Magnetism and Disorder Effects on $\mu$SR Measurements of the Magnetic Penetration Depth in Iron-Based Superconductors

J.E. Sonier,1,2 W. Huang,1 C.V. Kaiser,1, C. Cochrane,1 V. Pacradouni,1 S.A. Sabok-Sayr,1 M.D. Lumsden3, B.C. Sales3, M.A. McGuire3, A.S. Sefat3 and D. Mandrus1,4
1 Department of Physics, Simon Fraser University, Burnaby, British Columbia V5A 1S6, Canada
2 Canadian Institute for Advanced Research, Toronto, Canada
3 Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
4 Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee 37996, USA
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It is shown that attempts to accurately deduce the magnetic penetration depth $\lambda$ of overdoped BaFe$_{12}Co_{0.18}As_2$ single crystals by transverse-field muon spin rotation (TF-$\mu$SR) are thwarted by field-induced magnetic order and strong vortex-lattice disorder. We explain how substantial deviations from the magnetic field distribution of a nearly perfect vortex lattice by one or both of these factors is also significant for other iron-based superconductors, and this introduces considerable uncertainty in the values of $\lambda$ obtained by TF-$\mu$SR.

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TF-$\mu$SR is routinely used to determine the magnetic penetration depth $\lambda$ of type-II superconductors in the vortex state for the purpose of obtaining indirect information on the energy gap structure [1]. The magnetic field distribution $n(B)$ in the sample is determined by detecting the decay positrons from implanted positive muons that locally probe the internal fields, and $\lambda$ is subsequently determined by modeling the contribution of the vortex lattice (VL) to $n(B)$. However, even in conventional superconductors the VL contribution is not known a priori, and one must rely on phenomenological models to deduce what is really an “effective” penetration depth $\lambda$. One reason for this is that only cumbersome microscopic theories account for the effects of low-energy excitations on $n(B)$ [2]. Extrapolating low-temperature measurements of $\lambda$ to zero field to eliminate the effects of intervortex transfer of quasiparticles, as well as nonlocal and nonlinear effects, has been demonstrated to be an accurate way of determining the “true” magnetic penetration depth $\lambda$ [3, 4]. Yet an underlying assumption is always that the VL is highly ordered and that other contributions to $n(B)$ are relatively minor. The purpose of this Letter is to point out that this is not the case in many of the recently discovered iron-based superconductors, making a reliable determination of $\lambda$ by TF-$\mu$SR extremely difficult.

Here we report on representative TF-$\mu$SR measurements of BaFe$_{12}Co_{0.18}As_2$ ($T_c = 21$ K) single crystals grown from a FeAs flux, as described elsewhere [3]. High-statistics TF-$\mu$SR spectra of 20 million muon decay events were collected in magnetic fields $H = 0.02$ T to 0.5 T applied transverse to the initial muon spin polarization $P(t=0)$, and parallel to the $c$-axis of the crystals. The TF-$\mu$SR signal is the time evolution of the muon spin polarization, and is related to $n(B)$ as follows

$$P(t) = \int_0^\infty n(B) \exp(i\gamma_\mu B t) dB,$$

where $\gamma_\mu$ is the muon gyromagnetic ratio. Generally, the TF-$\mu$SR signal is fit in the time domain, with the inverse Fourier transform or “TF-$\mu$SR line shape” given by

$$n(B) = \int_0^\infty P(t) \exp(-i\gamma_\mu B t) dt,$$

providing a visual approximation of the internal field distribution. The field distribution of a perfectly ordered VL is characterized by sharp cutoffs at the minimum and maximum values of $B(\mathbf{r})$, and a sharp peak at the saddle-point value of $B(\mathbf{r})$ [1]. These features are not observed in polycrystalline samples, where the orientation of the crystal lattice varies with respect to $H$, but are observed in single crystals when a highly-ordered VL exists and other contributions to $n(B)$ are relatively minor.

We have attempted to fit the TF-$\mu$SR spectra to a theoretical polarization function $P(t)$ that has been successfully applied to a wide variety of type-II superconductors, and utilized in some of the experiments on iron-based superconductors. The spatial variation of the field, from which $n(B)$ is calculated, is modeled by the analytical Ginzburg-Landau (GL) function [1]

$$B(\mathbf{r}) = B_0(1 - b^4) \sum_G \frac{e^{-i\mathbf{G} \cdot \mathbf{r}} u K_1(u)}{\xi^2 G^2},$$

where $G$ are the reciprocal lattice vectors of an hexagonal VL, $b = B/B_{c2}$ is the reduced field, $B_0$ is the average internal magnetic field, $K_1(u)$ is a modified Bessel function, and $u^2 = 2\xi^2 G^2(1 + b^4)|1 - 2b(1 - b)|$. As explained later, $P(t)$ is multiplied by a Gaussian de polarization function $\exp(-\sigma^2 t^2)$ to account for the effects
of nuclear dipolar fields and frozen random disorder. We stress that the fitting parameters $\xi$ and $\lambda$ can deviate substantially from the “true” coherence length and magnetic penetration depth if other contributions to $n(B)$ are significant. An important feature of Eq. (3) is that it accounts for the finite size of the vortex cores, by generating a “high-field” cutoff in $n(B)$. The GL coherence length $\xi_{ab} \sim 26 \, \text{Å}$ calculated from the upper critical field $H_{c2} \sim 50 \, \text{T}$ of BaFe$_{1.82}$Co$_{0.18}$As$_2$ with $H \parallel c$, represents a lower limit for the vortex core radius $\xi$. The core size can be much larger if there are spatially extended quasiparticle core states associated with either the existence of a second smaller superconducting gap $\Delta_2$ or a single anisotropic gap $\Delta_1$. Yet fits of the TF-$\mu$SR spectra of BaFe$_{1.82}$Co$_{0.18}$As$_2$ using Eq. (3), show no sensitivity to the vortex cores at any field and converge with values of $\xi$ approaching zero. Fig. 1 shows that even at 0.5 T where the vortex density is highest, a high-field cutoff is not discernible in the TF-$\mu$SR line shape. We next discuss reasons for this insensitivity to the vortex cores.

**Magnetism**—The effective field $B_{e}$ experienced by the muon is a vector sum of various contributions that may be static or fluctuating in time. With correlation times generally much longer than the muon life time, the nuclear moments constitute a dense static moment system that cause a Gaussian-like depolarization of the TF-$\mu$SR spectrum. Yet as shown in Fig. 2(a), BaFe$_{1.82}$Co$_{0.18}$As$_2$ exhibits an exponential depolarization above $T_c$ that is typical of dilute or fast fluctuating electronic moments. The latter interpretation is consistent with the observation of a paramagnetic (PM) shift of the average internal field $\langle B_{\mu} \rangle$ sensed by the muons below $T_c$. This is evident in Fig. 2(b), where we show representative Fourier transforms of $P(t)$ at $H = 0.02 \, \text{T}$. Instead of the expected diamagnetic shift imposed by the superconducting state, $\langle B_{\mu} \rangle$ exceeds $H$. The magnitude of the PM shift increases with increasing $H$ and/or decreasing $T$.

The occurrence of a PM shift in the superconducting state of BaFe$_{2-x}$Co$_x$As$_2$ and SrFe$_{2-x}$Co$_x$As$_2$ has been reported by others [11][12], and implies an enhancement of $\langle B_{\mu} \rangle$ from magnetic order occupying a large volume of the sample. Magnetic order exists in underdoped samples at $H = 0$ [10], and is apparently induced in overdoped samples by the applied field. Yet the effects of magnetism on the line width and functional form of $n(B)$ have not been considered. A strong relaxation of the TF-$\mu$SR signal occurs even in long-range magnetically ordered systems, and with decreasing temperature there must be an increased broadening of $n(B)$ associated with the growth.
of the correlation time for spin fluctuations.

Accounting for such magnetism is non-trivial because of the spatially-varying superconducting order parameter and the likelihood that the field-induced magnetism occurs in a nematic phase \[13\]. Even so we have achieved excellent fits of the TF-\(\mu\)SR spectra of \(\text{BaFe}_{1.82}\text{Co}_{0.18}\text{As}_{2}\) to polarization functions that incorporate enhanced magnetism in the vortex core region (\(e.g\). commensurate spin-density wave, ferromagnetism, spin-glass), where superconductivity is suppressed. Here we describe typical results for one form of magnetism: To account for line broadening from magnetism at higher temperatures, \(P(t)\) is multiplied by an exponential depolarization function \(\exp(-\Delta t)\), as observed above \(T_c\). In addition, enhanced magnetic order in the vortex cores is modeled by adding the following term to Eq. 3.

\[
B_{AF}(r) = B_{AF}e^{-\frac{1}{2}(r/\xi_{AF})^2} \sum_{K} \left( e^{-iK\cdot r} - e^{-iK\cdot r'} \right).
\]

The \(K\) sum is the reciprocal lattice of an antiferromagnetic (AF) square iron sublattice of spacing \(a = 2.8 \text{ Å}\), \(B_{AF}\) is the field amplitude, \(\xi_{AF}\) governs the radial decay of the field amplitude from the core center, and \(r\) and \(r'\) are the position vectors for ‘up’ and ‘down’ spins, respectively. This kind of magnetic order has the effect of smearing the high-field cutoff, and can even introduce a low-field tail in \(n(B)\) \[14\].

As indicated by the value of \(\xi\) in Fig. 1(b), fits to this model are sensitive to the vortex cores. With decreasing temperature, the magnetism-induced relaxation evolves from exponential to Gaussian, and the magnetic order in the vortex cores is enhanced. Consistent with the results Ref. \[12\], fits of TF-\(\mu\)SR spectra of overdoped \(\text{BaFe}_{2-x}\text{Co}_{x}\text{As}_{2}\) to a model that does not include magnetism and is insensitive to the vortex cores (\(i.e\). \(\xi\) fixed to 5 \(\text{Å}\)) yield an unusual linear temperature of \(1/\lambda^2\) immediately below \(T_c\), and a saturation of \(\lambda\) at low \(T\). In contrast, fits assuming magnetic order exhibit a linear temperature dependence well below \(T_c\) that is suggestive of gap nodes. However, these results simply demonstrate the ambiguity in modeling such data. Without knowledge of the precise form of the magnetism, our model cannot be deemed rigorously valid. Furthermore, as we explain next, VL disorder is a serious concern.

**Disorder**—Thus far TF-\(\mu\)SR has been applied to iron-based superconductors under the assumption that one is probing a fairly well-ordered hexagonal VL. Yet, to date this has been observed only in K\(\text{Fe}_2\text{As}_2\) \[13\]. Vortex imaging experiments on the \(R\text{Fe}_3\text{As}(O_1-x\text{F}_x)\), \(A_1-x\text{B}_x\text{Fe}_2\text{As}_2\) and \(A\text{Fe}_2-x\text{Co}_x\text{As}_2\) families all show a highly disordered VL indicative of strong bulk pinning \[10\] \[21\]. In Fig. 1 we show the effect of such disorder on the ideal \(n(B)\). Using a radial distribution function closely resembling that observed in overdoped \(\text{BaFe}_{1.81}\text{Co}_{0.19}\text{As}_2\) \[20\], we have used molecular dynamics to simulate \(n(B)\) of the disordered VL. Although the disordered line shape in Fig. 1(b) is asymmetric, it is strongly smeared with a field variation greatly exceeding that of the perfect VL.

Small perturbations of the VL by random pinning can be handled by convoluting the ideal theoretical line shape with a Gaussian distribution of fields \[22\]. This causes a Gaussian depolarization \(\exp(-\sigma^2t^2)\) of \(P(t)\). But for polycrystalline samples, \(n(B)\) of the perfect VL is nearly symmetric and cannot be isolated from a symmetric distribution of disorder. Consequently, VL disorder has not been accounted for in TF-\(\mu\)SR studies of polycrystalline or powdered iron-based superconductors \[23\] \[25\]. Given the severity of disorder in these materials and no knowledge about how this disorder evolves with temperature or doping, the accuracy of information deduced about \(\lambda\) is questionable. Since disorder of rigid flux lines broaden \(n(B)\), such studies certainly underestimate \(\lambda\).

While small perturbations of \(B(r)\) by vortex pinning may be accounted for in measurements on single crystals, a Gaussian convolution of the ideal \(n(B)\) becomes increasingly inadequate as the degree of disorder is enhanced \[20\]. In Fig. 1(b) we show that Gaussian broadening of the ideal line shape does not precisely re-
produce \( n(B) \) of the disordered VL. More importantly, however, because the large disorder-induced broadening smears out the high-field cutoff, the fitting parameters \( \lambda \) and \( \xi \) are ambiguous. This is illustrated in Fig. 4(c), where a nearly identical Gaussian broadened line shape is obtained for very different values of these parameters. Hence disorder introduces considerable uncertainty even in measurements on single crystals [11, 12, 27, 30].

In summary, the effects of magnetic order and/or random frozen disorder of the VL in iron-based superconductors introduce considerable uncertainty in values of \( \lambda \) obtained by TF-\( \mu \)SR. Unfortunately, these effects cannot be modeled in a reliable way. Compounding the problem is a lack of information on how the magnetism and VL disorder evolve with temperature. Consequently, caution is warranted in drawing conclusions about the anisotropy of the superconducting gap in these materials from TF-\( \mu \)SR measurements.

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