# Effect of hydrostatic pressure on the superconducting transition temperature and superfluid density of $SmFeAsO_{0.85}$ and $PrFe_{0.925}Co_{0.075}AsO$ superconductors

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(Received 21 October 2010; published 13 December 2010)

We have measured magnetic susceptibility of iron pnictide superconductors $SmFeAsO_{0.85}$ and $PrFe_{0.925}Co_{0.075}AsO$ under hydrostatic pressure up to 1.15 GPa. The superconducting transition temperature ($T_C$) deceases linearly and the Meissner signal size also decreases with increasing pressure for $SmFeAsO_{0.85}$. In contrast, the $T_C$ of $PrFe_{0.925}Co_{0.075}AsO$ initially increases with pressure then saturates above ~0.8 GPa. Meanwhile its Meissner signal exhibits the similar pressure dependence. Our results indicate that the pressure dependences of $T_C$ and superfluid density in both systems are positively correlated which suggests that these quaternary iron-based superconductors are not conventional BCS ones.

DOI: 10.1103/PhysRevB.82.212506 PACS number(s): 74.62.Fj, 74.25.Ha, 74.70.Xa

Iron-arsenic superconductors have attracted much interest since the discovery of 26 K superconductivity in F-doped LaFeAsO.[1] The superconducting transition temperature $T_C$ was then quickly raised up to 55 K,[2–6] well exceeding the McMilan limit (about 40 K by weak-coupling BCS theory). First principle calculation suggests that the $T_C$ of multiband iron-based superconductor may be enhanced under pressure by increasing orbital degeneracy in the underdoping regime and suppressed at optimal-doping and overdoping regions due to the decrease in orbital degeneracy,[7] namely, based on the BCS theory the sign of pressure ($P$) coefficient of $T_C$, $dT_C/dP$, of the iron-based superconductors is due to the pressure-induced change in the density of state at the Fermi level. On the other hand, another multiband superconductor $MgB_2$ shows a negative pressure coefficient of $T_C$ which is attributed to the pressure-induced reduction in the electron-phonon coupling[8] within the BCS framework. Experimentally, a positive $dT_C/dP$ is found in optimally doped LaFeAs(O,F) for $P<4$ GPa and it then becomes negative for $P>4$ GPa.[9–11] A negative $dT_C/dP$ is also observed in optimally doped and overdoped SmFeAs(OF) samples.[12–14] In this work, we study the pressure effects on $T_C$'s and Meissner signals of iron-based superconductors $SmFeAsO_{0.85}$ and $PrFe_{0.925}Co_{0.075}AsO$ by magnetic measurements under pressures. Magnetic susceptibility measurement is capable of extracting information of the penetration depth, as the magnetic susceptibility is a function of penetration depth in granular superconductors.[15] Furthermore, since the square of penetration depth is inversely proportional to the ratio of superfluid density to effective mass ($n_s/m^*$), the pressure dependence of the susceptibility thus turns out to be an effective probe for monitoring the change in $n_s/m^*$ under pressure. Since the pressure dependences of both $T_C$ and Meissner signal can be simultaneously tracked for each given sample, the high-pressure study is superior to the doping dependence study due to being free of the possible sample complications resulting from the differences both in types and in quantities of the dopants and the difference in sample quality due to sample preparations. In other words, as a thermodynamics parameter, a moderate pressure generally tunes the electronic states in the system without introducing other extrinsic effects. We find that $SmFeAsO_{0.85}$ and $PrFe_{0.925}Co_{0.075}AsO$ exhibit the same pressure dependences of their $T_C$ and Meissner signal, i.e., either increasing or decreasing simultaneously with pressure. We argue that the positive or negative sign of the pressure coefficient of $T_C$ we observed in these two systems are closely related to the increase or decrease in the superfluid density under pressure. Therefore, our observation suggests that these iron-based superconductors are not of the conventional BCS type.

Nominal $SmFeAsO_{0.85}$ sample (Sm-55) with $T_C$ of 55 K, the highest record of $T_C$ observed in iron-based superconductors, was prepared under a high pressure of 6 GPa. Nominal $PrFe_{0.925}Co_{0.075}AsO$ (Pr-15) sample with $T_C$ of 15 K, the maximum $T_C$ obtained in $Pr(Fe_{1-x}Co_x)AsO$ system, was synthesized under ambient condition. The details of synthesis and characterizations were reported elsewhere.[16,17] The grains of both polycrystalline samples are randomly orientated and the grain size is of the order of about $14\times6\times\sim1\ \mu m^3$ and $20\times20\times1\ \mu m^3$ for Sm-55 and Pr-15,[17,18] respectively. The sample was mounted in a commercially available pressure cell (Mcell 10), which was specially designed for Quantum Design magnetic property measurement system (MPMS) for magnetic measurements under pressure. Hydrostatic pressure was loaded at room temperature and monitored *in situ* by recording the $T_C$ shift in a small piece of Sn mounted in the Mcell 10.[9] To confirm the reproducible pressure behavior of the samples, the pressure was applied in a nonmonotonic order: the loading sequence is of 0.28 GPa, 1.1 GPa, 0.82 GPa, 0.65 GPa, 0.39 GPa, and 0.07 GPa for Sm-55 sample and of 0.25 GPa, 0.80 GPa, 1.15 GPa, and 0.50 GPa for Pr-15 sample, respectively. Zero-field cooling (ZFC) and FC dc susceptibilities were measured under a field of 10 Oe using MPMS-XL1. Background signals from the pressure cell and holders were subtracted simultaneously during the measurements.[9]

The pressure-dependent superconducting transition of the optimally doped Sm-55 sample was tracked by dc susceptibility, $4\pi\chi=4\pi\,(M-M_{60\text{ K}})/H$, as shown in Fig. 1(a). Here $M$ denotes the magnetization, $M_{60\text{ K}}$ the magnetization at 60

 

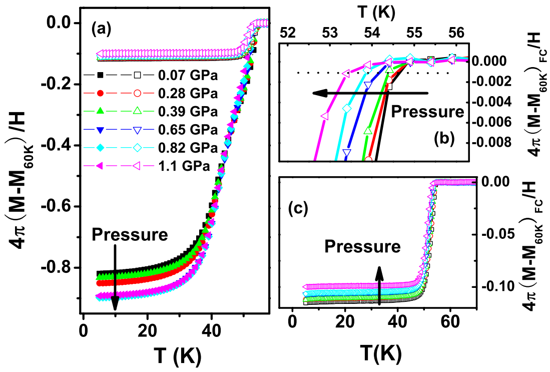


FIG. 1. (Color online) (a) ZFC (solid symbols) and FC (hollow symbols) susceptibilities as function of temperature under pressures for Sm-55 sample. (b) The enlargement of the FC susceptibility curves in the vicinity of $T_C$. The temperatures at points intersected by the dashed line are defined as the superconducting critical temperatures under various pressures in this study. (c) An expanded view of the FC susceptibilities as function of temperature under pressures for Sm-55.

K, and $H$ the applied field. The data in the vicinity of $T_C$ are expanded in Fig. 1(b). Opposite to the naive thinking that pressure would increase the carrier density via lattice shrinking so that it would enhance $T_C$, the superconducting transition shifts toward lower temperature with pressure, indicating a negative pressure coefficient of $T_C$. Here the superconducting transition temperature $T_C$ is defined as the temperature where the field cooling $4\pi\chi_{FC}$ decreases down to $-1\times10^{-3}$ [as indicated by the dashed line in Fig. 1(b)]. As plotted in Fig. 3(c), the $T_C$ decreases from 54.6 to 53.4 K with hydrostatic pressure increasing from 0.07 to 1.1 GPa, at an almost constant rate $dT_C/dP\approx-1.1$ K/GPa. Furthermore, the FC susceptibility below $T_C$ also shows a negative pressure dependence as indicated in Fig. 1(c) by the arrow. Illustrated in Fig. 3(d) is the curve of Meissner signal size taken as the FC susceptibility at 5 K, $4\pi\chi_{5\ K}$, which decreases linearly with increasing pressure at a rate of $d(4\pi\chi_{5\ K})/dP\approx-0.014$/GPa.

Now turn to the case of Pr-15 sample. We note that although both samples share the same crystal structure of 1111 family, the charge carriers in Pr-15 are directly introduced within FeAs layer by Co substitution while the carriers in Sm-55 sample are produced by O vacancy out of the FeAs layer. As mentioned above, the $T_C$ of Pr-15 is 15 K, which is the highest $T_C$ reachable in Co-doped PrFeAsO system indicating that this sample is optimally doped one. As shown in Fig. 2, the superconducting transition of Pr-15 shifts toward higher temperature under pressure. Plotted in Fig. 3(a) is the quantitative pressure dependence of $T_C$ where $T_C$ is determined in the same way as that for Sm-55 sample as shown in the inset of Fig. 2. It can be seen that the $T_C$ rises linearly when pressure increases from 0.25 to 0.8 GPa then it becomes saturated up to the highest pressure, 1.15 GPa, achievable in this work. One may find from Fig. 2 that the Meissner signal size is also somewhat lifted by hydrostatic pressure. Plotted in Fig. 3(b) are the data of Meissner signal taken as the FC susceptibility at 4.5 K. Interestingly, the Meissner signal also first rises from 0.16 to 0.17 with pressure then

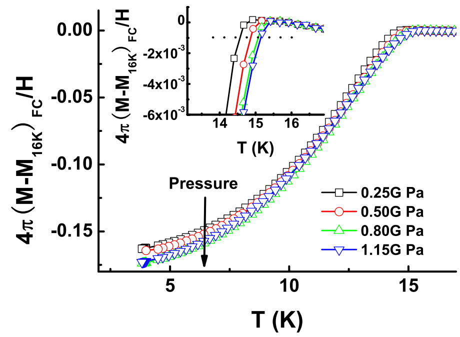


FIG. 2. (Color online) FC susceptibilities as function of temperature under various pressures for Pr-15 sample. In the inset is the enlargement of the susceptibility curves in the vicinity of $T_C$. The dashed line has the same meaning as in Fig. 1(b).

remains almost unchanged when pressure increases from 0.8 GPa up to 1.15 GPa, showing a similar pressure dependence as that of $T_C$. A clear comparison can thus be made from Fig. 3 for the pressure effects of Pr-15 and Sm-55 samples. Both samples exhibit a close correlation between the pressure dependences of their $T_C$'s and Meissner signals: they either simultaneously increase or decrease or keep almost constant with applied pressure.

When the average grain size $2r$ in the direction perpendicular to applied field is comparable to magnetic penetration depth $\lambda$, the ideal diamagnetic susceptibility of a granular superconductor is a function of $\lambda$ and $r$,[15] $4\pi\chi_I=-1+\frac{3\lambda}{r}\coth\frac{r}{\lambda}-\frac{3\lambda^2}{r^2}$. In this study, grains in the polycrystalline samples are randomly oriented, the overall susceptibility should be reduced to one third of the ideal value, i.e., $4\pi\chi=-\frac{1}{3}+\frac{\lambda_{ab}}{r}\coth\frac{r}{\lambda_{ab}}-\frac{\lambda_{ab}^2}{r^2}$, when considering the anisotropy of penetration depth. Here $\lambda_{ab}$ denotes the in-plane penetration depth. We may take the in-plane penetration depth $\lambda_{ab}$ under ambient condition as about 200–350 nm for 1111 iron-based superconductors.[19–23] Since the average grain size in present samples is about 6–20 $\mu$m as mention above, the values of

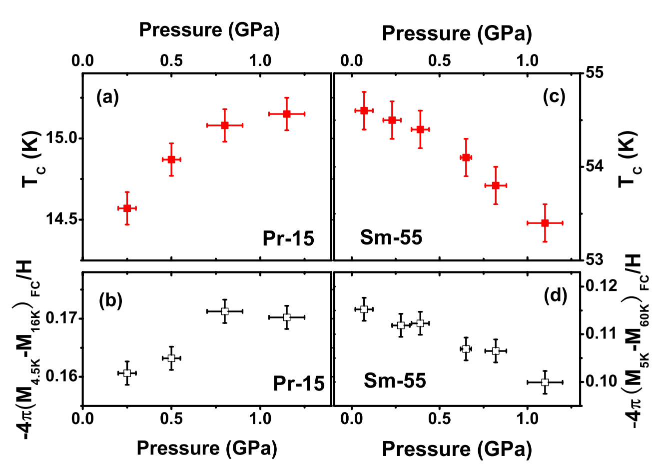


FIG. 3. (Color online) The pressure dependences of $T_C$ (a) and of the Meissner signal size taken at 4.5 K (b) for Pr-15 sample. The linear pressure effects on $T_C$ with a pressure coefficient $dT_C/dP\approx-1.1$ K/GPa (c) and on the Meissner signal size at 5 K with a $d(4\pi\chi_{5\ K})/dP\approx-0.014$/GPa (d) for Sm-55 sample.

$4\pi\chi$ can thus be estimated as between about $-0.23$ and $-0.31$ according to this formula. This is in the same order of the ambient $4\pi\chi$'s we observed for Sm-55 and Pr-15 samples, which are about $-0.12$ and $-0.15$, respectively. The lower values of measured Meissner signal than the calculated ones imply that the pinning effects are significant, which are most likely due to the existence of the amorphous impurities and grain-boundary cracks as revealed by backscattered electron imaging and orientation imaging microscopy.[18] Fortunately, in the present work where the maximum pressure is only $\sim$1 GPa, it can be safely assumed that the pressure effect on the pinning, if any, is not strong enough to change the trends of the pressure dependences of Meissner signal. This is also evidenced by the fact that different pressure dependences, positive or negative, of the Meissner signal can be revealed for Pr-15 and Sm-55 samples. Hence the above formula can be properly used to make an estimation in present study, which describes that the diamagnetic susceptibility value monotonically decreases when the square of penetration depth increases and vice versa. As it has been mentioned above that the square of penetration depth is inversely proportional to the ratio of superfluid density $n_s$ to effective mass $m^*$, Fig. 3 therefore clearly indicates that the increase or decrease in $T_C$ is associated with the parallel increase or decrease in $n_s/m^*$. The facts that the $T_C$ may increase and decrease with pressure[9–11] suggest that the contribution to the pressure effect should mainly come from $n_s$ instead of $m^*$. We thus propose that the pressure dependences of $T_C$ shown in Figs. 3(a) and 3(c) are positively correlated with the variation in the superfluid density under pressure.

It is worth mentioning that $\mu$SR experiments have revealed that the $T_C$ of most iron-based superconductors nearly linearly depends on the superfluid density,[24] following the Uemura relation which was first observed in high-temperature cuprate superconductors suggesting the non-BCS feature in the cuprates.[25] This is qualitatively consistent with our results of the iron-based superconductors under pressure. In addition, we also notice that in *s*-wave BCS-type superconductor $RbOs_2O_6$, the penetration depth derived from susceptibility measurements is independent of pressure.[26] This is in opposite to our present findings so that it implies that, indeed, iron-based superconductor is more like cuprate superconductors than ordinary BCS ones. Besides, the $T_C$ of iron-based superconductors is also found correlated with the change in chemical bond and lattice dimension (see, for example, Refs. 27 and 28), which may be attributed to the pressure effect due to inner chemical pressure associated with chemical composition.

In summary, by diamagnetic susceptibility measurements under pressures on nominal $SmFeAsO_{0.85}$ and $PrFe_{0.925}Co_{0.075}AsO$, we have shown the direct correlation between the pressure effects on $T_C$'s and Meissner signals. Our experiments demonstrate that the variation in superfluid density under pressure is responsible for the different signs of $dT_C/dP$ observed in each individual sample. The advantage of pressure-dependence study on a given sample is that it excludes the influences of the extrinsic factors on the experimental measurements, such as impurities, disorders, and different doping elements which may vary from sample to sample. The positive correlation between $T_C$ and superfluid density under pressure distinguishes these iron-based superconductors from the conventional BCS superconductors. However, current study cannot reliably establish the quantitative relationship between $T_C$ and superfluid density. Further studies are required to pin down the detailed features in the iron-based systems.

We would like to thank P. H. Hor (University of Houston) and W. M. Liu (Institute of Physics, CAS) for valuable discussions. This work is supported by the NSF of China (Projects No. 10874211 and No. 10734120) and the National Basic Research Program of China (Project No. 2007CB925002).

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