Physica C 469 (2009) 1958–1961

Contents lists available at ScienceDirect

Physica C

journal homepage: www.elsevier.com/locate/physc

Divergency of SDW and structure transition in $Fe_{1-x}Ni_xSe_{0.82}$ superconductors

S.B. Zhang [a], H.C. Lei [a], X.D. Zhu [a], G. Li [a], B.S. Wang [a], L.J. Li [a], X.B. Zhu [a], W.H. Song [a], Z.R. Yang [a], Y.P. Sun [a,b,*]

[a] Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei, China
[b] High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, China

ARTICLE INFO

Article history:
Received 26 March 2009
Received in revised form 26 May 2009
Accepted 13 July 2009
Available online 16 July 2009

PACS:
74.70.Ad
74.25.Fy
74.25.Ha

Keywords:
FeSe
Iron-based superconductor
Superconductivity
Substituted effect

ABSTRACT

The effect of the Ni-substitution on the superconductivity of α-$FeSe_{0.82}$ system has been investigated. It is found that the superconductivity of α-$FeSe_{0.82}$ can be quickly suppressed when a small quantity of Fe is substituted by Ni element. The resistivity and moment of the sample at normal state increase with increasing Ni-substitution content. Moreover, it is found that the Ni-substitution does not change the structure transition temperature, but increases the spin density wave (SDW) order temperature. It is suggested that the quick suppression of superconductivity of α-$FeSe_{0.82}$ caused by the Ni-substitution is related to the increase of SDW order temperature. Therefore, Fe atom plays a key role for the appearance of the superconductivity in FeSe system.

1. Introduction

In the past, it was considered to be difficult for the occurrence of superconductivity in iron-based compounds, because of the magnetism of Fe. The recent discovery of the Fe-based high temperature superconductors has attracted a world-wide attention [1–5]. It is the first system where Fe plays the key role for superconductivity. Many following study works have focused on the searching for the new superconductors in other Fe-based compounds. Very recently, the binary iron–selenium system, which has a similar but simpler layered structure than the quaternary arsenide, was found to be superconducting with $T_C \sim 8$ K [6]. For α-$FeSe_x$ crystals grown by flux method, it is found that its T_C is about 10 K, which is little higher than that of polycrystals [7]. The finding of iron–selenium superconductor has opened a new route for the study of Fe-based superconductors and the search for new superconductors. Subsequent work revealed that a huge enhancement of superconducting transition temperature T_C was observed under high pressure. And the onset temperature was as high as 27 K at 1.4 GPa [8,9]. The tetragonal FeTe compound with an analogous structure to α-FeSe does not show superconducting transition and occurs a structural phase transition from tetragonal to orthorhombic around 80 K [10,11]. In $FeSe_{1-x}Te_x$ system, superconducting phase is suppressed when the sample composition approaches the end member $FeTe_{0.82}$, which exhibits an incommensurate antiferromagnetic order. Superconductivity in this class of Fe-based compounds is associated with magnetic fluctuations [12,13]. However, the superconductivity has been observed in S-substituted FeTe compounds with $T_C \sim 8$ K [14]. FeSe systems including α-FeSe with PbO-structure, β-phase with NiAs-structure, ferromagnetic hexagonal Fe_7Se_8 and monoclinic Fe_3Se_4 were widely studied in the past [15–17]. The tetragonal phase PbO-type α-FeSe has a Fe based planar sublattice equivalent to layered iron-based oxypnictides, which has a layered crystal structure belonging to the *P*4/nmm space group. The crystal of α-FeSe is composed of a stack of edge-sharing $FeSe_4$-tetrahedra layer by layer [6]. An enhancement of T_C was observed with the substitution of S or Te for Se. And the structural and electronic properties of FeSe which is substituted at Fe site have been studied [18]. The effect of the stoichiometry variation of Fe in $Fe_{1+\delta}Se$ system has also been studied, in which the superconductivity of $Fe_{1+\delta}Se$ system reveals an extreme sensitivity to the stoichiometry of Fe [19]. The density functional calculation indicates that doped FeSe is not a conventional electron-phone superconductor, which is similar to the Fe–As superconducting system. And both FeSe and FeTe show

* Corresponding author. Address: Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, China. Tel.: +86 551 5592757; fax: +86 551 5591434.
E-mail addresses: sbzhang@issp.ac.cn (S.B. Zhang), ypsun@issp.ac.cn (Y.P. Sun).

0921-4534/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.physc.2009.07.003

spin-density wave (SDW) ground states, while FeS is closed to instability [20]. In order to understand further the importance of the Fe atom, and the relation of the SDW and structural transition in the binary iron–selenium system, in this work, we have systemically studied the structural, electronic transport and magnetic properties of Ni-substituted $Fe_{1-x}Ni_xSe_{0.82}$ polycrystals.

2. Experimental

$Fe_{1-x}Ni_xSe_{0.82}$ polycrystalline samples with $x = 0$. 0.01, 0.02, 0.03, 0.04 were prepared using a solid state reaction method. High purity powder of selenium (99.99%), iron (99.9%) and nickel (99.8%) with appropriate stoichiometry were mixed and ground with an agate mortar and pestle in a vacuum glove box. Then the ground powders were sealed into different quartz tube and heated at 700 °C for 24 h. After furnace cooling to room temperature, the slight melted samples were reground and pressed into small pellets. The pellets were all sealed into quartz tube and sintered again at 700 °C for 24 h, and finally annealed at 400 °C for 36 h.

The crystal structure of the samples was determined by X-ray diffraction pattern (XRD) using a Philips X' pert PRO X-ray diffractometer with Cu K_α radiation at room temperature. The resistivity of the sample was measured by the standard four-probe method using a Quantum Design Physical Property Measurement System (PPMS) ($1.8\ K \le T \le 400\ K$, $0\ T \le H \le 9\ T$). Magnetization measurements as a function of temperature were performed in a Quantum Design Superconducting Quantum Interference Device (SQUID) system ($1.8\ K \le T \le 400\ K$, $0\ T \le H \le 5\ T$).

3. Results and discussion

Fig. 1 shows the X-ray diffraction (XRD) patterns of $Fe_{1-x}Ni_xSe_{0.82}$ ($x = 0$, 0.01, 0.02, 0.03 and 0.04) samples. All the samples trace amount of possible impurity phases including iron oxide and selenium. β-phase FeSe has also been observed, and the peaks of β-phase have been suppressed with increase of Ni-substitution content. It is suggested that the Ni-doping helps to synthesize PbO-type α-FeSe phase. We indexed all peaks with a tetragonal lattice *P*4/nmm (Fe/Ni, 2a (0.75, 0.25, 0); Se 2b (0.25, 0.25, 0.265)) by standard Rietveld method. And the fitting result of $Fe_{0.97}Ni_{0.03}Se_{0.82}$ has been shown in Fig. 2 as an example. The lattice constants of *a*(Å), *c*(Å) and unit cell volume *V*(Å^3) obtained from fitting results have been shown in Fig. 3. It reveals that the lattice constants *a*, *c* and unit cell volume *V* have a decreased trend with increasing Ni-substitution content corresponding to

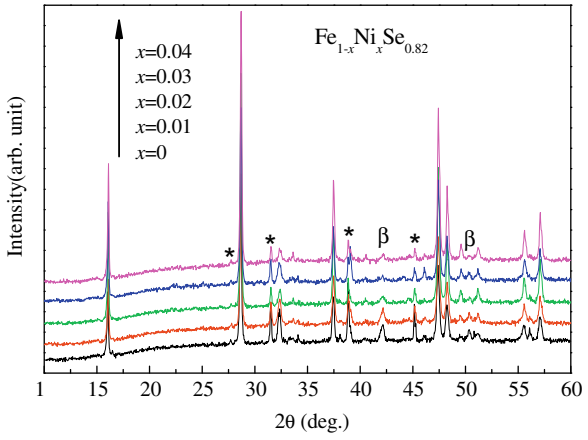

Fig. 1. X-ray diffraction patterns of $Fe_{1-x}Ni_xSe_{0.82}$ ($x = 0$, 0.01, 0.02, 0.03, 0.04). Asterisks indicate the impurity phases and β indicate hexagonal phase.

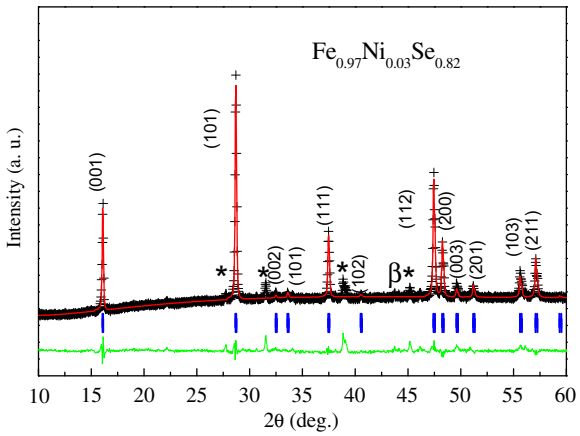

Fig. 2. X-ray diffraction pattern of $Fe_{0.97}Ni_{0.03}Se_{0.82}$. Crosses indicate the experimental data and the calculated data is the continuous line overlapping them. The lowest curve shows the difference between experimental and calculated patterns. The vertical bars indicate the expected reflection positions. Asterisks indicate the impurity phases and β indicate hexagonal phase.

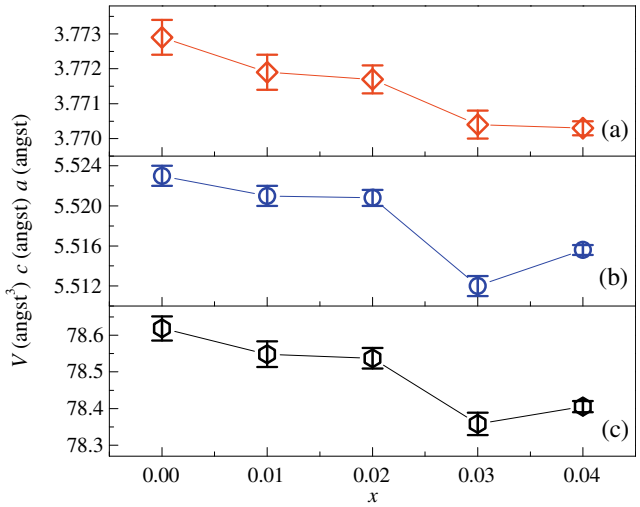

Fig. 3. Ni-substitution dependence of lattice constants (a) *a*, (b) *c* and unit cell volume (c) *V*.

the smaller ionic radii of Ni^{2+} (0.55 Å) than Fe^{2+} (0.64 Å). This is not completely agreement with the results reported in Ref. [15], in which *a* increased with Ni-substitution.

Fig. 4 shows the temperature dependence of resistivity for $Fe_{1-x}Ni_xSe_{0.82}$ at zero magnetic field. The inset shows the magnification plot of resistivity in low temperature region of $FeSe_{0.82}$. The onset temperature T_C^{onset} and zero resistance transition temperature T_{C0} of $FeSe_{0.82}$ are 11.33 K and 4.33 K, respectively. Fig. 4 indicates that Ni-substitution quickly suppresses the superconductivity of the sample. Only a small Ni-substitution content of $x = 0.01$, the T_{C0} of the sample is below 2 K. As $x = 0.04$, no superconducting transition is observed over 2 K. In fact, it is very surprise that the superconductivity of the sample can be suppressed by Ni-substitution from the viewpoint of pressure effect. Previously, the phenomenon of the superconductivity enhancement of α-$FeSe_x$ has been observed [8,9]. For our Ni-substituted samples, the structure fitting results indicates that the lattice parameters *a*, *c* and unit cell *V* decrease with Ni-substitution content, implying the introduction of a positive chemical pressure. That is to say, the chemical pressure is not always favorable for superconductivity of FeSe system, although T_C has been improved by S-substitution [18]. From

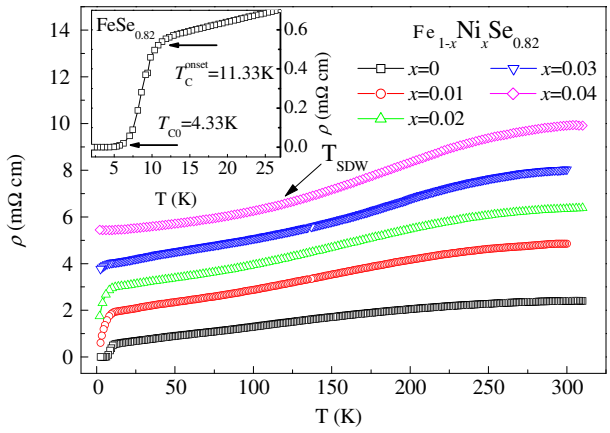

Fig. 4. Temperature dependence of resistivity for α-$Fe_{1-x}Ni_xSe_{0.82}$ at zero magnetic field. The inset shows the magnification plot of resistivity in low temperature region of α-$FeSe_{0.82}$.

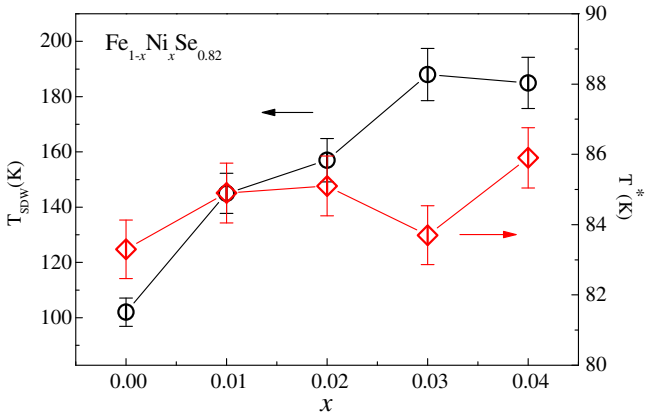

Fig. 6. Ni-substitution dependence of T_{SDW} and T^* for α-$Fe_{1-x}Ni_xSe_{0.82}$.

Fig. 4, it is found that all $Fe_{1-x}Ni_xSe_{0.82}$ samples exhibit metallic behavior and the resistivity at normal state increase with increasing Ni-substitution content. Furthermore, a broad bump at $T_{SDW} \approx 150$ K for no Ni-substitution sample is observed and shifts to high temperature with increasing Ni-substitution content. This kind of bump of resistivity curve in the normal state is also observed in Fe-based oxypnictide $La(O_{1-x}F_x)FeAs$, in which the bump is attributed to the SDW order. From optical measurements and theoretical calculations, it has been argued that LaOFeAs has an antiferromagnetic SDW instability below 150 K and the appearance of superconductivity is suggested to arise from the suppression of this SDW order [21,22]. Recently, the density functional calculation indicates that FeSe has a SDW ground state [20]. Therefore, it is suggested that the bump observed in the curve of temperature dependence of resistivity in the normal state may also originate from SDW order. The SDW order temperature T_{SDW} has been defined by differentiating from the temperature dependence of resistivity as shown in Fig. 5. It is noticed that T_{SDW} increases with the Ni-substitution content as shown in Fig. 6. We consider that the enhancement of the SDW order caused by Ni-substitution should be the intrinsic reason for the suppression of superconductivity.

Fig. 7 shows the temperature dependence of magnetic susceptibility for $Fe_{1-x}Ni_xSe_{0.82}$ measured in a 10 Oe magnetic field. The zero field cooling (ZFC) and field cooling (FC) susceptibility are almost temperature independent above a magnetic abnormality at about T^*, which indicates that these samples are Pauli paramagnet over T^*. The abnormality of susceptibility, especially pronounced in FC measurement, has been attributed to the structural transition from the tetragonal ($P4/nmm$) to triclinic ($P-1$) symmetry [6]. The transitional temperature T^* of all $Fe_{1-x}Ni_xSe_{0.82}$ samples almost do not depend on the Ni-substitution content and the results have been shown in Fig. 6. In other words, Ni-substitution does not influence the structural transition temperature of α-FeSe system. Impurity in $Fe_{1-x}Ni_xSe_{0.82}$ samples may becomes another reason which leads to the abnormality of susceptibility. Higher quality samples are needed in order to study the intrinsic mechanism.

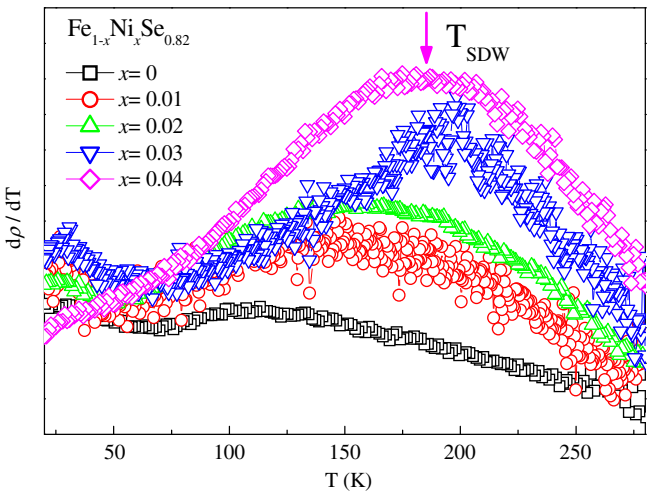

Fig. 5. Temperature dependence of $d\rho/dT$ for α-$Fe_{1-x}Ni_xSe_{0.82}$ at zero magnetic field.

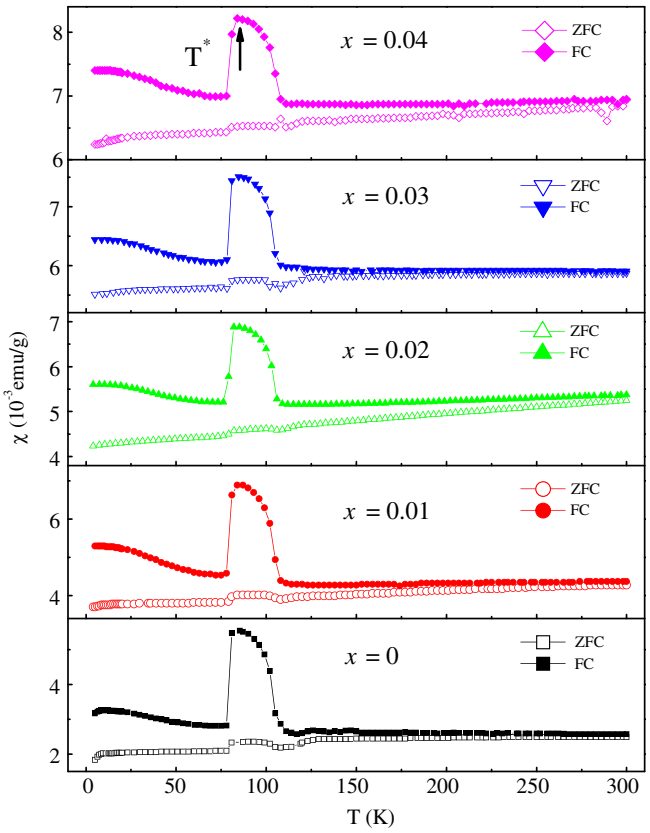

Fig. 7. Temperature dependence of magnetic susceptibility $\chi(T)$ for α-$Fe_{1-x}Ni_xSe_{0.82}$ measured at 10 Oe.

4. Conclusions

In summary, Ni-substituted $Fe_{1-x}Ni_xSe_{0.82}$ polycrystalline samples have been synthesized. The transport and magnetic properties of the sample have been investigated. It is found that the superconductivity of the sample is very sensitive to the Ni-substitution and is quickly suppressed. It is suggested that the enhancement of the SDW order caused by Ni-substitution may be the intrinsic reason for the suppression of superconductivity, though a positive chemical pressure is introduced by Ni-substitution. Moreover, Ni-substitution does not change the structural transition temperature. The result reveals that Fe plays a crucial role for superconductivity in this binary iron–selenium alloy.

Acknowledgements

This work was supported by the National Key Basic Research under Contract Nos. 2006CB601005, 2007CB925002, and the National Nature Science Foundation of China under Contract Nos. 10774146, 10774147, Director's Fund of Institute of Solid State Physics and Director's Fund of Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei.

References

[1] Y. Kamihara, T. Watanabe, M. Hirano, H. Hosono, J. Am. Chem. Soc. 130 (2008) 3296.
[2] X.H. Chen, T. Wu, G. Wu, R.H. Liu, H. Chen, D.F. Fang, Nature 453 (2008) 761.
[3] G.F. Chen, Z. Li, D. Wu, G. Li, W.Z. Hu, J. Dong, P. Zheng, J.L. Luo, N.L. Wang, Phys. Rev. Lett. 100 (2008) 247002.
[4] Z.A. Ren, J. Yang, W. Lu, Y. Wei, X.L. Shen, Z.C. Li, G.C. Che, X.L. Dong, L.L. Sun, F. Zhou, Z.X. Zhao, Europhys. Lett. 82 (2008) 57002.
[5] P. Cheng, L. Fang, H. Yang, X.Y. Zhu, G. Mu, H.Q. Luo, Z.S. Wang, H.H. Wen, Sci. China Ser. G – Phys. Mech. Astron. 51 (2008) 719.
[6] F.C. Hsu, J.Y. Luo, K.W. Yeh, T.K. Chen, T.W. Huang, P.M. Wu, Y.C. Lee, Y.L. Huang, Y.Y. Chu, D.C. Yan, M.K. Wu, Proc. Nat. Acad. Sci. USA 105 (2008) 14262.
[7] S.B. Zhang, Y.P. Sun, X.D. Zhu, X.B. Zhu, B.S. Wang, G. Li, H.C. Lei, X. Luo, Z.R. Yang, W.H. Song, J.M. Dai, Supercond. Sci. Techol. 22 (2009) 015020.
[8] Y. Mizuguchi, F. Tomika, S. Tsuda, T. Yamaguchi, Y. Takano, Appl. Phys. Lett. 93 (2008) 152505.
[9] L. Li, Z.R. Yang, M. Ge, L. Pi, J.T. Xu, B.S. Wang, Y.P. Sun, Y.H. Zhang, arXiv:/0809.0128.
[10] K.W. Yeh, T.W. Huang, Y.L. Huang, T.K. Chen, F.C. Hsu, P.M. Wu, Y.C. Lee, Y.Y. Chu, C.L. Chen, J.Y. Luo, D.C. Yan, M.K. Wu, Europhys. Lett. 84 (2008) 37002.
[11] W. Bao, Y. Qiu, Q. Huang, M.A. Green, P. Zajdel, M.R. Fitzsimmons, M. Zhernenkov, M. Fang, B. Qian, E.K. Vehstedt, J. Yang, H.M. Pham, L. Spinu, Z.Q. Mao, arXiv:/0809.2058.
[12] M.H. Fang, H.M. Pham, B. Qian, T.J. Liu, E.K. Vehstedt, Y. Liu, L. Spinu, Z.Q. Mao, Phys. Rev. B 78 (2008) 224503.
[13] M.H. Fang, B. Qian, H.M. Pham, J.H. Yang, T.J. Liu, E.K. Vehstedt, L. Spinu, Z.Q. Mao, arXiv:/0811.3021.
[14] Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, Y. Takano, arXiv:/0811.0711.
[15] S. Margadonna, Y. Takabayashi, M.T. McDonald, K. Kasperkiewicz, Y. Mizuguchi, Y. Takano, A.N. Fitch, E. Suard, K. Prassides, Chem. Commun. 43 (2008) 5607.
[16] A. Okazaki, K. Hirakawa, J. Phys. Soc. Jpn. 11 (1956) 930.
[17] A. Okazaki, J. Phys. Soc. Jpn. 14 (1959) 112.
[18] Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, Y. Takano, arXiv:/0811.1123.
[19] T.M. McQueen, Q. Huang, V. Ksenofontov, C. Felser, Q. Xu, H.W. Zandbergen, Y.S. Hor, J. Allred, A.J. Williams, D. Qu, J. Checkelsky, N.P. Ong, R.J. Cava, arXiv:/0811.1613.
[20] A. Subedi, L.J. Zhang, D.J. Singh, M.H. Du, Phys. Rev. B 78 (2008) 134514.
[21] C. de la Cruz, Q. Huang, J.W. Lynn, J.Y. Li, W. Ratcliff II, J.L. Zarestky, H.A. Mook, G.F. Chen, J.L. Luo, N.L. Wang, P.C. Dai, Nature 453 (2008) 899.
[22] J. Dong, H.J. Zhang, G. Xu, Z. Li, G. Li, W.Z. Hu, D. Wu, G.F. Chen, X. Dai, J.L. Luo, Z. Fang, N.L. Wang, Europhys. Lett. 83 (2008) 27006.