Exceptional Suppression of Flux-Flow Resistivity in FeSe$_{0.4}$Te$_{0.6}$ by Back-Flow from Excess Fe Atoms and Se/Te Substitutions

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We measured the microwave surface impedance of FeSe$_{0.4}$Te$_{0.6}$ single crystals with- and without external magnetic fields. The superfluid density exhibited a quadratic temperature dependence, indicating a strong pair-breaking effect. The flux-flow resistivity behaved as $\rho_f(B < B_{c2})/\rho_n = \alpha B / B_{c2}$. The observed $\alpha$ value of $\approx 0.66$ was considerably smaller than that of other Fe-based materials ($\alpha > 1$) and was attributed to the back-flow current produced by the disturbed motion of magnetic vortices by disorder. This is the first-time observation of the back-flow effect in multiple-band superconductors.

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Introduction—Following the discovery of superconductivity in LaFeAsO$_{1-x}$F$_x$ [1], Fe-based superconductors (Fe-SCs) have been extensively investigated worldwide. Fe-SCs exhibit multiple bands/gaps; thus, it has been predicted that the superconducting order parameter could change sign among different sheets of the Fermi surface [2, 3], and various gap structures have been observed experimentally [4]. To elucidate the mechanism of such novel and diverse SCs, the gap structure of each material should be systematically investigated, and the essential characteristics of the Fe-SCs should be extracted from the accumulated data.

In addition to the conventional probes that are sensitive to low-energy excitations, such as the temperature-dependent magnetic penetration depth, $\lambda(T)$, the magnetic field dependence of the flux-flow resistivity, $\rho_f(B)$, is known to be sensitive to the superconducting gap structure, since $\rho_f$ is induced by quasiparticles excited inside the vortex core reflecting the gap function. For most SCs, the flux-flow resistivity at low fields behaves as $\rho_f(B)/\rho_n \approx \alpha B / B_{c2}$, where $\rho_n$ and $B_{c2}$ are the normal state resistivity and the upper critical field, respectively. The structure of the superconducting gap is reflected in the gradient $\alpha$. Specifically, the $\alpha$ values of conventional SCs with an isotropic gap are almost unity [5], which are explained by the Bardeen-Stephen (B-S) theory [6]. However, unconventional SCs with p-wave [7], d-wave [8, 9], and anisotropic s-wave [10] symmetry exhibit $\alpha$ values above unity. Kopnin and Volokov (K-V) [11] justified the empirical relationship in which $\alpha$ increases with the anisotropy of the gap function by accounting for the bound states inside the vortex cores. A large $\alpha$ value has also been found for the $\rho_f(B)$ of two-band SCs [12, 13].

Novel phenomena have been predicted for multiple-band SCs such as the dissociation of a fractional flux quantum [14] and a time-reversal-symmetry-breaking state [15]. Thus, it is both interesting and significant to experimentally investigate the characteristics of the vortices of multiple-band SCs. To determine how novel features of Fe-SCs appear in the flux-flow, thus far, we have investigated the $\rho_f(B)$ of several Fe-based materials, such as LiFeAs (Li111) [17], LiFeAs$_{0.97}$P$_{0.03}$ [18], NaFe$_{0.97}$Co$_{0.03}$As (Co-Na111) [19], SrFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ (P-Sr122) [20], and BaFe$_2$(As$_{0.55}$P$_{0.45}$)$_2$ (P-Ba122) [21]. The primary contributions of these studies were that the observed $\alpha$s were significantly different from each other and that the gradient, $\alpha$, tended to increase when at least one highly anisotropic gap was present, which is somewhat similar to the behavior of single-band SCs. We recently confirmed this tendency in Li111 and P-Ba122 by quantitatively evaluating the relation between $\alpha$ and the gap anisotropy by extending the K-V model to two-band systems [22]. Based on those systematic studies for $\rho_f(B)$ of the Fe-SCs, the gap-anisotropy scenario is probably common to all of the Fe-SCs. However, the behavior of $\rho_f(B)$ for Fe-SCs with strong impurity scattering remains unclear because the existing flux-flow data for Fe-SCs have mostly been obtained for fairly clean materials, and there is no theoretical research as for the effect of strong disorder on vortices of multiple-band SCs. Although we have already determined that Co-Na111 exhibits gapless superconductivity, we have not determined the relation between the $\alpha$ value and the amount/strength of the impurity. To elucidate the role of impurity scattering for $\rho_f(B)$, we focused on the FeSe$_{1-x}$Te$_x$ system. It is well known that excess Fe atoms can enter Fe (II) sites easily and act as magnetic impurities [23, 24]. Therefore, FeSe$_{1-x}$Te$_x$ is an appropriate materials for investigating the $\rho_f(B)$ of Fe-SCs with strong impurity scattering.

In this letter, we report on the microwave surface impedance measurements of FeSe$_{0.4}$Te$_{0.6}$ single crystals both in the zero-field limit and under finite magnetic fields. The results for both $\lambda(T)$ and the parameter for the pinning of vortices indicated that FeSe$_{0.4}$Te$_{0.6}$ was a SC in the dirty limit. We observed that the $\alpha$ value of the $\rho_f(B)$ data was exceptionally small because of the considerable back-flow current that was generated in SCs with strong disorder.

Experiment—Single crystals of FeSe$_{1-x}$Te$_x$ were grown using a method that has been described elsewhere.

\[ \begin{align*}
\text{Exceptional Suppression of Flux-Flow Resistivity in FeSe}_{0.4}\text{Te}_{0.6} & \text{ by Back-Flow from Excess Fe Atoms and Se/Te Substitutions} \\
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\text{We measured the microwave surface impedance of FeSe}_{0.4}\text{Te}_{0.6} \text{ single crystals with- and without external magnetic fields. The superfluid density exhibited a quadratic temperature dependence, indicating a strong pair-breaking effect. The flux-flow resistivity behaved as } \rho_f(B < B_{c2})/\rho_n = \alpha B / B_{c2}. \text{ The observed } \alpha \text{ value of } \approx 0.66 \text{ was considerably smaller than that of other Fe-based materials (} \alpha > 1 \text{) and was attributed to the back-flow current produced by the disturbed motion of magnetic vortices by disorder. This is the first-time observation of the back-flow effect in multiple-band superconductors.} \\
\text{PACS numbers: 74.25.nn, 74.25.Ld, 74.70.Xa}
\end{align*} \]
A composition analysis using energy dispersive X-ray spectroscopy (EDX) was performed on samples with a nominal composition of Fe : Se : Te = 1 : 0.4 : 0.6. The corresponding actual ratios were found to be 1.00 ± 0.04 : 0.37 ± 0.05 : 0.63 ± 0.02. Henceforth, we denote this composition by FeSe$_{0.4}$Te$_{0.6}$.

Figure 1 shows the temperature dependence of the dc-resistivity, $\rho_{dc}(T)$, which was measured using the four-probe method: the superconducting transition appeared at $T_c \approx 14$ K. The residual resistivity of $\approx 300 \mu\Omega$cm was estimated by extrapolating the normal state resistivity. This result was consistent with our previous result $^{27}$ and much larger than that of clean Fe-SCs such as Li111 ($\approx 30 \mu\Omega$cm) and P-Sr122 ($\approx 50 \mu\Omega$cm), indicating strong impurity scattering in this material. We also determined the dc-magnetization using a superconducting quantum interference device magnetometer: the temperature-dependent dc-magnetic susceptibility, $\chi(T)$, indicated a bulk superconductivity of $T_c \approx 14$ K with a superconducting volume fraction of $\approx 100\%$ (which is not shown in this letter). To measure the surface impedance, single crystals were cut into a small piece with typical dimensions of $a \times b \times c = 0.5 \times 0.5 \times 0.2$ mm$^3$.

The microwave surface impedance $Z_s = R_s + iX_s$, where $R_s$ and $X_s$ denote the surface resistance and the surface reactance, respectively, was measured using the cavity perturbation technique $^{28}$ with a cylindrical oxygen-free Cu cavity resonator that was operated in the TE$_{011}$ mode. The resonant frequency and the quality factor (Q-factor) of the resonator, and the filling factor of the sample were $\omega/2\pi \approx 19$ GHz, $Q \gtrsim 6 \times 10^4$, and $F \approx 6 \times 10^{-6}$, respectively. Both an external field, $B_{dc}$, of up to 8 T, and a microwave field, $B_w$, were applied parallel to the $c$-axis of the sample (a schematic is shown in the inset of Fig. 2. The magnitude of $Z_s$ was determined by assuming the Hagen-Rubens limit in the normal state. The details of this procedure are described elsewhere $^{3, 17, 20, 28}$. The reproducibility of results described below was checked by measuring four specimens cut from different batches of single crystals.

We analyzed the flux-flow resistivity using the Coffey-Clem (C-C) model, where $Z_s$ induced by the vortex motion is calculated $^{29}$. The flux creep and the thermal fluctuations were negligibly small at sufficiently low temperatures; therefore, the C-C model yielded the relation $Z_s = -i\mu_0\omega\sqrt{1 + \rho_f(\mu_0\omega)^2}[1 - i\omega(T_c/\omega)^2]$, where $\mu_0$ is the vacuum permeability and $\omega(T_c/\omega)^2$ is the crossover frequency that characterized the crossover between the resistive response ($\omega > \omega(T_c)$) and the reactive response ($\omega < \omega(T_c)$). Consequently, at $T < T_c$, we could directly obtain $\rho_f(B, \omega)$, $\omega(T_c(B, \omega)$, and $\lambda(T, B = 0) = X_s(T, B = 0)/\mu_0\omega$ from $R_s(T, B)$ and $X_s(T, B)$.

Results and discussion— Figure 2 shows the temperature dependence of $\lambda^{-2}$, which was proportional to the superfluid density $\lambda(T)$, that was obtained from the data taken in the zero-field limit. It can be clearly seen that $n_s(T)$ changed as $n_s(T) \propto \lambda^{-2}(0) - A(T/T_c)^n$, with an exponent of $n \approx 2$. The two-dimensionality of the Fermi surface makes the existence of point nodes unlikely in FeSe$_{0.4}$Te$_{0.6}$. Thus, the $T^2$-dependence shows that gapless superconductivity was induced by the pair-breaking effect in this material. The results for the $T^2$-dependence and $\lambda(0) \approx 500$ nm were consistent with previous reports $^{21, 30}$.

Figure 3 shows that the crossover frequency, $\omega(T_c/2\pi)$, decreased as $B$ and $T$ increased. These behaviors were consistent with the conventional understanding that increasing the driving force and thermal fluctuations weaken vortex pinning and have also observed in other Fe-SCs $^{17, 18, 20}$. The observed value for $\omega(T_c/2\pi) \approx 30$ GHz was much larger than that observed in LaFeAsO$_{0.9}$F$_{0.1}$ ($\approx 6$ GHz) $^{31}$ and Li111 ($\approx 3$ GHz) $^{17}$, suggesting that FeSe$_{0.4}$Te$_{0.6}$ has a very strong pinning nature, which is quantitatively consistent with a large critical current density $^{32, 33}$.

Figure 4 shows the $B$-dependence of the flux-flow resistivity, $\rho_f(B)$, which was measured at $T = 2$ K. The vertical axis was normalized by the normal-state resistiv-
Using the value of \( T_c \) for fairly clean Fe-SCs are also shown as a comparison.

The upper critical field line in Fig. 1, and the horizontal axis was normalized by the superconducting region (which is shown as an orange cores occupy the entire sample, i.e., \( \alpha \) is defined by its value in the orbital limit where the vortex strongly affected by the Pauli paramagnetic effect, i.e., \( \alpha \) is value should be considered carefully because it is considerably different from previously reported values for other Fe-SCs, i.e., \( \alpha_{\text{Co-Na111}} \approx 1.1 \), \( \alpha_{\text{Li111}} \approx 1.4 \), \( \alpha_{\text{P-Sr122}} \approx 3.3 \), and \( \alpha_{\text{P-Ba122}} \approx 3.2 \). Previous flux-flow studies on cuprates, two-band systems, and Fe-SCs have shown that (i) the sign-change of the gap function is not essential for \( \rho_f(B) \approx \frac{\rho^{\text{orb}}(0)}{T} \), (ii) the multiple gaps of the SCs result in an \( \alpha \) value above unity \( 12–14, 21 \), and (iii) an anisotropic gap function also results in an \( \alpha \) value above unity \( 17, 21 \). Thus, the observed small gradient \( \alpha_{\text{FeSe0.4Te0.6}} < 1 \) is hard to be understood by these features.

One possible explanation for the small gradient value is the effect of disorder. The obtained results for (i) the large residual dc-resistivity, (ii) the \( T^2 \)-dependence of the superfluid density, and (iii) the large crossover frequency indicate that FeSe\(_{0.4}\)Te\(_{0.6}\) contains a large amount of disorder, even as single crystals. This characteristic is in sharp contrast to those of fairly clean Fe-SCs such as Li111, P-Sr122, and P-Ba122. Thus, we consider that this characteristic of FeSe\(_{0.4}\)Te\(_{0.6}\) causes the observed small \( \alpha_{\text{FeSe0.4Te0.6}} \). In fact, a small gradient (or a corresponding steep enhancement just below \( B_{c2} \)) of \( \rho_f(B) \) has been observed experimentally in superconducting alloys with a high concentration of disorder, such as Nb-Ta, Ti-V, Al-In, and Pb-In systems.

Several theoretical studies \( 40–46 \) have been conducted on the dissipation of the mixed state of single-band SCs with a high concentration of disorder. We highlight a series of calculations by Hu and Thompson \( 12, 43, 46 \) in which the time-dependent Ginzburg-Landau theory was used to model SCs with a high concentration of impurities, where the back-flow current around the vortex core was shown to be highly significant. Since the back-flow current decreases the electric field inside the vortex core, \( \rho_{\text{back}} \), the flux-flow resistivity, \( \rho_f = \rho_{\text{core}}/J \) (where \( J \) is the current applied to the sample), should be smaller than that predicted by the B-S model in which the back-flow current is neglected. Therefore, we believe that the smaller \( \alpha \) than the B-S prediction is due to the back-flow current and is a manifestation of SCs with large amount of disorder. Returning to the case of FeSe\(_{1-x}\)Te\(_x\), it is well known that excess Fe atoms act as magnetic impurities \( 23 \). Thus, it is expected that FeSe\(_{1-x}\)Te\(_x\) with excess Fe atoms behaves similarly to conventional SCs with paramagnetic impurities, and we consider that the observed small \( \alpha \) of FeSe\(_{0.4}\)Te\(_{0.6}\) also originates from the back-flow phenomenon. The magnetic vortex in multiple-band SCs is not understood even theoretically because of the complexity of the system. Therefore, this first experimental observation of the back-flow phenomenon in these SCs is highly significant.

\[ \text{FIG. 4. (Color online) The magnetic field dependence of the flux-flow resistivity of FeSe}_{0.4}\text{Te}_{0.6} \text{ (blue solid circle) measured at } T/T_c \approx 0.13 \text{. For comparison, the same plots of Li111 (gray open circle, } T/T_c \approx 0.11 \text{), P-Sr122 (gray solid circle, } T/T_c \approx 0.08 \text{), and the B-S's prediction (dotted line) are also shown.} \]
Finally, we consider the difference between Co-Na111 and FeSe0.4Te0.6. For a negligibly small back-flow, the gradient \( \alpha \) of these materials should be larger than unity because Co-Na111 has multiple bands with almost isotropic electronic states \(^{48}\), and FeSe0.4Te0.6 has multiple bands with moderately anisotropic nodeless gaps \(^{49, 50}\). In reality, the back-flow current Co-Na111 and FeSe0.4Te0.6 is not negligible, and we observed that the \( \alpha \) values of these materials were suppressed. These behaviors are shown in Fig. 8 as dashed lines (which correspond to the predicted behavior in the clean limit) and solid lines (which correspond to the experimentally observed behavior). Although both Co-Na111 and FeSe0.4Te0.6 exhibited gapless superconductivity, different \( \alpha \) values were observed for the two materials: \( \alpha_{\text{Co-Na111}} \approx 1 \) and \( \alpha_{\text{FeSe0.4Te0.6}} \approx 0.66 \). This difference could be attributed to the differences in the type and amount of impurities. In Ref. \(^{46}\), the \( \alpha \) small amount of magnetic impurities and a large amount of non-magnetic impurities in contrast to the weak suppression of \( \alpha_{\text{Co-Na111}} \) by a small amount of non-magnetic impurities (Co 3%). Although we do not as yet understand the explicit relationship between the amount of disorder of a sample and its \( \alpha \) value, this relationship could be elucidated by performing more systematic studies of \( \rho_f (B) \) for FeSe1-xTe_x with different amounts of excess Fe atoms and/or that of Co-Na111 containing magnetic impurities, such as Mn.

**Conclusions**—We measured the microwave surface impedance of FeSe0.4Te0.6 single crystals both in the zero-field limit and under finite magnetic fields. The superfluid density measured under a zero-external field behaved as \( n_s (T) - n_s (0) \propto (T/T_c)^2 \), indicating a strong pair-breaking effect in this material. The data obtained under finite magnetic fields showed that \( \omega_{c2}/2\pi \) for FeSe0.4Te0.6 was much larger than that of LiFeAs and of LaFeAs0.9Fe0.1, indicating very strong pinning in FeSe0.4Te0.6. The gradient of \( \rho_f (B \ll B_c) \) was \( \alpha_{\text{FeSe0.4Te0.6}} \approx 0.66 \), which is considerably smaller than that of other Fe-SCs (\( \alpha \geq 1 \)). We attributed this slow increase in the flux-flow resistivity to the back-flow current arising from the disturbed motion of vortices, which should provide valuable information on the understanding of vortices in multiple-band SCs.

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![Fig. 5](image-url) (Color online) Schematic image of \( \rho_f (B \ll B_c) \) of Co-Na111 and FeSe0.4Te0.6 (solid lines). The dashed lines are behaviors expected when the back-flow current is negligibly small, and the dotted lines are the B-S's prediction.

value of conventional SCs was calculated as a function of the spin-flip scattering rate \( \tau_s^{-1} \) and the total scattering rate \( \tau_1^{-1} \): if the pair-breaking by spin-flip scattering is not too strong, \( \alpha \) could be larger than unity when the total scattering rate is similar to that resulting from magnetic impurities \( (\tau_1^{-1} \approx \tau_s^{-1}) \) and becomes less than unity as the scattering rate by non-magnetic impurities becomes large \( (\tau_1^{-1} \gg \tau_s^{-1}) \). Although it is not clear whether these predictions are quantitatively valid, a similar trend is expected for multiple-band SCs. Recent scanning tunneling microscopy/spectroscopy studies on NaFe0.97-\( x \)Co0.03As \((T=\text{Cu, Mn})\) showed that Co atoms are non-magnetic or weak-magnetic impurities \(^{51}\), suggesting that the condition \( \tau_1^{-1} \approx \tau_s^{-1} \) is satisfied for Co-Na111. In contrast, excess-Fe atoms (i.e., corresponding to atomic concentrations below 4%) and doped Se/Te atoms (Se 37%, Te 63%) in FeSe0.4Te0.6 behaved as magnetic impurities and non-magnetic impurities, respectively. This finding most likely corresponds to the condition \( \tau_1^{-1} \gg \tau_s^{-1} \). Therefore, the strongly suppressed \( \alpha_{\text{FeSe0.4Te0.6}} \) may be attributed to the combination of a

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