海外特別研究員最終報告書

独立行政法人 日本学術振興会 理事長 殿

採用年度 平成30年度 受付番号 201860128 氏 名 井口雄介

(氏名は必ず自署すること)

海外特別研究員としての派遣期間を終了しましたので、下記のとおり報告いたします。 なお、下記及び別紙記載の内容については相違ありません。

記

- 1. 用務地(派遣先国名)<u>用務地: スタンフォード大学 (国名: アメリカ合衆国)</u>
- 研究課題名(和文)<u>※研究課題名は申請時のものと違わないように記載すること。</u>
 走査型 SQUID 顕微鏡を利用した非従来型超伝導体の局所的超伝導状態の観察
- 3. 派遣期間: 平成 30 年 4 月 1 日 ~ 令和 2 年 3 月 31 日
- 4. 受入機関名及び部局名

Department of Physics and Applied Physics Stanford University, CA, USA

- 5. 所期の目的の遂行状況及び成果…書式任意 **書式任意 (A4 判相当 3 ページ以上、英語で記入も可)** (研究・調査実施状況及びその成果の発表・関係学会への参加状況等)
- (注)「6. 研究発表」以降については様式10-別紙1~4に記入の上、併せて提出すること。

Project title: Observation of local superconducting states in unconventional superconductors by using scanning SQUID microscopy

Scanning SQUID (Superconducting QUantum Interference Device) microscopy, which can obtain the local susceptibility by measuring an absolute value of magnetic flux, is very unique and strong scanning magnetic probe. The big purpose of this project is to reveal the local superconducting states of unconventional superconductors, such as chiral superconductor candidates, by using the scanning SQUID microscope. I have obtained the following results by measuring an iron-based superconductor FeSe, a chiral *d*-wave symmetry superconductor candidate URu₂Si₂, and a disordered charge density wave material Pd-intercalated ErTe₃.

- 1. In a nematic superconductor FeSe, the local vortex pinning potential has two-fold rotational symmetry along the twin boundary of the nematic domains.
- 2. In a chiral *d*-wave superconductor candidate URu₂Si₂, the local vortex pinning potential has four-fold rotational symmetry and two-fold rotational symmetry. The ferromagnetic impurity domain which appears at 16.1 K coexists with the superconducting phase. In the clean area, the local superfluid density has linear temperature dependence in low temperature, which suggests nodal gap structure such as chiral *d*-wave symmetry.
- 3. In a disordered charge density wave material Pd-intercalated ErTe₃, the superconducting phase transition temperature depends on the Pd concentration, but the superconducting superfluid response is homogeneous. The local superfluid density has non BCS-like temperature dependence.

[1] Imaging anisotropic vortex dynamics in FeSe

Iron-based superconductor FeSe shows the nematic phase transition at 90 K and the superconducting phase coexisting with the nematic phase below $T_c = 9$ K. The scanning SQUID microscopy has detected the increase of superfluid density around the twin boundary(TB) in the similar iron-based superconductor $Ba(Fe_{1-x}Co_x)_2As_2$. Here we observed the anisotropic vortex dynamics strongly depending on TBs. We studied the local vortex dynamics by using the scanning SQUID microscopy on the single crystal FeSe in the BLUEFORS dry refrigerator. The cleaved single crystals of FeSe were prepared by Prof. Ian R. Fisher's group. Figures 1(a) and (b) show the schematic image that our scanning SQUID microscope detects local dc magnetic flux ϕ , ac magnetic susceptibility M. These were obtained by applying ac magnetic field and include vortex dynamics. The measurement of an isolated vortex dynamics provides local vortex pinning potentials by simulating with a simple quadratic pinning potential $U(x,y) = 1/2(k_xx^2 + k_yy^2)$, where k_x and k_y are the spring constants. We observed the anisotropic susceptibility images over isolated vortices, which is consistent with the vortex dynamics with anisotropic pinning potential. The spring constants k_s and k_w ($k_s > k_w$) are in the order of >0.1 μ N/m and 0.01 μ N/m, respectively, which are three orders of magnitude weaker than that in Pb. We induced many vortices by cooling with a large magnetic field as shown in Fig. 1(c). The vortices were trapped along the TBs. The orientations of the anisotropic vortex dynamics were also along the TBs. Our results suggested that the scanning SOUID microscopy is very sensitive tool for the local rotational symmetry of vortex pinning potential.

We reported this result in the march meeting 2019 of the American Physics Society at Boston and published it in the peer reviewed journal Physical Review B (I.P. Zhang *et al.*, Phys. Rev. B 100, 024514(2019)).



Fig 1: (a),(b) Schematic image of scanning SQUID microscopy measuring local magnetic flux ϕ over an isolated vortex, local susceptibility M due to the vortex dynamics in anisotropic (a) and isotropic (b) local vortex pinning potentials U. (c),(d) Simultaneous large-area magnetic flux (c) and susceptibility (d) images showing vortices pinned on TBs and corresponding anisotropic vortex dynamics. Dashed lines mark the inferred domain boundaries. The orientations of the anisotropic vortex dynamics are oriented parallel to the TBs.

[2] Local observation of inhomogeneous superconductivity and ferromagnetism in heavy fermion superconductor URu₂Si₂

The heavy fermion superconductor URu₂Si₂ has been extensively studied to reveal the order parameters of the enigmatic hidden order phase (HO) ($T_{\rm HO} = 17.5$ K) and the coexisting unconventional superconducting phase (SC) ($T_c = 1.5$ K). The HO phase has the electron nematic order, but the tetragonal crystal structure is also weakly forced to transform into an orthorhombic in ultra-pure samples or at high hydrostatic pressure at $T_{\rm HO}$. On the other hand, recent studies have suggested that the chiral d-wave symmetry gap function could be considered as the most likely candidate to serve as the superconducting order parameter of URu₂Si₂. The nodal gap function was suggested by thermal conductivity and electron specific heat measurements. Spontaneous time-reversal symmetry breaking was observed by the polar Kerr effect measurement. Spin singlet state was suggested by Knight shift measurements and Pauli limit shown in higher critical field H_{c2} . Although the recent studies have been measured in high quality samples, there is no microscopic measurement at the superconducting phase in URu₂Si₂. We studied the local superfluid density and magnetism in the HO and SC phases by using the scanning SQUID microscopy on the single crystal URu₂Si₂ in the BLUEFORS dry dilution refrigerator. The cleaved single crystals of URu₂Si₂ were prepared by Dr. Eric D. Bauer and Dr. Filip Ronning. We have locally observed the isotropic and anisotropic vortex dynamics in the superconducting phase of single crystals of URu₂Si₂ by using our scanning SQUID microscope as shown in Fig. 2. Our results suggested that the vortex pinning potential had locally four-fold symmetry and two-fold rotational symmetry due to lattice defects, because it was not ordered like FeSe as shown in Fig. 2(e).



Fig 2: (a)-(e) Observed anisotropic (a) and isotropic (c) *M* over an isolated vortex at T = 1.0 and 1.2 K, respectively, where the color range is $0.7 \Phi_0/A$. Simulated *M* were obtained by using anisotropic $k_s = 107.7 \text{ nN/m}$, $k_w = 19.9 \text{ nN/m}$ (b), and isotropic k = 17.6 nN/m (d) for (a) and (c), respectively. (e) Local susceptibility image at T = 1.2 K. Open circles and double ended arrows indicate the isotropic and anisotropic vortex dynamics, respectively, which were observed in each position at 1 K in different cooling cycles.

In addition, we observed the inhomogeneous superconductivity and coexisting ferromagnetic domains as shown in Figs. 3(a)-(l). There are no apparent correlations between the ferromagnetic domains and superconductivity. We revealed that the ferromagnetic domains were localized, because the positive susceptibilities were only observed at the local area as shown in Fig. 3(e). We also obtained the temperature dependence of the local normalized superfluid density from the local penetration depth measurement as shown in Figs. 3(m) and (n). The obtained linear temperature dependence is consistent with the fits by semiclassical expression for *s*, $d_x^{2-y^2}$, and chiral *d*-wave symmetry gap functions and a multiband model (α -model) which includes chiral *d*-wave symmetry gap function. In particular, α -model provides the most similar temperature dependence to our results in high temperature region. Our results clearly suggested that URu₂Si₂ has the *d*-wave symmetry gap function including chiral *d*-wave.

I reported this result in the march meeting 2019 of the American Physics Society at Boston, the 75th annual meeting (2019) of the Physical Society of Japan at Kyushu university, and the 32nd International Symposium on Superconductivity 2019 at Kyoto as the invited talk. I am writing the manuscript to submit it to peer reviewed journals.



Fig 3: (a)-(l) Local susceptibility (a)-(f) and magnetic flux (g)-(l) images at T = 0.36-16.4 K at URu₂Si₂. (m) Temperature dependence of the penetration depth calculated from the fitting of the height dependence of susceptibility measured at P2 (blue triangles), P3(magenta triangles) and P4 (green circles). (n) Temperature dependence of normalized superfluid density estimated by using the penetration depths shown in (m) with $\lambda(0) = 0.95 \mu m$. Dash, dot, dashdot, and solid lines are the theoretical calculations by using *s*, d_x^2 - $_y^2$, and chiral *d*-wave symmetry gap functions and α -model, respectively.

[3] Inhomogeneity of superconductivity in a disordered charge density wave material Pdintercalated ErTe₃

The rare-earth tri-tellurides RTe₃ (R=La-Pr, Sm, Gd-Tm) are quasi-2D metals that show unidirectional incommensurate charge density wave (CDW) states. Pd intercalation introduces significant disorder to the crystal lattice, suppressing the CDW formation and leading to a superconducting ground state. The material presents an opportunity to explore in detail the interplay between superconductivity and CDW formation in the presence of disorder (see the phase diagram in Fig. 4(a)). Here we study the spatial variance of the superfluid response as a function of Pd concentration in Pd_xErTe_3 (0.003<x<0.054) by using a scanning SQUID microscope with sub-micron spatial resolution. We quantify the inhomogeneity of the emergent superconductivity as the CDW is progressively disordered by the Pd intercalation. We studied the local superfluid density in the SC phase by using the scanning SQUID microscopy on the single crystal Pd-intercalated ErTe₃ in the BLUEFORS dry dilution refrigerator. The cleaved single crystals of Pd-intercalated ErTe₃ were prepared by Prof. Ian R. Fisher's group. We observed quite homogeneous susceptibilities at x=0.003-0.054 as shown in Fig. 4(b). However the critical temperature, which were obtained by the temperature dependence of the superfluid density, depends on Pd concentration. The temperature dependences of the normalized superfluid density are not consistent with the isotropic s-wave symmetry and the BCS weak coupling. We need more theoretical study to understand our results, but. the larger superfluid density than BCS model at high temperature might imply that the Cooper pair has stronger attractive interaction.

I was supposed to report this result in the march meeting 2020 of the American Physics Society at Denver, but it was canceled due to the COVID-19 pandemic. I uploaded my slides on the abstract web page of the meeting to share our results. I am writing the manuscript to submit it to peer reviewed journals.



Fig 4: (a) Phase diagram of Pd-intercalated ErTe₃ obtained from (Ref) Phys. Rev. B 99, 235138(2019) and (this work) the temperature dependence of superfluid density. (b) Susceptibility images of x=0.3-5.4 % at T = 0.6-3.0 K. (c) Temperature dependence of normalized superfluid density estimated by using the penetration at x=0.3-5.4 % with $\lambda(0) = 0.7$ -1.0 µm. Dash line is the theoretical calculations by using *s*-wave symmetry gap function.