

Supplementary information: Parity-doublet spin interactions in ultracold polyatomic molecules

Junheng Tao,^{1,2} Paige Robichaud,^{1,2} Christian Hallas,^{1,2} Giseok Lee,^{1,2} and John M. Doyle^{1,2}

¹*Department of Physics, Harvard University, Cambridge, MA 02138, USA*

²*Harvard-MIT Center for Ultracold Atoms, Cambridge, MA 02138, USA*

(Dated: August 3, 2026)

S1. DIPOLAR EXCHANGE INTERACTION

The two stretched parity-doublet states $|\uparrow\rangle \equiv |N=1, J=3/2, F=2, m_F=2, p=+\rangle$ and $|\downarrow\rangle \equiv |N=1, J=3/2, F=2, m_F=2, p=-\rangle$ define a pseudospin-1/2, with $s_i^z = (|\uparrow\rangle_i\langle\uparrow| - |\downarrow\rangle_i\langle\downarrow|)/2$, raising and lowering operators $s_i^+ = |\uparrow\rangle_i\langle\downarrow|$, $s_i^- = (s_i^+)^{\dagger}$, and transverse components $s_i^x = (s_i^+ + s_i^-)/2$, $s_i^y = -i(s_i^+ - s_i^-)/2$. The two states share all angular-momentum quantum numbers except parity and are split by the parity-doublet gap Δ as shown in Fig. 1c.

Parity forbids a permanent dipole in either state, $\langle\uparrow|\mathbf{d}|\uparrow\rangle = \langle\downarrow|\mathbf{d}|\downarrow\rangle = 0$, so at zero external field the molecules carry no lab-frame dipole moment. The two states are instead connected by a transition dipole $d_{\uparrow\downarrow} = \langle\uparrow|\mathbf{d}|\downarrow\rangle = 0.73$ D. Because $|\uparrow\rangle$ and $|\downarrow\rangle$ share the same projection $m_F = 2$, the Wigner-Eckart theorem ($\Delta m_F = 0$) allows only the space-fixed $q = 0$ (i.e. z) spherical component of $\mathbf{d}$ between them. The dipole operator projected onto the manifold is therefore off-diagonal in the parity basis and aligned with the quantization axis,

$$\hat{\mathbf{d}}_i \longrightarrow d_{\uparrow\downarrow} \hat{\mathbf{z}} (|\uparrow\rangle_i\langle\downarrow| + |\downarrow\rangle_i\langle\uparrow|) \propto 2d_{\uparrow\downarrow} s_i^x \hat{\mathbf{z}}. \quad (\text{S1})$$

Two molecules separated by $\mathbf{r}$ interact through the electric dipole-dipole Hamiltonian

$$H_{\text{dd}} = \frac{1}{4\pi\epsilon_0 r^3} [\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2 - 3(\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{r}})(\hat{\mathbf{d}}_2 \cdot \hat{\mathbf{r}})]. \quad (\text{S2})$$

With $\hat{\mathbf{z}} \cdot \hat{\mathbf{r}} = \cos \alpha$, where α is the angle between the intermolecular axis and the quantization field, projecting both dipoles into the parity-doublet manifold gives

$$\hat{H}_{\text{dd}}^{(ij)} = \frac{d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 r_{ij}^3} (1 - 3\cos^2 \alpha_{ij}) (s_i^+ + s_i^-) (s_j^+ + s_j^-). \quad (\text{S3})$$

After the rotating wave approximation

$$\hat{H}_{\text{dd,RWA}}^{(ij)} = \frac{d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 r_{ij}^3} (1 - 3\cos^2 \alpha_{ij}) (s_i^+ s_j^- + s_i^- s_j^+). \quad (\text{S4})$$

Using $s_i^+ s_j^- + s_i^- s_j^+ = 2(s_i^x s_j^x + s_i^y s_j^y)$, the interaction becomes $\hat{H}_{\text{XY}} = \frac{1}{2} \sum_{i \neq j} V_{ij} (s_i^x s_j^x + s_i^y s_j^y)$ with

$$V_{ij} = \frac{2d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 r_{ij}^3} (1 - 3\cos^2 \alpha_{ij}). \quad (\text{S5})$$

S2. COLLISIONAL STUDY

S2.1. Collision Hamiltonian and basis

The parity-doublet states are the opposite-parity stretched states $|\uparrow\rangle = |N=1, J=3/2, F=2, m_F=2, p=+\rangle$, and $|\downarrow\rangle = |N=1, J=3/2, F=2, m_F=2, p=-\rangle$ with splitting $\nu_{\text{pd}} = 43$ MHz. At zero dc electric field, parity forbids a diagonal laboratory-frame dipole moment. Within the qubit manifold, the projected dipole operator is

$$P\hat{\mathbf{d}}_iP = d_{\uparrow\downarrow} \hat{\mathbf{z}} \sigma_i^x, \quad d_{\uparrow\downarrow} = 0.73 \text{ D}. \quad (\text{S6})$$

The projected two-molecule interaction is therefore

$$\hat{V}_{\text{dd}}(R, \theta) = \frac{d_{\uparrow\downarrow}^2}{4\pi\epsilon_0 R^3} (1 - 3\cos^2 \theta) \sigma_1^x \sigma_2^x. \quad (\text{S7})$$

In the Ramsey experiment, the initial state is symmetric upon particle exchange. Hence we choose the symmetric internal basis

$$|q\rangle \in \{|\uparrow\rangle, |T_0\rangle, |\downarrow\rangle\},$$

where

$$|\uparrow\rangle = |\uparrow\uparrow\rangle, \quad |T_0\rangle = \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}}, \quad |\downarrow\rangle = |\downarrow\downarrow\rangle.$$

In this basis,

$$\sigma_1^x \sigma_2^x|_{\text{sym}} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix} \equiv X.$$

Taking the threshold of the mixed pair state as the zero of energy gives $\epsilon_q/h = (+\nu_{\text{pd}}, 0, -\nu_{\text{pd}})$.

The channel basis is

$$|q; LM_L\rangle. \quad (\text{S8})$$

As the internal pair state is symmetric, and CaOH is bosonic, only even partial waves occur. In atomic units, the channel Hamiltonian is

$$H_{qL,q'L'}^{(M_L)}(R) = \left[\epsilon_q + \frac{L(L+1)}{2\mu R^2} \right] \delta_{qq'} \delta_{LL'} + \frac{d_{\uparrow\downarrow}^2}{R^3} X_{qq'} A_{LL'}^{(M_L)}, \quad (\text{S9})$$

with reduced mass $\mu = 28.5$ u, and angular matrix

$$A_{LL'}^{(M)} = \int d\Omega Y_{LM}^*(\Omega) (1 - 3\cos^2\theta) Y_{L'M}(\Omega)$$

that couples L and $L \pm 2$. We also define the dipole length and energy

$$a_d = \mu d_{\uparrow\downarrow}^2 = 4.29 \times 10^3 a_0, \quad E_d/k_B = 0.165 \text{ } \mu\text{K}. \quad (\text{S10})$$

The large dipole length motivates both the high partial-wave cutoff and a large matching radius.

S2.2. Coupled-channel scattering calculation

For total energy E_{tot} , the matrix of radial solutions $F(R)$ obeys

$$\left[-\frac{1}{2\mu} \frac{d^2}{dR^2} \mathbf{I} + \mathbf{H}(R) - E_{\text{tot}} \mathbf{I} \right] \mathbf{F}(R) = 0 \quad (\text{S11})$$

where $\mathbf{H}(R)$ is the channel Hamiltonian. We propagate the logarithmic derivative

$$Y(R) = F'(R)F^{-1}(R) \quad (\text{S12})$$

from the inner radius $R_0 = 30a_0$ to the asymptotic matching radius $R_m = 10^4 a_0$. These radii follow the numerical choices used in Ref. [1].

Similar to Ref. [1], we impose a universal loss short-range boundary condition at R_0 [2]. To do this, we diagonalize the channel Hamiltonian at

$$\mathbf{H}(R_0)\mathbf{U} = \mathbf{U} \text{diag}(U_\alpha). \quad (\text{S13})$$

The corresponding local radial wave numbers are

$$k_\alpha = [2\mu(E_{\text{tot}} - U_\alpha)]^{1/2}. \quad (\text{S14})$$

For a locally open channel, a wave traveling from large to small has the form

$$F_\alpha(R) \propto \exp[-ik_\alpha(R - R_0)]. \quad (\text{S15})$$

Its logarithmic derivative at the inner boundary is therefore $-ik_\alpha$. Transforming this condition back to the asymptotic channel basis gives $\mathbf{Y}(R_0) = \mathbf{U} \text{diag}(-ik_\alpha) \mathbf{U}^\dagger$. For locally closed channels, the branch is chosen with

$$\text{Re } k_\alpha \geq 0, \quad \text{Im } k_\alpha \geq 0. \quad (\text{S16})$$

S2.3. Asymptotic matching

At R_m the dipolar couplings are negligible compared with the channel kinetic energies. We first partition $\mathbf{Y}$ into open and closed blocks and eliminate the asymptotically closed channels. In the remaining open subspace $\tilde{\mathbf{Y}}_{oo}$, let $\mathbf{H}^-(R)$ and $\mathbf{H}^+(R)$ denote diagonal matrices of incoming and outgoing spherical Riccati-Hankel functions. A scattering solution can be written as

$$\mathbf{F}_o(R) = \mathbf{H}^-(R) - \mathbf{H}^+(R)\mathbf{S}^{(q)}. \quad (\text{S17})$$

Matching the logarithmic derivatives

$$\mathbf{S}^{(q)} = \left[\mathbf{H}^{+'} - \tilde{\mathbf{Y}}_{oo}\mathbf{H}^+ \right]^{-1} \left[\mathbf{H}^{-'} - \tilde{\mathbf{Y}}_{oo}\mathbf{H}^- \right]. \quad (\text{S18})$$

Primes denote radial derivatives. Its flux-normalized matrix element

$$S_{q'L'M_L, qLM_L}^{(q)} \quad (\text{S19})$$

is the amplitude for an incoming channel (q, L, M_L) to emerge in the open channel (q', L', M_L) . Conservation of M_L is written explicitly. The scattering matrix would be unitary if every outgoing channel were included and no short-range loss were present. Its missing norm is the probability of short-range capture.

S2.4. Partial-wave sums, cross sections, and rate coefficients

We include every even incident and outgoing partial wave satisfying

$$|M_L| \leq L \leq L_{\max}. \quad (\text{S20})$$

The calculations for $\pm M_L$ are degenerate so we propagate only nonnegative projections and account for the degeneracy using

$$g_M = \begin{cases} 1, & M_L = 0, \\ 2, & M_L > 0. \end{cases} \quad (\text{S21})$$

For an incident channel $i = (q, L, M_L)$, the probability missing from the open-channel scattering matrix is

$$P_i^{\text{cap}} = 1 - \sum_{f \in \text{open}} |S_{fi}|^2. \quad (\text{S22})$$

The probability for inelastic scattering into a different retained internal pair channel, excluding short-range loss, is

$$P_i^{\text{inel}} = \sum_{\substack{f \in \text{open} \\ q_f \neq q}} |S_{fi}|^2. \quad (\text{S23})$$

The initial product Ramsey state has pair-state populations

$$w_{\uparrow} : w_{T_0} : w_{\downarrow} = \frac{1}{4} : \frac{1}{2} : \frac{1}{4}, \quad (\text{S24})$$

the population-averaged cross sections are

$$\sigma_p(E_c) = \frac{2\pi}{k^2} \sum_q w_q \sum_{M_L \geq 0} g_M \sum_{\substack{L \text{ even} \\ L \geq M_L}} P_{qLM_L}^p(E_c), \quad p \in \{\text{cap}, \text{inel}\}. \quad (\text{S25})$$

Multiplying a cross section by the relative speed

$$v(E_c) = \frac{\hbar k}{\mu} = \sqrt{\frac{2E_c}{\mu}} \quad (\text{S26})$$

gives the corresponding two-body rate coefficient,

$$K_p(E_c) = v(E_c)\sigma_p(E_c). \quad (\text{S27})$$

These quantities use only probabilities and are sufficient for short-range capture and inelastic rate coefficients within the retained basis. Ramsey coherence is phase sensitive: it requires the complex scattering amplitudes themselves, as described next.

S2.5. Scattering map

A total elastic cross section measures how much outgoing flux differs from an unscattered plane wave. Ramsey contrast is reduced only if the outgoing motion, final internal state, or loss reservoir contains information about which component of the incoming spin superposition was present. We therefore construct an internal-state map from the complex scattering amplitudes while retaining the labels of the unobserved outgoing motional channels.

Consider an incoming internal pair state

$$|\chi\rangle = \sum_q c_q |q\rangle \quad (\text{S28})$$

and an incident orbital channel $a = (L, M_L)$. Each component enters with the same kinetic energy E_c .

The amplitude that contributes to an integrated scattering cross section is proportional to the transition matrix

$$\mathbf{T}^{(q)} = \mathbf{I} - \mathbf{S}^{(q)}. \quad (\text{S29})$$

For an incoming channel, the scattered state can be organized as

$$|\Psi_{\text{sc}}^{(a)}\rangle = \sum_{\lambda} [A_{\lambda a}(E_c)|\chi\rangle] \otimes |\lambda\rangle. \quad (\text{S30})$$

The label

$$\lambda = (E_{\text{out}}, L', M_L) \quad (\text{S31})$$

specifies an unobserved outgoing motional channel: its kinetic energy, outgoing partial wave, and conserved orbital projection.

The cross-section-weighted operator acting on the internal pair space is

$$[A_{\lambda a}(E_c)]_{q'q} = \left(\frac{2\pi g_M}{k^2}\right)^{1/2} \left[\delta_{q'q} \delta_{L'L} - S_{q'L',qL}^{(q)}(E_c) \right] \chi_{\lambda}(E_c + \epsilon_q - \epsilon_{q'}). \quad (\text{S32})$$

The indicator enforcing equality of the outgoing kinetic energy and orbital channel is

$$\chi_{\lambda}(E) = \begin{cases} 1, & E = E_{\text{out}} \text{ and the outgoing orbital state is } (L', M_L), \\ 0, & \text{otherwise.} \end{cases} \quad (\text{S33})$$

In the simulation, channels are placed in the same energy group when their outgoing kinetic energies agree. Eq. (S32) specifies which amplitudes must be added coherently. For elastic scattering, $E_{\text{out}} = E_c$ for every component of the initial superposition. Those amplitudes therefore lead to the same outgoing motional state and must be combined coherently inside one operator $A_{\lambda a}$. By contrast, amplitudes with different outgoing kinetic energies or orbital states multiply orthogonal motional states and are combined only after tracing over motion.

Tracing over the unresolved outgoing motion gives the scattered contribution to the internal pair density matrix,

$$\Phi_a^{\text{sc}}[\rho] = \sum_{\lambda} A_{\lambda a} \rho A_{\lambda a}^{\dagger}. \quad (\text{S34})$$

Here, ρ is the incoming internal pair density matrix, and Φ_a^{sc} denotes the scattered-state map for incident orbital channel a .

S2.6. Lindblad dissipator and short-range loss

During a short interval dt , the probability for a molecule to encounter partners in a cross section σ is

$$n v(E_c) \sigma dt, \quad (\text{S35})$$

where n is the local density. Combining the jump map in Eq. (S34) with the corresponding no-jump amplitude gives

$$\rho(t + dt) - \rho(t) = n dt \mathcal{D}_{E_c}[\rho] + O(dt^2), \quad (\text{S36})$$

where

$$\mathcal{D}_{E_c}[\rho] = v(E_c) \sum_{a,\lambda} \left[A_{\lambda a} \rho A_{\lambda a}^\dagger - \frac{1}{2} \left\{ A_{\lambda a}^\dagger A_{\lambda a}, \rho \right\} \right]. \quad (\text{S37})$$

A complete scattering master equation also contains coherent forward scattering. We denote by

$$W_{\text{fs}}(E_c) \quad (\text{S38})$$

the Hermitian pair-spin operator obtained from the real part of the zero-angle scattering amplitude. Its contribution is

$$\left. \frac{d\rho}{dt} \right|_{\text{fs}} = -\frac{i}{\hbar} [n W_{\text{fs}}(E_c), \rho]. \quad (\text{S39})$$

This term produces a coherent density-dependent phase or frequency shift. We do not include it in the reported Ramsey contrast-decay rate coefficient, because coherent long-range spin evolution is already propagated by DTWA. The dissipator in Eq. (S37) is also not added to the DTWA rate: in the weak-deflection regime, averaging spin-dependent scattering phases over unresolved outgoing channels and averaging the same interaction over sampled classical trajectories are overlapping descriptions.

The open-channel matrix is subunitary because the inner boundary absorbs flux. We define a matrix of short-range loss cross sections Σ_{loss} , whose diagonal element is the total capture cross section for an incoming internal pair state

$$[\Sigma_{\text{loss}}]_{qq} = \sum_a \frac{2\pi g_M}{k^2} \left[1 - \sum_{f \in \text{open}} \left| S_{f,qa}^{(q)} \right|^2 \right]. \quad (\text{S40})$$

The capture contribution to the unnormalized surviving pair state is

$$\mathcal{L}_{E_c}^{\text{cap}}[\rho] = -\frac{v(E_c)}{2} \{ \Sigma_{\text{loss}}, \rho \}. \quad (\text{S41})$$

The energy-resolved evolution used to calculate the reported initial decay is therefore

$$\left. \frac{d\rho}{dt} \right|_{E_c} = n [\mathcal{D}_{E_c}[\rho] + \mathcal{L}_{E_c}^{\text{cap}}[\rho]]. \quad (\text{S42})$$

S2.7. Ramsey contrast decay

The initial pair state is

$$|\psi_0\rangle = | + x, + x \rangle = \frac{1}{2} \left(| \uparrow \uparrow \rangle + \sqrt{2} | T_0 \rangle + | \downarrow \downarrow \rangle \right), \quad \rho_0 = |\psi_0\rangle \langle \psi_0|. \quad (\text{S43})$$

Write the surviving pair density matrix in the ordered basis $(\uparrow, T_0, \downarrow)$, and tracing over the second molecule gives

$$\rho^{(1)} = \text{Tr}_2 \rho = \begin{pmatrix} \rho_{\uparrow\uparrow} + \rho_{T_0 T_0}/2 & (\rho_{\uparrow T_0} + \rho_{T_0 \downarrow})/\sqrt{2} \\ (\rho_{T_0 \uparrow} + \rho_{\downarrow T_0})/\sqrt{2} & \rho_{\downarrow\downarrow} + \rho_{T_0 T_0}/2 \end{pmatrix}. \quad (\text{S44})$$

Its trace

$$N_s = \text{Tr} \rho^{(1)} = \text{Tr} \rho \quad (\text{S45})$$

is the surviving norm. The normalized Ramsey contrast along x is

$$C_x = \frac{\text{Tr} [\sigma_x \rho^{(1)}]}{N_s} = \frac{2 \text{Re} [\rho_{\uparrow\downarrow}^{(1)}]}{\text{Tr} \rho^{(1)}}. \quad (\text{S46})$$

Differentiating the observable

$$\frac{dC_x}{dt} = \frac{2 \operatorname{Re} [\dot{\rho}_{\uparrow\downarrow}^{(1)}]}{N_s} - C_x \frac{\dot{N}_s}{N_s}. \quad (\text{S47})$$

The second term is essential when loss is present. Take $t = 0$

$$\left. \frac{dC_x}{dt} \right|_{t=0} = 2 \operatorname{Re} [\dot{\rho}_{\uparrow\downarrow}^{(1)}]_{t=0} - \dot{N}_s(0). \quad (\text{S48})$$

The familiar phase-shift result is recovered as a special limit. Suppose the scattering matrix is diagonal in all pair-spin channels, there is no loss, and only one outgoing motional state is relevant

$$S_q = e^{2i\delta_q}. \quad (\text{S49})$$

where δ_q is the real elastic scattering phase shift in pair channel q . Directly applying these phases and tracing over one molecule gives

$$C'_x = \cos(\delta_\downarrow - \delta_\uparrow) \cos(\delta_\downarrow + \delta_\uparrow - 2\delta_{T_0}). \quad (\text{S50})$$

When the two aligned channels have the same phase, $\delta_\uparrow = \delta_\downarrow \equiv \delta_\parallel$, the contrast change per collision becomes

$$\Delta C = 1 - C'_x = 2 \sin^2(\delta_\parallel - \delta_{T_0}). \quad (\text{S51})$$

This is the single-phase-difference expression used in simplified collision models [3, 4]. The full scattering map is required here because the CaOH scattering matrix can couple orbital and internal channels, so a single diagonal phase shift is not sufficient.

Finally, we define the energy-resolved initial Ramsey contrast-decay rate coefficient

$$K_R(E_c) = -\frac{1}{nC_x(0)} \left. \frac{dC_x}{dt} \right|_{t=0, E_c}. \quad (\text{S52})$$

S2.8. Thermal and trap averaging

For a nondegenerate three-dimensional gas, the relative collision-energy distribution is

$$p_T(E) = \frac{2}{\sqrt{\pi}} \frac{\sqrt{E}}{(k_B T)^{3/2}} \exp\left(-\frac{E}{k_B T}\right). \quad (\text{S53})$$

The thermal rate coefficients are

$$\langle K_p \rangle_T = \int_0^\infty dE p_T(E) K_p(E), \quad p \in \{R, \text{cap}, \text{inel}\}. \quad (\text{S54})$$

Energy-correlated quantities are averaged before any factorization. For example,

$$\langle K \Delta C \rangle_T = \int_0^\infty dE p_T(E) K(E) \Delta C(E), \quad (\text{S55})$$

For a Gaussian density profile, the initial density experienced by a randomly selected molecule is

$$\bar{n} = \frac{\int d^3\mathbf{r} n^2(\mathbf{r})}{\int d^3\mathbf{r} n(\mathbf{r})} = \frac{n_0}{2^{3/2}}, \quad (\text{S56})$$

where n_0 is the peak density. The initial cloud-averaged rate is therefore

$$\Gamma_p(0) = \bar{n} \langle K_p \rangle_T. \quad (\text{S57})$$

S2.9. Simulations and results

We evaluated the scattering calculation described above without fitting any collision parameter to the Ramsey data. For each collision energy E_c , we propagated the three distinct incoming internal thresholds $q \in \{\uparrow, T_0, \downarrow\}$ in every M_L block. Within each block, all even incident and outgoing partial waves satisfying $|M_L| \leq L \leq L_{\max}$ were included. A single propagation at fixed q , M_L , and E_c supplies all incident- L columns of that block.

The fundamental radial solution and its derivative were integrated on 120 equal radial segments between $R_0 = 30a_0$ and $R_m = 10^4 a_0$. Within each segment we used an eighth-order adaptive Dormand-Prince integrator with relative and absolute tolerances 10^{-8} and 10^{-10} , respectively.

Closed-channel solutions contain exponentially growing components that can make the fundamental solution matrix $\mathbf{F}$ ill-conditioned. After each radial segment, we factorize $\mathbf{F} = \mathbf{Q}\mathbf{R}$ and replace $(\mathbf{F}, \mathbf{F}')$ by $(\mathbf{Q}, \mathbf{F}'\mathbf{R}^{-1})$. This leaves the log derivative invariant $(\mathbf{F}'\mathbf{R}^{-1})(\mathbf{F}\mathbf{R}^{-1})^{-1} = \mathbf{F}'\mathbf{F}^{-1}$, while restoring orthonormal columns and preventing exponentially growing closed-channel components from erasing the numerically smaller independent solutions. The calculation used the parameters in Table S1.

Parameters	Values
reduced mass (μ)	28.5 u
parity-doublet splitting (ν_{pd})	43 MHz
transition dipole	0.73 D
inner boundary (R_0)	$30 a_0$
matching radius (R_m)	$10^4 a_0$
radial QR segments	120
thermal partial waves ($L_{\max}$)	20
collision-energy grid	12 logarithmic points, 2–600 μK
temperature	60 μK
short-range boundary	universal absorption loss

TABLE S1. Scattering calculation parameters.

For each energy, the complex amplitudes were assembled into the operators $A_{\lambda a}$. We grouped amplitudes coherently only when they had the same outgoing kinetic energy, L' , and M_L . The resulting dissipator was applied to the initial pair state, and the derivative of the normalized one-body contrast was evaluated.

With these we obtain

$$\langle K_R \rangle_T = 1.67 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$$

and

$$\langle K_{\text{cap}} \rangle_T = 4.83 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}.$$

The direct internal-state-changing coefficient among the parity-doublet channels is approximately

$$\langle K_{\text{inel}} \rangle_T \simeq 9.5 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}.$$

The Ramsey coefficient arises almost entirely from spin-dependent elastic scattering where different spin components produce distinguishable outgoing motional states. The direct inelastic collision contribution is negligible.

The collision model and DTWA rates agree closely within uncertainty, as shown in Fig. S1. The predicted molecule number loss rate is also close to the experimental observations and that in Ref. [1]. This is an important consistency check, but it does not identify an additional decay channel that should be added to DTWA. Both calculations use the same $1/r^3$ parity-doublet interaction and the same thermal relative motion. DTWA samples the coherent spin phase along classical trajectories and averages over those trajectories. The scattering calculation encodes the same two-body evolution in asymptotic motional channels and traces over the outgoing motion. In the present weak-deflection semiclassical regime, these are overlapping descriptions of long-range dipolar dephasing.

For illustration, adding the two interaction-induced rates would substantially exceed either independent calculation as well as experimental data. We therefore use DTWA as the many-body prediction in the main text and the collision model as an independent two-body cross-check. A combined calculation would require a matched kinetic treatment that separates the coherent self-energy from the dissipative collision integral, or a joint propagation of spin and motion.

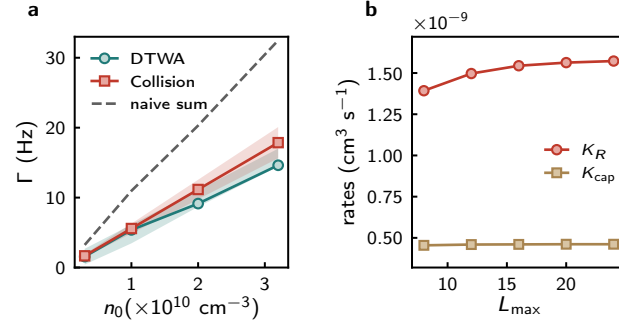


FIG. S1. Two-body scattering prediction for Ramsey contrast decay and numerical convergence. **a** Interaction-induced Ramsey decay rate Γ versus peak molecular density n_0 . Green circles show the many-body DTWA prediction, and red squares show the initial rate obtained from the two-body collision after thermal averaging. Shaded bands denote the propagated uncertainties of the respective calculations. The grey dashed line is their naive sum, which substantially overpredicts the decay; The two approaches contain overlapping descriptions of long-range dipolar dephasing. **b** Partial-wave convergence at $E_c/k_B = 60 \mu\text{K}$ of the Ramsey-decay coefficient K_R and number loss capture coefficient K_{cap} . The Ramsey coefficient approaches convergence with increasing L_{max} , while K_{cap} is stable.

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