Robust storage qubits in ultracold polar molecules

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Quantum states with long-lived coherence are essential for quantum computation, simulation and metrology. The nuclear spin states of ultracold molecules prepared in the singlet rovibrational ground state are an excellent candidate for encoding and storing quantum information. However, it is important to understand all sources of decoherence for these qubits, and then eliminate them, in order to reach the longest possible coherence times. Here, we fully characterise the dominant mechanisms for decoherence of a storage qubit in an optically trapped ultracold gas of RbCs molecules using high-resolution Ramsey spectroscopy. Guided by a detailed understanding of the hyperfine structure of the molecule, we tune the magnetic field to where a pair of hyperfine states have the same magnetic moment. These states form a qubit, which is insensitive to variations in magnetic field. Our experiments reveal a subtle differential tensor light shift between the states, caused by weak mixing of rotational states. We demonstrate how this light shift can be eliminated by setting the angle between the linearly polarised trap light and the applied magnetic field to a magic angle of $\arccos(1/\sqrt{3}) \approx 55^\circ$. This leads to a coherence time exceeding 5.6 s at the 95% confidence level.

Quantum coherence is a key resource [1], underpinning many prominent applications in quantum science and technology. These range from precision tests of fundamental physics [2], quantum metrology [3] and state-of-the-art atomic clocks [4] to quantum information processing [5], quantum simulation [6] and quantum thermodynamics [7]. Understanding the limits on quantum coherence is therefore of fundamental interest and technological importance. Cooling matter into the ultracold regime leads to long interrogation times coupled with exquisite experimental control, enabling quantum coherence to be investigated with incomparable precision.

Ultracold polar molecules [8, 9] have unique advantages over alternative ultracold systems, such as those comprised of neutral atoms [10] or trapped ions [11]. First, polar molecules possess rich internal structure arising from the addition of molecular vibration and rotation. This makes it possible to construct high-dimensional qudits [12] or synthetic dimensions with a large Hilbert space [13, 14], and furthermore allows the precise measurements of energies self-consistently across optical, microwave, and radio-frequency domains. Second, polar molecules combine this rich internal structure with access to controllable long-range dipole-dipole interactions [15]. These properties have stimulated a diverse range of proposed applications spanning the fields of quantum computation [12, 16–19], quantum simulation [20–23], quantum-state controlled chemistry [24–26], and precision tests of fundamental physics [27–29]. To realise many of these applications, we need to understand how to engineer long-lived quantum coherence in ultracold polar molecules.

In this work, we use high-precision Ramsey spectroscopy to investigate the sources of decoherence in an optically trapped ultracold gas of ^{87}Rb^{133}Cs molecules (hereafter RbCs). We focus on superpositions of nuclear spin states of the singlet rovibrational ground state. Such superpositions are expected to be relatively insensitive to magnetic dephasing, as the magnetic moments of the nuclear spins are small in comparison to electronic magnetic moments. Furthermore, the nuclear spin states are expected to experience near-identical AC Stark shifts in an optical trap, so that dephasing associated with the nonuniform optical potential is also suppressed. These properties point to the possibility of long-lived coherence and make the nuclear spin states of ultracold polar molecules excellent candidates for robust storage qubits in quantum computing architectures [17, 18, 30]. In such proposals, gate operations may be performed using resonant dipole-dipole interactions [20, 22] following microwave excitation to an excited rotational state, while single-qubit rotations can be performed using two-photon microwave pulses [31–34]. Previously, Park *et al.* performed a Ramsey experiment using a pair of nuclear spin states in an ultracold gas of fermionic NaK molecules. Over an interrogation time of ~ 0.6 s they measured a coherence time of 0.7(3) s [30]. However, the various decoherence mechanisms were not fully explored. Here, we completely characterise all experimentally relevant decoherence mechanisms. This new understanding allows us to construct a storage qubit which exhibits no signs of decoherence over an interrogation time of 1.2 s, and we can place a lower limit on the coherence time $T_2^* > 5.6$ s at the 95% confidence level, paving the way for the use of ultracold molecules as a platform for quantum computation.

To begin, we seek to identify pairs of nuclear spin

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states with identical magnetic moments that connect to a common excited rotational state, by calculating the rotational and hyperfine structure of the RbCs molecule in externally applied magnetic and optical fields [33–36]. We construct the Hamiltonian in a fully uncoupled basis set $|N, M_N\rangle\,|i_{\mathrm{Rb}}, m_{\mathrm{Rb}}\rangle\,|i_{\mathrm{Cs}}, m_{\mathrm{Cs}}\rangle$, as shown in Methods, where N represents the angular momentum of the molecule with its projection along the quantisation axis M_N, and $i_{\mathrm{Rb}} = 3/2, i_{\mathrm{Cs}} = 7/2$ denote the nuclear spins of Rb and Cs respectively, with their projections $m_{\mathrm{Rb}}, m_{\mathrm{Cs}}$. However, typical magnetic fields in our experiments are not sufficiently high to decouple the rotational and nuclear angular momenta. Even when the laser is polarised along the magnetic field, the only good quantum number that can be used to describe a given hyperfine sublevel is $M_F = M_N + m_{\mathrm{Rb}} + m_{\mathrm{Cs}}$. To uniquely identify each hyperfine state, we label them by $(N, M_F)_k$ where k is an index counting up the states with given N and M_F in order of increasing energy. There are 32 nuclear spin states in the $N = 0$ rotational ground state of RbCs, with energies E shown in Fig. 1(a). The magnetic moments $\mu = dE/dB$ for a selection of the states are plotted as a function of magnetic field B in Fig. 1(b). There are a number of state combinations which display crossings where the difference in magnetic moments is zero. These crossings indicate turning points in the energy difference between states, where the energy difference becomes insensitive to magnetic field noise. Our experiment produces an optically trapped ultracold gas of up to 3000 RbCs molecules with a peak density of $\sim 10^{11}\,\mathrm{cm}^{-3}$ by association from a pre-cooled atomic mixture [37, 38], using a procedure (described in Methods) that initialises the molecules in the state $(0, 4)_1$; full state compositions are given in Section I and Table I of the Supplementary Information. For simplicity, we therefore select the qubit states to be $|0\rangle \equiv (0, 4)_1$ and $|1\rangle \equiv (0, 3)_0$ which are predicted to have the same magnetic moment when $B = 154.524\,\mathrm{G}$, as shown in Fig. 1(c), where we plot $\mu_{01} = \mu_{|1\rangle} - \mu_{|0\rangle}$.

We investigate the dependence of the coherence time T_2 on μ_{01} by performing a series of Ramsey measurements to measure T_2 as a function of B, see Fig. 1(d) and Methods. The results are shown in Fig. 1(e). We observe the longest coherence time when the difference in magnetic moments between the two states is zero, as expected. We fit the magnetic field variation of T_2^* with

$$T_2^* = \left(\frac{|\mu_{01}|\,\Delta B}{h} + \frac{1}{T_2^{(\mathrm{AC})}}\right)^{-1}, \tag{1}$$

where ΔB and T_2^* are fitting parameters. A derivation of Eq. 1 is given in Section II of the Supplementary Information. μ_{01} is calculated from the molecular Hamiltonian as shown in Fig. 1(c). ΔB describes the magnitude of variation in magnetic field over the duration of the measurement which contributes to the decoherence. We find $\Delta B = 34(5)\,\mathrm{mG}$, which is consistent with the expected stability of the magnetic fields in our experiments. The term $T_2^{(\mathrm{AC})}$ accounts for other sources of decoherence, which we show below to be dominated by differential tensor light shifts. For these measurements we find $T_2^{(\mathrm{AC})} = 1.3(4)\,\mathrm{s}$, for trap light polarised with $\beta = 0^\circ$ and intensity $I = 15.8\,\mathrm{kW\,cm^{-2}}$. Fig. 1(f) shows Ramsey fringes recorded close to the $\mu_{01} = 0$ condition and contrasts the behaviour with that seen at $B = 217.39\,\mathrm{G}$ where, although the difference in magnetic moments is still small, magnetic dephasing limits the coherence time.

To show that the remaining decoherence $T_2^{(\mathrm{AC})}$ is dominated by differential tensor light shifts, we perform a series of Ramsey measurements using different optical trap intensities. Each Ramsey measurement allows us to precisely determine the difference in energy between $|0\rangle$ and $|1\rangle$. For these experiments the two microwaves fields differ in frequency by 76 kHz, and so by measuring the frequency of the Ramsey fringes δ, we determine the difference in energy between the states equal to $h \times (76\,\mathrm{kHz} + \delta)$. The sign of δ is found by comparison with additional Ramsey measurements with intentionally different two-photon detunings. We measure δ for a range of trap laser intensities, and find an intensity-dependent energy shift between the two states as shown in Fig. 2.

The differential light shift arises from terms off-diagonal in N which cause mixing between states with the same parity. The largest contributions to the light shift are second-order terms

$$\langle N = 0, M_N = 0|\, H_{\mathrm{AC}}^{(2)}\, |2, 0\rangle\, \langle 2, 0|\, H_{\mathrm{quad}}\, |0, 0\rangle\,, \tag{2}$$

where $H_{\mathrm{AC}}^{(2)}$ represents the rank-2 tensor component of the AC Stark interaction and and H_{quad} represents the nuclear electric quadrupole interaction, as described under Methods. The matrix elements of $H_{\mathrm{AC}}^{(2)}$ and H_{quad} are shown graphically in Figs. 2(a) and (b). These second-order terms lead to components with $N > 0$ in the state composition of $|0\rangle$ and $|1\rangle$ with coefficients $< 10^{-5}$. This results in tensor light shifts in the rotational ground state which scale with the anisotropic polarisability $\alpha^{(2)}$ [35] and depend on M_F and the laser polarisation. This is analogous to the tensor light shifts that arise in ground-state alkali atoms due to hyperfine structure [39]. The terms in Eq. 2 are all diagonal in M_N and are proportional to $P_2(\cos\beta) = \frac{1}{2}(3\cos^2\beta - 1)$, where β is the angle between the linearly polarised electric field of the trap light and the applied magnetic field which forms the quantisation axis. As a result, the light shift changes the observed two-photon detuning according to

$$\delta = (\alpha_{01} I)/2h\epsilon_0 c + \delta_0, \tag{3}$$

where

$$\alpha_{01} = X(B)\alpha^{(2)} P_2(\cos\beta), \tag{4}$$

is the difference in the effective differential polarisability between the states. Here, I is the average intensity experienced by the molecules, δ_0 is the two-photon detuning in free space, and $X(B)$ is a numerical factor which is determined from the molecular Hamiltonian and depends upon the magnetic field. We validate the form of Eq. 3 and Eq. 4 by comparing to a full model of the rotational and hyperfine structure in Section IV of the Supplementary Information.

In addition to the tensor light shift, each state also experiences a much larger scalar light shift. However, as the scalar shift is identical for all states, this does not contribute to decoherence. The largest differential tensor light shift we measure is $1.01995(6)\,\mathrm{Hz\,kW^{-1}\,cm^2}$, for $\beta = 0°$. This is caused by individual tensor lights shifts for each of the states which we calculate to be $-1.45\,\mathrm{Hz\,kW^{-1}\,cm^2}$ for $|0\rangle$, and $-2.47\,\mathrm{Hz\,kW^{-1}\,cm^2}$ for $|1\rangle$. In contrast the scalar light shift for both states is $-41.2\,\mathrm{kHz\,kW^{-1}\,cm^2}$; this is 4 orders of magnitude larger than the tensor light shifts.

We compare our calculations with the behaviour observed in experiments. For a magnetic field of $B \approx 154.50\,\mathrm{G}$ where magnetic decoherence is minimised, we calculate the prefactor $X(B) = 4.00(4) \times 10^{-5}$. We plot the differential light shift measured at this magnetic field in the optical trap for fixed laser polarisations $\beta = 0°, 54°, 90°$ in Fig. 2(c). The solid lines indicate a fit to the results using Eq. 3, with $\alpha^{(2)}$ and δ_0 as free parameters. We find excellent agreement between our model and the experiment, with $\alpha^{(2)}/4\pi\epsilon_0 = 545(4)\,a_0^3$ and $\delta_0 = 983.0(2)\,\mathrm{Hz}$. The uncertainties shown are the statistical uncertainties found in the fitting. Additional systematic uncertainties in $\alpha^{(2)}$ are given in Section V of the Supplementary Information. The value of δ_0 indicates the two-photon detuning in free space, and so we determine the free-space energy difference between the states of $h \times (76\,\mathrm{kHz} + \delta_0) = h \times 76.983\,0(2)\,\mathrm{kHz}$. This is in excellent agreement with a calculation from the molecular Hamiltonian which predicts an energy difference between the states of $h \times 77.0(7)\,\mathrm{kHz}$, where the uncertainty results from the current precision with which the strength of the scalar nuclear spin-spin interaction (c_4) and the magnitude of the nuclear magnetic moments are known for RbCs [33, 34].

To test our understanding of the origin of the differential light shift further, we explore different magnetic fields as shown in Fig. 2(d). For higher magnetic fields, the measurements are performed for $\beta = 0°$ only. The increased uncertainties arise from the magnetic dephasing restricting the measurement time. The variation with magnetic field arises from the numerical prefactor $X(B)$ in Eq. 4. We find good qualitative agreement between theory and experiment, with α_{01} rising with magnetic field. For calculations over a broader range of magnetic fields see Section VI of the Supplementary Information. Our theory appears to underestimate the increase in α_{01} slightly. We attribute the discrepancy to uncertainties in the parameters of the molecular Hamiltonian which combine in a non-trivial way in the calculation of $X(B)$. Ramsey measurements of the type presented here should permit further refinement of these parameters. This will be the focus of future work.

The tensor light shifts we observe are proportional to $P_2(\cos\beta)$. This allows us to engineer a magic polarisation trap, as $P_2(\cos\beta) = 0$ for the magic angle $\beta_{\mathrm{magic}} = \arccos\sqrt{1/3} \approx 55°$. This magic angle has been exploited previously to maximise coherence times for superpositions of rotational states in optically trapped ultracold polar molecules [31, 40]. We experimentally verify this angle dependence for the nuclear spin-states in Fig. 2(e) using a single beam of the dipole trap. We see that the polarisation dependence of the experimentally measured δ is well described by our model and that $\delta \approx \delta_0$ when $\beta \approx 55°$, indicating that the tensor light shift is zero. This is further confirmed using the measurements in Fig. 2(c), where all δ measured in the trap for $\beta = 55°$ are consistent with the free-space value, and the gradient of δ as a function of I is zero.

The optimal configuration to maximise coherence time is where $\mu_{01} = 0$ and $\alpha_{01} = 0$, which is realised in our experiments for $B \approx 154.5\,\mathrm{G}$ and $\beta \approx 55°$. We perform a Ramsey experiment using these optimal parameters as shown in Fig. 3. The maximum Ramsey time available is limited by collisional loss of the molecules [41, 42] with $T_1 = 0.61(4)$ seconds, which reduces our signal at long times. We measure Ramsey fringes out to $T = 1.2\,\mathrm{s}$. We find the energy difference between the states in the trap to be $h \times 76,982.733(16)\,\mathrm{Hz}$; this is a precision of 1 part in 10^7.

Over the 1.2 s interrogation time shown in Fig. 3, we observe no evidence of decoherence. An accurate value for the coherence time cannot be extracted from the data by least-squares fitting because the sum of squares of residuals decreases continuously as $T_2^* \to \infty$, as shown in Supplementary Figure 7. Instead, we estimate the lower bound of the coherence time with confidence intervals constructed using the Feldman and Cousins approach [43]. We find $T_2^* > 5.6\,\mathrm{s}$ at the 95 % confidence level as described in Section VII of the Supplementary Information; this is an order of magnitude longer than previous work [30].

It is also instructive to consider calculated limits on the coherence resulting from residual magnetic and optical dephasing. In the case of magnetic dephasing, the limit on the coherence time at $B = 154.52\,\mathrm{G}$ due to magnetic field noise of $\Delta B = 35\,\mathrm{mG}$ is $\sim 2.0 \times 10^3\,\mathrm{s}$. In terms of optical dephasing, laser polarisation of $\pm 0.5°$ from the magic angle results in a limit on the coherence time of $\sim 40\,\mathrm{s}$. Details of these calculations are given in sections II and VIII of the Supplementary Information.

Our measurements do not indicate any other detectable mechanisms for decoherence. We see no evidence for collisional energy shifts, which would be observed by a change in the energies of the states when the density reduces over the course of each Ramsey measurement, as shown in Section IX of the Supplementary Information. This is consistent with previous observations [30], and the absence of collisional energy shifts or decoherence may be expected as short-range collisions in the gas lead to loss of molecules with high probability [41, 42, 44, 45]. Measurements of the coherence out to longer times will require confinement of the molecules to a 3D optical lattice [46], optical tweezers [47–49], or the use of alternative trapping techniques such as a blue-detuned optical trap [50] to avoid losses from the optical excitation of two-molecule collision complexes [42, 51]. The creation of controlled arrays of molecules is also a key component of the proposed quantum computing protocols where storage qubits have applications; our method of using a magic-polarisation trap is compatible with the confinement of molecules to arrays of optical tweezers or a 3D optical lattice [52]. Our findings are broadly applicable to all $^1\Sigma$ molecules in their rovibrational ground state, including the range of bialkali molecules currently under investigation.

Robust storage qubits are a key component of the proposed architectures for quantum computing with ultracold polar molecules [17, 18, 30]. We have previously demonstrated single-qubit rotations in our apparatus using two-photon microwave pulses [34]. To complete the architecture, we will need to demonstrate high-fidelity gate operations between individually addressable molecules. This can be achieved using resonant dipole-dipole interactions [20, 22] between pairs of molecules in different rotational states, provided the rotational coherence in the trap is longer than the gate time. A promising direction is to use rotationally magic trapping wavelengths [50, 53] to confine molecules in optical tweezers [47, 49, 54, 55]. Our robust storage qubits can be combined with this approach to realise a platform for quantum computation using ultracold polar molecules.

METHODS

Hamiltonian used to calculate the rotational and hyperfine structure

We calculate the energy level structure of RbCs in the electronic and vibrational ground state by diagonalising the relevant Hamiltonian. We extract the energy levels and eigenstates of the Hamiltonian by numerical diagonalisation. The hyperfine constants for all our calculations are given in Table II of the Supplementary Information. We give details of the relevant Hamiltonian here, as well as in our previous publications [33–36]. In the presence of externally applied magnetic and off-resonant optical fields, the Hamiltonian (H_{RbCs}) can be decomposed into rotational (H_{rot}), hyperfine (H_{hf}), Zeeman (H_{Zeeman}), and AC Stark (H_{AC}) contributions [56, 57],

$$H_{\mathrm{RbCs}} = H_{\mathrm{rot}} + H_{\mathrm{hf}} + H_{\mathrm{Zeeman}} + H_{\mathrm{AC}}. \tag{5}$$

The rotational contribution is

$$H_{\mathrm{rot}} = B_v \boldsymbol{N}^2 - D_v \boldsymbol{N}^2\boldsymbol{N}^2, \tag{6}$$

where $\boldsymbol{N}$ is the rotational angular momentum operator, and B_v and D_v are the rotational and centrifugal distortion constants, respectively. The hyperfine contribution has four terms,

$$H_{\mathrm{hf}} = H_{\mathrm{quad}} + H_{II}^{(0)} + H_{II}^{(2)} + H_{NI}, \tag{7}$$

where

$$H_{\mathrm{quad}} = \sum_{j=\mathrm{Rb,Cs}} e\boldsymbol{Q}_j \cdot \boldsymbol{q}_j, \tag{8a}$$

$$H_{II}^{(0)} = c_4 \boldsymbol{I}_{\mathrm{Rb}} \cdot \boldsymbol{I}_{\mathrm{Cs}}, \tag{8b}$$

$$H_{II}^{(2)} = -c_3\sqrt{6}\boldsymbol{T}^2(C) \cdot \boldsymbol{T}^2\left(\boldsymbol{I}_{\mathrm{Cs}}, \boldsymbol{I}_{\mathrm{Rb}}\right), \tag{8c}$$

$$H_{NI} = \sum_{j=\mathrm{Rb,Cs}} c_j \boldsymbol{N} \cdot \boldsymbol{I}_j. \tag{8d}$$

H_{quad} describes the interaction between the nuclear electric quadrupole ($e\boldsymbol{Q}_j$) of nucleus j with the electric field gradient at the nucleus ($\boldsymbol{q}_j$). $H_{II}^{(0)}$ and $H_{II}^{(2)}$ are the scalar and tensor nuclear spin-spin interactions, with strengths determined by the coupling constants c_3 and c_4 respectively. The second-rank tensors $\boldsymbol{T}^2$ describe the anisotropy of the spin-spin interaction [58]. H_{NI} describes the interaction between the magnetic field generated by the rotating molecule and the nuclear magnetic moments, with coupling constants c_j.

The interaction of the molecule with the external magnetic field ($\boldsymbol{B}$) is described by the Zeeman contribution,

$$\begin{aligned} H_{\mathrm{Zeeman}} = &- g_{\mathrm{r}}\mu_{\mathrm{N}}\boldsymbol{N} \cdot \boldsymbol{B} \\ &- \sum_{j=\mathrm{Rb,Cs}} g_j\left(1-\sigma_j\right)\mu_{\mathrm{N}}\boldsymbol{I}_j \cdot \boldsymbol{B}, \end{aligned} \tag{9}$$

where μ_{N} is the nuclear magneton. The first term is the contribution from the magnetic moment associated with the rotation of the molecule, which is characterised by the g-factor g_{r}. The second term describes the contribution from the nuclear spins, characterised by the nuclear g-factors g_j and shielded isotropically by the factor σ_j [57]. In our calculations, the magnetic field vector is directed parallel to the space-fixed z axis with magnitude B.

The presence of the optical trap gives rise to the AC Stark effect, which results from the interaction between the off-resonant oscillating electric field of the trap light $\boldsymbol{E}_{\mathrm{AC}}$ and the frequency-dependent molecular polarisability tensor $\boldsymbol{\alpha}$ [35]. This contributes

$$H_{\mathrm{AC}} = H_{\mathrm{AC}}^{(0)} + H_{\mathrm{AC}}^{(2)} = -\frac{1}{2}\boldsymbol{E}_{\mathrm{AC}} \cdot \boldsymbol{\alpha} \cdot \boldsymbol{E}_{\mathrm{AC}}. \tag{10}$$

Here, $H_{\mathrm{AC}}^{(0)}$ and $H_{\mathrm{AC}}^{(2)}$ are written explicitly to show that H_{AC} can be decomposed into scalar and tensor components respectively. The scalar component produces the same shift in energy for all (N, M_N).

The terms H_{quad} and $H_{\mathrm{AC}}^{(2)}$ have components which are off-diagonal in N, and are the dominant contributions to the tensor light shifts which we observe. To be explicit, the matrix elements for these terms are included in Section III of the Supplementary Information.

Production of ultracold RbCs molecules

We produce ground-state RbCs molecules from an optically trapped ultracold mixture of ^{87}Rb and ^{133}Cs atoms using a two-step process. First, we use magnetoassociation on an interspecies Feshbach resonance at 197 G [59]. Following this, the remaining atoms are removed from the trap using the Stern-Gerlach effect. Second, the magnetic field is set to 181.6 G, where the molecules are transferred to a single hyperfine sublevel of the $X^1\Sigma\,(v=0, N=0)$ rovibrational ground state using stimulated Raman adiabatic passage (STIRAP) [37, 38, 60]. We set the STIRAP to initialise the molecules in $|0\rangle \equiv (0,4)_1$, and the transfer is performed with the trap light switched off to avoid spatially varying AC Stark shifts which otherwise limit the efficiency [38]. Following STIRAP, the molecules are recaptured in a crossed optical dipole trap at $\lambda = 1550$ nm; see Section X of the Supplementary Information for details. Both beams are linearly polarised at an angle β with respect to the magnetic field. The molecules have a typical temperature of $0.7\,\mu$K, and a peak density of $\sim 1\times10^{11}\,\mathrm{cm}^{-3}$. We detect molecules by reversing the association process and performing absorption imaging on the resulting atomic clouds. As such, we only image molecules which occupy $|0\rangle$.

Ramsey measurement protocol

To measure the coherence time of the qubit, we perform Ramsey spectroscopy. To couple the qubit states, we use two microwave fields to form a 3-level lambda system, where both qubit states are coupled to a common rotationally excited state $|E\rangle$, which is chosen to have significant transition dipole moment to both $|0\rangle$ and $|1\rangle$. The excited states used throughout this work are tabulated in Table I of the Supplementary Information. Technical details of the microwave apparatus we use are given in Section XI of the supplementary information.

For the measurements presented in Fig. 1, we prepare an equal superposition of $|0\rangle$ and $|1\rangle$ by applying a $\pi/2$ pulse on the $|0\rangle \leftrightarrow |E\rangle$ transition followed by a π pulse on the $|E\rangle \leftrightarrow |1\rangle$ transition. The optical trap is briefly switched off during any microwave pulses in order to avoid varying AC Stark shifts of the transitions across the thermal spatial distribution of molecules. The typical duration for each pulse is $\sim 100\,\mu$s. We project the phase of the superposition onto the population of the states by reversing this pulse sequence after a hold time T as shown in Fig.1(d). During T the molecules are confined to the crossed optical dipole trap, and a DC magnetic field is applied in the vertical z direction. With the two microwave frequencies fixed, we observe Ramsey fringes in the form of an oscillating number of molecules in the initial state $|0\rangle$ as a function of T.

For the measurements presented in Fig. 2 and Fig. 3, we find that there is a strong transition from $|0\rangle$ to $(1,4)_4$ just 20 kHz detuned from the $|E\rangle \leftrightarrow |1\rangle$ transition frequency. We therefore use a modified Ramsey sequence as shown in the inset to Fig. 3 to avoid off-resonantly driving the population out of $|0\rangle$ during the Ramsey pulses.

The frequency of the Ramsey fringes is equal to the two-photon detuning of the microwaves. We fit a model to the fringes, which is derived in Section XII of the Supplementary Information, that accounts for both two-body collisional loss of molecules and decoherence of the superposition,

$$N(T) = \frac{N_i}{2}\left(\frac{1}{1+\frac{T}{T_1}[e-1]}\right)\times\left[e^{-T/T_2^*}\cos(2\pi(\delta T+\phi))+1\right]. \tag{11}$$

Here, N_i is the initial total number of molecules, T_1 is the $1/e$ lifetime for molecules in the trap, T_2^* is the $1/e$ coherence time, and δ and ϕ are the frequency and phase of the Ramsey fringes.

To set the magnetic field for a given measurement, we jump the magnetic field to its target value immediately after the STIRAP and then hold 5 ms before the start of the Ramsey sequence. After the Ramsey sequence is completed, the magnetic field is jumped back to 181.6 G and held for 5 ms before the return STIRAP and imaging.

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II. DATA AVAILABILITY

The data associated with this work are available at: **DOI:10.15128/r25m60qr93w**.

III. CODE AVAILABILITY

The python code for hyperfine structure calculations can be found at **DOI:10.5281/ZENODO.3755881**.

IV. AUTHOR CONTRIBUTIONS

P.D.G., J.A.B., S.L.B., and S.L.C. conceived the idea and planned the experiments. P.D.G. and J.A.B. performed the experiments and carried out the data analysis. J.A.B. wrote the code for calculating the rotational and hyperfine structure of the molecules with guidance from J.M.H.. All authors contributed to interpreting the results and writing the manuscript.

V. COMPETING INTERESTS

The authors declare no competing interests.

[1] A. Streltsov, G. Adesso, and M. B. Plenio, Colloquium: Quantum coherence as a resource, Rev. Mod. Phys. **89**, 041003 (2017).

[2] M. S. Safronova, D. Budker, D. DeMille, D. F. J. Kimball, A. Derevianko, and C. W. Clark, Search for new physics with atoms and molecules, Rev. Mod. Phys. **90**, 025008 (2018).

[3] V. Giovannetti, S. Lloyd, and L. Maccone, Advances in quantum metrology, Nature Photon **5**, 222 (2011).

[4] A. D. Ludlow, M. M. Boyd, J. Ye, E. Peik, and P. O. Schmidt, Optical atomic clocks, Rev. Mod. Phys. **87**, 637 (2015).

[5] T. Ladd, F. Jelezko, R. Laflamme, Y. Nakamura, C. Monroe, and J. O'Brien, Quantum computers, Nature **464**, 45 (2010).

[6] I. M. Georgescu, S. Ashhab, and F. Nori, Quantum simulation, Rev. Mod. Phys. **86**, 153 (2014).

[7] S. Vinjanampathy and J. Anders, Quantum thermodynamics, Contemporary Physics **57**, 545 (2016).

[8] L. D. Carr, D. DeMille, R. V. Krems, and J. Ye, Cold and ultracold molecules: science, technology and applications, New J. Phys. **11**, 055049 (2009).

[9] J. L. Bohn, A. M. Rey, and J. Ye, Cold molecules: Progress in quantum engineering of chemistry and quantum matter, Science **357**, 1002 (2017).

[10] A. W. Young, W. J. Eckner, W. R. Milner, D. Kedar, M. A. Norcia, E. Oelker, N. Schine, J. Ye, and A. M. Kaufman, Half-minute-scale atomic coherence and high relative stability in a tweezer clock, Nature **588**, 408 (2020).

[11] P. Wang, C.-Y. Luan, M. Qiao, M. Um, J. Zhang, Y. Wang, X. Yuan, M. Gu, J. Zhang, and K. Kim, Single ion qubit with estimated coherence time exceeding one hour, Nat. Comms. **12**, 233 (2021).

[12] R. Sawant, J. A. Blackmore, P. D. Gregory, J. Mur-Petit, D. Jaksch, J. Aldegunde, J. M. Hutson, M. R. Tarbutt, and S. L. Cornish, Ultracold polar molecules as qudits, New J. Phys. **22**, 013027 (2020).

[13] B. Sundar, B. Gadway, and K. R. A. Hazzard, Synthetic dimensions in ultracold polar molecules, Sci. Rep. **8**, 3422 (2018).

[14] B. Sundar, M. Thibodeau, Z. Wang, B. Gadway, and K. R. A. Hazzard, Strings of ultracold molecules in a synthetic dimension, Phys. Rev. A **99**, 013624 (2019).

[15] B. Yan, S. A. Moses, B. Gadway, J. P. Covey, K. R. A. Hazzard, A. M. Rey, D. S. Jin, and J. Ye, Observation of dipolar spin-exchange interactions with lattice-confined polar molecules, Nature **501**, 521 (2013).

[16] D. Demille, Quantum computation with trapped polar molecules, Phys. Rev. Lett. **88**, 067901 (2002).

[17] S. F. Yelin, K. Kirby, and R. Côté, Schemes for robust quantum computation with polar molecules, Phys. Rev. A **74**, 050301 (2006).

[18] K.-K. Ni, T. Rosenband, and D. D. Grimes, Dipolar exchange quantum logic gate with polar molecules, Chem. Sci. **9**, 6830 (2018).

[19] M. Hughes, M. D. Frye, R. Sawant, G. Bhole, J. A. Jones, S. L. Cornish, M. R. Tarbutt, J. M. Hutson, D. Jaksch, and J. Mur-Petit, Robust entangling gate for polar molecules using magnetic and microwave fields, Phys. Rev. A **101**, 062308 (2020).

[20] R. Barnett, D. Petrov, M. Lukin, and E. Demler, Quantum magnetism with multicomponent dipolar molecules in an optical lattice, Phys. Rev. Lett. **96**, 190401 (2006).

[21] A. Micheli, G. K. Brennen, and P. Zoller, A toolbox for lattice-spin model with polar molecules, Nat. Phys. **2**, 341 (2006).

[22] A. V. Gorshkov, S. R. Manmana, G. Chen, E. Demler, M. D. Lukin, and A. M. Rey, Quantum magnetism with polar alkali-metal dimers, Phys. Rev. A **84**, 033619 (2011).

[23] S. R. Manmana, E. M. Stoudenmire, K. R. A. Hazzard, A. M. Rey, and A. V. Gorshkov, Topological phases in ultracold polar-molecule quantum magnets, Phys. Rev. B **87**, 081106 (2013).

[24] R. V. Krems, Cold controlled chemistry, Physical Chemistry Chemical Physics **10**, 4079 (2008).

[25] N. Balakrishnan, Perpective: Ultracold molecules and the dawn of cold controlled chemistry, Journal of Chemical Physics **145**, 150901 (2016).

[26] M.-G. Hu, Y. Liu, M. A. Nichols, L. Zhu, G. Quéméner, O. Dulieu, and K.-K. Ni, Nuclear spin conservation enables state-to-state control of ultracold molecular reactions, Nature Chemistry 10.1038/s41557-020-00610-0 (2020).

[27] T. Zelevinsky, S. Kotochigova, and J. Ye, Precision test of mass-ratio variations with lattice-confined ultracold molecules, Phys. Rev. Lett. **100**, 043201 (2008).

[28] J. J. Hudson, D. M. Kara, I. J. Smallman, B. E. Sauer, M. R. Tarbutt, and E. A. Hinds, Improved measurement of the shape of the electron, Nature **473**, 493 (2011).

[29] The ACME Collaboration, Improved limit on the electric dipole moment of the electron, Nature **562**, 355 (2018).

[30] J. W. Park, Z. Z. Yan, H. Loh, S. A. Will, and M. W. Zwierlein, Second-scale nuclear spin coherence time of ultracold ^{23}Na^{40}K molecules, Science **357**, 372 (2017).

[31] B. Neyenhuis, B. Yan, S. A. Moses, J. P. Covey, A. Chotia, A. Petrov, S. Kotochigova, J. Ye, and D. S. Jin, Anisotropic polarizability of ultracold polar ^{40}K^{87}Rb molecules, Phys. Rev. Lett. **109**, 230403 (2012).

[32] S. A. Will, J. W. Park, Z. Z. Yan, H. Loh, and M. W. Zwierlein, Coherent microwave control of ultracold ^{23}Na^{40}K molecules, Phys. Rev. Lett. **116**, 225306 (2016).

[33] P. D. Gregory, J. Aldegunde, J. M. Hutson, and S. L. Cornish, Controlling the rotational and hyperfine state of ultracold ^{87}Rb^{133}Cs molecules, Phys. Rev. A **94**, 041403(R) (2016).

[34] J. A. Blackmore, P. D. Gregory, S. L. Bromley, and S. L. Cornish, Coherent manipulation of the internal state of ultracold ^{87}Rb^{133}Cs molecules with multiple microwave fields, Phys. Chem. Chem. Phys. **22**, 27529 (2020).

[35] P. D. Gregory, J. A. Blackmore, J. Aldegunde, J. M. Hutson, and S. L. Cornish, ac stark effect in ultracold polar ^{87}Rb^{133}Cs molecules, Phys. Rev. A **96**, 021402(R) (2017).

[36] J. A. Blackmore, R. Sawant, P. D. Gregory, S. L. Bromley, J. Aldegunde, J. M. Hutson, and S. L. Cornish, Controlling the ac Stark effect of RbCs with dc electric and magnetic fields, Phys. Rev. A **102**, 053316 (2020).

[37] P. K. Molony, P. D. Gregory, Z. Ji, B. Lu, M. P. Köppinger, C. R. Le Sueur, C. L. Blackley, J. M. Hutson, and S. L. Cornish, Creation of ultracold ^{87}Rb^{133}Cs molecules in the rovibrational ground state, Phys. Rev. Lett. **113**, 255301 (2014).

[38] P. K. Molony, P. D. Gregory, A. Kumar, C. R. Le Sueur, J. M. Hutson, and S. L. Cornish, Production of ultracold ^{87}Rb^{133}Cs in the absolute ground state: complete characterisation of the STIRAP transfer, ChemPhysChem. **17**, 3811 (2016).

[39] P. G. H. Sandars, Differential polarizability in the ground state of the hydrogen atom, Proc. Phys. Soc. **92**, 857 (1967).

[40] F. Seeßelberg, X.-Y. Luo, M. Li, R. Bause, S. Kotochigova, I. Bloch, and C. Gohle, Extending rotational coherence of interacting polar molecules in a spin-decoupled magic trap, Phys. Rev. Lett. **121**, 253401 (2018).

[41] P. D. Gregory, M. D. Frye, J. A. Blackmore, E. M. Bridge, R. Sawant, J. M. Hutson, and S. L. Cornish, Sticky collisions of ultracold RbCs molecules, Nat. Comms **10**, 3104 (2019).

[42] P. D. Gregory, J. A. Blackmore, S. L. Bromley, and S. L. Cornish, Loss of ultracold ^{87}Rb^{133}Cs molecules via optical excitation of long-lived two-body collision complexes, Phys. Rev. Lett. **124**, 163402 (2020).

[43] G. J. Feldman and R. D. Cousins, Unified approach to the classical statistical analysis of small signals, Physical Review D **57**, 3873 (1998).

[44] X. Ye, M. Guo, M. L. González-Martínez, G. Quéméner, and D. Wang, Collisions of ultracold ^{23}Na^{87}Rb molecules with controlled chemical reactivities, Sci. Adv. **4**, eaaq0083 (2018).

[45] J. F. E. Croft, J. L. Bohn, and G. Quéméner, Unified model of ultracold molecular collisions, Phys. Rev. A **102**, 033306 (2020).

[46] B. Zhu, B. Gadway, M. Foss-Feig, J. Schachenmayer, M. L. Wall, K. R. A. Hazzard, B. Yan, S. A. Moses, J. P. Covey, D. S. Jin, J. Ye, M. Holland, and A. M. Rey, Suppressing the loss of ultracold molecules via the continuous quantum zeno effect, Phys. Rev. Lett. **112**, 070404 (2014).

[47] L. Anderegg, L. W. Cheuk, Y. Bao, S. Burchesky, W. Ketterle, K.-K. Ni, and J. M. Doyle, An optical tweezer array of ultracold molecules, Science **365**, 1156 (2019).

[48] L. W. Cheuk, L. Anderegg, Y. Bao, S. Burchesky, S. S. Yu, W. Ketterle, K.-K. Ni, and J. M. Doyle, Observation of collisions between two ultracold ground-state caf molecules, Phys. Rev. Lett. **125**, 043401 (2020).

[49] J. T. Zhang, Y. Yu, W. B. Cairncross, K. Wang, L. R. B. Picard, J. D. Hood, Y.-W. Lin, J. M. Hutson, and K.-K. Ni, Forming a single molecule by magnetoassociation in an optical tweezer, Phys. Rev. Lett. **124**, 253401 (2020).

[50] R. Bause, M. Li, A. Schindewolf, X.-Y. Chen, M. Duda, S. Kotochigova, I. Bloch, and X.-Y. Luo, Tune-out and magic wavelengths for ground-state ^{23}Na^{40}K molecules, Phys. Rev. Lett. **125**, 023201 (2020).

[51] Y. Liu, M.-G. Hu, M. A. Nichols, G. D. D., T. Karman, H. Guo, and K.-K. Ni, Photo-excitation of long-lived transient intermediates in ultracold reactions, Nat. Phys. **16**, 1132 (2020).

[52] S. Kotochigova and D. DeMille, Electric-field-dependent dynamic polarizability and state-insensitive conditions for optical trapping of diatomic polar molecules, Phys. Rev. A **82**, 063421 (2010).

[53] Q. Guan, S. L. Cornish, and S. Kotochigova, Magic conditions for multiple rotational states of bialkali molecules in optical lattices, Phys. Rev. A **103**, 043311 (2021).

[54] W. B. Cairncross, J. T. Zhang, L. R. B. Picard, Y. Yu, K. Wang, and K.-K. Ni, Assembly of a rovibrational ground state molecules in an optical tweezer, Phys. Rev. Lett. **126**, 123402 (2021).

[55] R. V. Brooks, S. Spence, A. Guttridge, A. Alampounti, A. Rakonjac, L. A. McArd, J. M. Hutson, and S. L. Cornish, Preparation of one ^{87}Rb and one ^{133}Cs atom in a single optical tweezer, arXiv:2104.05760 (2021).

[56] J. M. Brown and A. Carrington, Rotational Spectroscopy of Diatomic Molecules (Cambridge University Press, 2010).

[57] J. Aldegunde, B. A. Rivington, P. S. Zuchowski, and J. M. Hutson, Hyperfine energy levels of alkali-metal dimers: Ground-state polar molecules in electric and magnetic fields, Phys. Rev. A **78**, 033434 (2008).

[58] J. Aldegunde and J. M. Hutson, Hyperfine structure of alkali-metal diatomic molecules, Phys. Rev. A **96**, 042506 (2017).

[59] M. P. Köppinger, D. J. McCarron, D. L. Jenkin, P. K. Molony, H. W. Cho, S. L. Cornish, C. R. Le Sueur, C. L. Blackley, and J. M. Hutson, Production of optically trapped ^{87}RbCs Feshbach molecules, Phys. Rev. A **89**, 033604 (2014).

[60] P. D. Gregory, P. K. Molony, M. P. Köppinger, A. Kumar, Z. Ji, B. Lu, A. L. Marchant, and S. L. Cornish, A simple, versatile laser system for the creation of ultracold ground state molecules, New J. Phys. **17**, 055006 (2015).

FIG. 1. **Effect of magnetic fields on the coherence of the storage qubit.** **(a)** Zeeman structure of the nuclear spin states in the rotational ground state of RbCs. **(b)** Magnetic moments μ/μ_{N} of the states as a function of magnetic field B. We construct the qubit from the states $|0\rangle \equiv (0,4)_1$ and $|1\rangle \equiv (0,3)_0$, which are chosen as they possess identical magnetic moments when $B = 154.524\,\mathrm{G}$ (indicated by the vertical dashed line). **(c)** The differential magnetic moment $\mu_{01} = \mu_{|1\rangle} - \mu_{|0\rangle}$ as a function of magnetic field. **(d)** Configuration of states and microwave pulse sequence used to perform Ramsey spectroscopy. The energy separation between the qubit states and the rotationally excited state $|E\rangle$ is $2B_v \approx 980\,\mathrm{MHz}$. **(e)** Measured coherence time T_2^* as a function of magnetic field, with error bars indicating 1σ uncertainties. The line shows a model for the decoherence as described in the text. Fitting to the results indicates a magnetic field variation of 34(5) mG over the course of the measurement contributes to the observed decoherence, and a peak coherence time $T_2^{(\mathrm{AC})} = 1.3(4)\,\mathrm{s}$ when $\mu_{01} \approx 0$. **(f)** Example Ramsey measurement performed at $B = 154.50\,\mathrm{G}$, where $\mu_{01} \approx 0$. The y-axis indicates the number of molecules remaining in state $|0\rangle$ measured as a function of the Ramsey time T. The red shaded region indicates the maximum and minimum of the Ramsey fringes observed when $B = 217.39\,\mathrm{G}$, where $\mu_{01} \approx 0.17\,\mu_{\mathrm{N}}$ and the coherence time is correspondingly much shorter. The trap light has polarisation $\beta = 0^\circ$ and intensity $I = 15.8\,\mathrm{kW\,cm^{-2}}$ for all measurements shown.

FIG. 2. **Differential tensor light shifts between nuclear spin states in the rotational ground state.** We measure the differential light shifts between $|0\rangle \equiv (0,4)_1$ and $|1\rangle \equiv (0,3)_0$ by Ramsey spectroscopy. We show the Hamiltonian matrix elements for **(a)** the nuclear electric quadrupole interaction H_{quad} and **(b)** the tensor AC Stark effect $H_{\mathrm{AC}}^{(2)}$ graphically for the uncoupled $|N, M_N, m_{\mathrm{Rb}}, m_{\mathrm{Cs}}\rangle$ basis. The states are split by dashed lines into groups according to N and by minor ticks into multiples of 32 basis states with the same M_N. The block diagonal elements in N are labelled for $N > 0$. The color coding indicates the value of the matrix element in units of frequency. Note the off-diagonal elements connecting states with $\Delta N = 2$ shaded in yellow, which lead to tensor light shifts proportional to the anisotropic polarisability. The mathematical expressions describing the matrix elements are given in Section III of the Supplementary Information. $H_{\mathrm{AC}}^{(2)}$ is calculated for an intensity of 16 $\mathrm{kW\,cm^{-2}}$ and a polarisation angle of $\beta = 0^\circ$. **(c)** Two-photon detuning δ as a function of the trap intensity I. The linear polarisation of each beam is set to an angle β of (i) 0° (ii) 55° (iii) 90° with respect to a 154.50 G magnetic field. The coloured lines indicate a fit to the results, following the model given in Eq. 3. We find $\alpha^{(2)}/(4\pi\epsilon_0) = 545(4)\,a_0^3$, and $\delta_0 = 983.0(2)\,\mathrm{Hz}$. **(d)** Differential polarisability between the states as a function of magnetic field, measured for $\beta = 0^\circ$. The line is the expectation from the molecular Hamiltonian described in Methods. **(e)** δ measured in a single beam of the dipole trap with fixed intensity $I = 15.3\,\mathrm{kW\,cm^{-2}}$, as a function of β. The dashed horizontal line indicates δ_0. The solid line is calculated using Eq.3 with the parameters found in (c). All error bars indicate 1σ uncertainties.

FIG. 3. **Robust coherence of the storage qubit.** Ramsey experiment with the qubit states $|0\rangle \equiv (0,4)_1$ and $|1\rangle \equiv (0,3)_0$, using the optimal configuration $B \approx 154.50\,\mathrm{G}$, $\beta \approx 55^\circ$. The y-axis indicates the number of molecules remaining in $|0\rangle$, following the Ramsey sequence shown inset, as a function of the hold time T. The shaded region in the upper plot indicates the maximum and minimum of the Ramsey fringes as a function of time; the spacing of the fringes is too small to plot at this scale. The lower plots show the Ramsey fringes observed at 400 ms intervals.