Microwave shielding of ultracold polar molecules

Tijj Karman and Jeremy M. Hutson
Joint Quantum Centre (JQC) Durham-Newcastle, Department of Chemistry, Durham University, South Road, Durham, DH1 3LE, United Kingdom
(Dated: June 13, 2018)

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 cancelation of the long-range dipole-dipole interaction that is vital to many applications.

A variety of polar molecules have now been produced at [14,15], or cooled down to [6,7], ultracold temperatures. Potential applications include quantum simulation [8,9], quantum computing [10,11] and the creation of novel quantum phases [12,13]. All these applications require high densities, where collisional losses becomes 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 [14,15]. These losses can be suppressed by preventing molecules from coming close together. In this paper, we use microwaves to engineer repulsive long-range interactions that shield molecular collisions. Our approach does not require confinement to 2 dimensions as in Refs. [12,16].

Fig. 1(a) shows the shielding mechanism schematically in the low-intensity limit. Microwave radiation is blue detuned by $\Delta$ from the $n=0 \to 1$ rotational transition of the molecule. The field-dressed state with one molecule rotationally excited ($n=1$) is energetically below the undressed ground ($n=0$) state 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 coupling is neglected. The repulsive $K=1$ states cross the undressed ground state at the Condon point, which moves inwards as $\Delta$ increases. This crossing is avoided by $2\hbar \Omega$, where $\Omega$ is the Rabi frequency. The upper adiabatic curve is repulsive and provides shielding. This is closely analogous to optical blue-shielding for atoms [17].

Molecules typically have weaker resonant dipole interactions than atoms and need larger values of $\Omega$ 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 $\sigma^+$ polarization and $m_n$ is the projection of $n$ onto the microwave propagation axis. There are also dark states $|0\rangle$ corresponding to $|1,0,-1\rangle$ and $|1,-1,-1\rangle$. This produces 5 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, $\Omega$ and $\Delta$, 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, as well as electron and nuclear spin states. A full description of the Hamiltonian and examples of the resulting adiabatic curves are given in the supplemental material [18].

We perform numerically exact coupled-channels scattering calculations of two different types of loss. The coupled-channel approach is essential, because semiclassical approximations such as Landau-Zener break down when the 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 [19], and apply both capture boundary conditions at short range and S-matrix boundary conditions at long range [20]. Flux that reaches the capture radius may be lost if there are 3-body processes or chemical reactivity. We calculate the probability of reaching short range (RSR) and the corresponding rate coefficient. 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 \( \Delta \) and \( \Omega \). This calculation is for RbCs+RbCs collisions in the presence of circularly polarized \( (\sigma^+) \) microwaves, without static fields or hyperfine interactions. For large \( \Omega \) and comparable or smaller \( \Delta \), 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; these 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 [18].

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 significant 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 STIRAP 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 [18]. Considerably shorter times may be achieved with ramps that are slower at low intensity.

For ultracold collisions, the strong dependence of the scattering length on the position of the least-bound state usually precludes \textit{ab initio} calculation of elastic cross sections \( \sigma_{el} \) [21, 22]. In the presence of shielding, however, the molecules never experience the inaccurately known short-range interactions, and the calculated \( \sigma_{el} \) is quantitatively predictive. For RbCs molecules, shielded as above with \( \Delta = 1 \) MHz and \( \Omega = 10 \) MHz, we obtain \( \sigma_{el} = 3.6 \times 10^{-10} \) 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 [23] 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.

We next consider the effect of hyperfine interactions. These can cause losses for molecules that are not present for atoms, because atomic hyperfine splittings are much larger than \( \Omega \) and \( \Delta \). We carry out coupled-channel calculations in a full basis set including nuclear spin functions [24]. We initially consider \( ^{87}\text{Rb}\text{Cs} \) 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 \( \sigma^+ \) polarization, to the spin-stretched \( f = 6, m_f = 6 \) rotationally excited \( n = 1 \) state [25]. At low magnetic fields, the additional chan-
Figure 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 G and 200 G magnetic fields, respectively. Panel (c) shows the dependence of the RSR and microwave-induced loss rates on magnetic field for fixed $\Omega = 20$ MHz and $\Delta = 1$ MHz. The rate for RSR is small, as in the hyperfine-free case. 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. Similar or better shielding should be achievable for other polar bialkali molecules, where the hyperfine interactions are typically weaker than for RbCs. The supplemental material [18] 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 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.

Figure 4. The dependence of the microwave-induced loss on 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 supplemental material [19] 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 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. [27] proposed a different mechanism for microwave shielding in the presence of a static electric field. For a given electric field, they chose $\Omega$ and $\Delta$ 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 suggested an electric field $b_{\text{ext}}/\mu$, which optimizes the repulsive $R^{-6}$ shield, and estimated loss rates using a semiclassical model of the nonadiabatic transitions. We have calculated RSR probabilities and microwave-induced loss rates at their optimal field, which is 0.9 kV/cm for RbCs, using our coupled-channels approach; the results are shown in Fig. 5. The cancellation of the first-order interaction occurs for $\Omega/\Delta = 0.95$.
which is indicated by the black line. It may be seen that the optimum shielding is obtained for values of $\Omega$ and $\Delta$ that are far from this line. It occurs at much higher values of $\Omega/\Delta$, where there is no cancelation of the dipole-dipole interaction. Microwave shielding can thus 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 2-body and 3-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 section, 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. [27] and do not require cancelation of the field-induced dipole-dipole interaction.

**ACKNOWLEDGEMENT**

This work was supported by the U.K. Engineering and Physical Sciences Research Council (EPSRC) Grants No. EP/P008275/1, EP/N007085/1 and EP/P01058X/1.
[24] J. Aldegunde, B. A. Rivington, P. S. Żuchowski, and J. M. Hutson, Phys. Rev. A 78, 033434 (2008).

[25] P. D. Gregory, J. Aldegunde, J. M. Hutson, and S. L. Cornish, Phys. Rev. A 94, 041403(R) (2016).

[26] J. Aldegunde and J. M. Hutson, Phys. Rev. A 96, 042506 (2017).

[27] A. V. Gorshkov, P. Rabl, G. Pupillo, A. Micheli, P. Zoller, M. D. Lukin, and H. P. Büchler, Phys. Rev. Lett. 101, 073201 (2008).