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A search for field-induced ordering in the optimally doped $Ba(Fe, Co)_2As_2$ superconductor

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A ^{57}Fe Mössbauer search for field-induced magnetic order in optimally doped $Ba(Fe_{1-x}Co_x)_2As_2$ in fields of up to 6 T showed no changes that could be attributed to field-induced order. We also observed no difference between the normal (30 K) and superconducting states (5 K). Any field-induced order is certainly less than 1% of the order present in the parent $BaFe_2As_2$. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795421]

I. INTRODUCTION

Transverse-field muon spin relaxation (TF − μSR) is routinely used to determine the magnetic penetration depth (λ) of type-II superconductors. The superconducting sample is placed in an applied field, the field distribution (n(B)) within the sample is measured, and then the contribution of the vortex lattice is modelled to obtain λ. An underlying weakness of this method is that the actual geometry of the vortex lattice, the vortex structure, and vortex interactions may not be known with sufficient certainty. Furthermore, one must assume that other contributions to n(B) are absent. Recent work on the iron-based superconductors $Sr(Fe_{0.91}Co_{0.09})_2As_2$[1,2] and $Ba(Fe_{0.91}Co_{0.09})_2As_2$[2,3] has led to the suggestion that the applied field may induce magnetic order in these systems, and that the presence of field-induced order in the vortex cores, where superconductivity has been suppressed, may affect the derived penetration depth.[3] Such field-induced ordering may be possible as the parent $BaFe_2As_2$ exhibits spin density wave ordering at 143 K,[4] and magnetic order persists in Co-doped $Ba(Fe_{1-x}Co_x)_2As_2$ as far as $x = 5.5\%$.[5,6]

As a frequency-domain technique, many variants of μSR are most suited to the observation of small fields and moments and therefore tend to be carried out with small applied fields (typically tens of mT). The observed field shifts leading to the suggestion of field-induced order are also small (of order a few percent of the applied field in most cases). Mössbauer spectroscopy, being an energy-domain technique, is more sensitive to larger fields and the ordering in the parent phase is quite significant.[7] Furthermore, since any induced moment is likely to be associated with the iron atoms, a direct probe of these sites should be more sensitive than an impurity-based technique like μSR. We therefore set out to investigate whether any evidence of field-induced order could be found in optimally doped $Ba(Fe_{1-x}Co_x)_2As_2$ at larger applied fields.

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II. EXPERIMENTAL METHODS

Single-crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ with $x = 0\%$, 4.7%, 7.4%, and 7.7%, were grown using a self-flux method.[8] The crystals were roughly square plates, with dimensions 5 mm × 5 mm × 0.5 mm and the crystal c-axis normal to the plate surface. Susceptibility and magnetisation measurements using a quantum design physical property measurement system confirmed that the three cobalt-doped samples were superconducting and that all magnetic and superconducting transitions were consistent with the earlier reports.[8–10] The samples with $x = 7.4\%$ and 7.7% were found to be close to optimally doped with superconducting transition temperatures of ∼21 K.

The samples for the Mössbauer work were prepared by cleaving the crystals into thin sheets along the *ab*-plane with a razor blade and making a 1.5–2 cm^2 mosaic, with tungsten-loaded GE-7031 varnish used to fill any gaps between the crystals. Low temperature ^{57}Fe Mössbauer spectra were obtained using a helium flow cryostat equipped with a 7 T split-coil superconducting solenoid. The 50 mCi ^{57}Co **Rh** source was located at a null point in the field, with the drive, source, and sample mounted vertically and operated in sine mode. Spectra were initially fitted using a simple first-order perturbation code; however, the effects of a small, unresolved quadrupole splitting led us to use a full solution to the Hamiltonian for the $m_I = \frac{3}{2} \rightarrow \frac{1}{2}$ transition, modified for single-crystals. No attempt was made to include the effects of the incommensurate spin density wave ordering of $BaFe_2As_2$ in fitting its spectrum.[7]

Zero-field spectra taken at 5 K and shown in Figure 1 confirm that magnetic order is lost as cobalt is added. The two optimally doped samples show no magnetic splitting at 5 K.

III. RESULTS

Spectra taken in large applied magnetic fields show a clear magnetic splitting for both the 7.4% (Figure 2) and the 7.7% (Figure 3) samples. As the field was applied parallel to the γ-beam (and the crystal c-axis) the two $\Delta m_I = 0$ transitions (normally lines 2 and 5 of the 6-line Zeeman pattern for ^{57}Fe in a magnetic field) have zero intensity and only

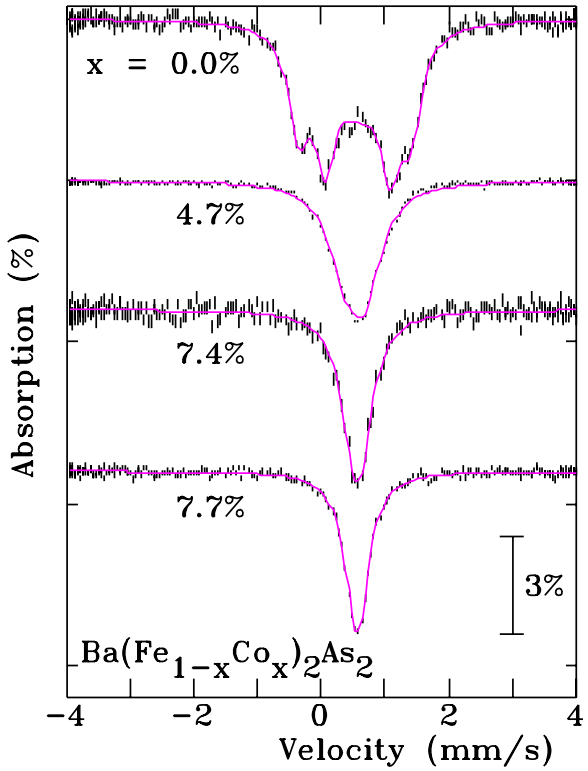

FIG. 1. Mössbauer spectra for a single-crystal mosaic of $Ba(Fe_{1-x}Co_x)_2As_2$ at 5 K in zero applied field. There is no magnetic splitting for x = 7.4% and 7.7%.

four lines (1–3–4–6) are seen. There is also a clear asymmetry to line intensities in the spectra, most evident in the central doublets (lines 3 and 4) in the middle field range (1–4 T) spectra. This is the result of a small, unresolved quadrupole

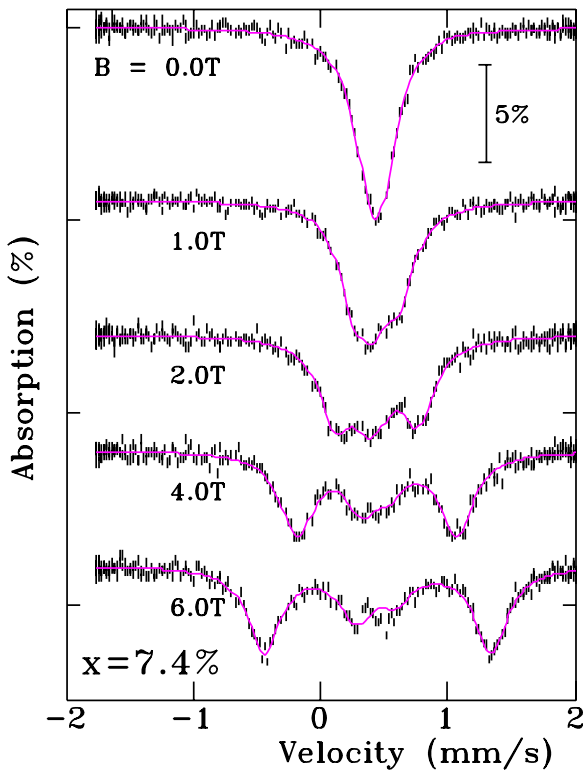

FIG. 2. Mössbauer spectra for a single-crystal mosaic of $Ba(Fe_{0.926}Co_{0.074})_2As_2$ (x = 7.4%) at 5 K in several applied fields. The slight asymmetry in the spectra is due to an unresolved quadrupole contribution.

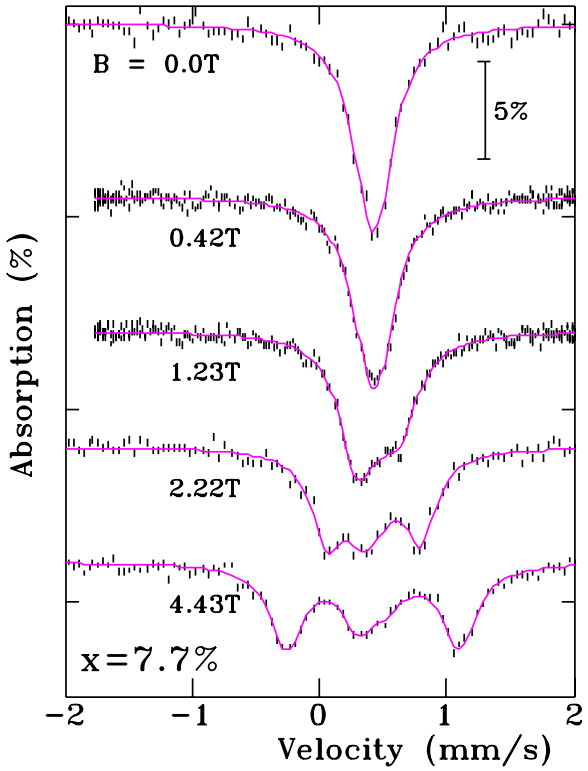

FIG. 3. Mössbauer spectra for a single-crystal mosaic of $Ba(Fe_{0.923}Co_{0.077})_2As_2$ (x = 7.7%) at 5 K in several applied fields. The slight asymmetry in the spectra is due to an unresolved quadrupole contribution.

splitting of about 0.05(1) mm/s, and the spectra had to be fitted using a solution to the full Hamiltonian to account for this effect. A comparable contribution is evident in the 5 K spectrum of the un-doped sample of $BaFe_2As_2$ shown in Figure 1.

The observed hyperfine field (B_{hf}) is plotted as a function of the applied field in Figure 4. A linear fit gives a slope of 0.96(2) with an intercept −0.02(5) T, i.e., fully consistent with a line of slope 1 passing through the origin.

Two conclusions can be reached immediately. The first conclusion is that the applied field clearly penetrates the bulk

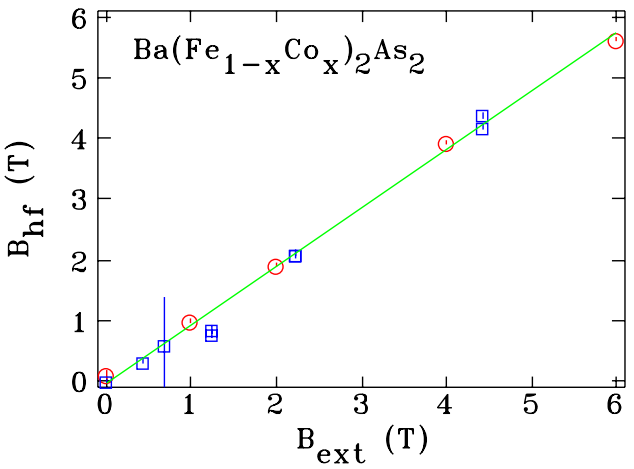

FIG. 4. Measured hyperfine fields (B_{hf}) as a function of applied field for $Ba(Fe_{1-x}Co_x)_2As_2$ with x = 7.4% (○) and 7.7% (□) at 5 K. The fitted line has a slope of 0.96(2), consistent with there being no induced field in either sample.

of the sample as we are unable to resolve any zero-field component. This is not surprising given the geometry (field applied perpendicular to the crystal sheets) and fields (many Tesla, much greater than $\mu_0 H_{c1}$) used here. The sample is expected to be well into the vortex state with only a small fraction excluding the applied field.

The second conclusion is that there is no evidence for induced magnetic order. Antiferromagnetic ordering as seen in the parent $BaFe_2As_2$[4,11,12] would have many of the same effects as ferromagnetic ordering (with the obvious exception of a demagnetising field) but is unlikely to be induced by a uniform external field and will not be considered further. The hyperfine field associated with any field-induced ferromagnetic moment would lead either to a step offset (positive intercept in Figure 4) if the moment were induced by a small field, or some curvature if the moment were induced progressively and then saturated. The demagnetising field associated with ferromagnetic order would initially act to exclude the applied field, especially if the order were induced by a small applied field. This would shift the trend line in Figure 4 down but could not make the intercept negative as the iron hyperfine field is always much larger than the magnetisation, even in densely magnetic systems such as metallic iron where the ratio is more than 16. Here, with much of the volume occupied by non-magnetic barium and arsenic, the ratio would be much higher. A cancellation or compensation of these two contributions is not possible. Our fit to the data in Figure 4 is inconsistent with a positive offset from the origin and even the uncertainty of ~50 mT puts the upper limit for an induced moment at less than 1% of that seen in the parent $BaFe_2As_2$, or much less than 0.01 μ_B/Fe.

The final possibility is that the induced moment is a linear function of the applied field, so that no curvature or offset occurs. As the hyperfine field at the iron nucleus is antiparallel to the local moment, the two contributions (from the local moment and the external field) act against each other and this could account for the slight (4(2)%) departure from unity in the fitted slope. If we scale the hyperfine field from the parent $BaFe_2As_2$ where the moment $(0.87\mu_B)$[4] and hyperfine field (5.5 T)[7,11] are both known, then the fitted slope puts a limit on the field-induced moment of $6(3) \times 10^{-3} \mu_B$/T.

The μSR-based studies that invoked field-induced magnetism to explain their observations all show a strong temperature dependence[1–3] with the effect disappearing near the superconducting transition. We checked for this in the $x = 7.7\%$ sample at 2.22 T and 4.43 T by measuring at 5 K and 30 K (well above T_{sc} of ~21 K) and found no significant difference. Indeed, while the measured hyperfine fields were consistent within error (~30 mT), in both cases, the fitted values were larger at 30 K than at 5 K, the reverse of the effect reported from μSR work.

IV. CONCLUSION

We studied single-crystal mosaics of self-flux grown $Ba(Fe_{1-x}Co_x)_2As_2$ ($x = 0\%$, 4.4%, 7.4%, and 7.7%) in fields of up to 6 T applied parallel to the crystal c-axis and perpendicular to the crystal plates. In both superconducting compositions ($x = 7.4\%$ and 7.7%), we observed complete penetration of the applied field, seeing only the applied field at the ^{57}Fe nuclei with no shift that could be attributed to field-induced order. Furthermore, we observed no difference in the field at the ^{57}Fe nuclei between 30 K (the normal state) and 5 K (where the samples were superconducting in zero field). Any field-induced magnetic ordering in superconducting $Ba(Fe_{1-x}Co_x)_2As_2$ is certainly less than 1% of the order present in the parent $BaFe_2As_2$.

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