Neutron diffraction study of the magnetic and structural phase transitions in $BaFe_2As_2$

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Since the discovery of superconductivity in the iron arsenides in February 2008, the field has witnessed an explosion of research into this newly discovered high temperature superconducting (high-T_c) phase [1]. It was quickly uncovered that a key similarity between the iron pnictide high-T_c phase and that of the well known cuprates is close proximity of a magnetic instability in their respective phase diagrams [2,3,4]. The undoped parent compounds of both the cuprates and the iron pnictides are long-range ordered antiferromagnets; however the seemingly itinerant, semimetallic nature of the iron pnictide parent materials stand in stark contrast to the strongly localized, Mott insulating parent systems of the high-T_c cuprates. The nearby suppression of antiferromagnetism (AF) and the persistence of strong, remnant spin fluctuations in both classes of high temperature superconductors are thought to play a key role in the formation of their superconducting condensates.

One of the pivotal first steps in understanding the role of magnetism within the superconducting phase of the iron pnictides is to first explore the intrinsic nature of the magnetic phase of their undoped parent compounds. The bilayer AFe_2As_2 (A = Ba, Sr, Ca, …) class of pnictide superconductors constitute the most suitable system for studying the detailed magnetism in these systems due to the relative ease in obtaining high quality single crystals. For this reason, we chose to study the $BaFe_2As_2$ system (Ba-122), and, for our experiment, we successfully grew one large high-quality crystal with a mass of ~ 600 mg and with a T_N = 136 K determined by bulk susceptibility and resistivity measurements [5]. The sample was aligned within the [H, 0, L] scattering plane on the N5 neutron beamline and mounted within a closed cycle refrigerator with exchange gas to ensure an accurate control of sample temperature. We used flat PG-002 crystals for both monochromator and analyzer with a fixed final energy of E_f = 3.52 THz. A PG filter was mounted in the scattered side to eliminate higher order wavelengths from the beam. In addition a liquid nitrogen cooled sapphire filter was used in the main beam to reduce fast neutron background. Collimations were set to [none, 1°, 0.85°, 1.2°].

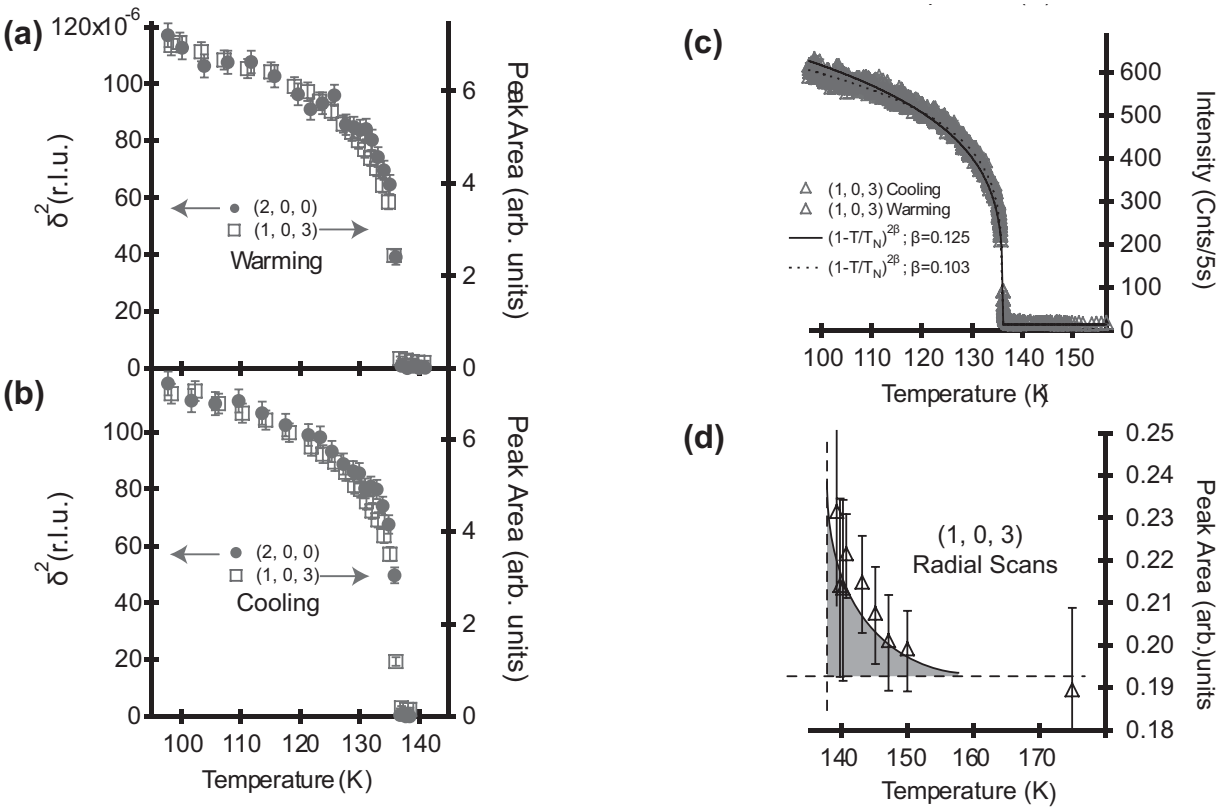

Fig 1. *(a) and (b) show the magnetic order parameter squared (open squares) and the structural order parameter squared (solid circles) overplotted as the system is respectively warmed and cooled through T_N. (c) Magnetic order parameter of Ba-122 upon warming and cooling through T_N. Dashed line shows the 2D Ising fit to the order parameter. (d) Plot of 3D critical scattering above T_N. Open triangles show the integrated area of the (1,0,3) magnetic peak above T_N while the dashed line denotes the backgrounmd from λ/2 contamination.*

After verifying that the AF ordered spin structure in our Ba-122 crystal matched those from previous reports [6], the initial focus of our experiment was to explore the antiferromagnetic phase transition within the critical regime. The sample was slowly cooled and warmed through T_N while the strongest **Q** = (1, 0, 3) [r.l.u.] magnetic reflection was measured (see Fig. 1). Both the peak intensity and integrated peak area of this reflection were tracked, and neither showed any hysteresis in strong contrast to earlier experiments where strong first order behavior has been reported [7]. Instead, we observe a continuous evolution of the magnetic order parameter (taken as the intensity of the (1, 0, 3) magnetic Bragg reflection) in the critical regime which is well modeled by a simple power law with an exponent of $\beta = 1/8$ as shown in Fig. 1c. This is the exponent predicted for phase transitions in a 2D Ising model, and it immediately suggests that 2D magnetic fluctuations may be driving the magnetic phase transition in this Ba-122 system. The agreement between the simple $(1\text{-}T/T_N)^{2\beta}$ with $\beta = 1/8$ fit and the evolution of the magnetic order parameter over a remarkable range in reduced temperature is also reminiscent of the power law behavior observed in other well known 2D Ising magnets such as K_2NiF_4 [8].

In order to further explore the critical magnetism in Ba-122, we measured magnetic correlations above T_N about the **Q** = (1, 0, 3) reflection and observed 3D critical fluctuations extending from T = 136K to approximately T = 145K (Fig. 1d). We were unable to observe any diffuse, 2D critical fluctuations anticipated by the $\beta = 1/8$ critical exponent of the magnetic order parameter; however, due to the small sample volume of our Ba-122 crystal, it is difficult to preclude their presence. This implies that, at the minimum, there is a crossover to 3D critical behavior close to T_N due to the finite coupling between the FeAs layers.

One of our key findings is the observed relationship between the structural and magnetic order parameters in this system [5]. We tracked the evolution of the structural order parameter from the high temperature tetragonal phase to the low temperature orthorhombic phase as the system was cooled through T_N. Consistent with earlier reports, there appeared a simultaneous structural distortion to the orthorhombic phase concomitant with the onset of AF order [6]; however we also observed an unexpected correlation between the structural and magnetic order parameters in the critical regime. As the system is cooled into the antiferromagnetic phase, the structural order parameter (the peak width observed in longitudinal scans around (2, 0, 0) structural Bragg reflection) squared directly tracks the magnetic order parameter (the peak width observed in longitudinal scans around (1, 0, 3) magnetic Bragg reflection) squared (Figs. 1a and b). This implies a biquadratic coupling term between the structural and magnetic order parameters in the Landau free energy expansion for this Ba-122 system. Such a biquadratic coupling immediately implies that the structural order parameter is a distinct, primary order parameter as opposed to a secondary order parameter simply driven through a linear-biquadratic coupling to the magnetic order parameter through the system's strain field. This observation stands in strong contrast to current picture of the structural distortion as merely arising through strain-induced coupling to the magnetic order parameter and instead implies that two coupled, yet independent, structural and magnetic phase transitions coexist in Ba-122.

For systems with two coupled primary ordered parameters, the simplest free energy expansion leads to the formation of a tetracritical point in their phase diagrams [9]. In the case of Ba-122, this implies that this iron pnictide parent system resides close to the proximity of such a tetracritical point in its phase diagram. One general expectation from this picture is that as the system is tuned away from the tetracritical point, the onset of magnetic and structural phase transitions should separate. This is precisely what is observed in Co-doped Ba-122 [10]; however further studies are required to fully explore the nature of the separated phase transitions in these doped samples.

The next step in our study of this compound is to coalign a substantially larger sample volume of Ba-122 crystals in search of any potential 2D critical fluctuations and to measure the temperature evolution of their instantaneous correlation lengths. We also plan to map out the inelastic magnetic spectrum of this system in an effort to generate a comprehensive understanding of the spin behavior in this bilayer iron arsenide parent system.

References

[1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
[2] J. Zhao et al., Nature Mater. 7, 953 (2008).
[3] M. Rotter et al., Phys. Rev. Lett. 101, 107006 (2008).
[4] R. J. Birgeneau, C. Stock, J. M. Tranquada, and K. Yamada, J. Phys. Soc. Jpn. 75, 111003 (2006).
[5] Stephen D. Wilson, Z. Yamani, C. R. Rotundu, B. Freelon, E. Bourret-Courchesne, and R. J. Birgeneau, Phys. Rev. B 79, 184519 (2009).
[6] Q. Huang, Y. Qiu, W. Bao, M. A. Green, J. W. Lynn, Y. C. Gasparovic, T. Wu, G. Wu, and X. H. Chen, Phys. Rev. Lett. 101, 257003 (2008).
[7] M. Kofu, Y. Qiu, W. Bao, S.-H. Lee, S. Chang, T. Wu, G. Wu, and X. H. Chen, New J. Phys., 11, 055001 (2009).
[8] R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B 1, 2211 (1970).
[9] K.-S. Liu and M. E. Fisher, J. Low Temp. Phys. 10, 655 (1973).
[10] J.-H. Chu, J. G. Analytis, C. Kucharczyk, and I. R. Fisher, Phys. Rev. B 79, 014506 (2009).