nature portfolio

Peer Review File

Signal amplification in a solid-state sensor via asymmetric many-body echo

Corresponding Author: Professor Mikhail Lukin

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

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)
The pursuit of improved metrology methods is a central theme in modern physics, as precision measurements underpin fundamental science and technological advancements. Quantum-enhanced sensing, leveraging many-body correlations and entanglement, has been a key focus in AMO systems, where techniques such as spin squeezing have enabled measurement sensitivities beyond classical limits. However, translating these advances to solid-state platforms presents unique challenges, including stronger decoherence, complex disorder, and technical noise. Solid-state quantum sensors, particularly those based on defect centers in diamond, offer practical advantages over AMO systems due to their robustness, scalability, and operation under ambient conditions. Methods that enhance metrological performance in such systems, such as the signal amplification approach explored in this study, are essential for extending quantum-enhanced sensing to real-world applications. This is particularly relevant for high-resolution magnetic imaging, materials characterization, and biological sensing, where solid-state platforms can provide nanoscale spatial resolution and direct integration with technological devices. The development of new protocols that exploit many-body dynamics for metrological gain in these settings represents an important step toward practical quantum technologies.

This work presents a systematic study of how many-body interactions in a disordered ensemble of nitrogen-vacancy (NV) centers in diamond can be harnessed for quantum-enhanced sensing. A key achievement is the engineering of one- and two-axis twisting (TAT) dynamics, which are known to generate metrologically useful quantum correlations. In naturally occurring dipolar spin ensembles, such interactions are difficult to exploit due to their anisotropic nature and the presence of disorder, which tends to average out their beneficial effects. The authors overcome these challenges through a combination of dynamical control of the quantization axis and Floquet engineering, allowing them to induce uniform-sign twisting interactions in a two-dimensional NV ensemble at room temperature. These engineered interactions provide a practical route toward entanglement-enhanced sensing in solid-state platforms. Building on this capability, the authors investigate how these engineered interactions can be used to amplify weak sensing signals. While traditional quantum metrology approaches focus on reducing measurement noise (e.g., via spin squeezing), this work takes a different route by amplifying the measured signal itself. Specifically, the authors employ an echo-based scheme, where TAT interactions are applied both before and after the sensing step. This approach enhances the distinguishability of small phase shifts encoded in the spin ensemble, making the sensing signal more robust to readout noise. A particularly striking result of the study is the discovery that maximal signal amplification occurs when the time-reversed TAT evolution after signal imprint is twice as long as the forward evolution. This asymmetric echo sequence departs from conventional symmetric echoes, where the forward and backward evolution times are equal. The authors attribute this unexpected enhancement to a time-reversal mirror symmetry in the system's dynamics, which ensures that the chosen asymmetry leads to optimal amplification. This insight not only improves the performance of signal amplification in NV-based sensors but may also be relevant for other quantum metrology schemes relying on many-body interactions. Overall, the paper introduces a novel approach to engineering and exploiting many-body interactions in a solid-state quantum sensor, demonstrating experimentally that these interactions can be tuned and optimized for practical metrology applications. The results contribute to the broader effort of making quantum-enhanced sensing more accessible in realistic, room-temperature environments, bridging the gap between idealized AMO-based quantum sensors and robust solid-state implementations.

The manuscript is well-organized and presents its results in a clear and convincing manner. The logical structure guides the reader through the motivation, methodology, experimental results, and theoretical interpretation in a coherent and accessible way. The arguments are well-supported by both experimental data and theoretical insights, making the claims compelling. The figures are well-designed and effectively illustrate key concepts, experimental setups, and results, providing intuitive visual support for the discussion. The data presentation is thorough, with careful analysis that strengthens the credibility of the findings. Overall, the clarity of exposition and the strong connection between theory and experiment contribute to a high-quality presentation of the work.

The results presented in this work are highly relevant to the field of quantum sensing and many-body quantum metrology. The successful engineering of one- and two-axis twisting interactions in a disordered solid-state spin ensemble represents a significant advance in harnessing many-body effects for practical sensing applications. Moreover, the discovery that an asymmetric echo sequence—rather than the conventional symmetric approach—yields maximal signal amplification is conceptually unexpected and provides new insight into the role of time-reversal symmetry in many-body dynamics. This novel mechanism is not only of interest to specialists in NV-based sensing but also has broader implications for quantum metrology and quantum control. Given its relevance to both fundamental quantum physics and practical applications, the study is well-suited for publication in a high-impact journal such as Nature. While the manuscript is already strong, we encourage the authors to consider the following question and list of comments:

1. A central aspect of the manuscript is the demonstration of signal amplification, with the susceptibility derived from Kubo's formula serving as the primary figure of merit. This contrasts with other studies on echo sequences, such as Refs. [12] and [37], which focus on the net metrological gain in terms of the signal-to-noise ratio (SNR). Given this difference in perspective, an important question arises: What can be said about the noise properties in the proposed sequence? Specifically, is it to be expected that the amplification observed in the susceptibility directly translates into an equivalent enhancement in the SNR? Furthermore, what is the dominant noise source in the experimental measurements on the NV ensemble, and would this noise be affected by the application of TAT dynamics? Addressing these points would help clarify the practical implications of the proposed amplification scheme for real-world quantum sensing applications.

Additionally, we suggest the authors address the following minor points:
2. The nomenclature in Eq. (1) differs from that used in the underlying calculations in the Supplementary Information. Please unify the notation for consistency.
3. Is it possible to provide an analytical connection between the coefficients of the dipolar TAT Hamiltonian in Eq. (3) and the OAT Hamiltonian in Eq. (1), which was used to engineer the TAT Hamiltonian? A clearer analytical link would improve the theoretical understanding of the transition between these interaction regimes.
4. Please clarify what is meant by the term "semi-classical flow" (line 138). A brief explanation would make the description more accessible.
5. The formulation in lines 157–158, "… a sensing rotation … is sandwiched between a pair of time-reversed TAT Hamiltonians," may be misleading. Please consider rewording to make it explicit that the second TAT Hamiltonian is time-reversed compared to the first one.
6. In line 219, it is stated that "quantum projection noise would become relevant in such cases." Please elaborate on why quantum projection noise is not currently relevant and under what conditions it would start to play a role.

Referee #2

(Remarks to the Author)
This referee co-reviewed the paper with one of the other reviewers.

Referee #3

(Remarks to the Author)
The manuscript by Gao et al. reports experimental demonstration of quantum sensing enhanced by quantum correlations in the ensemble of coupled NV centers. Interactions between NV centers have previously been considered as a source of decoherence in diamond magnetometers. The manuscript demonstrates that coherent control methods allow to increase the performance of NV based sensors using coupling between NV spins. Although the signal amplification is relatively low (less than 10 per cent), the manuscript shows a very significant result in the field of quantum sensing and metrology. Remarkable, careful engineering of a coherent control protocol allows amplification in positionally disordered NV centers coupled by long range dipolar interactions.
The manuscript is the first demonstration of the use of quantum correlations for enhanced sensing. I recommend it for publication after addressing the following points
Authors use nearly two-dimensional layers of NV centers. I recommend including SIMS data in the additional material section.
The concentration of substitutional nitrogen (P1 centers) is not given. Although EPR measurements are not possible for such a thin layer, they can be detected by double electron resonance. I expect that these electron spins could influence the dynamics of the NV centres.
I also recommend to include details on the determination of NV centers concentration in the doped layer.
The authors discuss the effects of laser power drift and signal normalization. Is there also an effect of the charge instability of the NV centres observed for the sample, and if so, how does this affect the measurement?

Referee #4

(Remarks to the Author)
The paper by Gao and coworkers discusses whether the spin-spin interactions in a solid-state spin ensemble can be exploited for improved sensing performance, compared to the case of an equivalent non-interacting spin ensemble. The authors demonstrate that this is indeed possible, but highly challenging because of the anisotropy of the dipolar spin-spin couplings and the random locations of the spin defects. The authors address these challenges by confining the spins to a two-dimensional layer and aligning them with the out-of-plane direction, reducing dipolar anisotropy, and by refocusing the time evolution to produce an echo signal, mitigating the large spread in couplings that comes with the random spin locations. Both ingredients are key for achieving a net signal improvement. They report a signal improvement by 6.7%.

In my view, the significance of the reported work lies in demonstrating a net improvement in sensing performance, even if the improvement is marginal in a practical sense. As far as I know this is the first demonstration of signal enhancement in a solid-state spin ensemble. Since solid-state spin systems have many practical applications in metrology (see Refs. 1-7), this is a significant result. Once this has been said, I have two major reservations with regard to publication in Nature. First, I have to admit that the paper is very challenging to read and understand, especially the discussion of the time-reversed echo. While this is advanced physics, I find the presentation unnecessarily complicated for the general reader. In its present form, the paper would be better suited for a specialist physics paper. The second reservation I have is regarding the impact of the signal amplification beyond the basic demonstration. Even in a most optimistic scenario, the amplification will only be a small factor. In practical uses of solid-state quantum sensors, many other parameters influence sensitivity (material quality, environment in which sensor is operated, optimization of optical readout, etc.), and those usually impact sensitivity much more than a subtle 6.7% improvement. Therefore, while the authors should be congratulated on achieving a positive gain, I question the optimistic outlook on practical advances.

Technically, the experimental body of work and the analysis look sound, although I cannot judge this in depth. The approach of confining the spins to a thin two-dimensional layer combined with toggling spins alignment between 111 and 100 is neat and innovative. The demonstration of Fig. 2b (main plot vs. inset) is convincing in confirming the twisting dynamics.

Further comments:

1) Could the authors briefly motivate the choice of bias field? Why 890 Gauss instead of another value?

2) On line 172, the amplification by 6.7% is claimed from Fig. 4c. How are these 6.7% obtained? Does this correspond to the relative increase (of the blue curve?) at t+ + t- >0 compared to the red trace at t=0? Or does this correspond to the peak value of the green curve in Fig. 4d?

3) Although the time-reversed echo is explained at length, I still do not fully capture the mechanism. Is the t- = 2t+ asymmetry a general feature of any type of qubit pair interaction, or does it only apply to spin-spin interactions that are dipolar in character (where the coupling constant varies between -J and +2J depending on the angle between the spins' axes and the

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)
The revised manuscript provides an improved presentation which, in our opinion, addresses all points raised in our report and, we believe, also those raised by the other referees. Major changes in the revision include a more intuitive, pictorial explanation—supported by an additional figure (Fig. 4e)—of the new echo protocol for signal amplification based on time-reversed mirror symmetry, as well as an expanded discussion of the broader perspectives of the current work in the manuscript's outlook. Both changes strengthen the presentation and enhance its accessibility to non-expert readers. As expressed in our initial report, we believe the authors present a strong, surprising, and important result that demonstrates a novel concept in quantum metrology within solid-state systems and warrants publication in Nature. We recommend acceptance of the revised manuscript.

We provide the following remarks for the authors' consideration regarding the new or revised sections of the manuscript:

• Lines 204–206: In the sentence "Specifically, the dynamics ...", it remains unclear what dynamics are depicted in Fig. 4(e). Adding "the dynamics under the TAT Hamiltonian" and rephrasing the sentence—for example, starting with "As illustrated in Fig. 4(e)..."—could improve clarity for the reader.
• Lines 204–206: Later in the sentence, the authors write "… under the reflection about the YZ plane (R)." From Fig. 4(e), it is not immediately clear to the reader that the reflection plane is the YZ plane rather than the XZ plane. Adding axis labels to the figure could help clarify this point.
• Check if articles are missing in line 219 ("by the TAT Hamiltonian") and line 222 ("by the refocusing").
• Line 226: In the sentence "This conclusion is further supported by …" it is claimed that a local minimum is obtained for a modified (XYZ instead of TAT) Hamiltonian. From Extended Data Figure 6 it is not immediately clear why this results in a minimum.
• Lines 242–245: The sentence "While positional disorder …" is rather technical. It may help to rephrase in order to make it

more accessible.
• Line 249: “explore” is used twice in the same sentence.
• In Eq. (6) of the Supplementary, the “dynamical susceptibility” χ is defined without an absolute value, while an absolute value is used in the main text (line 213). Moreover, different terms are used in the main text (“amplification amplitude” in line 212) and in the Supplementary (“dynamical susceptibility” in line 555).
• Line 569, typo: “on” should be “in”.
• In line 3 of the proof of Eq. (9), the operator $\mathcal{R}$ is missing between $M(-t_+)]$ and $|\psi_0\rangle$.
• Caption of Fig. 4(e): “the” is repeated.
• Add legend to Extended Data Fig. 3.

Remarks on text from the first version of the manuscript:

• In the main text, you write “… we decompose the XXZ Hamiltonian – derived from the secular approximation (see Supplement) …”. We could find this calculation in the Supplement only for an XYZ Hamiltonian. Is this a typo?

Referee #2

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Referee #4

(Remarks to the Author)
This is my second report on the revised paper by Gao et al. Overall, I do appreciate the efforts undertaken by the authors to make the manuscript more accessible and clarify a number of questions by myself and the other referees. On the other hand, I remain skeptical about the outlook toward practical use in metrology, which is a key aspect and selling point of solid-state sensors like NV centers. While the authors do give an outlook, even they call it “optimistic”. Thus, I remain of the opinion that the work will remain an academic demonstration of spin dynamics with a very narrow real-world perspective. This statement should not distract from the science and quality of the work, but it limits in my view its significance and suitability for a journal like Nature.

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We would like to thank all Referees for careful review of our manuscript and many helpful comments and suggestions. Based on these comments, we have made a number of revisions, which in our view, helped to improve our manuscript. In what follows we respond to all comments point by point and indicate revisions where appropriate.

All line numbers in our responses/revisions refer to lines in the revised draft. The revisions are indicated in red.

Comments of Referee #1:

The pursuit of improved metrology methods is a central theme in modern physics, as precision measurements underpin fundamental science and technological advancements. Quantum-enhanced sensing, leveraging many-body correlations and entanglement, has been a key focus in AMO systems, where techniques such as spin squeezing have enabled measurement sensitivities beyond classical limits. However, translating these advances to solid-state platforms presents unique challenges, including stronger decoherence, complex disorder, and technical noise. Solid-state quantum sensors, particularly those based on defect centers in diamond, offer practical advantages over AMO systems due to their robustness, scalability, and operation under ambient conditions. Methods that enhance metrological performance in such systems, such as the signal amplification approach explored in this study, are essential for extending quantum-enhanced sensing to real-world applications. This is particularly relevant for high-resolution magnetic imaging, materials characterization, and biological sensing, where solid-state platforms can provide nanoscale spatial resolution and direct integration with technological devices. The development of new protocols that exploit many-body dynamics for metrological gain in these settings represents an important step toward practical quantum technologies.

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The results presented in this work are highly relevant to the field of quantum sensing and many-body quantum metrology. The successful engineering of one- and two-axis twisting interactions in a disordered solid-state spin ensemble represents a significant advance in harnessing many-body effects for practical sensing applications. Moreover, the discovery that an asymmetric echo sequence—rather than the conventional symmetric approach—yields maximal signal amplification is conceptually unexpected and provides new insight into the role of time-reversal symmetry in many-body dynamics. This novel mechanism is not only of interest to specialists in NV-based sensing but also has broader implications for quantum metrology and quantum control. Given its relevance to both fundamental quantum physics and practical applications, the study is well-suited for publication in a high-impact journal such as Nature. While the manuscript is already strong, we encourage the authors to consider the following question and list of comments:

We thank the Referee for their careful reading of the manuscript and their support for publication.

1. A central aspect of the manuscript is the demonstration of signal amplification, with the susceptibility derived from Kubo's formula serving as the primary figure of merit. This contrasts with other studies on echo sequences, such as Refs. [12] and [37], which focus on the net metrological gain in terms of the signal-to-noise ratio (SNR). Given this difference in perspective, an important question arises: What can be said about the noise properties in the proposed sequence? Specifically, is it to be expected that the amplification observed in the susceptibility directly translates into an equivalent enhancement in the SNR? Furthermore, what is the dominant

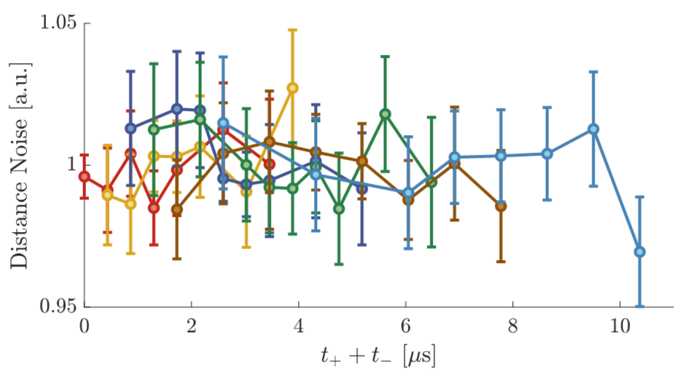
noise source in the experimental measurements on the NV ensemble, and would this noise be affected by the application of TAT dynamics? Addressing these points would help clarify the practical implications of the proposed amplification scheme for real-world quantum sensing applications.

The observed amplification is indeed expected to directly translate into an equivalent enhancement of the SNR, because the total noise strength does not change significantly under spin dynamics. This is because the readout noise of NV ensembles is typically dominated by photon shot noise (by at least an order of magnitude, see Ref. [24]), rather than the quantum projection noise of the spin states. Using the number $\sigma_R \sim 20$ in Ref. [24] (i.e. the photon shot noise is 20 times stronger than the quantum projection noise of a coherent spin state), we can do a simple estimation of how much the total noise would change when the quantum projection noise is doubled under spin dynamics. Assuming a composition rule of the noise $\sigma_{\text{Total}} = \sqrt{\sigma_{\text{Shot}}^2 + \sigma_{\text{Quantun}}^2}$, the total noise for a coherent spin state is $\sqrt{20^2 + 1^2} = 20.025$, while the total noise after doubling the quantum projection noise is $\sqrt{20^2 + 2^2} = 20.100$, which is only 0.4% larger than the former and thus negligible compared to the observed amplification. The above theoretical intuition is also supported by experimental data (see the newly added Extended Data Fig. 3, which is also attached below), where we explicitly see that the noise remains a constant (i.e. within the errorbar and smaller than the observed 6.7% amplification) despite the complicated spin dynamics.

To make the above points more explicit to readers, we added a sentence after line 176:

"Such amplification would directly translate into an equivalent enhancement in the signal-to-noise ratio (SNR), as the readout noise of NV ensembles is dominated by photon shot noise, which is not affected by spin dynamics (Extended Data Fig. 3)."

We also added Extended Data Fig. 3 to show the noise data for interested readers.



We also note that we have moved the Kubo formula to SI in response to critiques of Referee 4. In the main text, this discussion is replaced with a less mathematical description of the same physics.

Additionally, we suggest the authors address the following minor points:

2. The nomenclature in Eq. (1) differs from that used in the underlying calculations in the Supplementary Information. Please unify the notation for consistency.

It is our understanding that the Referee refers to the use of $\hat{\sigma}^{x,y,z}$ in Eq. (1) and $\hat{S}^{x,y,z}$ in the calculation in SI. This is an intentional choice because we use $\hat{S}^{x,y,z}$ for the full spin-1 operators of the NV electronic ground states, while using $\hat{\sigma}^{x,y,z}$ for the effective qubit defined on the subspace spanned by $\{|\tilde{0}\rangle, |\widetilde{-1}\rangle\}$.

To avoid such confusion, we explicitly stated this in line 806 and line 814. We also changed the notations for the full spin-1 operators from $\hat{S}^{x,y,z}$ to $\hat{J}^{x,y,z}$, which is less likely to cause confusion.

3. Is it possible to provide an analytical connection between the coefficients of the dipolar TAT Hamiltonian in Eq. (3) and the OAT Hamiltonian in Eq. (1), which was used to engineer the TAT Hamiltonian? A clearer analytical link would improve the theoretical understanding of the transition between these interaction regimes.

There is indeed a subtlety here as the Heisenberg part in Eq. (1) is not the same as the Heisenberg part in Eq. (3). This is because we conceptually decompose Eq. (3) into a Heisenberg part and a traceless part, while decomposing Eq. (1) into a Heisenberg part and an Ising part (which is not traceless). To emphasize the relation between them, we note that the trace (i.e. $J_{XX} + J_{YY} + J_{ZZ}$) of an XYZ Hamiltonian is an invariant under Floquet engineering (see Ref. [10]). Following this rule, the trace of Eq. (1) ($3J^{\text{Heis}} + J^{\text{Twist}}$) leads to a Heisenberg part in Eq. (3) that equals $J^{\text{Heis}'} = J^{\text{Heis}} + \frac{1}{3}J^{\text{Twist}}$.

We added this explicit relation in line 134 to improve the rigorousness of Eq. (3).

4. Please clarify what is meant by the term "semi-classical flow" (line 138). A brief explanation would make the description more accessible.

In line 139-140, we added the explanation "(i.e. mean-field spin dynamics under the corresponding classical Hamiltonian)" to address this.

5. The formulation in lines 157–158, "… a sensing rotation … is sandwiched between a pair of time-reversed TAT Hamiltonians," may be misleading. Please consider rewording to make it explicit that the second TAT Hamiltonian is time-reversed compared to the first one.

We changed the words to "… a sensing rotation … is sandwiched between a pair of forward and backward evolutions under the TAT Hamiltonian" to improve the precision of the sentence.

6. In line 219, it is stated that "quantum projection noise would become relevant in such cases." Please elaborate on why quantum projection noise is not currently relevant and under what conditions it would start to play a role.

We hope that our response to Referee 1's first point also addresses this question.

To improve the precision of language we changed the sentence to "quantum projection noise would become comparable to technical noise in such cases".

Referee #2 (Remarks to the Author):

This referee co-reviewed the paper with one of the other reviewers.

Referee #3 (Remarks to the Author):

The manuscript by Gao et al. reports experimental demonstration of quantum sensing enhanced by quantum correlations in the ensemble of coupled NV centers. Interactions between NV centers have previously been considered as a source of decoherence in diamond magnetometers. The manuscript demonstrates that coherent control methods allow to increase the performance of NV based sensors using coupling between NV spins. Although the signal amplification is relatively low (less than 10 per cent), the manuscript shows a very significant result in the field of quantum sensing and metrology. Remarkable, careful engineering of a coherent control protocol allows amplification in positionally disordered NV centers coupled by long range dipolar interactions.
The manuscript is the first demonstration of the use of quantum correlations for enhanced sensing. I recommend it for publication after addressing the following points

We thank the Referee for their careful reading of the manuscript and their support for publication.

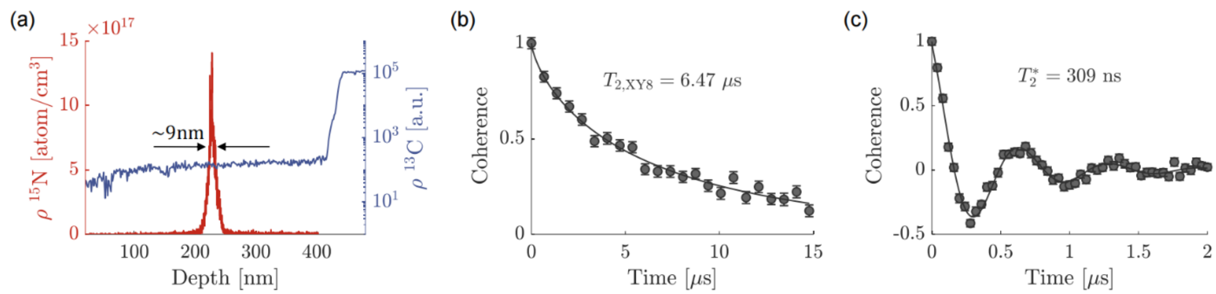
Authors use nearly two-dimensional layers of NV centers. I recommend including SIMS data in the additional material section.

This is added as Extended Data Fig. 2(a).

The concentration of substitutional nitrogen (P1 centers) is not given. Although EPR measurements are not possible for such a thin layer, they can be detected by double electron resonance. I expect that these electron spins could influence the dynamics of the NV centres.
I also recommend to include details on the determination of NV centers concentration in the doped layer.

Unfortunately, we have not done DEER measurements on the sample, but as a proxy, we have Ramsey measurements that provide conceptually similar information. Assuming that the on-site disorder felt by the NVs are dominated by interaction with P1s (which is typically the case in these high-density samples), the P1 concentration can be calculated based on the measured T_2^*. Considering other sources of on-site disorder, this method provides an upper bound on the P1 density. The NV centers' concentration is characterized with similar method, by measuring the decay timescale T_2 under XY8 sequence, where the decay is dominated by dipolar interaction between the NVs.

These discussions are added to Methods (line 402-411), and relevant data are presented in Extended Data Fig. 2(b-c), which is also attached below.



The authors discuss the effects of laser power drift and signal normalization. Is there also an effect of the charge instability of the NV centres observed for the sample, and if so, how does this affect the measurement?

We have not observed any charge instability in this sample, but we designed our experiment in a way robust against it in case it has any effects. Specifically, we use a fixed laser illumination duty cycle across all data points to keep the equilibrium concentration of NV^- and NV^0 constant. Moreover, even if there is some residual fluctuation of charge state concentration (which leads to a fluctuation in the optical spin contrast), it is further accounted for by the full contrast measurements that are interleaved with the main measurements.

The above discussion is added in the manuscript in line 460-462.

Referee #4 (Remarks to the Author):

The paper by Gao and coworkers discusses whether the spin-spin interactions in a solid-state spin ensemble can be exploited for improved sensing performance, compared to the case of an equivalent non-interacting spin ensemble. The authors demonstrate that this is indeed possible, but highly challenging because of the anisotropy of the dipolar spin-spin couplings and the random locations of the spin defects. The authors address these challenges by confining the spins to a two-dimensional layer and aligning them with the out-of-plane direction, reducing dipolar anisotropy, and by refocusing the time evolution to produce an echo signal, mitigating the large spread in couplings that comes with the random spin locations. Both ingredients are key for achieving a net signal improvement. They report a signal improvement by 6.7%.

In my view, the significance of the reported work lies in demonstrating a net improvement in sensing performance, even if the improvement is marginal in a practical sense. As far as I know this is the first demonstration of signal enhancement in a solid-state spin ensemble. Since solid-state spin systems have many practical applications in metrology (see Refs. 1-7), this is a significant result.

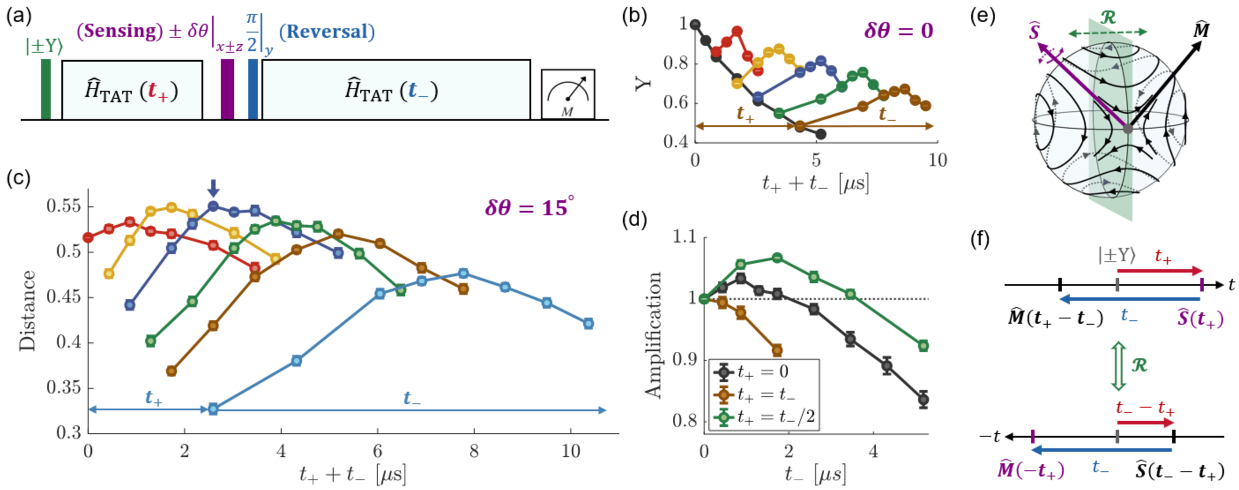
We thank the Referee for their careful reading of the manuscript and their appreciation of the innovations in this work.

Once this has been said, I have two major reservations with regard to publication in Nature. First, I have to admit that the paper is very challenging to read and understand, especially the discussion of the time-reversed echo. While this is advanced physics, I find the presentation unnecessarily complicated for the general reader. In its present form, the paper would be better suited for a specialist physics paper.

We thank the Referee for this helpful suggestion regarding the presentation. Upon revisiting the manuscript, we agree that the discussion of the time-reversed echo was unnecessarily complicated/technical. To address this, we added a new panel (f) in Fig. 4 (which is also attached below) to visually explain the implications of the time-reversed mirror symmetry. We also rewrote this section to describe a simple physical picture with minimal equations (the original discussion is moved to SI and expanded upon there). With the revised presentation, we believe it is now clear that the mechanism under discussion is conceptually novel and broadly applicable across a wide range of many-body Hamiltonians and quantum sensing platforms.

The inclusion of the new panel enables a more intuitive explanation of the mechanism. In panel (e), we visually see that mirror reflection around the YZ plane ($\mathcal{R}$) preserves the flow lines, up to a reversed flow direction. Therefore, when combined with time reversal ($t \leftrightarrow -t$), the flow direction is restored, and the resulting combined transformation forms a symmetry of the underlying Hamiltonian (which justifies the term "time-reversed mirror symmetry"). As a combined transformation of time reversal and mirror reflection, this symmetry flips the sign of time ($t \leftrightarrow -t$) and exchanges the sensing ($\hat{S}$) and measurement ($\widehat{M}$) operators (as illustrated in panel (e)). Applying these two rules to the asymmetric echo protocol in panel (f), the sensing operator at time t_+ (i.e. $\hat{S}(t_+)$) is mapped to $\widehat{M}(-t_+)$, and the measurement operator at $t_+ - t_-$ (i.e. $\widehat{M}(t_+ - t_-)$) is mapped to $\hat{S}(t_- - t_+)$. This transformation links the original asymmetric echo to an equivalent protocol with a modified forward evolution time $t_+' \equiv t_- - t_+$(see panel (f)), ensuring that the amplification amplitude – defined as $\chi \equiv \left|\mathrm{d}\langle\widehat{M}\rangle/\mathrm{d}(\delta\theta)\right|$ – remains the same between them (i.e. $\chi(t_+, t_-) = \chi(t_- - t_+, t_-)$). At the time $t_+ = \frac{t_-}{2} + \epsilon$, this relation transforms to $\chi\left(\frac{t_-}{2} + \epsilon, t_-\right) = \chi\left(\frac{t_-}{2} - \epsilon, t_-\right)$, guaranteeing a symmetry of amplification around $t_+ = t_-/2$ and thus an extremum at this point.

To enhance accessibility to general readers, we have moved the abstract discussion – including the Kubo formula and the mathematical definition of the time-reversed mirror symmetry – to SI. In the main text, we focus on the above discussions (line 203-216), emphasizing the visual representation of time-reversed mirror symmetry (panel (e)) and the key physical intuition that links this symmetry to the optimal condition $t_+ = t_-/2$ (panel (f)). Additionally, we separated the generic result that $t_+ = t_-/2$ is an extreme point from the more specific discussion explaining why this extremum is a local maximum, presenting them in distinct paragraphs (line 217-231) to improve the logical structure. With this revised presentation, which integrates intuitive explanations and visual aids, we believe the discussion is now accessible to a broader readership.



The second reservation I have is regarding the impact of the signal amplification beyond the basic demonstration. Even in a most optimistic scenario, the amplification will only be a small factor. In practical uses of solid-state quantum sensors, many other parameters influence sensitivity (material quality, environment in which sensor is operated, optimization of optical readout, etc.), and those usually impact sensitivity much more than a subtle 6.7% improvement. Therefore, while the authors should be congratulated on achieving a positive gain, I question the optimistic outlook on practical advances.

We agree that the currently achieved 6.7% amplification is marginal for practical applications, but we are optimistic about future improvements. We identify two main routes that have the potential to enable significant improvements in the near future:

（1） It is theoretically predicted (Refs. [41] and [42]) that scalable spin squeezing (and signal amplification) can be achieved by engineering interactions that support finite temperature XY ferromagnetic ordering. Although the presence of positional disorder may shrink the ordered phase boundary (Ref. [43]) and thus tighten the constraints for scalability in the thermodynamic limit, our numerical simulations at intermediate system size (N=200, Extended Data Fig. 9(a)) predict an amplification exceeding 3 (i.e. 9.5 dB), with consideration of positional disorder and realistic 2D layer thickness (9nm). Moreover, even when we consider a realistic spin polarization (80%, dashed lines in Extended Data Fig. 9(a)), an amplification approaching 2 (i.e. 6dB) is predicted. Given recent advances in NV coherence time (Ref. [56]) and in CVD growth of (111)-oriented diamonds (Ref. [29]), such opportunities can be explored experimentally in the near future.

（2） As confirmed by our numerical simulations (Extended Data Fig. 8), the dominant limiting factor in the system is the positional disorder of spins, for which we know two potential ways to overcome:

a) There are studies on controlled NV creation with direct laser writing that achieve a spatial accuracy of 33 nm in combination of a near-unity yield (Ref. [44]). Further advances in such technique may enable interacting NV ensembles on a lattice, which would lead to much larger amplification, as predicted by our numerics (Extended Data Fig. 8).

b) Even without advances in material growth, we recently proposed a method based on magnetic field gradients that is fundamentally robust against spin dimers (Ref. [45]). This method relies on a spin spiral initial state (created using magnetic field gradient) evolving under a Heisenberg Hamiltonian $H = J_{ij}\vec{S}_i \cdot \vec{S}_j$. Intuitively, this method is robust against spin dimers because the dimers – which live close in space – point in roughly the same direction on the Bloch sphere in a spin spiral initial state. Such a state (i.e. two spins pointing in the same direction) is an approximate eigenstate of the Heisenberg Hamiltonian, leading to suppressed contribution of dimers to the many-body dynamics. Our numerics show that this method allows us to achieve scalable spin squeezing (and amplification) despite the positional disorder, and we are now actively exploring this route experimentally.

Beyond the optimistic outlook on realizing larger amplification, we also want to point out that a significant innovation in this work is the generic mechanism of asymmetric echo based on time-reversed mirror symmetry. Such discovery is conceptually unexpected and provides new insights into the role of time-reversal symmetry in many-body dynamics. Therefore, we believe that this work will have broader interest beyond NV-based quantum sensing, and be relevant to quantum metrology and quantum information science in general.

Regarding the manuscript, we agree that the sentence (in line 233-236) "Our numerical analysis shows that by reducing the positional disorder towards a 2D lattice, suppressing the dynamical on-site disorder by going to lower temperature, and improving the spin polarization, an improvement of the amplification above 10 (i.e. 20~dB) is expected theoretically" is very optimistic as it neglects many experimental imperfections (positional disorder, decoherence and polarization limitations). We have replaced it with a more realistic outlook pointing to near-future opportunities as discussed above. In addition, we also acknowledge other methods that improve practical sensitivity (which are in principle compatible with signal amplification demonstrated in this work) in the outlook (line 250-251), and point to future works for integration of our approach with them.

The revised outlook reads as follows:

Our results can be extended along several directions. A natural goal is to realize larger, scalable (i.e. improve with system size N) amplification, to achieve practical benefits. One promising approach is to engineer interactions that support XY ferromagnetic ordering [41, 42]. While positional disorder may shrink the ordered phase boundary [43] and thus tighten the constraints for scalability in the thermodynamic limit, our numerical simulations at intermediate system size

($N = 200$, Extended Data Fig. 9(a)) supports a significant enhancement of amplification. This can be explored experimentally by using recent advances in CVD growth of (111)-oriented diamonds [29] (see Supplement and Extended Data Fig. 9). Additionally, overcoming positional disorder – the primary limitation in the system (Extended Data Fig. 8) – presents another opportunity for improvement. This challenge motivates exploring controlled NV creation [44], or integration with strong magnetic field gradients to explore mechanisms robust to positional disorder [45]. From a practical standpoint, another interesting direction is to combine our approach with other sensitivity-enhancing techniques (e.g. improved spin readout [24, 46]). Such combination could enable exploring new opportunities beyond signal amplification, since quantum projection noise would become comparable to technical noise in such cases. Ultimately, combined with other features of solid-state quantum sensors, such as the capability of bringing the sensors to close proximity with sensing targets [6] and the injection of sensor-containing nanoparticles for in vivo measurements [7, 47], signal amplification can lead to practical advances in nanoscale imaging technologies for novel condensed matter materials and biological structures.

In SI, we updated Extended Data Fig. 9 (previously Extended Data Fig. 11) to include the same simulation with a realistic spin polarization of 80%, making it more relevant to near-future experiments. We also moved it to be exactly after Extended Data Fig. 8, where experimental limitations to the amplification are discussed, to improve the logical connection.

Technically, the experimental body of work and the analysis look sound, although I cannot judge this in depth. The approach of confining the spins to a thin two-dimensional layer combined with toggling spins alignment between 111 and 100 is neat and innovative. The demonstration of Fig. 2b (main plot vs. inset) is convincing in confirming the twisting dynamics.

Further comments:

1) Could the authors briefly motivate the choice of bias field? Why 890 Gauss instead of another value?

We chose this value to maximize the qubit frequency within the constraints of achievable strength of the pulsed field. Higher qubit frequency is in general better as it allows better rotating wave approximation (RWA) and faster pulses. However, as shown in Fig. 1(b), higher qubit frequency (i.e. length of the purple arrow $\eta_{\text{eff.}}$) requires a stronger pulsed field (red arrow), which requires a larger current on the wires below the diamond. Such technical constraints limit the qubit frequency and lead to this choice of the B field.

In the manuscript, we added the brief explanation "890 G, chosen to maximize the qubit frequency within technical constraints" in line 77-78.

2) On line 172, the amplification by 6.7% is claimed from Fig. 4c. How are these 6.7% obtained?

Does this correspond to the relative increase (of the blue curve?) at t+ + t- >0 compared to the red trace at t=0? Or does this correspond to the peak value of the green curve in Fig. 4d?

It is both, as Fig. 4(d) is a replot of Fig. 4(c).

To make it clearer to readers, we removed all downward arrows in panel (c) except the blue one, which points to the location at which maximum amplification is achieved. We also added one sentence in the caption "The downward blue arrow indicates the point with maximum amplification, relative to the non-interacting reference at $t_+ = t_- = 0$".

3) Although the time-reversed echo is explained at length, I still do not fully capture the mechanism. Is the t- = 2t+ asymmetry a general feature of any type of qubit pair interaction, or does it only apply to spin-spin interactions that are dipolar in character (where the coupling constant varies between -J and +2J depending on the angle between the spins' axes and the

We hope our response to Referee 4's first point makes this clear. To summarize:

（1） The extreme value of amplification at $t_+ = t_-/2$ is a generic consequence of the time-reversed mirror symmetry (which is shared across a wide range of Hamiltonians, such as OAT, TAT, and LMG, as shown in Extended Data Fig. 4). But such symmetry does not necessarily guarantee that the extremum is a local maximum (although a maximum is observed in most scenarios, see Extended Data Fig. 4).

（2） When the system is dimer dominated (as in this work), the observed local maximum can be explained by dimer refocusing under the TAT Hamiltonian, requiring a microscopic model beyond general symmetry arguments (see SI and Extended Data Fig. 5). When the Hamiltonian deviates from the TAT point, the local maximum change into a local minimum (Extended Data Fig. 4(d)), but the symmetry around $t_+ = t_-/2$ is kept, as dictated by time-reversed mirror symmetry.

（3） When the system is not dimer dominated, we generically observe a local maximum in our numerics (Extended Data Fig. 4(b-c)). We have a heuristic argument (Extended Data Fig. 4(e-f)) for this observation, but the exact conditions for a local maximum required further investigations.

（4） The asymmetric echo is not related to -J and +2J dependence on the angle, because all spin pairs are "head-to-head" (i.e. spin separation perpendicular to the quantization axis, leading to the "-J" coupling) under the out-of-plane quantization axis $\eta_{\text{eff.}}$ In other words, the system does not even "know" that there exists a direction with "+2J" coupling, because there are no spin pairs along this direction (at least in an ideal 2D plane).

In the manuscript, we improved the logical structure of relevant discussion. Firstly, we split the general consequence of time-reversed mirror symmetry from the more specific discussions in the dimer dominated regime, presenting them in distinct paragraphs to improve the logical flow. We also moved the sentence "Although such symmetry only guarantees an extreme value of amplification at $t_+ = t_-/2$ instead of a maximum value, we show that a maximum is expected in most cases" from line 263-265 to line 229-231 (with some rewording), to put it close to relevant discussions.

We would like to thank all Referees for careful review of our revised manuscript and many helpful comments and suggestions. In what follows we respond to all comments point by point and indicate revisions where appropriate. The revisions are indicated in red.

Comments of Referee #1:

The revised manuscript provides an improved presentation which, in our opinion, addresses all points raised in our report and, we believe, also those raised by the other referees. Major changes in the revision include a more intuitive, pictorial explanation—supported by an additional figure (Fig. 4e)—of the new echo protocol for signal amplification based on time-reversed mirror symmetry, as well as an expanded discussion of the broader perspectives of the current work in the manuscript's outlook. Both changes strengthen the presentation and enhance its accessibility to non-expert readers. As expressed in our initial report, we believe the authors present a strong, surprising, and important result that demonstrates a novel concept in quantum metrology within solid-state systems and warrants publication in Nature. We recommend acceptance of the revised manuscript.

We thank the Referee for their careful reading of the manuscript and their support for publication.

We provide the following remarks for the authors' consideration regarding the new or revised sections of the manuscript:

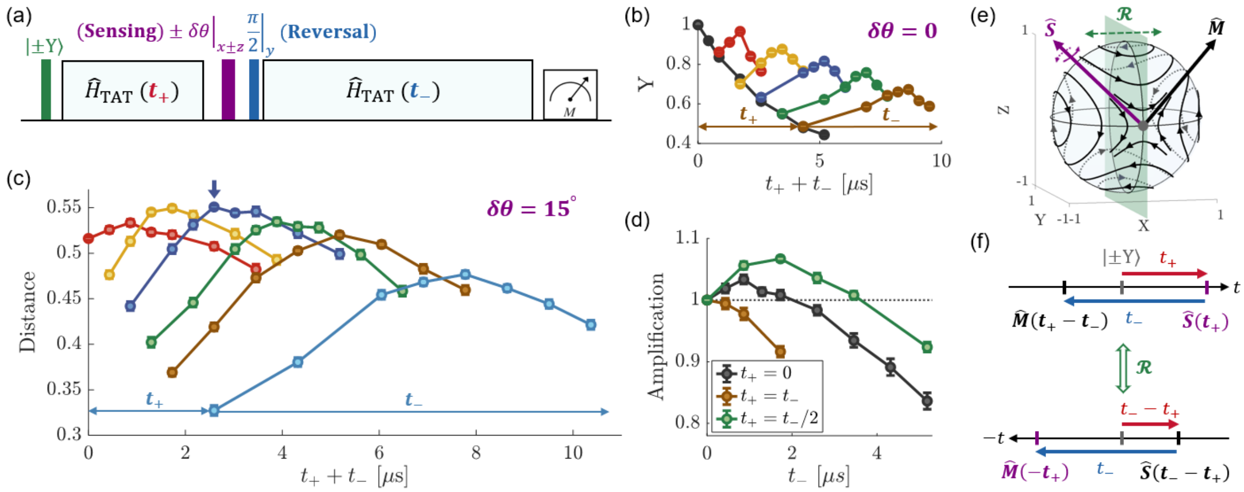
• Lines 204–206: In the sentence "Specifically, the dynamics ...", it remains unclear what dynamics are depicted in Fig. 4(e). Adding "the dynamics under the TAT Hamiltonian" and rephrasing the sentence—for example, starting with "As illustrated in Fig. 4(e)..."—could improve clarity for the reader.

We thank the referee for pointing this out. The sentence has been rephrased to "As illustrated in Fig. 4(e), the dynamics under the TAT Hamiltonian remain invariant - up to a reversal of direction - under reflection about the YZ plane (the green shade, $\mathcal{R}$)."

• Lines 204–206: Later in the sentence, the authors write "… under the reflection about the YZ plane (R)." From Fig. 4(e), it is not immediately clear to the reader that the reflection plane is the YZ plane rather than the XZ plane. Adding axis labels to the figure could help clarify this point.

We tried adding axis label to the figure (see below), but we think the extra labels make the figure look busy, as there are already many labels and arrows in the panel (e) and (f). Because of this, we decided to make the point clear by explicitly stating in the text that the YZ plane is the green shaded plane:

"As illustrated in Fig. 4(e), the dynamics under the TAT Hamiltonian remain invariant - up to a reversal of direction - under reflection about the YZ plane (the green shade, $\mathcal{R}$)."



• Check if articles are missing in line 219 ("by the TAT Hamiltonian") and line 222 ("by the refocusing").

We added articles at these two locations.

• Line 226: In the sentence "This conclusion is further supported by …" it is claimed that a local minimum is obtained for a modified (XYZ instead of TAT) Hamiltonian. From Extended Data Figure 6 it is not immediately clear why this results in a minimum.

The presence of the local minimum is because the original maximum at $t_+ = t_-/2$ splits into two distinct maxima under the modified refocusing conditions (as discussed in Supplement and shown in Extended Data Fig. 5(g-h) – which was previously Extended Data Fig. 6). Therefore, the original location $t_+ = t_-/2$ – which now sit between two local maxima – becomes a local minimum. To make this logic clearer to readers, we changed the sentence to "This conclusion is further supported by modifying the Hamiltonian such that a local minimum is obtained, as the original maximum splits into two peaks under the modified refocusing conditions (Extended Data Fig. 5(g-h))."

• Lines 242–245: The sentence "While positional disorder …" is rather technical. It may help to rephrase in order to make it more accessible.

The essential information we want to convey in this sentence is that although scalability in the thermodynamic limit can be negatively affected by the presence of positional disorder, we still expect a significant enhancement at intermediate system size. We agree that the details "While positional disorder may shrink the ordered phase boundary and thus tighten the constraints for scalability in the thermodynamic limit" is unnecessary for general readers, and have rephrased the sentence as "While positional disorder may pose a challenge for realizing scalability in the thermodynamic limit, our numerical simulations at intermediate system size (N=200, Extended Data Fig. 7(a)) supports a significant enhancement of amplification."

• Line 249: "explore" is used twice in the same sentence.

We replaced the second "explore" by "investigate".

• In Eq. (6) of the Supplementary, the "dynamical susceptibility" χ is defined without an absolute value, while an absolute value is used in the main text (line 213). Moreover, different terms are used in the main text ("amplification amplitude" in line 212) and in the Supplementary ("dynamical susceptibility" in line 555).

We thank the Referee for pointing out this subtle point. To keep notation and language consistent, we changed the notation for "amplification amplitude" in the main text from χ to $|\chi|$. Relatedly, we rephrased the sentence in SI to point out the relation between "amplification amplitude" and "dynamical susceptibility" as "the amplification amplitude is measured by the absolute value of the dynamical susceptibility, which is quantified by the Kubo formula".

• Line 569, typo: "on" should be "in".

We fixed the typo.

• In line 3 of the proof of Eq. (9), the operator $\mathscr{R}$ is missing between $M(-t_+)]$ and $|\psi_0\rangle$.

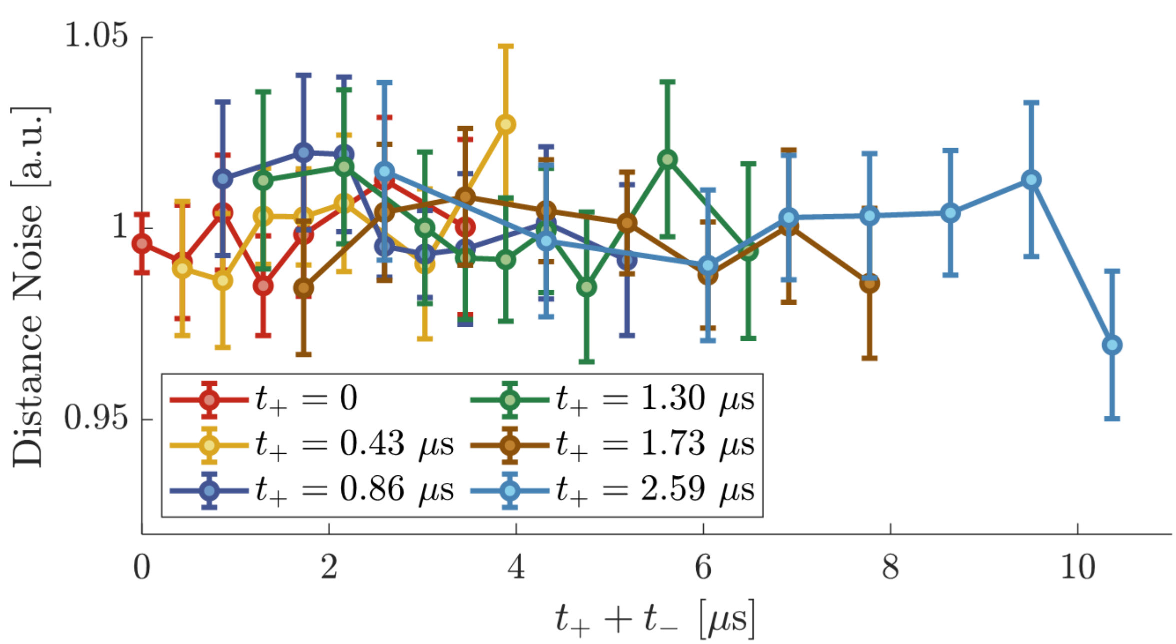
The problem comes from skipping a step in the derivation: $\mathcal{R}|\psi_0\rangle = |\psi_0\rangle$ according to Eq. (7). We agree that this can lead to confusion for interested readers, and have fixed it by including this step as a separate line in the derivation.

• Caption of Fig. 4(e): "the" is repeated.

We fixed the problem.

• Add legend to Extended Data Fig. 3.

We added the legend (see below).



Remarks on text from the first version of the manuscript:

• In the main text, you write "… we decompose the XXZ Hamiltonian – derived from the secular approximation (see Supplement) …". We could find this calculation in the Supplement only for an XYZ Hamiltonian. Is this a typo?

This is not a typo. In the SI section "calculation of the effective Hamiltonian", we calculated the effective Hamiltonian in the qubit subspace, whose XX coefficient equals to its YY coefficient, and therefore is an XXZ Hamiltonian.

To make this section easier to find for interested readers, we changed its title to "calculation of the effective XXZ Hamiltonian" and built a table of content at the beginning of SI.

Referee #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Referee #4 (Remarks to the Author):

This is my second report on the revised paper by Gao et al. Overall, I do appreciate the efforts undertaken by the authors to make the manuscript more accessible and clarify a number of questions by myself and the other referees. On the other hand, I remain skeptical about the outlook toward practical use in metrology, which is a key aspect and selling point of solid-state sensors like NV centers. While the authors do give an outlook, even they call it "optimistic". Thus, I remain of the opinion that the work will remain an academic demonstration of spin dynamics with a very narrow real-world perspective. This statement should not distract from the science and quality of the work, but it limits in my view its significance and suitability for a journal like Nature.

We thank the Referee for their careful reading of the manuscript and their appreciation of the innovations in this work.