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London penetration depth in iron-based superconductors

R Prozorov\(^1, 2\) and V G Kogan\(^1\)

\(^1\) The Ames Laboratory, Ames, IA 50011, USA
\(^2\) Department of Physics & Astronomy, Iowa State University, Ames, IA 50011, USA

E-mail: prozorov@ameslab.gov and kogan@ameslab.gov

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Abstract

Measurements of London penetration depth, a sensitive tool to study multiband superconductivity, have provided several important insights into the behavior of Fe-based superconductors. We first briefly review the ‘experimentalist-friendly’ self-consistent Eilenberger two-band model that relates the measurable superfluid density and temperature dependences of the superconducting gaps. Then we focus on BaFe\(_2\)As\(_2\)-derived materials, for which the results are consistent with (1) two distinct superconducting gaps; (2) development of strong in-plane gap anisotropy with departure from optimal doping; (3) development of gap nodes along the c direction in a highly overdoped regime; (4) significant pair-breaking, presumably due to charge doping; (5) fully gapped intrinsic behavior (exponential at low temperatures) at optimal doping if the scattering is removed (probed in the ‘self-doped’ stoichiometric LiFeAs); (6) competition between the magnetically ordered state and superconductivity, which do coexist in underdoped compounds. Overall, it appears that while there are common trends in the behavior of Fe-based superconductors, the gap structure is non-universal and is quite sensitive to the doping level. It is plausible that the rich variety of possible gap structures within the general \(s_\pm\) framework is responsible for the observed behavior.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

After the initial discoveries of superconductivity in Fe-based superconductors, first in LaFePO with \(T_c \approx 4\) K [1] and then in LaFeAs(O\(_{1-x}\)F\(_x\)) with \(T_c \approx 26\) K [2], \(T_c\) as high as 55 K has been reported in SmFeAs(O\(_{1-x}\)F\(_x\)) [3]. These findings have triggered intense research aimed at understanding the fundamental physics governing this new family of superconductors. Since the original discoveries, at least seven different classes of iron-based superconductors have been identified, all exhibiting unconventional physical properties [4–7]. Among these, perhaps the most diverse family is based on (AE)Fe\(_2\)As\(_2\) parent compounds (where AE=alkaline earth, denoted as the ‘122’ system) in which electron [8–11], hole [12, 13] and isovalent [14, 15] dopants induce a superconductivity ‘dome’ with maximum \(T_c\) achieved at fairly
high concentrations of the dopant (up to 45% in the case of BaK122) and showing coexistence of superconductivity and long-range magnetic order on the underdoped side.

After more than three years into the intense study of Fe-based superconductors, several experimental conclusions supported by a large number of reports were drawn. Among those, relevant to our discussion of London penetration depth are the following: (1) two distinct superconducting gaps of magnitude ratio of about 2; (2) power-law behavior of thermodynamic quantities at low temperatures presumably due to pair-breaking scattering and gap anisotropy; (3) doping-dependent three-dimensional gap structure possibly with nodes. For some reviews of various experiments and theories the reader is referred to [4–7, 11, 16–19]. Here we focus on London penetration depth as a sensitive tool to study superconducting gap structure, albeit averaged over the Fermi surface [20].

2. Measurements of London penetration depth

There are several ways to measure magnetic penetration depth in superconductors [20]. These include muon-spin rotation (μSR) [21–23], frequency-dependent conductivity [24, 25], magnetic force and superconducting quantum interference device (SQUID) microscopy [26, 27], measurements of the first critical field using either global or local probes [28, 29], microwave cavity perturbation [32–36], mutual inductance (especially suitable for thin films) [37] and the self-oscillating tunnel-diode resonator (TDR) [28, 34, 38–43]. Each technique has its advantages and disadvantages, and only by a combination of several different measurements performed on different samples and under different experimental conditions can one obtain a more or less objective picture. In the case of Fe-based superconductors, most of the reports agree in general, but still differ on the details. Most difficulties come from the necessity to resolve very small variations of the penetration depth at the lowest temperatures (below 1 K) while maintaining extreme temperature resolution and the ability to statistically measure a sufficient number of data points over the full temperature range. In addition, it is important to look at a general picture by measuring many samples of each composition and over an extended doping regime. This review is based on the results obtained with a self-oscillating TDR that has the advantage of providing stable resolution of about 1 part per 10⁹ [20]. Indeed, comparison of TDR with other techniques, especially bulk probes, such as thermal conductivity, is important to come up with a consensus regarding the gap structure in pnictide superconductors [44–46].

2.1 Tunnel-diode resonator

2.1.1 Frequency-domain measurements. When a non-magnetic conducting sample is inserted into a coil of a tank circuit of quality factor Q and resonating at a frequency \( f_0 = 1/2\pi \sqrt{L_0C} \), it causes changes in the resonant frequency and the quality factor. For a flat slab of width 2\( w \) and volume \( V_i \) in parallel field [47],

\[
\frac{\Delta f}{f_0} = \frac{f_0 - f(\alpha)}{f_0} = \frac{V_i}{2V_c} \left( 1 - \frac{\tanh(\alpha w)}{\alpha w} \right),
\]

\[
\Delta \left( \frac{1}{Q} \right) = \frac{V_i}{V_c} 2 \alpha\left( \frac{\tanh(\alpha w)}{\alpha w} \right),
\]

where \( \alpha = (1 - i)/\delta \) in a normal metal and \( \alpha = 1/\lambda \) in a superconductor. Here, \( \delta \) is the normal skin depth and \( \lambda \) is the London penetration depth. \( V_i \) is volume of the inductor. Solving explicitly, we have for a normal metal,

\[
\frac{\Delta f}{f} = \frac{V_i}{2V_c} \left( 1 - \frac{\delta}{2w} \frac{\sinh(\frac{3w}{2}) + \sin(\frac{\delta w}{2})}{\cosh(\frac{3w}{2}) + \cos(\frac{\delta w}{2})} \right)
\]

and in a superconductor, we obtain the known ‘infinite slab’ solution:

\[
\frac{\Delta f}{f} = \frac{V_i}{2V_c} \left( 1 - \frac{\lambda}{w} \frac{\tanh(\frac{w}{\lambda})}{\lambda} \right),
\]

which allows us to measure both superconducting penetration depth and normal-state skin depth (which gives the contactless measure of resistivity).

In the case of a finite sample with magnetic susceptibility \( \chi \), we can still write \( \Delta f = -4\pi T \chi(T)G \), where \( G \approx f_0 V_i/2V_c(1 - N) \) is a geometric calibration constant that includes the effective demagnetization factor \( N \) [48]. Constant \( G \) is measured directly by pulling the sample out of the coil [20, 48]. The susceptibility in the Meissner state can be written in terms of London penetration depth \( \lambda(T) \) and normal state paramagnetic permeability (if local-moment magnetic impurities are present), \( \mu(T) \), as [48]

\[
4\pi T \chi = \frac{\sqrt{\mu(T)} \lambda(T)}{R} \tanh \left( \frac{\sqrt{\mu(T)} R}{\lambda(T)} \right) - 1,
\]

where \( R \) is the characteristic sample dimension. For a most typical slab geometry in an experiment with \( 2b \geq 2a \) planar dimensions and thickness \( 2d \) (magnetic field excitation is along the \( d \)-side), \( R \) is approximately given by [48]

\[
\frac{w}{R} \approx 2 \left[ 1 + \left( 1 + \frac{2d}{w} \right)^2 \right] \arctan \left( \frac{w}{2d} - \frac{2d}{w} \right)
\]

with \( w \approx ab/(a + b) \).

Unlike the microwave cavity perturbation technique that requires scanning of the frequency [47], TDR is a self-oscillating resonator always locked onto its resonant frequency [49]. The 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 (and of the coil) is stabilized at \( \pm 1 \) mK. This is essential for the stability of the 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 a few angstroms. The ac magnetic excitation field, \( H_{ac} \), in the coil is about 20 mOe, which is small enough to ensure that no vortices are created and the London penetration depth is measured.

2.1.2 Measurements of the absolute value of \( \lambda(T) \). The described TDR technique provides precise measurements of
the variation of the penetration depth, $\Delta \lambda(T)$, but not the absolute value for reasons described in detail elsewhere [50]. However, the TDR technique can be extended to obtain the absolute value of the penetration depth, $\lambda(T)$. The idea is to coat the entire surface of the superconductor with a thin film of a conventional superconductor with lower $T_c$ and a known value of $\lambda(0)$. In this work we used aluminum, which is convenient, since its $T_c^{Al}$ is quite low for most of the discussed materials, so it is possible to extrapolate to $T = 0$ and obtain $\lambda(0)$. While the Al film is superconducting it screens the magnetic field and the effective penetration depth is [39]

$$\lambda_{\text{eff}}(T) = \lambda_{Al}(T) \frac{\lambda(T) + \lambda_{Al}(T) \tanh \left( \frac{\pi T}{2 \lambda_{Al}(T)} \right)}{\lambda_{Al}(T) + \lambda(T) \tanh \left( \frac{\pi T}{2 \lambda_{Al}(T)} \right)} \quad (7)$$

However, when it becomes normal, the Al layer does not cause any screening because its thickness, $t$, is much less than the normal-state skin depth at the TDR operating frequency of 14 MHz, where $\delta_{Al} \approx 75 \mu m$ for $\rho_{Al}=10 \mu \Omega \cdot cm$ [51]. By measuring the frequency shift upon warming from the base temperature, $T = T_{\text{min}}$, to $T = T_c^{Al}$ where $\lambda_{Al} \to \infty$ and equation (7) gives $\lambda_{\text{eff}}(T_c^{Al}) = t + \lambda(T_c^{Al})$, we can calculate $\lambda(T_c^{Al})$. We also note that if aluminum coating is damaged (i.e. cracks and/or holes), the result will underestimate $\lambda(0)$. This method, therefore, provides the lower boundary estimate.

2.1.3. Out-of-plane penetration depth. Finally, the TDR technique can be used to obtain the out-of-plane component of the penetration depth, $\lambda_c$. For the excitation field $H_{ac}$, screening currents flow only in the $ab$-plane and $\Delta f$ is only related to the in-plane penetration depth, $\Delta \lambda_{ab}$. However, when the magnetic field is applied along the $ab$-plane ($H_{ac}$), screening currents flow both in the plane and between the planes, along the $c$-axis. In this case, $\Delta f^\perp$ contains contributions from both $\Delta \lambda_{ab}$ and $\Delta \lambda_c$. For a rectangular sample of thicknesses $2t$, width $2w$ and length $l$, $\Delta f^\perp$ is approximately given by

$$\frac{\Delta f^\perp}{\Delta f^\parallel} \approx \frac{\Delta \lambda_{ab} t}{l} + \frac{\Delta \lambda_c}{w} = \frac{\Delta \lambda_{\text{min}}}{R_b}, \quad (8)$$

where $R_b$ is the effective dimension that takes into account finite size effects [48], equation (6). Knowing $\Delta \lambda_{ab}$ from the measurements with $H_{ac}$ and the sample dimensions, one can obtain $\Delta \lambda_c$ from equation (8). However, because $2w \geq 4 \times 2t$ in most cases, $\Delta f^\perp$ is in general dominated by the contribution from $\Delta \lambda_{ab}$. The subtraction of $\Delta \lambda_c$ becomes therefore prone to large errors. An alternative, more accurate approach is to measure the sample twice [52]. After the first measurement with the field along the longest side $l$ ($H_{ac}$), the sample is cut along this $l$ direction into two halves, so that the width (originally $2w$) is reduced to $w$. Since the thickness $2t$ remains the same, we can now use equation (8) to calculate $\Delta \lambda_c$ without knowing $\Delta \lambda_{ab}$. Note that the length $l$ and width $w$ are in the crystallographic $ab$-plane, whereas the thickness $2t$ is measured along the $c$-axis. In our experiments, both approaches to estimate $\Delta \lambda_c(T)$ produced similar temperature dependence, but the former technique had a larger data scatter, as expected.

3. London penetration depth and superconducting gap

In order to describe the London penetration depth in multigap superconductors and to take into account pair-breaking scattering, we use the weak coupling Eilenberger quasi-classical formulation of the superconductivity theory that holds for a general anisotropic Fermi surface and for any gap symmetry [53]. This method, suitable for the analysis of the experimental data, is described in a few publications of one of us [40, 54–56]; here we briefly summarize the scheme.

In the clean case, the Eilenberger equations read

$$v \Pi f = 2 \Delta g / h - 2 \omega f, \quad (9)$$

$$-v \Pi^* f^* = 2 \Delta^* g / h - 2 \omega f^*, \quad (10)$$

$$g^2 = 1 - f f^*, \quad (11)$$

$$\Delta(r, \omega) = 2 \pi T N(0) \sum_{\alpha=0}^\infty \left\langle V(v', v) J(f(v', r, \omega)) \right\rangle_{v'}, \quad (12)$$

$$j = -4 \pi |e| N(0) T \sum_{\omega>0} \left\langle |e| \right\rangle_{\omega} \left\langle g \right\rangle_{\omega} \quad (13)$$

Here, $v$ is the Fermi velocity, $\Pi = \nabla + 2 \pi i A / \phi_0$, $\phi_0$ is the flux quantum. $\Delta(r)$ is the gap function (the order parameter) which may depend on the position $k_F$ at the Fermi surface (or on $v$). Eilenberger functions $f(r, v, \omega)$, $f^*$ and $g$ originate from Gor’kov’s functions integrated over the energy variable near the Fermi surface to exclude fast spatial oscillations on the scale $1/k_F$; instead $f$ and $g$ vary on the scale of the coherence length $\xi$ relevant for superconductivity. Functions $f$ and $f^*$ describe the superconducting condensate, whereas $g$ represents normal excitations. $N(0)$ is the total density of states at the Fermi level per spin. The Matsubara frequencies are defined by $\omega = \pi T (2n+1)$ with an integer $n$, and $o_D$ being the Debye frequency (for phonon-mediated superconductivity or a relevant energy scale for other mechanisms). The averages over the Fermi surface weighted with the local density of states $\propto 1/|v|$ are defined as

$$\langle X \rangle = \int \frac{d^2 k_F}{(2\pi)^2} h N(0) |v| X. \quad (14)$$

The order parameter $\Delta$ is related to $f$ in the self-consistency equation (12). Often, the effective coupling $V$ is assumed to be factorizable [57], $V(v, v') = V_0 \Omega(v) \Omega(v')$; this assumption is not always justifiable but it makes the algebra much simpler. One looks for the order parameter in the form $\Delta(r, T; \omega) = \Psi(r, T) \Omega(\omega)$. Then, the self-consistency equation (12) takes the form

$$\Psi(r, T) = 2 \pi T N(0) V_0 \sum_{\omega>0} \left\langle \Omega(v) J(f(v, r, \omega)) \right\rangle_{v}. \quad (15)$$

The function $\Omega(v)$ (or $\Omega(k_F)$), which describes the variation of $\Delta$ along the Fermi surface, is conveniently normalized [58]:

$$\langle \Omega^2 \rangle = 1. \quad (16)$$
In the absence of currents and fields $\Pi = 0$, and the Eilenberger equations give, for the uniform ground state,

$$f_0 = f'_0 = \frac{\Delta_0}{\beta}, \quad g_0 = \frac{\hbar \omega}{\beta}, \quad \beta^2 = \Delta_0^2 + \hbar^2 \omega^2. \tag{17}$$

Note that in general, both $\Delta_0 = \Omega_0(k_F)$ and $\beta$ depend on the position $k_F$ at the Fermi surface.

Instead of dealing with the effective microscopic electron–electron interaction $V$, one can use, in this formal scheme, the measurable critical temperature $T_c$, utilizing the identity

$$\frac{1}{N(0)V_0} = \ln \frac{T_c}{T} + 2\pi T \sum_{\omega > 0} \frac{1}{h\omega}, \tag{18}$$

which is equivalent to the famous relation $\Delta(0) = \pi T_c e^{-\gamma} = 2\hbar \omega_D \exp(-1/N(0)V_0)$; $\gamma = 0.577$ is the Euler constant. Substituting equation (18) into equation (15) and replacing $\omega_D$ with infinity due to fast convergence we obtain

$$\frac{\Psi}{2\pi T} \ln \frac{T}{T_c} = \sum_{\omega > 0} \left( \frac{\Psi}{h\omega} - \left( \Omega f \right) \right). \tag{19}$$

Now equation for $\Psi(T)$ reads

$$\frac{1}{2\pi T} \ln \frac{T_c}{T} = \sum_{\omega > 0} \left( \frac{1}{h\omega} - \left( \Omega^2 \right) / \left( \sqrt{\omega^2 + \hbar^2 \omega^2} \right) \right). \tag{20}$$

3.1. London penetration depth

Within microscopic theory, the penetration of weak magnetic fields into superconductors is treated perturbatively. Weak supercurrents and fields leave the order parameter modulus unchanged, but cause the condensate, i.e. $\Delta$ and the amplitudes $f$, to acquire an overall phase $\theta(r)$. Using the method of perturbations, one obtains corrections to $f_0, g_0$ among which we need only

$$g_1 = i\hbar \frac{\Delta_0^2}{2\beta^3} \vec{v} \cdot (\vec{\nabla} \theta + 2\pi \vec{A} / \phi_0). \tag{21}$$

Substituting this in the general expression (13) for the current density, one obtains the London relation between the current and the ‘gauge invariant vector potential’ $a = \phi_0 \nabla \theta / 2\pi + \vec{A}$:

$$4\pi J_i / c = - \langle (\lambda^2)^{-1} \rangle a_k. \tag{22}$$

Then, the general expression (13) for the current gives the inverse tensor of the squared penetration depth:

$$\langle (\lambda^2)^{-1} \rangle_k = \frac{16\pi^2 e^2 T}{c^2} N(0) \sum_{\omega} \left( \frac{\Delta_0 v_i v_k}{\beta^3} \right). \tag{23}$$

This result holds at any temperature for clean materials with arbitrary Fermi surface and order parameter anisotropies [54]. The temperature dependence of $\Delta_0 = \Omega_0$ and $\beta$ should be obtained by solving equation (20).

Thus, the general scheme for evaluation of $\lambda(T)$ in clean superconductors consists of two major steps: first evaluate the order parameter $\Delta_0(T)$ in uniform zero-field state for a given gap anisotropy $\Omega(v)$, then use equation (23) with a proper averaging over the Fermi surface. The sum over Matsubara frequencies is rapidly convergent and is easily done numerically (except in a few limiting situations for which analytic evaluation is possible).

3.1.1. Isotropic $\Delta$ on a general Fermi surface. For the majority of materials with electron–phonon interaction responsible for superconductivity, the relevant phonons have frequencies on the order of $\omega_D$. One can see that in exchanging such phonons, the electron momentum transfer is of the order of $\hbar k_F$; this leads to considerable smoothing of $\Delta(k_F)$ [59]. Indeed, in such materials $\Delta(k_F)$ is nearly constant along the Fermi surface, the strong possible anisotropy of the latter notwithstanding. In this case, commonly called the ‘$s$-wave’, $\Delta$ is taken as a constant at the Fermi surface:

$$\langle (\lambda^2)^{-1} \rangle_k = \frac{8\pi e^2 N(0) v_i v_k}{c^2} 2\pi T \Delta^2 \sum_{\omega} \frac{1}{\beta^3}. \tag{24}$$

We obtain in the Ginzburg–Landau domain,

$$\langle (\lambda^2)^{-1} \rangle_k (T \rightarrow T_c) = 2\langle (\lambda^2)^{-1} \rangle (0)(1 - T/T_c), \quad t = T/T_c. \tag{25}$$

At low temperatures,

$$\frac{\Delta(T)}{T_c} = \frac{\Delta(0)}{T_c} - \sqrt{\frac{2\pi T \Delta(0)}{T_c}} e^{-\Delta(0)/T} \approx 1.764 - 3.329 \sqrt{\pi} e^{-1.764/4}. \tag{26}$$

The low-temperature behavior of the penetration depth is given by

$$\langle (\lambda^2)^{-1} \rangle_k (\lambda^2)^{-1} (0) \left( 1 - 2\sqrt{\frac{\pi \Delta(0)}{T}} e^{-\Delta(0)/T} \right). \tag{27}$$

3.1.2. Two-dimensional $d$-wave. As an example of anisotropic $\Delta$, let us take a relatively simple but important case of a $d$-wave order parameter on the two-dimensional cylindrical Fermi surface: $\Omega = \Omega_0 \cos 2\varphi$, where $\varphi$ is the properly chosen azimuth angle on the Fermi cylinder. With this choice, the gap nodes are at $\varphi = \pm \pi/4, \pm 3\pi/4$. The normalization equation (16) gives $\Omega_0 = \sqrt{2}$, so that $\Delta_0 = \sqrt{2} \cos 2\varphi$. The order parameter at $T = 0$ is now given by

$$\frac{\Delta_{\text{max}}(0)}{T_c} = \frac{2\pi}{e^{\varphi + 0.5}} \approx 2.139 \tag{28}$$

and at $T \ll T_c$,

$$\frac{\Delta_{\text{max}}(T)}{T_c} \approx 2.139 - 0.927 t^3. \tag{29}$$

After averaging over the Fermi cylinder we obtain, for the in-plane penetration depth,

$$\lambda^{-2}(T) = \lambda^{-2}(0) \left( 1 - \sqrt{\frac{T}{\Delta_{\text{m}}}} \right), \tag{30}$$

$$\lambda^{-2}(0) = \frac{4\pi e^2 N(0) v^2}{c^2}. \tag{31}$$
3.2. Eilenberger two-gap scheme: the γ-model

The full-blown microscopic approach based on the Eliashberg theory is quite involved and not easy for data analysis [60–63]. Hence, there arose a need for a relatively simple but justifiable, self-consistent and effective scheme experimentalists could employ. The weak-coupling model is such a scheme. Over the years, the weak-coupling theory has been proven to describe well a multitude of superconducting phenomena. Similar to the weak coupling is the ‘renormalized BCS’ (renormalized Bardeen–Cooper–Schrieffer) model [63] that incorporates the Eliashberg corrections in the effective coupling constants. We call our approach a ‘weak-coupling two-band scheme’ and refer the reader to original papers where it is clarified that the applicability of the model for the analysis of the superfluid density and specific heat data is broader than the traditional weak coupling [54, 56].

The Eilenberger approach can be used to describe a two-gap situation self-consistently, in which

\[ \Delta(k) = \Delta_{1,2}, \quad k \in F_{1,2}, \]  

where \( F_1 \) and \( F_2 \) are two separate sheets of the Fermi surface. Denoting the densities of states on the two parts as \( N_{1,2} \), we have for a quantity \( X \) constant at each Fermi sheet

\[ (X) = (X_1 N_1 + X_2 N_2)/N(0) = n_1 X_1 + n_2 X_2, \]  

where \( n_{1,2} = N_{1,2}/N(0) \); clearly, \( n_1 + n_2 = 1 \).

The self-consistency equation (12) now takes the form

\[ \Delta_v = 2\pi T \sum_{\mu=1,2} n_\mu \lambda_{v\mu} f_\mu = \sum_{\mu} n_\mu \lambda_{v\mu} \Delta_\mu \sum_\omega \frac{2\pi T}{\beta_\mu} \]  

where \( v = 1, 2 \) is the band index and \( \lambda_{v\mu} = N(0) V(v, \mu) \) are dimensionless effective interaction constants. Note that the notation commonly used in the literature for \( \lambda_{v\mu} \) differs from ours: \( \lambda_{v\mu}^{(a)} = n_\mu \lambda_{v\mu} \).

Turning to the evaluation of \( \Delta_v(T) \), we note that the sum over \( \omega \) in equation (34) is logarithmically divergent. To deal with this difficulty, we employ Eilenberger’s idea of replacing \( \hbar \omega_0 \) with the measurable \( T_c \). Introducing dimensionless quantities

\[ \delta_v = \frac{\Delta_v}{2\pi T} = \frac{\Delta_v}{T_c} \frac{1}{2\pi t}, \]  

with \( t = T/T_c \), we obtain

\[ \delta_v = \sum_{\mu=1,2} n_\mu \lambda_{v\mu} \delta_\mu \left( \frac{1}{\lambda} + \ln \frac{T_c}{T} - A_\mu \right), \]  

\[ A_\mu = \sum_{n=0}^{\infty} \left( \frac{1}{n + 1/2} - \frac{1}{\sqrt{\delta_\mu + (n + 1/2)^2}} \right), \]  

where \( \lambda \) is defined as

\[ 1.76 T_c = 2\hbar \omega_0 \exp(-1/\lambda), \]  

or

\[ \frac{1}{\lambda} = \ln \frac{T}{T_c} + \sum_\omega \frac{2\pi T}{\hbar \omega_0}. \]

In terms of \( \lambda_{v\mu} \), \( \lambda \) is expressed as

\[ \lambda = \frac{2n_1 n_2 \lambda_{11} \lambda_{22} - \lambda_{12}^2}{n_1 \lambda_{11} + n_2 \lambda_{22} - \sqrt{(n_1 \lambda_{11} - n_2 \lambda_{22})^2 - 4n_1 n_2 \lambda_{12}^2}}. \]  

For the given coupling constants \( \lambda_{v\mu} \) and densities of states \( n_v \), the system, (36) and (37), can be solved numerically for \( \delta_v \) and therefore provides the gaps \( \Delta_v = 2\pi T \delta_v(t) \). Example calculations are shown in figure 1. The first graph in the top row is calculated assuming no interband coupling. Naturally, we obtain material with two different transition temperatures. The second graph shows the gaps with \( \lambda_{12} = 0.05 \), which features a single \( T_c \) and quite non-single-BCS-gap temperature dependence of the smaller gap. A single BCS gap is shown for comparison.

Superfluid density. Having formulated the way to evaluate \( \Delta(T) \), we turn to the London penetration depth given for general anisotropies of the Fermi surface and of \( \Delta \) by equation (23) [54]. We consider here only the case of currents in the \( ab \)-plane of uniaxial or cubic materials with two separate Fermi surface sheets, for which the superfluid density, \( \rho = \lambda_{ab}^2/(\lambda_{ab}^2 + \lambda_{12}^2) \) (T), is [56]

\[ \rho = \gamma \rho_1 \rightleftharpoons (1 - \gamma) \rho_2, \]

\[ \rho_v = \delta_v \sum_{n=0}^{\infty} \left[ \delta_v + (n + 1/2)^2 \right]^{-3/2}, \]

\[ \gamma = \frac{n_1 v_1^2}{n_1 v_1^2 + n_2 v_2^2}, \]  

where \( v_v \) are the averages of the in-plane Fermi velocities over the corresponding band.

With the discovery of two-gap superconductivity in a number of materials, including MgB\(_2\) [64, 65], NbSe\(_2\) [52], V\(_3\)Si [66], Lu\(_2\)Fe\(_2\)Si\(_2\) [67] and ZrB\(_2\) [68] one of the most popular approaches to analyze the experimental results has been the so-called ‘\( \alpha \)-model’ [64]. Developed originally to renormalize a single weak-coupling BCS gap to account for strong-coupling corrections [69], it was used to introduce two gaps, \( \Delta_{1,2} \), each having a BCS temperature dependence, but different amplitudes [64]. This allowed for a simple way to fit the data on the specific heat [64] and the superfluid density, \( \rho = \rho_1 + (1 - \rho_2) \rho_2 \) [20, 65]. Here, \( \rho_1 \) is evaluated with \( \Delta_{1,2} \), taking into account the relative band contributions. The fitting is usually quite good (thanks to a smooth and relatively ‘featureless’ \( \rho(T) \)) and the parameters \( \alpha \) were found to be one larger and one smaller than unity (unless they are both equal to 1 in the single-gap limit). Although the \( \alpha \)-model has played an important and timely role in providing convincing evidence for two-gap superconductivity, it is intrinsically inconsistent as applied to the full temperature range. The problem is that one cannot assume \( \alpha \)-model temperature dependences for the gaps in the presence of however weak interband coupling (required to have single \( T_c \)). In an unlikely situation of zero interband coupling, two gaps would have single-gap BCS-like \( T \)-dependences, but would have two different transition temperatures. The formal similarity in terms of additive partial superfluid densities
prompted us to name our scheme the ‘γ-model’. We note, however, that these models are quite different: our γ that determines partial contributions from each band is not just a partial density of states \( n_1 \) of the α-model, instead it involves the band’s Fermi velocities. The gaps, \( \Delta_{1,2}(T) \), are calculated self-consistently during the fitting procedure.

The γ-models can be simplified for a compensated metal, such as a clean stoichiometric superconductor LiFeAs [41]. To reduce the number of fitting parameters, yet capturing the compensated multiband structure, we consider the simplest model of two cylindrical bands with the mass ratio, \( \mu = m_1/m_2 \), whence the partial density of states of the first band, \( n_1 = \mu/(1 + \mu) \). The total superfluid density is \( \rho_s = \gamma \rho_1 + (1 - \gamma) \rho_2 \) with \( \gamma = 1/(1 + \mu) \). We also use the Debye temperature of 240 K [70] to calculate \( T_c \), which allows the fixing of one of the in-band pairing potentials, \( \lambda_{11} \). This leaves three free fit parameters: the second in-band potential, \( \lambda_{22} \), interband coupling, \( \lambda_{12} \), and the mass ratio, \( \mu \).

Indeed, we found that \( \rho_s(T) \) can be well described in the entire temperature range by this clean-limit weak-coupling BCS model [41]. Figure 2 shows the fit of the experimental superfluid density to the γ-model. The insets show temperature-dependent superconducting gaps obtained as a solution of the self-consistency equation, equation (34), and the lower inset shows the gap ratio as a function of temperature. Evidently, the smaller gap does not exhibit a

Figure 1. Top row: self-consistent gaps calculated for the indicated \( \lambda_{\mu \nu} \) in a compensated metal with \( \mu = 1 \). The first graph is for no interband pairing, \( \lambda_{12} = 0 \). The second for \( \lambda_{12} = 0.05 \). Bottom row: corresponding partial and total superfluid densities.

Figure 2. Symbols represent superfluid density, \( \rho_s(T) = (\lambda(0)/\lambda(T))^2 \), in a single crystal of LiFeAs calculated from the measured \( \lambda(T) \) with \( \lambda(0) = 200 \) nm. Solid lines represent the fit to a two-gap γ-model, \( \rho_s = \gamma \rho_1 + (1 - \gamma) \rho_2 \). The dashed line is a single-gap BCS solution. Upper inset: superconducting gaps, \( \Delta_1(T) \) and \( \Delta_2(T) \) calculated self-consistently during the fitting. Lower inset: \( \Delta_1/\Delta_2 \) as a function of temperature.
Figure 3. Gap ratio, $\gamma_\Delta = \Lambda_1(0)/\Lambda_2(0)$, for indicated $\mu$ (to check the effect of different partial densities of states) and $\lambda_{12}$. The magnitude of $\gamma_\Delta$ is given by the intensity shown on the right. Red lines in each graph show $\gamma_\Delta = 2$, which corresponds to the experimental situation in the pnictides.

BCS temperature dependence emphasizing the failure of the commonly used $\alpha$-model.

Lastly, we note that in order to have two distinct gaps, as observed in many experiments [4], one has to have significant in-band coupling constants, $\lambda_{11}$ and $\lambda_{12}$. To support this conclusion we calculate the gap ratio $\gamma_\Delta$ for fixed $\lambda_{12}$ and varying $\lambda_{11}$ and $\lambda_{22}$ (see figure 3). Red lines in each graph show $\gamma_\Delta = 2$. For all cases, one needs significant $\lambda_{12}$ and varying $\lambda_{11}$ to reach this gap ratio. Therefore, the original simplified $s^{\pm}$ model, with two identical Fermi surfaces and only interband coupling, $\lambda_{12} \neq 0$ and $\lambda_{11} = \lambda_{22} = 0$, does not describe the experimentally found two distinct gaps in Fe-based superconductors [71].

### 4. Effects of scattering

Scattering by impurities strongly affects the London penetration depth even in the simplest case of non-magnetic impurities in materials with an isotropic gap parameter and for scattering processes which can be characterized by the scalar (isotropic) scattering rate. The $\Delta$ anisotropy and magnetic impurities complicate the analysis of $\lambda(T)$, among other reasons due to the $T_c$ suppression in these cases. For an extended treatment the reader is referred to [40, 55] and here we only summarize properties related to the discussion of our results. Introducing the non-magnetic scattering rate, $1/\tau$, and magnetic scattering rate (spin-flip), $1/\tau_m$, the London penetration depth is expressed as [56]

$$\lambda_{ik}^{-1} = \frac{16\pi^2e^2TN(0)\langle v_i v_k \rangle}{c^2} \sum_{\omega} \frac{f_0^2}{\Delta/\hbar \pm h/2\pi}. \quad (42)$$

Here $1/\tau^\pm = 1/\tau \pm 1/\tau_m$. Clearly, the evaluation of $\lambda(T)$ in the presence of magnetic impurities is quite involved. There is one limit, however, for which we have a simple analytical answer.

**Gapless limit.** This is the case when $\tau_m$ is close to $2\hbar/\Delta_0(0)$, the critical value for which $T_c = 0$, i.e. $\tau_m \Delta \ll 1$. The resulting expression for the order parameter is remarkably simple [72]:

$$\Delta^2 = 2\pi^2(T^2 - T_c^2) \quad (43)$$

The result for superfluid density is valid in the entire temperature domain, $T < T_c$ [55],

$$\lambda_{ik}^{-1} = \frac{8\pi^2e^2N(0)\langle v_i v_k \rangle}{c^2(\rho^-)^2} \left( \ln \frac{2\rho_m}{\rho^+} + \frac{\rho^-}{2\rho_m} \right) (1 - t^2), \quad (44)$$

where $\rho = \hbar/(2\pi T_c \tau)$, $\rho_m = \hbar/(2\pi T_c \tau_m)$ and $\rho^\pm = \rho \pm \rho_m$. For a short transport mean-free path $\rho \gg \rho_m$ we have Abrikosov–Gor’kov’s result:

$$\lambda_{ik}^{-1} = \frac{8\pi^2e^2N(0)\langle v_i v_k \rangle}{c^2\rho \rho_m} (1 - t^2). \quad (45)$$
5. Experimental results

Ba(Fe\textsubscript{1−x}T\textsubscript{x})\textsubscript{2}As\textsubscript{2} is one of the most studied systems among all Fe-based superconductors and we have collected extensive data that illustrate general features often common to many other members of the diverse pnictide family. Here we focus on the electron-doped Ba(Fe\textsubscript{1−x}T\textsubscript{x})\textsubscript{2}As\textsubscript{2} with \(T = \text{Co and Ni}\). One reason for choosing this series is the fact that large and high-quality single crystals are available [11]. All samples were grown from the self-flux and were extensively characterized by transport, structural, thermal and magneto-optical analyses. They all exhibited uniform superconductivity at least on the 1 µm scale and dozens of samples were screened before entering into the resulting discussion [11, 75]. To demonstrate the sample quality we show magneto-optical images in figures 4 and 5. Details of this visualization technique are described elsewhere [76]. In the images, the intensity is proportional to the local magnetic induction. All samples show excellent Meissner screening [76]. Figure 4 shows the penetration of the magnetic field into the optimally doped sample with \(x = 0.07\) at 20 K. The distinct 'Bean oblique wedge' shape [77] of the penetrating flux with a current turn angle of 45° (implying isotropic in-plane current density) is observed.

Furthermore, to look for possible mesoscopic faults and inhomogeneities, we show the trapped magnetic field obtained after cooling in a 1.5 kOe magnetic field and turning the field off. The vortex distribution is quite homogeneous indicating robust uniform superconductivity for various doping levels. This is shown in figure 4 for four different doping levels.

The parent compound, BaFe\textsubscript{2}As\textsubscript{2}, is a poor metal [78] having a high-temperature tetragonal phase with no long-range magnetic order and undergoes structural and magnetic transitions around 140 K into a low-temperature orthorhombic phase with long-range antiferromagnetic spin density wave (SDW) order [11]. Transition metal doping onto the iron site serves to suppress the structural and magnetic transition temperatures and superconductivity emerges after magnetism has sufficiently been weakened. Doping into the barium site with potassium results in hole-doped superconductivity. The properties of this hole-doped system, at least as far as penetration depth is concerned, are quite similar to the electron-doped FeT122 [32, 79]. On the other hand, the properties of materials obtained by isovalent doping of phosphorus into the arsenic site seem to induce superconductivity without introducing significant scattering and seem to result in a superconducting gap with line nodes [80].

Figure 4. Magnetic flux penetration at 20 K into a Ba(Fe\textsubscript{0.95}Co\textsubscript{0.05})\textsubscript{2}As\textsubscript{2} crystal. The last frame shows a schematic overlay of the expected 'Bean oblique wedge' shape with isotropic in-plane current density.
also discuss the case of a substantial variation of \(\lambda(T)\) between various samples, probably due to the edge effect.

In the following analysis we use two ways to represent the power-law behavior: \(\lambda(T) = \lambda(0) + A(T/T_c)^2\) at low temperatures (below 0.3 \(T_c\)) with \(A\) being the only free parameter, because at a gross level, all samples follow the \(\lambda(T) \sim T^2\) behavior rather well and \(\lambda(T) = \lambda(0) + CT^n\), leaving the exponent \(n\) as a free parameter to analyze its evolution with doping or artificially introduced defects. In the case of vertical line nodes, we expect a variation from \(n = 1\) to \(n = 2\) upon increase of pair-breaking scattering [73], but in the case of a fully gapped \(s_{\pm}\) state we expect an opposite trend to approach \(n = 2\) in the dirty limit from clean-limit exponential behavior [92–95]. If, however, nodes are formed predominantly along the \(c\)-axis in the extended \(s\)-wave scenario, the effect of scattering on the in-plane penetration depth would be the opposite—starting from roughly \(n = 2\) in the clean limit and approaching exponential in the dirty limit [96].

5.1. In-plane London penetration depth

Figure 6 shows the normalized differential TDR magnetic susceptibility of several single crystals of \(\text{Ba(Fe}_{1-x}\text{Co})_2\text{As}_2\) across the superconducting ‘dome’. All but one sample were grown under the same conditions and with similar starting chemicals. All but one had thicknesses in the range 100–400 nm. One of the samples (\(x = 0.074\), denoted as batch 2) was cleaved for the irradiation experiments and had a thickness of 20 nm. We use it for comparison with the ‘thick’ batch 1 and, also, to study the effects of deliberately induced defects. It turns out that the edges of the thicker samples are not quite smooth and, when imaged by a scanning electron microscope (SEM), look like a used book (see figure 11(a)). Since calibration of the TDR technique relies on the volume penetrated by the magnetic field, the thinner samples should be closer to the idealization of the sample geometry (top and bottom surfaces are always very flat and mirror-like), thus producing a more reliable calibration. On the other hand, this would only lead to a change in the amplitude (due to geometric mis-calibration) of the penetration depth variation (i.e. pre-factor \(A\)) and would not change its functional temperature dependence (i.e. the exponent \(n\)). Thinner samples, on the other hand, have a better chance to be more chemically uniform, thus have reduced scattering. We observed these effects comparing samples from different batches.

The low-temperature variation of the London penetration depth is presented in figure 7 as a function of \((T/T_c)^n\) obtained by fitting the data to \(\Delta\lambda(T) = \lambda(0) + C(T/T_c)^n\). Each curve reveals a robust power-law behavior with the exponent \(n\) shown in the inset in figure 7. The fitted exponent \(n\) varies from \(n = 2 \pm 0.1\) for underdoped samples to \(n = 2.5 \pm 0.1\) for the overdoped samples within batch 1 and reaches \(n = 2.83\) in batch 2. If the superconducting density itself follows a power law with a given \(n\), then \(C \sim f_2(c/\omega_p)S\), where \(f_2\) 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 multi-gap character of the superconductivity, possible nodal structure, unitary impurity scattering strength, etc) and \(\omega_p\) is the plasma frequency.

Clearly the sample of batch 2 shows behavior much closer to exponential compared with that of batch 1. As discussed above, this could be due to the variation of scattering between the batches. We analyze low-temperature \(\lambda(T)\) in figure 8. We attempted to fit the data with three functions: the power law with free pre-factor \(C\) and exponent \(n\), the standard
Figure 7. Low-temperature behavior of $\Delta\lambda(T)$ versus $(T/T_c)^n$ for all studied concentrations. The inset shows exponent $n$ as a function of concentration. The sample from batch 2 is shown for comparison.

Figure 8. Low-temperature behavior of $\Delta\lambda(T)$ for the batch 2 sample. Solid lines are the fits to three functions described in the text. The inset shows full temperature variation indicating very sharp superconducting transition.

Figure 9. Low-temperature behavior of $\Delta\lambda(T)$ versus $(T/T_c)^{2.83}$ for the sample from batch 2. Marks show actual reduced temperature. The inset shows the behavior below the commonly accepted ‘low-temperature limit’ of $T_c/3$.

5.2 Absolute value of the penetration depth

To further investigate the effects of doping and the difference between the batches, we use the method described in section 2.1.2, which involves measuring the sample, coating it with a uniform layer of Al and re-measuring [39, 50]. The Al film was deposited onto each sample while it was suspended from a rotating stage by a fine wire in an argon atmosphere of a magnetron sputtering system. Film thickness was checked using a scanning electron microscope in two ways, both of which are as shown in figure 11. The first method involved breaking a coated sample after all measurements had been performed to expose its cross section. After this, it was mounted on an SEM sample holder using silver paste, as shown in figure 11(a). The images of the broken edge are shown for

single-gap BCS behavior, equation (27), with a fixed value of $\lambda(0) = 200$ nm and $\Delta(0)$ as a free parameter and to a BCS-like function where both $\lambda(0)$ and $\Delta(0)$ were free parameters. The resulting values are shown in figure 8. The power-law fit yields quite high exponent $n \approx 2.83$ and the best fit quality. The BCS-like fit yields a reasonable fit quality, but produces impossible values of both $\lambda(0) \approx 48$ nm and $\Delta(0) \approx 0.78 T_c$ (the latter cannot be less than a weak coupling BCS value of 1.76). Finally, the fixed $\lambda(0)$ BCS fit does not agree with the data and also produces unreasonable $\Delta(0) \approx 1.25 T_c$. One strong conclusion follows from this exercise—we are dealing with a multi-gap superconductor.

In order to understand the validity of the empirical power-law behavior, figure 9 shows the low-temperature behavior of $\Delta\lambda(T)$ versus $(T/T_c)^{2.83}$ for the sample from batch 2. Arrows show actual reduced temperature. The inset zooms to below the commonly accepted ‘low-temperature limit’ of $T_c/3$.

We now summarize the observed power-law behavior of the in-plane penetration depth for the electron-doped 122 family of superconductors. Figure 10 shows the experimental low-temperature limit power-law exponent for different dopants on the Fe site and in different doping regimes. The shaded areas show the expectations for the pair-breaking scattering effects in $s$- and $d$-wave scenario. It seems that statistically $d$-wave pairing (more generally vertical line nodes) cannot explain the in-plane variation of the penetration depth. However, if the nodes appear somewhere predominantly along the $c$-axis, they may induce an apparent power-law behavior of $\lambda_{ab}(T)$ with the effective $n \approx 2$ in the clean limit and going towards exponential behavior with an increase in the scattering rate [96].
Figure 10. Power-law exponent of the low-temperature variation of in-plane $\lambda_{ab}(T)$ is several electron-doped Ba122 superconductors at various doping levels. Shaded areas show the influence of pair-breaking scattering with $n = 2$ being the limiting value of $n$ approaching from either the $s$-wave side (nodeless exponential) or $d$-wave side (vertical line nodes). In the case of extended $s$-wave with nodes predominantly along the $c$-axis, effective $n$ increases from 2 in the clean limit towards the exponential behavior when the nodes are lifted by scattering.

Figure 11. Scanning electron microscope images of the Al-coated samples. (a) Large scale view. The broken side is on top. (b) and (c) are zoomed in on the Al film on the edge of the broken side. (d) A trench produced by a focused ion beam (FIB). (e) Close-up view of the FIB trench showing the Al film and its thickness.

two different zoom levels in figures 11(b) and (c). The second method used a focused ion beam (FIB) to make a trench on the surface of a coated sample, with the trench depth being much greater than the Al coating thickness, shown in figure 11(d). The sample was then tilted and imaged by the SEM that is built into the FIB system, as shown in figure 11(e).

Examples of the penetration depth measurements before and after coating are shown in figure 12. Notice how small is the effect of Al coating when presented on a large scale of a full superconducting transition of the coated sample. However, the TDR technique is well suited to resolve the variation due to the aluminum layer [39, 50].

Obtained values of $\lambda_{ab}(0)$ are summarized in the top panel of figure 13 for doping levels, $x$, across the superconducting region of the phase diagram shown schematically in the bottom panel of figure 13. The size of the error bars for the $\lambda_{ab}(0)$ points was determined by considering the film thickness to be $t = 100 \pm 10$ nm and $\lambda_{Al}(0) = 50 \pm 10$ nm. The scatter in the $\lambda_{ab}(0)$ values shown in the upper panel of figure 13 has an approximately constant value of $\pm 0.075 \mu$m for all values of $x$, which probably indicates that the source of the scatter is the same for all samples. For comparison, figure 13 also shows $\lambda_{ab}(0)$ obtained from $\mu$SR measurements (red stars) [22], the magnetic force microscopy (MFM) technique (open stars)
Given statistical uncertainty these measurements are consistent with our results within the scatter. It may also be important to note that the $\lambda_{ab}(0)$ values from other experiments are all on the higher side of the scatter that exists within the TDR $\lambda_{ab}(0)$ data set. As discussed above, our TDR techniques give a low bound of $\lambda(0)$, consistent with this observation.

In order to provide a more quantitative explanation for the observed increase in $\lambda_{ab}(0)$ as $x$ decreases in the underdoped region, we have considered the case of $s^\pm$ superconductivity coexisting with itinerant antiferromagnetism [98]. For the case of particle hole symmetry (nested bands), the zero temperature value of the in-plane penetration depth in the region where the two phases coexist is given by [39, 98]:

$$\lambda_{ab}^{SC+SDW}(0) = \lambda_{ab}(0) \sqrt{1 + \frac{\Delta_{AF}^2}{\Delta_0^2}},$$

where $\lambda_{ab}(0)$ is the value for a pure superconducting system with no magnetism present, and $\Delta_{AF}$ and $\Delta_0$ are the zero temperature values of the antiferromagnetic and superconducting gaps, respectively. Deviations from particle hole symmetry lead to a smaller increase in $\lambda_{ab}^{SC+SDW}(0)$, making the result in equation (46) an upper estimate [98].

The three blue dashed lines shown in the top panel of figure 13, which were produced using equation (46), show the expected increase in $\lambda_{ab}(0)$ in the region of coexisting phases below $x \approx 0.047$ by normalizing to three different values of $\lambda_{ab}(0)$ in the pure superconducting state, with those being 120, 180 and 270 nm to account for the quite large dispersion of the experimental values. This theory does not take into account changes in the pure superconducting state, so for $x > 0.047$ the dashed blue lines are horizontal. These theoretical curves were produced using parameters that agree with the phase diagram in the bottom panel of figure 13 [98, 99]. While the exact functional form was not provided from any physical motivation and merely serves as a guide to the eye, the solid gray line in figure 13 is a fit of the TDR $\lambda_{ab}(0)$ data to a function of the form $A + B/x^n$, which does indeed show a dramatic increase of $\lambda_{ab}(0)$ in the coexistence region and also a relatively slight change in the pure superconducting phase. It should be noted that a dramatic increase in $\lambda_{ab}(0)$ below $x \approx 0.047$ cannot be explained by the impurity scattering, which would only lead to relatively small corrections in $\lambda(T)$.

With the experimental values of $\lambda(0)$ we can now analyze the superfluid density. In general $\lambda^{-2}(T)$ is given by equation (23) and depends on the averaging over the particular Fermi surface. For example, in the simplest cylindrical case, equation (30) and $\lambda^{-2}(0) = 4\pi e^2 N(0) v^2 / c^2$, where $N(0)$ is the density of states at the Fermi level and $v$ is the Fermi velocity. However, it is instructive to analyze the behavior from a two-fluid London theory point of view looking at the density of superconducting electrons, $n_s = (mc^2/4\pi e^2)\lambda^{-2}$ as a function of temperature. The zero value, $n_s(0)$, will in general be less than the total density of electrons due to pair-breaking scattering, so the magnitude of $n_s$ is useful when comparing different samples.

Figure 14 shows the data for underdoped, optimally doped and overdoped samples from batch 1 and also a sample from batch 2 for comparison. For this sample 2 $\lambda(0) = 200$ nm is used. There is a clear, but expected, asymmetry with respect to doping. Underdoped samples show quite low density, because not all electrons participate in the formation of the

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**Figure 12.** Main panel: full superconducting transition of an optimally doped FeCo122 crystal from batch 1 before and after Al coating. Inset: zoomed-in low-temperature region, $T_{min} \approx T \lesssim T_{AI}$, before (green triangles) and after (brown circles) the Al coating on the same sample. (Notice how small the effect is at full scale in the main panel.)

**Figure 13.** Top panel: the zero temperature London penetration depth, $\lambda_{ab}(0)$, as a function of the Co concentration, $x$. The three dashed blue lines are theoretical curves obtained using equation (46) for three different values of $\lambda_{ab}(0)$ in the pure superconducting state. The solid gray line is a fit to the TDR data only of the form $A + B/x^n$. Also shown are values of $\lambda_{ab}(0)$ obtained by other experiments for comparison, explained in the text. Bottom panel: schematic phase diagram for FeTi122 system showing the coexisting region [98, 99].
Cooper pairs and parts of the Fermi surface are gapped by the SDW as was discussed above. The overdoped sample, $x = 0.01$, despite having smaller $T_c$ than the ones with $x = 0.074$, shows the highest $n_s$. Obviously, the data scatter is significant. Therefore, the only reliable conclusion is that penetration depth increases dramatically upon entering the coexisting region. The overdoped side has to be studied more to acquire enough statistics. Furthermore, comparing two samples with $x = 0.074$ from two different batches reveals an even more striking difference. Not only does the sample from batch 2 have larger $n$, but the temperature dependence of $\rho_s$ in the full temperature range is also quite different. The pronounced convex shape (positive curvature) of $\rho_s(T)$ observed in all samples from batch 1 at the elevated temperatures becomes much less visible in sample 2. The bottom panel of figure 14 clearly demonstrates this difference, which is hard to understand based purely on geometrical considerations (different thicknesses). It seems that thicker samples of batch 1 had a higher chance of being chemically inhomogeneous across the layers. On the other hand, the convex shape of $\rho_s(T)$ at elevated temperatures is a sign of two-gap superconductivity [56], which depends sensitively on the interaction matrix, $\lambda_{ij}$, see section 3.2 and equation (34). This feature becomes more pronounced when the interband coupling, $\lambda_{12}$, becomes smaller compared with the in-band coupling potentials, $\lambda_{11}$ and $\lambda_{22}$. If our interpretation that the difference between $x = 0.074$ samples from batch 1 and batch 2 is due to enhanced pair-breaking scattering in batch 1, this would indicate that this scattering is primarily of interband character, so it disrupts the interband pairing. Additional information regarding the role of scattering in determining the anomalous convex behavior was obtained from the microwave cavity perturbation measurements on FeSe$_{0.4}$Te$_{0.6}$ single crystals [36]. Both real and imaginary parts of complex conductivity allowed the determination of $\lambda(T)$ and temperature-dependent scattering time from which a crossover from dirty to clean limit upon cooling has been proposed. All these results provide indirect leverage to the $s_\pm$ scenario where interband coupling plays the major role.

The normalized superfluid density for sample 2 is analyzed in figure 15 using the two-band $\gamma$-model described in section 3.2. Symbols show the data and the solid lines represent partial and total $\rho(T)$ obtained from a fit to equation (41). Also shown are the clean (dashed gray lines) and dirty (dotted gray lines) single-gap $s$- and $d$-wave cases. The inset shows superconducting gaps obtained self-consistently during the fitting, see equation (37).
we do not know the absolute value of $\gamma_\lambda$ and the results are presented in figure 16. The problem is that the gaps fully determine the experimental $\rho_s$ and this is the main result. We find that $\Delta_1(0) = 1.883 T_c = 3.73$ meV and $\Delta_2(0) = 0.754 T_c = 1.49$ meV, which are in good agreement with specific heat [85,100] and $\mu$SR penetration depth measurements [91] done on the samples of similar composition. Also shown in figure 15 are the clean (dashed gray lines) and dirty (dotted gray lines) single-gap $\lambda_s$ and $\lambda_d$-wave cases. (Note that while the gap does not depend on the non-magnetic impurities in isotropic $s$-wave case (Anderson theorem), the superfluid density does.) Clearly, $\rho_s(T)$ for sample 2 (and, of course for the samples of batch 1) does not even come close to any of these single-gap scenarios.

### 5.3. Anisotropy of London penetration depths

Let us now discuss the electromagnetic anisotropy in the superconducting state, parametrized by the ratio $\gamma_\lambda = \lambda_c/\lambda_{ab}$. To determine $\lambda_c$, we used the method described in section 2.1.3 and the results are presented in figure 16. The problem is that we do not know the absolute value of $\lambda_c(0)$, so we could only obtain $\Delta \lambda_c(T)$ with the help of knowing $\Delta \lambda_{ab}(T)$, which was measured on the same crystal independently. To find the total $\lambda_c$ we use the fact that close to $T_c$ (in the region of validity of Ginzburg–Landau theory), we should have [54]

$$\gamma_\lambda(T_c) = \sqrt{\gamma_\rho(T_c)},$$  \hspace{1cm} (47)

where anisotropy of normal state resistivity, $\gamma_\rho = \rho_c/\rho_{ab}$, is taken right above $T_c$. With $\gamma_\rho \approx 4 \pm 1$ [101], so that $\gamma_\lambda(T_c) \approx 2$. The results are shown in the inset in figure 16. Of course, there is some ambiguity in determining the exact anisotropy value, but the qualitative behavior does not change—the anisotropy increases upon cooling. With our estimate it reaches a modest value of 5 at low temperatures, which makes pnictides very different from high-$T_c$ cuprates. This is in contrast to a two-gap superconductor MgB$_2$ where $\gamma_\lambda$ decreases upon cooling [65,102], which may be due to different dimensionality of the Fermi sheets. In any case, temperature-dependent $\gamma_\lambda$ can only arise in the case of a multi-gap superconductor.

Next we examine the anisotropy of $\lambda(T)$ at different doping levels. This study was performed on FeNi122 samples and is reported in detail elsewhere [42]. Figure 17(a) summarizes the $T(a)$ phase diagram showing structural/magnetic ($T_{sm}$) and superconducting ($T_c$) transitions. The inset shows TDR measurements in a full temperature range for all concentrations used in this study. Figure 17(b) shows the low-temperature ($T \leq 0.3T_c$) behavior of $\lambda_{ab}(T)$ for several Ni concentrations. The data plotted versus $(T/T_c)^2$ are linear for underdoped compositions and show a clear deviation towards a smaller power-law exponent (lower temperatures marked by arrows in figure 17(b)) for overdoped samples. While at moderate doping levels the results are fully consistent with our previous measurements in FeCo122 [86,87], the behavior in
the overdoped samples is clearly less quadratic. It should be noted that in order to suppress $T_c$ by the same amount, one needs a two times lower Ni concentration compared with Co. In the previous FeCo122 study [86], the samples never reached highly overdoped compositions equivalent to $x = 0.072$, $T_c = 7.5$ K. The inset shows similar data for the underdoped $x = 0.033$, $T_c = 15$ K.

Now we apply a technique described in section 2.1.3 to estimate $\lambda_c(T)$. Figure 18 shows the effective penetration depth, $\lambda_{\text{mix}}$ (see equation (8)), for overdoped, $x = 0.072$ (main panel), and underdoped, $x = 0.033$ (inset), samples before (A) and after (B) cutting in half along the longest side (l-side) as illustrated schematically at the top of the figure. Already in the raw data, it is apparent that the overdoped sample exhibits a much smaller exponent $n$ compared with the $\lambda_{ab}(T)$, while underdoped samples show a tendency to saturate below 0.13$T_c$. Using equation (8) we can now extract the true temperature-dependent $\Delta\lambda_c(T)$. The result is shown in figure 19 for two different overdoped samples of the same composition, $x = 0.072$ having $T_c = 7.5$ K and $T_c = 6.5$ K, and for an underdoped sample with $x = 0.033$ having $T_c = 15$ K. Since the thickness of the sample is smaller than its width, we estimate the resolution of this procedure for $\Delta\lambda_c$ to be about 10 nm, which is much lower than 0.2 nm for $\Delta\lambda_{ab}$. Nevertheless, the difference between the samples is obvious. The overdoped samples show a clear linear temperature variation up to $T_c/3$, strongly suggesting nodes in the superconducting gap. The average slope is large, about $d\lambda_c/dT \approx 300$ nm K$^{-1}$, indicating a significant amount of thermally excited quasiparticles. By contrast, in the underdoped sample the inter-plane penetration depth saturates indicating a fully gapped state. If fitted to the power-law the exponent in the underdoped sample $2 \leq n \leq 3$, depending on the fitting range.

Nodes, if present somewhere on the Fermi surface, affect the temperature dependence of both components of $\Delta\lambda(T)$. However, the major contribution still comes from the direction of the supercurrent flow, thus placing the nodes in the present case at or close to the poles of the Fermi surface. The nodal topologies that are consistent with our experimental results are latitudinal circular line nodes located at the finite $k_z$ wave vector or a point (or extended area) polar node with a nonlinear $(\Delta(\theta) \sim \theta^p, p > 1)$ variation of the superconducting gap with the polar angle, $\theta$. It is interesting to note a close similarity to the results of thermal conductivity measurements in overdoped FeCo122 that have reached the same conclusions—the in-plane state is anisotropic, but nodeless [45], whereas the out-of-plane response is nodal [46]. Still, we emphasize that the apparent power-law behavior of the in-plane penetration depth, $\lambda_{ab}(T)$, in a heavily overdoped samples could be induced by the out-of-plane nodes [96, 103]. To summarize, it appears that not only is the gap not universal across different pnictide families [80], it is not universal even within the same family over an extended doping range. A similar conclusion has been reached for the hole-doped BaK122 pnictides [104–106].

5.4. Pair-breaking

Although the natural variation in the scattering rates between samples provides a good hint towards the importance of pair-breaking scattering, for more quantitative conclusions we need to introduce an additional disorder. This can be achieved with the help of heavy-ion irradiation. To examine the effect of
irradiation, $\sim 2 \times 0.5 \times 0.02 - 0.05 \text{ mm}^3$ single crystals were selected and then cut into several pieces preserving the width and thickness. We compare sets of samples, where the samples in each set are parts of the same original large crystal and have identical temperature-dependent penetration depth in the unirradiated state. (These samples are what we call batch 2 in this review with the unirradiated reference piece appearing in the discussion and figures of the previous sections). Irradiation with 1.4 GeV $^{208}\text{Pb}^{56+}$ ions was performed at the Argonne Tandem Linear Accelerator System (ATLAS) with an ion flux of $\sim 5 \times 10^{11}$ ions s$^{-1}$ m$^{-2}$. The actual total dose was recorded in each run. Such irradiation usually produces columnar defects or the elongated pockets of disturbed superconductivity along the ion propagation direction. The density of defects, $d$, per unit area is usually expressed in terms of the so-called ‘matching field’, $B_\phi = \Phi_0 d$, which is obtained assuming one flux quanta, $\Phi_0 \approx 2.07 \times 10^{-7}$ G cm$^{-2}$ per ion track. Here we studied samples with $B_\phi = 0.5 \text{T}, 1.0 \text{T}$ and $2.0 \text{T}$ corresponding to $d = 2.4 \times 10^{10}$ cm$^{-2}, 4.8 \times 10^{10}$ cm$^{-2}$ and $9.7 \times 10^{10}$ cm$^{-2}$. The sample thickness was chosen in the range $\sim 20-50 \mu\text{m}$ to be smaller than the ion penetration depth, 60–70 $\mu\text{m}$. The same samples were studied by magneto-optical imaging. Strong Meissner screening and large uniform enhancement of pinning have shown that the irradiation has produced uniformly distributed defects [107].

Indeed, to see the effect, we need to start with the best (largest exponent) sample we have. We irradiated the sample designated as batch 2 with $n = 2.83$, which was discussed in detail above (see figures 8, 9 and 15). To analyze the power-law behavior and its variation with irradiation, we plot $\Delta \lambda$ versus $(t = T / T_c)$ in figure 20, where the $n_0$ values for FeCo122 and FeNi122 were chosen from the best power-law fits of the unirradiated samples (see figure 21). While the data for unirradiated samples appear as almost perfect straight lines showing robust power-law behavior, the curves for irradiated samples show downturns at low temperatures indicating smaller exponents. This observation, emphasized by the plots of the derivatives $d \Delta \lambda(t)/dt$ in the inset of figure 20, points to a significant change in the low-energy excitations with radiation. The variations of $T_c$ and $n$ upon irradiation are illustrated in figure 21. Dashed lines and circles show FeCo122, while solid lines and triangles show FeNi122. The upper panel shows the variation of $T_c$ and the width of the transition. Since $B_\phi$ is directly proportional to the area density of the ions, $d$, we can say that $T_c$ decreases roughly linearly with $d$. The same trend is evident for the exponent $n$ shown in the lower panel of figure 21.

The influence of impurities, assuming $s_\pm$ pairing, has been analyzed numerically in a T-matrix approximation [88]. Figure 22(a) shows calculated superfluid densities for different values of the scattering rate. Figure 22(b) shows corresponding densities of states. Finally, figure 22(c) shows the central result: the correlation between $T_c$ and $n$. Note that these two quantities are obtained independent of each other. Assuming that the unirradiated samples have some disorder due to doping, and scaling $T_c^{un}$ to lie on the theoretical curve, we find that the $T_c(B_\phi)$ of the irradiated samples also follows

**Figure 20.** Detailed comparison of the functional form of $\Delta \lambda(T)$ for irradiated FeCo122 and FeNi122. In the main panels $\Delta \lambda(T)$ is plotted versus $(t = T / T_c)^n$ with the exponents $n$ taken from the best fits of unirradiated samples: $n_0 = 2.8$ and 2.5 for FeCo122 and FeNi122, respectively. It is apparent that irradiation causes low-temperature deviations, which are better seen in the derivatives, $d \Delta \lambda(t)/dt$, plotted in the insets.

**Figure 21.** Top panel: the suppression of $T_c$ with disorder relative to unirradiated $T_c^{un}$. The vertical bars denote the width of the transition corresponding to the diamagnetic signal change from 90% (onset) to 20% (end). Symbols are shown at the mean values between the onset and end of the transition. Lower panel: exponent $n$ versus $B_\phi$. 
Figure 22. (a) Superfluid density and (b) the density of states, computed for the $s_\pm$ state with sign-changing isotropic gaps and strong interband impurity scattering, between the Born and unitary limits. The dashed line in (a) is an example of a power-law fit $\rho(T)/\rho_0 = \rho(0)/\rho_0 - a(T/T_{c0})^n$ for $0 < T < 0.4T_{c0}$; (b) as the impurity concentration $n_{\text{imp}} \sim \Gamma$ increases, the band of mid-gap states approaches the Fermi level and the exponent $n$ is reduced; (c) $T_c$ versus power $n$, from the theoretical model (triangles) and experiment (squares and circles).

this curve. The assumption of similarity between doping and radiation-induced disorder, implied in this comparison, while not unreasonable, deserves further scrutiny. A more recent discussion on the variation of $T_c$ with disorder is found in [108].

6. Conclusions

It was not possible to include many interesting results obtained for various members of the diverse family of iron-based superconductors in this review. However, we may provide some general conclusions based on our work as well as on the results by others.

(a) The superconducting gap in optimally doped pnictides is isotropic and nodeless.

(b) Pair-breaking scattering changes the clean-limit low-temperature asymptotics (exponential for nodeless and $T$-linear for line nodes) to a $\sim T^2$ behavior. Therefore, additional measurements (such as deliberately introduced disorder) are needed to make conclusions about the order parameter symmetry. In anisotropic superconductors in general and in $s_\pm$, in particular, even the non-magnetic impurities are pair breaking.

(c) The materials can be described within a self-consistent two-band $\gamma$-model with two gaps with a ratio of magnitudes of about $\Delta_1(0)/\Delta_2(0) \approx 2-3$.

(d) Upon doping, the power-law exponent $n$ for the in-plane penetration depth, $\lambda_{ab}(T)$, decreases reaching values below 2 signaling the development of significant anisotropy, whereas out-of-plane $\lambda_c(T)$ shows a linear-$T$ behavior signaling line nodes with Fermi velocity predominantly in the $c$-direction.

(e) There is a fairly large region of coexisting superconductivity and long-range magnetic order, albeit with suppressed superfluid density.

(f) Overall, the observed behavior is consistent with the $s_\pm$ pairing, but realistic three-dimensional calculations are required to achieve agreement with experiments.

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