Journal of Applied Physics

Pressure effects on Tc in superconducting (Tl, Cs)1−xFe2−ySe2

S. C. Chen, K. J. Syu, H. H. Sung, W. H. Lee, C. C. Li et al.

Citation: J. Appl. Phys. **113**, 153903 (2013); doi: 10.1063/1.4802661
View online: http://dx.doi.org/10.1063/1.4802661
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v113/i15
Published by the American Institute of Physics.

JOURNAL OF APPLIED PHYSICS **113**, 153903 (2013)

Pressure effects on T_c in superconducting $(Tl, Cs)_{1-x}Fe_{2-y}Se_2$

S. C. Chen,[1] K. J. Syu,[1] H. H. Sung,[1] W. H. Lee,[1,a)] C. C. Li,[2] and Y. Y. Chen[2]
[1]*Department of Physics, National Chung Cheng University, Ming-Hsiung, Chia-Yi 62199, Taiwan*
[2]*Institute of Physics, Academia Sinica, Taipei 115, Taiwan*

(Received 22 February 2013; accepted 5 April 2013; published online 18 April 2013)

Hydrostatic-pressure (up to 0.96 GPa) dependence of T_c in a newly discovered Fe-based superconductor $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ ($T_c = 28$ K, at ambient pressure) is reported. Static magnetization measurements under pressure indicate that the linear increase in T_c is initially rapid ($dT_c/dP \sim 9.9\ K \cdot GPa^{-1}$) but slows down to $dT_c/dP \sim 2.5\ K \cdot GPa^{-1}$ for P ≧ 0.18 GPa. The T_c of the superconducting phase is 32 K at pressure P = 0.96 GPa. The simple rigid band model or the Bardeen, Cooper, Schrieffer theory may not be sufficient to account for our observations, if one assumes that the lattice parameters would be linearly decreased with pressure. © *2013 AIP Publishing LLC* [http://dx.doi.org/10.1063/1.4802661]

I. INTRODUCTION

The high-T_c selenide superconducting family derived from the parent compound $TlFe_2Se_2$, which crystallizes in the $ThCr_2Si_2$-type structure (*I*4/*mmm*), has attracted much interest during the past three years. These related systems (so-called 122 phases) allow significant vacancies on the Fe, Se, or Tl sublattice while still keeping its tetragonal symmetry.[1–8] Some superconducting examples reported in the literature are as follows: $K_{0.8}Fe_2Se_2$ ($T_c \sim 31$–33 K),[2,3] $Cs_{0.8}(FeSe_{0.98})_2$ ($T_c = 29.6$ K),[4] $Rb_{0.8}Fe_2Se_2$ ($T_{c,onset} = 31$ K),[5] $Rb_{0.78}Fe_2Se_{1.78}$ ($T_c = 32$ K),[6] $Tl_{0.4}K_{0.3}Fe_{2-y}Se_2$ ($T_{c,onset} = 27.7$ K),[7] $Tl_{0.4}Rb_{0.4}Fe_{2-y}Se_2$ ($T_{c,onset} = 31.8$ K),[5] and $Tl_{0.58}Rb_{0.42}Fe_{1.72}Se_2$ ($T_{c,onset} = 32$ K).[8] Recently, superconductivity in $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ with T_c around 28 K at ambient pressure has been reported by Syu *et al.*[9] In order to further explore the extensive examination on the theory of Fe-based superconductor, in this work, the hydrostatic pressure dependence of T_c measurements for this compound $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ have been made. It is noted that the isostructural superconducting phase of 122 Fe-based compounds like $Tl_{0.6}Rb_{0.4}Fe_{1.67}Se_2$, $K_{0.8}Fe_{1.7}Se_2$ etc. have been found to be sensitive to the application of pressure.[10]

II. EXPERIMENT

Samples investigated were prepared as described previously.[9] The superconducting phase was estimated to have the nonstoichiometric compositions $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$. The refined lattice parameters as determined by the least squares fit method for the $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ phase are $a = 0.3881(8)$ nm, and $c = 1.4057(9)$ nm. The decrease (increase) in the lattice parameter a (c) of $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$, as compared with $TlFe_2Se_2$ ($a = 0.3883$ nm, $c = 1.4022$ nm),[11] unambiguously results from the vacancies and larger ionic radius of Cs^{+1}.

A customer designed high pressure cell made by BeCu alloy and ZrO_2 ceramic rods (here after "BeCu cell") is used to create high pressure environment. BeCu cell is a clamp type hydrostatic pressure cell. Powder samples are placed in a sealed Teflon capsule (1.6 mm in diameter and 5 mm for height) along with silicon oil (as pressure medium). The applied force would transfer to sample through opposite rod placed beside the sample capture. The pressure at low temperature was determined *in situ* by measuring the T_c shift in an applied magnetic field of 10 Oe of a small piece of high purity (99.9999%) Pb placed alongside the sample. The dc magnetic susceptibility measurements were made in a magnetic field of 10 Oe between 5 and 50 K with Quantum Design Magnetic Properties Measurement System (MPMS) system. The background signal from the cell was removed under the same experimental conditions with an empty cell.

III. RESULTS AND DISCUSSION

Fig. 1(a) presents the temperature dependence of the zero-field-cooling (ZFC) and field-cooling (FC) magnetization data measured in a field of 10 Oe between 5.0 and 40 K for the sample $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ at ambient pressure. Measurements were performed on a bulk sample of about 0.1 g mass. The ZFC curve for the sample shows clear transition from paramagnetic state to superconducting state around 28 K. The quite wide transition width is a manifestation of the inhomogeneous superconducting phase. It is implied that the inhomogeneity of the sample due to the disordered Tl and Cs atoms arranged in 2 a position of the space group *I*4/*mmm* and the random distribution of vacancy in 4 d Fe sites results in a broad transition. However, this sample shows large shielding signal, ∼−0.146 emu/g, at T = 5 K. Precluding any correction for demagnetization effects, size effects, and non-superconducting impurity phase to our sample, we calculate a diamagnetic effect (from ZFC data at 5 K) of 110% of an ideal value of χ_{dc} for a long cylinder ($-1/4\pi$). This superconducting volume fraction is presumably able to constitute bulk superconductivity. The low field positive Meissner flux expulsion (from FC data) is a characteristic of bulk magnetic superconductors. The FC data will become negative if the powder sample is used, as will be shown in Fig. 2(b). The resistivity data between 5 and 38 K

[a)]Author to whom correspondence should be addressed. E-mail: phywhl@ccu.edu.tw. Tel.: +886-5-2720586. Fax: +886-5-2720587.

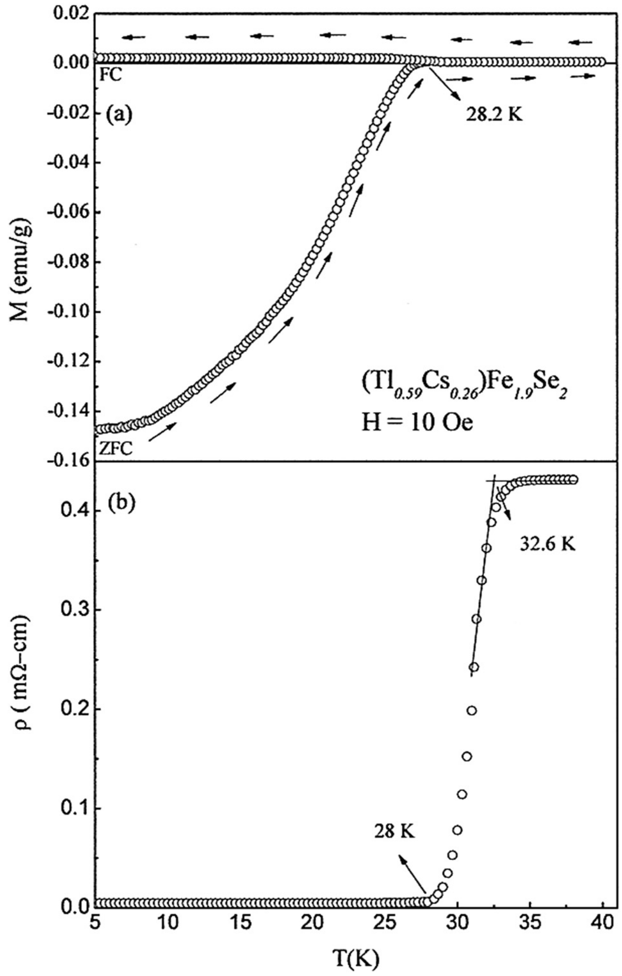

FIG. 1. (a) Temperature dependence of ZFC and FC magnetization measured at 10 Oe under ambient pressure, the $T_{c,onset}$ is 28.2 K. This data are reproduced from Ref. 9. (b) Electrical resistivity (ρ) versus temperature. Resistivity begins to drop dramatically below 32.6 K and zero resistivity appears at 28 K.

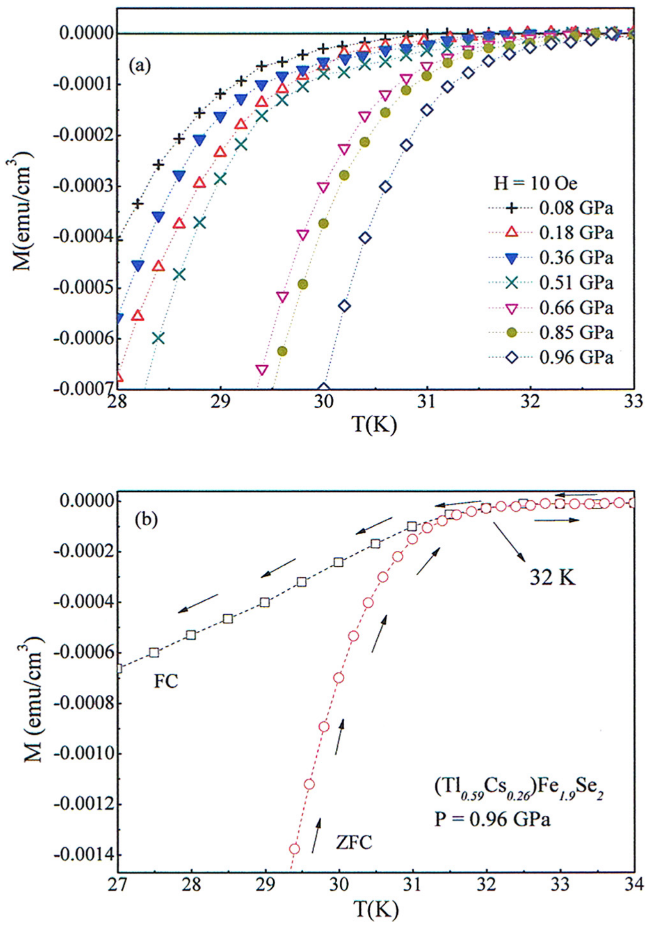

FIG. 2. (a) Temperature dependence of ZFC magnetization data for the sample $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ measured in a field of 10 Oe between 28 and 33 K under different pressures. Seven symbols (+, △, ▼, ×, ▽, ●, ◇) correspond to the pressures P = 0.08, 0.18, 0.36, 0.51, 0.66, 0.85, and 0.96 GPa. (b) The splitting point of ZFC and FC curves for $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ under P = 0.96 GPa is at T = 32 K.

for the sample investigated are depicted in Fig. 1(b). As depicted in Fig. 1(b), the $T_{c,onset}$, a point taken as the intersection of the normal state line and the superconducting transition line, appears at 32.6 K and the zero resistivity temperature $T_{c,zero}$ occurs at 28 K. it is found that the superconducting transition temperatures as determined by either electrical or magnetic method are well consistent in this work. Figure 2(a) demonstrates the temperature dependent ZFC magnetization of polycrystalline $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ for applied pressures ranging from atmospheric to approaching 1.0 GPa. It is seen that the shielding curves for constant field (10 Oe) shift clearly toward higher temperature region with increasing hydrostatic pressure. This result is different from what is observed in the superconducting system $LaO_{1-x}F_xFeAs$ ($x = 0.11$) in which the onset T_c shows no obvious change when the pressure is increased up to 1.03 GPa.[12] The T_c value is taken to be the onset of superconductivity. A good way to determine the onset T_c value for the system with strong pinning effect is to take the splitting point of the ZFC and FC curves. Since the primary cause of flux trapping is attributed to the presence of lattice defects within the body of the superconductor, the strong pinning effect in $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ is predicted due to its structure defects. Fig. 2(b) is the representative example for determining the onset T_c value of $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ under P = 0.96 GPa. Obviously, the T_c value of the superconducting phase is 32 K at pressure P = 0.96 GPa for $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$. The hydrostatic pressure dependence of T_c for

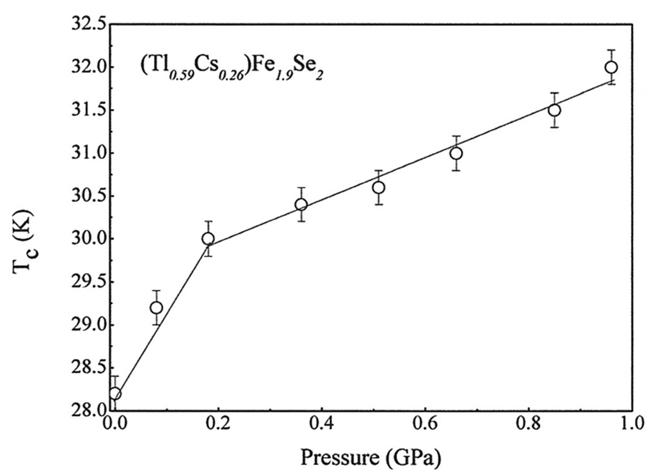

FIG. 3. The pressure dependence of the superconducting phase transition temperature T_c for $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$.

$(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ is then plotted in Fig. 3. These data shown in Fig. 3 are remarkable in that there are two very pressure dependent features. The first feature is the fast increase in T_c ($dT_c/dP \sim 9.9\,K \cdot GPa^{-1}$) initially. The second conspicuous feature is that the increasing rate slows down to $dT_c/dP \sim 2.5\,K \cdot GPa^{-1}$. It was suggested that the electronic structures of this compound should depend on more than just the valence electron density and the lattice parameters if the lattice parameters of the unit cell are linearly decreased with the hydrostatic pressure and the conventional BCS (Bardeen, Cooper, Schrieffer) theory does not fail. Thus, for the interpretation of the results one first needs to know the equation of state, and second the volume dependence of the electronic structure of the sample. Experience may tell us that for an electron-phonon weakly coupled superconductor, e.g., LaNiSi,[13] its pseudo-ternary system $La(Pt_{1-x}Ni_x)Si$ exhibits monotonic T_c functions of x or lattice parameters of the unit cell.[14] In fact, 3d magnetic ion (Fe, Co, Ni) based superconductors are always found to be challengeable to the conventional BCS theory of superconductivity. For the noncentrosymmetric superconductor $LaNiC_2$,[15–17] the increase in T_c is initially rapid ($dT_c/dx = 12$ K) but slows down ($dT_c/dx = 1$ K) for $x \geqq 0.2$ in the pseudo-ternary system $La(Ni_{1-x}Cu_x)C_2$.[18] As to the Co-based superconductor Zr_2Co, the superconducting phase diagrams of $Zr_2(Co_{1-x}Ni_x)$[19] and $Zr_2(Co_{1-x}Ga_x)$[20] exhibited an explicit maximum T_c close to $x = 0.1$ and $x = 0.05$, respectively. Takekuni *et al.* further employed the nuclear-magnetic-resonance (NMR) technique to ponder ^{59}Co in the normal state of $Zr_2(Co_{1-x}Ni_x)$ and to explore the itinerant nearly antiferromagnetic behavior in superconducting $Zr_2(Co_{1-x}Ni_x)$. They identified that the superconducting transition temperature related more to the spin density fluctuations around q = Q (Q being an AF wave vector) than to the density of states at the Fermi level.[21]

IV. CONCLUSIONS

From the present study, magnetic measurements under hydrostatic pressure up to 0.96 GPa for $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$ showed a two-step increase in T_c. The simple rigid band model and the conventional BCS theory may not be sufficient to account for our observations, if one assumes that the lattice parameters of the unit cell are linearly decreased with pressure. We have taken note of the isostructural 122 superconducting systems $Tl_{0.6}Rb_{0.4}Fe_{1.67}Se_2$, $K_{0.8}Fe_{1.7}Se_2$ and $K_{0.8}Fe_{1.78}Se_2$ in which T_c drops from ~32 K to ~5 K with pressure from 1 GPa to ~10 GPa and re-emerges second superconducting phase at 48 K above 11.5 GPa.[10] To further investigate the superconducting behavior in $(Tl_{0.59}Cs_{0.26})Fe_{1.9}Se_2$, an interesting study will be directed toward measurements under higher pressure (>1 GPa) using diamond anvil technique to see the changes of superconductivity.

ACKNOWLEDGMENTS

This work was supported by the National Science Council of the Republic of China under Grant Nos. NSC-99-2112-M-194-006-MY3 and NSC-101-2811 -M-194-016.

[1] L. Häggström, A. Seidel, and R. Berger, J. Magn. Magn. Mater. **98**, 37 (1991).
[2] J. Guo, S. Jin, G. Wang, S. Wang, K. Zhu, T. Zhou, M. He, and X. Chen, Phys. Rev. B **82**, 180520(R) (2010).
[3] Y. Mizuguchi, H. Takeya, Y. Kawasaki, T. Ozaki, S. Tsuda, T. Yamaguchi, and Y. Takano, Appl. Phys. Lett. **98**, 042511 (2011).
[4] Z. Shermadini, A. Krzton-Maziopa, M. Bendele, R. Khasanov, H. Luetkens, K. Conder, E. Pomjakushina, S. Weyeneth, V. Pomjakushin, O. Bossen, and A. Amato, Phys. Rev. Lett. **106**, 117602 (2011).
[5] C. H. Li, B. Shen, F. Hang, X. Zhu, and H. H. Wen, Phys. Rev. B **83**, 184521 (2011).
[6] A. F. Wang, J. J. Ying, Y. J. Yan, R. H. Liu, X. G. Luo, Z. Y. Li, X. F. Wang, A. Zhang, G. J. Ye, P. Cheng, Z. J. Xiang, and X. H. Chen, Phys. Rev. B **83**, 060512 (2011).
[7] R. H. Liu, X. G. Luo, M. Zhang, A. F. Wang, J. J. Ying, X. F. Wang, Y. J. Yan, Z. J. Xiang, P. Cheng, G. J. Ye, Z. Y. Li, and X. H. Chen, EPL **94**, 27008 (2011).
[8] H. Wang, C. Dong, Z. Li, S. Zhu, Q. Mao, C. Feng, H. Q. Yuan, and M. Fang, EPL **93**, 47004 (2011).
[9] K. J. Syu, H. H. Sung, and W. H. Lee, Physica C **471**, 591 (2011).
[10] L. Sun, X. J. Chen, J. Guo, P. Gao, Q. Z. Huang, H. Wang, M. Fang, X. Chen, G. Chen, Q. Wu, C. Zhang, D. Gu, X. Dong, L. Wang, K. Yang, A. Li, X. Dai, H. K. Mao, and Z. X. Zhao, Nature **483**, 67 (2012).
[11] R. Berger and C. F. Van Bruggen, J. Less-Common Met. **113**, 291 (1989).
[12] W. Lu, J. Yang, X. L. Dong, Z. A. Ren, G. C. Che, and Z. X. Zhao, New J. Phys. **10**, 063026 (2008).
[13] W. H. Lee, F. A. Yang, C. R. Shih, and H. D. Yang, Phys. Rev. B **50**, 6523 (1994).
[14] W. H. Lee, Solid State Commun. **94**, 425 (1995).
[15] W. H. Lee, H. K. Zeng, Y. D. Yao, and Y. Y. Chen, Physica C **266**, 138 (1996).
[16] A. D. Hillier, J. Quintanilla, and R. Cywinski, Phys. Rev. Lett. **102**, 117007 (2009).
[17] I. Bonalde, R. L. Ribeiro, K. J. Syu, H. H. Sung, and W. H. Lee, New J. Phys. **13**, 123022 (2011).
[18] H. H. Sung, S. Y. Chou, K. J. Syu, and W. H. Lee, J. Phys.: Condens. Matter **20**, 165207 (2008).
[19] Z. Kakutani, Y. Murakami, K. Nishimura, and K. Mori, Ann. Rep. Low Temp. Toyama Univ. **8**, 26 (1996).
[20] K. J. Syu, C. H. Wu, S. C. Chen, and W. H. Lee, Solid State Commun. **151**, 404 (2011).
[21] M. Takekuni, H. Sugita, and S. Wada, Phys. Rev. B **58**, 11698 (1998).