­­­­Dear Editor:

We are grateful for having received positive comments and constructive criticisms on our manuscript from two very knowledgeable reviewers. We have carefully considered all of their comments and addressed them by implementing significant changes to the manuscript. We have revised the emphasis and presentation of the paper to provide a more focused motivation, a more complete discussion of the scientific context, and a more simplified physical picture for understanding. We have also included a thorough discussion of the impact of this work to previous results in a new Supplementary Material section. Below is a detailed list of our responses and revisions covering every comment from both reviewers. With this thoroughly revised manuscript we hope that Reviewer A may now join Reviewer B for recommending the publication of this work in PRL.

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.*”

Our Response: We agree with this synopsis as it pertains to our original manuscript, but we have rearranged our emphasis in the new manuscript, motivated by the reviewer’s comments. Please 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*.”

Our Response: We appreciate the reviewer’s opinion that we have not presented new insights in a convincing manner in the original manuscript. To correct this, we have thoroughly revamped our discussion of the mechanism and made this the primary focus of the revised manuscript. We believe that we now successfully present 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 providing a clear and intuitive physical 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 presented a much more explicitly calculation of this scaling law in Sec. C of the new 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)].*”

Our response: We thank the reviewer 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 this new study. 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 that we now highlight and clarify in the paper. 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." The two new references we cite here, [41,42], are the two papers pointed out by this reviewer.

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*.”

Our response: 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 considering the importance of magnetic trapping of cold molecules, it is timely to provide a more complete and rigorous treatment of this effect, supported for the first time by an experiment designed specifically to clarify this point, not as a side effect. We elaborate on these points further.

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.”

We believe that these new, improved understandings are important for us to share with the wide scientific community. We are no longer presenting a side point on the spin-flip loss that could be deconvolved, but an important effect that must be completely addressed by anyone working on trapping cold molecules in similar regimes.

“*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*:”

Our response: We respectfully disagree with the reviewer on this point, although we thank the reviewer for challenging us to make a stronger case. The work on this new type of trap goes beyond a simple demonstration of trapping molecules, and it provides a clear physical picture of the behavior of molecules in combined electro-magnetic fields in the context of trapping. We have specifically designed this trap not necessarily as our next generation trap but as an important testbed for spin-flip losses.

The reviewer appropriately notes that the work “will certainly be of interest to specialists in the field of cold molecules.” We concur and we wish to emphasize it reaches further. 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 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?*”

Our response: We agree with the reviewer and have now provided much more details about the connection between this work and previous studies, both in the main text and in the new Supplementary Materials. Indeed it is our desire for more complete and rigorous investigations that has motivated the new research presented in this manuscript. The current work is strongly connected with that of Ref. [29]. We refer the reviewer to the detailed discussions presented in Sec. A of the Supplementary Materials. The summary is that the effects previously attributed to inelastic collision loss are mostly accounted for by the spin flip loss. But there are remaining differences that will need to be explored in a separate experiment that more clearly separates the collisional and the spin-flip loss effects. We are constructing a new apparatus to address this.

“*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.*”

Our response: We have now added a complete description of our current best understanding in Sec. B of the Supplementary Materials, including new data that were taken after the publication of Ref. [22] to reproduce the previous experimental results. Essentially, the new interpretation does weaken the evaporative cooling result, especially by undermining the hypothesis of cold samples building up in the low magnetic field region 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 shows such enhancements for evaporation sequences designed to achieve a twofold temperature reduction.” We have also provided a paragraph on the 10-fold enhancement of phase space density under this experimental condition.

Furthermore, we have added descriptions of independent experiments on the comparison of forward and backward evaporation, as well as evaporation results under different initial temperature conditions, in the Supplementary Material. The direct experimental evidence for evaporation stands.   
  
“*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}?*”

Our response: Yes, this is exactly what is meant by H\_{E perp B}, we are grateful for the reviewer’s question for clarity. We now 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.*”

Our response: Perhaps the reviewer 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 picture is in fact 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 effect. The spin-rotational coupling is in general quite large, and usually more significant than hyperfine effects.

To remove any remaining confusions, we have reworked the section, so hopefully our conclusions come across more clearly. Please 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.*”

Our response: We agree with reviewer that this description was written too densely. To address this, we have moved the scaling law derivation to Sec. C of the new Supplementary Materials, and we have 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. With this expanded discussion and new material, we believe the explanation of the scaling law is now much easier to follow.

“*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.*”

Our response: 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 ΔM=3. This discussion was actually presented in an earlier publication by Stuhl *et al*., (B. K. Stuhl, M. Yeo, B. C. Sawyer, M. T. Hummon, and J. Ye, Physical Review A 85, 033427 (2012)), which we cite in this paper. This effective 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 Schrödinger 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?*”

Our response: 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.*”

Our response: 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.*”

Our response: In the revised manuscript, we have more clearly disambiguated between the scaling law calculation, where we simply choose the surface where P=1/e and determine its area, and the full numerical 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. Sec. A, Eqn. 2 of our Supplementary [44] includes the full integral expression. We perform these integrations for OH over the velocity distribution in a 2 T/cm magnetic quadrupole… .”

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.*”

Our response: We thank the reviewer for providing this useful analogy. The connection is clear, but there is also a key difference between our trap and the Ioffe configuration. 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, and thus it becomes challenging to confine molecules that are relatively hot compared with typical cold atoms. In our trap, the bias field does not need to provide any confinement, the electric field does this. So unusually small bias fields are possible- we achieve a two-order-of-magnitude loss suppression with a bias field that is only 4% of the trap depth. We now make this point 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*.”

Our response: We thank the reviewer for the strong support of our work. We also appreciate the comment for further clarification, which fits well with our revised focus of the paper that emphasizes the experiment-theory agreement on the spin flip loss removal. We agree with the reviewer that the removal of this loss is a significant advance, and we conclude based on experimental evidence 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*.”

Our response: We thank the reviewer again for raising this important point. We have indeed done such a calculation. If we plot the trap potential after application of the bias field as in Fig. 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, and the efficacy of our new approach for suppressing it.

“*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.*”

Our response: 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*.”

Our response: 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"*.”

Our response: With the goal of providing greater clarity, we have basically rewritten this section based on an intuitive picture for the loss effect. We believe this section is now free of sign and direction related ambiguities. Please see the paragraph near the top of page 2 beginning, “We begin with an intuitive picture…” We hope that it will be much easier to follow.

“*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*.”

Our response: We thank the reviewer for reminding us to provide more complete information on the table. 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].” Partly due to the important connections between our current study and previous publications, we have decided to keep this table. Its contents now feature further discussions in the Supplementary Material describing the impact of this spin-flip loss on the interpretation of evaporation and E-field induced inelastic collision experiments.

“*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?*”

Our response: We agree that turning off the field may be the best decision in some cases, but we wish to point out that one of the key advantages to working with dipolar molecules 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 first at low fields and then higher fields". I believe a word "decreases" is missing, right?*”

Our response: Thanks for the careful observation by this reviewer. To provide a more clear presentation of our work, 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 the 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*.”

Our response: 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.