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Peer Review File

Electric field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling

Corresponding Author: Professor Tomasz Dietl

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Professor Dietl,

Thank you for submitting your manuscript, "Electric field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of interest, some important points are raised, particularly by Reviewer #3. We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

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-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type ‘Revised Manuscript - Marked Up’ when uploading the manuscript file to our online system.

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We hope to receive your revised paper within three months; please let us know if you aren’t able to submit it within this time so that we can discuss how best to proceed. If we don’t hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Oleksandr Pylypovskyi
Editorial Board Member
Communications Materials
orcid.org/0000-0002-5947-9760

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript titled by “Electric-field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling”, reported a comprehensive study of magnetization changes in ferromagnetic GaMnN grown by molecular beam epitaxy when exposed to an electric field. Two phenomena were identified in this work: (1) The magnetoelectric signal is odd and reversible over a wide magnetic field range, attributed to the inverse piezoelectric effect altering trigonal single-ion anisotropy. (2) In the hysteresis regime, the effect becomes non-linear and irreversible with a time-dependent electric field, which can reorient the magnetization. The authors state that the experimental results support theories of time-dependent perturbations in non-equilibrium glasses with an enhanced effective temperature. In addition, the conclusion of this work is supported by two complementary theoretical approaches: (i) a macrospin model, which describes changes in magnetic anisotropy through magnetizing or demagnetizing anisotropy fields, depending on the orientation of the external magnetic field relative to the easy axis; and (ii) an atomistic Landau-Lifshitz-Gilbert model for interacting classical spins, with parameters fitted to our M(H) data.
The experimental results and theoretical models presented are highly impressive. The investigation of electric-field control over magnetic anisotropy is a timely and significant topic in spintronics. The paper is well-written, with clear and instructive descriptions of the results. I strongly recommend its publication in a high-impact journal in its current form.
The authors may consider the following minor technical issues:
1. The sign “÷” appears couple times in the paper on my computer screen. I think it is “-”, it shows wrongly due to font problem.
2. Regarding the non-linear magnetoelectric effect, since it occurs in the hysteresis regime, it should be naturally related to

the domain texture of the GaMnN system. Could the authors comments on this? Is it possible to simulate this non-linear effect via LLG-based micromagnetic simulations?

Reviewer #2 (Remarks to the Author):

The manuscript presents several intriguing aspects of (Ga,Mn)N, with the potential for significant impact on the evolving field of synaptic materials. I would recommend its publication, provided the authors satisfactorily address the following points:

1. The authors assert that the magnetoelectric responses of the reported (Ga,Mn)N samples are well-supported by both the macrospin model and the atomistic LLG theory. However, Figure 3 does not clearly demonstrate how accurately these theories align with the experimental data. The authors should clarify the discrepancies between the theoretical predictions and the experimental findings and provide plausible explanations for these differences.

2. The discovery that (Ga,Mn)N is a piezoelectric ferromagnetic homogeneous compound is particularly noteworthy. While the magnetic measurements are thorough and convincing, the manuscript would greatly benefit from a discussion on potential electric probes for practical applications. Suggesting plausible methods for electric probing would significantly enhance the impact of these findings.

3. The term "glass" can be somewhat ambiguous in the context of materials science. Although the hysteresis responses to time-dependent stimuli are well-demonstrated, the manuscript would benefit from a more precise definition and explanation of the glass phase as it pertains to this study.

Reviewer #3 (Remarks to the Author):

In the manuscript titled “Electric-field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling,” the authors present their experimental findings and model interpretation of the magnetoelectric effect in Mn-doped GaN. Using a piezoelectric-driven strain, they demonstrate the alteration of magnetocrystalline anisotropy and its impact on the (ferro)magnetic properties of epitaxially grown GaMnN thin films. This work can be viewed as a natural continuation of the authors' previous studies on the same system (Ref 8), where they also used the piezoelectric effect to alter magnetocrystalline anisotropy, but that time in the paramagnetic state, above the Curie transition temperature (Tc). In essence, the primary mechanism underlying the observed magnetoelectric effect was elucidated in Reference 8, and the present work extends this understanding to the ferromagnetic state of GaMnN.

Overall, the experimental work and theoretical analysis are well structured and clearly presented in the manuscript. The experimental methods are appropriate and carefully planned. In particular, the measurements of magnetism in response to the electrical stimuli are carried out thoroughly, using the specialized setup and procedure (as presented in Reference 8), which contributes significantly to the quality of the results. The results presented in the manuscript would significantly contribute to the field of magnetoelectrics, offering the scientific community valuable insights into the phenomena of magnetoelectric coupling in diluted magnets.

To summarize the main phenomenon discussed in this work, the magnetoelectric effect results from piezoelectric-driven strain applied to the tetrahedral coordination of anions surrounding Mn, which alters the single-ion uniaxial magnetic anisotropy along the c-axis. Based on this mechanism, the general characteristics of the magneto-electric measurements can be interpreted in a fairly straightforward way. At this point, however, it would be beneficial for the reader to have more information about the assumptions the authors made in their modeling and data analysis.

A) Judging from the shape of the magnetization loops (Fig.1), there seems to be little difference between the parallel and perpendicular directions. Does this mean that the magnetocrystalline anisotropy (with the easy axis along the c-axis) is relatively weak here, or perhaps not measurable with standard SQUID measurements?

B) Fig. 1b shows the simulated curves for two models. It seems that both models assume a single magnetic domain scenario and do not consider a multidomain case. When compared to the actual data, the models do not seem to adequately describe the measured results. The measured curves seem to suggest a shape indicative of a multidomain case, or at least a distribution of coercive fields within the sample. Which scenario applies here?

C) Regarding point B, did you try to measure a virgin M(H) curve?

D) Also regarding point B, why is a multidomain case completely excluded from your consideration? Please discuss this briefly in the manuscript.

E) If a multidomain state cannot be definitively excluded, then the discussion of the switching mechanism, especially in the hysteretic regime, should be approached with caution. In the presence of domain walls, one can easily imagine a magnetoelectric effect based on a single-ion anisotropy, domain nucleation, pinning and switching.

F) Considering hypothetical magnetic domain switching and dynamics, the effects observed in Figures 3 and 6 could also be convincingly explained. For example, in the multidomain picture, the observed irrevesibilities could simply be related to domain nucleation or (de)pinning mechanisms.

For example, in Figure 3, the modeled curves are for the single-domain case. Perhaps the real system is multi-domain, and that's why there is some discrepancy in the magnitude and shape of the modeled and simulated curves (at least a partial explanation)?

In my opinion, it would be advisable to critically examine the possibility of the multi-domain versus single-domain scenario to improve the overall completeness of this work. Of course, in the multi-domain state, the physical picture becomes much more complicated (probably too complicated for quantitative analysis).

In summary, I recommend the manuscript for publication after addressing the above comments and providing the necessary clarifications.

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Version 1:

Decision Letter:

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Dear Professor Dietl,

Your manuscript titled "Electric field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials.

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

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We hope to hear from you within two weeks; please let us know if the process may take longer.

Best regards,

Oleksandr Pylypovskyi
Editorial Board Member
Communications Materials
orcid.org/0000-0002-5947-9760

REVIEWERS' COMMENTS:

Reviewer #3 (Remarks to the Author):

In my opinion, the authors have adequately addressed the reviewers' concerns in the revised manuscript.
Therefore, I recommend the manuscript for publication in its current form.

Ref.: manuscript COMMSMAT-24-0533-T
D. Sztenkiel et al. "Electric-field manipulation …"

We would like to thank the Reviewers for their time and expertise devoted to the evaluation of our manuscripts and their kind comments: Reviewer #1 stated: "The experimental results and theoretical models presented are highly impressive … I strongly recommend its publication in a high-impact journal in its current form". Similarly, Reviewer #2 specified: "I would recommend its publication, provided the authors satisfactorily address the following points". Finally, according to Reviewer #3: "Overall, the experimental work and theoretical analysis are well structured and clearly presented in the manuscript. The experimental methods are appropriate and carefully planned." And "In summary, I recommend the manuscript for publication after addressing the above comments and providing the necessary clarifications."

Below, we provide a point-by-point reply to the Reviewers' comments and the list of corresponding changes introduced to the manuscript.

Reviewer #1

The manuscript titled by "Electric-field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling" reported a comprehensive study of magnetization changes in ferromagnetic GaMnN grown by molecular beam epitaxy when exposed to an electric field. Two phenomena were identified in this work: (1) The magnetoelectric signal is odd and reversible over a wide magnetic field range, attributed to the inverse piezoelectric effect altering trigonal single-ion anisotropy. (2) In the hysteresis regime, the effect becomes non-linear and irreversible with a time-dependent electric field, which can reorient the magnetization. The authors state that the experimental results support theories of time-dependent perturbations in non-equilibrium glasses with an enhanced effective temperature. In addition, the conclusion of this work is supported by two complementary theoretical approaches: (i) a macrospin model, which describes changes in magnetic anisotropy through magnetizing or demagnetizing anisotropy fields, depending on the orientation of the external magnetic field relative to the easy axis; and (ii) an atomistic Landau-Lifshitz-Gilbert model for interacting classical spins, with parameters fitted to our M(H) data.

The experimental results and theoretical models presented are highly impressive. The investigation of electric-field control over magnetic anisotropy is a timely and significant topic in spintronics. The paper is well-written, with clear and instructive descriptions of the results. I strongly recommend its publication in a high-impact journal in its current form.

The authors may consider the following minor technical issues:

1. The sign "÷" appears couple times in the paper on my computer screen. I think it is "-" it shows wrongly due to font problem.

Our reply:

The symbol "÷" has been removed.

2a. Regarding the non-linear magnetoelectric effect, since it occurs in the hysteresis regime, it should be naturally related to the domain texture of the GaMnN system. Could the authors comments on this?

Our reply:

The question of the domain structure was also raised by Reviewer # 3 and also, to some extent, by Reviewer #2. In order to clarify this crucial issue we have supplemented the manuscript by a new paragraph in the second column of p. 8 and by the virgin curve added to magnetic hystereses presented in Fig. 4a. The new paragraph reads:

"To explain the origin of that universal behaviour, we first recall that despite magnetic dilution, a well-defined magnetic domain structure was found in (Ga,Mn)As, the fact explained quantitatively in terms of a long-range character of spin-spin interactions in that type of ferromagnets[49]. In order to address this question in (Ga,Mn)N, we show in Fig. 4a experimental data on virgin magnetization obtained after cooling the sample down to 2 K under near-zero-field conditions ($H \approx 0.15$ Oe). The measured form of $m(H)$ corresponds neither to the domain-wall motion nor to the domain-wall pinning in a typical multidomain case for a ferromagnet. In the former case one would expect a rapid increase of m, nearly along the y-axis, in the latter the initial $m(H)$ would be horizontal along the x-axis. Actually, the observed behaviour is consistent with or even supports the view that in the case of short-range interactions, driven here by superexchange, the virgin magnetic properties are dominated more by superparamagnetic regions present in the percolation picture than by a regular domain structure which is formed by dipole interactions in dilute ferromagnets with long-rage exchange couplings[49]."

The above new paragraph is followed by slightly modified previous statements:

"More specifically, we expect in both $(Bi,Sb,Cr)_2Te_3$ and (Ga,Mn)N, a co-existence, at $T_C > T > 0$, of an infinite ferromagnetic percolating clusters with superparamagnetic regions, characterized by a broad spectrum of blocking temperatures and relaxation times. With no doubts, the presence of long-range dipole interactions, responsible for the domain formation in materials like (Ga,Mn)As, contributes to the complexity of spin dynamics."

We also highlighted this point in the following parts of the manuscript:

(p. 1, right column): "In those semi-insulating layers, Mn^{3+} ions are coupled by a short-range ferromagnetic (FM) superexchange[16,19–21], and FM ordering is realized in a percolation-like fashion[22–24]." and
(p. 2, left column): "Second, the percolating ferromagnetism means the presence of an infinite FM cluster at $T \leq T_C$. This cluster encompasses all spins at $T = 0$ but only about 20% of spins at T approaching T_C (ref. 22). Thus, at any non-zero temperature, a large portion of the Mn ions resides in finite clusters of different sizes. Such a magnetic system is expected to exhibit a broad spectrum of relaxation times and, in the hysteretic regime, constitutes a glass far from thermal equilibrium." and
(p. 3, left column): "We see that at that temperature $T \approx T_C/4$ and the time scale of the experiment, magnetic hysteresis involves only a part of spins, substantiating expectations of the percolation model[22–24]"

2b. Is it possible to simulate this non-linear effect via LLG-based micromagnetic simulations?

Our reply:

In order to address this question we have modified a statement in p. 8 (right column, bottom) to the form:

"It is worth noting that the standard Langevin dynamics does not capture the effects of time-dependent perturbations, so that the nonlinear and irreversible effects do not show up in our LLG simulations."

Reviewer #2 (Remarks to the Author):

The manuscript presents several intriguing aspects of (Ga,Mn)N, with the potential for significant impact on the evolving field of synaptic materials. I would recommend its publication, provided the authors satisfactorily address the following points:

1. The authors assert that the magnetoelectric responses of the reported (Ga,Mn)N samples are well-supported by both the macrospin model and the atomistic LLG theory. However, Figure 3 does not clearly demonstrate how accurately these theories align with the experimental data. The authors should clarify the discrepancies between the theoretical predictions and the experimental findings and provide plausible explanations for these differences.

Our reply:

We now tackle with this challenge in modified statements in p. 7 (right column), in which we directly refer to Fig. 3:

"As seen in Fig. 3, the LLG theory is successful only qualitatively, i.e., predicts properly the sign and shape of $M_E(H_j)$. The obtained results bring us to the conclusion that the LLG approach, when applied to dilute ferromagnets, predicts too large magnitudes of remanence in the case of both $M(H_j)$ and $M_E(H_j)$. We assign this outcome to the time scale of our experiment, which is sufficiently long to allow demagnetization of most of the Mn spins outside the percolation cluster. Such a demagnetization process does not occur in the necessary much shorter time frame of the LLG simulations, so that most of the Mn spins contribute to the theoretical remanence, as shown by the solid lines in Fig. 1."

2. The discovery that (Ga,Mn)N is a piezoelectric ferromagnetic homogeneous compound is particularly noteworthy. While the magnetic measurements are thorough and convincing, the manuscript would greatly benefit from a discussion on potential electric probes for practical applications. Suggesting plausible methods for electric probing would significantly enhance the impact of these findings.

Our reply:

We agree with the Reviewer that we should add such a discussion to the main text. Therefore we changed the following sentences of the main text (p. 9, left column):

“The spectrum of (Ga,Mn)N functionalities has recently been enriched by the demonstration of efficient generation and detection of spin currents in Pt/(Ga,Mn)N structures up to 50 K (ref. 51).”

to

“The spectrum of (Ga,Mn)N functionalities has recently been enriched by the demonstration of efficient generation and detection of spin currents in Pt/(Ga,Mn)N structures up to 50 K (ref. 51) using spin Hall magnetoresistance (SMR) effect[52]. The same SMR could be then employed to electrically probe the magnetic state of the insulating (Ga,Mn)N, paving the way towards more practical applications. “

3. The term "glass" can be somewhat ambiguous in the context of materials science. Although the hysteresis responses to time-dependent stimuli are well-demonstrated, the manuscript would benefit from a more precise definition and explanation of the glass phase as it pertains to this study.

Our reply:

Yes, despite decades of studies on spin glasses in dilute systems with random or antiferromagnetic interactions, still controversies exist on the nature of spin arrangement and their description, in particular, on whether the droplet model or mean-field model with long-range interactions is more appropriate. In the response to Reviewer #1’s 2a comment we discuss this issue in some detail for the case of dilute ferromagnets. However, we should admit that microscopic information is still lacking. In view of the above we kindly ask the Reviewer to relax this requirement.

Reviewer #3 (Remarks to the Author):

In the manuscript titled “Electric-field manipulation of magnetization in an insulating dilute ferromagnet through piezoelectromagnetic coupling” the authors present their experimental findings and model interpretation of the magnetoelectric effect in Mn-doped GaN. Using a piezoelectric-driven strain, they demonstrate the alteration of magnetocrystalline anisotropy and its impact on the (ferro)magnetic properties of epitaxially grown GaMnN thin films. This work can be viewed as a natural continuation of the authors' previous studies on the same system (Ref 8), where they also used the piezoelectric effect to alter magnetocrystalline anisotropy, but that time in the paramagnetic state, above the Curie transition temperature (Tc). In essence, the primary mechanism underlying the observed magnetoelectric effect was elucidated in Reference 8, and the present work extends this understanding to the ferromagnetic state of GaMnN.

Overall, the experimental work and theoretical analysis are well structured and clearly presented in the manuscript. The experimental methods are appropriate and carefully planned. In particular, the measurements of magnetism in response to the electrical stimuli are carried out thoroughly, using the specialized setup and procedure (as presented in Reference 8), which contributes significantly to the quality of the results. The results presented in the manuscript would significantly contribute to the field of magnetoelectrics, offering the scientific community valuable insights into the phenomena of magnetoelectric coupling in diluted magnets.

To summarize the main phenomenon discussed in this work, the magnetoelectric effect results from piezoelectric-driven strain applied to the tetrahedral coordination of anions surrounding Mn, which alters the single-ion uniaxial magnetic anisotropy along the c-axis. Based on this mechanism, the general characteristics of the magneto-electric measurements can be interpreted in a fairly straightforward way. At this point, however, it would be beneficial for the reader to have more information about the assumptions the authors made in their modeling and data analysis.

A) Judging from the shape of the magnetization loops (Fig.1), there seems to be little difference between the parallel and perpendicular directions. Does this mean that the magnetocrystalline anisotropy (with the easy axis along the c-axis) is relatively weak here, or perhaps not measurable with standard SQUID measurements?

Our reply:

As detailed in ref. 8 and also in a new ref. 67, we have elaborated a method to quantify magnetocrystalline anisotropy of (Ga,Mn)N thin films by SQUID magnetometry. In the present study the samples are chosen to exhibit a very weak total magnetic anisotropy. The following sentence states this in the manuscript (p. 3, left column):

"The data indicate also that magnetocrystalline anisotropy is close to zero for the Mn content and strain in question."

In following, we modified a part of the Methods to read (p. 13, left column):

"All the magnetic measurements are performed using a Quantum Design MPMS XL superconducting quantum interference device (SQUID) magnetometer following a procedure elaborated earlier[8,64,65] and adjusted for measurements of ultra-thin (Ga,Mn)N films[66] deposited on sapphire substrates[67]. As shown previously for MBE-grown epilaers[17,18,20], and checked for the present samples, our high-resolution magnetic data do not reveal the presence of any high-T_C ferromagnetic precipitatates."

B) Fig. 1b shows the simulated curves for two models. It seems that both models assume a single magnetic domain scenario and do not consider a multidomain case. When compared to the actual data, the models do not seem to adequately describe the measured results. The measured curves seem to suggest a shape indicative of a multidomain case, or at least a distribution of coercive fields within the sample. Which scenario applies here?

C) Regarding point B, did you try to measure a virgin M(H) curve?

D) Also regarding point B, why is a multidomain case completely excluded from your consideration? Please discuss this briefly in the manuscript.

F) Considering hypothetical magnetic domain switching and dynamics, the effects observed in Figures 3 and 6 could also be convincingly explained. For example, in the multidomain picture, the observed irrevesibilities could simply be related to domain nucleation or (de)pinning mechanisms.

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For example, in Figure 3, the modeled curves are for the single-domain case. Perhaps the real system is multi-domain, and that's why there discrepancy in the magnitude and shape of the modeled and simulated curves (at least a partial explanation)?
In my opinion, it would be advisable to critically examine the possibility of the multi-domain versus single-domain scenario to improve the overall completeness of this work. Of course, in the multi-domain state, the physical picture becomes much more complicated (probably too complicated for quantitative analysis).
```

Our reply:

We are indebted to the Reviewer for bringing the question of domains up which indeed was missing in the original manuscript, as also noted by other Reviewers. Prompted by the above suggestions, we have supplemented the manuscript by the results of the virgin magnetization measurements that are presented in Fig. 4a of the revised manuscript. We have also added a new paragraph addressing the question of domains, as already noted in our reply to concerns of Reviewer #1. This new paragraph reads:

"To explain the origin of that universal behaviour, we first recall that despite magnetic dilution, a well-defined magnetic domain structure was found in (Ga,Mn)As, the fact explained quantitatively in terms of a long-range character of spin-spin interactions in that type of ferromagnets[49]. In order to address this question in (Ga,Mn)N, we show in Fig. 4a experimental data on virgin magnetization obtained after cooling the sample down to 2 K under near-zero-field conditions ($H \approx 0.15$ Oe). The measured form of $m(H)$ corresponds neither to the domain-wall motion nor to the domain-wall pinning in a typical multidomain case for a ferromagnet. In the former case would expect a rapid increase of m, nearly along the Y-axis, in the latter the initial $m(H)$ would be horizontal along the X-axis. Actually, the observed behaviour is consistent with or even supports the view that in the case of short-range interactions, driven here by superexchange, the virgin magnetic properties are dominated more by superparamagnetic regions present in the percolation picture than by a regular domain structure which is formed by dipole interactions in dilute ferromagnets with long-rage exchange couplings[49]."

The above new paragraph is followed by slightly modified previous statements:

"More specifically, we expect in both $(Bi,Sb,Cr)_2Te_3$ and (Ga,Mn)N, a co-existence, at $T_C > T > 0$, of an infinite ferromagnetic percolating clusters with superparamagnetic regions, characterized by a broad spectrum of blocking temperatures and relaxation times. Therefore, these systems in the metastable regime of the hysteresis can be regarded as a glass that is far from thermal equilibrium. With no doubts the presence of long-range dipole interactions, responsible for the domain formation in materials like (Ga,Mn)As, contributes to the complexity of glass dynamics."

We also highlighted this point in the following parts of the manuscript:

(p. 1, right column): "In those semi-insulating layers, Mn^{3+} ions are coupled by a short-range ferromagnetic (FM) superexchange[16,19–21], and FM ordering is realized in a percolation-like fashion[22–24]." and

(p. 2, left column): "Second, the percolating ferromagnetism means the presence of an infinite FM cluster at $T \leq T_C$. This cluster encompasses all spins at $T = 0$ but only about 20% of spins at T approaching T_C (ref. 22). Thus, at any non-zero temperature, a large portion of the Mn ions resides in finite clusters of different sizes. Such a magnetic system is expected to exhibit a broad spectrum of relaxation times and, in the hysteretic regime, constitutes a glass far from thermal equilibrium." and
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In summary, I recommend the manuscript for publication after addressing the
above comments and providing the necessary clarifications.
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We would like to thank all Reviewers for carefully examining our manuscript and putting forward constructive suggestions how to improve it. We hope that the revised manuscript fulfills the quality criteria expected for papers in *Communications Materials*.

Sincerely yours,

for authors:
Dariusz Sztenkiel
Maciej Sawicki
Tomasz Dietl