of a magnetic field on the neutron spin resonance in FeSe

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We use inelastic neutron scattering to study the effect of a magnetic field on the neutron spin resonance ($E_r = 3.6$ meV) of superconducting FeSe ($T_c = 9$ K). While a field aligned along the in-plane direction broadens and suppresses the resonance, a c-axis aligned field does so much more efficiently, consistent with the anisotropic field-induced suppression of the superfluid density from the heat capacity measurements. These results suggest that the resonance in FeSe is associated with the superconducting electrons arising from orbital selective quasiparticle excitations between the hole and electron Fermi surfaces.

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Conventional Bardeen-Cooper-Schrieffer (BCS) superconductivity in materials such as aluminum and tin emerges from the pairing of electrons through phonon-mediated attractions and is associated with the opening of an isotropic superconducting gap in reciprocal space below T_c [1]. Although there is no consensus for a microscopic theory, high-transition-temperature (high-T_c) superconductivity in copper- and iron-based materials, derived from their antiferromagnetic (AF) ordered parent compounds [2,3], is believed to arise from interactions between itinerant electrons mediated by spin fluctuations [4]. One of the key signatures is the appearance of a neutron spin resonance mode, a collective spin excitation with an intensity tracking the superconducting order parameter below T_c [4–6]. The energy of the resonance, E_r, in different superconductors is proportional to either T_c or superconducting gap amplitude [7–9].

In the weak-coupling itinerant electron picture, the resonance is a bound state (spin exciton) appearing below the particle-hole continuum at a momentum transfer **Q** that connects parts of the Fermi surface exhibiting a sign change in the superconducting order parameter [5]. For copper oxide superconductors, which are single-band superconductors with d-wave gap symmetry [10–12], the resonance peaks at the in-plane AF wave vector $\mathbf{Q}_{\mathrm{AF}} = (0.5, 0.5)$ and displays hourglasslike dispersion around $\mathbf{Q}_{\mathrm{AF}}$ consistent with expectations of the spin-exciton picture [13–16]. In the absence of (or for very weak) spin-orbit coupling (SOC) [17], the resonance is isotropic in spin space and arises from the spin-1 singlet-triplet excitations of the electron Cooper pairs [5,18]. When a magnetic field is applied, the spin-1 of the resonance should split into three energy levels following the Zeeman energy $E_\pm = E_r \pm g\mu_B B$ (at energies $E_r - g\mu_B B$, E_r, and $E_r + g\mu_B B$) [Fig. 1(a)], where $g = 2$ is the Landé factor, B is the magnitude of the field, and μ_B is the Bohr magneton. On the other hand, if superconductivity coexists with AF order or there is large SOC-induced anisotropy, the resonance can be a doublet where the application of a magnetic field will split the mode into two peaks [Fig. 1(b)] [18], as seen in the heavy fermion superconductor $CeCoIn_5$[19,20]. However, the application of a 14 T magnetic field approximately along the c axis in cuprate superconductors, where T_c and the superconducting gap is two orders of magnitude larger, only slightly suppresses the intensity of the resonance with no evidence for Zeeman splitting [21,22].

In the case of iron-based superconductors [23], where electrons in an Fe 3d t_{2g} band with d_{xz}, d_{yz}, and d_{xy} orbitals are near the Fermi level, superconductivity may occur in multiple orbitals through the hole-electron Fermi surface nesting [24]. As a consequence, the resonance can have more than one component in energy [25,26] and be anisotropic in spin space due to SOC [27–29]. Since the effect of Zeeman energy for a maximum possible applied field of 14 T is still small compared with the intrinsic energy width of the resonance for optimally doped iron pnictide/chalcogenide superconductors [30,31], there is no confirmed evidence of Zeeman field-induced triplet splitting of the resonance [32–35]. Nevertheless, a c-axis aligned magnetic field suppresses the intensity of the mode much more efficiently than for an in-plane field [31,32]. These results are consistent with lower upper critical fields required to suppress superconductivity in c-axis aligned fields [23], suggesting that the intensity of the resonance is a measure of superconducting electron pairing density [36].

To further test if the resonance in iron-based superconductors is a spin exciton and associated with singlet-triplet or singlet-doublet transition [Figs. 1(a) and 1(b)], we need

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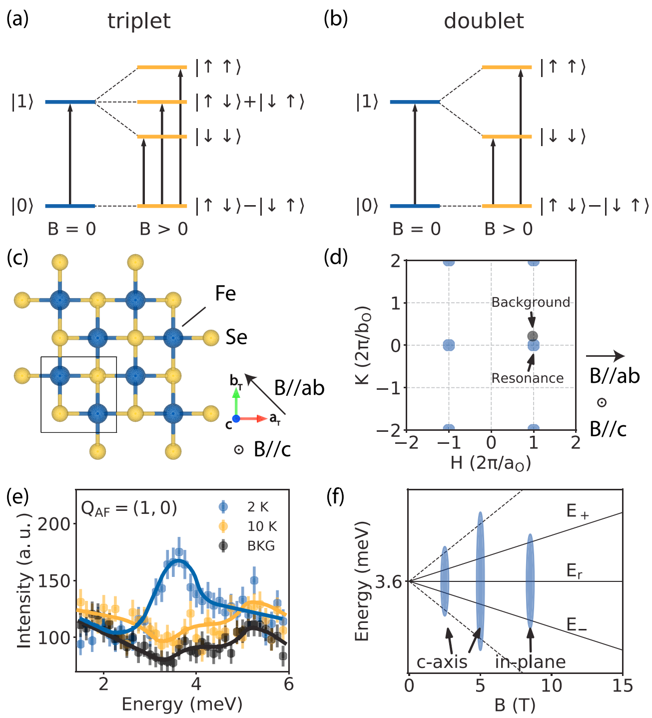

FIG. 1. (a) Schematic illustration of the Zeeman splitting of the spin exciton from singlet $|0\rangle$ to triplet $|1\rangle$ excited states. (b) Schematic illustration of a singlet-to-doublet excitation. (c) Crystal structure of FeSe. (d) Reciprocal space where the blue dots represent the $\mathbf{Q}_{\rm AF} = (1, 0)$ wave vector. The background position at $\mathbf{Q}_{\rm bkgd} = (0.977, 0.213, 0)$ is marked as a small circle. (e) PANDA measurements of the energy dependence of the scattering below (blue circles) and above (yellow circles) T_c at $\mathbf{Q}_{\rm AF} = (1, 0)$. The background scattering is shown as black circles. The error bars indicate statistical errors of one standard deviation. (f) Schematic of normalized peaks and excitation positions of the resonance in FeSe as a function of increasing magnetic field. Solid lines are $E_\pm = E_r \pm 2\mu_B B$ and E_r. Dashed lines are guides to the eye for a c-axis aligned field.

to find a clean material with relatively low T_c and a sharp resonance in energy without multiorbital effects. FeSe, which undergoes a tetragonal-to-orthorhombic structural transition at $T_s = 90$ K, forms a nematic phase below T_s, and becomes superconducting at $T_c = 9$ K [37–40], is an excellent choice for several reasons [Figs. 1(c) and 1(d)]. First, the compound is known to be extremely clean and has a relatively low resonance energy of $E_r = 3.6$ meV [41]. Second, superconductivity in FeSe is orbital selective and occurs mostly through hole-electron Fermi surface nesting of quasiparticles with d_{yz} orbital characters [42], resulting in a resonance only at the in-plane AF wave vector $\mathbf{Q}_{\rm AF} = (1, 0)$ [43]. Third, neutron polarization analysis of the resonance reveals that the mode is anisotropic in spin space and essentially c-axis polarized due to SOC [44], suggesting that a magnetic field cannot split the mode into triplets. Finally, the upper critical fields to suppress superconductivity in FeSe are around 16 and 28 T for the c-axis and in-plane aligned fields, respectively [44,45], meaning that an applied magnetic field will have a larger impact on superconductivity compared with that of optimally doped iron pnictides.

We carried out inelastic neutron scattering experiments to study the effect of a magnetic field on the resonance of FeSe using the multiaxis crystal spectrometer (MACS) at NIST Center for Neutron Research [46], and the cold neutron triple-axis spectrometer PANDA at Heinz Maier-Leibnitz Zentrum, Germany [47]. The c-axis aligned magnetic field experiments were performed on MACS with a fixed $E_f = 3.7$ meV and PANDA with a fixed $E_f = 5.1$ meV. The vertical magnetic fields were aligned along the [0,0,1] direction perpendicular to the $[H, K, 0]$ scattering plane. The in-plane magnetic field experiment was performed on MACS with the same instrumental setup, while the sample was aligned in the $[H, 0, L]$ scattering plane with the field along the [0,1,0] direction. Since an in-plane magnetic field will not produce orbital current within the FeSe layer, its effect on the resonance will be mostly the Zeeman effect.

At zero field, superconductivity in FeSe induces a resonance at $E_r \approx 3.6$ meV and a spin gap of about 2.8 meV as shown from data obtained on PANDA [Figs. 1(e) and 1(f)] [41,43,44]. The effect of an 8.5 T in-plane magnetic field on the resonance and low-energy spin excitations is shown using data obtained on MACS. Figures 2(a)–2(d) show constant-energy scans along the [1,0] direction at different energies with 8.5 T and zero magnetic fields in the superconducting state ($T = 2$ K). At $E = 2.5$ meV, an 8.5 T field induces magnetic scattering near $\mathbf{Q}_{\rm AF} = (1, 0)$ above the flat background, indicating a reduction in spin gap energy [Fig. 2(a)]. Near the resonance around $E = 3.5$ [Fig. 2(b)], the field suppresses the resonance as expected. Above the resonance energy at $E = 4.5$ and 5.5 meV, the applied field has little effect on the magnetic scattering [Figs. 2(c) and 2(d)]. Figures 2(e) and 2(f) show the two-dimensional (2D) wave-vector-energy images of the resonance above background scattering at zero and 8.5 T field, respectively [48]. The effect of an 8.5 T in-plane magnetic field is to weaken and broaden the resonance, with no convincing evidence for the splitting of the mode. Figure 2(g) is a cut along the energy direction at $\mathbf{Q}_{\rm AF} = (1, 0)$, which reveals the resonance at 0 T field. The identical cut at 8.5 T is shown in Fig. 2(h).

Figure 3 illustrates the effect of a 5 T c-axis aligned magnetic field on the resonance. Figures 3(a)–3(d) show constant-energy scans along the [1,0] direction with different energies in 5 T and zero magnetic fields in the superconducting state ($T = 2$ K). At $E = 2$ meV, a 5 T c-axis aligned field induces magnetic scattering near $\mathbf{Q}_{\rm AF}$, which is 1.6 meV below the spin resonance energy E_r. Off the resonance energy at $E = 3$ and 4 meV and above the resonance energy at $E = 5$ meV, the applied field has slight effect on the resonance. Figures 3(e) and 3(f) compare the 2D images of the wave-vector and energy dependence of the spin resonance in 0 and 5 T, respectively. For a c-axis aligned magnetic field, the upper critical field $B_{c2}(\perp)$ is around 16 T, meaning that a 5 T field is already ∼31% of B_{c2}, which is similar to the fraction of 30% achieved for the 8.5 T in-plane oriented field given the 28 T critical field. Although qualitatively the broadening in energy is similar to that of the in-plane field, the amplitude of the broadening is more significant. Figures 3(g) and 3(h) show the constant-$\mathbf{Q}$ cuts at the $\mathbf{Q}_{\rm AF}$ position from (e) and (f), respectively. We see that an applied field shifts the magnetic spectral weight to lower energies. By comparing Figs. 2(g),

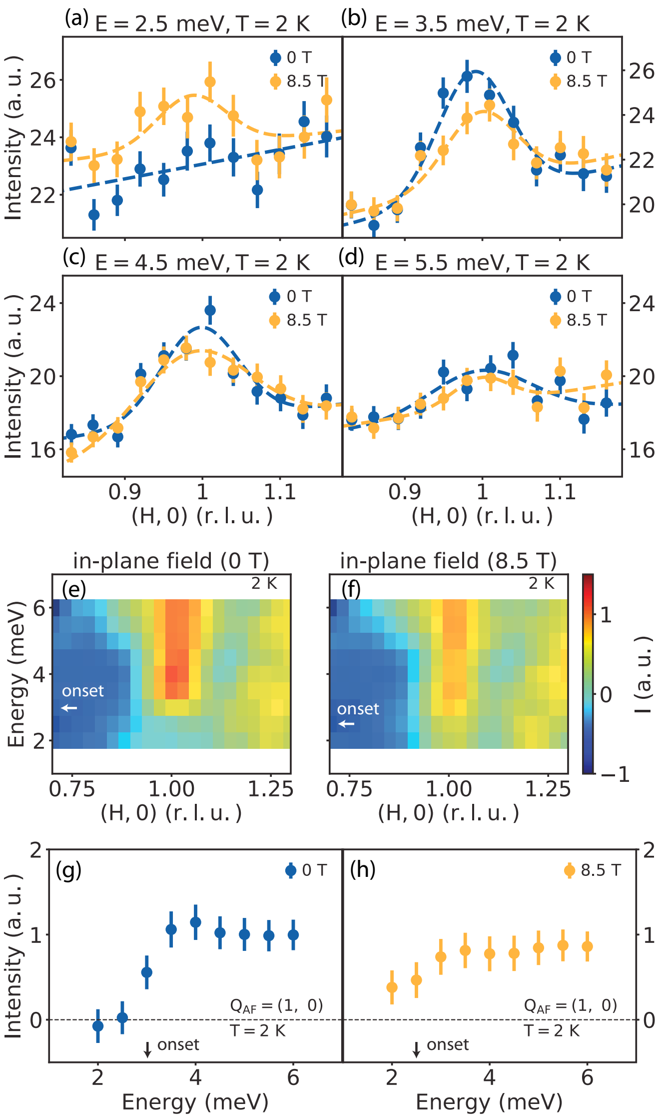

FIG. 2. (a)–(d) Constant-energy scans along the [1,0] direction at $E = 2.5$, 3.5, 4.5, and 5.5 meV in zero and 8.5 T in-plane magnetic fields at $T = 2$ K. (e) and (f) 2D images of wave-vector and energy dependence of the spin fluctuations in 0 and 8.5 T in-plane magnetic fields at $T = 2$ K. (g) and (h) are constant-**Q** cuts at the $\mathbf{Q}_{AF}$ position from (e) and (f), respectively. They have been smoothed two times by the 2D data processing method in the DAVE-MSLICE program at NCNR. The arrows in (e)–(h) indicate the lowest energy where a Gaussian can be fit to the data. The scattering of an assembly of Al plates coated with CYTOP, as well as a constant adjusted to force the scattering at $E = 2.4$ meV and $\mathbf{Q}_{AF}$ to be zero [Fig.4(b)], was subtracted as background in (e)–(h) [48]. The monitor counts in (e)–(h) are normalized to an arbitrary unit (a.u.) and can be compared directly. L is integrated in all panels, because spin fluctuations have no c-axis modulations in FeSe. The error bars indicate statistical errors of one standard deviation.[1]

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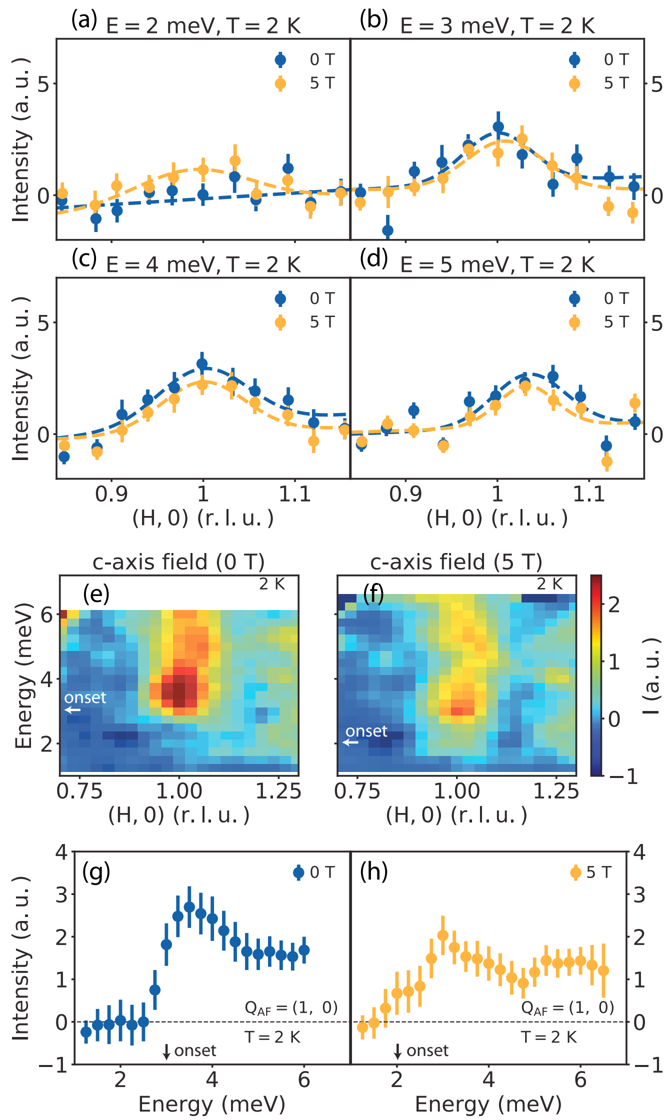

FIG. 3. (a)–(d) Constant-energy scans along the [1,0] direction at $E = 2$, 3, 4, and 5 meV in zero and 5 T c-axis aligned magnetic fields at $T = 2$ K. (e) and (f) 2D images of wave-vector and energy dependence of the resonance in zero and 5 T magnetic fields at 2 K. The background subtraction process is similar to that of Fig. 2 [48]. (g) and (h) are constant-**Q** cuts at the $\mathbf{Q}_{AF}$ position from (e) and (f), respectively. They have been smoothed two times by the 2D data processing method in the DAVE-MSLICE program at NCNR. The arrows in (e)–(h) indicate the lowest energy where a Gaussian can be fit to the data. The error bars indicate statistical errors of one standard deviation.

2(f), 3(g), and 3(f), we conclude that a 5 T c-axis aligned field has a larger impact on the resonance than that of an 8.5 T in-plane field.

To determine if the broadening of the resonance in the c-axis aligned magnetic field follows expectations from the field-induced Zeeman effect, we carried out additional measurements on PANDA. Figures 4(a) and 4(b) show the evolution of the magnetic scattering at $\mathbf{Q}_{AF} = (1, 0)$ in the

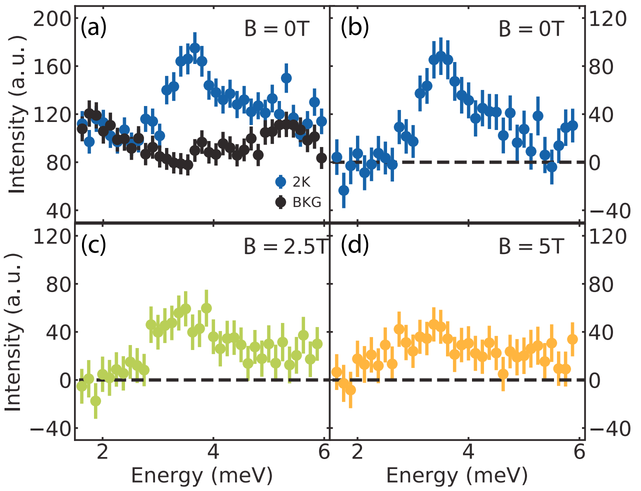

FIG. 4. (a) Constant-**Q** scans at $\mathbf{Q}_{\mathrm{AF}}$ and the off-peak background positions at 2 K in zero field as shown in Fig. 1(d). (b)–(d) Constant-**Q** scans with background subtracted in 0, 2.5, and 5 T *c*-axis aligned fields. The error bars indicate statistical errors of one standard deviation.

superconducting state before and after correcting for the background scattering, respectively. As expected, we see a well-defined spin gap below 2.8 meV and a resonance peaked at $E_r = 3.6$ meV [Fig. 4(b)]. Upon application of a 2.5 T field, the resonance broadens and weakens, but still seems to be centered around $E_r = 3.6$ meV [Fig. 4(c)]. At 5 T, the magnetic scattering is broadened and weakened further [Fig.4(d)].

To understand these results, we first consider the effect of Zeeman energy $\pm g\mu_B B$ on the resonance. For an isotropic resonance with weak SOC, such as for cuprate superconductors [17], the application of a magnetic field is expected to split the mode into a triplet [49]. This is similar to the magnetic field effect on quantum magnets such as $TlCuCl_3$ [50] and $Sr_{14}Cu_{24}O_{41}$ [51], in which the ground state is a singlet and the excited state is a triplet, and the system undergoes a so-called magnon Bose-Einstein condensation (BEC) in magnetic fields [52]. When SOC becomes important, as in the case of iron-based superconductors [53], low-energy spin excitations become anisotropic in spin space [54]. In the case of FeSe, neutron polarization analysis suggests that the resonance is highly anisotropic in spin space [44]. As a consequence, the resonance should not be split by a Zeeman field into a triplet. If the resonance is a magnonlike excitation, two polarizations perpendicular to the applied field are needed to form a doublet. Since the resonance is reported to be mostly polarized along the *c* axis [44], a Zeeman field should be unable to split the mode into a doublet.

Figure 1(f) compares the expected broadening of the resonance assuming that the mode splits into three peaks in the applied magnetic fields via the Zeeman effect. Taking $g = 2$, the field-induced Zeeman splitting equals to 0.58 and 0.98 meV in 5 and 8.5 T, respectively. In the 8.5 T in-plane magnetic field, the lowest energy where excitation can be observed at $\mathbf{Q}_{\mathrm{AF}}$ is 2.5 meV, which is 1.1 meV below the peak of the resonance at a zero field. For a 5 T field along the *c* axis, the magnetic signal can be observed down to 2 meV. Since the Zeeman splitting should have no field directional dependence, the wider in-plane field-induced resonance must be due to field-induced orbital current that suppresses superconductivity. As a function of the increasing magnetic field along the *c* axis, the intensity of the resonance is gradually suppressed and broadened, qualitatively consistent with the field-induced suppression of superconductivity and superfluid density [55]. Indeed, as previously noted the experiments correspond to applying essentially the same 30% fraction of the upper critical field for both field directions.

In recent electric and thermal transport measurements [55], it was argued that FeSe is in a BCS-BEC crossover regime, and a large magnetic field along the *c* axis might induce a new superconducting phase coexisting with magnetic order, possibly the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [55–58]. To study if this phase has field-induced magnetic order as suggested from the field-induced broadening of the resonance, we carried out neutron diffraction experiments using the 2-axis-diffractometer E4, HZB, Germany [59]. We aligned about 200 pieces of FeSe single crystals in the $[H, K, 0]$ scattering plane and mapped out one quadrant of the zone with a wave vector between 0.14 and 1.54 reciprocal lattice unit [48]. However, we did not find any observable difference between data at different temperatures (0.25 and 3 K) or at base temperature (0.25 K) with different fields (0, 12, 14, and 14.5 T) along the *c* axis, suggesting no observable field-induced magnetic order up to 14.5 T [48]. However, thermal conductivity data indicated the FFLO phase might exist for an ∼24 T in-plane magnetic field [58]. Unfortunately, currently available neutron spectrometers cannot access such a high DC field.

Assuming that the resonance is directly associated with superconducting electron pairs [36], we can estimate the upper critical fields for *c*-axis and in-plane fields using field-induced suppression of the resonance. If the spin gap energy below the resonance decreases linearly with the applied field, we estimate that the lowest energy position of the spin gap to $E = 0$ meV in the *c*-axis and in-plane magnetic fields corresponds to fields of 12 and 30 T, respectively. These values are close to the measured upper critical fields (B_{c2}) of 16 and 28 T. The field directional dependence of the spin resonance is also consistent with that of the superfluid density from heat capacity measurements [60], implying that the resonance is associated with superconducting electrons arising from the orbital selective hole-electron quasiparticle excitations [42,43].

In summary, we determined the effect of *c*-axis and in-plane magnetic fields on the neutron spin resonance of FeSe. We find that an in-plane magnetic field increases the width of the resonance following the field-induced Zeeman effect. A *c*-axis aligned field suppresses and broadens the resonance much more effectively than the in-plane field, clearly related to the orbital effect and vortex currents induced by the *c*-axis field. The data indicates that rather than the absolute applied field, it is the ratio of the applied field to the upper critical field that controls changes in the magnetic excitation spectrum. Our results are consistent with the hypothesis that the resonance is associated with electron pairing density in FeSe superconductors.

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