Probing pairing symmetry of $Sm_{1.85}Ce_{0.15}CuO_4$ via highly-sensitive voltage measurements: Evidence for strong impurity scattering

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Using a highly-sensitive home-made mutual-inductance technique, temperature profiles of the magnetic penetration depth $\lambda(T)$ in the optimally-doped $Sm_{1.85}Ce_{0.15}CuO_4$ thin films have been extracted. The low-temperature behavior of $\lambda(T)$ is found to be best-fitted by linear $\Delta\lambda(T)/\lambda(0) = \ln(2)k_BT/\Delta_0$ and quadratic $\Delta\lambda(T)/\lambda(0) = \Gamma^{-1/2}\Delta_0^{-3/2}T^2$ laws above and below $T = 0.22T_C$, respectively, which clearly indicates the presence of d-wave pairing mechanism dominated by strong paramagnetic scattering at the lowest temperatures. The best fits produce $\Delta_0/k_BT_C = 2.07$ and $\Gamma/T_C = 0.25(T_C/\Delta_0)^2$ for the estimates of the nodal gap parameter and impurity scattering rate.

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INTRODUCTION

It is well established that most of the conventional low-$T_C$ superconductors have s-wave pairing symmetry [1]. As for high-$T_C$ cuprates (HTC), the study of pairing symmetry in these materials still remains one of the most polemical and active fields of research. A number of recent experiments, including phase-sensitive measurements [2,3], the angle resolved photoemission (ARPES) [4,5], and the Raman spectroscopy [6] have revealed that electron-doped HTC with nearly optimal doping have predominantly $d_{x^2-y^2}$ pairing symmetry. In particular, Kokales et al. [7], Prozorov et al. [8] and Snezhko et al. [9] showed that the low temperature superfluid density of Ce-based magnetic superconductors $Pr_{2-x}Ce_xCuO_4$ (PCCO) and $Nd_{2-x}Ce_xCuO_4$ (NCCO) varies quadratically with temperature in the whole range of doping, in agreement with the theoretical prediction for a d-wave superconductor with impurity scattering. Moreover, recently remeasured [10] magnetic-field dependence of the low-temperature specific heat of optimally-doped ($x = 0.15$) PCCO give further evidence in favor of d-wave-like pairing symmetry in this material at all temperatures below 4.5K. We should also mention very interesting result [11] on anomalous change in the field dependence of the electronic specific heat in PCCO crystals from linear (at $T = 2K$) to nonlinear (at $T = 3K$) temperature behavior which can provide plausible explanation for the previous conflicting experimental results on the pairing symmetry in the electron-doped cuprates.

At the same time, much less is known about such electron-doped material as $Sm_{2-x}Ce_xCuO_4$ (SCCO). Since $Sm$ has a larger ion size than $Ce$, $Pr$ and $Nd$, it is expected that paramagnetic scattering contribution to low-temperature behavior of SCCO should be much stronger than in PCCO and NCCO. Indeed, the penetration depth measurements on SCCO single crystals [12] have indicated that this magnetic superconductor exhibits a rather strong enhancement of diamagnetic screening below 4K which (by analogy with PCCO and NCCO) could be responsible for a d-wave pairing scenario with rather strong impurity scattering.

To shed more light on the pairing symmetry of electron-doped magnetic superconductors, in this Letter we present a study on the optimally-doped $Sm_{1.85}Ce_{0.15}CuO_4$ (in the form of thin films grown by the pulsed laser deposition technique) by using a high-sensitivity homemade mutual-inductance bridge to extract their penetration depths with high precision.

EXPERIMENTAL PROCEDURE AND EXTRACTION METHOD

A few SCCO thin films ($d = 200nm$ thick) grown by pulsed laser deposition on standard $LaAlO_3$ substrates [7] were used in our measurements. All samples showed similar and reproducible results. The structural quality of the samples was verified through X-ray diffraction and scanning electron microscopy together with energy dispersive spectroscopy technique. To account for a possible magnetic response from substrate, we measured several stand alone pieces of the substrate. No tangible contribution due to magnetic impurities was found. The critical temperature $T_C$ was determined via the measured complex voltage output $V_{AC} = V' + iV''$ as the temperature where $V' = 0$ (see Fig.1).
The experimental bridge used in this work is based on the mutual-inductance method. To measure samples in the shape of thin films, the so-called screening method has been developed [13]. It involves the use of primary and secondary coils, with diameters smaller than the dimension of the sample. When these coils are located near the surface of the film, the response (i.e., the complex voltage output $V_{AC}$) does not depend on the radius of the film or its properties near the edges. In the reflection technique [14], an excitation (primary) coil coaxially surrounds a pair of counter-wound (secondary) pick-up coils. If we take the current in the primary coil as a reference, $V_{AC}$ can be expressed via two orthogonal components, i.e., $V_{AC} = V_L + iV_R$. The first one is the inductive component, $V_L$ (which is in phase with the time-derivative of the reference current) and the second one is the quadrature resistive component, $V_R$ (which is in phase with the reference current). It can be easily demonstrated that $V_L$ and $V_R$ are directly related to the average magnetic moment and the energy losses of the sample, respectively [15]. When there is no sample in the system, the net output from the secondary coils is close to zero because the pick up coils are identical in shape but are wound in opposite directions. The sample is positioned as close as possible to the set of coils, to maximize the induced signal in the pick-up coils. An alternate current sufficient to create a magnetic field of amplitude $h_{AC}$ and frequency $f$ is applied to the primary coil by an alternating voltage source, $V_{in}$. The output voltage of the secondary coils $V_{AC}$ is measured through the usual lock-in technique [16].

To extract the profile of the penetration depth within the discussed here method, one should resolve the following equation relating the measured output voltage $V_{AC}$ to the $\lambda(T)$ sensitive sample features [14]:

$$V_{AC} = V' + iV'' = i\omega I_p \int_0^\infty dx \frac{M(x)}{1 + 2x/Q}$$  \hspace{1cm} (1)

where $Q = i\omega GM_0 (h_p + h_s)$. Here, $I_p$ and $\omega = 2\pi f$ are respectively the amplitude and the frequency of the current in the primary coil, $h_p$ ($h_s$) is the distance from the primary (secondary) coil to the sample, $G$ is the total conductance of the sample, and $M(x)$ is a geometrical factor [14]. Since the total impedance of the sample is given by [17]

$$Z = R + i\omega L_k,$$

the expression for the sample’s total conductance reads:

$$G = \frac{1}{R + i\omega L_k}$$ \hspace{1cm} (2)

Here $L_k$ and $R$ are the kinetic inductance and resistance of the sample, respectively. From the above equations it follows that by measuring $V_{AC}(T)$ we can numerically reproduce the temperature dependencies of both $L_k$ and $R$.

Finally, from the two-fluid model, the relation between $L_k$ and $\lambda(T)$ for thin films (with film thickness $d \ll \lambda$) is given by [1,17,18]:

$$L_k = \mu_0 \lambda \coth \left( \frac{d}{\lambda} \right) \approx \mu_0 \lambda \left( \frac{\lambda}{d} \right)$$ \hspace{1cm} (3)

It is worth mentioning that instead of the tabulation-based procedure used before [19], in the present study we have simultaneously determined $G(T)$ from Eq.(1) and extracted both $L_k$ and $R$ using Eq.(2). Then from the temperature dependence of $L_k$ we have obtained the seeking temperature dependence of $\lambda(T)$.

**RESULTS AND DISCUSSION**

Fig.1 shows the typical results for the temperature dependence of the voltage output $V_{AC}(T)$ in a typical SCCO thin film with $T_C = 20.2K$. Fig.2 extracted the obtained $\lambda^2(T)/\lambda^2(0)$ for all temperatures obtained by using Eqs. (1)-(3).

Turning to the discussion of the obtained results, recall that for conventional BCS-type superconductors with s-wave pairing symmetry the superfluid fraction $x_s(T) = \lambda^2(0)/\lambda^2(T)$ saturates exponentially as $T$ approaches zero. On the other hand, for a superconductor with a line of nodes, $x_s(T)$ will show a power-like behavior at low temperatures. More precisely, for tetragonal symmetry (and neglecting dispersion in the $c$-axis direction), the simple $d_{x^2-y^2}$ pairing state predicts a linear dependence [20] $\Delta \lambda(T)/\lambda(0) = AT$ for the low-temperature variation of in-plane penetration depth $\Delta \lambda(T) = \lambda(T) - \lambda(0)$. Here $A = \ln(2)k_F/\Delta_0$ with $\Delta_0$ being the amplitude of the zero-temperature value of the $d$-wave gap parameter. At the same time, in the presence of strong enough impurity scattering the linear $T$ dependence changes to a quadratic dependence $\Delta \lambda(T)/\lambda(0) = BT^2$ where $B = \Gamma^{-1/2}\Delta_0^{-3/2}$ with $\Gamma$ being the (unitary limit) scattering rate, which is proportional to the impurity concentration of the sample [21]. By trying many different temperature dependencies (including both exponential and power-like), we found that above and below $T = 0.22T_C$ our high-quality SCCO films are best-fitted by a linear (see Fig.3a) and quadratic (see Fig.3b)
dependencies, respectively. What is important, the fits produce physically reasonable values of both d-wave node gap parameter $\Delta_0/k_BT_C = 2.07$ and paramagnetic impurity scattering rate $\Gamma/T_C = 0.25(T_C/\Delta_0)^3$. Hence, our results confirm a universal pairing mechanism in electron-doped magnetic superconductors with d-wave nodal symmetry dominated by paramagnetic impurity scattering at the lowest temperatures (for comparison, a strong-coupling BCS behavior with $\Delta_0 = 2.07k_BT_C$ is shown in Fig.3 by dotted line). It is also interesting to notice that boundary temperature ($T = 0.22T_C$) which demarcates two scattering mechanisms (pure and impure) lies very close to the temperature where strong enhancement of diamagnetic screening in SCCO was observed [12] attributed to spin-freezing of Cu spins. Moreover, the above crossover temperature remarkably correlates with the temperature where an unexpected change in the field dependence of the electronic specific heat in PCCO crystals was found [11] attributed to the symmetry change from nodal to gapped. However, to make a more definitive conclusion regarding this correlation and the origin of the crossover, it is necessary to measure the field-dependent contribution to electronic specific heat in our SCCO films. And finally, comparing the above estimates of the node gap parameter and impurity scattering rate deduced from our data for SCCO with similar parameters reported for best PCCO crystals [7] (with $2\Delta_0/k_BT_C = 3.9$ and $\Gamma/T_C = 0.13(T_C/\Delta_0)^3$), we conclude that as expected larger Sm ion indeed produces larger contribution to impurity scattering.

**SUMMARY**

By using a high-sensitivity home-made mutual-inductance technique we extracted with high accuracy the temperature profiles of penetration depth $\lambda(T)$ in high-quality optimally-doped $Sr_{1.85}Ca_{0.15}CuO_4$ (SCCO) thin films. The low-temperature fits of our data clearly demonstrated that this electron-doped magnetic superconductor possesses a d-wave pairing symmetry. More precisely, SCCO was found to follow a clean limit (with linear temperature dependence of $\lambda(T)$) for $T > 0.22T_C$ while dominated by strong paramagnetic impurity scattering (with quadratic temperature dependence of $\lambda(T)$) for $T < 0.22T_C$.

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[1] C.P. Poole Jr., H.A. Farach and R.J. Creswick, *Superconductivity*, Academic Press (1995).
[2] D.J. Van Harlingen, *Rev. Mod. Phys.* 67 (1995), p. 515.
[3] C.C. Tsuei and J.R. Kirtley, *Rev. Mod. Phys.* 72 (2000), p. 969.
[4] T. Sato, T. Kamiyama, T. Takahashi, K. Kurahashi and K. Yamada, *Science* 291 (2001), p. 1517.
[5] N.P. Armitage, D.H. Lu, D.L. Feng, C. Kim, A. Damascelli, K.M. Shen, F. Ronning, Z.-X. Shen, Y. Onose, Y. Taguchi and Y. Tokura, *Phys. Rev. Lett.* 86 (2001), p. 1126.
[6] G. Blumberg, A. Koitzsch, A. Gozar, B.S. Dennis, C.A. Kendziora, P. Fournier and R. L. Greene, *Phys. Rev. Lett.* 88 (2002), p. 107002.
[7] J.D. Kokales, P. Fournier, L.V. Mercaldo, V.V. Talanov, R.L. Greene and S.M. Anlage, *Phys. Rev. Lett.* 85 (2000), p. 3696.
[8] R. Prozorov, R.W. Giannetta, P. Fournier and R.L. Greene, *Phys. Rev. Lett.* 85 (2000), p. 3700.
[9] A. Snezhko, R. Prozorov, D.D. Lawrie, R.W. Giannetta, J. Gauthier, J. Renaud and P. Fournier, *Phys. Rev. Lett.* 92 (2004), p. 157005.
[10] W. Yu, B. Liang and R.L. Greene, *Phys. Rev. B* 72 (2005), p. 212512.
[11] Hamza Bali and R.L. Greene, *Phys. Rev. Lett.* 93 (2004), p. 067001.
[12] R. Prozorov, D.D. Lawrie, I. Hetel, P. Fournier and R.W. Giannetta, *Phys. Rev. Lett.* 93 (2004), p. 147001.
[13] A. F. Herbad and A.T. Fiory, *Phys. Rev. Lett.* 44 (1980), p. 291.
[14] B. Jeanneret, J.L. Gavilano, G.A. Racine, Ch. Leeman and P. Martinoli, *Appl. Phys. Lett.* 55 (1989), p. 2336.
[15] F.M. Araujo-Moreira, P. Barbara, A.B. Cawthorne and C.J. Lobb. In: A. Narlikar, Editor, *Studies of High Temperature Superconductors*, vol. 43, Nova Science, New York (2002), p. 227.
[16] Magnetic Susceptibility of Superconductors and Other Spin Systems, edited by R.A. Hein, T.L. Francavilla, and D.H. Liebenberg, Plenum Press, New York (1992).
[17] T.P. Orlando and K.A. Delin, *Foundations of Applied Superconductivity*, Addison-Wesley, New York (1991).
[18] M. Tinkham, *Phys. Rev. Lett.* 61 (1988), p. 1658.
[19] A. J. C. Lanfredi, J. P. Rino and F. M. Araujo-Moreira, *J. of Magn. Magnetic Materials* 226-230 (2001), p. 288.
[20] A. Zimmers, R.P.S.M. Lobo, N. Bontemps, C.C. Homes, M.C. Barr, Y. Dagan and R.L. Greene, *Phys. Rev. B* 70 (2004), p. 132502.
[21] P. J. Hirschfeld, W. O. Putikka and D. J. Scalapino, *Phys. Rev. B* 50 (1994), p. 10 250.
FIG. 1: Temperature behavior of the typical output voltages, $V_{AC}$, measured for a typical SCCO thin film ($T_C = 20.2K$) under an alternate magnetic field of amplitude $h_{AC} = 100 \text{ mOe}$ and frequency $f = 55 \text{ kHz}$.

FIG. 2: Extracted from the output voltages $V_{AC}(T)$ variation of $\lambda^2(T)/\lambda^2(0)$ for SCCO thin film as a function of the reduced temperature using Eqs.(1)-(3).
FIG. 3: Low temperature fits of the extracted variation of the penetration depth $\Delta \lambda(T)/\lambda(0)$ in SCCO film for two temperature regions: (a) $T > 0.22T_C$ and (b) $T < 0.22T_C$. The dotted line shows a strong-coupling BCS behavior with $\Delta_0 = 2.07k_B T_C$. 