London penetration depth in single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ spanning underdoped to overdoped compositions

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The London penetration depth $\lambda(T)$ has been measured in single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ using the tunnel diode resonator technique. The measured doping levels of $x=0.038$, 0.047, 0.058, 0.074, and 0.10 range from underdoped to overdoped concentrations. The measurements have shown that the density of carriers participating in superconductivity decreases sharply in the underdoped regime but the penetration depth as a function of temperature exhibits a robust power law, $\Delta\lambda(T) \sim T^n$, for all measured dopings with n being about 2 in underdoped samples and 2.5 in overdoped samples. We discuss the implications of these results and possible interpretations of such a robust behavior.

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The structure and symmetry of the superconducting order parameter is of crucial importance for determining the pairing mechanism in the newly discovered Fe-based pnictide superconductors. A useful method of probing the gap structure is to measure the magnetic penetration depth in single crystals. The two parent systems for which a considerable amount of effort has been put forth to study are (RE)FeAsO (1111) and (AE)Fe₂As₂ (122), where RE is a rare earth and AE is an alkali earth.

In the fluorine doped or oxygen deficient 1111 system, the majority of experiments indicate a fully gapped Fermi surface (FS). Measurements of the London penetration depth, $\lambda(T)$, using a tunnel diode resonator (TDR) technique on $NdFeAsO_{0.9}F_{0.1}$ (Ref. [1]) and $SmFeAsO_{1-x}F_y$ (Ref. [2]) as well as microwave cavity perturbation on $PrFeAsO_{1-y}$ (Ref. [3]) have found an exponential temperature dependence of $\lambda(T)$ at low temperatures. Similar conclusions have been reached by muon spin relaxation (μSR) studies and most point contact Andreev reflection measurements,[4,5] although some of them have been interpreted in terms of nodal gaps.[6] Knight shift measurements indicate spin singlet superconductivity. The spin-lattice relaxation rate, $1/T_1 \sim T^3$, is characteristic of nodal superconductivity but can be reconciled with the extended $s_{\pm}$ model by pair-breaking scattering under particular assumptions about its strength and defect concentration.[7]

The situation in the 122 system is somewhat more controversial. TDR measurements on $Ba(Fe_{0.93}Co_{0.07})_2As_2$ have shown a clear nonexponential behavior of $\lambda(T)$ (Ref. [8]), whereas microwave measurements on $Ba_{1-x}K_xFe_2As_2$ were interpreted in terms of two fully opened superconducting gaps.[9] In this Rapid Communication, we focus on penetration depth studies of large single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ with different Co dopings. We find that the penetration depth exhibits a robust power law, $\Delta\lambda(T) \approx C(T/T_c)^n$, for all x. There is a clear change of regime at $x \sim 0.06$, where (i) the orthorhombic/antiferromagnetic–tetragonal/nonmagnetic phase boundary crosses the superconducting phase boundary (Fig. 1), (ii) n changes from 2.0 ± 0.1 to 2.4 ± 0.1, and (iii) the coefficient C suddenly drops by an order of magnitude (Fig. 5). Unless there is a sudden change in the impurity scattering around $x=0.06$, this strongly suggests that the exponent values and the power law itself are not due to impurities, but rather are an intrinsic characteristic related to the proximity to the magnetic ordering/structural transition. One of our separate studies shows a $\lambda(T) \sim T^2$ behavior in a hole-doped 122 system, $(Ba_{1-x}K_x)Fe_2As_2$, as well.[10]

Single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ were grown out of self-flux.[11] The actual cobalt concentration was determined by wavelength dispersive x-ray spectroscopy in the electron probe microanalyzer of a JEOL JXA-8200 Superprobe. Magneto-optical imaging has revealed homogeneous Meissner screening. Slabs with sizes of $\sim 1 \times 1 \times 0.2$ mm³ and mirrorlike surfaces were cleaved with a razor blade from larger crystals.

The in-plane London penetration depth, $\lambda(T)$, has been measured using a self-oscillating TDR.[12] A sample to be studied is mounted on a sapphire rod and inserted into the inductor coil of the tank circuit. Throughout the measurement the temperature of the circuit is stabilized at $5.00 \text{ K} \pm 0.01$ K. This is essential for stability in the measured resonant frequency, which is resolved to about 0.01 Hz. This translates to the ability to detect changes in $\lambda(T)$ in the range of about 1 Å. The ac magnetic excitation field in the coil is about 20 mOe, which is small enough to ensure that no vortices are present. The sample, with magnetic susceptibility $\chi(T)$, leads to a change in the resonant frequency of $\Delta f \equiv f(T) - f_0 = -G 4\pi\chi(T)$, where $f_0 = 1/2\pi\sqrt{LC} \approx 14$ MHz and $G \simeq f_0 V_s / 2V_c (1-N)$ is a geometrical calibration factor defined by the coil characteristics and the sample volume V_s. G is measured directly by pulling the sample out of the coil at the lowest temperature.[13] The susceptibility of a rectangular superconducting slab in the Meissner state can be written in terms of $\lambda(T)$ and a characteristic dimension R, as $4\pi\chi(T) = \lambda/R \tanh(R/\lambda) - 1$ (Ref. [13]).

Microscopic, thermodynamic, and transport measurements of the $Ba(Fe_{1-x}Co_x)_2$ crystals used in this study have shown superconductivity coexists with the orthorhombic phase in the underdoped regime in this particular system.[11] Our TDR measurements reveal similar features. Figure 1(a)
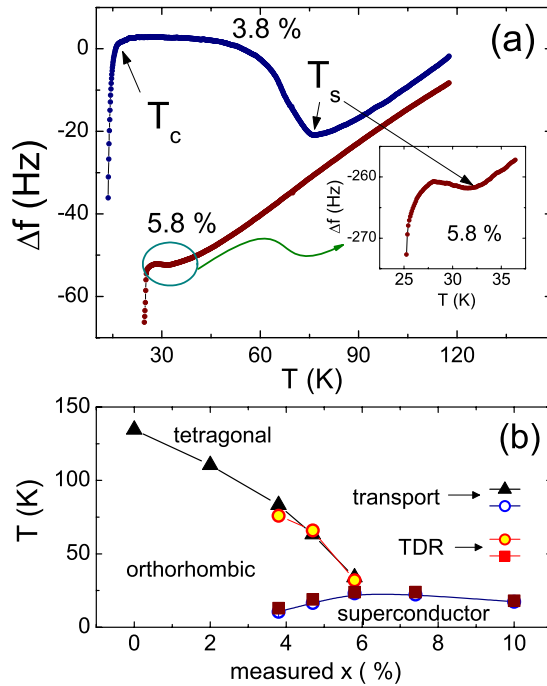
FIG. 1. (Color online) (a) Raw data for $x=0.038$ (underdoped) and $x=0.058$ (near optimal doping, the data have been divided by a factor of 5 for clarity). The inset emphasizes a magnetic/structural transition. (b) Phase diagram showing structural, T_s, and superconducting, T_c, transitions determined from transport (Ref. [11]) and TDR measurements.

FIG. 3. (Color online) Low-temperature behavior of $\Delta\lambda(T)$ for all studied concentrations. Solid lines are the fits to $\Delta\lambda \propto C(T/T_c)^n$ with C and n as free parameters.

shows TDR frequency shifts as a function of temperature for scans running from below T_c to ≈ 120 K for two samples with $x=0.038$ and 0.058. The data for the $x=0.058$ sample have been divided by a factor of 5 for clarity. In the normal state, the magnetic penetration depth is limited by the skin depth, which depends on the normal-state resistivity. The overall variation of Δf over the transition region is about 20 Hz, which corresponds to a variation of about 45 nm in the skin depth. This should be compared to the 13300 Hz change corresponding to the superconducting transition of the sample. The temperature of the structural/magnetic transition, T_s, was defined to be at the position of the minimum in
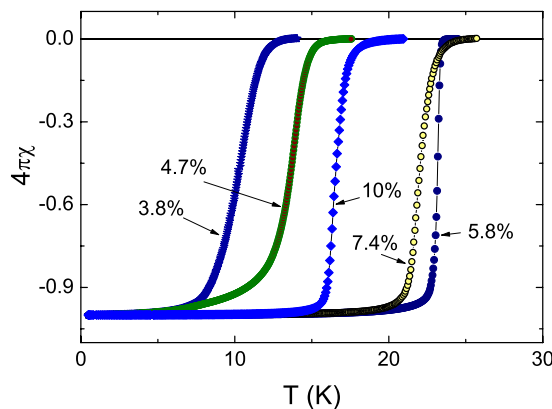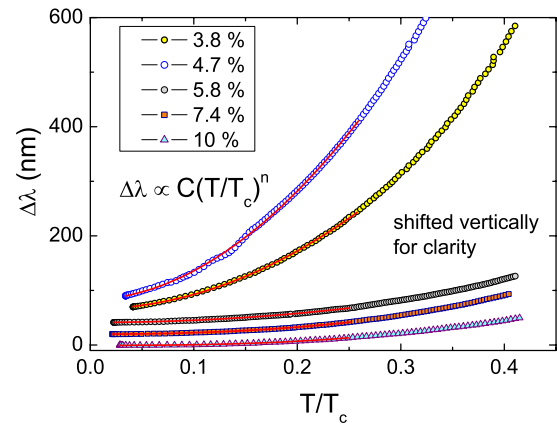
$\Delta f(T)$ (see Fig. 1). The superconducting T_c was defined at the onset. Both temperatures are in excellent agreement with transport measurements,[11] as shown in Fig. 1(b). Figure 2 shows the rf susceptibility constructed from the TDR frequency shifts in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ for $x=0.038$, 0.047, 0.058, 0.074 and 0.10, which cover the range from underdoped to overdoped. Optimal doping for this series occurs for a concentration between $x=0.058$ and 0.074.

The low-temperature variation of the penetration depth is examined in Fig. 3. For all superconducting samples we observe a power law $\Delta\lambda(T) \propto CT^n$. The fitted exponent n varies from $n=2\pm0.1$ for underdoped samples to $n=2.5\pm0.1$ for the overdoped samples. If the superconducting density itself follows a power law with a given n, then $C=f_s(c/\omega_p)S$, where f_s is the superconducting fraction at zero temperature, c is the speed of light, and S is defined by the fraction of the Fermi surface that is gapless (which may reflect a multigap character of the superconductivity, possible nodal structure, unitary impurity scattering strength, etc.) and ω_p is the plasma frequency.

To examine how close the overall power-law behavior is to quadratic, we plot $\Delta\lambda[(T/T_c)^2]$ in Fig. 4 scaled by the prefactor A obtained from the fitting of the data to $\Delta\lambda(T) \propto A(T/T_c)^2$ at low temperatures (below 0.3 T_c) with A being the only free parameter. At a gross level, all samples follow the $\lambda(T) \sim T^2$ behavior rather well.

FIG. 2. (Color online) $4\pi\chi(T)$ in single crystals of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ for different x.
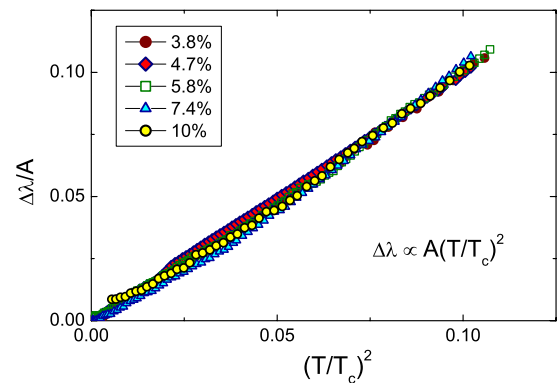

FIG. 4. (Color online) Scaled $\Delta\lambda(T)/A$ vs $(T/T_c)^2$ where A is obtained from the fitting, $\Delta\lambda \propto A(T/T_c)^2$.
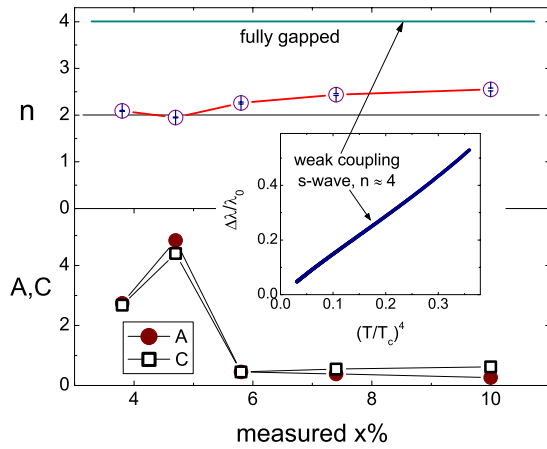
FIG. 5. (Color online) Doping dependence of the exponent n (upper panel) and of the fitting prefactors A and C (lower panel). The inset shows that the exponential behavior described by Eq. (1) is well represented by the power law with $n \approx 4$.

To summarize the observed power-law behavior, the upper panel of Fig. 5 shows the exponent n that was obtained from the best fit with two free parameters, $\Delta\lambda(T) \propto CT^n$. The lower panel of Fig. 5 shows the prefactor C obtained from the above fit as well as a prefactor obtained by fitting to a pure quadratic behavior, $\Delta\lambda(T) \propto AT^2$. There is a clear change of regime at $x \sim 0.06$: (i) at lower x the coexistence of antiferromagnetism and orthorhombicity with superconductivity has been inferred[11] (also, see Fig. 1), (ii) at $x \sim 0.06$, n changes from 2.0 ± 0.1 to 2.4 ± 0.1, and (iii) the coefficient C suddenly drops by an order of magnitude (Fig. 5).

Let us now discuss the implications of these observations. If the FS is fully gapped, the penetration depth of a homogeneous and clean superconductor in the local limit exhibits the well-known exponential temperature dependence:

$$\Delta\lambda(T) = \lambda(0)\sqrt{\frac{\pi\Delta_{min}}{2k_B T}}\exp\left(-\frac{\Delta_{min}}{k_B T}\right), \qquad (1)$$

where Δ_{min} is the minimal value of the gap. This holds roughly until $k_B T \lesssim \Delta_{min}/5$. Our data are not consistent with this behavior. In what follows we comment on the viability of various scenarios that yield power-law behavior of $\lambda(T)$. If a material is clean, in the local limit, anisotropic pairing with line or point nodes of the pairing gap leads to a linear or quadratic temperature dependence of $\Delta\lambda(T)$, respectively. Thus, the most direct interpretation of our data would be in terms of point nodes of the gap,[14,15] as for example in PrOs$_4$Sb$_{12}$ (Ref. 15). However, this is only correct if the system is clean and in the local limit. $\Delta\lambda(T) \propto T^2$ is consistent with line nodes[16] of the gap if one includes scattering by impurities[17] or nonlocal corrections.[18] Unitary impurity scattering creates a state with a quadratic behavior below some characteristic temperature[17] $k_B T^* \sim \Gamma$, where Γ is the impurity scattering rate. In our case the requirement would be $\Gamma \gtrsim k_B T_c/3$. This explanation requires relatively strong impurity scattering[19] with a substantial unitary component, consistent with the fact that our samples are doped in the active plane, but implying that in clean samples, i.e., samples doped

away from the Fe-As planes, a linear behavior should be restored. A quadratic T dependence of $\Delta\lambda$ may also be the result of strong impurity scattering in a pairing state that is fully gapped in the clean limit. The exponential behavior of Eq. (1) transforms to quadratic if the gap is driven into a gapless or near-gapless regime by impurity scattering. As in Ref. 7, it would require a scattering rate Γ of the order of the smallest gap Δ_{min} and a relatively fine balance between the unitary and Born scattering regimes. In clean samples, i.e., those doped away from the Fe-As planes, an exponential, or respectively, linear behavior should be restored. Indeed, the existing data for the 1111 system[1,2,20] surprisingly do not show a quadratic dependence; namely, an exponential one in the As-based compounds[1,2] but a linear one in the P-based system.[20] Furthermore, a problem with this explanation is that we do not see any systematic dependence of the $\Delta\lambda(T)$ characteristics on impurity concentration, but rather an abrupt change in regime as we cross the structural/magnetic phase transition.

Another mechanism that may transform the linear behavior in a state with line nodes into a quadratic one at $T < T^* \simeq \Delta(0)\xi_0/\lambda(0)$ is due to nonlocal effects,[18] where ξ_0 is the coherence length. In the Fe pnictides, however, T^* would be less than 1 K.

The power-law behavior of $\Delta\lambda$ may also be a consequence of the material being inhomogeneous. While the observation of a smearing of the jump in heat capacity at T_c in under- and overdoped samples[11] may be considered a hint for such a scenario, we do see very homogeneous Meissner screening in magneto-optical measurements. Also the jump in C_p is doping dependent and it is unlikely that any inhomogeneity can explain the universal behavior shown in Fig. 3 for all concentrations.

In view of this discussion, it is tempting to look for an explanation on a phenomenological level that would not rely on impurity scattering as a crucial element changing the functional dependence of λ. Indeed, any excitation coupled to electrons with an energy larger than $\sim 2\pi T$ is pairbreaking, including regular phonons. Moreover, for an $s^{\pm}$ or a d-wave state even phonons with arbitrarily small energies can be pairbreaking. The same holds for the coupling to other collective bosonic modes, such as antiferromagnetic spin fluctuations. Since thermally excited bosons are needed for pairbreaking, the scattering rate Γ becomes T dependent (Ref. 21). In the case of line nodes, where a T-independent Γ yields an exponent $n=2$, strong scattering off of the thermally excited bosons would always yield a smaller exponent. Given the special role that the AFM critical point seems to play, the possibility exists that the pair-breaking fluctuations are associated with an intermediate range dynamic ordering, like the dynamic domains speculated in Ref. 22. These will have very small energy and a potential to be strong scatterers. A clear derivation of the exponent n that results from such a picture is still missing.

To summarize, we have measured the temperature dependence of the penetration depth in single crystalline Co-doped BaFe$_2$As$_2$. The main observations are as follows: (1) the superconducting density in Ba(Fe$_{1-x}$Co$_x$)$_2$ changes quadratically with temperature to at least $T_c/3$ and so exponential or linear behavior can be safely excluded; (2) there is a sharp

change in the T dependence of the penetration depth (and probably in the absolute value at zero temperature), which occurs near the same concentration at which the magnetic ordering in the normal state disappears; (3) there is no visible sample quality effect on the exponent and amplitude of the temperature-dependent part of the penetration depth.

The observed behavior is compatible with neither fully gapped nor line node superconductivity; "accidental" point nodes within the $s^{\pm}$ model can be excluded based on weak concentration dependence of the power-law exponent, but could still be considered for alternative pairing mechanisms. Impurity-driven quadratic behavior is possible, but seems somewhat problematic even for a line-nodal state, and especially for a nodeless state, given no visible dependence on the impurity concentration. The possible influence of the proximity to antiferromagnetic ordering suggests an intriguing interpretation in terms of pair-breaking defects of magnetic nature whose concentration is controlled by the temperature and not by the Co concentration.

Note added in proof. Recent data on Nd and La 1111 crystals[25] show that the apparent flattening of the penetration depth in Nd-1111 is due to the paramagnetic contribution from Nd ions.

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