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## Neutron spin resonance as a probe of the superconducting energy gap of $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ superconductors

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We use inelastic neutron scattering to show that for the optimally electron-doped  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  ( $T_c = 20$  K) iron arsenide superconductor, application of a magnetic field that partially suppresses the superconductivity and superconducting gap energy also reduces the intensity and energy of the resonance. These results demonstrate that the energy of the resonance is intimately connected to the electron pairing energy, and thus indicate that the mode is a direct probe for measuring electron pairing and superconductivity in iron arsenides.

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The discovery of high-transition temperature (high- $T_c$ ) superconductivity near antiferromagnetism in iron arsenides raised the possibility of an unconventional superconducting mechanism.<sup>1-6</sup> In one class of unconventional microscopic models,<sup>4-6</sup> electron pairing in iron-arsenide superconductors is mediated by quasiparticle excitations between sign reversed hole and electron Fermi pockets.<sup>7,8</sup> Although the presence of a neutron spin “resonance”<sup>9-13</sup> is consistent with this picture,<sup>14-17</sup> much is unknown about the microscopic origin of the mode. In this Rapid Communication, we show that for the  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  [ $T_c = 20$  K, Fig. 1(c)] superconductor, application of a magnetic field that partially suppresses the superconducting gap energy also reduces the intensity and energy of the resonance. These results demonstrate that the energy of the resonance is intimately connected to the electron pairing energy, and thus indicate that the mode is a direct probe for measuring electron pairing and superconductivity in iron arsenides.

Soon after the discovery of superconductivity in  $\text{LaFeAsO}_{1-x}\text{F}_x$  (Ref. 1), band-structure calculations of Fermi surfaces for these materials found two hole cylinders around the  $\Gamma$  point and two electron cylinders around the  $M$  point.<sup>14</sup> Electron pairing arises from quasiparticle excitations from the hole pocket to electron pocket [inset in Fig. 1(c)] that induces a resonance peak at the antiferromagnetic (AF) ordering wave vector  $Q = (0.5, 0.5, 0)$  in the spin excitations spectrum [Fig. 1(b)].<sup>14-17</sup> The energy of the resonance is at (or slightly less than) the addition of hole and electron superconducting gap energies ( $\hbar\omega = |\Delta(k+Q)| + |\Delta(k)|$ ).<sup>15-17</sup> Although the resonance observed by inelastic neutron scattering in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  (Ref. 9) and  $\text{BaFe}_{2-x}(\text{Co}, \text{Ni})_x\text{As}_2$  (Refs. 10-13) are consistent with this picture, the microscopic origin of the mode is still unknown. One way to resolve this problem is to study the effect of a magnetic field on spin excitations. A magnetic field suppresses  $T_c$  and reduces the magnitude of the superconducting energy gap. If the resonance is associated with the superconducting energy gaps,<sup>4-6</sup> application of a magnetic field that partially suppresses the superconducting gaps should also reduce the en-

ergy of the resonance, just like increasing temperature can reduce the superconducting gap and resonance energy.<sup>13</sup> We find this is indeed the case for  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  (Fig. 1) and our results thus provide the most compelling evidence that electron pairing in iron-arsenide superconductors is directly correlated with magnetic excitations.

In the undoped state, the parent compounds of iron-arsenide superconductors are nonsuperconducting antiferromagnets with a spin structure as shown Fig. 1(a) (Ref. 3). Upon doping to induce optimal superconductivity, the static AF order is suppressed and magnetic excitations in the superconducting state are dominated by a resonance above the spin gap energy.<sup>9-13</sup> For  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  with  $T_c = 20$  K [Fig. 1(c)], the resonance occurs near  $\hbar\omega \approx 8$  meV at  $Q = (0.5, 0.5, 0)$  reciprocal lattice unit (rlu) above a  $\hbar\omega \approx 3$  meV spin gap at 4 K (Refs. 11 and 12) and is purely magnetic as shown by polarized neutron-scattering experiments.<sup>18</sup> We used inelastic neutron scattering to study the effect of a 14.5 T  $c$  axis aligned magnetic field on the resonance and spin gap using the IN22 thermal triple-axis spectrometer at the Institut Laue-Langevin, Grenoble, France (Fig. 1).<sup>19</sup> We coaligned 5.5 g of single crystals of  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  grown by self-flux (with in-plane mosaic of  $2^\circ$ ) and define the wave vector  $Q$  at  $(q_x, q_y, q_z)$  as  $(H, K, L) = (q_x a / 2\pi, q_y b / 2\pi, q_z c / 2\pi)$  in rlu, where  $a = b = 3.963$  and  $c = 12.77$  Å are the tetragonal unit-cell lattice parameters.<sup>11</sup> Our samples are aligned in the  $(H, K, 0)$  horizontal scattering plane inside a 14.5-T vertical field magnet. The final neutron energy was fixed at 14.7 meV with a pyrolytic graphite filter before the analyzer. Field was always applied in the normal state at 25 K.

At zero field, energy scans at  $T = 25$  K show clear gapless continuum of scattering at the signal  $Q = (0.5, 0.5, 0)$  position above the background  $Q = (0.62, 0.62, 0)$  [(red) filled and open circles in Fig. 1(d)]. On cooling to  $T = 2$  K, a spin gap gradually opens below  $\hbar\omega \approx 3$  meV and the low-energy spectral weight is transferred into the resonance at  $\hbar\omega \approx 8$  meV.<sup>11,12</sup> While imposition of a 14.5-T magnetic field has little effect on the background [Fig. 1(d)] and normal

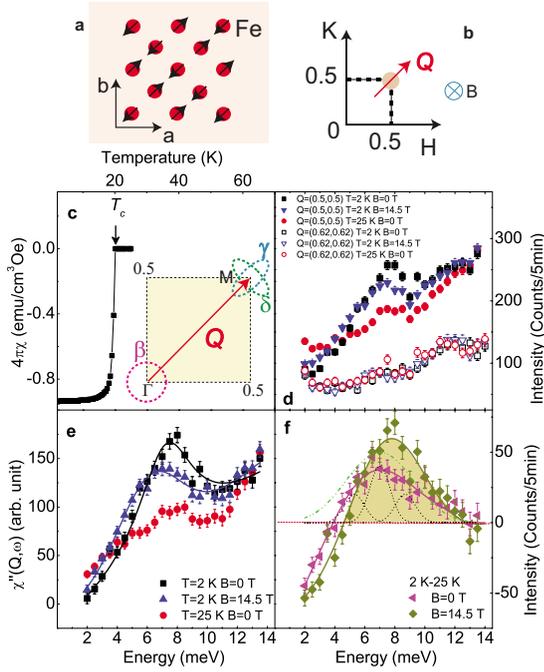


FIG. 1. (Color online) (a) Schematic of the Fe spin ordering in  $\text{BaFe}_2\text{As}_2$ . (b) Reciprocal space probed and the direction of applied field in tetragonal notation (Ref. 10). (c) Susceptibility of our sample indicating  $T_c=20$  K. The inset shows schematic of how the resonance is produced by quasiparticle excitations between the hole and electron pockets. (d) Energy scans at the signal  $Q=(0.5,0.5,0)$  and background  $Q=(0.62,0.62,0)$  rlu positions for various fields and temperatures. For clarity, the 14.5-T data at 25 K are not shown. (e) Temperature and field dependence of  $\chi''(Q, \omega)$  at  $Q=(0.5,0.5,0)$ . The black and (blue/grey) solid lines are Lorentzian fits to the resonance on sloped linear backgrounds. Horizontal bar indicates instrumental energy resolution. (f) Difference spectra of the neutron intensity between  $T=2$  K ( $<T_c$ ) and  $T=25$  K ( $T_c+5$  K) at  $Q=(0.5,0.5,0)$  for  $B=0$  and a 14.5-T  $c$ -axis-aligned field. The black dotted lines indicate the expected Zeeman splitting under 14.5-T field using the corresponding instrumental resolution. The green line is the expected resonance scattering profiles at 14.5 T.

state scattering at  $Q=(0.5,0.5,0)$ , the resonance peak in the superconducting state is clearly suppressed and shifted to a lower energy [(blue) triangles in Fig. 1(d)]. Figure 1(e) plots the temperature dependence of the imaginary part of the dynamic susceptibility  $\chi''(Q, \omega)$ , obtained by subtracting the background scattering and correcting for the Bose population factor  $\chi''(Q, \omega) = \{1 - \exp[-\hbar\omega/(k_B T)]\} S(Q, \omega)$ , where  $k_B$  is the Boltzmann constant. Inspection of the figure reveals that application of a 14.5-T magnetic field shifted the energy of the resonance from  $\hbar\omega=7.3 \pm 0.2$  to  $6.0 \pm 0.3$  meV and broadened the mode only slightly. Comparison of the temperature difference plots at zero and 14.5 T in Fig. 1(f) confirms the shift in energy of the mode. In addition, the data suggest that superconductivity-induced resonance intensity gain [the shaded area in Fig. 1(f)] decreases about 23% from zero to 14.5 T.

Figure 2 summarizes  $Q$  scans at energies  $\hbar\omega=0, 2, 3, 8$  meV which corresponds to elastic scattering, below and near spin-gap energy, and at the resonance energy,

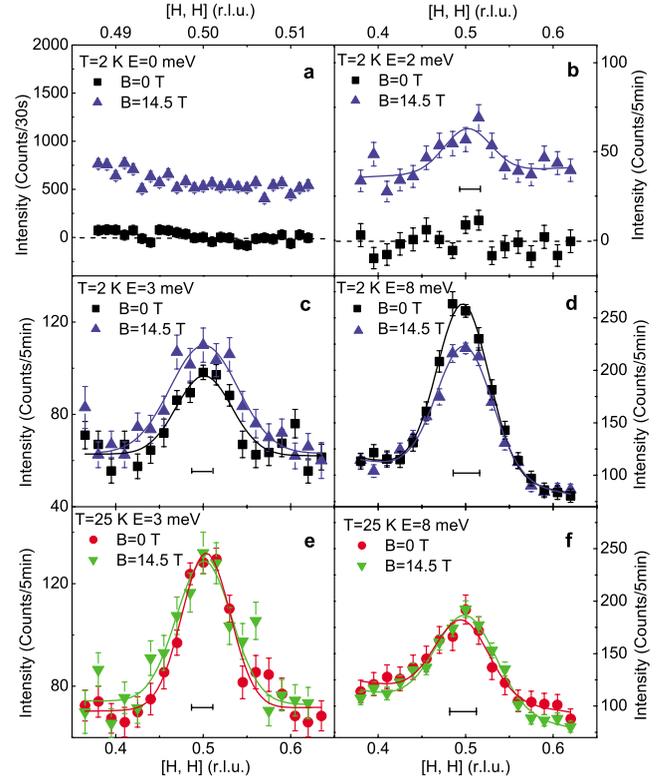


FIG. 2. (Color online) (a)  $\hbar\omega=0$  meV and (b)  $\hbar\omega=2$  meV. Spin-spin correlation length at 2 K and 14.5 T is  $\xi=64 \pm 16$  Å. Note that the vertical scales for the  $B=0$ -T data in (a) and (b) were offset for clarity; (c)  $\hbar\omega=3$  meV at 2 K. At zero field,  $\xi=65 \pm 10$  Å. At 14.5 T,  $\xi=47 \pm 10$  Å; (d)  $\hbar\omega=8$  meV at 2 K. At zero field,  $\xi=57 \pm 2$  Å. At 14.5 T,  $\xi=53 \pm 3$  Å; (e)  $\hbar\omega=3$  meV at 25 K. At zero field,  $\xi=62 \pm 5$  Å. At 14.5 T,  $\xi=54 \pm 6$  Å; (f)  $\hbar\omega=8$  meV at 25 K. At zero field,  $\xi=55 \pm 5$  Å. At 14.5 T,  $\xi=49 \pm 4$  Å. The solid lines are Gaussian fits to the data on linear backgrounds and horizontal bars in (b)–(f) are the instrumental resolution.

respectively. At  $\hbar\omega=0$  meV and 2 K, the scattering are featureless at zero and 14.5 T [Fig. 2(a)], indicating that such a field does not induce AF long-range static order. For  $\hbar\omega=2$  meV, the scattering at zero field show no peak, which is consistent with the presence of a spin gap at 2 K.<sup>11,12</sup> However, the identical  $Q$  scan at 14.5 T shows a clear peak at  $Q=(0.5,0.5,0)$ , suggesting a field-induced scattering due to the decreasing value of the zero field spin gap [Figs. 1(e) and 2(b)]. Similarly, a 14.5-T field enhances the zero field  $\hbar\omega=3$  meV peak near  $Q=(0.5,0.5,0)$  at 2 K [Fig. 2(c)] but has no effect at 25 K [Fig. 2(e)]. In contrast, imposition of a 14.5-T field at 2 K partially suppresses the resonance intensity at  $\hbar\omega=8$  meV [Fig. 2(d)]. The same field again has no effect at 25 K [Fig. 2(f)]. Fourier transforms of the Gaussian peaks at  $\hbar\omega=8$  meV and 2 K in Fig. 2(d) give spin-spin correlation lengths of  $\xi=57 \pm 2$  Å and  $\xi=53 \pm 3$  Å for 0 and 14.5 T, respectively. Whereas a field can change the energy and intensity of the resonance, it has small effect on spin-spin correlation length similar for copper-oxide superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$  (Ref. 20).

Figure 3 compares temperature dependence of the scattering at  $Q=(0.5,0.5,0)$  for  $\hbar\omega=2$  and 8 meV at zero and

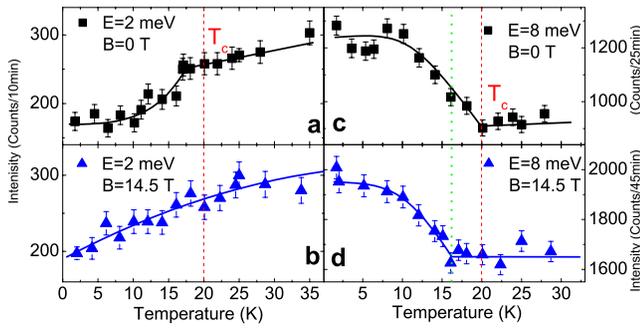


FIG. 3. (Color online) (a) Temperature dependence of the scattering at  $\hbar\omega=2$  meV and zero field shows the opening of a spin gap slightly below  $T_c$  (Refs. 15 and 16). (b) The same temperature dependence at 14.5 T. (c) Temperature dependence of the scattering at  $\hbar\omega=8$  meV and zero field displays order parameter like intensity increase below  $T_c=20$  K. (d) Application of a 14.5-T field suppresses  $T_c$  to  $\sim 16$  K.

14.5 T, respectively. Consistent with previous work,<sup>11,12</sup> we find that a spin gap opens at  $\hbar\omega=2$  meV [Fig. 3(a)] and the scattering at the resonance energy ( $\hbar\omega=8$  meV) shows a superconducting order parameterlike increase below  $T_c$  [Fig. 3(c)]. Under 14.5-T field, the kink in zero field<sup>12</sup> at  $\hbar\omega=2$  meV slightly below  $T_c$  disappears [Fig. 3(b)] and the scattering shows no observable anomaly. On the other hand, temperature dependence of the scattering at 8 meV shows a clearly depressed  $T_c$  of  $\sim 16$  K at 14.5 T from  $T_c=20$  K at zero field [Figs. 3(c) and 3(d)]. Since an applied magnetic field that suppresses  $T_c$  also decreases the superconducting gap energy, these results demonstrate that the resonance energy and its temperature dependence are directly correlated with the superconducting gap energy and electron pairing strength.

Figures 4(a) and 4(b) show the magnetic field dependence of the scattering at the resonance energy at 2 and 25 K, respectively. While the normal-state spin excitations have no observable field effect up to 14.5 T [Fig. 4(b)], the scattering at the resonance energy clearly decreases with increasing field [Fig. 4(a)]. The solid line is a linear fit to the data using  $I/I_0=1-B/B_{char}$  with  $B_{char}\approx 32$  T, where intensity of the resonance is suppressed to the normal-state value. The dotted line represents a fit assuming  $I/I_0=1-(B/B_{char})^{1/2}$  where  $B_{char}\approx 66$  T (Ref. 20). Since the energy of the resonance is decreasing with increasing field, it is difficult to compare  $B_{char}$  with the  $c$  axis upper critical field  $B_{c2}$  of  $\sim 43$  to  $\sim 50$  T for  $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$  samples ( $T_c\approx 22$ – $25.3$  K).<sup>21,22</sup>

The total momentum sum rule states that the magnetic structure factor  $S(Q, \omega)$  when integrated over all wave vectors and energies, i.e.,  $\int_{-\infty}^{\infty} d\omega \int dQ S(Q, \omega)$ , should be a temperature- and field-independent constant.<sup>23</sup> To see if this is true at zero and 14.5 T, we plot in Fig. 4(c) experimentally measured difference spectrum,  $S(Q, \omega, B=0 \text{ T}) - S(Q, \omega, B=14.5 \text{ T})$ , at  $Q=(0.5, 0.5, 0)$  and 2 K. We find that the spectral weight loss of the resonance under a 14.5-T field is approximately compensated by the field-induced subgap intensity gain, suggesting that the sum rule is satisfied within our probed  $Q$ -energy space.

In previous work on copper-oxide superconductors such

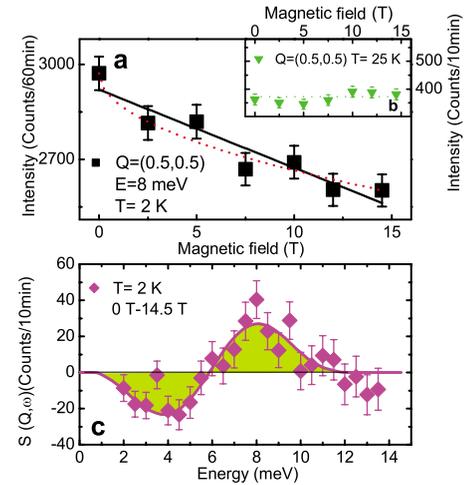


FIG. 4. (Color online) (a) The magnetic field dependence of the scattering at  $\hbar\omega=8$  meV,  $Q=(0.5, 0.5, 0)$ , and 2 K. While the solid line is a fit using  $I/I_0=1-B/B_{char}$  with  $B_{char}\approx 32$  T, the dotted line represents  $I/I_0=1-(B/B_{char})^{1/2}$ , where  $B_{char}\approx 66$  T. (b) The scattering at 25 K has no observable field dependence. (c) The difference spectrum of the neutron-scattering intensities between zero and 14.5-T field at 2 K and  $Q=(0.5, 0.5, 0)$ .

as  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (Ref. 19),  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  (Refs. 20 and 24), and  $\text{Pr}_{0.88}\text{LaCe}_{0.12}\text{CuO}_4$  (Ref. 25), application of a magnetic field was found to suppress the intensity of the resonance<sup>19,20</sup> and induce AF order at the expense of the resonance.<sup>24,25</sup> However, the energy of the mode is magnetic field independent.<sup>19,20,24,25</sup> Theoretically, several effects of a magnetic field on the resonance and spin excitations have been considered within the random-phase approximation:<sup>26</sup> first, the supercurrents circulating around the field-induced vortices may slightly broaden the resonance in energy without changing its  $Q$ -energy integrated weight; second, a field-induced uniform suppression of the superconducting gap magnitude should cause the resonance to shift to lower energy and decrease in intensity; third, the effect of field-induced suppression of the superconducting coherence factor might lead to suppression of the spectral weight and causing the resonance to shift to higher energy; and finally, suppression of the resonance within the field-induced vortex cores could result in reduced resonance intensity without shifting its position.<sup>19,20</sup> Since we observed a clear field-induced resonance energy and intensity reduction in  $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$  (Figs. 1–4), our data are most consistent with a field-induced suppression of the superconducting gap energy.

If this picture is correct, we can use data in Figs. 1–4 to estimate the  $B_{c2}$  and expected resonance energy shift at 14.5-T field. In Ginzburg-Landau theory, which is the best phenomenological theory to describe superconductors, magnetic field dependence of the superconducting gap  $\Delta(B)$  is related to the zero field gap  $\Delta(0)$  via  $\Delta(B)/\Delta(0)=\sqrt{1-B/B_{c2}}$  (Ref. 26). Since superconducting gap is proportional to  $T_c$  (i.e.  $2\Delta\propto k_B T_c$ , Refs. 7 and 8), we estimate  $B_{c2}=40.3$  T using the measured  $T_c$  ( $\approx 16$  K) at 14.5 T in Fig. 3(d) and  $B_{c2}=B/\{1-[T_c(14.5 \text{ T})/T_c(0 \text{ T})]^2\}$ . This value is very close to the measured  $B_{c2}=43$ – $50$  T for  $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$  (Refs. 21 and 22). Since the resonance en-

ergy equals to  $\hbar\omega = |\Delta(k+Q)| + |\Delta(k)|$ , one should expect the mode energy to shift from  $\hbar\omega \approx 7.3 \pm 0.3$  meV at zero field to  $\hbar\omega(14.5 \text{ T}) = [T_c(14.5 \text{ T})/T_c(0 \text{ T})]\hbar\omega(0 \text{ T}) \approx 5.84$  meV. Inspection of Fig. 1(e) shows that this is indeed the case with  $\hbar\omega(14.5 \text{ T}) = 6.0 \pm 0.3$  meV. This is the most compelling evidence that the resonance is related to superconducting gap energy.

To test if the resonance directly probes the electron spin singlet-to-triplet transition (from singlet spin  $S=0$  for Cooper pairs to triplet spin  $S=1$ ) arising from the sign reversed electron and hole pockets scattering [Fig. 1(c)], we note that a triplet excitation should be split in the presence of a magnetic field via the Zeeman energy splitting  $\Delta E_{\text{Zeeman}} = \pm g\mu_B B$  (Refs. 20 and 27). Assuming the Lande factor  $g=2$  and  $S=1$ , the Zeeman magnetic energy splitting for a 14.5-T field is  $\Delta E_{\text{Zeeman}} = \pm 1.7$  meV. Experimentally, the energy widths of the resonance in Fig. 1(e) assuming Lorentzian line shapes change from  $4.8 \pm 0.6$  meV full width at half maximum (FWHM) at zero field to  $5.6 \pm 1$  meV FWHM at 14.5 T as shown in the black and

blue solid lines, respectively. For unpolarized neutron-scattering experiments on isotropic triplet excitations, the single peak at zero field should split into three peaks separated by Zeeman energy with the integrated intensity of the unshifted peak equals to the sum of the two shifted side peaks.<sup>28</sup> Given the finite energy width of the resonance and instrumental resolution [Fig. 1(e)], we cannot determine the Zeeman splitting of the mode [Fig. 1(f)]. Therefore, while our data support the notion that the resonance is directly correlated with the superconducting electron energy gap, it is unclear whether the mode is the long-sought singlet-to-triplet transition.

*Note added.* Recently, we became aware of the two recent magnetic field effect works on the  $\text{FeTe}_{1-x}\text{Se}_x$  superconductors.<sup>29,30</sup>

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