

Molecular Spin-Flip Loss and a Dual Quadrupole Trap

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Doubly dipolar molecules exhibit complex internal spin-dynamics when electric and magnetic fields are both applied. 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. We conclusively demonstrate and address this with OH molecules in a trap geometry where spin-flip losses can be tuned from over 200 s^{-1} to below our 2 s^{-1} vacuum limited loss rate with only a simple external bias coil and with minimal impact on trap depth and gradient.

The ultracold regime extends toward molecules on many fronts [1]. KRb molecules have reached lattice quantum degeneracy [2] and other bialkalis continue to progress [3–6]. Creative and carefully engineered laser cooling strategies are tackling certain nearly vibrationally diagonal molecules [7–13]. A diverse array of alternative strategies have succeeded to greater or lesser extents on other molecules [14–20]. All of these molecules will require secondary strategies like evaporation or sympathetic cooling to make further gains in phase space density [21–23]. 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 [24]. Spin flips were directly observed near $50 \mu\text{K}$ and overcome with a time-orbiting potential trap [25] and a plugged dipole trap [26], famously enabling the first production of Bose-Einstein condensates. Motivated by the interest in dipolar molecules in mixed fields for quantum chemistry, precision measurement and many-body physics, we previously investigated loss of magnetically trapped hydroxyl radicals (OH) with applied electric field [27]. This trap loss occurred for sub-states of OH's $X^2\Pi_{J=3/2}$ ground state manifold other than the most well trapped one (positive parity and full spin polarization, $|f, m_J=3/2\rangle$, blue in Fig. 1). Due to the closely spaced parity doublet, a general feature of Hund's case (a), these states intersect opposite parity states at non-zero B-fields, where E-field can open avoided crossings and cause trap loss. We now identify internal spin-dynamics leading to trap loss near $B=0$ even for the most well trapped state and even at 50 mK .

These internal spin-dynamics are subtle, having eluded three previous investigations of note: In [28] the analogues of atomic spin-flip loss for molecules in mixed fields were modeled, and a magnetic quadrupole trap for OH molecules with superposed E-field was specifically addressed. It was concluded that no significant loss enhancement due to electric field would be evident.

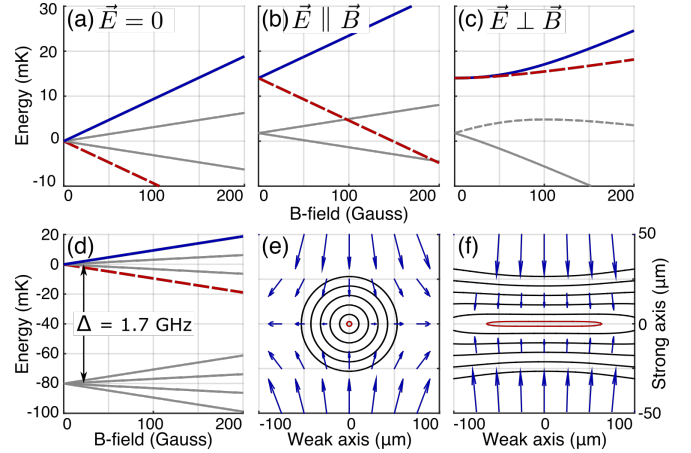


FIG. 1. A uniform E-field, added to magnetically trapped molecules for dipolar studies or other purposes, can lead to spin-flip losses. Four Zeeman split lines in OH's $X^2\Pi_{3/2}$ manifold are shown (a-c), with the trapped $|f, 3/2\rangle$ state in blue and its spin-flip partner $|f, -3/2\rangle$ in dashed red. These states are shown with no E-field (a), with $E = 150 \text{ V/cm}$ and $E \parallel B$ (b), and with $E \perp B$ (c). Note the vastly reduced red-blue splitting in the latter case. The gray close dashes (c) denote the $|f, 1/2\rangle$ state, which is briefly discussed in the main text. The opposite parity ($|e\rangle$) manifold is split by Δ (d). Energy splitting contours are shown every 40 MHz near the zero of a 2 T/cm magnetic quadrupole trap for OH molecules [27] without E-field (e), and with uniform $E = 150 \text{ V/cm}$ along the strong axis of the quadrupole (f). The vectors are $\mu_{\text{eff}}\vec{B} \pm d_{\text{eff}}\vec{E}$, where the sign is positive above the horizontal centerline and negative below, which represents the proper quantization axis for magnetically trapped molecules. Note the drastic widening of the lowest contour (red), the culprit for molecular spin-flip loss enhancement.

This is true only for the approximate $^2\Pi_{1/2}$ Hamiltonian used in that study. In [29] E-fields were applied in our magnetic quadrupole trap to study E-field induced collisions. Although an initial approximation was made of the spin-dynamical effect, subsequent investigations have revealed it to be a threefold underestimate, enough to render deconvolution of any remaining collisional effect difficult. The specific implications for these previous

results are discussed further in supplementary materials. Finally, in [30] it was correctly noted that Hund's case (a) molecules maintain a quantization axis in mixed fields. The states of the molecule were shown to align with one of the two quantization axes given by the vectors $\mu_{\text{eff}}\vec{B} \pm d_{\text{eff}}\vec{E}$, μ_{eff} and d_{eff} the effective dipole moments of the molecule in uncombined fields. It was asserted that this would maintain quantization near the zero of a quadrupole trap and avoid spin-flip loss, but as we now describe the loss is actually enhanced.

We begin with an intuitive explanation of the loss enhancement based on molecular orientation. Consider a magnetic quadrupole trap, where a weak-field seeking molecule remains trapped insofar as it adiabatically follows the field direction. When molecules pass near the trap center, they experience a rotating field direction, causing spin-flips. When electric field is added, it dominates in the trap center where the magnetic field is weakest. As correctly noted in [30], this causes a quantization axis to be maintained, either of $\mu_{\text{eff}}\vec{B} \pm d_{\text{eff}}\vec{E}$. Now suppose a molecule initially follows the sum quantization axis and is in a region where E and B are nearly parallel before passing by the trap center. After passing the trap center, B rotates nearly 180° , so that E and B are nearly anti-parallel. Now the length of the sum quantization axis, which is proportional to the field induced energy shift of the molecule, actually decreases with increasing $|B|$. Further away from the trap center the molecule is then magnetically strong field seeking and is lost. To be more general, we define the relative orientation of the fields as the sign of $\phi = \vec{E} \cdot \vec{B}$. When ϕ is negative, the trapped state must have the vector difference quantization axis, and vice versa, so that an increase in magnitude of the magnetic field increases its potential energy. Whenever ϕ changes sign, i.e. where $\phi = 0$ and $\vec{E} \perp \vec{B}$, there will be a chance of spin-flip associated with the molecule's propensity to maintain its quantization axis rather than remain in the well-trapped state. Since ϕ is a continuous scalar defined in 3D, $\phi = 0$ is one of its contour levels and thus always corresponds to a 2D surface; in the case of a magnetic quadrupole with homogeneous E it is a plane.

This intuition is in agreement with a more rigorous analysis of the energy splittings between the trapped state and its spin-flip partner. By exactly diagonalizing the approximate eight state ground molecular Hamiltonian for OH, subtracting the relevant state energies and Taylor expanding, we find:

$$H(\mathcal{B}_\perp, \mathcal{B}_\parallel, \mathcal{E}) = \mathcal{B}_\parallel + \mathcal{B}_\perp^3 \frac{\Delta^2}{\mathcal{E}^4} + \mathcal{O}(\mathcal{B}_\parallel^2, \mathcal{B}_\perp^4) \quad (1)$$

Here $\mathcal{B} = \mu_{\text{eff}}\vec{B}$ and $\mathcal{E} = d_{\text{eff}}\vec{E}$, the magnetic field is considered in parallel and perpendicular components relative to the electric field, and Δ is the lambda doubling. The relevant splitting between spin flip partner states reaches a deep minimum whenever B_\parallel goes to zero and $\vec{E} \perp \vec{B}$,

TABLE I. Enhancements (η) and loss rates (Γ) for OH with typical applied fields. Zero field values are equivalent to atomic spin-flip loss. E-field is required during evaporation and spectroscopy to open avoided crossings for $|e\rangle$ parity states [22, 27], or applied for polarization of the molecules to study collisions [29]. Background loss is 2 s^{-1} , experiment length 100 ms.

E (V/cm)	55 mK		5 mK		Purpose
	η	$\Gamma (\text{s}^{-1})$	η	$\Gamma (\text{s}^{-1})$	
0	1	0.02	1	1.3	Zero Field
300	5	0.1	9	11	Evaporation
550	17	0.3	40	50	Spectroscopy
3000	1000	19	1600	2000	Polarizing

where the remaining Zeeman splitting is reduced from linear to cubic in magnetic field (Fig. 1a-c). This reduction in the Zeeman splitting from linear to cubic is in fact a well known phenomenon in the precision measurement community[31, 32], and experimentalists have exploited it for the suppression of unwanted influence from magnetic fields in measurements of the electron electric dipole moment. However, in the case of applying mixed fields during trapping, this suppression is not beneficial but rather detrimental, and the reduced splitting creates a broad plane in which spin-flips can occur (Fig. 1e-f). The extent of the loss enhancement is best addressed as discussed below, but a reasonable scaling law can be formed by comparing the surface area of energy contours such as those shown in red in Fig. 1e-f. For κ the energy scale of hops without any application of electric field, electric field enhances the loss rate by $\eta = (d_{\text{eff}}E/\sqrt{\kappa}\Delta)^{8/3}$.

We can better quantify the extent of the expansion by considering molecular trajectories in light of the Landau Zener formula:

$$P_{\text{hop}} = e^{-\delta^2/\hbar\dot{H}} \quad (2)$$

Which relates the probability of diabatically hopping between two states P_{hop} to the energetic coupling between the states δ and the rate of approach of the energy levels \dot{H} . Any molecules whose trajectory passes through the $\vec{E} \perp \vec{B}$ surface experience a minimum energy splitting δ at the point of intersection with the plane. To a good approximation, the hopping probability for a molecule can be computed knowing only its crossing point and orthogonal velocity, and the Landau-Zener loss flux through the plane can be numerically integrated. This loss-flux integration is an important improvement relative to our previous work [29], as discussed further in our supplementary. We perform such integrations for OH in our previous 2 T/cm magnetic quadrupole [33] under various electric fields of interest (Tab. I).

Unfortunately, experimental verification of this model and these loss-flux calculations is difficult in a magnetic quadrupole trap with superimposed electric field because

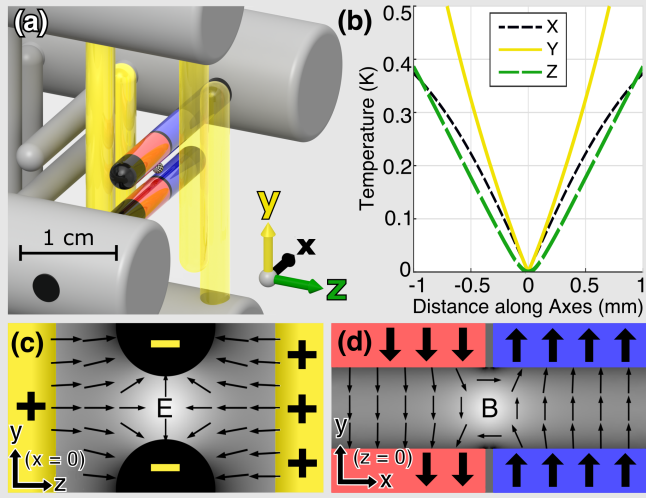


FIG. 2. The last six pins of our Stark decelerator [33] form the trap (a), which is 0.45 K deep with trap frequency $\nu \approx 4$ kHz (b). Along y the trap is bounded by the 2 mm pin spacing. The yellow pins are positively charged and the central pin pair negatively, which forms a 2D electric quadrupole trap with zero along the x -axis. This is shown for the $x=0$ plane (c), with yellow pins artificially projected for clarity since they don't actually intersect the plane. The central pins are magnetized, with two domains each. Blue indicates magnetization along $+\hat{y}$, red along $-\hat{y}$. These domains produce a magnetic quadrupole trap with zero along the z -axis, shown in the $z=0$ plane (d).

of an important competing effect: the enhanced field dependent collision cross-section. Furthermore, our prior results on the collisional effect [29] might be largely explained by the enhanced spin-flip loss, which makes experimental verification especially important. Alarmed by this possibility, we developed a new trap where the enhanced molecular spin-flip loss can be conclusively experimentally demonstrated without any convolved collisional effects, and where our loss-flux calculations could be clearly tested.

Our idea is to use a pair of 2D quadrupole traps, one magnetic and the other electric, with orthogonal centerlines (Fig. 2):

$$\vec{B} = B'x\hat{y} - B'y\hat{x} \quad \vec{E} = E'y\hat{y} - E'z\hat{z} \quad (3)$$

We achieve these fields in a geometry that matches our Stark decelerator [16]. This geometry has $\vec{E} \perp \vec{B}$ in both the $x=0$ and $y=0$ planes, and $\mu_{\text{eff}}B < d_{\text{eff}}E$ in a large cylinder surrounding the z -axis— a perfect recipe for large spin-flip losses. However, by adding a small magnetic field $\vec{B} = B_{\text{coil}}\hat{z}$ along the centerline of the magnetic quadrupole with an external bias coil, a dramatic change can be made to the surfaces where $\vec{E} \perp \vec{B}$ with only a tiny change to the trapping potential.

B_{coil} morphs the $\vec{E} \perp \vec{B}$ surface from a pair of planes into a hyperbolic sheet, pushing it away from the z -axis

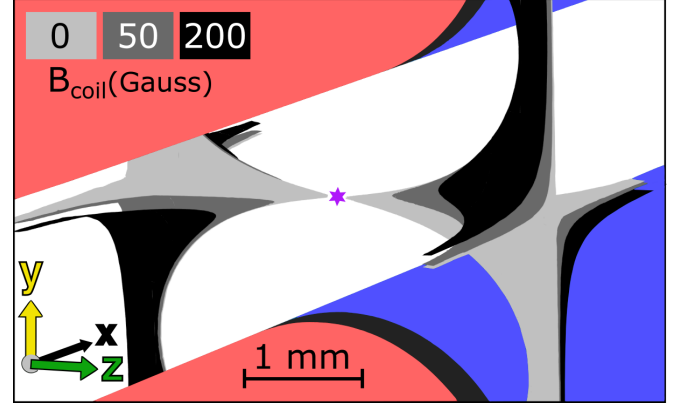


FIG. 3. Surfaces where spin-flips can occur ($\vec{E} \perp \vec{B}$, $\mu_{\text{eff}}B < d_{\text{eff}}E$) are shown for three values of B_{coil} in light gray, dark gray, and black. The magnetic pins are shown as in Fig. 2 for context. The purple star marks the trap center, to which molecules are confined within a ~ 1 mm diameter.

where the magnetic field is smallest:

$$\vec{E} \cdot \vec{B} = B'E'xy - B_{\text{coil}}E'z = 0 \Rightarrow x \cdot y = z \cdot B_{\text{coil}}/B' \quad (4)$$

In Fig. 3, the surfaces where $\vec{E} \perp \vec{B}$ for several B_{coil} magnitudes are calculated wherever the splitting there is small enough that a molecule of thermally average velocity orthogonal to the surface would have a $1/e$ or greater hopping probability. The loss regions ought to be tuned far enough from the trap center that molecules cannot access them. This is indeed what we observe, note the striking difference in trap lifetimes in Fig. 4a. With only 200 G bias field (the trap is 5 kG deep) the loss is suppressed below that due to background gas.

In order to stringently verify our calculations of loss by integration of molecule fluxes across $\vec{E} \perp \vec{B}$ surfaces, we perform these calculations for a diverse collection of loss surfaces obtained by translation of the magnetic pins in their mounts. This translation serves to disrupt the idealized 2D magnetic quadrupole by adding a small trapping field $\vec{B} \propto B'z\hat{z}$, which dramatically alters the $\vec{E} \perp \vec{B}$ surfaces and the magnitude of the fields and the hopping probability across them. We perform this same translation in situ, and obtain a striking agreement (Fig. 4b). This is particularly remarkable given that this direct integral calculation assumes a purely thermal distribution and doesn't involve the computation of any actual trajectories. The fits are performed using only temperature as a free parameter, which enters the calculation via the thermal distribution used for integration, and fits to 170 ± 20 mK. [34]. An intuitive explanation for the intriguing double well structure in population versus B_{coil} is that B_{coil} first translates the magnetic zero along the z -axis, overlapping it with larger electric fields at first before moving it out of the trap.

With strong experimental confirmation of the molecular spin-flip loss enhancement, we can move on to gen-

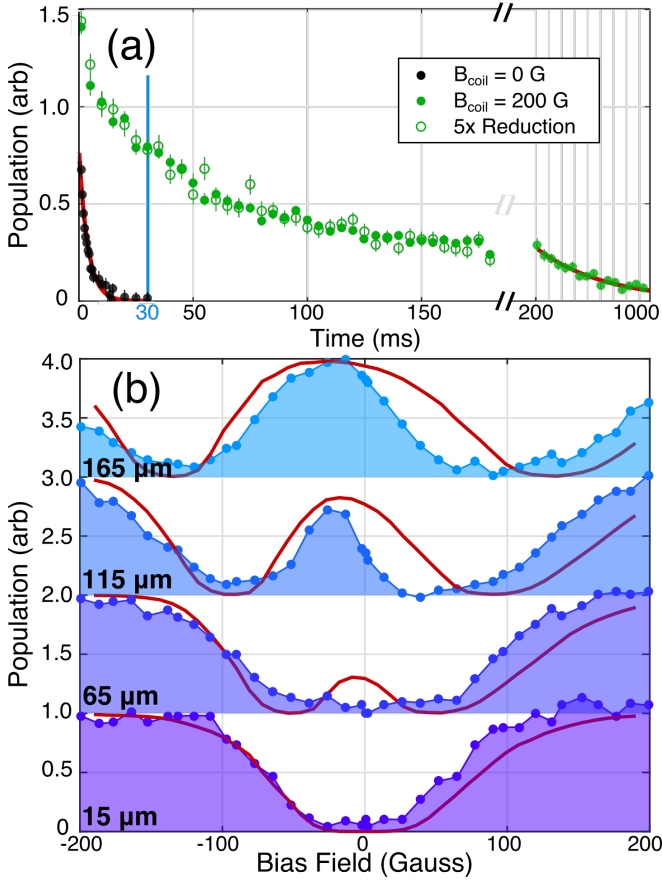


FIG. 4. Time traces (a) without bias field (black), with bias field (green dots), and with modulated density (green circles). One body fits (red) give loss rates of 200 s^{-1} without bias field and 2 s^{-1} with full bias field at long times, in agreement with our background gas pressure. At the fixed time 30 ms, population is shown as a function of both pin translation and bias field (b), for several values of pin translation, labeled relative to perfect alignment. Fits (red) are calculated by integrating the molecule flux of a thermal ensemble through surfaces where $\vec{E} \perp \vec{B}$.

eralize beyond OH. Hund’s case (a) states are most susceptible in the sense that smaller electric fields are sufficient to cause a significant problem, but with enough electric field any state exhibiting competition between E-fields and B-fields for alignment of the molecule or atom will be susceptible. 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, $\Lambda = 0$) [30]. In these states, which include most laser-cooled molecules thus far, the E-fields and B-fields couple to rotation and spin respectively, which are only related by the spin-rotation coupling constant γ . Since γ is usually in the tens of MHz [23], molecular spin-flip loss remains quite significant. The inclusion of hyperfine requires a careful case-by-case investigation. For OH, it would initially seem to add an extra

splitting that could protect from spin-flips, but in fact the lossy plane is only shifted slightly away from $\vec{E} \perp \vec{B}$ but retains the same area. For YO [35], certain hyperfine states can avoid spin-flip loss entirely when E-field is applied. These states are characterized by significant electron-spin-to-nuclear-spin dipolar coupling, which results in a protective gap regardless of field orientation.

We can also generalize to arbitrary geometries with a simple loss suppression strategy: avoid $\mu_{\text{eff}} B < d_{\text{eff}} E$ where $\vec{E} \perp \vec{B}$. This is maximally violated by the magnetic quadrupole, where in a suitably small region even the smallest stray electric field dominates the magnetic. One trivial tactic would be to use only one field, but we note that one of the main attractions of working with dipolar species is to utilize both. Another way is to trap with E-field and superpose B-field. Unlike the Zeeman effect, the Stark effect is never linear, and some zero field parity splitting always prevents the orientation reversing spin-flips that we have been discussing for magnetic traps. However, this same splitting pushes all states with the same sign of m_J very close to one another, leading to a different type of loss [36]. Intriguingly, this kind of electric trap loss is not necessarily worsened by the addition of magnetic field but can be totally mitigated by it [37]. Another option is to trap with both fields and keep zeros precisely overlapped. This was realized for OH with a superposed magnetic quadrupole and electric hexapole [38].

The present trap, in addition to providing our desired experimental testing ground for molecular spin-flip loss, also manages to provide large 5 T/cm trap-gradients useful for maintaining high densities and collision rates. Moreover, with loss removed, we observe a population trend whose initially fast decay rate decreases over time (Fig. 4a, green dots), suggesting collisions. To test this, we implement a five-fold reduction in initial population [39] and scale the resulting trend by five (green circles). If collisions had contributed, this new trend would show less decay, but we observe no significant change. This absence of collisions differs from our earlier work [22], but could be attributed to the significantly higher initial temperature in this trap. However, the results of this paper introduce a need for new interpretation of other aspects of this evaporation work, since the electric fields used for the RF knife ought to have caused a significant effect to the distribution. We discuss the implications further in supplementary materials. An alternative hypothesis for the population trend is the existence of chaotic trap orbits with long escape times [40]. Moving forward, we aim to increase the density by means of several improvements [41, 42].

Molecule enhanced spin-flip loss arises in mixed electric and magnetic fields due to a competition between field quantization axes. We conclusively demonstrate and suppress this effect using our dual magnetic and electric quadrupole trap, which is also an ideal setting for further

progress in collisional physics thanks to its large trap gradient. Our calculation of the magnitude of spin-flip loss via flux through surfaces where $\vec{E} \perp \vec{B}$ enables detailed predictions of how its location and magnitude ought to scale with bias field and trap alignment, which we experimentally verify. Our results correct existing predictions about molecular spin-flips in mixed fields and pave the way toward further improvements in molecule trapping and cooling.

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