Two-gap and paramagnetic pair-breaking effects on upper critical field of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals

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Abstract

We investigated the temperature dependence of the upper critical field [$H_{c2}(T)$] of fluorine-free SmFeAsO$_{0.85}$ and fluorine-doped SmFeAsO$_{0.8}$F$_{0.2}$ single crystals by measuring the resistive transition in low static magnetic fields and in pulsed fields up to 60 T. Both crystals show that $H_{c2}(T)$'s along the $c$ axis [$H_{c2}^c(T)$] and in an $ab$-planar direction [$H_{c2}^{ab}(T)$] exhibit a linear and a sublinear increase, respectively, with decreasing temperature below the superconducting transition. $H_{c2}(T)$'s in both directions deviate from the conventional one-gap Werthamer-Helfand-Hohenberg theoretical prediction at low temperatures. A two-gap nature and the paramagnetic pair-breaking effect are shown to be responsible for the temperature-dependent behavior of $H_{c2}^c$ and $H_{c2}^{ab}$, respectively.

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I. INTRODUCTION

The upper critical field, $H_{c2}$, is one of the most important superconducting parameters, providing a valuable insight into the pairing mechanism and information on fundamental superconducting properties such as coherence length scales. The temperature dependence of the upper critical field $H_{c2}(T)$ and its anisotropy are sensitive to the details of the underlying electronic structures and reflect the dimensionality of superconductivity. Furthermore, $H_{c2}$ is related to the critical current density, which is an important material parameter for application purposes.

After the discovery of iron-pnictides-based superconductors [1], REFeAsO$_{1-x}$F$_x$ (RE = rare-earth elements), with relatively high superconducting transition temperature ($T_c$), many efforts have been made to investigate their $H_{c2}(T)$. REFeAsO$_{1-x}$F$_x$ has a layered structure with alternating stacks of insulating REO and conducting FeAs layers. Despite the presence of the two-dimensional nature in the materials, most of studies on $H_{c2}(T)$ of REFeAsO$_{1-x}$F$_x$ have been limited to polycrystals [2, 3] because of the difficulty with growing REFeAsO$_{1-x}$F$_x$ single crystals. Therefore, recent investigations of single crystals have been more focused on AEFe$_2$As$_2$ (AE = alkaline-earth elements) [4, 5, 6] single crystals, which can be grown with relative ease in the ambient conditions. High-field measurements on electron- and hole-doped AEFe$_2$As$_2$ single crystals and films [7, 8, 9, 10, 11] showed that $H_{c2}(T)$ in a $c$-axis field [$H_{c2}^c(T)$] and in an $ab$-planar field [$H_{c2}^{ab}(T)$] increases almost linearly and sublinearly with decreasing temperature below $T_c$, respectively, regardless of the doping level and the degree of disorder. The resultant $H_{c2}^{ab}(T)$ approaches the value of $H_{c2}^c(T)$ at $T$ far below $T_c$, leading to the almost isotropic superconductivity in the zero-temperature limit. Such a quasi-isotropic property of $H_{c2}(T)$ in a layered structure is quite intriguing and has been attributed to the multi-band effect [9, 10, 11].

Recently single crystalline REFeAsO$_{1-x}$F$_x$ (RE = Sm, Nd, Pr) [12, 13, 14, 15] were successfully grown by using the flux-growth technique or the high-pressure-high-temperature technique. There is, however, a single report on the $H_{c2}(T)$ of REFeAsO$_{1-x}$F$_x$ single crystals for NdFeAsO$_{0.7}$F$_{0.3}$ at temperatures far below $T_c$, where $H_{c2}(T)$ was traced by high-field resistivity measurements using pulsed magnetic field up to 60 T [16]. $H_{c2}^c(T)$ in the study exhibited a pronounced upturn curvature at low temperatures [16]. By contrast, $H_{c2}^{ab}(T)$ showed a downturn curvature in the low-temperature range [16]. Apparently, the tempera-
ture dependence of $H_{c2}^c$ for NdFeAsO$_{1-x}$F$_x$, despite being common iron-ar senic compounds, appears to be quite different from that of AEFe$_2$As$_2$. Thus, it is highly required to examine any common or dissimilar $H_{c2}(T)$ behavior of different compounds of REFeAsO$_{1-x}$F$_x$, both in c-axis and in ab-planar fields, by adopting high-quality single crystals.

In this paper, we present $H_{c2}(T)$ for the magnetic fields along the c-axis ($H_{||c}$) and in the ab-plane ($H_{||ab}$) for single crystals of oxygen-deficient SmFeAsO$_{0.85}$ and fluorine-doped SmFeAsO$_{0.8}$F$_{0.2}$. $H_{c2}(T)$ were determined from the resistive transition in pulsed (static) magnetic fields up to 60 T (6.9 T). The sublinear increase of $H_{c2}^{ab}(T)$ with decreasing temperature below $T_c$, as previously seen in a NdFeAsO$_{0.7}$F$_{0.3}$ single crystal [16] and in AEFe$_2$As$_2$ compounds [7, 8, 9, 10, 11], was also observed in our crystals. On the other hand, $H_{c2}^c(T)$’s of our SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ crystals linearly increase with decreasing temperature near $T_c$ but tend to be saturated far below $T_c$. This temperature dependence of $H_{c2}^c$ is in contrast to the linear temperature dependence found in AEFe$_2$As$_2$ in all the temperature range below $T_c$ [7, 8, 9, 10, 11] and to the significant upturn behavior in NdFeAsO$_{0.7}$F$_{0.3}$ single crystals below $T_c$ [16]. A deviation of $H_{c2}(T)$ from the conventional one-gap Werthamer-Helfand-Hohenberg (WHH) prediction is found in our crystals. This feature, along with the reduction of its anisotropy with lowering temperatures, turns out to be common to iron-pnictides superconductors. Its detailed temperature dependence and the anisotropy, however, can be quite different depending on the compounds, which indicates the complex interplay of a multi-band nature and the paramagnetic effect.

II. EXPERIMENT

Single crystals of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ with nominal compositions were grown using the self-flux method under high-temperature and high-pressure condition. Stoichiometric starting compounds of SmAs, Fe$_2$O$_3$, and Fe for SmFeAsO$_{0.85}$ single crystals and SmAs, FeAs, Fe$_2$O$_3$, Fe, and SmF$_3$ for SmFeAsO$_{0.8}$F$_{0.2}$ single crystals were used. A pellet sealed in a boron-nitride container was placed in a cubic pyrophylite cell equipped with a carbon heater. A 14-mm cubic multi-anvil-type press was used to pressurize the whole assembly. Heat treatment at 1350-1450 °C was done for 8-10 h under a constant pressure of 3.3 GPa, which was then followed by rapid cooling to room temperature. Details of the single-crystal growth are described elsewhere [17]. After the pressure was released, the final
bulk was mechanically crushed to separate the single crystals from the flux.

Thus-grown crystals have plate-like shapes. X-ray diffraction reveals that the crystal surface is normal to the $c$-axis with the plate-shaped surface along the $ab$-plane. In-plane resistive transition of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals was measured using the standard four-probe technique. Contact leads were prepared by using photolithography on the plate-like sample surface. The upper insets of Figs. 1(a) and (b) show optical microscopic images of the four-probe patterned SmFeAsO$_{0.85}$ [dimensions: $\sim$80×50×10 $\mu$m$^3$] and SmFeAsO$_{0.8}$F$_{0.2}$ [dimensions: $\sim$60×50×10 $\mu$m$^3$] single-crystal specimens, respectively.

The resistive transition $[R(T)]$ was measured in low applied magnetic fields up to 6.9 T. Resistance as a function of fields $[R(H)]$ up to 60 T was also measured at different temperatures in pulsed-field facilities at Hochfeld-Magnetlabor Dresden. During the measurements, magnetic fields were applied along the $c$ axis and in the $ab$ plane while maintaining the current flow of 1 mA normal to the magnetic field.

III. RESULTS

As shown in the lower insets of Figs. 1(a) and (b), the superconducting transitions in zero field are very sharp for both SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ crystals. The onset of the superconducting transition, defined by the deviation from the linear $R(T)$ above $T_c$, occurs at about $T_{c,\text{onset}}$=50.5 K for SmFeAsO$_{0.85}$ and about 42 K for SmFeAsO$_{0.8}$F$_{0.2}$. The transition width $\Delta T_c$, determined by adopting the criterion of 10-90% of the normal-state resistance $R_n$, is $\sim$ 0.5 K for SmFeAsO$_{0.85}$ and $\sim$0.8 K for SmFeAsO$_{0.8}$F$_{0.2}$. $\Delta T_c$’s for both crystals are much narrower than the reported values of 2–4 K for single crystalline REFeAsO$_{1-x}$F$_x$ (RE = Sm, Nd) \[14, 15, 18\], indicating good quality of our samples. As Figs. 1 (a) and (b) show, the residual resistivity ratio $RRR\equiv\rho(300 \text{ K})/\rho(T_{c,\text{onset}})$ of $\sim$4.5 for SmFeAsO$_{0.85}$ is larger than the value of 2.5 seen previously for NdFeAsO$_{0.82}$F$_{0.18}$ single crystals \[14, 18\], while $RRR$ of SmFeAsO$_{0.8}$F$_{0.2}$ is $\sim$2.2, which is somewhat smaller but still comparable to that of the previous report. This indicates that the impurity scattering effect in our fluorine-free SmFeAsO$_{0.85}$ single crystal is less than the fluorine-doped REFeAsO$_{1-x}$F$_x$ single crystal. According to the recent report \[19\], fluorine does not fully substitute for oxygen and, thus, some oxygen vacancies remain in the crystal. The resulting additional scattering centers in REFeAsO$_{1-x}$F$_x$ may have enhanced the impurity scattering and led to a smaller $RRR$ value.
than in SmFeAsO$_{1-x}$.

Figures 2(a) and (b) present temperature dependence of resistance [$R(T)$] of SmFeAsO$_{0.85}$ single crystal in low magnetic fields from 0 to 6.9 T for $H||c$ and $H||ab$, respectively. The corresponding $R(T)$ of SmFeAsO$_{0.8}$F$_{0.2}$ single crystal is displayed in Figs. 3(a) and (b). Upon increasing magnetic fields, the resistive transition in $H||c$ becomes broader and the onset of superconductivity shifts to lower temperatures. The trend is more conspicuous in $H||c$ than in $H||ab$. These behaviors of $R(T)$ in low magnetic fields of $H||c$ and $H||ab$ in both crystals are similar to what was previously reported for SmFeAsO$_{0.7}$F$_{0.25}$ [15] and NdFeAsO$_{0.82}$F$_{0.18}$ [14, 16] single crystals. However, details of the magnetic field dependence are notably different from the previous observation. In our crystals, the resistive tail is more clearly observed, for $H||c$ in particular, with a gradual extension to lower temperatures with increasing fields [see Fig. 2(a) for SmFeAsO$_{0.85}$ and Fig. 3(a) for SmFeAsO$_{0.8}$F$_{0.2}$]. The tail of $R(T)$ was also observed in high $H||c$ in cuprates [20, 21, 22, 23] and in YNi$_2$B$_2$C [24], where both have layered structure with CuO$_2$ and Ni$_2$B$_2$ conducting planes, respectively. It has been known that such a resistive tail can be explained in terms of the vortex-glass phase [25]. Therefore, the observation of the $R(T)$ tail in our crystals indicates the possible formation of the vortex-glass phase for $H>2$ T. In the same magnetic-field region of $H||c$, the formation of vortex-liquid phase was also confirmed in NdFeAsO$_{1-x}$F$_x$ single crystals [26]. Oxygen vacancies in both of our SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ crystals may have acted as random intrinsic point defects and induced the vortex-glass state. Detailed analysis on the vortex dynamics in our crystals will be presented separately [27].

The magnetic-field dependence of resistance [$R(H)$] of our SmFeAsO$_{0.85}$ [Figs. 2(c)-(d)] and SmFeAsO$_{0.8}$F$_{0.2}$ [Figs. 3(c)-(d)] crystals was measured in pulsed magnetic fields (both $H||c$ and $H||ab$) up to 60 T at various temperatures. The upper critical fields, $H_{c2}^{c}$ for $H||c$ and $H_{c2}^{ab}$ for $H||ab$, were obtained by adopting different criteria; 90%, 50%, and 10% of $R_n$. The normal-state resistance $R_n$ was determined by linearly extrapolating the normal-state behavior above the onset of superconductivity in $R(T)$ and $R(H)$ curves separately. Thus-determined values of $H_{c2}^{c}$ and $H_{c2}^{ab}$ are shown in Figs. 4(a) and (b) for SmFeAsO$_{0.85}$ [in Figs. 4(c) and (d) for SmFeAsO$_{0.8}$F$_{0.2}$]. In both crystals, $H_{c2}(T)$ obtained from $R(T)$ (in Figs. 2 and 3) at low static magnetic fields (open symbols) is in line with those from $R(H)$ curves (in Figs. 2 and 3) at high pulsed magnetic fields (solid symbols). With lowering temperature, $H_{c2}^{c}(T)$ exhibits a slight upturn variation for the 10% criteria but it turns
gradually into a slight downturn curvature as one moves to 90% criteria, in particular for SmFeAsO$_{0.8}$F$_{0.2}$ sample. Similar behavior has also been observed in cuprates [28, 29, 30, 31, 32]. The criteria-dependent discrepancy arises from the fact that the region near 10% of $R_n$ is related to the vortex-liquid phase while the region near 90% of $R_n$ is affected by the superconducting fluctuation [29, 30, 31, 32]. Thus, we adopt the 50%-$R_n$ criterion to determine $H_{c2}(T)$. The resultant $H_{c2}(T)$ for $H_{\parallel c}$ and $H_{\parallel ab}$ is summarized in Fig. 5(a) for SmFeAsO$_{0.85}$ [in Fig. 5(b) for SmFeAsO$_{0.8}$F$_{0.2}$]. The values of $H_{c2}(T)$ for $H_{\parallel c}$ and $H_{\parallel ab}$ of our SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals are in the same range as reported for the corresponding polycrystals, with the similar value of $T_c \sim 50$ K and $\sim 40$ K, respectively, for the two crystals [3, 33].

According to a recent report on high-field resistivity measurements in a NdFeAsO$_{0.7}$F$_{0.3}$ single crystal up to 60 T [16], $H_{c2}(T)$ for $H_{\parallel c}$ exhibits a pronounced upturn in the entire ranges of magnetic field 60 T and temperature below $T_c \sim 45$ K. This result is similar to the earlier report for polycrystalline samples [2, 3], where the upturn shape of $H_{c2}^c(T)$ was suggested to be an intrinsic property of iron pnictides and was explained in terms of the two-band model [2, 3, 16]. However, our SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals show linear increase of $H_{c2}^c(T)$ with decreasing temperature near $T_c$ but tend to be saturated far below $T_c$ [see Figs. 5(a) and (b)]. This temperature dependence of $H_{c2}^c$ is in contrast to the linear and upturn temperature dependences in AEFe$_2$As$_2$ [7, 8, 9, 10, 11] and in NdFeAsO$_{0.7}$F$_{0.3}$ single crystals [16], respectively, below $T_c$.

IV. DISCUSSION

First, we compare the $H_{c2}(T)$ data of our crystals with the conventional WHH theory [34], which is based on the orbital effect arising from the Lorentz force acting on paired electrons with opposite momenta as the main cause of pair breaking. In addition, the theory is extended to include the effects of spin paramagnetism ($\alpha$) and spin-orbit scattering ($\lambda_{so}$). Here, we assume that the spin-orbit scattering due to impurities is negligible ($\lambda_{so}=0$) [35]. As shown in the Fig. 5, the data points of $H_{c2}(T)$ for $H_{\parallel c}$ and $H_{\parallel ab}$ in both crystals do not well follow the WHH model for $\alpha=0$ (solid lines). $H_{c2}^c(T)$’s for both crystals are enhanced compared to the WHH prediction, while $H_{c2}^{ab}(T)$’s are suppressed below the WHH curve with a flattening behavior.
Using the WHH theory for $\alpha=0$ and $\lambda_{so}=0$, we estimate the $H_{c2}$ value at $T=0$ [$H_{c2,WHH}(0)$]. $H_{c2,WHH}(0)\approx84~T$ [47 T] and $H_{c2,WHH}^{ab}(0)\approx378~T$ [280 T] for SmFeAsO$_{0.85}$ [SmFeAsO$_{0.85}$F$_{0.2}$] are obtained using the relation, $H_{c2,WHH}(0)=0.69T_c|dH_{c2}/dT|_{T_c}$. The values of $|dH_{c2}/dT|_{T_c}$ are presented in Table I. It is noteworthy to compare these $H_{c2,WHH}(0)$ values with the paramagnetic limiting field due to Zeeman paramagnetic pair breaking, $H_p(T=0)\approx(1+\lambda_{ep})H_{p}^{BCS}(T=0)$. Here, $H_{p}^{BCS}(T=0)=1.84T_c(H=0)$ [36] is the Pauli or Clongston-Chandrasekhar-limit field for isotropic s-wave pairing in the absence of spin-orbit scattering in weakly coupled superconductors. $\lambda_{ep}$ is introduced to take into account the strong electron-boson (i.e., phonon) coupling in the system. If we take $\lambda_{ep}=0.6$, $H_p(0)$‘s for SmFeAsO$_{0.85}$ and SmFeAsO$_{0.85}$F$_{0.2}$ are estimated to be about 145 T and 120 T, respectively. In both crystals, the values of $H_{c2,WHH}(0)$ are much smaller than the corresponding values of $H_p(0)$. This indicates that the $H_{c2}$ is determined dominantly by the orbital effect rather than the paramagnetic effect. By contrast, $H_{c2,WHH}^{ab}(0)$ is much larger than $H_p(0)$. In fact, the $H_{c2}^{ab}(T)$’s of both crystals have a tendency to be suppressed below the WHH curve for $\alpha=0$ and thus the actual $H_{c2}^{ab}(0)$ is expected to be much smaller than $H_{c2,WHH}^{ab}(0)$ estimated based on the paramagnetic effect. For $H_{c2}^{c}(T)$ where the orbital effect is dominant, on the other hand, its enhancement compared to the WHH curve with $\alpha=0$ cannot be explained in terms of the conventional one-gap WHH theory.

In order to understand the detailed temperature dependence of $H_{c2}(T)$, we consider the multi-band nature of iron-pnictides. It has been well-known that there are two different coexisting groups of Fermi surfaces: one with electron and the other with hole character [38, 39, 40, 41, 42]. Using the two-gap dirty-limit model of $H_{c2}(T)$ [43], we can fit the experimental data as shown in Figs. 6(a) and (b). The equation of $H_{c2}(T)$ for $H_{||c}$ considering orbital pair breaking is given by $a_0[\ln t+U(h)][\ln t+U(\eta h)]+a_2[\ln t+U(\eta h)]+a_1[\ln t+U(h)]=0$, where $t=T/T_c$, $U(x)=\Psi(1/2+x)-\Psi(x)$, $\Psi(x)$ is the Euler digamma function, $\eta=D_2/D_1$, $D_{1,2}$ are diffusivities of the bands 1 and 2, and $h=H_{c2}D_1/(2\phi_0T)$. $a_{0,1,2}$ are constants described with intraband- and interband- coupling constants $\lambda_{11,22}$ and $\lambda_{12,21}$ in the bands 1 and 2, respectively. Precise definitions of $a_{0,1,2}$ can be found in Ref. [43].

The equation of $H_{c2}(T)$ can be generalized to the case of a field inclined by angle $\theta$ with respect to the $ab$ plane by adopting angle-dependent diffusivities, $D_{1,2}(\theta)=[(D_{1,2}^{ab})^2\cos^2\theta+D_{1,2}^{ab}D_{1,2}^{c}\sin^2\theta]^{1/2}$ [43]. Therefore, $D_{1,2}$ are given by $D_{1,2}^{ab}$ for $H_{||c}$ and $[D_{1,2}^{ab}D_{1,2}^{c}]^{1/2}$ for $H_{||ab}$, where $D_{1,2}^{ab}$ ($D_{1,2}^{c}$) are the in-plane (out-of-plane) electron diffusivities of the bands 1 and
2. We discuss two different cases; (1) dominant intraband coupling \(w > 0\) and (2) dominant interband coupling \(w < 0\), where \(w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21}\). Here, we take three sets of \(\lambda\) for \(w > 0\) [(1) \(\lambda_{11} = 0.8, \lambda_{22} = 0.3, \lambda_{12,21} = 0.18\), (2) \(\lambda_{11,22} = 0.5, \lambda_{12,21} = 0.25\), (3) \(\lambda_{11,22} = 0.7, \lambda_{12,21} = 0.5\)] and two sets of \(\lambda\) for \(w < 0\) [(4) \(\lambda_{11,22} = 0.49, \lambda_{12,21} = 0.5\), (5) \(\lambda_{11,22} = 0.5, \lambda_{12,21} = 0.55\)]. Due to the lack of microscopic theory of pairing mechanism, we choose the values of \(\lambda\) close to the ones adopted in earlier reports [9, 16].

First, we consider the case of \(H_{||c}\). As shown in Fig. 6(a) for SmFeAsO\(_{0.85}\), the \(H_{c2}(T)\) predicted by the two-gap theory can reproduce nicely the experimental data taken up to 60 T for all cases. Depending on the sign of \(w\), however, the theoretical curves have different curvatures beyond the field range of measurements. Near \(T = 0\), the \(H_{c2}\) curves saturate to the values of \(H_{c2}^c(0) \sim 110\) T for (1) and \(\sim 135-142\) T for (2) and (3) in \(w > 0\), but still rapidly increase with upturn curvatures toward \(H_{c2}^c(0) \sim 220-300\) T for (4) and (5) in \(w < 0\). In these cases, \(\eta_{||c} = D_2^{ab}/D_1^{ab}\) is in the range of \(\sim 5-9\) for \(w > 0\) and \(\sim 19-36\) for \(w < 0\). In contrast, for SmFeAsO\(_{0.8}\)F\(_{0.2}\), the different sets of fitting parameters lead to almost identical curves, well fitting the \(H_{c2}(T)\) data [Fig. 6(b)] with \(H_{c2}^c(0) \sim 50\) T, \(H_{c2}^{ab}(0) \sim 208\) T, and \(\eta_{||c} \sim 2.2-3.7\). This indeterminacy of the sign of \(w\) for a better fit to the \(H_{c2}^c\) of SmFeAsO\(_{0.8}\)F\(_{0.2}\) may stem from the higher inhomogeneity of the crystal.

If we take into account the difference in the average Fermi velocities [44] of hole and electron sheets, the difference in the intraband diffusivities \(D_2^{ab} \sim (19-36)D_1^{ab}\) for \(w < 0\) looks too high. In addition, since \(H_{p}(0) \sim 145\) T was estimated for SmFeAsO\(_{0.85}\), the parameter set of (1) for \(w > 0\), which gives \(H_{c2}^c(0) \sim 110\) T, is more reasonable to explain the experimental data. Thus, in the reasonable range of \(\eta_{||c}\), the \(H_{c2}^c(T)\) curves of both crystals do not show the pronounced upturn behavior in the whole field and temperature range. This is somewhat different from the result [10] of NdFeAsO\(_{0.7}\)F\(_{0.3}\) single crystal, showing the significant upturn of \(H_{c2}^c(T)\) at the field up to 60 T. In Ref. [10], such a pronounced upturn of \(H_{c2}^c(T)\) is explained in terms of a two-band model, assuming a large difference in \(D_2^{ab}\) and \(D_1^{ab}\) with \(\eta_{||c}\) \(\sim 10-100\). It is not clear yet whether such a huge difference in \(D_2^{ab}\) and \(D_1^{ab}\) is intrinsic. As pointed out in Ref. [10], the strong upturn in \(H_{c2}^c(T)\) can be due to scatterings at magnetic impurities. In any case, the strong deviation from the WHH model is a common feature of iron-pnictides, which reflects the multi-gap nature of the materials.

Next, we consider the case of \(H_{||ab}\). In both crystals, the various sets of fitting parameters using the two-gap model lead to an identical curve of \(H_{c2}^{ab}(T)\). As shown in Figs. 6(a)
TABLE I: Superconducting parameters of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals obtained from the analysis of $H_{c2} (T)$. The $c$-axis and the $ab$-plane coherence length, $\xi_c (0)$ and $\xi_{ab} (0)$, respectively, are estimated with the Ginzburg-Landau relations for the upper critical field of $H_{c2}^c = \Phi_0 / 2 \pi \xi_c^2 (0)$ and $H_{c2}^{ab} = \Phi_0 / 2 \pi \xi_{ab} (0) \xi_c (0)$.

|          | $T_{c,\text{onset}}$ | $\frac{dH_{c2}^c}{dT} |_{T_c//c}$ | $\frac{dH_{c2}^c}{dT} |_{T_c//ab}$ | $H_{c2}^c (0)^d$ | $H_{c2}^{ab} (0)^e$ | $\xi_{ab} (0)$ | $\xi_c (0)$ |
|----------|----------------------|-----------------------------|-----------------------------|----------------|----------------|----------------|----------------|
| SmFeAsO$_{0.85}$ | 50.5 | 2.5 | 11 | 110 | 150 | 17 | 3.6 |
| SmFeAsO$_{0.8}$F$_{0.2}$ | 42 | 1.7 | 9.9 | 50 | 100 | 26 | 3.6 |

$^dH_{c2}^c (0)$ is determined from the analysis with two-band model.

$^eH_{c2}^{ab} (0) \equiv H_{c2,WHH}^p (T = 0)$ is estimated with the WHH theory including paramagnetism.

and (b), the fitting curves of two-band model cannot capture the flattening behavior with decreasing temperatures in both crystals. As discussed above, for $H_{||ab}$ we expect that the paramagnetic limiting plays an essential role for determining $H_{c2}^{ab} (T)$. In the framework of the WHH theory, such a spin-paramagnetic effect can be taken into account by introducing the so-called Maki parameter, $\alpha$. With $\alpha=2.3$ and 2.7, the $H_{c2}^{ab} (T)$ data of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ crystals are nicely fitted by the WHH model [see Fig. 7]. It has been known that the Maki parameter $\alpha$ becomes larger as the system is disordered [35]. A slightly larger value of $\alpha$ for SmFeAsO$_{0.8}$F$_{0.2}$ than for SmFeAsO$_{0.85}$ is consistent with its smaller $RRR$ value. The values of $H_{c2,WHH}^p (0)$ obtained by considering the Pauli paramagnetism with $\alpha \neq 0$ in the WHH theory are estimated to be $\approx 150$ T for SmFeAsO$_{0.85}$ and $\approx 100$ T for SmFeAsO$_{0.8}$F$_{0.2}$. Since $H_{c2,WHH}^p (0) \geq H_p (0)$ in our crystals, the data of $H_{c2}^{ab} (T)$ are strongly affected by the spin paramagnetic effect rather than by the two-band nature.

Due to the quasi-two-dimensional Fermi-surface topology, for a $H_{||c}$, the cross-section of the Fermi-surface produces closed current loops that form vortices [43, 46, 47, 48, 49]. Thus, for $H_{||c}$, the orbital pair-breaking mechanism plays a dominant role in destroying the superconductivity in high magnetic fields. Thus, the two-gap theory, taking into account the orbital pair-breaking effect, well describes our $H_{c2}^c (T)$ data. For a $H_{||ab}$, however, closed loops cannot be easily formed because the cross-sectional area of the Fermi-surface is almost fully open [50] with negligible orbital effect, thus resulting in a rapid increase of $H_{c2} (T)$ near $T_c$. Therefore, the spin-paramagnetic effect is a more dominant factor in reducing the
increase rate of $H_{c2}^{ab}(T)$ with decreasing temperature in our crystals.

In Figure 8, the anisotropy of $H_{c2}^2$, $\gamma \equiv H_{c2}^{ab}/H_{c2}^c$, is plotted as a function of reduced temperature $t= T/ T_c$ for SmFeAsO$_{0.85}$ (circles) and SmFeAsO$_{0.8}$F$_{0.2}$ (diamonds). The value of $\gamma$ for SmFeAsO$_{0.85}$ (SmFeAsO$_{0.8}$F$_{0.2}$) crystal is in the range of about 3–6 (4–7), at the temperature region of $T= (0.75–1) T_c$. SmFeAsO$_{0.8}$F$_{0.2}$ has a somewhat larger $\gamma$ than SmFeAsO$_{0.85}$. These values are similar in magnitude to the ones reported in other REFeAsO$_{1-x}$F$_x$ (RE = Sm and Nd) single crystals [14, 15, 16, 18, 51, 52]. The $\gamma$ has temperature dependence, which is distinct from that of the conventional single-band superconductivity. The decreasing $\gamma$ with decreasing temperature in both crystals results from the enhanced $H_{c2}^c(T)$ and the suppressed $H_{c2}^{ab}(T)$ compared to the WHH for $\alpha=0$ as shown in Figs. 5 (a) and (b). Therefore, the temperature dependence of $\gamma$ originates from the combined effect of two-band nature and spin paramagnetism.

V. SUMMARY

This study reports on $H_{c2}^c(T)$ and $H_{c2}^{ab}(T)$ of fluorine-free SmFeAsO$_{0.85}$ and fluorine-doped SmFeAsO$_{0.8}$F$_{0.2}$ single crystals, investigated by measuring the resistive transition at high magnetic fields up to 60 T. In contrast to the strong upturn curvature of $H_{c2}^c(T)$ reported earlier in NdFeAsO$_{1-x}$F$_x$ single crystal, $H_{c2}^c(T)$’s in both of our crystals increase linearly with decreasing temperature near $T_c$ and tends to be saturated at low-enough temperatures. We confirm that the temperature dependences of $H_{c2}^c(T)$’s well follow the two-gap dirty-limit prediction while they deviate from the one-gap WHH prediction, regardless of inclusion of the spin-paramagnetic effect. On the other hand, $H_{c2}^{ab}(T)$ of our crystals show the downturn curvature, consistent with the earlier observation in NdFeAsO$_{1-x}$F$_x$ single crystal and AEFe$_2$As$_2$ compounds. The importance of paramagnetic effect on the downturn curvature in $H_{c2}^{ab}(T)$ has already been pointed out for NdFeAsO$_{1-x}$F$_x$ single crystal and AEFe$_2$As$_2$ compounds, but $H_{c2}^{ab}(T)$ data were analyzed only within the two-gap model [9, 10, 11, 16]. In this study, the temperature dependences of $H_{c2}^{ab}(T)$’s are analyzed in terms of two-gap model and the WHH theory including the paramagnetic effect. Our analysis clearly indicates that the flattening of $H_{c2}^{ab}(T)$ is governed mainly by the paramagnetic pair-breaking effect rather than the two-gap effect. This study shows that the upper critical field in Sm-based iron-pnictides is determined by the complex interplay of a two-band nature and the
paramagnetic effect depending on the direction of magnetic field application with respect to the crystal axes. We believe this is the generic characteristics of different families of iron-pnictide compounds.

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FIG. 1: Temperature dependence of the resistance $R(T)$ of (a) SmFeAsO$_{0.85}$ and (b) SmFeAsO$_{0.8}$F$_{0.2}$ single crystals in zero magnetic field. The upper insets of (a) and (b) show the microscopic images of the four-probe patterned crystals used for the transport measurements. The lower insets of (a) and (b) show a magnified view near the superconducting transition. The onset of superconducting transition, $T_{c, \text{onset}}$, defined by the deviation from the linearity of $R(T)$, is indicated in the lower insets.
FIG. 2: Temperature dependence of the resistance $R(T)$ of SmFeAsO$_{0.85}$ single crystal measured at the various static fields from 0 to 6.9 T for (a) $H_{||c}$ (0, 0.2, 0.4, 0.6, 0.8, 1, 1.5, 2, 3, 4, 5, 6, 6.9 T) and (b) $H_{||ab}$ (0, 0.5, 1, 2, 3, 4, 5, 6, 6.9 T). Field dependence of the resistance $R(H)$ of SmFeAsO$_{0.85}$ single crystal measured at various temperatures in pulsed magnetic fields up to 60 T for (c) $H_{||c}$ and (d) $H_{||ab}$. 
FIG. 3: Temperature dependence of the resistance $R(T)$ of SmFeAsO$_{0.8}$F$_{0.2}$ single crystal measured in the various static fields from 0 to 6.9 T for (a) $H_{//c}$ (0, 0.2, 0.4, 0.7, 1, 1.5, 2, 3, 4, 5, 6, 6.9 T) and (b) $H_{//ab}$ (0, 0.5, 1, 2, 3, 4, 5, 6, 6.9 T). Field dependence of the resistance $R(H)$ of SmFeAsO$_{0.8}$F$_{0.2}$ single crystal measured at various temperatures in pulsed magnetic fields up to 60 T for (c) $H_{//c}$ and (d) $H_{//ab}$.
FIG. 4: (color online) Temperature dependence of the upper critical fields $H_{c2}(T)$ of SmFeAsO$_{0.85}$ for (a) $H \parallel c$ and (b) $H \parallel ab$ and of SmFeAsO$_{0.8}$F$_{0.2}$ for (c) $H \parallel c$ and (d) $H \parallel ab$. $H_{c2}(T)$ values are extracted from 10%, 50%, and 90% of the normal-state resistance $R_n$ determined by the linear extrapolation above the onset of superconductivity of $R(T)$ (open symbols) and of $R(H)$ (closed symbols) curves.
FIG. 5: $H_{c2}(T)$ for $H_{\parallel c}$ (circles) and $H_{\parallel ab}$ (triangles) of (a) SmFeAsO$_{0.85}$ and (b) SmFeAsO$_{0.8}$F$_{0.2}$ extracted from 50% of $R_n$ shown in Figs. 4(a)-(d). The open and closed symbols indicate the values of $H_{c2}(T)$ obtained from $R(T)$ and $R(H)$ curves, respectively. The experimental data were analyzed in terms of the WHH theory, which is exhibited by solid lines without the spin-paramagnetic effect ($\alpha=0$).
FIG. 6: Analysis of $H_c^2(T)$ for $H_{||c}$ and $H_{||ab}$ of (a) SmFeAsO$_{0.85}$ and (b) SmFeAsO$_{0.8}$F$_{0.2}$ single crystals using the two-gap theory in different pairing scenario: for $w>0$, (1) $\lambda_{11}=0.8$, $\lambda_{22}=0.3$, $\lambda_{12,21}=0.18$, (2) $\lambda_{11,22}=0.5$, $\lambda_{12,21}=0.25$, and (3) $\lambda_{11,22}=0.7$, $\lambda_{12,21}=0.5$), and for $w<0$, (4) $\lambda_{11,22}=0.49$, $\lambda_{12,21}=0.5$, and (5) $\lambda_{11,22}=0.5$, $\lambda_{12,21}=0.55$, where $w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21}$. The best fit is obtained for different values of $\eta_{||c}=D_2^{ab}/D_1^{ab}$ in the inset and an identical value of $\eta_{||ab}=[D_2^{ab}D_2^{ab}/D_1^{ab}D_1^{ab}]^{1/2}=1$. 
FIG. 7: Analysis of $H_{c2}(T)$ for $H_{||ab}$ by using the WHH theory including spin-paramagnetic effect ($\alpha \neq 0$). The flattening of $H_{c2}(T)$ for $H_{||ab}$ of SmFeAsO$_{0.85}$ and SmFeAsO$_{0.8}$F$_{0.2}$ single crystals, which can be clearly observed by a deviation from the dotted lines of the WHH theory for $\alpha=0$, is well explained by the WHH theory for $\alpha=2.3$ and 2.7 (solid lines), respectively, rather than by the two-band model (dash-dotted lines).

FIG. 8: Anisotropy ratio of $H_{c2}$, $\gamma=H_{c2}^{ab}/H_{c2}^{c}$, as a function of reduced temperature $t=T/T_c$ for SmFeAsO$_{0.85}$ (circles) and SmFeAsO$_{0.8}$F$_{0.2}$ (diamonds) single crystals.