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## PAPER

Pressure induced evolution of superconductivity and magnetic hourglass dispersion in  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$ 

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Iron based high temperature superconductors have several common features with superconducting cuprates, including the square lattice and the proximity to an antiferromagnetic phase. The magnetic excitation spectrum below  $T_c$  of  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  shows an hourglass-shaped dispersion with a resonance around the commensurate point. In a previous inelastic neutron scattering study, we showed that the hourglass-shaped dispersion is most likely a prerequisite for superconductivity, while the consequences are the opening of a gap and a shift of spectral weight. In this paper we follow the evolution of the hourglass shaped dispersion under applied pressure up to 12 kbar. Our results show that the pressure-induced 37% increase of  $T_c$  is concomitant with a change in the magnetic excitation spectrum, with an increase of the hourglass energy by 38%.

**1. Introduction**

Iron based chalcogenides and oxypnictides started attracting attention with the discovery of high temperature superconductivity in the doped iron compound  $\text{RFeAsO}$  [1, 2], further fuelled by their striking similarities to the high  $T_c$  cuprates which go beyond a layered structure with a square lattice and a common proximity of magnetic and superconductive state [3]. As in the cuprates (for a review see [4]), a spin resonance is observed in the inelastic excitation spectrum below  $T_c$  in both iron chalcogenide and pnictides compounds [5].  $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$  has the simplest layered crystallographic structure among the iron-based superconductors and displays a spin excitation spectrum with a spin resonance, an hour-glass shape dispersion and a spin gap which accompany the onset of superconductivity. References [6–8] in addition to the Te/Se ratio as a tuning parameter, the excess of iron (parameter  $y$ ) located on interstitial sites strongly influences both the magnetic and the superconducting properties of  $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$  [9] and its study provides new insights into the superconducting mechanism. In addition, parent compounds show peculiar magnetic behavior upon application of pressure such as pressure-induced ferromagnetism in antiferromagnetic  $\text{Fe}_{1+y}\text{Te}$  [10], indicating a strong correlation between the crystal structure and the magnetic properties. Further investigations of the interplay between magnetism and unconventional superconductivity are thus possible by the application of pressure, a parameter with the advantage of tuning superconductivity properties [11, 12] without sample composition changes, avoiding potential changes in doping-induced inhomogeneity and the hard to control level of excess iron.

In a previous study [8], we reported the magnetic excitation spectrum in single crystals of  $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$  at zero applied pressure.  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  with  $y = 0.02$  is a bulk superconducting material with  $T_c \sim 10\text{K}$ . We found a magnetic hour glass dispersion with the constriction of the incommensurate spectrum towards the

commensurate wavevector  $Q = (0.5, 0.5, 0)$  occurring at an energy of  $E_g = 5.3(5)$  meV. This commensuration clearly developed above  $T_c$ , while the temperature dependence of the spin gap showed a rapid depletion of intensity below  $T_c$ . This implies that the commensuration would be a prerequisite for superconductivity, while the spin gap is a consequence. This was further confirmed by the incommensurate spectrum of the non superconducting sample  $\text{Fe}_{1.05}\text{Te}_{0.7}\text{Se}_{0.3}$  ( $y = 0.05$ ), where no hourglass shape was observed in the magnetic spectrum. We characterized the magnetic spectrum by three energies:  $E_{\text{hg}} = 5.3(5)$  meV the energy of the commensurate hourglass position;  $E_{\text{gap}} = 3.7(5)$  meV, below which the spectral weight is depleted in the SC state; and  $E_{\text{max}} = 7.5(5)$  meV the energy of the maximum spectral weight in the SC state. We suggest that  $E_{\text{hg}}$  sets an upper limit for  $T_c$  and in this particular case the transition temperature of  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  is close to that limit. Here we investigate how respectively  $T_c$  and the magnetic excitation spectrum develops under the application of pressure in the SC sample  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$ .

## 2. Experimental method and samples

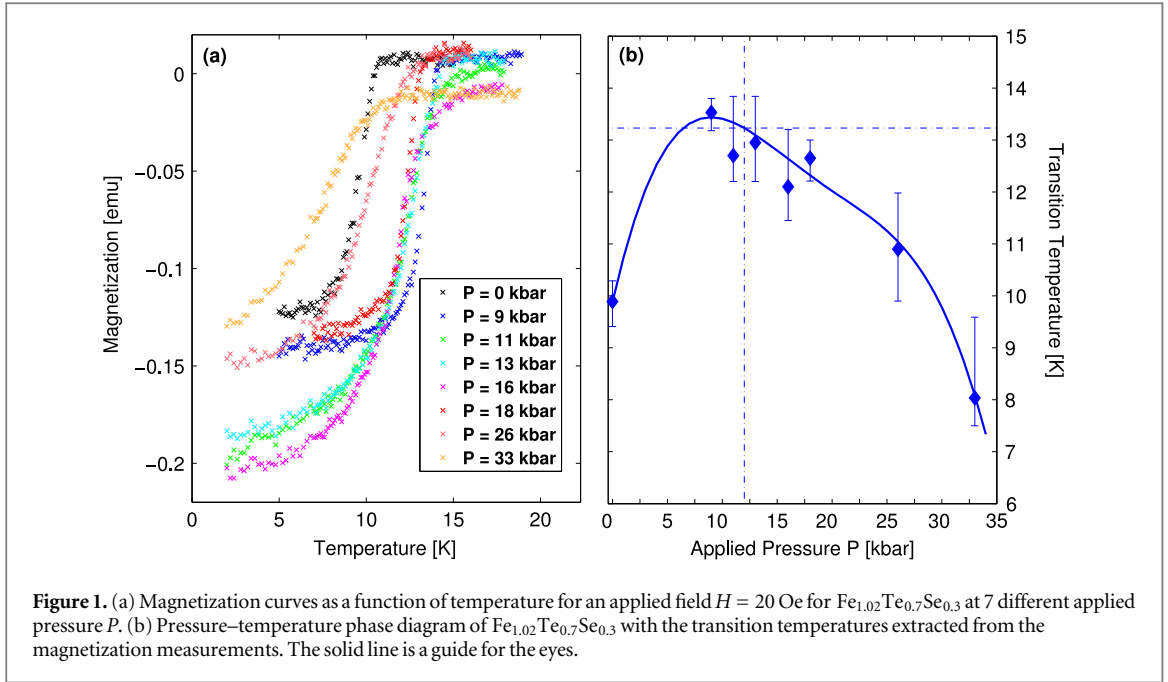
Magnetization measurements were performed in a SQUID magnetometer (quantum design) with the sample loaded in a diamond anvil pressure cell. The  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  sample was cut with a laser to a diameter of  $300 \mu\text{m}$  and a thickness of  $100 \mu\text{m}$ , which corresponds to the maximum size of the gasket hole. The magnetization of  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  was measured with  $H = 20$  Oe for eight applied pressures in the range 0–33 kbar. The background signal of the pressure cell was measured by a temperature scan of the empty cell at  $H = 20$  Oe and subtracted from the obtained data. As the signal from the sample is small when compared to the background, the variation in absolute value of the magnetization as a function of pressure cannot be quantitatively analysed.

Inelastic neutron scattering was performed on the high-flux thermal neutron spectrometer IN8 at the ILL, France in the same configuration as in our previous measurement [8]. A single crystal of  $\sim 30 \text{ mm}^3$ , from the same batch as the magnetization measurements, was encapsulated in Pb powder which was confined inside a circular TiZr gasket. The gasket was loaded between Cd shielded boron nitride anvils of a Paris–Edinburgh cell, which was mounted inside a modified cryostat of the ILL ‘orange’ type. The base temperature of  $T = 5$  K was reached by first filling the sample chamber with liquid nitrogen, pumped out at  $T = 77$  K, and further cooling with the help of a cold head employing the Gifford–McMahon refrigeration cycle. The pressure was applied *in situ* by pressing the anvils with an externally connected He compressor stage. The Pb served on the one hand as hydrostatic pressure transmitting medium, on the other hand as gauge for the pressure calibration at the sample position by following the pressure dependent scattering angle shift of the (111) nuclear reflection of Pb. This experimental setup gave satisfying signal to noise ratios even for the small sample volume in use. Nevertheless, acquisition times of approximately 1 h/point were necessary to reach comparable statistics as in our previous experiment, therefore severely limiting the number of scans that could be collected. Measurements were performed at  $P = 12$  kbar and  $T < 6$  K. These results were compared with a former experiment on IN8 at zero applied pressure (detailed method in [8]).

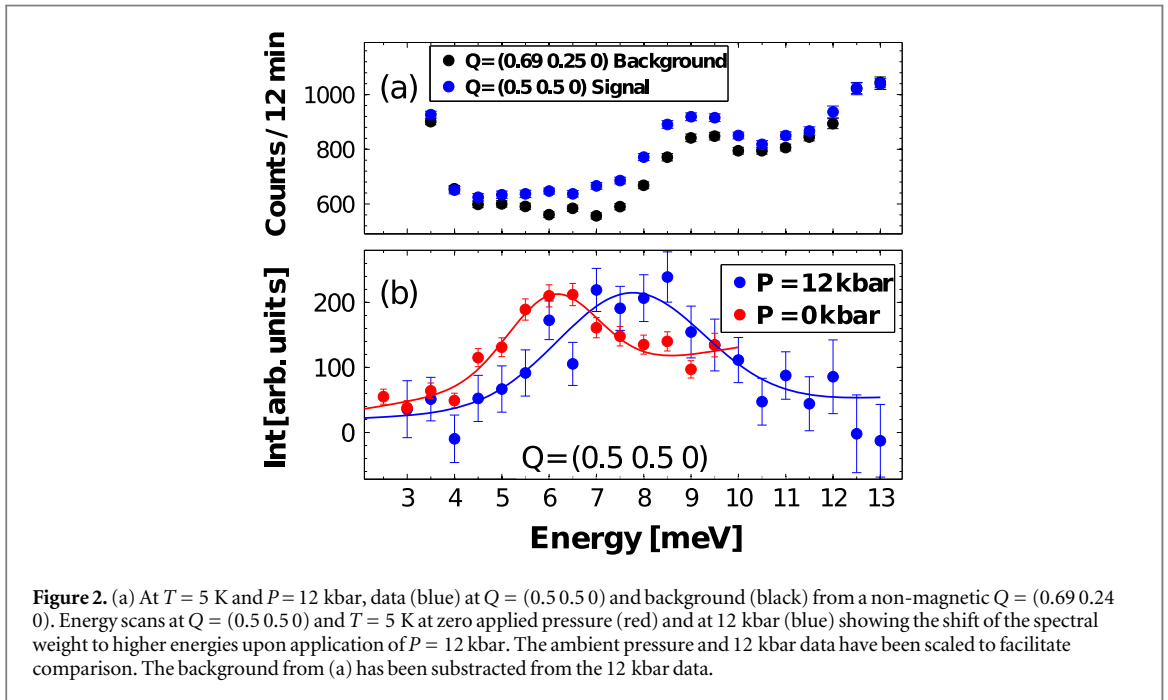
## 3. Results

Figure 1(a) shows the magnetization curves as a function of temperature for eight applied pressures from which the superconducting transition temperatures were extracted. The resulting Pressure–temperature phase diagram presented in figure 1(b) shows the rise of the superconducting transition temperature from  $T_c = 9.7$  K at zero applied pressure to a maximum of  $T_c = 13.3 \pm 0.4$  K at  $P \sim 10$  kbar (shown by the dashed lines) and then a decrease of  $T_c$  upon further application of pressure. A comparable Pressure–temperature phase diagram was obtained for a richer Se composition in [11], with a stronger enhancement of superconductivity with pressure. This is to be expected, as magnetic order is strengthened by pressure in Te-rich compositions [13], and the competition between magnetism and superconductivity can be tuned by lowering the Te/Se ratio. Similar behaviour was found for other members of the iron Chalcogenide family such as undoped FeSe [14, 15], where pressure enhances spin fluctuations and drives  $T_c$  to a much higher value [16], and in  $\text{FeTe}_{0.6}\text{Se}_{0.4}$  [17], where the authors attribute the shift of the SC phase transition with applied pressure to an increased hybridization and weakening of the coupling strength.

In order to study the evolution of the three energy scales in the excitation spectrum with pressure,  $P = 12$  kbar was applied to the  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  sample. This pressure is slightly above  $P_{\text{max}}$  determined from magnetization measurements, but  $T_c$  is very close to the maximum value. Figure 2 shows neutron energy scans at the commensurate position  $Q = (0.5, 0.5, 0)$  at 5 K for both zero applied pressure and 12 kbar. In the zero applied pressure case, the maximum of the spectral weight at the commensurate position is found at  $\sim 6$  meV, just above  $E_{\text{hg}}$ . Under the application of a 12 kbar pressure, the commensurate magnetic peak is shifted towards higher energy transfers with the maximum of the spectral weight at  $\sim 7.5$  meV.



**Figure 1.** (a) Magnetization curves as a function of temperature for an applied field  $H = 20$  Oe for  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  at 7 different applied pressure  $P$ . (b) Pressure–temperature phase diagram of  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  with the transition temperatures extracted from the magnetization measurements. The solid line is a guide for the eyes.

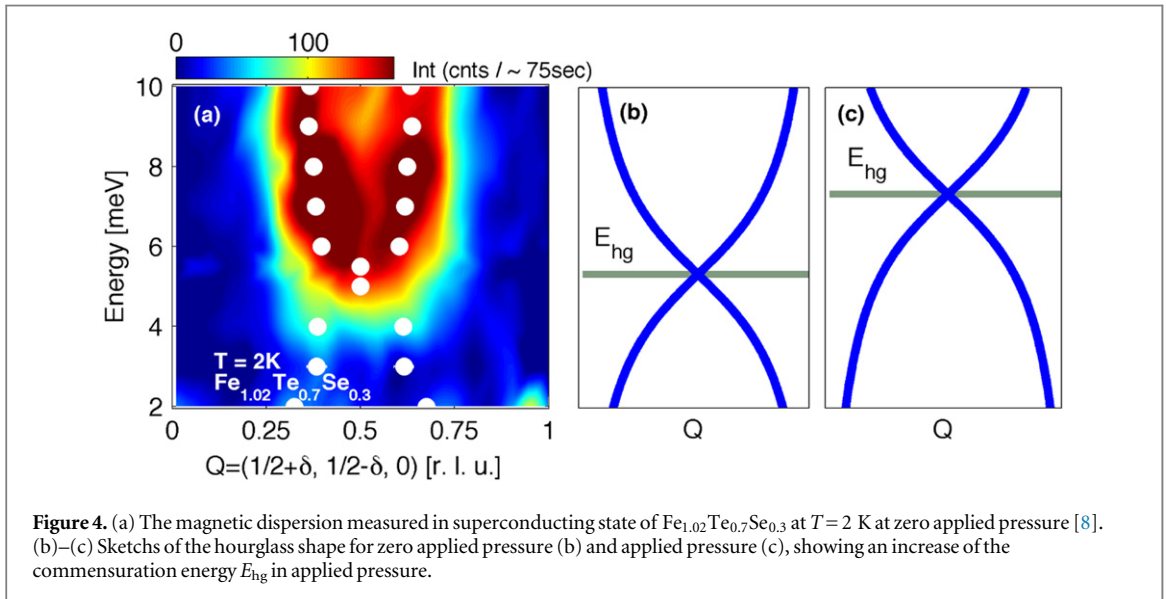
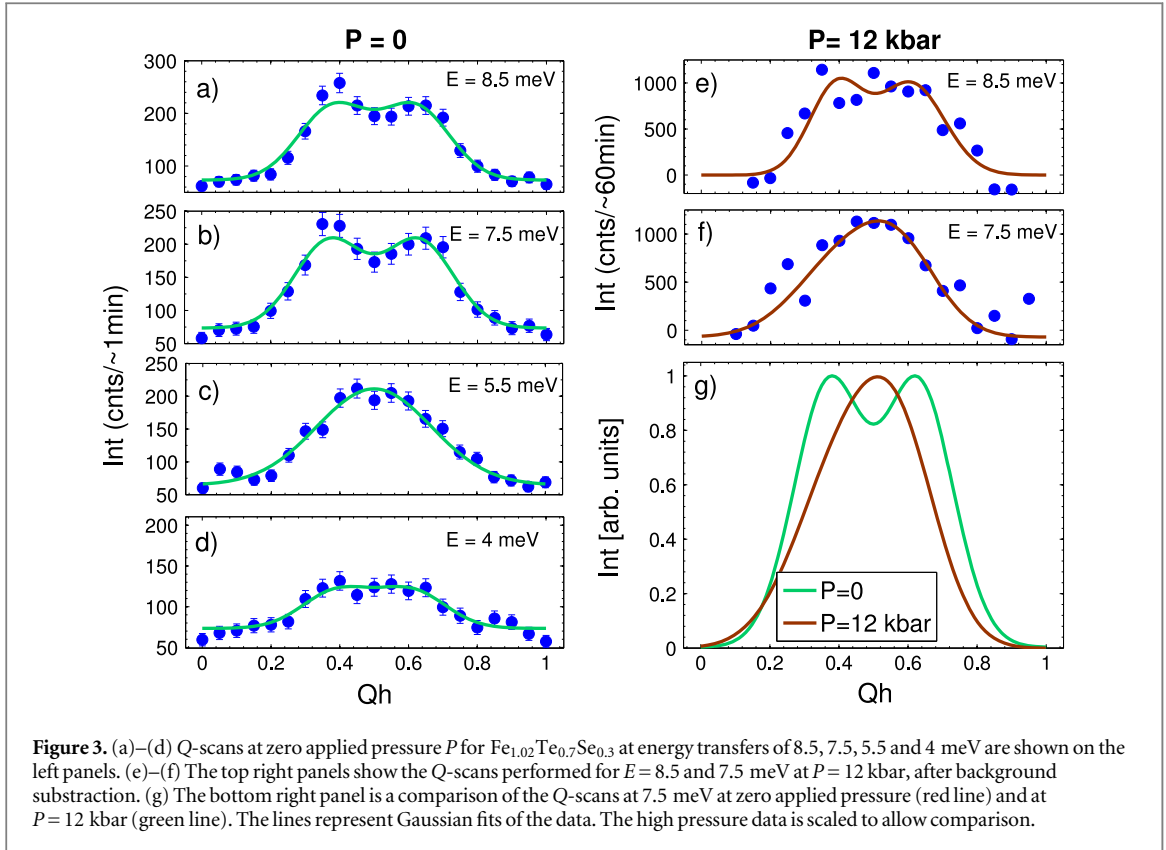


**Figure 2.** (a) At  $T = 5$  K and  $P = 12$  kbar, data (blue) at  $Q = (0.5, 0.5, 0)$  and background (black) from a non-magnetic  $Q = (0.69, 0.24, 0)$ . Energy scans at  $Q = (0.5, 0.5, 0)$  and  $T = 5$  K at zero applied pressure (red) and at 12 kbar (blue) showing the shift of the spectral weight to higher energies upon application of  $P = 12$  kbar. The ambient pressure and 12 kbar data have been scaled to facilitate comparison. The background from (a) has been subtracted from the 12 kbar data.

Further inelastic  $Q$ -scans show magnetic excitations at the incommensurate positions  $Q = (1/2 \pm \delta, 1/2 \mp \delta, 0)$ , for both the sample at zero applied pressure and the sample at 12 kbar. Figures 3(a)–(d) show  $Q$ -scans for the zero applied pressure data. We observe two distinct incommensurate peaks dispersing towards each other only merging at  $E = 5.5$  meV, which corresponds to the resonance.

Figures 3(e)–(f) shows  $Q$ -scans performed at  $E = 7.5$  meV and  $E = 8.5$  meV at 12 kbar pressure. While at  $E = 8.5$  meV, a two peak incommensurate spectrum with  $\delta = 0.09$  is clearly observed, only one broad gaussian models the data at  $E = 7.5$  meV. A direct comparison of the  $Q$ -scans at  $E = 7.5$  meV performed at zero applied pressure and at applied pressure  $P = 12$  kbar are also shown on panel (g) after scaling.

Using the zero applied pressure results [8] to interpret the 12kbar pressure data, we obtain the schematic picture of the pressure-induced change to the excitation spectrum, shown in figure 4. The magnetic dispersion measured in the superconducting state of  $\text{Fe}_{1.02}\text{Te}_{0.7}\text{Se}_{0.3}$  at zero applied pressure and  $T = 2$  K is shown in figure 4(a). The schematic evolution of the hourglass shape with applied pressure illustrates the increase of  $E_{\text{hg}}$ ,



corresponding to a shift of the hourglass dispersion towards higher energy transfers. By combining the energy and Q-scans to the  $P = 0$  data, we estimate the 12 kbar commensurate energy  $E_{\text{hg}} = 7.3 \pm 0.6$  meV.

#### 4. Discussion

From the zero pressure study [8], we interpreted that because it exists above the transition temperature, the hourglass-shaped dispersion may be a necessary condition for the high-temperature superconductivity. One possible natural explanation is that the inwards dispersion allows the spin gap to shift spectral weight towards the commensurate point, thereby lowering the exchange energy [18]. A consequence of this interpretation is that the commensurate energy  $E_{\text{hg}}$  sets an upper limit for the possible spin gap and hence for  $T_c$ . This interpretation is

strongly supported by the current measurements. The observed 37% increase in  $T_c$  from 9.7 to 13.3 K is accompanied with an similar 38% increase in  $E_{hg}$  from 5.3(5) to 7.3(6) meV.

Naturally, the hourglass energy is not the only limitation to  $T_c$ , which is illustrated for example in the  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  family ( $T_c^{\text{max}} = 39$  K), where the hourglass energy is 40–50 meV [19], similar to  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  ( $T_c^{\text{max}} = 93$  K).  $T_c$  in LSCO is presumably suppressed by competing spin and charge stripe phases [20]. In this respect, the hourglass energy can indicate the upper limit that can be achieved in a family of high-temperature superconductors, hence providing guidance to whether searching for chemical composition variations may prove fruitful.

This leads us to conjecture a possible materials discovery strategy for finding new families of high-temperature superconductors. From transport or Meissner effect measurements alone it is easy to recognize if a new material is a superconductor, but difficult to know whether a material is close to being a superconductor. Screening new materials for incommensurate and hourglass shaped dispersions would allow to identify materials which may compositionally be close to superconducting compounds. Note that our hypothesis is that the hourglass dispersion is a necessary but not sufficient condition for superconductivity. For instance, the cobaltates show an hourglass spectrum [21], but remain insulating for all doping levels. An interesting challenge would be to find a way to mobilize carriers in this system. Evolution of modern neutron sources, especially the European Spallation Source, and new instruments specifically designed for small samples will render measurements possible on  $<1$  mm<sup>3</sup> samples [22] and could make such materials discovery strategies feasible.

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