GENERATING HIGH POWER CIRCULAR POLARIZED MICROWAVES

Implementing microwave shielding for CaF requires high power microwaves at 20.5 GHz with clean circular polarization. The microwave source must also have low phase noise to preserve the dressed state (see Dressed State Lifetime section). For shielding, microwaves are generated by mixing a 18.5 GHz (Keysight E8257D) source with a 2 GHz source (Windfreak SynthHD Pro locked to a 100 MHz low phase noise oscillator), Fig. S1. A pair of voltage controlled attenuators are used to control the 2 GHz microwave amplitude for preparation of the dressed states. Following the mixer, a bandpass filter selects the 20.5 GHz signal, which is then amplified. Next, the signal is split into four paths, each feeding a high power amplifier (Qorvo TGA4548-SM) producing ~5 W. After the amplifiers, mechanical phase shifters for each arm of the helical antenna array allow differential phase control which are used to steer the output beam and optimize the polarization in the tweezer volume. The antenna array itself consists of a 2 × 2 grid of helical antennas each rotated sequentially by 90 degrees. The helical antennas operate in axial mode creating circular polarization. The array has a copper backplane with a hole for the z-beam MOT light to pass through. A 90% transparency copper mesh covers this hole to allow light to pass through, but reflect microwaves. The antenna array is mounted inside of a re-entrant window tube just above the upper, in-vacuum, MOT coil. The end of the antenna is a few centimeters away from the molecules. Finite element analysis simulations show that reflections from the vacuum chamber vastly change the polarization cleanliness of the microwaves at the tweezer location. We optimize the polarization by adjusting the phase shifters to maximize the Rabi frequency of the transition driven by the $\sigma_-$ component of the field. The polarization is sensitive to differential phase shifts of approximately 10$^\circ$ between the 4 arms of the antenna. We use semi-rigid, phase stabilized SMA cables to minimize stress induced phase shifts from cable movements and temperature changes. The phase stability of the system was verified with a vector network analyzer.

MEASURING POLARIZATION

The polarization is characterized by driving Rabi oscillations on the three transitions between $|N = 1, J = 1/2, F = 0, m_f = 0\rangle$ and $|N = 0, F = 1, m_f = ±1, 0\rangle$, Fig. S1. We split the $F = 1$ manifold in a 13 Gauss magnetic field, sufficient to spectroscopically resolve the three $m_f$ projections. The matrix elements show a significant magnetic field dependence because a spin flip is required to drive the $\pi$ and $\sigma_+$ components of the transition. We calculate the matrix elements to be: 0.6 Debye for the $\sigma_+$ component, 1.0 Debye for $\pi$ and 1.4 Debye for $\sigma_-$ at 13 Gauss. The transition used for shielding has a dipole matrix element of 1.7 Debye and shows little dependence on magnetic field. The shielding transition is about 60 MHz higher in frequency than the transition used to measure polarization. We compare the ratios of Rabi frequencies on the shielding transition with the transition used to characterize the polarization and consistently find that the ratios agree with the matrix elements, suggesting that the 60 MHz frequency difference does not measurably change the polarization produced by the antenna.

To calculate the polarization ellipticity angle, $\xi$, in the plane containing the electric field vector, we first estimate the angle between the microwave k-vector and the z-axis magnetic field as

$$\theta = \tan^{-1}\left(\frac{\Omega_\pi}{\Omega_{\sigma_+} + \Omega_{\sigma_-}}\right).$$

The tilt angle is typically about 5–10°. We then estimate the polarization ellipticity angle in the plane containing the polarization ellipse from the $\sigma_{+,-}$ field components, defined as

$$\vec{E} = \epsilon_{\sigma_-} \cos(\xi) - \epsilon_{\sigma_+} \sin(\xi).$$

DRESSED STATE PREPARATION

The dressed states are adiabatically prepared by ramping the microwave power at a fixed detuning, where the sign of the detuning relative to the transition frequency selects either the upper or lower dressed state. The Rabi frequency is ramped up to the desired Rabi frequency in about 100 µs. The detuning remains fixed and is small with respect to the Rabi frequency (3 MHz when blue detuned and 7 MHz when red detuned when at the full microwave power (23 MHz Rabi frequency). The detuning is reduced for lower powers. Microwave shielding requires preparation of the upper dressed state. The microwaves couple the absolute ground state to the three $m_f$ projections of the $|N = 1, J = 1/2, F = 1, m_f = 0, \pm 1\rangle$ manifold. Calculations show that the upper dressed state remains clean as long as the initial detuning is blue of all transitions between the absolute ground state and $|N = 1, J = 1/2, F = 1, m_f = 0, \pm 1\rangle$. The lower dressed state, in contrast, hits the $m_f = 0$ level and adiabatically converts to the bare $|N = 1, J = 1/2, F = 1, m_f = 0\rangle$ state with only a small admixture of the ground state. For this reason, the direction of the magnetic field is set such that the microwave polarization drives the highest energy $m_f$ level. In order to measure the anti-shielding lifetime of the lower dressed state, we flip the magnetic field which makes the lowest $m_f$ level the transition most strongly driven by the microwave polarization. To prepare the lower dressed state,
the microwave detuning is set red of all nearby transitions, and then the power is ramped in the same way as for the shielding state preparation.

**Dressed State Lifetime**

The lifetime of the dressed states are measured by first transferring the population to the absolute ground state, then preparing the dressed state as described previously. We hold the dressing microwaves on at full power for a variable amount of time before reversing the power ramp. Population that remained in the original dressed state is returned to the absolute ground state while population that leaked into the wrong dressed state remains in \( N = 1 \). We then measure the population that remains in the \( N = 1 \) rotational level by direct imaging. Eventually, as the dressing time increases, the population becomes equally spread between the \( N = 0 \) and \( N = 1 \) states as the initially prepared dressed state becomes an incoherent mixture. We found that the lifetime of the dressed state was limited by phase noise at an offset frequency of 20.5 GHz microwave dressing time (calculated following [49]). The time dependent population in the undesired dressed state is

\[
P = \frac{1}{2} \left[ 1 - \exp \left( -\frac{\pi^2}{2} \Omega_R^2 S_\phi(\Omega_R) t \right) \right].
\]

Using the measured dressed state lifetime of \( >500 \) ms to back out the single side band phase noise at an offset frequency of 23 MHz, we extract a single side band phase noise of less than -150 dBc/Hz for the microwaves generated.

**Single Particle Lifetime**

The single particle lifetime was verified by post-selecting on data where a single molecule is loaded in the first image. Vacuum losses are canceled out by design as the experiential sequence for all collisions times is the same. As no decay is visible in Fig 2S, this shows that the two particle loss, shown in Figure 2, is dominated by two-body collisions.
FIG. S2. The single particle survival in the tweezers traps for the upper and lower dressed states as well as the ground state. The data set presented is the same as that presented in Figure 2 of the manuscript. The error bars represent the standard error.

Polarization Cleanliness Effect on Shielding

FIG. S3. The blue trace (64 ms) shows the shielded loss rate with $\zeta = 0.1$ while the orange trace (59 ms) shows $\zeta = 0.4$ a Rabi frequency of $\sim 23$ MHz, while in the upper dressed state. The error bars represent the standard error.

The effect of the shielding polarization cleanliness on the shielding efficiency was found to have little effect between an $\zeta = 0.1$ (100:1 in power) and $\zeta = 0.4$ (10:1 in power), a Rabi frequency of $\sim 23$ MHz, as predicted by the theory shown in Figure 3.

Tensor Stark Shifts

The states in the $N=1$ manifold used for shielding have a non-zero tensor polarizability, which we characterize with the following model

$$\hat{H}_{AC} = -I_{\alpha 2} P_2(\cos \theta),$$

where $\cos \theta$ is the angle between the molecular axis and the tweezer polarization direction and $-I_{\alpha 2} = 15$ MHz at the intensity used, as determined experimentally.

FIG. S4. Tensor Stark shifts of the $|N = 1, J = 1/2, F = 1, m_f = 0, \pm 1\rangle$ manifold versus tweezer polarization. In the z-polarized tweezer, the electric of the 780 nm light points parallel with the magnetic field. At B=0 Gauss, the $m_f = \pm 1$ states are degenerate. The xy-polarized tweezer has optical polarization pointing perpendicular to the magnetic field. From the multi-Gaussian fits shown we extract $-I_{\alpha 2} = 15$ MHz. The error bars represent the standard error.

We spectroscopically measure the the combined tensor AC Stark shifts from the tweezer light and Zeeman shifts from the applied magnetic field with the tweezer polarization parallel to the magnetic field and rotated 90° relative to the magnetic field (see Fig. S4). Shielding data was taken with the tweezer polarization parallel to the magnetic field for all data sets shown. At the tweezer depth used for shielding, there is a level crossing near 20 Gauss, where the $|N = 1, J = 1/2, F = 1, m_f = -1\rangle$ state crosses $|N = 1, J = 1/2, F = 1, m_f = 0\rangle$ to become the highest energy state. The ordering of $|N = 1, J = 1/2, F = 1, m_f = 0\rangle$ and $|N = 1, J = 1/2, F = 1, m_f = -1\rangle$ does not significantly change the dressed state preparation as long as the frequency of the microwave source remains blue detuned of all transitions from the ground state to the $|N = 1, J = 1/2, F = 1, m_f = 0, \pm 1\rangle$ manifold. The only significant effect of the $m_f = 0$ and $m_f = -1$ state ordering is to limit the minimum detuning from $m_f = -1$ when $m_f = 0$ is the higher energy state.

TRAP FREQUENCY

The trap frequency is measured by parametric heating, where the trap depth is modulated, resulting in strong heating when the modulation frequency is twice the trap frequency. We resolve the parametric heating feature at
a modulation frequency of 183 kHz for both the dressed and ground states. This corresponds to a trap frequency of \( \omega_r = 2\pi \times 91.5 \) kHz. From our calculations of the tensor AC Stark shifts in the trap, we expect the trap depth for the bare ground state and for the dressed state to be the same to within about 1%. The matching radial trap frequency, measured in the same tweezer, is enough to ensure that the trap depth is the same for dressed state and bare ground state, to within experimental error.

**TEMPERATURE**

The temperature of the trapped molecules is measured for both the dressed and ground states using the release and recapture method, where the trap is quickly switched off for the release time, then switched back on to recapture the molecules remaining in the trap volume. We use the 780 nm tweezer beam waist of 1.6 \( \mu \)m as well as the measured trap frequency as input parameters for a Monte-Carlo simulation of the release and re-capture dynamics [29]. Comparing to the results of the simulation, we fit a temperature of 96 (5) \( \mu \)K for both the bare ground state and for the dressed state molecules. The temperature measurement is then used to obtain the density in the tweezer and for thermal averaging of the loss rates in the coupled channels calculation.

**DRESSED STATE DENSITY**

In the experiment, we directly measure the lifetime of the molecule pairs, which requires knowing the density to calculate a 2-body loss rate. The trap depths and temperatures are measured to be the same, thus the density is the same for bare ground state and the upper dressed state. Equality of the density for both samples allows the “shielding factor” calculated in the main text to directly be the ratio of shielding lifetimes to the ground state lifetime as measured in the experiment. The loss rate \( \beta \) is given by \( \beta = \frac{1}{\rho \tau} \), where \( \rho \) is the average single particle density in the trap and \( \tau \) is the measured lifetime.

**COUPLED-CHANNELS CALCULATIONS**

We perform coupled-channels calculations to theoretically describe microwave shielding of CaF molecules. These calculations are very similar to those reported in Refs. [37–39], and we refer the reader to these works for a detailed description. Compared to Refs. [37–39] we include an additional term in the CaF Hamiltonian which is the tensor AC Stark shift due to the tweezer light, as described in section titled “Tensor Stark Shifts”.

Here, we briefly describe the computational details for completeness.

We propagate coupled-channels wavefunctions from \( R_{\text{min}} = 50 \) to 10 000 \( a_0 \) using the renormalized Numerov algorithm of Ref. [50], and match to fully absorbing universal boundary conditions at short range. In practice some flux of molecules does reach short range, which we consider to undergo loss with unit probability. Under efficient shielding conditions, negligible flux reaches short range such that the residual long-range microwave-induced loss can be described quantitatively. In practice some flux of molecules does reach short range, which we consider to undergo loss with unit probability. The channel basis functions are of the form

\[
n_{A,B}m_{j_{A,B}}^{(A,B)}|s_{A,B}s_{r_{A,B}}^{(A,B)}|i_{A,B}i_{r_{A,B}}^{(A,B)}|LM_L|N\rangle
\]  

(6)
where \( n, s = 1/2 \) and \( i = 1/2 \) are the rotational, electron spin, and nuclear spin angular momenta of molecules \( A \) and \( B \). \( L \) is the angular momentum associated with the end-over-end rotation of the molecules, and \( N \) is the number of microwave photons. The basis set is truncated by including only functions with \( n \leq 2 \), \( L \leq 10 \), and \( N = 0, -1, -2 \). Furthermore, the basis set is adapted to the permutation of identical bosonic CaF molecules. The interaction is limited to the dipole-dipole interaction and the molecular constants such as hyperfine couplings, rotational constants and dipole moments are taken from Ref. [51, 52]. Cross sections are computed on a logarithmically spaced grid of 21 energies between 0.1 \( \mu \)K and 1 mK, and subsequently averaged thermally to obtain elastic and loss rate coefficients at \( T = 100 \) \( \mu \)K. In all calculations reported here we assume the magnetic field and tweezer polarization are along the \( z \) axis, and the microwave polarization lies in the \( xy \) plane. We perform two sets of calculations, described below.

In the first set of calculations, we assume the microwave polarization is perfectly \( \sigma^- \) circular. In this case, there is no symmetry about the \( z \) axis such that the following quantity is conserved \( \mathcal{M} = m_j^{(A)} + m_j^{(B)} + M_L - N \), where \( m_j^{(X)} \) is the \( z \) projection of total angular momentum for molecules \( X = A, B \), \( M_L \) is the \( z \) projection of the end-over-end orbital angular momentum, and \( N \) is the number of \( \sigma^- \)-polarized photons. Limiting the basis set to a single value of \( \mathcal{M} \), equal to that of the initial channel, significantly reduces the computational costs and speeds up the calculation. This is exploited everywhere except in Fig. 3(b).

In the second set of calculations we account for the ellipticity of the microwave polarization. This means that the total projection quantum number \( \mathcal{M} \) is no longer conserved, and basis functions corresponding to different values of \( \mathcal{M} \) must be included in a single coupled calculation. As a result, the calculations are much more computationally intensive and we can no longer account for spin explicitly. Instead, we assume the electron \( s = 1/2 \) and nuclear spins \( i = 1/2 \) are approximately coupled to a singlet state \( j = 0 \), which is approximately true for low magnetic fields. We assume we can neglect coupling to the other hyperfine states, such that we are effectively left with a spin-free calculation. The only correction that we take into account is that the \( g \)-factor in the excited state is larger, modeled by a rotational magnetic moment of 0.15 \( \mu_B \).

In Fig. S7 we show the magnetic field dependence of the loss rate for calculations with and without explicit inclusion of spin, and with and without inclusion of the tensor AC Stark shift. Without tensor AC Stark effect, the approximation of treating spin implicitly is reasonably accurate at low magnetic field, where the spins are approximately coupled to a singlet, but fails to describe the increase of the loss with magnetic field, as the spins start to quantize along the magnetic field. At magnetic field strengths used experimentally, between 10 and 54 G, the implicit spin treatment is not accurate. Including the tensor AC Stark effect, this becomes the dominant source of enhancement of loss at low magnetic field strength, and the effect of explicit treatment of the spin degrees of freedom is smaller. At higher magnetic field, the loss rate drops if we consider spin implicitly, however in reality this competes with increased losses caused by the loss of singlet character. This interplay results in the weak dependence on \( B \) between 10 and 54 G shown in Fig. 4(b). Including the tensor AC Stark shift, the implicit treatment of spin is not perfect but reasonably accurate except at the highest magnetic field strength. We also illustrate its accuracy in Fig. 3(a). In the remainder of this work we account for spin implicitly except for when calculating the magnetic field dependence of the loss rates, shown in Fig. 4(b).
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