­­­­Dear editor and reviewers:

Thanks to the constructive criticism that we have received from you, we are pleased to present this thoroughly revised manuscript. In addition to carefully addressing all of your comments regarding the body of the text, we have also made significant changes to its emphasis and presentation. We have also included a thorough discussion of implications for previous results in a Supplementary Material section.

Sincerely,

David Reens, Hao Wu, Tim Langen, and Jun Ye.

Referee Comments:

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Report of Referee A -- LF16145/Reens  
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This paper presents a combined electric and magnetic trap for OH  
molecules designed to minimize losses due to non-adiabatic  
transitions. The design allows the loss rate to be tuned and to be  
suppressed to small values. Experimental data on trap loss is  
presented that seems consistent with the predictions of a model of  
loss due to non-adiabatic transitions.

We agree with this synopsis as it pertains to our previous manuscript, but we have rearranged our emphasis in the new manuscript, see from the bottom of the second paragraph: “But the electric field can also dramatically enhance spin-flip losses, due to internal spin-dynamics that we corroborate with direct experimental evidence for the first time in the present work. We achieve this with a novel trap geometry that also allows complete removal of the loss with minimal sacrifice of trap strength.”

In addition to presenting the new trap, the authors suggest that the  
paper brings new insights into the loss mechanism. I do not find this  
convincing.

We greatly appreciate the reviewer’s opinion about the lack of insight provided about the loss mechanism. We have thoroughly revamped our discussion of the mechanism and made this the primary focus of the manuscript. We believe that we now successfully bring new insights in the following ways:

Firstly, by corroborating with direct experimental evidence, which we now make a key point of the manuscript, as in sentences just quoted above.

Secondly, by giving a clear intuitive picture based on the internal spin dynamics of the molecule. See the first full paragraph on page 2: “We begin with an intuitive picture. In order to remain well trapped in combined fields, a molecule must remain weak field seeking with respect to both fields, i.e. doubly stretched. This means that its quantization axis, which is proportional to the field induced energy shift of the molecule, should have maximal length…”

Thirdly, by developing a scaling law for the effect, which demonstrates its relevance even at comparatively low electric field magnitudes, see the paragraph on the right column of page 2, which begins, “We have also developed an algebraic scaling law,” and goes on to say, “Crucially, it is not ∆ that sets the relevant scale, as one might naively suppose given that this is the energy beyond which the Stark effect is linear and the molecule is polarized. Instead it is √κ∆, which is in general much smaller.” We have also much more explicitly calculated this scaling law in Sec. C of our Supplementary Materials.

The mechanism rests on two ideas, outlined in the first  
two pages of the paper. The first is that the molecule is insensitive  
to small magnetic fields that are perpendicular to a strong applied  
electric field. This has been known for about fifty years and is a  
general result, not specific to any Hund’s case or even specific to  
polar molecules. It applies to atoms [e.g. J. Phys. B 3, 1620 (1970)]  
and to molecules with no orbital angular momentum [e.g. Phys. Rev.  
Lett. 89, 023003 (2002)].

We thank the referee for introducing us to this important connection with the existing literature related to the influence of magnetic fields on eEDM measurements. We believe this connection enhances the relevance of our work. It is indeed the case that the very same suppression of Zeeman influence over atomic and molecular eEDM measurements is related to the effect we describe. Nonetheless, there are important differences. To clarify these points in our manuscript we have added the following sentences in the text, "This Zeeman splitting suppression is in fact a known phenomenon in the precision measurement community [41, 42], and experimentalists have exploited it to suppress the influence of magnetic fields in electron EDM measurements. However, in the case of applying mixed fields during trapping, this suppression is not beneficial but rather detrimental."

We also note that the loss mechanism for molecules in traps results from the suppression of Zeeman influence in a plane where E and B are truly perfectly perpendicular, a situation not realized in eEDM searches but often unavoidable in a magnetically confining geometry for a trapped species. Finally, we point out that while the effect can certainly occur for all species, the strength of the electric field required is often greatly lessened for molecules.

The second idea is that, in regions where  
the electric and magnetic fields are perpendicular, and the magnetic  
field is weak, this suppression of the Zeeman splitting will lead to  
an enhancement of spin-flip losses. Reference [29] of the paper  
already discusses exactly this point, and in a way that is  
considerably clearer than in the present manuscript. In reference [29]  
the non-adiabatic loss rate is calculated as a function of electric  
field. I do not see anything in the present paper that differs from  
that calculation.

We agree that the essential theoretical details of the loss enhancement that we describe appear in the appendix of Ref [29], but we strongly believe that a separate publication is nonetheless warranted. Our reasons are threefold:

Firstly, we have indeed made a significant improvement to the calculation in Ref [29], which we had not previously mentioned due to our suspicion that the breadth of readership of Appendix A of Ref [29] may have been limited. However, we are very grateful for the reviewer’s careful attention in this regard, because we feel it has resulted in a much stronger manuscript. We now discuss our improvements explicitly in the main text, on the bottom of page 2: “the direct integration of flux is a key improvement relative to our previous work [37], where electric fields were applied to study collisions.” We have also discussed the improvements in a detailed fashion in Sec. A of our Supplementary Materials: “Our improvement relates to the next step, where an integral calculation for the loss rate is performed. In Ref. [1] the integration spans the entire 3D spatial distribution, weighted by the frequency of crossing of the center plane and the chance of loss for each crossing… A more accurate treatment that we use here is to perform an integration of flux through the loss plane directly…”

Secondly, we have corroborated these calculations with direct, un-convolved experimental evidence for the first time. We now discuss this explicitly, beginning near the bottom of page 3: “… enough to explain a significant portion of the effect previously attributed to collisions, see Sec. A of the Supplementary [44]. In light of this, it becomes especially important to perform direct, unconvolved experimental verification of both the magnitude of the loss effect and the validity of our loss-flux calculations. We now present the new trap where this is achieved.”

Finally, our message to the community is now completely altered relative to that in Ref. 29. We are no longer presenting a side point that can be deconvolved, but an important effect that must be completely addressed by anyone working in similar regimes earlier than previously realized.

In my view, the novelty in this paper is entirely the new trap design  
and the demonstration that it suppresses losses that were a problem in  
an earlier design. That is an incremental advance which, in my view,  
does not warrant publication in Physical Review Letters. The work will  
certainly be of interest to specialists in the field of cold  
molecules, especially to others who might be interested in trapping  
molecules using a combination of electric and magnetic fields. This  
paper would be suitable for a more specialized journal once the  
following points have been addressed:

We strongly disagree with this view of our trap as an incremental advance. We specifically designed this trap not necessarily as our next generation trap but as an important testbed for spin-flip losses. Nonetheless, we can see how this interpretation is supported by our previous manuscript, and we are very grateful for the reviewer’s feedback in this regard. We have overhauled our discussion and presentation to address this, in many of the ways already discussed. The message about the relevance of the new trap as an incremental advance in our experiment is no longer a key emphasis of the manuscript and is relegated to an outlook paragraph in the right column of page 4, which begins: “The present trap, in addition to providing the desired experimental testing ground for molecular spin-flip loss, produces large 5 T/cm trap gradients useful for maintaining high densities to facilitate collisional studies.”  
  
1. The paper seems to be correcting two earlier papers. Those  
corrections are important but they are described too briefly to  
understand their implications. The first is to reference [29]. The  
correction is mixed up with the introduction to the paper and appears  
as a single sentence “subsequent investigations have revealed it to be  
a threefold underestimate, enough to render deconvolution of any  
remaining collisional effect difficult.” The authors should give a  
fuller account here and give a clear statement about the implication  
of this correction. Does it mean that the effects previously  
attributed to collisions in [29] are now accounted for entirely by  
non-adiabatic losses leaving no remaining evidence for collisions?

We agree that a full account is needed, and indeed this desire for a full accounting motivated the investigations that have led to this manuscript. We agree with the reviewer’s observation of the brevity of the corrections, and we address this in several ways in this new manuscript, especially in the inclusion of detailed discussions in Sec. A of our Supplementary Materials. The summary is that the effects previously attributed to collisions are almost entirely accounted for, enough so that any remaining influence of collisions will need to be explored in a separate experiment that more clearly separates the collisional and the spin-flip loss effects.

The second correction is to reference [22]. The authors write “This new  
understanding modifies our interpretation of evaporation data for OH  
[22], especially at 5 mK where the loss rate is significantly enhanced  
by the E-fields used for RF knife purposes. We do find enhancements in  
normalized density at low fields for shallow RF knife cuts from 55 -  
30 mK.” The observation of evaporative cooling of OH is a very  
important result. The community should know whether or not there  
remains strong evidence for evaporative cooling under this modified  
interpretation. The statement currently given is not clear on this.

We agree that the implications of this new interpretation are important for the community. We have now added a complete description of our current best understanding in Sec. B of our Supplementary Materials. Essentially, the new interpretation does weaken the evaporative cooling result, especially by undermining the hypothesis of cold samples building up in the low regions of the trap where the spectroscopic technique was insensitive. From the bottom right paragraph of page 2 of the Supplementary Materials: “Some of the temperature fits performed in Fig. 3 of Ref. [2] relied on this assumption, which we now no longer use. We rely on only the directly experimentally accessible spectra, such as those shown in panels (a-c) (Fig. 3 of Ref. [2]). After taking similar measurements repeatedly, the depletion spectra are found to be useful to identify enhancements in density caused by the evaporation. Figure S3 show such enhancements for evaporation sequences designed to achieve a twofold temperature reduction.”  
  
2. H\_{E perp B} has not been defined. The context of the sentence  
implies that it refers to “the exact eigenenergies of OH”. But it  
seems from perturbation theory that the series expansion of the  
energies will have a non-zero second-order contribution, i.e. when a  
small magnetic field is applied perpendicular to the applied electric  
field, the energies scale as B2, not as B3. The energy splitting  
between the M=±3/2 states scales as B3. Is that what is meant by H\_{E  
perp B}?

Yes, this is exactly what is meant by H\_{E perp B}, we are grateful for the observation of this oversight. We address this by using the symbol $G$ to refer to this quantity and more clearly defining it. From page 2, left column, second full paragraph: “This intuition agrees with a more rigorous analysis of the energy splitting G between the trapped state and its spin-flip partner…”  
  
3. The paragraph about molecules in Hund’s case (b) seems incorrect,  
at least when hyperfine structure is included, which is often  
necessary. There will be a tensor Stark splitting which suppresses the  
Zeeman splitting in a perpendicular magnetic field, and this  
suppression is stronger for larger electric fields.

Perhaps the referee is saying the following… tensor Stark splittings suppress the Zeeman splitting in perpendicular magnetic fields, so that Hund’s case (b) molecules with hyperfine induced tensor Stark splittings still experience loss enhancement. This statement is in agreement with our assertions about Hund’s case (b), where we already explain that the spin-rotational coupling results in the Stark splitting still suppressing the Zeeman. The spin-rotational coupling is in general quite large, and usually more significant than hyperfine effects. We have reworked the section somewhat, so hopefully our claims come across more clearly, see the bottom of page 3 into page 4: “One way to avoid competition is for the fields to couple to unrelated parts of the Hamiltonian, which happens to a limited extent for Hund’s case (b) states without electron orbital angular momentum (Σ states, Λ = 0) [40]. In these states, which include most laser-cooled molecules thus far, the electric and magnetic fields couple to rotation and spin respectively, which are only related by the spin-rotation coupling constant. This constant is usually in the tens of MHz [30], so molecular spin-flip loss remains quite significant.”

4. The paragraph that begins “We can develop a scaling law…” is too  
densely written for readers to easily follow.

We agree that this description is written very densely. Too address this, we have moved the scaling law derivation to Sec. C of our Supplementary Materials, and been much more explicit about what is being done. We have also moved the subsections of this paragraph that had discussed the application of the Landau Zener formula, which is now explicitly provided in Eqn. 2, into their own separate paragraph, beginning with “To deduce the effect” on the right side of Page 2.

It is not clear why the  
expression given for kappa is the correct one. The expression is given  
in the context of the Landau-Zener formula. That formula contains the  
matrix element of a term in the Hamiltonian that connects the two  
states. The two relevant states here are stated: “the spin-flips we  
have discussed connect |f, 3/2> to |f,-3/2>”. But I do not think there  
is a term in the Hamiltonian that connects directly these two states  
and so it is unclear how the Landau-Zener theory is being applied.

Indeed, the Hamiltonian does not contain a term directly connecting |f, 3/2> to |f,-3/2>. However, in the vicinity of their avoided crossing an effective two state Hamiltonian could be perturbatively deduced at third order, since the states differ by 3*m*. This Hamiltonian would contain an off-diagonal coupling term with a value precisely equal to the half-gap between the states that can be measured by numerically diagonalizing the full Hamiltonian. We have actually taken great caution in making use of this assumption, and have even performed full time-dependent schrodinger solutions using the eight state Hamiltonian to ensure agreement with the hopping probability predicted by the Landau-Zener formula found in this manner.

It would be better to take some space to set out the theory clearly –  
what is the Hamiltonian, what are the adiabatic states being  
connected, what is the matrix element that goes into the Landau-Zener  
formula, and how does this then lead to the result given?

We have not explicitly included this discussion, but have alluded to it by stating: “We can also set κ to the minimum energy gap along the trajectory, which is found in the plane.” This occurs in the middle of the paragraph containing the Landau Zener formula, Eqn. 2.

The result  
given for eta is similarly obscure; again it would be better to  
explain more fully how to arrive at this result.

Supplementary Materials Sec. C now explains this much more fully.

5. It is unclear how the model results given in figure 4 are obtained.  
The paper gives a scaling law that can be used to estimate when the  
spin “will flip with probability at least 1/e”. A calculation of the  
actual loss rate does not seem to be given, yet it seems necessary to  
have it in order to obtain the results presented in figure 4 and to  
give corrections to previous work.

In the revised manuscript, we have more clearly disambiguated between the scaling law calculation, which simply picks the surface where P=1/e and finds its area, and the full calculations leading to Table I and Figure 4. These calculations are now more clearly described on page 2, halfway down in the right column: “This facilitates direct numerical computation of the loss rate (γ) by integrating the molecule flux through the plane for a thermal distribution, weighted by the hopping probability. See Sec. A, Eqn. 2 of our Supplementary [44] for the full integral expression.”

We also bring this up again in discussion of Fig. 4 near the end of Page 3: “We also compute loss rates for all values of pin translation and Bcoil by numerically integrating the loss flux through these unusual loss surfaces via the Landau-Zener formula, just as for the simpler quadrupole geometry discussed previously.”

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Report of Referee B -- LF16145/Reens  
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In LF16145, the authors report a major advance on trapping OH  
molecules. They manage to increase the lifetime of the trapped  
molecules from 5 to 500 ms. The long lifetime should enable future  
evaporative cooling of the molecules.  
  
The key idea here is to employ the Ioffe configuration to remove the  
Majorana spin flip when the magnetic field is vanishing at the trap  
center. A careful analysis of the trap configuration shows that this  
is bad when the external electric field is applied perpendicular to  
the magnetic field. A new trap configuration is presented with a bias  
field to eliminate the field zero, and indeed greatly suppressed trap  
loss is observed.

Yes, but there is a key difference between our configuration and the Ioffe. In Ioffe the bias also needs to provide axial confinement, and in general must be rather large to do so. Even when the smallest bias necessary is employed, the trap is considerably weakened. In our configuration, the bias field does not need to provide any confinement, the electric field does this. So unusually small bias fields are possible- we achieve our two orders of spin-flip loss suppression with a bias field that is only 4% of the trap depth. We now make this much more explicit, see the end of the top right paragraph on page 3, “With only 200 G bias field (the trap is 5 kG deep) the loss is suppressed below that due to background gas.”  
  
If such suppression of loss indeed comes from the removal of spin  
flips, I would very strongly recommend the publication of the paper in  
Physical Review Letters. It would represent a significant advance in  
extending the lifetime of a cold dense OH molecules, where evaporation  
and collisions have been reported. The only issue is that it is  
unclear whether the extended lifetime comes from the suppression of  
spin-flips or the sample in the new trap is just prepared at a much  
lower density. The authors should provide an argument that the long  
lifetime is indeed due to the removal of spin flips.

We thank the reviewer for this positive recommendation of our work. We agree that the removal of this loss is a significant advance, and we can confidently claim that the lifetime increase comes only from spin-flip suppression and not from density effects. In fact, clear disambiguation was a key motivation of our design, and we have decided to make this a point of emphasis in our manuscript, see the end of the first paragraph on the third page, “In light of this, it becomes especially important to perform direct, unconvolved (with collisional effects that is) experimental verification of both the magnitude of the loss effect and the validity of our loss-flux calculations. We now present the new trap where this is achieved.”  
  
Without more data taking, one possible check is to calculate the new  
trap potential similar to Figure 2 (b). I am sure the authors did or  
can do that. Combined with the 175mK measured in Fig. 5, one can  
estimate the density of the sample. As long as it is not too much  
lower than the previous work in Ref. [22], I will be satisfied.

We thank the reviewer again for raising this important point. Were we to plot the trap potential after application of the bias field as in figure 2 (b), the lines would not deviate noticeably from those without the bias field. The fact that the spin-flip loss can be alleviated without noticeable perturbation to the trapping geometry illustrates just how surprisingly magnified the spin-flip loss is relative to the familiar atomic case.

Other minor issues in the order of appearance in the manuscript  
  
1. page 1 intro: Molecules in the degeneracy regime have been reported  
before the KRb experiments, for example, by Deborah Jin on K2, Rudolf  
Grimm on Cs2 and Li2, Wolfgang Ketterle on Na2 and Li2 and so on.  
These works should be cited.

We have gladly incorporated these and some other citations: “Feshbach molecules at the BEC-BCS crossover have been studied [2–4], ground state alkali dimers continue to progress [5–13], and KRb polar molecules have reached quantum degeneracy in an optical lattice [14].”  
  
2. Fig. 1: Please put E=0 in panel (e) and E>0 in panel (f). The  
arrangement of the figures confused me since it suggests (b) and (e)  
are based on E//B; (c) and (f) are based on E \_|\_ B.

We have inserted these titles as suggested, and agree that this was confusing before.  
  
3. page 2: The sentence that "...the molecules must switch from the  
vector sum quantization axis to the vector difference quantization  
axis..." is confusing, since vector difference can be B-E or E-B. The  
latter E-B will still have the problem if E and B are are in the same  
direction. Please clarify the meaning of "vector difference".

With the goal of providing greater clarity, we have made significant changes to this section on an intuitive picture for the loss effect. We believe this section is now free of sign and direction related ambiguities. See the paragraph near the top of page 2 beginning, “We begin with an intuitive picture…”  
  
4. Table I. "experiment length 100 ms" needs clarification. This table  
contains a lot of information not discussed in the text. "Polarizing"  
is not explained, for example. The table is only mentioned briefly  
regarding a less important point, so may possibly go.

Partly due to the important ramifications of our manuscript for our previous publications, we have decided to keep this table. Its contents now feature further discussion in the supplementary material describing the implications of this spin-flip loss on the interpretation of evaporation and e-field induced collision experiments. Regarding the experiment length and the polarizing comment, we have modified the caption to read: “Enhancements (η) and loss rates (γ) for OH with typical applied fields. Zero field values are equivalent to traditional spin-flip loss. Electric field is required during evaporation and spectroscopy to open avoided crossings [29, 38], or applied to polarize the molecules and study collisions [37].”  
  
5. page 3: The sentence "Another possibility is to use one field only,  
but any experiment which aims to make use of the doubly dipolar nature  
of molecules cannot accept this compromise" is not understandable. The  
author should mention the advantage of having the E field trap.  
Otherwise, why can't the field be turned off if it only hurts the  
experiment?

We agree that turning off the field may be the best decision in some cases, but point out that one of the key advantages to working with a dipolar species is the utilization of both fields during the experiment. For the time being, we have actually removed this discussion of other possible geometries, instead focusing on how our geometry allows the loss to be tuned.  
  
6. page 4: "It is seen that increasing B\_coil increases population  
fist at low fields and then higher fields". I believe a word  
"decreases" is missing, right?

Thanks for your observation. This was a bit confusing. We have actually removed this section in order to accommodate greater clarity in our description of the details of molecule enhanced spin-flip loss, pursuant to the comments of our first reviewer.  
  
A final comment, high value of B\_coil is a bad idea for trapping since  
the confinement is greatly weakened than a quadrupole trap. Bare  
minimum Ioffe field just enough to remove spin flips is preferred to  
maintain a high molecular density at the center. In addition to weak  
confinement, strong B\_coil also gives a shallower trap, which is  
likely the origin of the fast initial loss seen in Fig. 4a. This  
picture is consistent with the observed absence of density-dependence.  
The initial loss in this case would be plain evaporation for hot  
molecules to escape.

We agree with these statements as far as the Ioffe-Pritchard trap lift is concerned, but this is not an issue in our geometry for the reasons discussed above. We reiterate our gratitude for the reviewer’s emphasis of this important distinction between density and single-particle effects. We believe these comments have enabled us to prepare a stronger manuscript.