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Magnetism and Disorder Effects on μSR Measurements of the Magnetic Penetration Depth in Iron-Arsenic Superconductors

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It is shown that attempts to accurately deduce the magnetic penetration depth λ of overdoped $BaFe_{1.82}Co_{0.18}As_2$ single crystals by transverse-field muon spin rotation (TF-μSR) 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-arsenic superconductors, and this introduces considerable uncertainty in the values of λ obtained by TF-μSR.

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TF-μSR is routinely used to determine the magnetic penetration depth λ of type-II superconductors in the vortex state, which provides indirect information on the energy gap structure [1]. The magnetic field distribution $n(B)$ in the sample is measured by detecting the decay positrons from implanted positive muons that locally probe the internal fields, and λ 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 $\tilde{\lambda}$. One reason is that only cumbersome microscopic theories account for the effects of low-energy excitations on $n(B)$ [2]. Extrapolating low-temperature measurements of $\tilde{\lambda}$ to zero field to eliminate intervortex quasiparticle transfer, nonlocal and/or nonlinear effects, has been demonstrated to be an accurate way of determining the "true" λ [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-arsenic superconductors, making a reliable determination of λ by TF-μSR extremely difficult.

Here we report on representative TF-μSR measurements of $BaFe_{1.82}Co_{0.18}As_2$ ($T_c = 21$ K) single crystals grown from a FeAs flux, as described elsewhere [5]. Magnetic susceptibility measurements at 20 Oe show a sharp superconducting transition and complete diamagnetic screening, and EDS X-ray measurements on different parts of the crystal indicate a uniform Co composition. High-statistics TF-μSR 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-μSR signal is the time evolution of the muon spin polarization, and is related to $n(B)$ as follows

$$P(t) = \int_0^\infty n(B) \exp(i\gamma_\mu Bt) dB \,, \tag{1}$$

where γ_μ is the muon gyromagnetic ratio. Generally, the TF-μSR signal is fit in the time domain, with the inverse Fourier transform or "TF-μSR line shape" providing a visual approximation of the internal field distribution. For a perfectly ordered VL, $n(B)$ 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 the VL is highly-ordered and other contributions to $n(B)$ are minor.

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

$$B(\mathbf{r}) = B_0(1 - b^4) \sum_{\mathbf{G}} \frac{e^{-i\mathbf{G}\cdot\mathbf{r}} \; u\, K_1(u)}{\tilde{\lambda}^2 G^2} \,, \tag{2}$$

where $\mathbf{G}$ 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, $u^2 = 2\tilde{\xi}^2 G^2 (1 + b^4)[1 - 2b(1 - b)^2]$, and $\tilde{\xi}$ is the coherence length. As explained later, $P(t)$ is multiplied by a Gaussian depolarization 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 $\tilde{\lambda}$ and $\tilde{\xi}$ can deviate substantially from the "true" λ and ξ if other contributions to $n(B)$ are significant. An important feature of Eq. (2) is that it accounts for the finite size of the

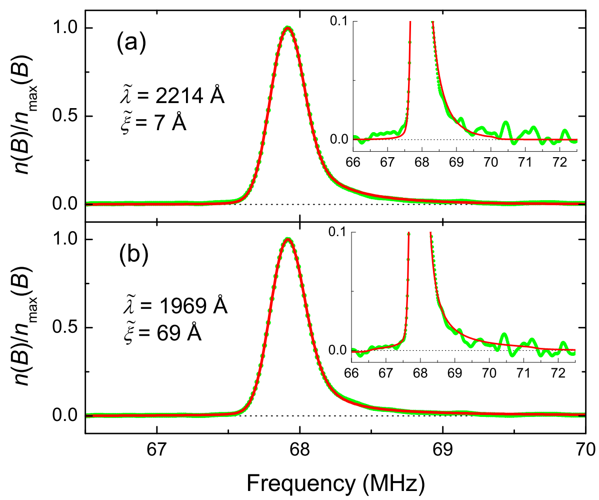

FIG. 1: (Color online) TF-μSR line shape of $BaFe_{1.82}Co_{0.18}As_2$ at $H = 0.5$ T and $T = 3.9$ K (green circles). (a) The red curve is the Fourier transform of a fit in the time domain assuming Eq. (2). In addition to the indicated values of $\tilde{\lambda}$ and $\tilde{\xi}$, the fit yields $\sigma = 0.265$ μs^{-1} and a PM shift of 8.6 G. (b) Fourier transform of a fit that assumes the model of field-induced AF order described in the main text (red curve). The fit yields $\sigma = 0.251$ μs^{-1} and a PM shift of 9.2 G. Other fit parameters are shown in Fig. 3.

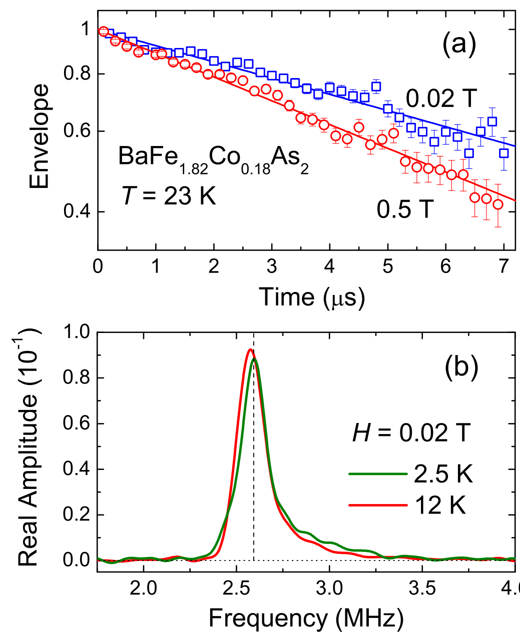

FIG. 2: (Color online) (a) Envelopes of TF-μSR spectra of $BaFe_{1.82}Co_{0.18}As_2$ in the normal state at $T = 23K$. The solid curves are fits to a single exponential relaxation function $G(t) = \exp(-\Lambda t)$, yielding $\Lambda = 0.081 \pm 0.003$ μs^{-1} and $\Lambda = 0.119 \pm 0.003$ μs^{-1} at $H = 0.02$ T and $H = 0.5$ T, respectively. (b) TF-μSR line shapes of $BaFe_{1.82}Co_{0.18}As_2$ below T_c at $H = 0.02$ T. The dashed vertical line corresponds to H.

vortex cores, by generating a "high-field" cutoff in $n(B)$. The GL coherence length $\xi_{ab} \sim 26$ Å calculated from the upper critical field $H_{c2} \sim 50$ T of $BaFe_{1.84}Co_{0.16}As_2$ with $\mathbf{H} \parallel c$ [9], represents a lower limit for the vortex core radius [3]. 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 [10] or a single anisotropic gap [11]. Yet fits of the TF-μSR spectra of $BaFe_{1.82}Co_{0.18}As_2$ using Eq. (2), show no sensitivity to the vortex cores at any field and converge with values of $\tilde{\xi}$ 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-μSR line shape. We next discuss two reasons for this:

Magnetism—The effective field $\mathbf{B}_\mu$ 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-μSR 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 [12]. The latter is consistent with the observation of a paramagnetic (PM) shift of the average internal field $\langle \mathbf{B}_\mu \rangle$ sensed by the muons below T_c. This is evident in Fig. 2(b), where we show representative Fourier transforms of $P(t)$ at $H = 0.02$ T. Instead of the expected diamagnetic shift imposed by the superconducting state, $\langle \mathbf{B}_\mu \rangle$ 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 $BaFe_{2-x}Co_xAs_2$ and $SrFe_{2-x}Co_xAs_2$ has been reported by others [6, 8], and implies an enhancement of $\langle \mathbf{B}_\mu \rangle$ from magnetic order occupying a large volume of the sample. Magnetic order exists in underdoped samples at $H = 0$ [13], 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-μSR 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 [14]. Even so we have achieved excellent fits of the TF-μSR spectra of $BaFe_{1.82}Co_{0.18}As_2$ to polarization functions that incorporate enhanced mag-

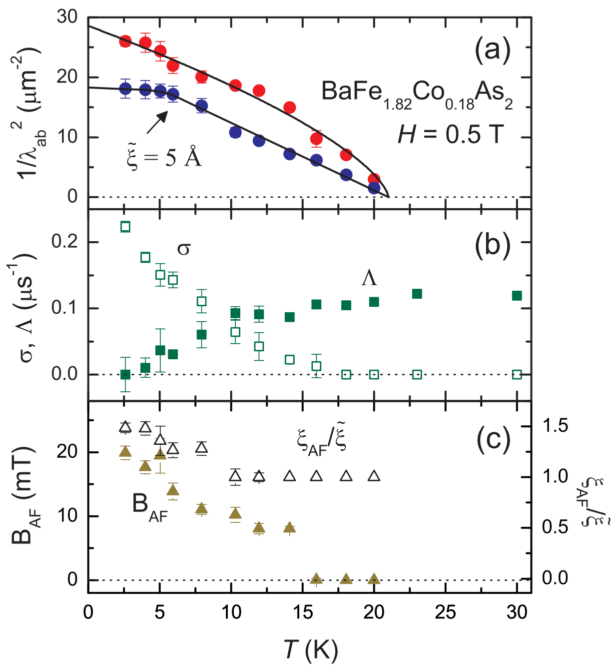

FIG. 3: (Color online) Results of fits of TF-μSR time spectra of $BaFe_{1.82}Co_{0.18}As_2$ at $H = 0.5$ T, assuming the model of magnetic order described in the main text. Temperature dependence of (a) $1/\tilde{\lambda}^2$, (b) the depolarization rates σ (Gaussian) and Λ (exponential), (c) $B_{\rm AF}$ and the ratio $\xi_{\rm AF}/\tilde{\xi}$. Also shown in (a) are results of fits without magnetic order, but with $\tilde{\xi}$ fixed to be 5 Å (blue circles).

netism 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 model of magnetism: First, $P(t)$ is multiplied by an exponential depolarization function $\exp(-\Lambda t)$, as observed above T_c. In addition, enhanced magnetic order in the vortex cores is modeled by adding the following term to Eq. (2)

$$B_{\rm AF}(\mathbf{r}) = B_{\rm AF} e^{-\frac{1}{2}(r/\xi_{\rm AF})^2} \sum_{\mathbf{K}} \left(e^{-i\mathbf{K}\cdot\mathbf{r}} - e^{-i\mathbf{K}\cdot\mathbf{r}'} \right) . \quad (3)$$

The $\mathbf{K}$ sum is the reciprocal lattice of an antiferromagnetic (AF) square iron sublattice of spacing $a = 2.8$ Å, $B_{\rm AF}$ is the field amplitude, $\xi_{\rm AF}$ governs the radial decay of $B_{\rm AF}$ from the core center, and $\mathbf{r}$ and $\mathbf{r}'$ 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 $n(B)$ [15]. As indicated by the large value of $\tilde{\xi}$ 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 (see Fig 3(b)), and the magnetic order in the vortex cores is enhanced (see Fig. 3(c)). Consistent with behavior deduced from TF-μSR measurements on $BaFe_{1.772}Co_{0.228}As_2$ [8], fits to a model without magnetism that is insensitive to the vortex cores (*i.e.* $\tilde{\xi}$ fixed to 5 Å) yield an unusual linear temperature dependence of $1/\tilde{\lambda}^2$ immediately below T_c, and a saturation of $\tilde{\lambda}$ at low T (see Fig. 3(a)). 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-μSR has been applied to iron-arsenic superconductors under the assumption that one is probing a fairly well-ordered hexagonal VL. Yet to date this has been observed only in KFe_2As_2 [16]. Vortex imaging experiments on the RFeAs($O_{1-x}F_x$), $A_{1-x}B_xFe_2As_2$ and $AFe_{2-x}Co_xAs_2$ families all show a highly disordered VL indicative of strong bulk pinning [17–22]. In Fig. 4 we show the effect of such disorder on the ideal $n(B)$. We used molecular dynamics to simulate $n(B)$ of the disordered VL. In particular, molecular dynamics iterations were performed until a radial distribution function closely resembling that observed in overdoped $BaFe_{1.81}Co_{0.19}As_2$ [21] was achieved (see Fig. 4(a)). The vortex configuration at this point was then assumed to be static and $n(B)$ was calculated. Although the line shape of the disordered VL in Fig. 4(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 [23]. This causes a Gaussian depolarization $\exp(-\sigma^2 t^2)$ of $P(t)$. But for polycrystalline samples, $n(B)$ is always nearly symmetric, so that the contribution from disorder cannot be isolated. Consequently, VL disorder has not been accounted for in TF-μSR studies of polycrystalline or powdered iron-arsenic superconductors [24–27]. 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 λ is questionable. Since disorder of rigid flux lines broaden $n(B)$, such studies certainly underestimate λ.

While small perturbations of $B(\mathbf{r})$ 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 [28]. In Fig. 4(b) we show that Gaussian broadening of the ideal line shape does not precisely reproduce $n(B)$ of the disordered VL. Moreover, because the large disorder-induced broadening smears out the high-field cutoff, the fitting parameters $\tilde{\lambda}$ and $\tilde{\xi}$ are ambiguous. This is illustrated in Fig. 4(c), where nearly identi-

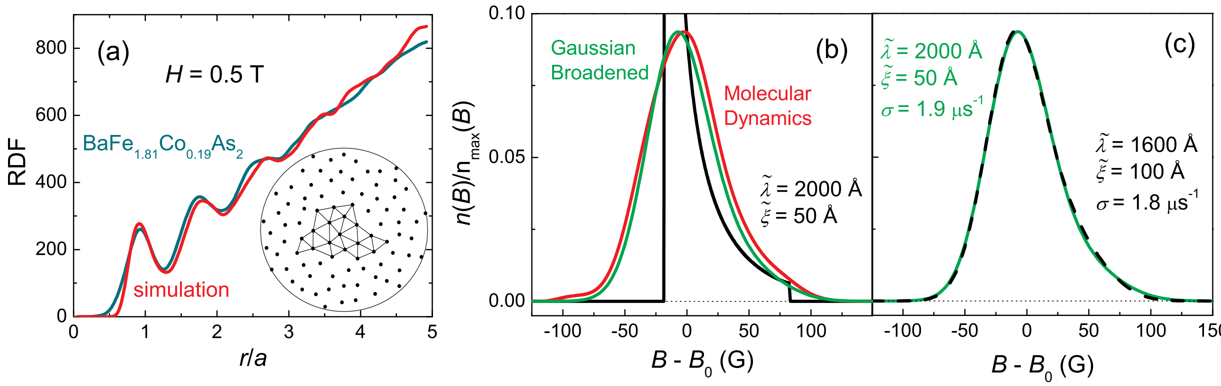

FIG. 4: (Color online) (a) Radial distribution function (RDF) of $BaFe_{1.81}Co_{0.19}As_2$ at $H = 0.5$ T from Ref. [21] and of the disordered VL shown in the lower right generated by molecular dynamics (MD). Note 5000 vortices were used in the MD simulation. The horizontal scale is normalized with respect to the intervortex spacing $a = 691$ Å of the perfect hexagonal VL. (b) Theoretical simulations of the TF-μSR line shape of the perfect VL (black curve) and of the disordered VL (red curve) corresponding to the RDF shown in (a). The green curve is the line shape of the perfect VL convoluted by a Gaussian distribution of fields, corresponding to $\sigma = 1.9\ \mu s^{-1}$. All three simulations assume $\tilde{\lambda} = 2000$ Åand $\tilde{\xi} = 50$ Å. (c) Same Gaussian-broadened line shape in (b) and a Gaussian-broadened ideal line shape with $\tilde{\lambda} = 1600$ Å, $\tilde{\xi} = 100$ Å, and $\sigma = 1.8\ \mu s^{-1}$. The heights of the line shapes in (b) and (c) are normalized with repect to the height $n_{\max}(B)$ of the ideal line shape.

cal Gaussian broadened line shapes are obtained for very different values of these parameters. Hence substantial disorder introduces considerable uncertainty even in measurements on single crystals [6–8, 29–31].

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

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