

# Molecular Spin-Flip Loss and a Dual Quadrupole Trap

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Doubly polar molecules exhibit complex internal spin-dynamics in mixed electric and magnetic fields. Near magnetic trap minima, these spin-dynamics lead to enhancements in Majorana spin-flip transitions by many orders of magnitude relative to atoms, and are thus an important obstacle for progress in molecule trapping and cooling. The effect is strongest for Hund's case (a) states but is quite significant for Hund's case (b) as well. We study these internal spin-dynamics with OH molecules and derive a trap geometry where spin-flip loss can be tuned from over  $200 \text{ s}^{-1}$  to complete removal with only a weak external bias cell and with no sacrifice of trap strength.

The ultracold regime extends toward molecules on many fronts [1]. KRb molecules have reached lattice quantum degeneracy [2] and other bi-alkalis continue to progress [3, 4]. Creative and carefully engineered laser cooling strategies are tackling certain nearly vibrationally diagonal molecules [5–9]. A diverse array of alternative strategies have succeeded to greater or lesser extents on other molecules [10–15]. All of these molecules will require secondary strategies like evaporation or sympathetic cooling to make further gains in phase space density. They also may face a familiar challenge: spin flip loss near the zero of a magnetic trap, but dramatically enhanced for many doubly dipolar molecules due to their internal spin dynamics in mixed electric and magnetic fields.

The knowledge of spin flips or Majorana hops as an eventual trap lifetime limit predates the very first magnetic trapping of neutrals [16]. Spin flips were directly observed near  $10 \mu\text{K}$  and overcome in the TOP trap [17], and soon later with a plugged dipole trap [18], ~~producing the first~~ Bose-Einstein condensates. In our earlier investigations, we observed OH trap loss with applied electric field [19]. This trap loss occurred for sub-states of OH's  $\Lambda^2\Pi_{3/2}$  ground state manifold other than the most well-trapped one (Fig. 1, panel (d)), and was attributable to avoided crossings that open at non-zero magnetic fields between levels of opposite parity. We have now identified trap loss at zero magnetic field with the application of electric field that affects even the most well-trapped substrate although it lacks any such crossings. We first suspected this during experiments in our previous trap geometry, a 3D permanent magnet quadrupole trap with homogeneous electric field [21], but it was difficult to disentangle it from competing effects. Our new trap geometry addresses this, but we use our previous geometry as our starting point to explain the internal spin-dynamics that lead to the enhanced loss.

These internal spin-dynamics are subtle; it has taken a concentrated several years to elucidate the effect with conclusive experimental evidence as reported here. The effect has also eluded three previous investigations of note: In [22] the analogues of atomic spin-flip loss for

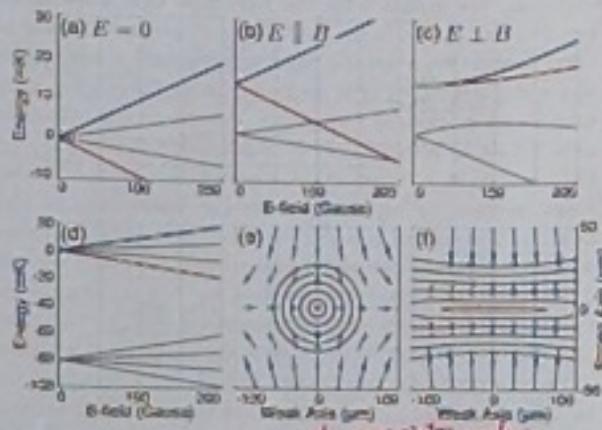


FIG. 1. (a) Four Zeeman split lines in OH's  $J = 3/2$  ground manifold, with the well-trapped state in blue and its spin-flip partner in red. (b) Again with  $E = 150 \text{ V/cm}$ , and  $E \parallel B$ ; (c) same with  $E \perp B$ . Note vastly reduced red-blue splitting. Other angles in appendix of [21]. (d) The opposite parity manifold of electrically strong field seeking substrates is  $\sim 80 \text{ mK}$  below, this is the lambda doublet splitting  $\Delta$ . (e) Energy splitting contours every 2 mK near the zero of a magnetic quadrupole trap with  $2 \text{ T/cm}$  gradient [19]. B-field vectors in blue. (f) Again with  $E = 150 \text{ V/cm}$ . Note dramatic widening of lowest contour (red). Vector direction gives the effective quantization axis of the trapped state,  $p_0 \theta \pm dE$  above (below) the horizontal centerline. Vector magnitude gives potential energy relative to trap center.

predicted (or stated in theory)

molecules in mixed fields were studied, and a magnetic quadrupole trap for OH molecules with superposed electric field was specifically addressed. It was concluded that no significant loss enhancement due to electric field would be evident. While this is true for the approximate  $^2\Pi_{1/2}$  Hamiltonian used in that study, it is not true for the actual  $^2\Pi_{3/2}$  ground state of OH. In [21] E-fields were applied in our magnetic quadrupole trap to study E-field induced collisions. Although an initial estimate of the effect was described, it was not made

estimate  
made

In this case

a factor of  $\pi$  underestimate of the neglect on-pole made decreasing of the loss mechanism and collision losses harder to separate.



the molecule align with one of two quantization axes: either the vector sum or the vector difference of the dipole moment weighted electric and magnetic fields. It was noted that this would maintain quantization near the zero of a quadrupole trap and avoid spin-flip loss. As we will explain, quantization is indeed maintained, but spin-flip loss is enhanced.

With only magnetic field, a molecule remains trapped insofar as it adiabatically follows the field direction. Near the trap center, the direction changes most rapidly, resulting in loss. When electric field is added, it dominates in the trap center where the magnetic field is weakest. Quantization is maintained but the quantization axis does not rotate with the magnetic field. Further away from the trap center the molecule is then magnetically strong field seeking and is lost. The molecule ought to have switched from the vector sum quantization axis to the vector difference quantization axis, so as to remain doubly weak field seeking despite the change in relative orientation of the fields.

But we don't need to have a weak field seeking site for  $E$ , since  $E$  can be applied in a uniform way.

It's all about having the partner axis fixed to the  $E$  direction, & thus  $E \parallel B$  represents a spin flip mostly.   
 define geometry than

This observation allows us to develop a scaling law for the loss enhancement in this geometry. For a given trap strength and sample temperature, there is a characteristic energy splitting  $\Delta$  below which spin-flips can occur calculated from the Landau-Zener formula. In our case  $\kappa = 5$  MHz. As shown in panel (c) of Fig. 1, E-field widens the  $\Delta$  valued energy contours near the trap zero,

in specific directions

We need to motivate the origin for this number (5 MHz): temp? gradient? etc.

TABLE I. Enhancements and loss rates for OH. Evaporation E-field detailed in [25]. Spectroscopic E-field is [19]. Background loss is  $2 s^{-1}$ , experiment length 100 ms.

$E$ (V/cm)	45 mK		5 mK		Purpose
	$v$	$\Gamma (s^{-1})$	$v$	$\Gamma (s^{-1})$	
0	1	0.02	1	1.3	No Field
300	5	0.1	9	11	Evaporation
550	17	0.3	40	50	Spectroscopy
3000	1000	10	1600	2000	Polarizing

greatly increasing the flux through this region. Note also that the energy gradient near the loss region, which also contributes to the Landau-Zener hopping probability, remains nearly identical in the  $x$ -direction between panels (a) and (f). Solving for  $B$  when  $H_{ELD}(B) = \kappa$  and dividing by the  $E = 0$  case gives the flux enhancement factor  $v = (d_E E / \sqrt{\kappa \Delta})^{1/3}$ . So E-fields beyond  $\sqrt{\kappa \Delta} / d_E$  lead to almost cubic enhancements in spin-flip loss. We can be more quantitative by integrating the velocity distribution and the flux through the plates, accounting for the velocity dependent Landau-Zener probability, Table I. The spin-flip loss is negligible at 50 mK, but relevant at the 5 mK targeted during evaporation [25]. Those values will thus require reinterpretation considering this effect [26]. With the goal of  $\mu$ K temperatures and below, it is clear spin flip loss must be addressed.

For Hund's case (a) states more generally, the Zeeman splitting when  $E \perp B$  is reduced from linear to order  $1/J$ , according to several test Hamiltonians we have diagonalized. This also agrees with the intuition that when  $\mu_B B < d_E E$  and  $E \perp B$ , the magnetic field must undo the electric field's coupling of opposite  $m_J$  number states, a test of order  $m_J - (-m_J) = 2J$ . Only  $J = 1/2$  states remain linear, but their vanishing g-factor renders them untrappable already. For Hund's case (b) the enhanced loss region is restricted to the trap energy regime where the spin-rotation coupling dominates. In this regime the state is effectively Hund's case (a). This can still be very significant, for example  $\gamma = 75$  MHz for SrF [27]. In preliminary investigations for Hund's case (b) molecules, which essentially consist of reproducing panels (a)-(c) of Fig. 1 for different Hamiltonians, we find large spin-flip loss enhancements for SrF's  $v=0, N=1$  magnetically trappable substates. Some Hund's case (b) molecular states such as YO's  $v=0, N=1$  manifold have a protected substate with  $m_F = 0$  and thus no hopping partner in the spin-rotation coupling regime that is nonetheless energetically separated from other state-crossings by the Lamb shift. This state is less strongly trappable due to the same  $m_F = 0$  feature, but is fully spin-flip immune.

We can generalize to arbitrary geometries and consider methods to avoid the loss using a simple strategy: avoid  $\mu_B B < d_E E$  where  $E \perp B$ . One way to achieve this is to trap with E-field and superpose B-field. The lambda

atom-like loss figures

new understanding  
mostly be worked  
out & done it  
50K,

$J$  is not defined yet here  
and  $2J$  is not an energy unit.

Caf?

YO  
ref

What does electric field coupling of opposite my come from?

It's hard to understand for general reader.

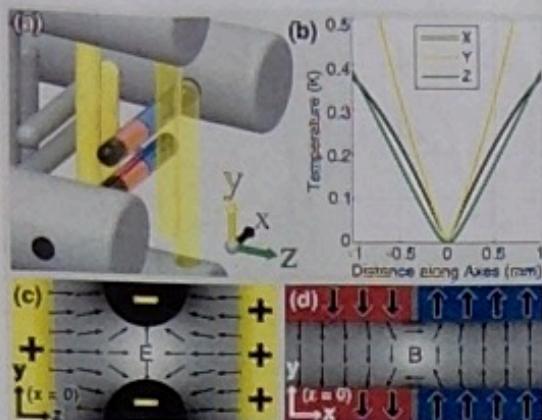


FIG. 2. (a) The trap consists of the last 6 pins of a Stark decelerator. The trap center is directly between the second to last pin pair. These pins have two magnetized domains each. The blue domains are magnetized along  $+y$ , the red along  $-y$ . These pins are grounded, while those in yellow are positively charged. OH is decelerated as in [20], except the slowing reaches almost zero velocity at the trap center. (b) Trap energy along axes.  $B' = 5 \text{ T/cm}$  and  $E' = 100 \text{ kV/cm}^2$ . Trap frequencies  $\nu_x = 3 \text{ kHz}$ ,  $\nu_y = 5 \text{ kHz}$ , and  $\nu_z = 4 \text{ kHz}$ . Pin-pairs are spaced 2 mm, which sets the maximum trap width in the  $y$  direction. (c) The electric 2D quadrupole in the  $x = 0$  plane. The magnetic pins intersect this plane and are shown as black circles. The other pins don't actually intersect this plane, but are projected onto it as yellow rectangles. (d) The magnetic 2D quadrupole in the  $z = 0$  plane.

~~the trap~~ significantly weakens doublet prevents flips in this configuration, but it does round the trap minimum considerably. Another option is to trap with both fields and keep zeros overlapped. This was once realized for OH with a superposed magnetic quadrupole and electric hexapole [28]. Such a scheme prevents spin-flip loss enhancement, but does not remove it entirely. It is also susceptible to misalignment induced loss enhancement. Another possibility is to use one field only. While this avoids spin-flip loss enhancement, any experiment which aims to make use of the doubly dipolar nature of molecules cannot accept this compromise.

We opt for a geometry that is distinct from these options: a pair of 2D quadrupole traps, one magnetic and the other electric, with orthogonal centerlines. We achieve these fields in a geometry that matches our Stark decelerator [12], as shown in Fig. 2. This approach is similar to the Ioffe-Pritchard strategy [29], where a 2D magnetic quadrupole is combined with an axial dipole trap. Axial and radial trapping interfere, resulting in significantly lower trap depths than the 3D quadrupole. We thwart this interference by using electric field for the third direction. Our geometry has  $E \perp B$  along both the  $xz$  and  $yz$  planes, with  $\mu_B B < d_E E$  in a large cylinder surrounding the  $z$ -axis. However, by adding magnetic

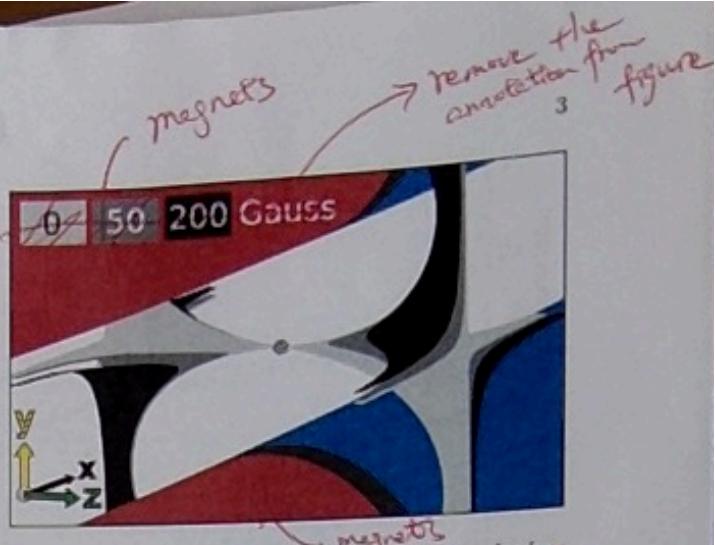
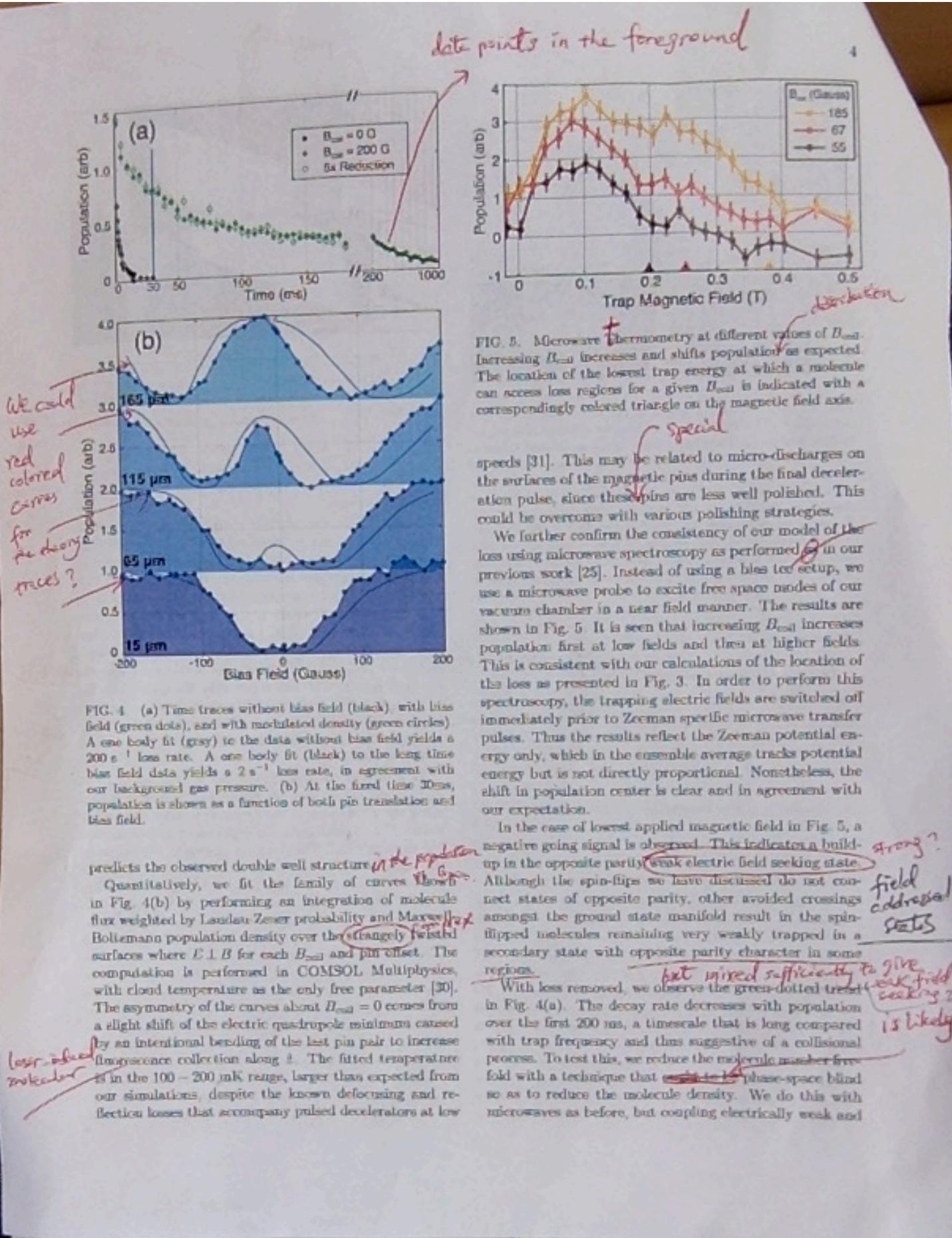


FIG. 3. Surfaces where spin-flips can occur for several values of  $B_{\text{cell}}$ . These surfaces are subsets of those where  $E \perp B$ , shown only where  $\mu_B B$  is small enough relative to  $d_E E$  to allow spin-flips.

The three diffuse shades of surface  
grey,  $B_{\text{cell}} = 0$ , dark gray,  $B_{\text{cell}} = 50$ , black  
 $B_{\text{cell}} = 200$  Gauss

Adding  $B_{\text{cell}}$  only slightly rounds the magnetic trapping potential, but it morphs the  $E \perp B$  surface from a pair of planes into a hyperbolic sheet, pushing it away from the  $z$ -axis where the magnetic field is smallest. This means that small magnitudes of  $B_{\text{cell}}$  are sufficient to avoid  $\mu_B B < d_E E$  where  $E \perp B$ . In Fig. 3, the surfaces where  $E \perp B$  for several  $B_{\text{cell}}$  magnitudes are shown whenever the splitting ~~there~~ is below the hopping threshold. Note how  $B_{\text{cell}}$  tunes the proximity of the loss regions ~~from?~~ the trap center. The loss regions are always visible, but they are tuned so far away that molecules accessing them have already escaped the trap. The striking difference in molecule trap lifetime with and without  $B_{\text{cell}}$  can be seen in Fig. 4, panel (a).

As a further confirmation of our  $E \perp B$  and  $\mu_B B < d_E E$  model of the loss, we translate our magnetic pins along the  $\hat{x}$  direction in their mounts to alter the surface where  $E \perp B$ , and compare experimental data against our expectations. The data are shown in Fig. 4, panel (b). Qualitatively, this translation serves to disrupt the otherwise perfectly 2D magnetic quadrupole by adding a small trapping field  $\vec{B} \propto B' \hat{z}$  along the  $z$ -axis. This means that  $B_{\text{cell}}$  no longer directly tunes the magnetic field magnitude along the  $z$ -axis. Instead,  $B_{\text{cell}}$  must first overcome the slight trapping field along the  $z$ -axis, translating a point of zero field along the  $z$  axis and eventually out of the trap. The point of zero magnetic field has  $\mu_B B < d_E E$  and lies on the  $\phi = \vec{E} \cdot \vec{B} = 0$  surface by definition, leading to strong loss unless it is aligned with the trap center, where  $E$  is also zero. This means that without any bias field, the loss should actually be a local minimum; as the field is increased in either direction the loss should first worsen and then improve when the zero leaves the trap. This qualitative explanation correctly



strong field seekers during the first half of deceleration. Spatial inhomogeneities on the order of the cloud size are unlikely given the 15 cm microwave wavelength, but any that exist are remixed during the remaining deceleration. The trend remains similar with this density-modulation, suggesting that single-particle physics is chiefly responsible. This discrepancy relative to our previous work is attributable to our warmer initial temperature. We hope soon to implement several density increasing measures and return to the collisional regime. The slowly varying ~~collisional~~ decay rate could be related to the existence of high energy chaotic orbits with long escape times, as seen in other exotically shaped trapping potentials [32].

Our dual quadrupole trap decisively overcomes molecule enhanced spin-flip loss by tuning it from an overwhelming rate to complete removal. Our explanation of the loss provides detailed predictions of how its location and magnitude ought to scale with bias field and trap alignment, which we have experimentally verified. Our results correct existing predictions about molecular spin-flips in mixed fields and we provide a consistent framework that explains this based on internal spin-dynamics. We have devised a viable trapping geometry in which spin-flip loss is fully mitigated without trap-depth sacrifice, paving the way toward further improvements in molecule trapping and cooling.

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D.R. and H.W. contributed equally. D.R. ~~is writing~~  
and trap design. H.W. ~~is experiment execution~~

For PRL We don't list individual author contributions.

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dark matter