Investigation of the superconducting gap structure in $SrFe_2(As_{0.7}P_{0.3})_2$ by magnetic penetration depth and flux flow resistivity analysis

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We measured the microwave surface impedances and obtained the superfluid density and flux flow resistivity in single crystals of a phosphor-doped iron-based superconductor $SrFe_2(As_{1-x}P_x)_2$ single crystals ($x = 0.30$, $T_c = 25$ K). At low temperatures, the superfluid density, $n_s(T)/n_s(0)$, obeys a power law, $n_s(T)/n_s(0) = 1 - C(T/T_c)^n$, with a fractional exponent of $n = 1.5$–1.6. The flux flow resistivity was significantly enhanced at low magnetic fields. These features are consistent with the presence of line nodes on at least one band and modulated nodeless gap with deep minimum on the same bands and/or other bands. The remarkable difference observed in the superconducting gap structure between $SrFe_2(As_{1-x}P_x)_2$ and $BaFe_2(As_{1-x}P_x)_2$ in our experiments is important for clarifying the mechanism of iron-based superconductivity in the 122 system.

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The symmetry of the superconducting (SC) order parameter is closely related to the pairing interaction and is an important problem for iron-based superconductors.[1] While nodeless $s\pm$-wave symmetry mediated by antiferromagnetic (AF) spin fluctuation has been considered to be a very likely candidate,[2,3] it has been revealed that many compounds have line nodes in their SC gap function. P-substituted systems are especially interesting in terms of the effect of the local structure parameter on the SC gap function. Kuroki *et al.* suggested that line nodes appear in the SC gap function when the pnictogen height from the Fe-As plane decreases.[4,5] One can experimentally control the pnictogen height and induce superconductivity through the isovalent substitution of As by P in the 122 system. $BaFe_2(As_{0.67}P_{0.33})_2$ ($T_c \sim 30$ K) is one of the most intensively studied P-substituted compounds.[6] London penetration depth measurement[7] provides an evidence of line nodes as other P-substituted iron-based SCs, such as LaFePO (Ref. 8) and LiFeP,[9] and the presence of closed nodal loops in the flat region of the electron Fermi surface was proposed based on an angle-resolved thermal conductivity measurement.[10] On the other hand, it has been theoretically suggested that P substitution warps the outer hole Fermi surface and weakens the nesting between the hole and electron Fermi surfaces. This action reduces the two dimensionality of the spin fluctuation, which results in the appearance of three-dimensional nodes on the outer hole sheet.[5] Therefore, there is no consensus on the detailed structure of the nodes. Another important consideration is to determine why the P-substituted Ba122 compounds have rather high T_c values, even in the presence of nodes. Therefore, it is of critical importance to investigate the SC gap structure in other P-doped systems.

In this paper, we focus on a different 122 system, namely the Sr122 system. The parent compound $SrFe_2As_2$ exhibits a maximum transition temperature of $T_c^{\max} = 32$ K at high pressures ($P = 5$ GPa).[11] Around the optimum pressure, nuclear magnetic resonance measurements have revealed the SC/AF hybrid state and the development of spin fluctuation toward T_c.[12] The chemical substitution is also effective in inducing superconductivity.[13–16] We measured the microwave surface impedances of $SrFe_2(As_{1-x}P_x)_2$ single crystals ($x = 0.30$, $T_c = 25$ K) to investigate the SC gap structure. We found that the low-temperature superfluid density n_s and the flux flow resistivity ρ_f at low fields are consistent with the presence of a SC gap that has line nodes. However, remarkable differences from $BaFe_2(As_{0.67}P_{0.33})_2$ were also found in the details of the SC gap, which is important for clarifying the mechanism of iron-based superconductivity in the 122 system.

Single crystals of $SrFe_2(As_{1-x}P_x)_2$ were grown using the SrAs self-flux method. Energy-dispersive x-ray analysis revealed that P substituted 30% of the As sites, which corresponds to $x = 0.30$. Figure 1 shows dc resistivity and dc magnetic susceptibility as functions of temperature of our $SrFe_2(As_{0.7}P_{0.3})_2$ single crystals. The dc resistivity exhibits a T-linear dependence over a wide temperature range, similar to the previous reports.[6,17] The SC transition temperature determined from the zero resistivity temperature is $T_c = 25$ K. The sharp transition and perfect shielding indicate that the crystals are of high quality.

The surface impedance $Z_s = R_s - iX_s$, where R_s is the surface resistance and X_s is the surface reactance, was measured using a cavity perturbation technique. We used two kinds of cylindrical oxygen-free Cu cavity resonators, operating in the TE_{011} mode at 19 and 44 GHz, which had quality factors Q of ~60 000 (19 GHz) and 26 000 (44 GHz), respectively. A piece of the crystal was mounted on a sapphire rod and was placed in the antinode of the microwave magnetic field H_ω. The shielding current flows in the ab planes because H_ω is parallel to the c axis. In this technique, one measures the changes of Q and the resonant frequency f of the resonator that result from the introduction of a sufficiently small sample. The shifts in the inverse of Q and f are proportional to R_s and X_s, respectively. The absolute values of Z_s were obtained by assuming the Hagen-Rubens limit ($\omega\tau \ll 1$), $R_s = X_s = (\mu_0\omega\rho/2)^{1/2} = \frac{1}{2}\mu_0\omega\delta$, above the T_c ($\omega = 2\pi f$ is the angular frequency, τ is the quasiparticle scattering time, μ_0 is the permeability in vacuum, and δ is the skin depth).

In the SC state effective penetration depth ($X_s = \mu_0\omega\lambda_{\rm eff}$). $\lambda_{\rm eff}$ coincides with the London penetration depth λ_L in the

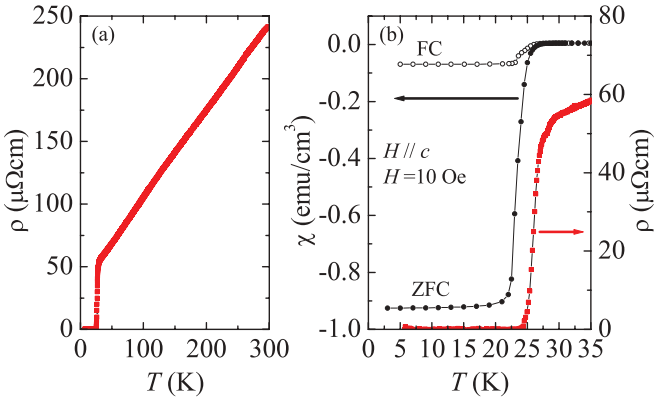

FIG. 1. (Color online) (a) The temperature dependence of the dc resistivity of a $SrFe_2(As_{0.7}P_{0.3})_2$ single crystal. (b) The dc resistivity and the dc magnetic susceptibility with both zero-field-cooled (ZFC) and field-cooled (FC) conditions at low temperatures.

clean local limit. Most iron-based superconductors are in this limit over the wide temperature range below T_c. Therefore, we can obtain the superfluid density $n_s(T)/n_s(0) = \lambda_L^2(0)/\lambda_L^2(T)$ via the London equation $\lambda_L^{-2} = \mu_0 n_s e^2/m^*$, which will provide information about the SC gap structure, particularly the presence or the absence of nodes in the gap.

Figure 2 shows the temperature dependence of microwave surface impedances of $SrFe_2(As_{0.7}P_{0.3})_2$. All crystals for Z_s measurements were cleaved from the same batch as that used for the dc resistivity and the dc magnetic susceptibility measurements, and formed so that they had dimensions about $0.5 \times 0.5 \times 0.1$ mm^3. The temperature dependence of R_s and X_s in the normal state are in good agreement with each other. Therefore, we were able to determine the absolute value of the London penetration depth with good reproducibility by

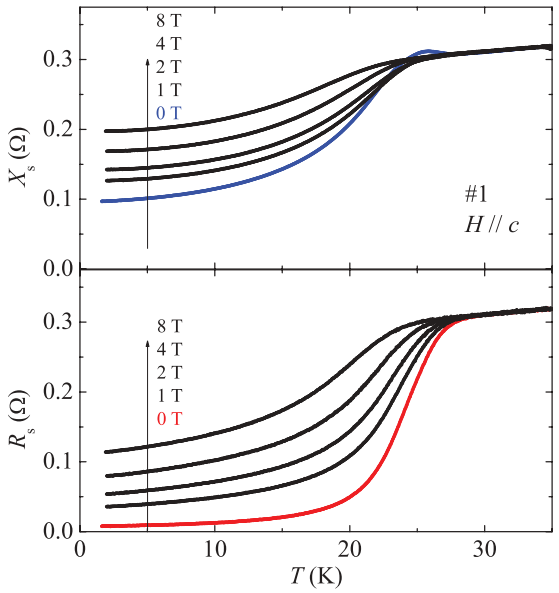

FIG. 2. (Color online) The temperature dependence of the microwave surface impedances of $SrFe_2(As_{0.7}P_{0.3})_2$ single crystals at 44 GHz. The hump appears at T_c in the X_s data because the characteristic field-penetration length changes from δ (in the normal state) to λ ($>2\delta$ in the SC state) within a narrow temperature range.

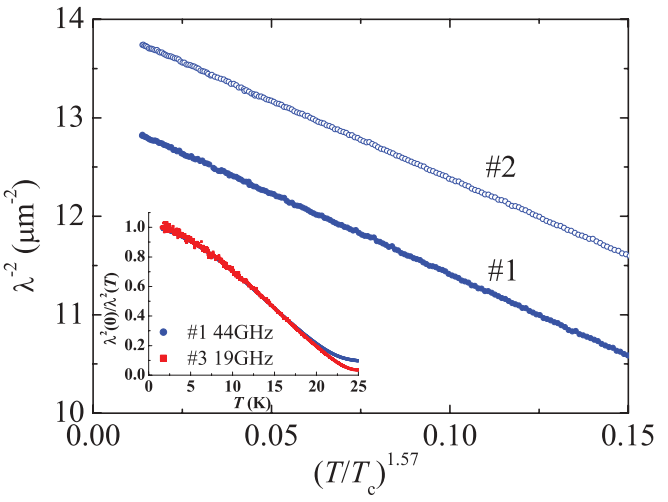

FIG. 3. (Color online) The magnetic penetration depth of $SrFe_2(As_{0.7}P_{0.3})_2$ single crystals at low temperature measured at 44 GHz. The data of crystal no. 2 are shifted vertically $+1$ μm^{-2} for clarity. The inset shows the temperature dependence of the superfluid densities $\lambda_L^2(0)/\lambda_L^2(T) = n_s(T)/n_s(0)$ measured at different frequencies.

assuming the Hagen-Rubens relation. The London penetration depth at 0 K is 270 ± 10 nm. This value is comparable to those of other optimally doped 122 compounds.[7,18] The superfluid density measured at two different frequencies (see the inset in Fig. 3) are in very good agreement, which indicates a good reliability of the absolute value of λ_L.

The temperature dependence of the superfluid densities, $n_s(T)/n_s(0)$, up to $0.3T_c$ is shown in Fig. 3. At low temperatures ($T \ll T_c/2$), $n_s(T)/n_s(0)$ is expressed for the nodeless superconductor as

$$\frac{n_s(T)}{n_s(0)} = 1 - \sqrt{\frac{2\pi\,\Delta_{\min}}{k_B T}}\exp\left(-\frac{\Delta_{\min}}{k_B T}\right), \qquad (1)$$

where $\Delta_{\min}$ is the minimum amplitude of the SC gap, and it is expressed for superconductors with line nodes, as

$$\frac{n_s(T)}{n_s(0)} = 1 - \alpha T, \qquad (2)$$

where α is a constant. We note that the discussion on the temperature dependence of n_s is essentially the same as that on $\delta\lambda_L(T) = \lambda_L(T) - \lambda_L(0)$ at low temperatures because $n_s(T)/n_s(0) \simeq 1 - 2\delta\lambda_L/\lambda_L(0)$ when $\delta\lambda_L/\lambda_L(0) \ll 1$. For $BaFe_2(As_{0.67}P_{0.33})_2$, T-linear $\lambda(T)$ is observed and it is considered to be an evidence of the presence of line nodes.[7] On the other hand, for $SrFe_2(As_{0.7}P_{0.3})_2$, the data were well fitted by a power law, $n_s(T)/n_s(0) = 1 - C(T/T_c)^n$ (C is a constant coefficient), with a fractional exponent, $n \simeq 1.6$, and n did not depend on the fitting temperature range. The power law has been observed in many iron-based compounds.[18] However, the values of the exponents are 2 or greater in most cases. For the case of $s\pm$-wave superconductors, the intrinsic exponential temperature dependence of $\lambda_L(T)$ changes to exhibit a power-law behavior when nonmagnetic impurity is introduced.[19] As impurity increases, the exponent decreases from $n > 3$ to $n \simeq 2$.[20,21] In other words, T^2 behavior appears

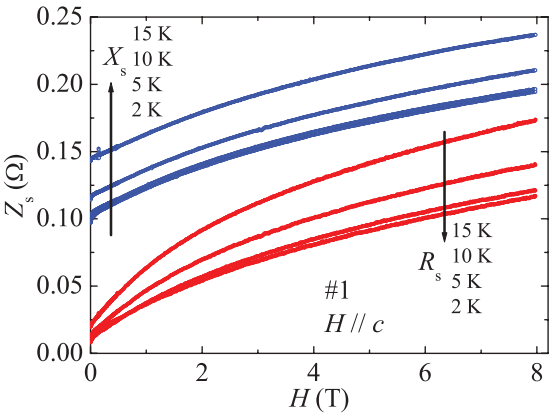

FIG. 4. (Color online) The magnetic-field dependence of the surface resistance R_s and the surface reactance Xs at 44 GHz under zero-field-cooled conditions.

in the disordered limit. However, the residual resistivity of approximately 50 $\mu\Omega$ cm in the $SrFe_2(As_{0.7}P_{0.3})_2$ is considerably smaller than that in other iron-based superconductors where T^2 behavior has been observed, for example in Ni-doped and Co-doped compounds.[20,22] This indicates that the exponent $n \simeq 1.6$ is induced by the intrinsic nature of the SC gap structure and not by impurity scattering. An exponent $n \simeq 1.6$, which is somewhat larger than the exponent values in other P-substituted systems, indicates that there is another channel of low-energy quasiparticle excitations in addition to the SC gap with line nodes.

To obtain further insights into the gap structure, we measured Z_s under finite magnetic fields and extracted the flux flow resistivity ρ_f. Figure 4 shows the magnetic-field dependence of Z_s at different temperatures. We performed experiments under both field-cooled and zero-field-cooled conditions and confirmed that there were no significant effects of vortex pinning on Z_s in a wide H range, except for very low fields below 0.5 T where a small degree of hysteresis appeared. In other words, the vortices are distributed homogeneously in the samples. We used the Coffey-Clem model to analyze Z_s.[23] In this model, at sufficiently lower temperature below T_c where the normal-fluid contribution and thermally excited flux creep are negligibly small, surface impedance in the mixed state ($H \gg H_{c1}$) can be expressed as

$$Z_s = -i\mu_0\omega\sqrt{\lambda_L^2 + (i/2)\tilde{\delta_f^2}(1 - i\omega_p/\omega)^{-1}}, \qquad (3)$$

where $\tilde{\delta_f} = \sqrt{2\rho_f/\mu_0\omega}$ is the flux flow skin depth. From this equation, we calculated the pinning frequency, ω_p, and ρ_f.

Figure 5(a) illustrates the ω_p of $SrFe_2(As_{0.7}P_{0.3})_2$. ω_p increases at low temperatures and at low fields, and it exceeds 30 GHz at 2 K and 1 T, which is considerably larger than the values of 6 GHz for $LaFeAsO_{0.9}F_{0.1}$ (Ref. 24) and 3 GHz for LiFeAs.[25] We can also estimate the vortex viscous drag coefficient $\eta = \mu_0 H\phi_0/\rho_f$. This parameter is related to the interval of discrete core bound states, $\omega_c \sim \Delta^2/E_F$ via $\eta = \pi\hbar n_c\omega_c\tau$, where τ is the quasiparticle relaxation time in the vortex core and n_c is the carrier density. $\omega_c\tau$ was observed to be on the order of 10^{-1}, which indicates that the vortex core is in the moderately clean regime.[26]

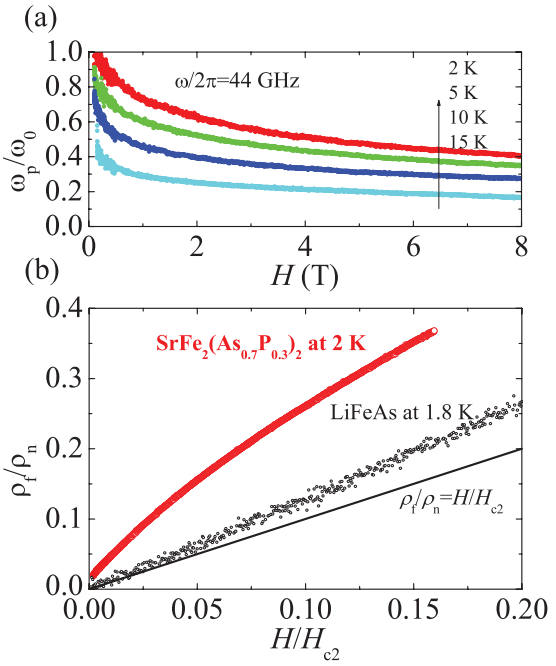

FIG. 5. (Color online) (a) The pinning frequency of $SrFe_2(As_{0.7}P_{0.3})_2$. (b) The magnetic-field dependencies of the flux flow resistivities of $SrFe_2(As_{0.7}P_{0.3})_2$ and LiFeAs (Ref. 25). The solid line indicates the BS relation, $\rho_f/\rho_n = H/H_{c2}$.

Figure 5(b) shows the magnetic-field dependence of ρ_f. It is clear that ρ_f exhibits a steeper increase at lower fields ($\rho_f/\rho_n > 2.5H/H_{c2}$ at $0.1H_{c2}$, where ρ_n is the resistivity in the normal state and H_{c2} is the upper critical field) than it does in the Bardeen and Stephen's (BS) model,[27] $\rho_f/\rho_n = H/H_{c2}$, and in the nodeless superconductor LiFeAs ($\rho_f/\rho_n \sim 1.3H/H_{c2}$).[25,28] Note that it is important to correctly evaluate $H_{c2}(2\text{ K})$; otherwise, ambiguity arises in the determination of the slope of $\rho_f(H)$. Here we use the value $H_{c2}(2\text{ K}) = 50$ T, because the H_{c2} of $SrFe_2(As_{0.7}P_{0.3})_2$ has been measured up to $H_{c2}(8\text{ K}) = 45$ T (Ref. 29) by other groups, which results in $H_{c2}(2\text{ K}) = 50$ T with the aid of Ginzburg-Landau theory. Because the $H_{c2}(T)$ of iron-based superconductors often exhibits a strong T dependence even at low temperatures, $H_{c2}(2\text{ K})$ may be greater than 50 T. In that case, the slope of ρ_f/ρ_n will be larger than that in our plot. Therefore, our plot provides the lower limit of the possible slope. Even if $H_{c2}(2\text{ K})$ is greater than 50 T, we can safely state that $\rho_f/\rho_n > 2.5H/H_{c2}$ at $0.1H_{c2}$.

According to the Kopnin and Volovik (KV) theory,[30] in the moderately clean regime, ρ_f can be expressed as

$$\frac{\rho_f}{\rho_n} = \frac{\omega_c^{\max}}{\langle\omega_c(\boldsymbol{k})\rangle_{FS}}\frac{H}{H_{c2}}, \qquad (4)$$

where $\langle\cdots\rangle_{FS}$ denotes an average over the Fermi surface. For an s-wave superconductor, this equation is reduced to the BS relation. However, when the SC gap has nodes, $\omega_c^{\max}/\langle\omega_c(\boldsymbol{k})\rangle_{FS}$ becomes larger than unity because ω_c depends on $\boldsymbol{k}$, which results in the enhancement of ρ_f. For example, $\rho_f/\rho_n \sim 2H/H_{c2}$ is observed in single-gap superconductors with line nodes.[31]

We would like to note two unusual features of ρ_f, which cannot be simply taken as the results of the presence of the line nodes confirmed by $\lambda_L(T)$. One is that the enhancement is considerably larger than that in the single-gap superconductor with line nodes. In the simplified phenomenological model of multigapped superconductors, quasiparticles excited at different Fermi surfaces contribute to the flux flow resistivity, similar to a parallel circuit, $\rho_f^{-1} = \rho_{f,1}^{-1} + \rho_{f,2}^{-1} + \cdots$.[32] Therefore, even if a nodal gap exists, the ρ_f associated with nodeless gaps should suppress the enhancement of the total ρ_f. The other unusual feature is the nonlinear H dependence. In both KV theory and the theory in Ref. 33, ρ_f is linear in H at low fields. In addition, we observed that $\rho_f \propto H$ in LiFeAs as shown in Fig. 5(b).[25] Nonlinear H dependence and a steep initial gradient are also found in ρ_f for MgB_2. MgB_2 has two distinct gaps where the gap amplitude ratio of the larger gap to the smaller gap is $\Delta_L/\Delta_S \sim 3$. Because the smaller gap is more sensitive to H, quasiparticles excited in the smaller gap rapidly increase with increasing H and enhance the flux flow resistivity at low fields.[33] Therefore, the contribution from the smaller gap saturates even far below H_{c2}. In such a situation, ρ_f may not be linear in H. Because $SrFe_2(As_{0.7}P_{0.3})_2$ is also a multigap superconductor, it is natural to conclude that the contribution from the very small gaps mentioned above produces unusual features. However, note that we cannot distinguish between the contribution from small gaps opening over an entire Fermi sheet and that from a deep gap minimum on a limited part of a Fermi sheet.

We now discuss the difference between $SrFe_2(As_{0.7}P_{0.3})_2$ and $BaFe_2(As_{0.67}P_{0.33})_2$ in terms of the low-energy quasiparticle excitation. The low-temperature $\lambda_L(T)$ exhibits different behaviors between these compounds: while $BaFe_2(As_{0.67}P_{0.33})_2$ exhibits nearly T-linear $\lambda_L(T)$,[7] we observed that $\lambda_L(T) \propto T^{1.6}$ in $SrFe_2(As_{0.7}P_{0.3})_2$. As we already discussed, both behaviors are consistent with the presence of a SC gap with line nodes. Therefore, we consider that the origin of the difference between these two compounds is related to the differences in the structure of the nodeless gaps. The significant differences in the low-energy quasiparticle excitation between these two compounds are also observed in the spin-relaxation rate and the heat capacity.[34] Both quantities exhibit a considerably larger residual density of states under magnetic fields in $SrFe_2(As_{1-x}P_x)_2$, which is consistent with the presence of the very small gaps that we observed in ρ_f. In the Sr122 system, additional quasiparticle excitations appear by replacing Ba^{2+} with Sr^{2+}. By this substitution, three dimensionality becomes more significant. It was suggested that three dimensionality can induce strong modulation of the superconducting gap along the k_z direction.[35] Therefore, we conclude that additional quasiparticle excitations observed in the above experiments are related to the strong modulation of the SC gap structure along the k_z direction rather than small gaps opening over a whole Fermi sheet. We also believe that the fractional power of $\lambda_L(T)$ is due to the combination of quasiparticle excitations in the nodal gap and the deep minimum in the nodeless gaps. In fact, Mishra *et al.*[36] calculated the magnetic penetration depth assuming the presence of three-dimensional nodes or horizontal nodes and found that n can be 1.5–2 when there is a nodeless gap with deep minimum on other Fermi surfaces.

In conclusion, we measured the microwave surface impedances of $SrFe_2(As_{0.7}P_{0.3})_2$ single crystals. The superfluid density at low temperatures obeys a power law, $n_s(T)/n_s(0) = 1 - C(T/T_c)^n$, with a fractional exponent of $n = 1.5$–1.6. The flux flow resistivity exhibits a very sharp increase with increasing H and a nonlinear field dependence. These results indicate the presences of line nodes on at least one band and modulated nodeless gap with deep minimum on the same bands and or other bands. We argued that the details of the SC gap structure depend on the size of the alkaline-earth ion between the Fe-As layers in the 122 system. This result indicates that the replacement of Ba^{2+} with Sr^{2+} mainly changes the part of the Fermi surface that does not significantly affect high T_c. In other words, it keeps the important part for realizing high-T_c superconductivity unchanged. Therefore, by comparing these two compounds in more detail, we will be able to clarify the mechanism of iron-based superconductivity.

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