Manifestation of itinerant magnetism in hole-doped iron arsenide superconductors

J. M. Allred,1,2 K. M. Taddei,1,3 D. E. Bugaris,1 M. J. Krogstad,1,3 S. H. Lapidus,4 D. Y. Chung,1 H. Claus,1 M. G. Kanatzidis,1,5 D. E. Brown,3 J. Kang,6 R. M. Fernandes,6 I. Eremin,7 S. Rosenkranz,1 O. Chmaissem,1,3 and R. Osborn1

1Materials Science Division, Argonne National Laboratory, Argonne, IL 60439-4845, USA
2Department of Chemistry, University of Alabama, Tuscaloosa, AL, 35487-0336, USA
3Physics Department, Northern Illinois University, DeKalb, IL 60115, USA
4Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439-4845, USA
5Department of Chemistry, Northwestern University, Evanston, IL 60208-3113, USA
6School of Physics and Astronomy, University of Minnesota, Minneapolis, MN 55455, USA
7Institut für Theoretische Physik III, Ruhr-Universität Bochum, 44801 Bochum, Germany

The nature of the magnetic ground state in iron-based superconductors is believed to be of paramount importance to the eventual understanding of their high-temperature superconducting mechanism, yet even the strength of the electronic correlations has remained contested since the discovery of superconductivity in iron-arsenides.

One way that this controversy is manifested originates from whether one assumes that the magnetic moments are quasi-local or not. In order to investigate this, the nuclear and magnetic structure of the ground states of the iron-based superconductors Sr1-xNa_xFe2As2 was investigated using a combination of powder x-ray and neutron diffractions, Mössbauer spectroscopy, and magnetic susceptibility. Samples were synthesized at compositions ranging from \( x = 0.1 \) to \( x = 0.6 \). The combination of techniques allowed for the distinction between compositional steps at least as small as \( \Delta x = 0.01 \). Along with the known magnetic dome of \( C_2 \) symmetry, another instance of the more recently discovered \( C_4 \) phase was uncovered (“\( C_2 \)” and “\( C_4 \)” phases, respectively).

This phase diagram was unique compared to our previous experiments, in that the bulk showed 100% phase fraction of the \( C_4 \) phase in several sample compositions. The aforementioned tools were used to probe the properties over a broad temperature range, and together elucidated the nature of the magnetic phase in the \( C_4 \) magnetic state. Rietveld refinements gave a very high-resolution picture of all phase transitions in the temperature domain, while Mössbauer data proves that half of the \( C_2 \) magnetic sites become nodal—that is, of zero net moment—while the remaining half increase their effective moments. Together this proves that the magnetic state is an itinerant spin density wave, where the \( C_4 \) phase is a superposition of two different \( Q \) vectors. This observation constrains the set of models that can naturally explain the magnetic properties in this family of materials by ruling out models that rely on localized spins. If indeed the magnetism and superconductivity are linked, this also narrows the plausible mechanisms for their high-temperature superconductivity.