

Topology meets time-reversal symmetry breaking in $\text{FeSe}_{1-x}\text{Te}_x$ superconductors

Corresponding Author: Professor Takasada Shibauchi

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors report a SR study of time-reversal symmetry breaking in the $\text{FeSe}_{1-x}\text{Te}_x$ system. Time-reversal symmetry breaking in this system has already been studied several times as the authors note. Thus, the statement at the bottom of page 7, "Most importantly we discovered the bulk TRSB superconductivity in the tetragonal phase at $x = 0.6$ " seems a bit of a stretch. Here the authors are using a bulk probe and therefore suggest the TRSB is a bulk phenomenon. The other studies used surface probes and suggested the observed TRSB is a surface phenomenon. I do not believe that the present study provides the last word on this discussion. I should explain.

Let us imagine that there is indeed a TRSB superconducting order parameter in the bulk. Further let us imagine that this is in the region of the phase diagram where we have a large spin-orbit component, as the authors suggest in their phase diagram. Now we note that there are several theoretical studies that suggest that the inversion symmetry breaking at the solid-vacuum interface combined with a large spin orbit interaction can allow, for instance, the mixing of singlet (their favored ordering at low doping) with triplet superconducting order parameters. I am not saying that such an interaction takes place but the present study certainly does not rule out such a possibility.

The authors present a phase diagram for the material. The phase diagram is probably perfectly valid. On the left-hand side, we see a continuation of the authors previously reported study of the $\text{FeSe}_{1-x}\text{S}_x$ system. Above a doping level of approximately $x = 0.4$, the topological phase enters the discussion as reported previously. Above approximately $x = 0.7$ the topological phenomenon starts to disappear and eventually all superconductivity is lost. The latter is consistent, as the authors note, with previous neutron scattering studies that show an AFM magnetic ordering at the high Te concentrations, the magnetic ordering, initially short range, competing with the TRSB superconductivity.

In summary, I think this paper contributes to the discussion on this interesting problem and should certainly be published. I do not think it offers the slam-dunk that the authors appear to believe.

Reviewer #2

(Remarks to the Author)

The report is in the attached file.

Reviewer #3

(Remarks to the Author)

The manuscript reports muon spin rotation measurements of the microscopic magnetic fields in a family of iron-chalcogenide superconductors (FeChSC), $\text{Fe}(1+y)\text{Se}(1-x)\text{Te}(x)$, $x = 0.35, 0.6, 0.7, 0.8$, and 0.9 . In these materials, non-trivial topological electronic band structure results in a surface state with potential for topological surface superconductivity and Majorana states. The goal of the reported measurements is to identify a time-reversal symmetry breaking (TRSB) superconductivity in the bulk of FeChSC , which has significant implications for the Dirac surface state. In particular, TRSB could explain the gap opening below T_c observed in previous experiments. In general, identifying superconductivity with TRSB is an important and timely problem of broad interest. In the presence of topological band structure TRSB may lead to new states such as quantum anomalous Hall insulator or an axion insulator. Hence, the motivation for the reported research is strong and well-argued, justifying the submission to a high-impact journal such as Nature Communications.

On the other hand, the experimental results presented in the manuscript in my opinion lack sufficient impact to justify publication in Nature Communications. In my opinion, many of the reported conclusions lack sufficient experimental substantiation, while some observations raise questions. One question of specific importance is that of chemical composition of the studied samples, which has not been carefully characterized. As previous studies have shown, both magnetic and superconducting properties of the $\text{Fe}_{1+y}\text{Se}_{1-x}\text{Te}_x$ family depend very sensitively on Fe off-stoichiometry, y . In the present work, however, authors simply rely on an assumption that Te-annealing allows to reduce y to zero. This is clearly insufficient: some sort of compositional characterization of the studied samples establishing their chemical composition is needed.

For example, authors do acknowledge that much of their observations in the $x=0.9$ sample where the magnetic volume fraction is estimated to be approximately 90% at low temperatures are similar to those reported in previous studies of $\text{Fe}_{1+y}\text{Se}_{1-x}$ with $\sim 3\%$ excess Fe, which exhibits incommensurate AFM order [44]. Magnetic susceptibility shown in the Supplementary also looks consistent with the incommensurate magnetism observed in Fe-rich samples. Is it possible that authors indeed observe incommensurate magnetism in their $x=0.9$ sample? Is it due to 10% Se doping or due to an uncharacterized excess Fe? How would the latter impact the superconducting phase?

More importantly, the central question of whether the microscopic magnetic fields which authors observe by μSR belong to a uniform superconducting phase with TRSB or perhaps result from a phase segregation into a superconducting and magnetic phase, is not clearly answered by the authors' results.

Here, aside from interleaved magnetic phases induced by interstitial Fe ions, authors need to rule out the possibility that nano-scale magnetic inclusions of ferromagnetic or super-paramagnetic Fe, or Fe_3Te_4 nano-particles are at the origin of the observed microscopic magnetic fields. Being embedded in a bulk superconducting phase, such magnetic nano-inclusions could induce screening currents and thus change magnetic field configurations in the sample at the superconducting transition. Convincingly ruling out such a possibility that observed behaviors result from chemically distinct magnetic phases or inclusions is needed in order for the manuscript to be reconsidered.

Finally, I have several technical remarks which I would like authors to address.

Page 4, first paragraph. "In this series, only the SC3 phase with the highest T_c maximum includes the topological region." - Perhaps, reference [35] would also be appropriate here.

Page 8, second paragraph. " $B_{\text{int}} = 0.09$ (0.62) G for $x \sim 0.35$ and 0.76 (1.53) G for $x = 0.6$, respectively" - do numbers in parentheses denote uncertainties, and if so, the last number would be consistent with zero? If these are results of the different, NSR and SR measurements, which seem to be indicated by follow-up text, each should be given with its own corresponding error. The difference between the two results is still large enough to question assertions based on comparing the internal fields from different measurements and therefore questions the relevance of entire discussion in this paragraph and elsewhere.

Similarly, "The present results that the magnitude of the TRSB field for $x \sim 0.35$ is almost identical to that of FeSe suggest that a similar TRSB superconducting state extends inside the nematic phase of $\text{FeSe}_{1-x}\text{Te}_x$." - how warranted is this statement given the large error bars?

Supplementary: Based on magnetization measurements, authors note that the $x = 0.6$ sample has almost 100% superconducting volume fraction, while for $x \geq 0.7$ samples it is lower ($\sim 60\%$). Are these values consistent with the μSR data? Can superconducting volume fractions be determined from the authors' μSR measurements?

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors respond to my comments in their response but not in the paper. In the context of the paper, the latter would be a bit of a challenge. As such I have no problem with the paper being published. It does bring new and interesting observations to the discussion.

Reviewer #2

(Remarks to the Author)

The revised manuscript has been improved, but I am not completely satisfied with the author's reply regarding LF. In particular, one can see in Fig.2f that in the normal state, the relaxation rate is indeed nearly completely suppressed by the application of 200G LF. In contrast, at the lowest measured temperature, the increase of the relaxation above the normal state value is nearly 50% of the ZF $\Delta\lambda = \lambda(T \rightarrow 0) - \lambda(T > T_c)$. This indicates that even 200G cannot decouple muon spins from fields associated with TRSB. This is a bit unexpected, given that the estimated internal field is $B_{\text{int}} = 0.62$ G only. So, to distinguish between the static or dynamic nature of the TRSB fields a more detailed study at different LF is required. These experiments would probably take a long time, given the muon facility schedule. Therefore, I think an alternative approach prior to publication would be to soften the paper's claims and discuss different possibilities on equal footing for the enhanced relaxation rate in the main text, including AFM fluctuations and the appearance of a weak FM on the surfaces and around defects. In particular, based on the detailed Kerr effect study, it was concluded that TRSB is related to weak surface FM state but not superconductivity [Phys. Rev. Lett. 130, 046702 (2023)]. The authors cite the paper but are absolutely silent about the contradictions in their interpretations.

Reviewer #3

(Remarks to the Author)

In the revised manuscript and in their responses, authors have addressed substantial fraction of concerns raised by the Reviewers. Commendably, they have characterized the composition of their samples, which clears large number of questions raised by the original manuscript and makes the presented work more accurate and comprehensive. I am, however, still hesitant to recommend the manuscript for publication because I am not convinced by the authors' response to one of my major concerns. Namely, the need to rule out a possibility that nano-scale magnetic inclusions of ferromagnetic or super-paramagnetic Fe, or Fe₃Te₄ nano-particles are at the origin of the observed microscopic magnetic fields. I noted that convincingly ruling out such a possibility, that observed behaviors result from chemically distinct magnetic phases or inclusions, is needed in order for the manuscript to be reconsidered. Authors' argument about putative Kondo screening is not applicable to phase inclusions rather than atomic magnetic impurities. Additionally, ferro-/ferrimagnetic phase inclusions such as Fe₃O₄ or Fe₃Te₄ would not lead to any sort of paramagnetic behaviors, so the absence of such paramagnetic effects would not rule out these. If authors could supply convincing evidence that microscopic magnetic fields observed in their muSR experiments do not arise from the combination of superconductivity and magnetic impurities, I would recommend the manuscript for publication in Nature Communications.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors partially followed my suggestions. In particular, they clearly acknowledged the observation of a weak FM state in the polar Kerr experiments. Future studies need to investigate how this state is related to the TRSB state in the bulk. Given the novelty of the data, I recommend the manuscript for publication.

Reviewer #3

(Remarks to the Author)

In their response to my concern about possible role of nano- or micro-inclusions of magnetic phases such as Fe₃O₄ or Fe₃Te₄, authors argue as follows. "In a muSR study on polycrystalline samples of FeSe_{0.5}Te_{0.5}, which are reported to contain ~0.5-1.0% volume of the Fe₃O₄ impurity phase, again show no change of ZF relaxation rate above and below T_c [P. K. Biswas et al., Phys. Rev. B 81, 092510 (2010)]. This also supports that the inclusions of magnetic nano-particles the reviewer mentioned do not affect the muSR relaxation at the superconducting transition. Thus, it is highly unlikely that the observed enhancement of ZF-muSR relaxation rate in the superconducting state of our well-characterized crystals arises from the combination of superconductivity and magnetic impurities." While the cited reference to a previous study alleviating my concern is somewhat convincing, I think this issue needs to be clarified in the main text and not in the supplementary where authors have addressed it. This is because potential influence of magnetic inclusions can undermine the credibility of the reported results and therefore is central to establishing this credibility. As such, it needs to be addressed in the main manuscript. I also recommend that some passages are revised to tone down statements that are not supported by the presented experimental data. For example, "Our findings provide a fundamental understanding of the complex relationship between the TRSB superconductivity, nematicity, topology, and magnetism in FeSe-based superconductors." A very similar statement is repeated later, in the conclusion, "... our findings provide fundamental information for our understanding of the complex relationship between the superconductivity, nematicity, topology, and magnetism in the iron-chalcogenide superconductors". I think these are overstatements in the context of the results presented in the manuscript, and also redundant. I also noticed a couple of typos in the manuscript, so another careful read would be warranted. Other than that, I think that the manuscript passes the bar and can be accepted for publication.

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Reply to Reviewer 1

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In summary, I think this paper contributes to the discussion on this interesting problem and should certainly be published. I do not think it offers the slam-dunk that the authors appear to believe.

We thank Reviewer #1 for evaluating our manuscript and for the positive comments that “I think this paper contributes to the discussion on this interesting problem and should certainly be published”. In the following, we address the issue on the surface vs bulk the reviewer raised.

We agree to the reviewer that at the surface, the inversion symmetry breaking can lead to the mixing of the even- and odd-parity order parameters and thus the superconducting state may be strongly different from the bulk. This point is indeed intriguing and deserves further studies. However, here we focus on the nature of superconducting state deep in the bulk, because our experimental condition with the muon energy of ~ 4.1 MeV implies that we are probing the region around an averaged stopping depth of ~ 320 μm from the incident surface. This depth is orders of magnitude longer than the coherence length (< 100 nm), and we used mosaic crystals with a total thickness of > 0.5 mm. Thus, the signals come from the bulk and essentially we have no information on the surface states in our measurements. We have added this information in the revised Supplementary Information.

We note that recent advances on the μSR measurement technique allow to use much lower energies down below 10 keV, from which information on the surface magnetization can be obtained [J. N. Graham *et al.*, Nat. Commun. **15**, 8978 (2024)]. The comments by the reviewer motivate us to go to this direction as our next target, but we believe that the present finding in our bulk measurement is an important input to this intriguing issue as the reviewer acknowledges.

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Reply to Reviewer 2

The paper presents the systematic μSR study of high-quality $\text{FeSe}_{1-x}\text{Te}_x$ single crystals with various doping levels. This system represents a large scientific interest due to the natural realization of topological superconductivity at $x \sim 0.5$. The central conclusion of the study is that the superconducting state breaks spontaneously time-reversal symmetry in a broad range of Te substitution levels, at least for samples with $x < 0.7$. I have found the data to be novel and of high quality, but I am not convinced that TRSB is indeed related to superconductivity for the following reasons.

We thank Reviewer 2 for the detailed review and helpful comments. We appreciate the reviewer's acknowledgement that our data are novel and of high quality. We take the reviewer's comments seriously and address them with a new figure as explained below. Please note that we made additional EDX measurements to determine the actual Te compositions in the Bridgman-grown crystals (see revised Supplementary Information), so we take the determined compositions for x values in the revised manuscript.

1. The relaxation rate gradually increases with temperature decrease, follows closely Curie-Weiss behaviour for $x = 0.35$ and 0.6 (see Fig. 1 below) instead of order parameter-like behaviour expected for SC TRSB state (as observed for S_2RuO_4 [Nature Vol. 394, 558-561 (1998)]).

To address the valid concern of the reviewer, we compare the relaxation rate data for $x \sim 0.35$ and $x = 0.64$ obtained by the zero-field (ZF) and transverse-field (TF) measurements in Fig. R1 (which is the same as the newly added Fig. S5 in the revised Supplementary Information). The TF data represents the temperature dependence of the superfluid density (an order parameter of superconductivity), which is found to be scaled roughly to that of the ZF relaxation rate. When TRS is broken in the superconducting state, the TRSB field is expected to appear near the defects or domain boundaries and the field profile is essentially determined by the supercurrent around them. Thus, the rough scaling between the superfluid density and ZF relaxation rate strongly supports that the observed temperature dependence of ZF data comes from the spontaneous TRSB field. Moreover, if the paramagnetic impurities are present to produce Curie-like $1/T$ dependence in the susceptibility, such dependence should be observed in the TF data above T_c , but the data show that the TF relaxation rate is independent of temperature, which rules out this possibility. This is reinforced by the absence of low- T upturn in the magnetization data for $x = 0.64$ in Fig. S3d.

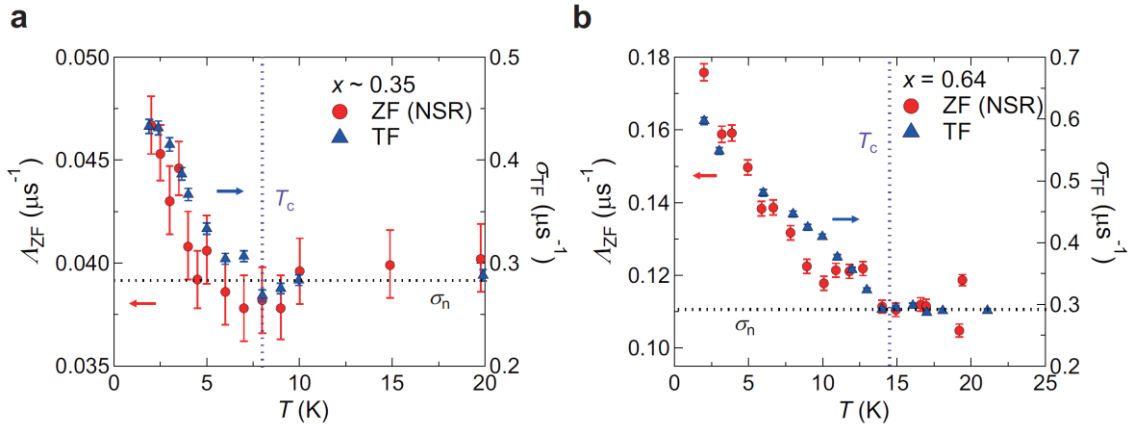


Fig. R1 Comparisons of temperature dependence of relaxation rate in the ZF and TF measurements for $x \sim 0.35$ (a) and $x = 0.64$ (b).

2. LF of 100G and 200G obviously do not decouple muon spin completely for $x = 0.35$ and $x = 0.6$. It is seen in Fig.2 of the manuscript that the relaxation rate is no-zero in LF; however, it is zero in Fig. 2e. Could the author explain how LF relaxation was obtained?

Usually, an even lower field is sufficient for decoupling such small internal fields [Nature Vol. 394, 558-561 (1998)].

The LF relaxation rate we have obtained from the data in Fig. 2a for $x \sim 0.35$ is $0.00054 \mu\text{s}^{-1}$ at 2 K, which is much lower than the ZF value and very close to the bottom (but not zero) in the scale of Fig. 2e. Detailed procedures of the fits have been described in Section IIIB in the Supplementary Information. For $x=0.64$, we obtained a larger LF relaxation rate of $\sim 0.030 \mu\text{s}^{-1}$ at 2 K (which is comparable to that reported in the LF measurements of Sr_2RuO_4 the reviewer mentioned), but it is much lower than the ZF value of $0.18 \mu\text{s}^{-1}$. This large difference between the ZF and LF rates for $x=0.64$ is in sharp contrast to the cases of $x=0.75$ and 0.83 , indicating that the ZF relaxation is dominated by the static field on the microsecond timescale. From these results, together with the discussion in the above reply to the first comment, we conclude that the temperature dependence of ZF relaxation rate for $x \sim 0.35$ and $x=0.64$ comes from the TRSB field in the superconducting state.

I think the authors need to provide stronger evidence before the manuscript can be published that the TRSB state is indeed related to superconductivity. One possibility is to demonstrate that the relaxation rate will be suppressed if superconductivity is suppressed by performing Knight shift measurements in strong magnetic fields, similar to the one reported in ref. [Nature Communications 153 (2023)].

We believe that we have addressed the issues raised by the reviewer. We thank for the reviewer's comments which we find valuable to improve our manuscript. We further thank the reviewer's suggestions on the Knight shift measurements in strong magnetic fields with suppressed superconductivity. Unfortunately, however, the upper critical field in $\text{FeSe}_{1-x}\text{Te}_x$ is very high: for $x=0.6$ it exceeds 45 T at low temperatures [K. Mukasa *et al.*, Phys. Rev. X **13**, 0011032 (2023)]. Thus, such measurements are not realistic in available conditions. We hope that the reviewer finds our revised manuscript suitable for a timely publication in Nat. Commun.

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Reply to Reviewer 3

The manuscript reports muon spin rotation measurements of the microscopic magnetic fields in a family of iron-chalcogenide superconductors (FeChSC), $\text{Fe}(1+y)\text{Se}(1-x)\text{Te}(x)$, $x = 0.35$,

0.6, 0.7, 0.8, and 0.9. In these materials, non-trivial topological electronic band structure results in a surface state with potential for topological surface superconductivity and Majorana states. The goal of the reported measurements is to identify a time-reversal symmetry breaking (TRSB) superconductivity in the bulk of FeChSC, which has significant implications for the Dirac surface state. In particular, TRSB could explain the gap opening below T_c observed in previous experiments. In general, identifying superconductivity with TRSB is an important and timely problem of broad interest. In the presence of topological band structure TRSB may lead to new states such as quantum anomalous Hall insulator or an axion insulator. Hence, the motivation for the reported research is strong and well-argued, justifying the submission to a high-impact journal such as Nature Communications.

We thank the reviewer for evaluating our paper and for the positive acknowledgments on the motivation of our work. To address the reviewer's comments, we have performed additional measurements on the chemical analysis of the samples. Below we reply to the comments in the same order as they appear in the report.

On the other hand, the experimental results presented in the manuscript in my opinion lack sufficient impact to justify publication in Nature Communications. In my opinion, many of the reported conclusions lack sufficient experimental substantiation, while some observations raise questions. One question of specific importance is that of chemical composition of the studied samples, which has not been carefully characterized. As previous studies have shown, both magnetic and superconducting properties of the $\text{Fe}_{1+y}\text{Se}_{1-x}\text{Te}_x$ family depend very sensitively on Fe off-stoichiometry, y . In the present work, however, authors simply rely on an assumption that Te-annealing allows to reduce y to zero. This is clearly insufficient: some sort of compositional characterization of the studied samples establishing their chemical composition is needed.

For example, authors do acknowledge that much of their observations in the $x=0.9$ sample where the magnetic volume fraction is estimated to be approximately 90% at low temperatures are similar to those reported in previous studies of $\text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x}$ with $\sim 3\%$ excess Fe, which exhibits incommensurate AFM order [44]. Magnetic susceptibility shown in the Supplementary also looks consistent with the incommensurate magnetism observed in Fe-rich samples. Is it possible that authors indeed observe incommensurate magnetism in their $x=0.9$ sample? Is it due to 10% Se doping or due to an un-characterized excess Fe? How would the latter impact the superconducting phase?

We agree with the reviewer that the compositional characterization is important in this system. We incorporated the reviewer's insightful suggestion and performed energy dispersive X-ray (EDX) spectroscopy in the samples used in this study. We measured 3 or 4 samples for each composition and 4 different spots for each sample. The averaged results with the standard deviations are shown in the revised Supplementary Information. We find no evident excess Fe in this analysis within the errors, confirming that our data are essentially free from the possible magnetism induced by excess Fe. Indeed, if we compare our results and the reported data in Ref. [44] using samples with ~3% excess Fe, the low-temperature magnetic volume fraction for $x=0.6$ is completely different: we see no magnetic signal whereas Ref. [44] reports more than 90% magnetic fraction. For the $x=0.9$ sample, the EDX analysis shows that the actual Te composition is closer to 0.94, where the magnetic transition temperature determined by the present μ SR and magnetization measurements is consistent with those reported for well-characterized annealed samples with excess Fe sufficiently removed in Refs. [31,32] and [Y. Sun *et al.*, Sci. Rep. **6**, 32290 (2016)]. In the revised manuscript, we have added the above discussion and modified x values based on the EDX analysis. We thank the reviewer again for the constructive comments, which helped to improve our paper.

More importantly, the central question of whether the microscopic magnetic fields which authors observe by μ SR belong to a uniform superconducting phase with TRSB or perhaps result from a phase segregation into a superconducting and magnetic phase, is not clearly answered by the authors' results.

Here, aside from interleaved magnetic phases induced by interstitial Fe ions, authors need to rule out the possibility that nano-scale magnetic inclusions of ferromagnetic or superparamagnetic Fe, or Fe_3Te_4 nano-particles are at the origin of the observed microscopic magnetic fields. Being embedded in a bulk superconducting phase, such magnetic nano-inclusions could induce screening currents and thus change magnetic field configurations in the sample at the superconducting transition. Convincingly ruling out such a possibility that observed behaviors result from chemically distinct magnetic phases or inclusions is needed in order for the manuscript to be reconsidered.

To address whether our results provide evidence for TRSB superconducting state, we focus on the results of $x=0.64$. Our scanning electron microscopy images do not find such inclusions (see new Fig. S1), which is consistent with the absence of magnetic signals in the high-field magnetization, low-field susceptibility and specific heat data shown in Fig. S2. Moreover, if magnetic inclusions should be present as the reviewer suggested, such inclusions act as magnetic impurities that lead to the Kondo effect, which is manifested as

the low-temperature upturn in the resistivity curve. However, we do not observe such a behavior in our $x=0.64$ samples. We note that in a high-field transport study using samples from the same source the normal-state resistivity with suppressed superconductivity remains metallic even below ~ 1 K [K. Mukasa *et al.*, Phys. Rev. X **13**, 011032 (2023)], supporting that no Kondo scattering is evident.

Finally, I have several technical remarks which I would like authors to address.

Page 4, first paragraph. "In this series, only the SC3 phase with the highest T_c maximum includes the topological region." - Perhaps, reference [35] would also be appropriate here.

We thank the reviewer for the suggestion. We have added the reference as suggested.

Page 8, second paragraph. " $B_{\text{int}} = 0.09$ (0.62) G for $x \sim 0.35$ and 0.76 (1.53) G for $x = 0.6$, respectively"- do numbers in parentheses denote uncertainties, and if so, the last number would be consistent with zero? If these are results of the different, NSR and SR measurements, which seem to be indicated by follow-up text, each should be given with its own corresponding error.

The difference between the two results is still large enough to question assertions based on comparing the internal fields from different measurements and therefore questions the relevance of entire discussion in this paragraph and elsewhere.

Similarly, "The present results that the magnitude of the TRSB field for $x \sim 0.35$ is almost identical to that of FeSe suggest that a similar TRSB superconducting state extends inside the nematic phase of $\text{FeSe}_{1-x}\text{Te}_x$." - how warranted is this statement given the large error bars?

The numbers in the parentheses are the results in the SR mode. We have modified the text to make it clearer. In the revised Fig. 4, we have added the error bars estimated from the lowest-temperature data in ZF- μ SR relaxation in Figs. 2e,f. For the SR mode, we have less data points due to limited machine time, which makes the error bigger especially for $x \sim 0.35$. We have also removed the sentence pointed out by the reviewer.

Supplementary: Based on magnetization measurements, authors note that the $x = 0.6$ sample has almost 100% superconducting volume fraction, while for $x \geq 0.7$ samples it is lower ($\sim 60\%$?). Are these values consistent with the μ SR data? Can superconducting volume

fractions be determined from the authors' uSR measurements?

In the TF- μ SR analysis we have shown in Fig. S4, we find two contributions, fast and slow relaxations for $x \geq 0.75$ samples, which is consistent with the non-zero contributions from the short-range AFM phase. More quantitative determination of the volume fraction is difficult from this analysis, but one rough estimate is to compare the weights of these two relaxations. From the fits we obtain the SC weights of about 43% for $x=0.75$ and 24 % for $x=0.83$, which are somewhat smaller than the SC volume fractions in the susceptibility and specific heat estimates. This discrepancy suggests that we cannot separate two contributions completely, which results in the ambiguity of the SC weight estimates. We emphasize, however, that for $x=0.64$ with 100% SC volume fraction we have single-component relaxation curves both in the TF- and ZF- μ SR measurements, which ensures that our data represents the SC signals.

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In summary, we believe that we have addressed the issues raised by the reviewers, who all acknowledged the importance of the present study in this intriguing system. With additional measurements, analyses, discussions provided in this revision, we trust that the Editor can find our revised manuscript suitable for a timely publication in *Nature Communications*.

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Reply to Reviewer 1

The authors respond to my comments in their response but not in the paper. In the context of the paper, the latter would be a bit of a challenge. As such I have no problem with the paper being published. It does bring new and interesting observations to the discussion.

We thank Reviewer #1 for recommendation for publication of our paper. We note that in the previous revision, we have added information that we are probing the bulk in Supplementary Information. In this revision we also added this in the main text. We also appreciate the reviewer for the positive comment that our paper brings new and interesting observations to the discussion.

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Reply to Reviewer 2

The revised manuscript has been improved, but I am not completely satisfied with the author's reply regarding LF. In particular, one can see in Fig.2f that in the normal state, the relaxation rate is indeed nearly completely suppressed by the application of 200G LF. In contrast, at the lowest measured temperature, the increase of the relaxation above the normal state value is nearly 50% of the ZF $\Delta\lambda = \lambda(T \rightarrow 0) - \lambda(T > T_c)$. This indicates that even 200G cannot decouple muon spins from fields associated with TRSB. This is a bit unexpected, given that the estimated internal field is $B_{int} = 0.62$ G only. So, to distinguish between the static or dynamic nature of the TRSB fields a more detailed study at different LF is required. These experiments would probably take a long time, given the muon facility schedule. Therefore, I think an alternative approach prior to publication would be to soften the paper's claims and discuss different possibilities on equal footing for the enhanced relaxation rate in the main text, including AFM fluctuations and the appearance of a weak FM on the surfaces and around defects. In particular, based on the detailed Kerr effect study, it was concluded that TRSB is related to weak surface FM state but not superconductivity [Phys. Rev. Lett. 130, 046702 (2023)]. The authors cite the paper but are absolutely silent about the contradictions in their interpretations.

We thank Reviewer #2 for the acknowledgment that our paper has been improved. The reviewer is correct in that the LF data for $x=0.64$ show a slight enhancement at low temperatures, which suggests the existence of other contribution than that from the static TRSB field in the ZF relaxation rate. However, the asymmetry spectra for $x>0.7$ (Figs. 2c,d)

showing almost no suppression under 200-G longitudinal fields are in clear contrast to those for $x=0.64$ (Fig. 2b), in which the LF relaxation is strongly suppressed compared with the ZF data. Namely, these results for $x=0.64$ are quite different from the results in the magnetic phase and thus imply that although there is a small amount of relaxation due to dynamic contributions, such as fluctuations, which are not suppressed by 200 G, they are not the dominant contribution. It is therefore natural to conclude that the observed increase in the ZF relaxation rate below T_c for $x=0.64$ ZF data contains a significant contribution from the TRSB field produced by the superconducting state. To incorporate the reviewer's suggestion, we reevaluate the error bar for the magnitude of the TRSB field estimated from the increase in the relaxation rate at ZF (the middle panel of Fig. 4) by considering the case that the contribution from the LF relaxation rate is subtracted.

As for the Kerr rotation study suggesting the surface effect, we have not discussed this simply because we do not obtain any information on the surface magnetism from our μ SR measurements. As we mentioned in Supplementary Information, the muon average stopping depth is $\sim 320 \mu\text{m}$, which is orders of magnitude longer than the coherence length ($< 100 \text{ nm}$), so the signal comes from the bulk, and we have essentially no information on the surface states in our measurements. The discussion of the surface magnetism in the paper the reviewer introduced (Ref. [36] in our manuscript) is based on some similarity with the van der Waals ferromagnetic material. However, we point out that the Kerr effect is a surface sensitive measurement and has little information on the bulk state. In other words, one cannot rule out the possibility that the bulk information is reflected on the surface, so it does not necessarily contradict our results.

We have added these discussions in the revised manuscript. We appreciate the reviewer's insightful comments which we find helpful to improve our manuscript.

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Reply to Reviewer 3

In the revised manuscript and in their responses, authors have addressed substantial fraction of concerns raised by the Reviewers. Commendably, they have characterized the composition of their samples, which clears large number of questions raised by the original manuscript and makes the presented work more accurate and comprehensive. I am, however, still hesitant to recommend the manuscript for publication because I am not convinced by the authors' response to one of my major concerns. Namely, the need to rule out a possibility

that nano-scale magnetic inclusions of ferromagnetic or super-paramagnetic Fe, or Fe₃Te₄ nano-particles are at the origin of the observed microscopic magnetic fields. I noted that convincingly ruling out such a possibility, that observed behaviors result from chemically distinct magnetic phases or inclusions, is needed in order for the manuscript to be reconsidered. Authors' argument about putative Kondo screening is not applicable to phase inclusions rather than atomic magnetic impurities. Additionally, ferro-/ferrimagnetic phase inclusions such as Fe₃O₄ or Fe₃Te₄ would not lead to any sort of paramagnetic behaviors, so the absence of such paramagnetic effects would not rule out these. If authors could supply convincing evidence that microscopic magnetic fields observed in their μ SR experiments do not arise from the combination of superconductivity and magnetic impurities, I would recommend the manuscript for publication in Nature Communications.

We thank Reviewer #3 for the positive statements that our revision clears large number of questions raised by the original manuscript and makes the presented work more accurate and comprehensive. To address the remaining issue raised by the reviewer, we point out that the effect of magnetic inclusions the reviewer worries can be ruled out from the comparisons between our results in well-characterized crystals and the previous μ SR results in polycrystals with impurity phases. For example, in undoped FeSe, the early-stage studies of zero-field (ZF) μ SR in powder samples of FeSe_{0.98} (with excess Fe) show no change in the relaxation above and below the superconducting transition temperature T_c [M. Bendele *et al.*, Phys. Rev. B **85**, 064517 (2012)]. However, more recent results on the very clean single crystals of FeSe grown by chemical vapor transport (which shows quantum oscillations with no sign of excess Fe from scanning tunnelling spectroscopy) clearly show the enhanced ZF- μ SR relaxation rate below T_c [K. Matsuura *et al.*, PNAS **120**, e2208276120 (2023)]. This difference has been attributed to differences in background contributions which may mask small signals of the TRSB field when the impurity phase exists. Importantly, this comparison demonstrates that the excess Fe impurity does not induce the apparent enhancement of relaxation rate below T_c . Similar situations can also be found in the optimal FeSe_{*x*}Te_{1-*x*}. In a μ SR study on polycrystalline samples of FeSe_{0.5}Te_{0.5}, which are reported to contain ~0.5-1.0% volume of the Fe₃O₄ impurity phase, again show no change of ZF relaxation rate above and below T_c [P. K. Biswas *et al.*, Phys. Rev. B **81**, 092510 (2010)]. This also supports that the inclusions of magnetic nano-particles the reviewer mentioned do not affect the μ SR relaxation at the superconducting transition. Thus, it is highly unlikely that the observed enhancement of ZF- μ SR relaxation rate in the superconducting state of our well-characterized crystals arises from the combination of superconductivity and magnetic impurities.

We have added the above discussion in the revised Supplementary Information (the last paragraph of section IV). We thank again Reviewer #3 for the in-depth review of our manuscript. We hope that the reviewer now finds our results merit timely publication in *Nature Communications*.

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Reply to Reviewer 2

The authors partially followed my suggestions. In particular, they clearly acknowledged the observation of a weak FM state in the polar Kerr experiments. Future studies need to investigate how this state is related to the TRSB state in the bulk. Given the novelty of the data, I recommend the manuscript for publication.

We thank Reviewer #2 for the recommendation for publication.

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Reply to Reviewer 3

In their response to my concern about possible role of nano- or micro-inclusions of magnetic phases such as Fe₃O₄ or Fe₃Te₄, authors argue as follows. "In a muSR study on polycrystalline samples of FeSe_{0.5}Te_{0.5}, which are reported to contain ~0.5-1.0% volume of the Fe₃O₄ impurity phase, again show no change of ZF relaxation rate above and below T_c [P. K. Biswas et al., Phys. Rev. B 81, 092510 (2010)]. This also supports that the inclusions of magnetic nano-particles the reviewer mentioned do not affect the muSR relaxation at the superconducting transition. Thus, it is highly unlikely that the observed enhancement of ZF-muSR relaxation rate in the superconducting state of our well-characterized crystals arises from the combination of superconductivity and magnetic impurities." While the cited reference to a previous study alleviating my concern is somewhat convincing, I think this issue needs to be clarified in the main text and not in the supplementary where authors have addressed it. This is because potential influence of magnetic inclusions can undermine the credibility of the reported results and therefore is central to establishing this credibility. As such, it needs to be addressed in the main manuscript. I also recommend that some passages are revised to tone down statements that are not supported by the presented experimental data. For example, "Our findings provide a fundamental understanding of the complex relationship between the TRSB superconductivity, nematicity, topology, and magnetism in FeSe-based superconductors." A very similar statement is repeated later, in the conclusion, "... our findings provide fundamental information for our understanding of the complex relationship between the superconductivity, nematicity, topology, and magnetism in the iron-chalcogenide superconductors". I think these are overstatements in the context of the results presented in the manuscript, and also redundant. I also noticed a couple of typos in the manuscript, so another careful read would be warranted. Other than that, I think that the manuscript passes the bar and can be accepted for publication.

We thank Reviewer #3 for the comments and recommendation for the acceptance. We have followed the reviewer's suggestions, and we move our discussion on the possible effect of magnetic impurities from Supplementary Information to the main text. We have also changed the wordings for the sentences pointed out by the reviewer and fixed typos. We believe that our manuscript is now ready for publication.