London penetration depth and strong pair-breaking in iron-based superconductors

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The low temperature variation of the London penetration depth for a number of iron-pnictide and iron-chalcogenide superconductors is nearly quadratic, \( \Delta \lambda(T) = \beta T^n \) with \( n \approx 2 \). The coefficient in this dependence shows a robust scaling, \( \beta \propto 1/T_c^2 \) across different families of these materials. We associate the scaling with a strong pair-breaking. The same mechanism has recently been suggested to explain the scalings of the specific heat jump, \( \Delta C \propto T_c \) [1], and of the slopes of the upper critical field, \( dH_c2/dT \propto T_c \) in these materials [2]. This suggests that thermodynamic and electromagnetic properties of the iron-based superconductors can be described within a strong pair-breaking scenario.

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Due to the unique electronic structure and, most likely, unconventional pairing mechanism, iron-based superconductors exhibit a number of uncommon properties. It has recently been reported [1] that across the whole family of iron-pnictides the specific heat jump, \( \Delta\varphi \), had been suggested [4, 7, 8, 13]. The symmetry of the structure [15, 16]. The Ferman surface (FS) average of the power-law behavior of \( \Delta \varphi \) 

\[
\Delta \varphi \propto T^2/T_c^3 ,
\]

(1)

Despite some initial disagreements in experimental reports, most precision measurements of the in-plane London penetration depth of iron-based superconductors had found the power-law behavior, \( \Delta \varphi(T) \propto T^n \) with \( n \approx 2 \) [3, 8]; for some compounds \( n \approx 1 \) is claimed [10–12]. Commonly, a non-exponential behavior is taken as evidence of unconventional order parameter, possibly having a nodal gap structure [3, 7, 8, 13]. However, such a direct correspondence between the nodes and the exponent \( n \) should exist only in clean materials. As a rule, scattering breaks this elegant connection. E.g., for d-wave superconductors, the linear low T dependence of \( \lambda \) in the clean case changes to \( T^2 \) in the presence of moderate scattering [14]. In fact, connection between the power-law behavior of \( \Delta \varphi(T) \) and scattering in pnictides had been suggested [4, 7, 8, 13]. The symmetry of the order parameter \( \Delta \) in multi-band iron-pnictides is not yet determined with certainty, however, many favor the \( s_\pm \) structure [15, 16]. The Fermi surface (FS) average of the order parameter in this model \( \langle \Delta \rangle \ll \Delta_{max} \). We then expect the penetration depth to behave like a “dirty” d-wave, i.e., to show the low-temperature variation \( \propto T^2 \).

The samples were plate-like single crystals with typical dimensions 1 x 1 x 0.2 mm\(^3\). Details of sample synthesis and characterization can be found elsewhere [17, 19]. The penetration depth measurements were performed with a self-resonating tunnel diode oscillator. Diamagnetic response of the sample causes shift of the resonant frequency, \( \Delta f = -G\chi(T) \), where \( \chi(T) \) is magnetic susceptibility determined by \( \lambda(T) \) in the Meissner state, \( -4\pi\chi(T) = [1 - (\lambda/R) \tan(h(\lambda/R))] \). The calibration constant \( G = f_0V_c/2V_c(1 - N) \) is measured directly by extracting the sample from the coil at the lowest temperature. Here \( f_0 \approx 14 \text{ MHz} \) is the empty resonator frequency, \( V_c \) and \( V_e \) are the sample and coil volumes, and \( N \) is the demagnetization factor. Details of measurements and of data analysis are described elsewhere [20, 21].

Figure 1(a) shows the linear behavior of \( \Delta \varphi \) versus \( (T/T_c)^2 \) for \( T < T_c/3 \) in iron-based compounds with \( T_c \) varying from \( \approx 12 \) to 23 K; the data are from Refs. 3–7 and 22. The exponent \( n \) in \( \Delta \varphi \propto T^n \) extracted by fitting the low temperature data is shown for six compounds in Fig 1(b). We see that \( \Delta \varphi(T) \propto T^2 \) holds for the (AE)/(Fe\(_{1-x}\)TM\(_x\))\(_2\)As\(_2\) (“122”), (RE)FeAs(O\(_{1-x}\)F\(_x\)) (“1111”), and FeTe\(_{1-x}\)Se\(_x\) (“11”) families; here AE stands for an alkali earth element, TM for a transition metal, RE for a rare earth. Thus, the four lines shown in Fig 1(a) are not merely for different doping levels of the same compound, but rather they belong to four different families of the iron-based materials. This universal behavior has prompted us to look for a universal cause; we offer below a strong pair-breaking as such a cause.

The theoretical tool we employ, the quasiclassical version of the weak-coupling Gor’kov theory, holds for a general anisotropic Fermi (F) surface and for any gap symmetry [23]. The formalism in the form convenient for our purpose is outlined in Ref. 2. We refer readers to this work for details. The theory is formulated in terms of functions \( f(r, k_F, \omega), f^+, \) and \( g \) which originate from Gor’kov’s Green’s functions and are normalized by \( g^2 + ff^+ = 1; \)
or equivalently simplification. For multi-band F-surfaces one may need not necessarily the AG spin-flip scattering. In each particular situation, the parameter $\rho_m$ must be properly defined. Here, without specifying the pair breaking mechanism, we apply the AG approach to show that the pair-breaking accounts for our data on the low temperature $\lambda(T)$ along with the earlier reported behavior of $H_{c2}$ slopes at $T_c$ and of the quite unusual dependence of the specific heat jump on $T_c$.

Evaluation of $\lambda(T; \tau, \tau_m)$ for arbitrary $\tau$’s and arbitrary anisotropy of $\Delta$ is difficult analytically. However, for a strong $T_c$ suppression, the problem is manageable. Within the microscopic theory, penetration of weak magnetic fields into superconductors is evaluated by first solving for the unperturbed zero-field state and then treating small fields as perturbations. It was shown by AG that for strong pair-breaking the formalism for the derivation of the Ginzburg-Landau equations near $T_c$ applies at all temperatures. Within the Eilenberger approach this means that $f \ll 1$ and $g \approx 1 - ff^+ / 2$ at all temperatures. The calculation then proceeds in a manner similar to that near $T_c$.

Within a two-band model for iron-based materials, the order parameter is believed to have a $\pm$ s structure, so that $(\Delta) \ll |\Delta_{max}|$. The problem is considerably simplifies if one assumes $(\Delta) = 0$; we use this assumption and expect the model to hold at least qualitatively. In the zero-field state, we look for solutions of Eilenberger equations as $f_0 = f^{(1)} + f^{(2)} + \ldots$ where $f^{(1)} \sim \Delta, f^{(2)} \sim \Delta^2$, etc. The Eilenberger equation for $f$ then yields:

$$f_0 = \frac{\Delta}{\omega_+} + \frac{\Delta}{2\omega_+} \left( \frac{\langle \Delta^2 \rangle}{\omega_+ - \omega_+^2} - \Delta^2 \right) + O(\Delta^3), \quad (4)$$

where $\omega_+ = \omega + 1 / 2 \tau_+$. One can see that even at low temperatures $f_{0,\text{max}} \sim \tau_+ T_c \sim 1 / \rho_+ \ll 1$ because for strong pair-breaking $T_c \to 0$. This is a quasiclassical justification for the AG statement that $f \ll 1$ at all $T$’s.

The $T$ dependence of $\Delta$ (or $\Psi$) is obtained with the help of the self-consistency equation (or the “gap equation”). For a strong pair-breaking, this equation takes the form [2]:

$$\Psi(1 - t^2) = \sum_{\omega > 0} \left( \Psi(\omega) - \langle \Omega f \rangle \right). \quad (5)$$

Substituting here $f$ of Eq. (4), we obtain the order parameter in the field-free state:

$$\Psi^2 = \frac{2\pi^2(T_c^2 - T^2)}{3\Omega^2 - 2} ; \quad (6)$$

this reduces to the AG form for $\Omega = 1$.

We can now consider the response to a small current

$$j = -4\pi|e|N(0)T \text{Im} \sum_{\omega > 0} \langle v g \rangle ; \quad (7)$$

It is well-known that the formal scheme of the seminal Abrikosov-Gor’kov (AG) work on magnetic impurities applies to various situations with different pair-breaking causes, not necessarily the AG spin-flip scattering. Within the microscopic theory, the Matsubara frequencies are $\omega = \pi T(2\nu + 1)$ with an integer $\nu$ and $h = k_B = 1$. The order parameter is taken in the form $\Delta(r, \k_F) = \Psi(r, \k_F) \Omega(\k_F)$ describes the variation of $\Delta$ along the F-surface and is conveniently normalized so that the average over the whole F-surface $(\Omega^2) = 1$. Hence, the model is a BCS-type weak-coupling approach providing a qualitative description at best.

The scattering in the Born approximation is characterized by two scattering times, the transport $\tau$ responsible for the normal conductivity and $\tau_m$ for processes breaking the time reversal symmetry (e.g., spin-flip):

$$1 / \tau_{\pm} = 1 / \tau \pm 1 / \tau_m. \quad (2)$$

Commonly, two dimensionless parameters are used:

$$\rho = 1 / 2 \pi T_c \tau \quad \text{and} \quad \rho_m = 1 / 2 \pi T_c \tau_m, \quad (3)$$

or equivalently $\rho_\pm = \rho \pm \rho_m$. This is of course a gross simplification. For multi-band F-surfaces one may need more parameters for various intra- and inter-band processes, which are hardly controllable and their number is too large for a useful theory. Our model is amenable for analytic work and may prove helpful, the simplifying assumptions notwithstanding.

![FIG. 1. (Color online) (a) $\Delta \chi$ versus $(T/T_c)^2$ for $\text{Ba(Fe}_{0.942}\text{Co}_{0.058})\text{As}_2$ marked by (1), $\text{Ba(Fe}_{0.941}\text{Ni}_{0.059})\text{As}_2$ (2), $\text{BaFe}_{2}0\text{.001Se}_{0.367}\text{Te}_{0.632}$ (3), and $\text{LaFeAsO}_{0.3}\text{Fe}_{0.7}$ (4). Inset: (a) $\Delta \chi$ for $\text{Ba(Fe}_{0.942}\text{Co}_{0.058})\text{As}_2$ marked by (1), $\text{Fe}_{2}0\text{.059}\text{Se}_{0.367}\text{Te}_{0.632}$ (2), $\text{Fe}_{2}0\text{.001Se}_{0.367}\text{Te}_{0.632}$ (3), and $\text{LaFeAsO}_{0.3}\text{Fe}_{0.7}$ (4). (b) Fitted exponent $n$ in $\Delta \chi \propto T^n$.](image-url)
$\lambda(0)$ is the density of states at the F-level per one spin. Weak supercurrents leave the order parameter modulus unchanged, but cause the condensate to acquire an overall phase $\theta(r)$. We then look for perturbed solutions as:

$$\Delta = \Delta_0 e^{i\theta}, \quad f = (f_0 + f_1) e^{i\theta}, \quad f^+ = (f_0 + f_1^*) e^{-i\theta}, \quad g = g_0 + g_1,$$

where the subscript 1 denotes small corrections to the uniform state $f_0, g_0$. In the London limit, the only coordinate dependence is that of the phase $\theta$, i.e., $f_1, g_1$ are $r$ independent. The Eilenberger equations provide the corrections among which we need only $g_1$:

$$g_1 = \frac{i f_0^2 v P}{2\omega_+} = \frac{i \Delta^2 v P}{2\omega_+^3}. \quad (9)$$

see [2]. Here $P = \nabla \theta + 2\pi A/\phi_0 = 2\pi a/\phi_0$ with the “gauged invariant vector potential” $a$.

We now substitute $g_0 + g_1$ in Eq. (7) and compare the result with $4\pi j_1/c = -(\lambda^2)_{ik}^{1/2} a_k$ to obtain:

$$\left(\lambda^2\right)_{ik}^{-1} = \frac{8\pi e^2 N(0)T_c}{c^2} \left\langle v_i v_k \Omega^2 \right\rangle \Psi^2 \sum_{\omega > 0} \frac{1}{\omega_+^3}. \quad (10)$$

The sum here is expressed in terms of the polygama function:

$$\sum_{\omega > 0} \frac{1}{\omega_+^3} = -\frac{1}{16\pi T^2} \psi'' \left(\frac{\rho^+}{2\pi} + \frac{1}{2}\right) \approx \frac{\tau^2}{\pi T}, \quad (11)$$

where $\rho^+ \gg 1$ has been used. Taking into account Eq. (5), one obtains:

$$\left(\lambda^2\right)_{ik}^{-1} = \frac{16\pi^3 e^2 N(0)k_B^2 T_c^2}{c^2 \hbar^2 (3\langle \Omega^4 \rangle - 2)} \left\langle v_i v_k \Omega^2 \right\rangle (T_c^2 - T^2) \quad (12)$$

in common units. It is now easy to obtain the low $T$ behavior of $\Delta \lambda_{ab} = \lambda_{ab}(T) - \lambda_{ab}(0)$ for a uniaxial material:

$$\Delta \lambda_{ab} = \eta T^2 \frac{T_c}{T^3}, \quad \eta = \frac{\hbar c}{8\pi k_B T_c} \sqrt{\frac{3\langle \Omega^4 \rangle - 2}{\pi e^2 N(0)\langle v_g \Omega^2 \rangle}}. \quad (13)$$

We stress that $\tau_c$ here is close to the critical value for which $T_c \rightarrow 0$. One readily obtains for $T = 0$,

$$\lambda_{ab}(0) = 2\eta/T_c. \quad (14)$$

Note: Eqs. (13) and (14) are derived for $\langle \Omega \rangle \approx 0$. One can show that they hold for $\langle \Omega \rangle \neq 0$ as well with, however, different coefficient $\eta$. We do not provide here this cumbersome calculation.

To examine the predicted scaling behavior, the factor $\beta$ in $\Delta \lambda = \beta T^2$ was obtained by fitting the low temperature $\Delta \lambda$ for the same 122, 1111 and 11 compounds of Fig.[3] with $\beta$ being the only fitting parameter. The $\beta$'s are plotted in the main panel of Fig.[2] versus $T_c$. The error bars on this graph reflect the fact that each sample studied has a certain transition width. The inset of Fig. 2 shows the convention adopted here for $T_c$ determination. The uncertainty of $T_c$ is the dominant source of error in determination of $\beta$. According to the strong pair breaking scenario, $\beta = \eta/T_c^3$. To compare experiment with theory, $\beta$ is plotted on a log-log scale in the main frame of Fig.[2] along with the line $\beta = (8.8 \pm 1.0)/T_c^3$ obtained by fitting the data. Moreover, by substituting $v \sim 10^7 \text{cm/s}$ and $N(0) \sim 10^{33} \text{erg}^{-1} \text{cm}^{-3}$ in Eq. (13) we roughly estimate $\tau_c \sim 3 \times 10^{-14} \text{s}$; this value corresponds to parameter $\rho^+ \approx 5$ for $T_c = 40 \text{K}$ and to larger values for lower $T_c$'s, an observation consistent with the major model assumption of $\rho^+ \gg 1$. The degree to which the experimental values follow the theory is remarkable, a substantial scatter of the data points notwithstanding.

The scalings of Eqs. (13) and (14) are obtained for a strong pair-breaking materials with the order parameter obeying $\langle \Omega \rangle \approx 0$. These conditions are likely to be satisfied in underdoped high-$T_c$ cuprates since underdoped materials are clearly disordered and the d-wave order parameter is suppressed by any scattering. Indeed, the surface resistance [26] and optical data [27] for YBa$_2$Cu$_3$O$_{6+x}$ samples with $T_c$ varying from 3 to 17 K show $1/\lambda_{ab}^2(0) \propto T_c^2$ in agreement with Eq. (14). This behavior differs from “Uemura scaling” $1/\lambda_{ab}^2(0) \propto T_c$ [28].

To our knowledge there is not yet sufficient data on $\lambda(0)$ for the iron-based materials to verify the scaling [11]. Similarly, we are not aware of a data set to check the scaling $H_{c2}(T)$ described in Ref. [2] are approximate by design since their derivati-
tion involves a number of simplifying assumptions. Still they are robust in showing that the pair-breaking is an important factor in superconductivity of iron-pnictides.

Many questions still remain; for example, why the Co doped 122 compounds deviate substantially from the general scaling behavior shown in Fig. 2, see also Ref. [2]. Another problem to address is how to reconcile the strong pair-breaking, which in the isotropic case leads to gapless superconductivity [24], with the in-plane thermal conductivity data showing $\kappa(0) = 0$ [25, 30]. At this point, we can say that (a) the strong pair-breaking model for anisotropic order parameters states that the total density of states $N(\epsilon)$ integrated over all pockets of the Fermi surface is finite at zero energy [2]; this does not exclude a possibility that $N = 0$ for some parts on the Fermi surface. And (b): in this work we are interested in the superfluid density $\propto 1/\lambda^2$ which depends only on the Fermi surface average $\langle \Delta \rangle$ so that our results are less sensitive to the $\Delta$ behavior on a particular set of directions (e.g., those in the $ab$ plane). The same qualitative argument shows that our scalings do not contradict the in-plane ARPES data [31].

To conclude, analysis of the low-temperature behavior of the London penetration depth shows that a strong pair-breaking is likely to be responsible for the nearly universal temperature dependence $\Delta_{ab} \propto T^2/T_c^3$, along with earlier reported $\Delta C \propto T_c^{-3}$ and $[dH_{c2}/dT]_c \propto T_c$, in nearly all iron-based superconductors.

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