Pis'ma v ZhETF, vol. 120, iss. 4, pp. 286 – 287

Synthesis and properties of 12442-family superconductor[1)]

I. V. Zhuvagin, V. A. Vlasenko, A. S. Usoltsev, A. A. Gippius, K. S. Pervakov, A. R. Prishchepa, V. A. Prudkoglyad, S. Yu. Gavrilkin, A. D. Denishchenko, A. V. Sadakov[2)]

Lebedev Physical Institute, Russian Academy of Sciences, 119991 Moscow, Russia

Submitted 6 June 2024
Resubmitted 25 July 2024
Accepted 26 July 2024

We report a synthesis of two members of recently discovered high-temperature superconductors of 12442 family, with formula $MCa_2Fe_4As_4F_2$ (M = Rb, K) and transition temperatures of 32.7 and 34.6 K, respectively. Quality of the samples was assessed using X-ray powder diffraction, superconducting transitions were identified through transport and magnetic experiments. The temperature dependence of the upper critical field and vortex activation energy was investigated under magnetic fields up to 19 T. Two distinct thermally activated flux flow regimes were observed in both systems. Field dependences of activation energy $U_0(H)$ indicate a change in the properties of vortex matter in these regimes and distinctly different dissipation mechanisms, reminiscent of cuprate HTSC.

DOI: 10.31857/S0370274X24080214, EDN: AWITRA

Iron-based superconductors (IBSCs) remain a dynamic field of research, particularly due to the discovery of new stoichiometric self-doped classes such as 1144 and 12442. This study presents a comprehensive examination of the synthesis pathways for novel iron-based superconductors within the 12442 class, alongside their characterization and an in-depth analysis of vortex states within the framework of thermally activated flux flow (TAFF) theory. Our findings reveal that the activation energy for the $KCa_2Fe_4As_4F_2$ and $RbCa_2Fe_4As_4F_2$ systems exhibits two distinct TAFF regimes, each characterized by unique field dependencies: a power-law behavior at higher temperatures and a logarithmic dependence at lower temperatures. This behavior indicates a dimensional crossover from three-dimensional to two-dimensional vortex liquid, reminiscent of high-temperature superconducting cuprates.

Experimental details. Transport measurements were conducted using Cryogenic CFMS-16 and 21 T systems, in magnetic fields up to 19 T. Magnetic ac-susceptibility characterization was carried out with MPMS XL-7 SQUID magnetometer. Current-voltage characteristics (IVCs) were obtained in a helium cryostat in the temperature range 2–40 K with variable temperature insert, ensuring temperature stabilization within ± 0.01 K, and using a custom-made low noise variable gain amplifier. For crystal structure and phase composition analysis by X-ray diffraction (XRD) we used a Rigaku MiniFlex 600 with Cu–K_α radiation in the 10–90 2θ angle region. The XRD patterns were analyzed employing the PDF-4+ database, Jana2006 software, and the Rietveld method for determining cell parameters.

Synthesis. Bulk samples of $KCa_2Fe_4As_4F_2$ and $RbCa_2Fe_4As_4F_2$ were synthesized using cryogenic mechanical grinding and solid-state reaction method. The initial reagents including K, Rb, Ca, As pieces, FeF_3 powder, and the powder of pre-synthesized FeAs were mixed in a stoichiometric ratio 3:6:2:2:10. The cell parameters, determined by XRD are: $a = 3.8656(4)$, $c = 30.997(4)$ Å with $R = 5.76\,\%$ (here, R is a commonly used measure of agreement between the amplitudes of the structure factors calculated from a crystallographic model and those from the original X-ray diffraction data) for the $KCa_2Fe_4As_4F_2$ sample and $a = 3.86978(18)$, $c = 31.6396(14)$ Å ($R = 7.67\,\%$), for the $RbCa_2Fe_4As_4F_2$ sample. Our XRD data is in a good agreement with the data reported in literature [1–3].

Results and discussion. The superconducting critical temperature values at zero magnetic field were found to be $T_c = 32.7$ K for $RbCa_2Fe_4As_4F_2$ and $T_c = 34.6$ K for $KCa_2Fe_4As_4F_2$. From resistive measurements in magnetic fields up to 19 T we obtained the temperature dependence of the upper critical field, $H_{c2}(T)$ (Fig. 1a, c). Using the single-band Werthamer–Helfand–Hohenberg (WHH) theory [4] in the dirty limit, we estimated the zero-field values of $H_{c2}(0)$ to be 97 and 129 T, for $RbCa_2Fe_4As_4F_2$ and $KCa_2Fe_4As_4F_2$ respectively.

A notable broadening of the superconducting transition suggests that there are significant thermal fluctuations in the system. The behavior of the vortex system, as described by the TAFF model [5], shows two

[1)]Supplementary materials are available for this article at DOI: 10.1134/S0021364024602021 and are accessible for authorized users.

[2)]e-mail: andrey.sadakov@gmail.com

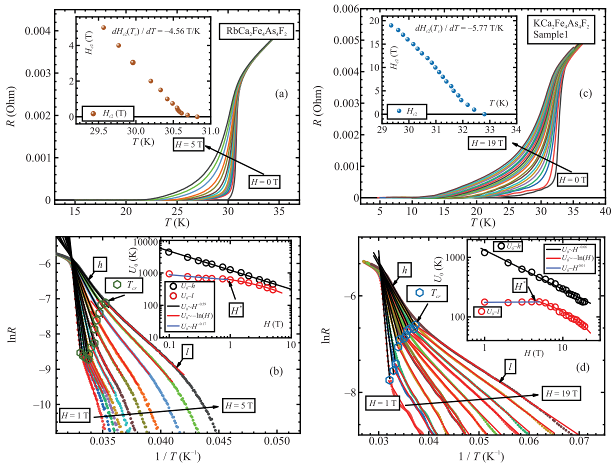

Fig. 1. (Color online) (a), (b) – Resistive transitions in different magnetic fields (inset shows the $H_{c2}(T)$ behavior) and Arrhenius plot (inset shows activation energy $U_0(H)$ field dependence) for $RbCa_2Fe_4As_4F_2$. Hexagons indicate crossover temperature from high (indicated by h) and low (indicated by l) temperature TAFF regions. (c), (d) – Resistive transitions in different magnetic fields (inset shows the $H_{c2}(T)$ behavior) and Arrhenius plot (inset shows activation energy $U_0(H)$ field dependence) for $KCa_2Fe_4As_4F_2$. Hexagons indicate the crossover temperature

linear regions in the $\ln\rho$ vs. 1/T plot, each with different slopes. Furthermore, the field-dependent activation energy $U_0(H)$, calculated from the Arrhenius plot, revealed two distinct dependencies on magnetic field: one following a power law and the other showing a logarithmic behavior (Figs. 1b, d). The presence of logarithmic behavior is commonly associated with low dimensionality in the vortex system and has been observed in various two-dimensional (2D) [6, 7] and quasi-2D systems [8]. We believe that the behavior of the activation energy is influenced by the appearance of edge dislocations or the emergence of vortex-antivortex pairs, similar to highly anisotropic cuprates.

There are two basic pinning mechanisms in type-II superconductors. The first is the pinning due to the randomly distributed spatial variations in the transition temperature T_c, commonly referred to as "δT_c pinning". The second pinning mechanism relates to spatial fluctuation of the charge-carrier mean free path, and is known as "δl pinning". These two pinning mechanisms demonstrate distinct dependencies of critical current on temperature in the single vortex-pinning regime. The temperature dependencies of the critical current density, $J_c(T)$, for the two studied systems exhibit similar shape and are more accurately characterized by the δT_c pinning model.

Conclusion. We presented a systematic synthesis of bulk samples of the 12442 phase using a combination of cryogenic mechanical grinding and solid-state reaction methods. The synthesis methods described in the literature yield satisfactory results only for compounds containing calcium in the $MCa_2Fe_4As_4F_2$ system. In contrast, compounds involving rare earth metals (such as Sm and Dy) could not be successfully synthesized using similar methods. The obtained superconductor critical parameters were consistent with those from other studies. The temperature-dependent vortex activation energy revealed two distinct thermally activated flux flow regimes in both systems. The field dependencies of the activation energy $U_0(H)$ suggest a shift in the characteristics of vortex matter in these regimes, indicating a dimensional crossover from 3D to 2D vortex liquid, reminiscent of copper-based high temperature superconductors.

Funding. This work was supported by the Russian Science Foundation (project # 23-12-00307).

Conflict of interest. The authors of this work declare that they have no conflicts of interest.

1. Z. C. Wang, C. Y. He, S. Q. Wu, Z. T. Tang, Y. Liu, A. Ablimit, C.-M. Feng, and G. H. Cao, J. Am. Chem. Soc. **138**(25), 7856 (2016); https://doi.org/10.1021/jacs.6b04538.
2. Z. Wang, C. He, Z. Tang, S. Wu, and G. Cao, Sci. China Mater. **60**, 83 (2017); https://doi.org/10.1007/s40843-016-5150-x.
3. X. Yi, M. Li, X. Xing, Y. Meng, C. Zhao, and Z. Shi, New J. Phys. **22**(7), 073007 (2020); https://doi.org/10.1088/1367-2630/ab9427.
4. N. R. Werthamer, E. Helfand, and P. C. Hohemberg, Phys. Rev. **147**, 295 (1966); https://doi.org/10.1103/PhysRev.147.295.
5. G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. **66**, 1125 (1994); https://doi.org/10.1103/RevModPhys.66.1125.
6. Z. Liu, C. Wang, C. Xu, M. Hao, H.-M. Cheng, W. Ren, and N. Kang, 2D Mater. **6**, 021005 (2019); https://doi.org/10.1088/2053-1583/ab0d2e.
7. C. Zhang, T. Hu, T. Wang, Y. Wu, A. Yu, J. Chu, H. Zhang, X. Zhang, H. Xiao, W. Peng, Z. Di, S. Qiao, and G. Mu, 2D Mater. **8**, 025024 (2021); https://doi.org/10.1088/2053-1583/abdaba.
8. H. Zhang, Y. Fang, T. Wang, Y. Liu, J. Chu, Zh. Li, D. Jiang, G. Mu, Z. Di, and F. Huang, Phys. Rev. B **103**, L180503 (2021); https://doi.org/10.1103/PhysRevB.103.L180503.