Microwave Surface-Impedance Measurements of the Magnetic Penetration Depth in Single Crystal Ba$_{1-x}$K$_x$Fe$_2$As$_2$ Superconductors: Evidence for a Disorder-Dependent Superfluid Density

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We report high-sensitivity microwave measurements of the in-plane penetration depth $\lambda_{ab}$ and quasiparticle scattering rate $1/\tau$ in several single crystals of hole-doped Fe-based superconductor Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x \approx 0.55$). While power-law temperature dependence of $\lambda_{ab}$ with the power $\sim 2$ is found in crystals with large $1/\tau$, we observe exponential temperature dependence of superfluid density consistent with the existence of fully opened two gaps in the cleanest crystal we studied. The difference may be a consequence of different level of disorder inherent in the crystals. We also find a linear relation between the low-temperature scattering rate and the density of quasiparticles, which shows a clear contrast to the case of d-wave cuprate superconductors with nodes in the gap. These results demonstrate intrinsically nodeless order parameters in the Fe-arsenides.

The discovery of high-$T_c$ superconductivity in Fe-pnictides$^1$ has attracted tremendous interests both experimentally and theoretically. The ‘mother’ materials have antiferromagnetic spin-density-wave order$^2$ and the superconductivity appears by doping charge carriers, either electrons or holes. Such carrier doping induced superconductivity resembles high-$T_c$ cuprates, but one of the most significant differences is the multiband nature of superconductivity in this new class of materials.

In this context, identifying the detailed structure of superconducting order parameter, particularly the presence or absence of nodes in the gap function, is of primary importance. In the electron-doped LnFeAs(O,F) or ‘1111’ (where Ln is Lanthanide ions), several experiments using polycrystals$^4,5$ suggest nodes in the gap, the tunnelling measurements$^6$, the magnetic penetration depth$^7$, and angle resolved photoemission (ARPES)$^8$ consistently indicate fully gapped superconductivity. In the hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ or ‘122’$^9$, however, experimental situation is controversial: ARPES$^{10,11,12}$ and lower critical field measurements$^{13}$ suggest the presence of line nodes in the gap function. Also, recent penetration depth measurements suggest superconductivity with point nodes in Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$$^{13}$. Such controversies may partly come from the different samples with various degrees of disorder in these reports. Indeed, in the unconventional $s_{\pm}$ state, the quasiparticle excitation spectrum is found to be sensitive to the pair breaking and interband scattering$^{16,17}$. Also, inhomogeneity or weak-links may affect the superfluid density$^{18}$, which is a direct measure of low-energy quasiparticle excitations. So experiments in single crystals with well-characterized quality are needed to elucidate how disorder affects the quasiparticle excitations and what is the intrinsic structure of the gap in the clean limit.

Here we present surface impedance measurements in several single crystals of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x \approx 0.55$). In this material, disorder may be caused by microscopic inhomogeneous content of K, which is reactive with moisture or oxygen. The degree of disorder can be quantified by the quasiparticle scattering rate $1/\tau$. We find that the temperature dependence of the in-plane penetration depth $\lambda_{ab}(T)$ is sample dependent, which can account for some of the controversies in the previous reports: crystals with large $1/\tau$ tend to exhibit power-law temperature dependence of $\lambda_{ab}(T)$. In the best crystal with the smallest $1/\tau$, we obtain strong evidence for the nodeless superconductivity having at least two different gaps.

Single crystals of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ were grown by a self-flux method$^{19}$. High purity starting materials were heated up to 1190°C under Ar atmosphere, and then cooled down at a rate of 4°C/hours, followed by a quench at 850°C. Energy dispersive X-ray (EDX) analysis reveals the doping level $x = 0.55(2)$$^{20}$, which is consistent with the c-axis lattice constant $c = 1.341(3)$ nm determined by X-ray diffraction$^{21}$. Bulk superconductivity is characterized by the magnetization measurements using a commercial magnetometer. In-plane mi-
crowave surface impedance \( Z_s = R_s + iX_s \) is measured in the Meissner state by using a sensitive superconducting cavity resonator \([7, 22, 23]\). In our frequency range \( \omega/2\pi \approx 28 \text{ GHz} \), the conductivity \( \sigma = \sigma_1 - i\sigma_2 \) can be extracted from \( Z_s(T) \) through the relation valid for the skin-depth regime:

\[
Z_s = R_s + iX_s = \left( \frac{i\mu_0\omega}{\sigma_1 - i\sigma_2} \right)^{1/2} .
\]

In the superconducting state, the surface reactance is a direct measure of the superfluid density \( n_s \) via \( X_s(T) = \mu_0\omega\lambda_{ab}(T) \) and \( \lambda_{ab}^{-2}(T) = \mu_0n_s(T)e^2/m^* \). In the normal state, \( \sigma_1 = ne^2\tau/m^* \gg \sigma_2 \) gives \( R_s(T) = X_s(T) = (\mu_0\omega/2\sigma_1)^{1/2} \) from Eq. (1), where \( n \) is the total density of carriers with effective mass \( m^* \). This equality can be used to determine \( X_s(0)/X_s(T_c) \), which allows us to determine \( \lambda_{ab}(T)/\lambda_{ab}(0) \) without any assumptions \([22]\). This also gives us estimates of the normal-state scattering rate \( 1/\tau = 1/\mu_0\sigma_1\lambda_{ab}^2(0) = 2\omega(X_s(T)/X_s(0))^2 \), which quantifies the degrees of disorder for each sample \([7]\).

Below \( T_c \), the real part of conductivity \( \sigma_1 \) is determined by the quasiparticle dynamics, and in the simple two-fluid model, which is known to be useful in cuprate superconductors \([21, 27]\), \( \sigma_1 \) is related to the quasiparticle scattering time \( \tau \) through \( \sigma_1 = (n-n_s)e^2\tau/m^*(1+\omega^2\tau^2) \).

Figure 1(a) shows the temperature dependence of magnetization \( M \). In the samples studied here, the low-temperature \( M \) (below \( \sim 10 \text{ K} \)) is independent of temperature, which ensures the bulk nature of superconductivity. However, the superconducting transition width varies slightly from sample to sample. Since this is likely related to the microscopic inhomogeneity of K content near the surface, which can be enhanced upon exposure to the air, we carefully cleave both sides of the surface of crystal #2 and cut into smaller size (crystal #3). For #3, the microwave measurements are done with minimal air exposure time, and another piece of cleaved crystal (#3') is used to measure \( M(T) \). In Fig. 1(b) we compare the temperature dependence of normal-state microwave resistivity \( \rho_1 = 1/\sigma_1 = 2R_s^2/\mu_0\omega \) [see Eq. (1)] for 3 samples. Note that such microwave measurements provides one of the most severe quality checks for superconductors \([18]\): additional quasiparticle excitations by the applied 28-GHz (~1.3 K) microwave should intrinsically broaden the transition width compared with the dc resistivity. It is now clear that the cleavage dramatically improves the sample quality, and crystal #3 exhibits the sharpest transition and the lowest \( 1/\tau \) [see Table I]. The ratio \( \rho_1(100\text{ K})/\rho_1(T_c) \) exceeds 4 [inset of Fig. 1(b)], which is larger than that of less doped crystals with \( x \lesssim 0.4 \) \([21]\).

In Fig. 2(a), we show typical temperature dependence of \( Z_s(T)/Z_s(40\text{ K}) \). As expected from Eq. (1), the normal-state temperature dependencies of real and imaginary parts are identical. The strong temperature dependence above \( T_c \) allows us to determine precisely the offset of \( X_s(0)/X_s(40\text{ K}) \), from which we are able to obtain \( n_s(0)/n_s(40\text{ K}) = \lambda_{ab}^2(0)/\lambda_{ab}^2(T) \) as demonstrated in Fig. 2(b). We find that crystals with large scattering rates exhibit strong temperature dependence of superfluid density at low temperatures, which mimics the power-law temperature dependence of \( n_s(T) \) in \( d \)-wave superconductors with nodes. As shown in the inset of Fig. 2(b), the low-temperature change in the penetration depth \( \delta\lambda_{ab}(T) = \lambda_{ab}(T) - \lambda_{ab}(0) \) can be fitted to \( T^2 \) and \( T^{2.4} \) dependence in crystal #1 and #2 respec-

![FIG. 1](color online). (a) Temperature dependence of dc magnetization in the zero-field-cooling condition under a 20-Oe field along the c axis. (b) Temperature dependence of the normalized 28-GHz microwave resistivity \( \rho_1(T)/\rho_1(40\text{ K}) \). Inset: the same plot for crystal #3 up to 100 K.

![FIG. 2](color online). (a) Temperature dependence of the surface resistance \( R_s \) and \( X_s \) in crystal #3. (b) Normalized superfluid density \( \lambda_{ab}^2(0)/\lambda_{ab}^2(T) \) for 3 samples with different normal-state scattering rates [see Table I]. Inset shows the low-temperature change in the penetration depth.

| sample | size (\( \mu m^3 \)) | \( T_c \) (K) | \( n_s \) \( (s^{-1}) \) | \( \lambda_{ab} \) \( (s^{-1}) \) |
|--------|----------------|-------------|----------------|----------------|
| #1     | \( 320 \times 500 \times 100 \) | \( 26.4(3) \) | \( 27(3) \times 10^{12} \) | \( 1.2(1) \times 10^{12} \) |
| #2     | \( 300 \times 500 \times 80 \) | \( 25.0(4) \) | \( 21(2) \times 10^{12} \) | \( 1.0(1) \times 10^{12} \) |
| #3     | \( 100 \times 180 \times 20 \) | \( 32.7(2) \) | \( 7.8(5) \times 10^{12} \) | \( 0.5(1) \times 10^{12} \) |
The solid line is a BCS calculation with $\frac{T}{T_c} \approx 0.55$. This indicates that the Fermi surface with the smaller gap $\Delta_1$ has a relatively large volume or carrier number, which is also in good correspondence with the ARPES results.

Next we discuss the low-energy quasiparticle dynamics. In Fig. 4 we show the temperature dependence of the quasiparticle conductivity $\sigma_1(T)/\sigma_1(35 \text{ K})$ in the cleanest sample #3. It is evident that below $T_c$, $\sigma_1(T)$ is enhanced from the normal-state values. Near $T_c$, the effects of coherence factors and superconducting fluctuations are known to enhance $\sigma_1(T)$. The former effect, known as a coherence peak, is represented by the solid line in Fig. 4(a). We note that in the $s_\pm$ pairing state, the coherence peak in the NMR relaxation rate can be suppressed by a partial cancellation of total susceptibility $\sum_q \chi(q)$ owing to the sign change between the hole and electron bands [28]. For microwave conductivity, the

\[
\frac{\delta \lambda_{ab}(T)}{\lambda_{ab}(0)} \approx \sqrt{\frac{\pi \Delta}{2k_B T}} \exp \left( -\frac{\Delta}{k_B T} \right)
\]
long wave length limit $q \to 0$ is important and the coherence peak can survive [29], which may explain the bump in $\sigma_1(T)$ just below $T_c$. At lower temperatures, where the coherence and fluctuation effects should be vanishing, $\sigma_1(T)$ shows a further enhancement. A similar but less pronounced enhancement has been observed in a 1111 system [30]. This $\sigma_1(T)$ enhancement can be attributed to the enhanced quasiparticle scattering time $\tau$ below $T_c$. The competition between increasing $\tau$ and decreasing quasiparticle density $n_n(T) = n - n_s(T)$ makes a peak in $\sigma_1(T)$. This behavior is ubiquitous among superconductors having strong inelastic scattering in the normal state [23-25]. Following the pioneering work by Bonn et al. [25], we employ the two-fluid analysis to extract the quasiparticle scattering rate $1/\tau(T)$ at low temperatures below $\sim 25$ K. In Fig. 1(b), the extracted $1/\tau(T)$ is plotted against the normalized quasiparticle density $n_n(T)/n$ and compared with the reported results in the $d$-wave superconductor YBa$_2$Cu$_3$O$_{6.95}$ [23]. It is found that the scattering rate scales almost linearly with the quasiparticle density in our 122 system, which is distinct from $1/\tau$ in cuprates that varies more rapidly as $\sim n_n^3$. Such cubic dependence in cuprates is consistent with the $T^3$ dependence of spin-fluctuation inelastic scattering rate expected in $d$-wave superconductors, which have $T$-linear dependence of $n_n$ [31]. In $s$-wave superconductors without nodes, $n_n(T)$ and $1/\tau(T)$ are both expected to follow exponential dependence $\sim \exp(-\Delta/k_B T)$ at low temperatures [31], which leads to the linear relation between $1/\tau(T)$ and $n_n(T)$. So this newly found relation further supports the fully gapped superconductivity in this system. We also note that such analysis yields residual scattering rates at low temperatures for each sample [see Table I], which reinforces the sample-dependent disorder.

In summary, we measured surface impedance in several Ba$_{1-x}$K$_x$Fe$_2$As$_2$ single crystals. The temperature dependence of the superfluid density depends on the samples having different scattering rates. In the cleanest sample with the smallest $1/\tau$, the superfluid density shows exponential behavior consistent with fully opened two gaps. The scattering rate analysis highlights the difference from the $d$-wave cuprates, which also supports the conclusion that the intrinsic order parameter in Fe-As superconductors is nodeless all over the Fermi surface.

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Note added—After submission, large quasiparticle thermal Hall conductivity in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ has been reported [32], consistent with the enhanced $\tau(T)$ below $T_c$.

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