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Microwave Shielding of Ultracold Polar Molecules

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We use microwaves to engineer repulsive long-range interactions between ultracold polar molecules. The resulting shielding suppresses various loss mechanisms and provides large elastic cross sections. Hyperfine interactions limit the shielding under realistic conditions, but a magnetic field allows suppression of the losses to below 10^{-14} cm^3 s^{-1}. The mechanism and optimum conditions for shielding differ substantially from those proposed by Gorshkov *et al.* [Phys. Rev. Lett. **101**, 073201 (2008)], and do not require cancellation of the long-range dipole-dipole interaction that is vital to many applications.

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A variety of polar molecules have now been produced at [1–6], or cooled down to [7–9], ultracold temperatures. Potential applications include quantum simulation [10,11], quantum computing [12,13], and the creation of novel quantum phases [14,15]. All these applications require high densities, where collisional losses become important. Even chemically stable molecules in their absolute ground state, which possess no two-body loss mechanisms, may undergo short-range three-body loss that is amplified by long-lived two-body collisions [16–18]. Short-range losses have been suppressed experimentally for fermionic molecules by a combination of strong electric fields and confinement [19]. However, this approach is not feasible for bosons [20]. In this Letter, we use microwaves to engineer repulsive long-range interactions that shield molecular collisions. Our approach does not require confinement to two dimensions as in Refs. [14,21], and can be applied to both bosonic and fermionic species.

Figure 1(a) shows the shielding mechanism schematically in the low-intensity limit. Microwave radiation is blue detuned by Δ from the $n = 0 \rightarrow 1$ rotational transition of the molecule. The field-dressed state with one molecule rotationally excited ($n = 1$) is energetically below the bare state with both molecules in the ground state ($n = 0$) by $\hbar\Delta$. The resonant dipole-dipole interaction splits the lower threshold into repulsive $|K| = 1$ and attractive $K = 0$ states. Here, K is the projection of the rotational angular momentum onto the intermolecular axis, which is a good quantum number when Coriolis and field-dependent couplings are neglected. The repulsive $K = 1$ states cross the bare ground state at the Condon point, which moves inwards as Δ increases. This crossing is avoided by $2\hbar\Omega$, where Ω is the Rabi frequency. The upper adiabatic curve is repulsive and provides shielding. This is closely analogous to optical blue shielding for atoms [22].

Microwave-dressed molecules typically have weaker resonant dipole interactions than optically dressed atoms and need larger values of Ω for optimum shielding. For high intensities, the individual monomer states are even and odd linear combinations $|\pm\rangle$ of the field-dressed states $|g\rangle = |0, 0, 0\rangle$ and $|g\rangle = |1, 1, -1\rangle$, with energies $\pm\hbar\Omega$. In the ket $|n, m_n, N\rangle$, N is the number of photons of σ^+ polarization and m_n is the projection of n onto the microwave propagation axis. There are also dark states $|0\rangle$

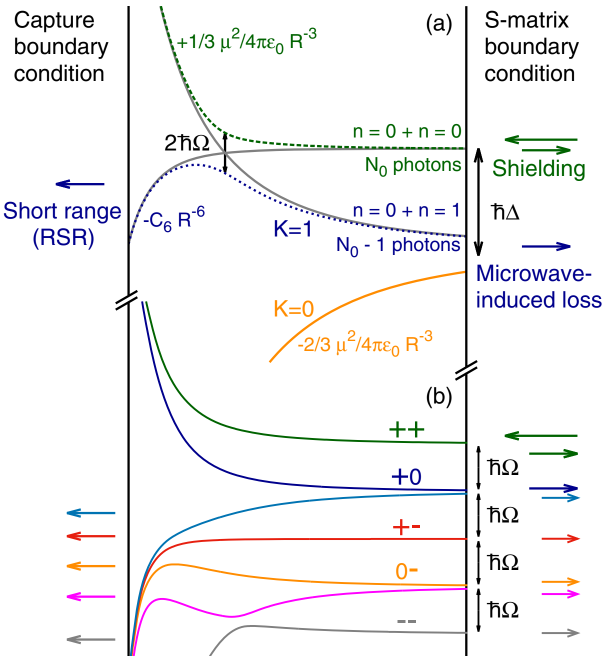

FIG. 1. Schematic representation of the potential curves relevant to microwave shielding. Panels (a) and (b) correspond to $\Omega \ll \Delta$ and $\Omega \gg \Delta$, respectively. The boundary conditions imposed in the coupled-channels calculations are indicated. Green arrows indicate incoming and elastically scattered flux, whereas the remaining arrows on the right- and left-hand sides indicate microwave-induced loss and reaching short range (RSR), respectively.

corresponding to $|1, 0, -1\rangle$ and $|1, -1, -1\rangle$. This produces five thresholds separated by approximately $\hbar\Omega$, as shown in Fig. 1(b). The top adiabatic curve is again repulsive and provides shielding.

Our goal is to find conditions, Ω and Δ, under which the collision dynamics is adiabatic and follows the repulsive shielding potential. We calculate the potential curves and couplings from a Hamiltonian that describes the molecules as rigid rotors interacting by dipole-dipole interactions. It also includes end-over-end rotation of the molecular pair (not included above) and interactions with electric, magnetic, and microwave fields, with hyperfine interactions where appropriate. We use a basis set consisting of symmetrized products of spherical harmonics for the rotation of both molecules and the end-over-end rotation, together with electron and nuclear spin states. A full description of the Hamiltonian and examples of the resulting adiabatic curves are given in the Supplemental Material [23].

We perform numerically exact coupled-channels scattering calculations of two different types of loss. The coupled-channels approach is essential, because semiclassical approximations such as Landau-Zener break down when the local wavelength is large compared to the width of the crossing. We propagate two sets of linearly independent solutions of the coupled-channels equations, using the renormalized Numerov method [35], and apply both capture boundary conditions at short range and S-matrix boundary conditions at long range [36–38]. We calculate the probability of reaching short range (RSR) and the corresponding rate coefficient. There is evidence that flux that reaches short range is lost with high probability, even for nonreactive molecules [18]. In addition, some of the reflected flux is lost, for example, by absorbing a microwave photon, accompanied by kinetic energy release. We also calculate the probabilities and rates for this microwave-induced loss. These two types of loss are illustrated in Fig. 1. The remaining flux is shielded and scatters elastically.

Figure 2 shows the probabilities and rates for RSR and microwave-induced loss as a function of Δ and Ω. This calculation is for RbCs + RbCs collisions in the presence of circularly polarized (σ^+) microwaves, without static fields or hyperfine interactions. For large Ω and comparable or smaller Δ, the probabilities for both RSR and microwave-induced loss are small, indicating that shielding is effective. Loss rates below 10^{-14} cm^3 s^{-1} can be achieved for feasible values of Ω; such rates are low enough to allow lifetimes of several seconds at densities that are high enough for Bose-Einstein condensation. Shielding is ineffective for linearly polarized microwaves, as shown in the Supplemental Material [23].

Microwave shielding of polar molecules is ineffective for $\Omega \ll \Delta$. This contrasts with blue shielding for ultracold atoms, and arises both because of the smaller transition dipoles for typical molecules and because of the strong rotational dispersion interaction. For $\Omega \gtrsim \Delta$ there is substantial state mixing even for the separated molecules, and the molecules must be prepared in the upper field-dressed state. This may be done either by forming molecules directly in the upper state by Stimulated Raman adiabatic passage or by switching on the microwave field adiabatically. For a linear intensity ramp, switching on the microwaves over 1 ms for $\Omega = 10$ MHz and $\Delta = 1$ MHz retains 99% in the upper adiabatic state, as described in the Supplemental Material [23]. Considerably shorter times may be achieved with ramps that are slower at low intensity.

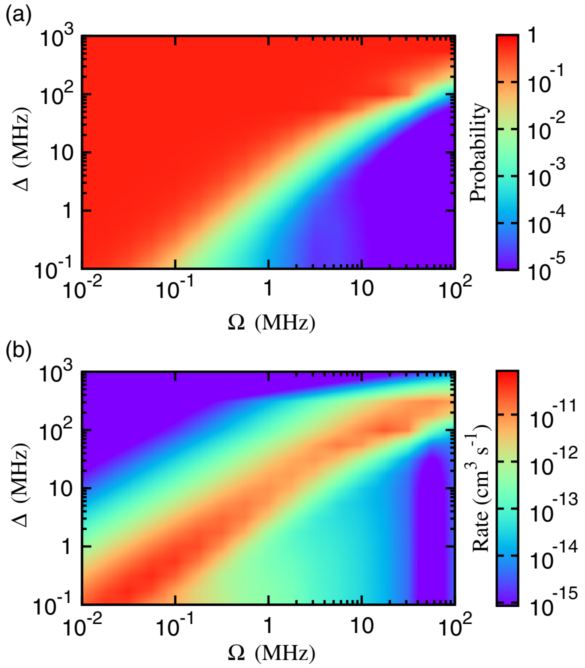

FIG. 2. Probability for RSR (a) and microwave-induced loss rate (b), as a function of Δ and Ω, for RbCs + RbCs collisions in circularly polarized microwaves, without static fields or hyperfine interactions. The color codings for probability and loss rate are equivalent and can be used to read either panel.

For ultracold collisions, the strong dependence of the scattering length on the position of the least-bound state usually precludes *ab initio* calculation of elastic cross sections σ_{el} [39,40]. In the presence of shielding, however, the molecules never experience the inaccurately known short-range interactions, and the calculated σ_{el} is quantitatively predictive. For RbCs molecules, shielded as above with $\Delta = 1$ MHz and $\Omega = 10$ MHz, we obtain $\sigma_{\mathrm{el}} = 3.6 \times 10^{-10}$ cm^2. This is large compared to the typical value expected for unshielded RbCs molecules, which is $4\pi\bar{a}^2 = 1.8 \times 10^{-11}$ cm^2. Here, $\bar{a}$ is the mean scattering length [41] that accounts for the rotational dispersion interaction. The combination of large elastic and suppressed inelastic cross sections may allow evaporative cooling of microwave-shielded polar molecules.

Next, we consider the effect of hyperfine interactions. These can cause losses for molecules that are not present for atoms, because atomic hyperfine splittings are much

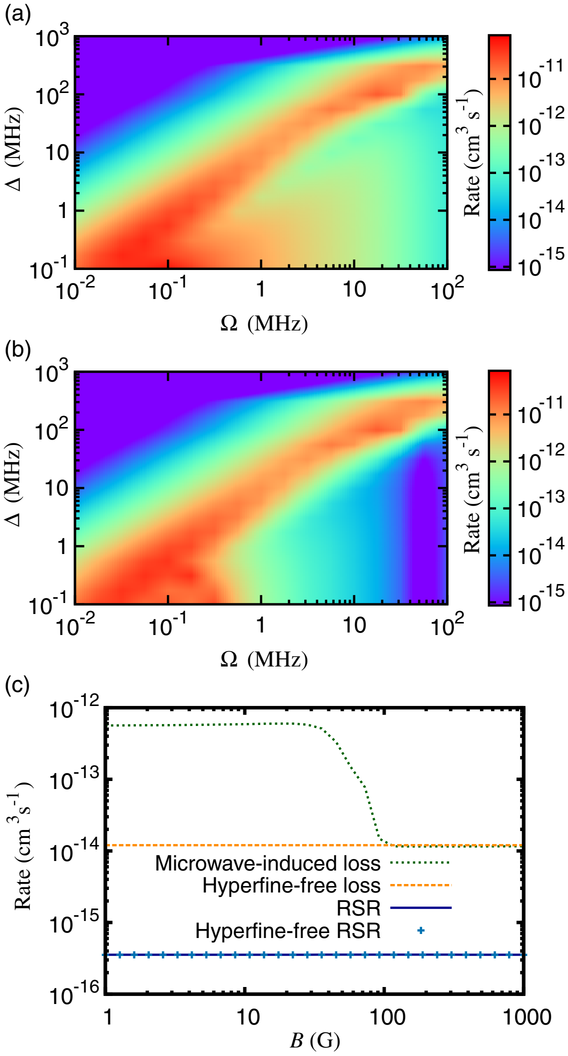

FIG. 3. Shielding of collisions of RbCs molecules in the spin-stretched state by circularly polarized microwaves, including hyperfine interactions. Panels (a) and (b) show the microwave-induced loss rate in 0 and 200 G magnetic fields, respectively. Panel (c) shows the dependence of the RSR and microwave-induced loss rates on the magnetic field for fixed $\Omega = 20$ MHz and $\Delta = 1$ MHz.

larger than Ω and Δ. We carry out coupled-channel calculations in a full basis set including nuclear spin functions [42]. Initially, we consider ^{87}Rb^{133}Cs molecules in the spin-stretched $f = 5$, $m_f = 5$ state for $n = 0$, which can be produced and trapped experimentally [2,3]. This state has the advantage that there is only one allowed microwave transition for σ^+ polarization, to the spin-stretched $f = 6$, $m_f = 6$ rotationally excited $n = 1$ state [43]. At low magnetic fields, the additional channels resulting from hyperfine coupling produce greater microwave-induced loss, as can be seen in Fig. 3(a). However, a magnetic field of 200 G parallel to the microwave propagation axis recovers the effective shielding obtained in the hyperfine-free case, as shown in Fig. 3(b). The transition between the low-field and high-field regimes is shown in Fig. 3(c) for fixed $\Omega = 20$ and $\Delta = 1$ MHz. The rate for RSR is small, as in the hyperfine-free case.

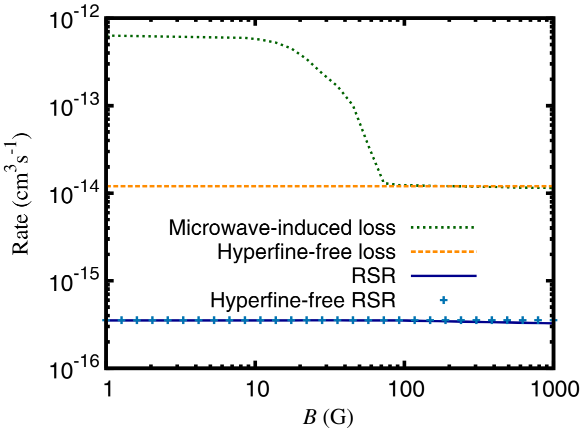

FIG. 4. The dependence of the microwave-induced loss on the magnetic field for fixed $\Omega = 20$ MHz and $\Delta = 1$ MHz, for collisions of RbCs molecules in the non-spin-stretched $f = 4$, $m_f = 4$ state, including hyperfine interactions.

The spin-stretched state becomes the absolute ground state at magnetic fields above 90 G. However, this is not a necessary or a sufficient condition for effective shielding. Figure 4 shows the microwave-induced loss for the non-spin-stretched $f = 4$, $m_f = 4$ state of RbCs as a function of magnetic field, for $\Omega = 20$ MHz and $\Delta = 1$ MHz. The loss reduces to the hyperfine-free value over much the same range of magnetic fields as for the spin-stretched state. The $f = 4$, $m_f = 4$ state is not the absolute ground state at any field; the suppression occurs because m_n becomes a nearly good quantum number at high fields. Microwave shielding may be achieved even for states that are not spin stretched and are not the absolute ground state.

Similar or better shielding should be achievable for other polar bialkali molecules, where the hyperfine interactions are typically weaker than for RbCs [42]. The Supplemental Material [23] gives results for the case of ^{39}K^{133}Cs, where the hyperfine couplings are weak enough that substantial shielding can be achieved even in zero magnetic field. The Supplemental Material [23] also considers the $^2\Sigma$ molecule CaF, where shielding is still effective but requires larger Rabi frequencies because of stronger couplings involving the unpaired electron spin.

Gorshkov *et al.* [43] proposed a different mechanism for microwave shielding in the presence of a static electric field. For a given electric field, they chose Ω and Δ to cancel the first-order dipole-dipole interaction. The dipole-dipole coupling then acts in second order, producing an R^{-6} interaction that is always repulsive for the upper adiabatic state. They estimated loss rates using a semiclassical model of the nonadiabatic transitions. We have calculated RSR probabilities and microwave-induced loss rates for RbCs at an electric field of 0.9 kV/cm, which optimizes the

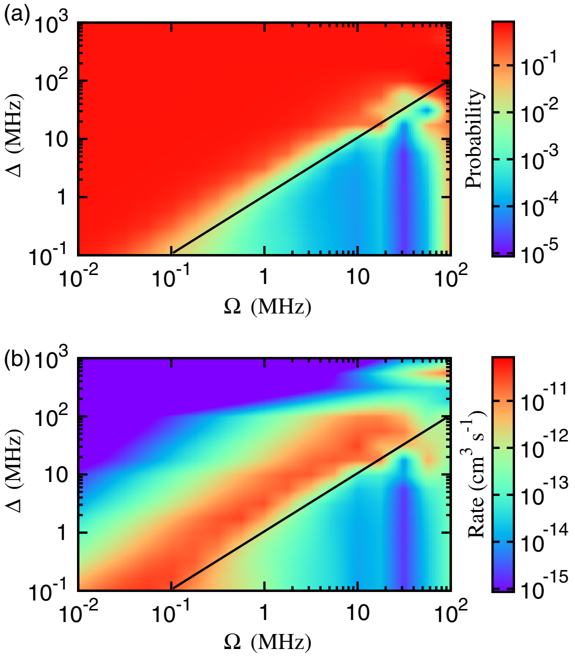

FIG. 5. Probability for RSR (a) and microwave-induced loss rate (b), as a function of Δ and Ω, for RbCs + RbCs collisions in circularly polarized microwaves and an electric field b_{rot}/μ. The black lines indicate Ω/Δ chosen so that the microwave-induced and field-induced dipole-dipole interactions cancel [43]. The optimum conditions for shielding are far from this line.

repulsive R^{-6} shield [43]. Our coupled-channels results are shown in Fig. 5. The black line at $\Omega/\Delta = 0.95$ shows where cancellation of the first-order interaction occurs. For this particular electric field, shielding starts to be effective at values of Ω close to the line, but this is coincidental and is not true for other electric fields. The optimum shielding is obtained for values of Ω and Δ that are far from the line. It occurs at much higher values of Ω/Δ, where there is no cancellation of the dipole-dipole interaction. Thus, microwave shielding can be realized in the presence of first-order dipole-dipole interactions, which play an essential role in most applications of ultracold polar molecules.

In conclusion, we have shown that collisions of ultracold polar molecules can be shielded by circularly polarized microwave radiation tuned close to a rotational transition. The microwaves prevent the collisions sampling the short-range region, where both two-body and three-body loss processes may occur. We have shown that hyperfine interactions may increase loss rates, but that this can be suppressed in a magnetic field. Loss rates can be suppressed to below 10^{-14} cm^3 s^{-1}, permitting lifetimes of seconds at densities sufficient for Bose-Einstein condensation. Shielding also produces large elastic cross sections, which combined with suppressed inelastic cross sections may allow evaporative cooling. Shielding is also effective in external electric fields, but the optimum parameters differ substantially from those proposed by Gorshkov *et al.* [43] and do not require cancellation of the field-induced dipole-dipole interaction.

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Note added.—Recently, we became aware of parallel work by Lassablière and Quéméner [44] that considers the effect of microwave radiation on molecule-molecule scattering lengths.

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[23] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.121.163401 for details of the Hamiltonians, molecular parameters, and theoretical methods used and for additional supporting calculations, including (a) comparison of the shielding for ^{87}Rb^{133}Cs with that for ^{39}K^{133}Cs and ^{40}Ca^{19}F and (b) estimates of the time needed to switch on microwave radiation adiabatically. The Supplemental Material includes Refs. [24–34].

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