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

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The low-temperature variation in 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, $\beta\propto 1/T_c^3$ 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, $\Delta C\propto T_c^3$ [Bud'ko *et al.*, Phys. Rev. B **79**, 220516(R) (2009)], and the slopes of the upper critical field, $dH_{c2}/dT\propto T_c$ in these materials [Kogan, Phys. Rev. B **80**, 214532 (2009)]. This suggests that thermodynamic and electromagnetic properties of the iron-based superconductors can be described within a strong pair-breaking scenario (this work).

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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-based superconductors, the specific-heat jump, ΔC, at the critical temperature T_c shows an extraordinary scaling $\Delta C\propto T_c^3$, whereas in conventional *s*-wave materials $\Delta C\propto T_c$. According to Ref. 2, this unusual scaling is caused by a strong pair breaking in materials with anisotropic order parameters; both transport and magnetic scattering in such materials suppress T_c and there are plenty of reasons for magnetic pair breaking in iron-based superconductors. Another consequence of this model, proportionality of slopes of the upper critical field $[dH_{c2}/dT]_{T_c}$ to T_c, has also been shown to hold for the data available.[2] In this work we show that the same idea can be applied to the low-temperature behavior of the London penetration depth $\lambda(T)=\lambda(0)+\Delta\lambda$, for which the pair breaking results in

$$\Delta\lambda \propto T^2/T_c^3. \tag{1}$$

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 $\Delta\lambda(T)\propto T^n$ with $n\approx 2$;[3–9] for some compounds $n\approx 1$ is claimed.[10–12] Commonly, a nonexponential behavior is taken as evidence of an unconventional order parameter, possibly having a nodal gap structure.[4,7,8,13] 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. For example, in *d*-wave superconductors, the linear low-T dependence of λ in the clean case changes to T^2 in the presence of moderate scattering.[14] In fact, the connection between the power-law behavior of $\Delta\lambda(T)$ and scattering in iron-based superconductors had been already suggested.[4,7,8,13] The symmetry of the order parameter Δ in multiband iron pnictides is not yet determined with certainty, however, many favor the $\pm s$ structure.[15,16] The Fermi-surface average of the order parameter in this model is such that $\langle\Delta\rangle \ll \Delta_{\max}$. 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 data were collected from our previous reports on different families of iron-based superconductors.[3–7,17] Details of sample synthesis and characterization can be found in Refs. 18–20. The low-temperature variation in the London penetration depth, $\Delta\lambda$, was measured by using a self-oscillating tunnel diode resonator described in detail elsewhere.[21,22] Fig. 1(a) shows examples of the quadratic variation in $\Delta\lambda$ that appear as straight lines when plotted versus $(T/T_c)^2$ for $T<T_c/3$ in compounds with T_c varying from $\approx$12 to 23 K. The exponent n in $\Delta\lambda\propto T^n$ extracted by fitting the low-temperature data shown for six compounds in Fig. 1(b). We

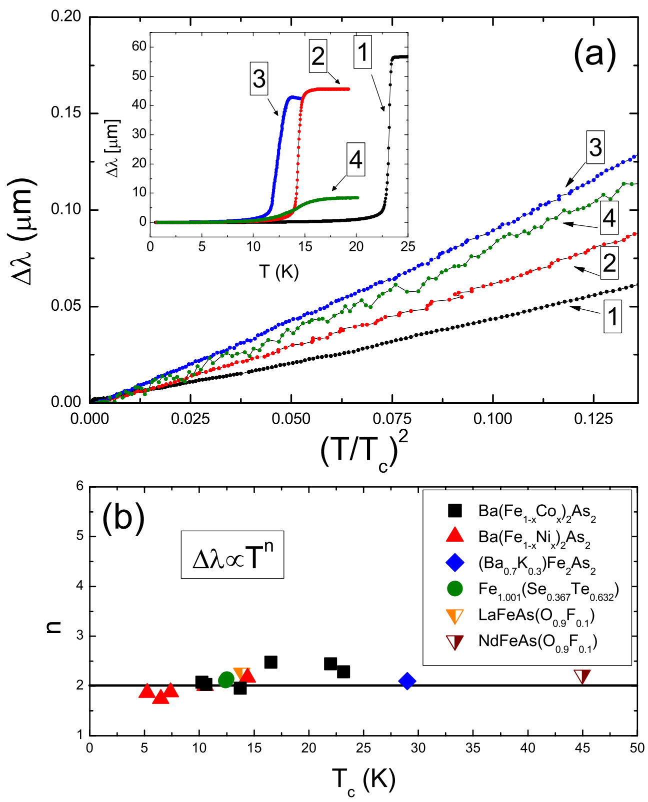

FIG. 1. (Color online) (a) $\Delta\lambda$ versus $(T/T_c)^2$ for $Ba(Fe_{0.942}Co_{0.058})_2As_2$ marked by (1), $Ba(Fe_{0.941}Ni_{0.059})_2As_2$ (2), $Fe_{1.001}Se_{0.367}Te_{0.632}$ (3), and $LaFeAsO_{0.9}F_{0.1}$ (4). Inset: (a) $\Delta\lambda$ in the full temperature range. (b) Fitted exponent n in $\Delta\lambda\propto T^n$.

see that $\Delta\lambda(T)\propto T^2$ holds for the $(AE)(Fe_{1-x}TM_x)_2As_2$ (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, and *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 belong to four different families of the iron-based materials. This common behavior has prompted us to look for a common 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 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 $f(\boldsymbol{r},\boldsymbol{k}_F,\omega), f^+$, and g which originate from Gor'kov's Green's functions and are normalized by $g^2+ff^+=1$; the Matsubara frequencies are $\omega=\pi T(2\nu+1)$ with an integer ν and $\hbar=k_B=1$. The order parameter is taken in the form $\Delta(\boldsymbol{r},\boldsymbol{k}_F)=\Psi(\boldsymbol{r},T)\Omega(\boldsymbol{k}_F)$ where $\Omega(\boldsymbol{k}_F)$ describes the variation in Δ along the Fermi surface and is conveniently normalized so that the average over the whole Fermi surface $\langle\Omega^2\rangle=1$. Hence, our model uses 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 τ responsible for the normal conductivity and τ_m for processes breaking the time-reversal symmetry (e.g., spin flip)

$$1/\tau_{\pm} = 1/\tau \pm 1/\tau_m. \tag{2}$$

Commonly, two dimensionless parameters are used

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

or equivalently $\rho_{\pm}=\rho\pm\rho_m$. This is of course a gross simplification. For multiband Fermi surfaces one may need more parameters for various intraband and interband 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.

It is well known that the formal scheme of the seminal Abrikosov-Gor'kov (AG) work on magnetic impurities[24] applies to various situations with different pair breaking causes, not necessarily the AG spin-flip scattering.[25] In each particular situation, the parameter ρ_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 in 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 τ's and arbitrary anisotropy of Δ 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 (Ref. 24) 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[15] so that $\langle\Delta\rangle\ll|\Delta_{\max}|$.[16] The problem simplifies considerably if one assumes $\langle\Delta\rangle=0$; 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 $f_0=f^{(1)}+f^{(2)}+\cdots$ where $f^{(1)}\sim\Delta$, $f^{(2)}\sim\Delta^2$, etc. The Eilenberger equation for f then yields[2]

$$f_0 = \frac{\Delta}{\omega_+} + \frac{\Delta}{2\omega_+^3}\left(\frac{\langle\Delta^2\rangle}{2\tau_+\omega_+} - \Delta^2\right) + \mathcal{O}(\Delta^5), \tag{4}$$

where $\omega_+=\omega+1/2\tau_+$. One can see that even at low temperatures $f_{0,\max}\sim\tau_+T_c\sim 1/\rho_+\ll 1$ 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 Δ (or Ψ) is obtained from the self-consistency equation (the "gap equation"). For a strong pair breaking, this equation takes the form[2]

$$\frac{\Psi(1-t^2)}{12\pi T\rho_+^2} = \sum_{\omega>0}^{\infty}\left(\frac{\Psi}{\omega^+} - \langle\Omega f\rangle\right). \tag{5}$$

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

$$\Psi^2 = \frac{2\pi^2(T_c^2-T^2)}{3\langle\Omega^4\rangle - 2} \tag{6}$$

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

We can now consider the response to a small current

$$\boldsymbol{j} = -4\pi|e|N(0)T\,\mathrm{Im}\sum_{\omega>0}\langle\boldsymbol{v}g\rangle \tag{7}$$

$N(0)$ is the density of states at the Fermi level per one spin. Weak supercurrents leave the order parameter modulus unchanged but cause the condensate to acquire an overall phase $\theta(\boldsymbol{r})$. We then look for perturbed solutions as

$$\Delta = \Delta_0 e^{i\theta}, \quad f = (f_0+f_1)e^{i\theta},$$

$$f^+ = (f_0+f_1^+)e^{-i\theta}, \quad g = g_0 + g_1, \tag{8}$$

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

$$g_1 = \frac{i f_0^2 \boldsymbol{vP}}{2\omega_+} = \frac{i\Delta^2 \boldsymbol{vP}}{2\omega_+^3} \tag{9}$$

see Ref. 2. Here $\boldsymbol{P}=\nabla\theta+2\pi\boldsymbol{A}/\phi_0 \equiv 2\pi\boldsymbol{a}/\phi_0$ with the "gauge invariant vector potential" $\boldsymbol{a}$.

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

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

The sum here is expressed in terms of the polygamma function

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

where $\rho_+ \gg 1$ has been used. Taking into account Eq. (6), one obtains

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

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

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

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

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

Note: Eqs. (1) 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 η. We do not provide here this cumbersome calculation.

To examine the predicted scaling behavior, the factor β in $\Delta\lambda=\beta T^2$ was obtained by fitting the low temperature $\Delta\lambda$ for 122, 1111, and 11 compounds of Fig. 1 with β being the only fitting parameter. The β'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 β. According to the strong pair-breaking scenario, $\beta=\eta/T_c^3$. To compare experiment with theory, β is plotted on a log-log scale in the main frame of Fig. 2 along with the line $\beta=(8.8\pm1.0)/T_c^3$ obtained by fitting the data. Moreover, by substituting $v\sim10^7$ cm/s and $N(0)\sim10^{33}$ erg^{-1} cm^{-3} in Eq. (1) we roughly estimate $\tau^+\sim 3\times10^{-14}$ s; this value corresponds to the parameter $\rho^+\approx 5$ for T_c=40 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 experimental values follow the theory is remarkable, a substantial scatter of the data points

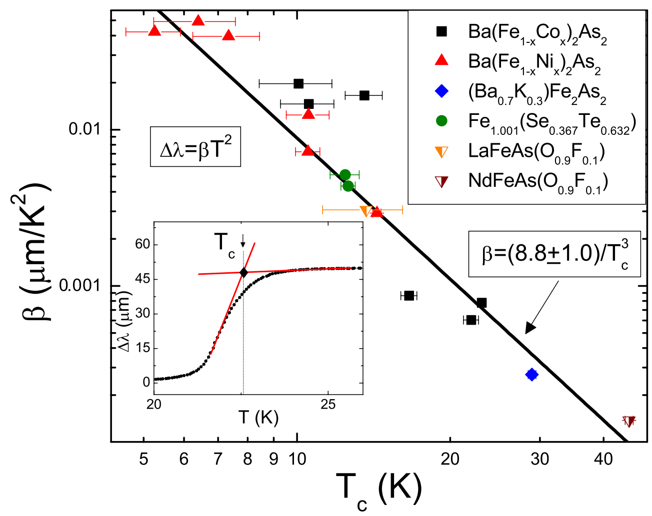

FIG. 2. (Color online) The factor β is obtained by fitting the data to $\Delta\lambda=\beta T^2$ and plotted versus T_c on a log-log scale. The solid line is a fit to $\beta=\eta/T_c^3$, motivated by Eq. (1) for a strong pair breaking.

notwithstanding. It is worth noting that $1/T_c^3$ scaling in $\Delta\lambda \propto T^2/T_c^3$ is a result of a strong pair breaking and—to our knowledge—does not follow from any other currently discussed model. On the other hand, we do not have yet a sufficient data set to verify the scaling in Eq. (14). Similarly, we are not aware of data to check the scaling $H_{c1}\propto T_c^2$ which follows from Eq. (14). We would like to stress that the penetration depth scalings discussed here as well as those for the specific-heat jump and for the slopes of $H_{c2}(T)$ described in Ref. 2 are approximate by design since their derivation involves a number of simplifying assumptions. Still they are robust in showing that the pair breaking is an important factor for superconductivity of iron pnictides.

The question arises whether or not one can have iron-based materials free of the pair-breaking scattering. If possible, these materials would have had much higher critical temperatures. For example, if $\rho_+\approx 5$ and $T_c\approx 20$ K, the "clean material" would have $T_{c0}=T_c\exp[\psi(\rho_+/2+1/2)-\psi(1/2)]$ in the range of room temperatures. We answer this question in negative. The pair-breaking scattering is probably inherent for the iron-based superconductors because the same interactions (presumably, spin fluctuations) cause both the pairing and the pair breaking and the full theory ought to consider both effects on the same footing.

Many other 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 $\kappa/T(T\to 0)=0$.[26,27] 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 the 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 averages so that our results are less sensitive to the Δ 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.[28]

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 $\Delta\lambda_{ab} \propto T^2/T_c^3$, along with earlier reported $\Delta C \propto T_c^{-3}$ and $[dH_{c2}/dT]_{T_c} \propto T_c$, in nearly all iron-based superconductors.

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