Excess Conductivity of Anisotropic Inhomogeneous Superconductors Above the Critical Temperature

T. I. Mogilyuk\textsuperscript{a, *}, P. D. Grigoriev\textsuperscript{b, c, d}, K. K. Kesharpu\textsuperscript{c}, I. A. Kolesnikov\textsuperscript{c}, A. A. Sinchenko\textsuperscript{e, f}, A. V. Frolov\textsuperscript{f}, and A. P. Orlov\textsuperscript{f}

\textsuperscript{a} National Research Center Kurchatov Institute, Moscow, 123182 Russia
\textsuperscript{b} Landau Institute for Theoretical Physics, Russian Academy of Sciences, Chernogolovka, Moscow oblast, 142432 Russia
\textsuperscript{c} National University of Science and Technology MISiS, Moscow, 119049 Russia
\textsuperscript{d} Lebedev Physical Institute, Russian Academy of Sciences, Moscow, 119993 Russia
\textsuperscript{e} Moscow State University, Moscow, 119991 Russia
\textsuperscript{f} Kotel’nikov Institute of Radio Engineering and Electronics, Russian Academy of Sciences, Moscow, 125009 Russia

\textsuperscript{*} e-mail: 5taras@mail.ru

Received April 15, 2019; revised April 22, 2019; accepted April 24, 2019

Abstract—The theoretical model of conductivity of a layered anisotropic normal metal containing small superconducting ellipsoidal granules with an arbitrary ratio of semiaxes is developed. Calculation data obtained under two simple approximations (self-consistent and Maxwell) are compared. The results may be applied in the analysis of the observed temperature dependence of the conductivity anisotropy in various anisotropic superconductors with the superconducting phase emerging in the form of isolated superconducting granules. The temperature dependence of the electric resistance along and across the conducting layers above and near the superconducting transition temperature is studied experimentally for bridge structures of a varying thickness. It is demonstrated that this resistance and even the effective superconducting transition temperature depend strongly on the bridge thickness (i.e., the number of layers through which the electric current flows). Note that significant differences were observed only for the resistance across the layers.

Keywords: conductivity, superconductors, anisotropy, inhomogeneity

DOI: 10.1134/S1063783419090166

1. INTRODUCTION

The search for ways to raise superconducting transition temperature $T_c$ in superconducting materials is one of the major objectives in condensed-matter physics. The majority of high-temperature superconductors (HTSCs) are metal alloys or complex oxide ceramics. Copper oxides [1–4] and iron-based superconductors [5–7] are of particular interest, since $T_c$ in these materials is the highest under atmospheric pressure and may be adjusted within a certain range by altering the chemical composition and the doping level. Since the dopant deposition is nonuniform, the initial spatially homogeneous compound turns into a heterogeneous material. Due to this heterogeneity, superconducting properties are often observed first in isolated regions of materials at $T > T_c$, which is one of the possible scenarios of formation of a pseudogap in cuprate HTSCs [8]. As temperature $T$ gets closer to $T_c$, small superconducting regions grow in size and their phases become coherent. At $T = T_c$, the entire volume becomes superconducting. This type of inhomogeneous evolution of superconducting properties above $T_c$ was called the Gossamer superconductivity (heterogeneous superconductivity) [9]. The presence of this heterogeneous phase was verified in the experiments on magnetic response with scan SQUID microscopy [10]. It was demonstrated theoretically in [5, 11] that the spatial heterogeneity of a superconductor may raise $T_c$. It also triggers superconducting effects above $T_c$. The relation between spatial heterogeneity and high-temperature superconductivity needs to be understood in order to synthesize novel materials with higher $T_c$ values.

It has been suggested recently that the excess conductivity of a layered anisotropic heterogeneous compound, which is attributable to isolated superconducting islands, is also strongly anisotropic with the effect maximized in the least conducting direction [9, 12]. A quantitative description of this effect in the Maxwell–Garnett approximation was used to analyze experimental data on conductivity and diamagnetic response...
in FeSe [9, 12]. The results of conductivity calculations in the self-consistent (see a brief review in [14, 15]) and Maxwell approximations for spheroidal superconducting granules were compared in [13]. The Maxwell approximation was applied in [16] to an anisotropic system with ellipsoidal superconducting inclusions.

FeSe has the simplest chemical composition in a large family of iron-based high-temperature superconductors, but its electron structure and properties are rather intriguing. The temperature of its transition increases to 40 K under pressure [17]. The observation of superconductivity with $T_c > 100$ K in a FeSe monolayer on an SrTiO$_3$ substrate [18] holds much promise. Bulk FeSe has an inhomogeneous microscopic structure and $T_c \approx 8$ K [19], which makes it a suitable material for comparison with our model of an anisotropic heterogeneous conductor. Such a comparison with the results of calculations in the Maxwell approximation for spheroidal superconducting inclusions was performed in [12].

2. EXPERIMENTAL

The temperature dependence of resistance along and across the layers was studied experimentally for samples of various thickness. A significant dependence was observed only for the resistance across the layers. The novel finding is that the observed anisotropy of excess conductivity above $T_c$ depends strongly on the sample (bridge) thickness. Figure 1 demonstrates how one may adjust the superconducting FeSe temperature by altering the thickness of the mesostructure or the bridge cut out in a FeSe single crystal.

The upper black curve in Fig. 1 corresponds to a sample thickness of 300 nm. This parameter was not determined in the process of etching for the other curves, since it is rather hard to do this for short bridges. However, the lateral geometry of samples is the same: $2 \times 2 \mu$m. Therefore, the resistance varies proportionally to the sample thickness. A thickness of 40–60 nm is thus obtained for the lower blue curve. These values are still much larger than the lattice constant of FeSe. Interestingly, the smaller the thickness (resistance), the higher the observed superconducting transition temperature. Even more intriguing is the fact that such results are impossible to obtain in the Maxwell approximation (even if the superconducting phase fraction depends on the thickness).

3. THEORY

The self-consistent approximation with ellipsoidal superconducting inclusions is used in this study. The proposed theoretical model agrees with the Maxwell approximation in the limit of small concentration of the superconducting phase and has a simple physical representation [9, 12]. According to this representation, two interlayer transport channels exist in an anisotropic conductor containing a small volume fraction of isolated superconducting inclusions. The first channel is the current flowing in the direction of the electric field (across the layers). The second channel is the current flowing along the conducting layers from one superconducting inclusion to another; it flows from one layer to a neighboring one only through superconducting inclusions, bypassing the nonsuperconducting phase.

Let us consider a homogeneous anisotropic conducting medium with conductivities $\sigma_{xx}$, $\sigma_{yy}$, and $\sigma_{zz}$. The continuity equation for the electric current is written as

$$-\nabla j = \sigma_{xx} \frac{\partial^2 V}{\partial x^2} + \sigma_{yy} \frac{\partial^2 V}{\partial y^2} + \sigma_{zz} \frac{\partial^2 V}{\partial z^2} = 0,$$

where $j$ is the current density and $V$ is the electrostatic potential. The coordinate substitution

$$x = x', \quad y = \sqrt{\mu} y', \quad z = \sqrt{\eta} z',$$

where

$$\mu = \frac{\sigma_{yy}}{\sigma_{xx}}, \quad \eta = \frac{\sigma_{zz}}{\sigma_{xx}},$$

Fig. 1. Temperature dependence of electric resistance $R(T)$ and superconducting transitions in FeSe revealed by this dependence in structures with different numbers of layers. The lateral dimensions are the same: $2 \times 2 \mu$m. The resistance is directly proportional to the structure thickness, which is in turn proportional to the number of layers. The upper black curve corresponds to a thickness of 300 nm; the lower blue curve, to 40–60 nm.
transforms Eq. (1) into the Laplace equation

\[
\sigma_{xx}^{\text{me}} \left( \frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} + \frac{\partial^2 V}{\partial z^2} \right) = 0.
\] (4)

If the inclusions are ellipsoidal (i.e., nonspherical) with semiaxes \(a, b = \beta a, \) and \(c = \gamma a\) and their axes are aligned with the coordinate ones, the semiaxes of these ellipsoidal inclusions change to

\[
a_x = a_0 \beta \sqrt{\mu}, \quad a_y = a_0 \gamma \sqrt{\eta}
\] (5)

after the coordinate transformation to the isotropic medium.

Next, we apply Eq. (18.19) from [15] to determine effective conductivity \(\sigma_e\) in the self-consistent effective medium approximation for the isotropic system

\[
\phi_s(\alpha_s - \alpha_m) \sigma_m + \phi_m(\alpha_m - \alpha_s) \sigma_{\text{me}} = 0,
\] (6)

where \(\sigma_m = I \sigma_l (l = (s, m)), \sigma_{s, m}\) is the conductivity of superconducting (s) and metallic (m) phases (\(\sigma_{s, m} = \sigma_{ss}^m\)), \(\phi_{s, m}\) is the volume fraction of superconducting (s) and metallic (m) phases, and the effective tensors of conductivity \(\sigma_e, \) and depolarization \(A^e\) are diagonal:

\[
\sigma_e = \begin{bmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{bmatrix}, \quad A^e = \begin{bmatrix} A_x & 0 & 0 \\ 0 & A_y & 0 \\ 0 & 0 & A_z \end{bmatrix}.
\] (7)

The ellipsoid depolarization tensor (see Eq. (6) in [20] or Subsection 17.1.2 in [15]; in our opinion, coefficient 2 in formula (17.25) should be raised to power 1 instead of \(j\) for condition (17.26) of equality of the tensor trace to unity to be satisfied) has principal values

\[
A_i = \frac{a_i a_j a_k}{2} \int_0^{+\infty} \frac{dt}{(t + a_1^2)(t + a_2^2)(t + a_3^2)}.
\] (8)

Integrals (8) were calculated analytically in [16]. Substituting the values of \(\sigma_m, A^e, \sigma_e, \) and \(\phi_m = (1 - \phi_s)\) into Eq. (6) and simplifying it on the assumption that conductivity \(\sigma_e\) of superconducting granules tends to infinity, we find the relation for the conductivity tensor

\[
\frac{\sigma_{ii}}{\sigma_m} = \frac{1}{1 - \phi_s / A_i}.
\] (9)

Reverting from the isotropic medium coordinates back to the initial anisotropic coordinates, we derive the resulting relation for the conductivity along three principal axes of an anisotropic heterogeneous medium with ellipsoidal superconducting inclusions

\[
\frac{\sigma_{ii}}{\sigma_m} = \frac{1}{1 - \phi_s / A_j}.
\] (10)

The obtained formulas contain an indirect temperature dependence mediated by the temperature dependence of superconducting phase fraction \(\phi_s.\) It may be derived from the diamagnetic response data (see, e.g., [9, 12, 16]). This \(\phi_s(T)\) dependence is shown in Figs. 3 and 4 in [12] for FeSe and in Fig. 3 in [16] for organic superconductor \(\beta_-(\text{BEDT-TTF})_2\text{I}_3;\) it remains similar to a power-law dependence in a wide range of temperatures \(T > T_c\) and drops sharply to zero at \(T \sim 5T_c\) (see Fig. 4d in [9]). Note that although these \(\phi_s(T)\) dependences for FeSe and \(\beta_-(\text{BEDT-TTF})_2\text{I}_3\) are qualitatively similar, they differ quantitatively, since function \(\phi_s(T)\) is defined by the properties and nature of inhomogeneity-inducing disorder, which differ from one compound to another (and even from one sample to another). If data on the diamagnetic response are not available, dependence \(\phi_s(T)\) may be determined using formulas (10) or (11) from the temperature dependence of excess conductivity along one axis (e.g., \(z\)). The temperature dependence along two other axes is derived from the obtained formulas and allows one to compare the results of model calculations with experimental data (see Fig. 2 in [16]).

4. DISCUSSION

Analytical expression (10) for the conductivity in anisotropic materials near the superconducting transition temperature with isolated superconducting inclusions was derived above within the classical model. Superconducting granules had ellipsoidal shapes and the same spatial orientation. The domain of applicability of the classical model was discussed in detail in [16]; it is applicable if granules are large compared to the superconductor coherence thickness or if the superconductivity proximity effects do not alter appreciably the shape of effective superconducting islands.

Equation (10) in the self-consistent approximation differs greatly from the expression

\[
\frac{\sigma_{ii}^n(\phi)}{\sigma_m} = 1 + \frac{\phi_s}{A_i(1 - \phi_s)}
\] (11)
derived [16] in the Maxwell approximation, although they agree in the \(\phi_s \ll A_i \ll 1 \) limit.\(^1\) Note that each of these two approximations (Maxwell and self-consistent) is accurate for a certain type of spatial inhomogeneity [15]. These approximations have several significant differences [15]. In our view, the most important is percolation threshold \(\phi_s^c,\) where the conductivity becomes infinite. In the Maxwell approximation, this threshold is the same in all directions: \(\phi_s^c = 1.\) In the self-consistent approximation, the value of \(\phi_s^c = A_i\)

---

\(^1\) \(A_i\) is always smaller than unity, since \(\sum A_i = 1\) and \(A_i > 0\) [19]. Therefore, condition \(\phi_s^c \ll A_i\) is more stringent than condition \(\phi_s^c \ll 1\) of applicability of the Maxwell approximation.
depends on the direction ($\phi$ along the lowest conductivity axis is much smaller). The possibility of existence of such an anisotropic percolation threshold in an infinite sample is debatable, although the results of several experiments [21] suggest that it exists in organic metal (TMTSF)$_2$PF$_6$. An anisotropic percolation threshold (superconducting transition) in a finite sample is possible, e.g., if the size of superconducting islands is comparable to one of the sample dimensions.

It should be noted that formulas (10) for the conductivity in an anisotropic inhomogeneous conductor in the self-consistent approximation and formulas (11) derived earlier in the Maxwell approximation do not provide a direct explanation for the observed dependence of the effective superconducting transition temperature on the sample thickness. However, the superconducting transition (percolation threshold), which defines transition temperature $T_c$, may shift relative to that in another direction and depends on the shape of superconducting inclusions. Therefore, if we assume that the shape of superconducting islands varies with thickness of the mesostructure, a probable explanation for the observed effect may be proposed. The observed dependence of the effective superconducting transition temperature on the sample thickness may also be explained in the self-consistent approximation by assuming that the volume fraction of the superconducting phase is higher in a thinner sample (due to surface effects). However, this assumption also requires microscopic justification.

**FUNDING**

T.I. Mogilyuk acknowledges support from the Russian Foundation for Basic Research (grant nos. 18-02-01022, 18-02-00280, 18-32-00205, and 19-02-01000). The work of P.D. Grigoriev was supported by state task no. 0033-2019-0001 “Development of Theory of Condensed Matter.” This study was supported in part by the BASIS Foundation for the Advancement of Theoretical Physics and Mathematics. A.A. Sinchenko acknowledges support from the Russian Foundation for Basic Research (grant no. 17-29-10007). The work of A.V. Frolov and A.P. Orlov was supported by the state task for the Kotel’nikov Institute of Radio Engineering and Electronics.

**CONFLICT OF INTEREST**

The authors declare that they have no conflicts of interest.

**REFERENCES**

1. K. Jung, Met. Mater. 2, 219 (1996).
2. G. Blumberg, M. Kang, M. V. Klein, K. Kadowaki, and C. Kendziora, Science (Washington, DC, U. S.) 278, 1427 (1997).
3. K. A. Müller, J. Phys.: Condens. Matter 19, 251002 (2007).
4. B. Keimer, S. A. Kivelson, M. R. Norman, R. Uchida, and J. Zaanen, Nature (London, U.K.) 518, 965 (2015).
5. M. Rotter, M. Pangerl, M. Tegel, and D. Johrendt, Angew. Chem. 47, 7949 (2008).
6. J.-H. Chu, J. G. Analytis, C. Kucharczyk, and I. R. Fisher, Phys. Rev. B 79, 014506 (2009).
7. Q. Si, R. Yu, and E. Abrahams, Nat. Rev. Mater. 1, 16017 (2016).
8. V. Z. Kresin, Y. N. Ovchinnikov, and S. A. Wolf, Phys. Rep. 431, 231 (2006).
9. A. A. Sinchenko, P. D. Grigoriev, A. P. Orlov, A. V. Frolov, A. Shakín, D. A. Chareev, O. S. Volkova, and A. N. Vasiliev, Phys. Rev. B 95, 165120 (2017).
10. I. Iguchi, T. Yamaguchi, and A. Sugimoto, Nature (London, U.K.) 412, 420 (2001).
11. I. Martin, D. Podolsky, and S. A. Kivelson, Phys. Rev. B 72, 060502 (2005).
12. P. D. Grigoriev, A. A. Sinchenko, K. K. Kesharpū, A. Shakín, T. I. Mogilyuk, A. P. Orlov, A. V. Frolov, D. S. Lyubshin, D. A. Chareev, O. S. Volkova, and A. N. Vasiliev, JETP Lett. 105, 786 (2017).
13. K. K. Kesharpū, P. D. Grigoriev, D. I. Lazeva, and T. I. Mogilyuk, J. Phys.: Conf. Ser. 1238, 012010 (2019).

https://doi.org/10.1088/1742-6596/1238/1/012010.

14. R. Landauer, AIP Conf. Proc. 40, 2 (1978).
15. S. Torquato, Random Heterogeneous Materials: Microstructure and Macroscopic Properties (Springer, New York, 2002).
16. S. S. Seidov, K. K. Kesharpū, P. I. Karpov, and P. D. Grigoriev, Phys. Rev. B 98, 014515 (2018).
17. S. Medvedev, T. M. McQueen, I. A. Troyan, T. Pałaszyk, M. I. Eremets, R. J. Cava, S. Naghavi, F. Casper, V. Ksenofontov, G. Wortmann, and C. Felsner, Nat. Mater. 8, 630 (2009).
18. J. F. Ge, Z. L. Liu, C. Liu, C. L. Gao, D. Qian, Q. K. Xue, Y. Liu, and J. F. Jia, Nat. Mater. 14, 285 (2015).
19. Yu. G. Naidyuk, G. Fuchs, D. A. Chareev, and A. N. Vasiliev, Phys. Rev. B 93, 144515 (2016).
20. S. Giordano, J. Electrostat. 58, 59 (2003).
21. N. Kang, B. Salameh, P. Auban-Senzier, D. Jerome, C. R. Pasquier, and S. Brazovskii, Phys. Rev. B 81, 100509(R) (2010).

Translated by D. Safin