Spin waves in the \((\pi, 0)\) magnetically ordered iron chalcogenide \(\text{Fe}_{1.05}\text{Te}\)

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We use inelastic neutron scattering to show that the spin waves in the iron chalcogenide \(\text{Fe}_{1.05}\text{Te}\) display novel dispersion clearly different from those in the related iron pnictide systems. By fitting the spin waves to a Heisenberg Hamiltonian, we extract magnetic exchange couplings that are dramatically different from both predictions by density functional calculations and measurements on the iron pnictide \(\text{CaFe}_2\text{As}_2\). While the nearest-neighbor exchange couplings in \(\text{CaFe}_2\text{As}_2\) and \(\text{Fe}_{1.05}\text{Te}\) are quite different, their next-nearest-neighbor exchange couplings are similar. These results suggest that superconductivity in the pnictides and chalcogenides share a common magnetic origin that is intimately associated with the next-nearest-neighbor magnetic coupling between the irons.

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All parent compounds of cuprate superconductors are antiferromagnetic (AF) Mott insulators characterized by the same local moment Heisenberg Hamiltonian \(^1\). For this reason, it is believed that magnetism is important for the high-\(T_c\) superconductivity \(^2\). The iron-based superconductors \(^2,4\) share many features in common with the cuprates, which leads many to conjecture that the magnetism present in these compounds is vital for the presence of superconductivity. The iron-based superconductors can be divided into two chemical classes, the iron pnictides such as \(\text{CaFe}_2\text{As}_2\) and iron chalcogenides \(\text{Fe}_{1+y}\text{Te}\). Many properties of the pnictides and chalcogenides are similar, including similar band-structure \(^5\) and magnetic excitations in the superconducting compositions \(^6\)\(^7\)\(^8\). Furthermore, the magnetism in the pnictide parent \(\text{CaFe}_2\text{As}_2\) (Fig. 1b) is consistent with first principle density functional calculations \(^9\). However, the parent compound \(^14,15\) of the iron chalcogenides, \(\text{Fe}_{1+y}\text{Te}\), possesses a different magnetic order (Fig. 1a). Therefore, it is important to determine if magnetism in these two systems can be described by a similar Hamiltonian. If the magnetic description between systems is entirely dissimilar, then it presents a serious challenge to many theories \(^10,16\) where superconductivity has a magnetic origin.

In this paper, by studying the spin-waves in \(\text{Fe}_{1.05}\text{Te}\), we have been able to compare the magnetic couplings within the pnictide and chalcogenide systems. We show that although the nearest neighbor couplings in the two systems are very different, the effective next nearest couplings \(J_2\) are very similar. Furthermore, the isotropic \(J_2\) we find in \(\text{Fe}_{1.05}\text{Te}\) is very different from the anisotropic \(J_2\) yielded from first principle density functional calculations \(^15\). Our results suggest that while the nearest-neighbor coupling may change, it is the next-nearest neighbor coupling that persists between different iron superconductors.

We have used time-of-flight inelastic neutron spectroscopy to determine the dispersion of spin-wave excitations in \(\text{Fe}_{1.05}\text{Te}\) (with AF ordering temperature \(T_N = 68\) K, see Fig. 1a) and ref. 20, the \(x = 0\) (non-superconducting) member of the isovalently substituted \(\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x\) iron chalcogenide superconductors \(^21,22\). By measuring spin-wave excitations in \(\text{Fe}_{1.05}\text{Te}\) throughout the Brillouin zone, we have used a Heisenberg Hamiltonian to determine the effective exchange couplings of the system. Our neutron scattering experiments were carried out on the HB-1 triple-axis spectrometer at high-flux-Isotope Reactor and on the ARCS chopper spectrometer at spallation-neutron-source, Oak Ridge National Laboratory, USA. We also used MAPS chopper spectrometer at ISIS, Rutherford-Appleton Laboratory, UK. For the experiment, we have co-aligned 6 grams of single crystals of \(\text{Fe}_{1.05}\text{Te}\). All data was collected at around 10 K \((\ll T_N)\) with incident neutron energies \(E_i = 55, 90, 180, 350, 500\) and 580 meV with the \(c\)-axis aligned along the incident beam direction. Since the spin-wave excitations have weak \(c\)-axis coupling, we integrate the excitations along the \(c\)-axis direction, and focus on spin waves in the \((h, k)\) plane.

For \(\text{Fe}_{1+y}\text{Te}\) with a small amount of excess iron \(y\), the magnetic structure is bi-collinear as shown in Fig. 1h (refs. 14, 15). This can be thought of as two AF sublattices as shown by darker and lighter colored atoms. We define the nearest-neighbors \((J_{1a}, J_{1b})\), the next-nearest-neighbors \((J_{2a}, J_{2b})\), and the next-next-nearest neighbor \((J_3)\) exchange interactions as shown inFig. 1k (ref. 19). The nearest-neighbor magnetic exchange couplings \(J_{1a}\) and \(J_{1b}\) are defined similarly to those of iron pnictides (Fig. 1b). However, the next-nearest-neighbor exchange couplings \(J_{2a}\) and \(J_{2b}\) in chalcogenides are directional.

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Our Fe$_{1.05}$Te samples (where $y$ was measured with inductively coupled plasma analysis [20]) were grown using Bridgeman technique as described before [20]. Fe$_{1+y}$Te$_{1-x}$Se$_2$ is tetragonal at high temperature and becomes orthorhombic or monoclinic (depending on $x$, refs. [14, 15, 21, 22]) below the AF ordering temperature $T_N$. The ab-plane lattice parameters for the various phases remain very similar, and on cooling into the low symmetry phase the sample becomes twinned. We therefore measure the wave vector in tetragonal $(h, k, l)$ reciprocal lattice units (rlu) $Q = h2\pi/a + k2\pi/b + l2\pi/c$, with in-plane lattice parameters $a = b = 3.80$ Å, and the out-of-plane $c = 6.23$ Å. In this notation, magnetic order in powder Fe$_{1+y}$Te has been found at $(0.5, 0, 0.5)$ for small $y$, and increasing $y$ will lead to incommensurate magnetic order [14, 15]. In the present single crystalline samples, the magnetic order was found to be centered very close to the commensurate position at $(0.485, 0, 0.5)$ rlu. However, we also observed a weaker peak at $(0.37, 0, 0.5)$ rlu attributed to a small portion of the sample with slightly different $y$. Figure 1I shows the temperature dependence of the magnetic Bragg intensity at $Q = (0.485, 0, 0.5)$ rlu confirming $T_N = 68$ K.

The magnetic excitations probed by neutron scattering in our Fe$_{1.05}$Te sample are summarized by representative constant energy slices in Figure 2. The data has been normalized to a vanadium standard and plotted in absolute units, without correction for the magnetic form factor, leading to a decrease in the intensity of the signal with increased $Q$. Each $E_i$ probes a different out-of-plane wave vector for each energy transfer, and it was found that data from different $E_i$’s were consistent, implying that the $L$-dependence of the data over the energy range probed was weak.

At low energy, 7.5 meV (Fig. 2A), magnetic excitations emerge from the magnetic zone center $(0, 0, 0)$ and other half-integer reciprocal lattice vectors, where AF Bragg peaks exist at the elastic position [in an untwinned sample, magnetic peaks would not appear at $(0, 0, 5)$, but twinning leads to an equal intensity domain rotated by 90° in-plane]. As the energy is increased, the response spreads out in $Q$ as expected for spin-wave excitations (Figs. 2B and 2C). As the energy is raised to around 60 meV (Fig. 2D), there are no longer peaks at half-integer positions, but instead rings of radius 0.5 rlu which are centered on integer reciprocal lattice points. This is unlike conventional spin waves, where the magnetic response up to the highest energies is centered on the magnetic zone center, with successively larger rings with increased energy. These rings are even clearer when the data is corrected for the magnetic form factor drop-off at high wave vector (see supplementary material).

As well as the surprise that the ring center is not the magnetic center, the dispersion of the rings in the present sample are novel. Rather than a linearly increasing radius with energy, the ring radii are close to 0.5 rlu when they first appear, and as energy is increased, the radius of rings around $(0, 0)$ expand and those around $(1, 0)$ contract (Figs. 2D-2E). Even at 115 meV a ring can be seen around $(1, 0)$, which by 225 meV contracts into a peak at $(1, 0)$ (Fig. 2G) before all response disappearing at
higher energies (Fig. 2h). Corresponding cuts along the
(h, 0) trajectory are shown in Fig. 4. A schematic of
the dispersion of the magnetic response is shown in Fig.
4i. The high-energy data in Fe\textsubscript{1.05}Te have similarities to
the high-energy spin excitations in Fe\textsubscript{T}e\textsubscript{1-x}Se\textsubscript{x} with x =
0.27, 0.49 [24].

In order to extract effective exchange energies, we fit
spin-wave data using an effective Heisenberg Hamiltonian
(see supplementary material for the model Hamiltonian)
with commensurate (0.5, 0.5) antiferromagnetism [23].
In order to yield the required commensurate magnetism,
there are constraints on the bounds of each parameter
\( J_{1a}, J_{1b}, J_{2a}, J_{2b}, \) and \( J_3 \). Because of the twinned
nature of the sample, the model used is the sum of two
equal sized domains rotated by 90°.

To determine the dispersion curves for spin waves, the
two-dimensional images in Fig. 2 were cut along the
(h, 0) and (1, k) directions as shown in Fig. 3. By fitting
Gaussians to many (h, 0) cuts of different energies
like those in Fig. 3, we obtain the dispersion plot in Fig.
4k using the fitted peak positions. Similarly, (1, k) cuts
were fitted to create Fig. 4i. These two dispersion plots
were simultaneously fitted to the dispersion of the model
\( J_{1a}, J_{1b}, J_{2a}, J_{2b}, \) and \( J_3 \). Because of the twinned
nature of the sample, the model used is the sum of two
equal sized domains rotated by 90°.

In the model, the intensity of the low energy band vanishes
to zero. In the displayed fit lines \( J_{2b} \) is fixed to zero.
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wave vector \((0, 5, 0.5)\) \cite{28, 29}. Similar isotropic AF \(J_2\) values in iron-pnictides and iron chalcogenides therefore naturally explain the experimentally observed neutron spin resonance within both classes of iron-based superconductors \cite{6–12}. First principle density functional calculations \cite{19} on Fe\(_{1.06}\)Te predict highly anisotropic next-nearest-neighbor exchange interactions which are not consistent with our data (see supplementary material), perhaps due to the complex nature of the orbital ordering \cite{30, 31} or itinerant magnetism \cite{32} in this material.

In summary, we have shown that spin-wave excitations in the iron chalcogenide Fe\(_{1.05}\)Te can be modeled by a Heisenberg Hamiltonian with anisotropic (dominantly) FM nearest-neighbor and isotropic AF next-nearest-neighbor exchange couplings. While the nearest-neighbor magnetic exchange couplings for Fe\(_{1.05}\)Te and CaFe\(_2\)As\(_2\) (ref. \cite{26}) are different, we find that the AF next-nearest-neighbor exchange couplings in these two classes of materials are not only similar in magnitude but also directional independent, even though they have different AF and crystalline lattice structures \cite{14, 15, 23}. Our findings suggest that superconductivity in both classes of iron-based superconductors shares a common magnetic origin that is intimately associated with the AF next-nearest-neighbor exchange couplings \cite{27}.

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Appendix A: Data corrected for Fe\(^{2+}\) magnetic form factor

To see more clearly the evolution of spin waves in Fe\(_{1.05}\)Te in Fig. 2 of the main text, we plot in Fig. S1 constant energy slices of the data in Fig. 2 converted into units of magnetic response, \(\chi'(q, \omega)\). This is related to the measured cross-section by

\[
\frac{k_i}{k_f} \frac{d^2\sigma}{d\Omega dE} = \frac{2(\gamma r_e)^2}{\pi g^2 \hbar^2 B} |F(Q)|^2 \frac{\chi''(q, \omega)}{1 - \exp(-\hbar \omega / kT)} \tag{A1}
\]

where \(F(Q)\) is the Fe\(^{2+}\) magnetic form factor, \((\gamma r_e)^2=0.2905\) \(\text{b sr}^{-1}\), \(k_i\) and \(k_f\) are initial and final neutron wave vector and the \(g\)-factor was assumed to be 2. The exponential term is from the Bose factor, which can be neglected at the energies of interest, as all measurements were taken at 10 K. The left hand side of this equation is the raw data as plotted in Fig. 2.

Appendix B: Heisenberg Hamiltonian

For a given set of exchange energies \((J_{1a}, J_{1b}, J_{2a}, J_{2b}, J_3, J_z)\) as shown in Fig. 1a, the dispersion of magnetic excitations from the Heisenberg Hamiltonian with magnetic order at \((0.5, 0.5)\) can be found by diagonalizing the Hamiltonian \(H\) \cite{25} for every miller index \((h, k, l)\) value:

\[
H = \begin{pmatrix}
A & J_{1b}D & -B & -J_{1a}C \\
J_{1b}C & A & -J_{1a}D & -B \\
B & J_{1a}C & A & -J_{1b}D \\
J_{1a}D & B & J_{1b}C & A
\end{pmatrix}
\tag{B1}
\]

where

\[
A = 2J_{2b}\cos(k_x - k_y) + 2(J_{1a} + J_{2a} - J_{1b} - J_{2b} + 2J_3 + J_z) \\
B = 2J_{2a}\cos(k_x + k_y) + 2J_3[\cos(2k_x) + \cos(2k_y)] + 2J_z \cos k_z
\]

\[
C = e^{ik_x} + e^{-ik_x} \\
D = e^{-ik_x} + e^{-ik_y}
\]

with

\[
k_x = \pi(h + k) \\
k_y = \pi(h - k) \\
k_z = 2\pi l
\]

For a twinned sample, the full dispersion must be calculated at both \((h, k, l)\) and \((-k, h, l)\).

Appendix C: The isotropy of \(J_2\) and the effect of \(J_3\) on the spin-wave fits

As described in the paper, the dispersion data in the \((h, 0)\) and \((1, k)\) directions were obtained from fit-
FIG. S1: Constant energy slices of data (from Fig. 2) converted into units of magnetic response \( \chi''(q, \omega) \) (after subtracting constant backgrounds). After the form factor correction, the magnetic signal no longer decreases in intensity at high wave vector transfer, and so the magnetic pattern is somewhat clearer than in the raw data. However, due to a slight increase in background at large wave vectors, the slices [notably (a)-(c)] show excessive intensity at the highest wave vectors.

FIG. S2: Dispersion extracted from data (solid symbols) and fits (or simulations) of the model dispersion (lines). Left panels show dispersion in \((h, 0)\) direction, and right panels show \((1, k)\) dispersion. (a)-(b) Fit performed with all parameters free. (c)-(d) Fit with fixed \( J_3 = 0 \) and \( J_{2a} = J_{2b} \). (e)-(f) Fit with fixed \( J_3 = 0 \) and \( J_{1a} = 0 \). (g)-(h) Simulation performed with theoretical values predicted by Han et al. \[19\].

Before discussing the necessity of the next-next-nearest neighbor term \( J_3 \), we will first briefly compare our dispersion with the theoretical exchange energies predicted by Han et al. \[19\]. Figures S2(g)-(h) shows our experimental data compared with the dispersion calculated for their theoretical values, and it is clear that the data and model are very dissimilar. The model cannot be reconciled with the data because in this theory \( J_{2b} = -J_{2a} \), whereas our data appears to be best described with \( J_{2a} = J_{2b} \).

Following our successful fit with \( J_{2a} = J_{2b} \), it was important to check whether the small next-next-nearest neighbor parameter \( J_3 \) could be fixed to zero in order to further decrease the number of parameters in the fit, and see if this extra small exchange interaction is actually necessary or not. The obvious starting place is to calculate the dispersion when using the above fit parameters but with \( J_3 \) set to zero. However, the model dispersion cannot be calculated in this case because this shifts...
by a factor of 1.5 in order to show the pattern more clearly.

The intensity of the model in (q)–(x) was magnified with best fit parameters shown in Fig. 5. This fit describes the low energy data well, but is not as successful at the high energies, underestimating the maximum energy of the band by around 50 meV. We interpret this fit as compensating the lack of an AF $J_3$ term by instead causing the $J_{1a}$ term to be AF. The compromise is that $J_{1a}$ cannot be too large or the high energy data cannot be described.

One may finally ask whether the data could be fitted better if $J_3$ was fixed to zero and $J_{2a}$ and $J_{2b}$ could vary independently. In this case $J_{1a}$ was found to stay small and so was fixed to zero for ease of fitting (results do not vary significantly with a fitted $J_{1a}$ term). The fit is shown in Fig. S2(c)–(f) with parameters $J_{1b} = -35.8 \pm 2.9; J_{2a} = 14.0 \pm 0.82; J_{2b} = 8.9 \pm 1.3$, and $J_{1a} = 0; J_3 = 0$ meV). In this fit it can be seen that (i) $J_{2a}$ and $J_{2b}$ are still quite similar, and (ii) the fit is also not as good as the original ($J_3 > 0$) fit, as it again underestimates the maximum energy.

We conclude that the data is best fit with an isotropic $J_2 (= J_{2a} = J_{2b})$. The addition of a $J_3$ exchange interaction also seems to be important to describe the highest energies whilst keeping the parameters in the regime with (0.5, 0, 0.5) magnetic order. For further insight into the similarity and dissimilarities between these different fits and models, we show instrument resolution convolved simulations with $J_3$ fixed to zero [see Fig. S3(a)–(h) for simulation with parameters from the fit in Fig. S2(c)–(d), and see Fig. S3(i)–(p) for simulation with parameters from the fit in Fig. S2(e)–(f)]. These alternative parameters show similar features to our data [compare with simulation of original ($J_3 > 0$) fit in Fig. 5], though do not extend high enough in energy. In Fig. S3(q)–(x) we have also included a simulation with the Han et al. [19] theoretical parameters [see Fig. S2(g)–(h)], which have $J_{2b} = -J_{2a}$. It is clear that these parameters do not describe the data well.

**Appendix D: The extended phase diagram**

In this section, we extend the phase diagram of the $J_1-J_2-J_3$ model (see ref. 23 and Hamiltonian described in a section above) applied to FeTe$_{1-x}$Se$_x$ system to the parameter region where $J_{1a,1b}$ can be negative (providing $J_{2a} \geq J_{2b} > 0$). First, it is only trivial to observe that the original model has the following two properties:

$$U(\hat{S})^\dagger H(J_{1a}, J_{1b}, J_{2a}, J_{2b}, J_3) U(\hat{S}) = H(-J_{1a}, -J_{1b}, J_{2a}, J_{2b}, J_3),$$  \hspace{1cm} (D1)

$$U(\hat{R})^\dagger H(J_{1a}, J_{1b}, J_{2a}, J_{2b}, J_3) U(\hat{R}) = H(J_{1b}, J_{1a}, J_{2a}, J_{2b}, J_3),$$  \hspace{1cm} (D2)

FIG. S3: Instrument resolution convolved simulation of model with various exchange energy parameters. (a)–(h) fit with $J_3$ fixed to zero (and $J_{2a} = J_{2b}$), with dispersion shown in Fig. S2(c)–(d). (i)–(p) fit with $J_3$ and $J_{1a}$ fixed to zero, with dispersion shown in Fig. S2(e)–(f). (q)–(x) parameters from the Han et al. [19] prediction, the dispersion of which was shown in Fig. S2(g)–(h). For comparison with the simulation with best fit parameters shown in Fig. 5, $J_3$ does not vary dispersion significantly, only the low energy intensity, and was therefore fixed for all plots to $J_3 = 1$ meV, the value chosen in Fig. 5. The intensity of the model in (q)–(x) was magnified by a factor of 1.5 in order to show the pattern more clearly.
where $\hat{S}$ is the sublattice symmetry operation defined by $\hat{S} : \hat{S}(i, j) \rightarrow (-)^{i+j} \hat{S}(i, j)$ and $\hat{R}$ is a lattice rotation along the $a$-axis of angle $\pi$, that is, $\hat{R} : \hat{S}(i, j) \rightarrow \hat{S}(j, i)$.

This fact tells us we only need to focus in the parameter region where $J_{1a} > |J_{1b}| > 0$, and the phases in other regions can be obtained by applying the appropriate symmetry operations on the ground state within this parameter region. For example, if one has $J_{1b} < J_{1a} < 0$, first we obtain the ground state for the Hamiltonian $H(-J_{1b}, -J_{1a}, ...)$ and then transform the state under the symmetry operation $\hat{S} \times \hat{R}$.

Second, as we have thoroughly discussed the phase diagram with $J_{1a,1b} > 0$ in the previous paper [22], we only need to discuss the case where $J_{1a} > 0$ while $J_{1b} < 0$ in this note. This discussion is very simple because in this parameter region only one parameter, $J_{2b}$, is frustrated in the AFM3 phase, therefore the only possible instability is the one toward the ICB phase, the transition line of which is given by

$$J_{1a} - J_{1b} = 4J_{2b} - 8J_3.$$  \hspace{1cm} (D3)

The full phase diagram is given in Fig. S4.

We note that the effective exchange couplings are very close to the phase boundary between the $(0, 5, 0)$ AF ordered state and two different incommensurate magnetic states with wave vectors $(0.5 - \delta, 0)$ and $(0.5 - \delta, 0.5 + \delta)$. This is also consistent with the fact that the transition from the $(0, 5, 0)$ AF state to the incommensurate $(0.5 - \delta, 0)$ magnetic state is observed in Fe$_{1+y}$Te by increasing $y$ (refs. 14, 15). There is also evidence that FeTe$_{1-x}$Se$_{x}$ can have $(0.5 - \delta, 0.5 + \delta)$ incommensurate spin excitations [24, 34, 35].