Antiferromagnetic fluctuations in Fe(Se$_{1-x}$Te$_x$)$_{0.92}$(x = 0.75, 1) observed by inelastic neutron scattering

Satoshi Iikubo$^1$*, Masaki Fujita$^2$, Seiji Niitaka$^3$, and Hidenori Takagi$^3$

$^1$WPI Advanced Institute for Materials Research, Tohoku University, Sendai, Miyagi 980-8577, Japan
$^2$Institute for Materials Research, Tohoku University, Sendai, Miyagi 980-8577, Japan
$^3$RIKEN (The Institute of Physical and Chemical Research), Wako, Saitama 351-0198, Japan

Motivated by the discovery of superconductivity in F-doped LaFeAsO, we investigated the magnetic fluctuations in a related compound Fe(Se$_{1-x}$Te$_x$)$_{0.92}$(x = 0.75, 1) using neutron scattering techniques. Non-superconducting FeTe$_{0.92}$ shows antiferromagnetic (AF) ordering with the ordering vector $|Q| = 0.97$ Å$^{-1}$ ($Q = (0.5,0,0.5)$) at $T_N \sim 70$ K and a structural transition between tetragonal and monoclinic phases. In the AF monoclinic phase, the low-energy spectral weight is suppressed, indicating a possible gap formation. On the other hand, in the paramagnetic tetragonal phase, we observed a pronounced magnetic fluctuation around at $|Q| \approx 0.92$ Å$^{-1}$, which is slightly smaller than the commensurate value. In Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, which does not show magnetic fluctuations and shows superconductivity at $T_c \approx 8$ K, we observed that a magnetic fluctuation is located at $|Q| \approx 0.9$ Å$^{-1}$ at low energies and shifts to a higher value of $|Q| \approx 1.2$ Å$^{-1}$ at higher energies. The latter value is close to a reciprocal lattice vector $Q = (0.5,0.5,0.5)$ or $(0.5,0.5,0.0)$, where the AF fluctuations are observed in other FeAs-based materials. The existence of this common characteristic in different Fe-based superconductors suggests that the AF fluctuations may play an important role in superconductivity.

KEYWORDS: Fe(Se$_{1-x}$Te$_x$)$_y$, magnetic fluctuation, neutron scattering, iron-based superconductor

The discovery of superconductivity in the F-doped oxy pnictide LaFeAsO with a superconducting transition temperature of $T_c = 26$ K has led to intensive studies on related superconductors including iron group. Soon after the discovery, the oxygen-free iron pnictides family of Fe-based superconductors, iron chalcogenides tetragonal to orthorhombic structure. Since the discovery parts commonly exhibit an antiferromagnetic (AF) order with the ordering vector $|Q| = 0.97$ Å$^{-1}$ ($Q = (0.5,0,0.5)$) at $T_N \sim 70$ K and a structural transition between tetragonal and monoclinic phases. In the AF monoclinic phase, the low-energy spectral weight is suppressed, indicating a possible gap formation. On the other hand, in the paramagnetic tetragonal phase, we observed a pronounced magnetic fluctuation around at $|Q| \approx 0.92$ Å$^{-1}$, which is slightly smaller than the commensurate value. In Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, which does not show magnetic fluctuations and shows superconductivity at $T_c \approx 8$ K, we observed that a magnetic fluctuation is located at $|Q| \approx 0.9$ Å$^{-1}$ at low energies and shifts to a higher value of $|Q| \approx 1.2$ Å$^{-1}$ at higher energies. The latter value is close to a reciprocal lattice vector $Q = (0.5,0.5,0.5)$ or $(0.5,0.5,0.0)$, where the AF fluctuations are observed in other FeAs-based materials. The existence of this common characteristic in different Fe-based superconductors suggests that the AF fluctuations may play an important role in superconductivity.

KEYWORDS: Fe(Se$_{1-x}$Te$_x$)$_y$, magnetic fluctuation, neutron scattering, iron-based superconductor

Motivated by the discovery of superconductivity in F-doped LaFeAsO, we investigated the magnetic fluctuations in a related compound Fe(Se$_{1-x}$Te$_x$)$_{0.92}$(x = 0.75, 1) using neutron scattering techniques. Non-superconducting FeTe$_{0.92}$ shows antiferromagnetic (AF) ordering with the ordering vector $|Q| = 0.97$ Å$^{-1}$ ($Q = (0.5,0,0.5)$) at $T_N \sim 70$ K and a structural transition between tetragonal and monoclinic phases. In the AF monoclinic phase, the low-energy spectral weight is suppressed, indicating a possible gap formation. On the other hand, in the paramagnetic tetragonal phase, we observed a pronounced magnetic fluctuation around at $|Q| \approx 0.92$ Å$^{-1}$, which is slightly smaller than the commensurate value. In Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, which does not show magnetic fluctuations and shows superconductivity at $T_c \approx 8$ K, we observed that a magnetic fluctuation is located at $|Q| \approx 0.9$ Å$^{-1}$ at low energies and shifts to a higher value of $|Q| \approx 1.2$ Å$^{-1}$ at higher energies. The latter value is close to a reciprocal lattice vector $Q = (0.5,0.5,0.5)$ or $(0.5,0.5,0.0)$, where the AF fluctuations are observed in other FeAs-based materials. The existence of this common characteristic in different Fe-based superconductors suggests that the AF fluctuations may play an important role in superconductivity.

KEYWORDS: Fe(Se$_{1-x}$Te$_x$)$_y$, magnetic fluctuation, neutron scattering, iron-based superconductor

*Present address: Kyushu Institute of Technology, Kitakyushu, Fukuoka 808-0196, Japan
The powder samples were sealed in an aluminum cell with He gas. The sample cell was mounted at the cold head of a closed-cycle He-gas refrigerator.

Figure 1 (a) shows the magnetic susceptibility of FeTe$_{0.92}$, with the AF ordering appearing as a sharp drop at $\sim 70$ K. This anomaly is also associated with a structural transition from a high-temperature tetragonal phase to a low-temperature monoclinic phase. The inset of Fig. 1 (a) displays the elastic neutron scattering profile of FeTe$_{0.92}$ that was recorded around $Q = (1,0,0)$ at 240 K and 10 K. At $T = 10$ K, we observe superlattice reflections, one of which is located at the low $q$ side of $(1,0,0)$. This observation corresponds to the AF ordering with the ordering vector $Q = (0.5,0,0.5)$. The ordered moment for Fe is estimated to be $\mu_{Fe} = 1.86 \pm 0.02 \mu_B$ and lies along the b-axis, which is consistent with a previous report. (Note that a small amount of impurity Fe$_3$O$_4$, which shows the anomaly in $\chi$ at $\sim 120$ K, is observed in our profiles as a second phase.)

Figure 1 (b) shows the magnetic susceptibility $\chi$ for Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, which shows superconductivity at $T_c \approx 8$ K. The inset of Fig. 1 (b) shows the elastic neutron scattering profile of Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$ observed at 10 K and 280 K. No superlattice peaks or peak splitting is observed at low temperatures, which implies that the system involves a tetragonal unit cell without magnetic ordering even at low temperature.

We first examine the inelastic scattering data for FeTe$_{0.92}$. Figure 2 (a) shows the $q$-dependence of the inelastic scattering of $\omega = 2$ meV for two different phases. For the paramagnetic tetragonal phase at $T = 89$ K, a strong low-energy magnetic excitation is present in the form of a broad scattering peak centered at $|Q| \approx 0.92$ Å$^{-1}$. The peak center is at 0.92 Å$^{-1}$, which is below 0.97 Å$^{-1}$ ($Q = (0.5,0.5)$) or 1.27 Å$^{-1}$ ($Q = (0.5,0.5,0.5)$) and above 0.82 Å$^{-1}$ ($Q = (0.5,0,0)$). If we estimate the peak center along the $(h,0,0)$ line, the value is assigned to an incommensurate $Q = (0.47,0,0.5)$. At $T = 10$ K, the spectral weight of the magnetic fluctuations decrease significantly. We can see a sharp intensity peaked at $|Q| \approx 1$ Å$^{-1}$, but it arises from a phonon. The magnetic Bragg peak appears at $|Q| \approx 0.97$ Å$^{-1}$ in the AF monoclinic phase, as seen in the inset of Fig. 1(a). It seems that the structural phase transition from tetragonal to monoclinic phase slightly changes the magnetic propagation vector from $|Q| \approx 0.92$ Å$^{-1}$ to $|Q| \approx 0.97$ Å$^{-1}$. A modification of the magnetic properties caused by the structural transition was also reported for the FeAs system. The magnetic excitation in the paramagnetic tetragonal phase is proposed to have a 2D vector $Q = (0.5,0,5)$, while the propagation vector in monoclinic phase is 3D vector $Q = (0.5,0.5,0.5)$. A remarkable difference between our Fe(Se$_{1-x}$Te$_x$)$_{0.92}$ system and the FeAs system is the a-b plane component of the propagation vector, which is $Q_{2D} = (0.5,0)$ for the former and $Q_{2D} = (0.5,0.5)$ for the latter.
represent Gaussian fittings, where the parameters used are the peak center, peak width, scale factor, and background counts that are assumed to be constant with a change in $q$. For the sake of clarity, a sharp spurious intensity peak near $1.1 \text{ Å}^{-1}$ was subtracted from these profiles. A weak but finite broad scattering peak centered at $|Q| \approx 0.9 \text{ Å}^{-1}$ appears at both 3.5 K and 100 K, indicating that the intensities are almost temperature independent except for the background. If the scattering intensities arose from a phonons, then the scattering intensities would be expected to be proportionate to the factor $(1 - \exp(-\hbar \omega/k_B T))^{-1}$, which increases by a factor of $\sim 3.4$ from 3.5 K to 100 K. Because this increase is not observed in the data, the most likely cause of these peaks is magnetic fluctuations. Figure 3 (b) shows the energy dependence of the $q$ integrated dynamical magnetic susceptibility $\chi''(\omega)$ below (3.5 K) and above (20 and 100 K) $T_c$. At the lower temperatures, 3.5 and 20 K, the observed $\chi''(\omega)$ shows a remarkable enhancement. Furthermore, the low-energy part of $\chi''(\omega)$ ($\omega < 6 \text{ meV}$) increases slightly at 3.5 K. In the case of FeAs-based materials, the enhancement of the peak intensity of $\chi'(q,\omega)$ below $T_c$ was interpreted to be a "resonance" as observed in high-$T_c$ Cu-oxides; however we could not find the anomalous enhancement in the data. These data simply seem to indicate that the superconducting gap opening affects the structure of the Fermi surface. We think that it is difficult to discuss the relationship between the observed enhancement of $\chi''(\omega)$ and the "resonance" in the absence of other effects on the electronic state near the Fermi surface.

The next, we will focus on the peak center for Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$. The energy evolution of the magnetic excitation spectra at 3.5 K is shown in Fig. 4 (a). For low energies ($\omega \leq 4 \text{ meV}$), the magnetic excitations of Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$ peak at a wave vector $|Q| \approx 0.9 \text{ Å}^{-1}$, which is close to the one for pure FeTe$_{0.92}$. However, as $\omega$ increase, the peak centers increase substantially up to $|Q| \approx 1.2 \text{ Å}^{-1}$ at $\omega \approx 10 \text{ meV}$. Figure 4 (b) shows the energy dependence of magnetic fluctuations for FeTe$_{0.92}$ at 65 K. Even above $6 \text{ meV}$, the inelastic scattering intensities retain their maximum at $|Q| \approx 0.9 \text{ Å}^{-1}$. (At 2 meV, the sharp intensity caused by a phonon at $|Q| \approx 1.0 \text{ Å}^{-1}$ makes it difficult to recognize the magnetic signal.) Figure 4 (c) shows the peak centers for both samples at several temperature collected at $2 \text{ meV} \leq \omega \leq 10 \text{ meV}$. In case of FeTe$_{0.92}$, the peak centers are almost constant as the function of $\omega$, which is consistent with what has been reported for other FeAs-based materials. For Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, the scattering intensities are located at $|Q| \approx 0.9 \text{ Å}^{-1}$ below $\omega \approx 4 \text{ meV}$. Above $\omega \approx 6 \text{ meV}$, the peak intensity lies at $|Q| \approx 1.2 \text{ Å}^{-1}$. This trend is nearly temperature independent. If we use a spin-wave model, we would expect the high-energy data to exhibit another intensity in the low $q$ side. This simple picture is different from the experimental results. In any case, the measurements reported here show conclusively that a significant evolution occurs in the magnetic excitation spectra when Se is substituted for Te.

At this point, we would like to note that the reciprocal lattice point $|Q| \approx 1.2 \text{ Å}^{-1}$ is close to the wave vectors $Q = (0.5,0.5,0)$ ($|Q| = 1.17 \text{ Å}^{-1}$) and $Q = (0.5,0.5,0.5)$.

---

**Fig. 3.** (a) Neutron scattering intensity for Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$ at 3.5 K (open circles) and 100 K (filled circles). (b) Energy dependence of the magnetic excitation spectra $\chi''(\omega)$ at 3.5 K ($T < T_c$), 20 K ($T > T_c$), and 100 K ($T > T_c$).
The substitution of Se for Te may cause the system to resemble that of the FeAs-based superconductors. If so, the spin excitation spectrum would resemble that of FeAs-based materials, consistent with what is observed around $\omega \approx 10$ meV. Because of the very weak intensity in Fe(Se$_{0.25}$Te$_{0.75}$)$_{0.92}$, it cannot be experimentally determined whether the regions $\omega \approx 10$ meV are continuous or separated. At the very least, however, our results show that substituting Se for Te may cause the system to possess characteristic magnetic fluctuations with the 2D nesting vector $Q = (0.5,0.5)$. We note that there is an independent experimental indication of AF fluctuation with a 2D $Q = (0.5,0.5)$ in FeSe$_{0.5}$Te$_{0.5}$. Our results on Fe(Se$_{1-x}$Te$_{x}$)$_2$ system may have significant implications on the physics of Fe-based superconductor from the comparison with FeAs system. The end compounds in both systems show quite different magnetic orderings from each other. Nevertheless the superconducting materials in both systems have the surprising common character of the proximity to an AF fluctuation with a 2D $Q = (0.5,0.5)$. This suggests that the mechanism of superconductivity in two systems may share some common features and especially that the AF correlation with a 2D $Q = (0.5,0.5)$ may play an important role in the mechanism of superconductivity in Fe-based superconductors. It is also interesting to see how the magnetic fluctuations change as the energy increases beyond $\omega = 10$ meV.

The authors thank K. Enoki and K. Ohoyama for their help in the neutron scattering measurements. We are grateful to K. Yamada and K. Horigane for useful discussions. This work was supported in part by a Grant-in-Aid for Young Scientists (B) (21740239) from the Japanese Ministry of Education, Culture, Sports, Science and Technology.

1) Y. Kamihara et al., J. Am. Chem. Soc. 130, 3296 (2008).
2) M. Rotter et al., Phys. Rev. B 78, 020503 (2008).
3) M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
4) F. Hsu et al., Proc. Natl. Acad. Sci. U.S.A. 105, 14262 (2008).
5) S. Margadonna et al., Chem. Commun. 5607 (2008).
6) M. H. Fang et al., Phys. Rev. B 78, 224503 (2008).
7) W. Bao et al., arXiv:0809.0051 (2008).
8) S. Li et al., Phys. Rev. B 79, 054503 (2009).
9) C. de la Cruz et al., Nature 453, 899 (2008).
10) Y. Qui et al., Phys. Rev. B 78, 052508 (2008).
11) M. Ishikado et al., J. Phys. Soc. Jpn. 78 043705 (2009).
12) A. D. Christiansen et al., Nature 456, 930 (2008).
13) R. A. Ewings et al., Phys. Rev. B 78, 220501(R) (2008).
14) R. J. McQueeney et al., Phys. Rev. Lett. 101, 227205 (2008).
15) J. Zhao et al., Phys. Rev. Lett. 101, 167203 (2008).
16) K. Matan et al., Phys. Rev. B 79, 054526 (2008).
17) Jon P. Wright et al., Phys. Rev. B 78, 020503 (2008).
18) P. Radhakrishna and J. W. Cable, Phys. Rev. B 54, 11940 (2008).
19) F. Ma et al., arXiv:0809.4732v1 (2008).
20) A. Subedi et al., Phys. Rev. B 78, 134514 (2008).
21) H. A. Mook et al., arXiv:0904.2178 (2009).