Ultracold coherent control of molecular collisions at a Förster resonance

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We show that the precise microwave preparation of a quantum superposition between three rotational states of an ultracold dipolar molecule generates controllable interferences in their two-body scattering dynamics and collisional rate coefficients, at an electric field that produces a Förster resonance. This proposal represents a feasible protocol to achieve coherent control on ultracold molecular collisions in current experiments. It sets the basis for future studies in which one can think to control the amount of each produced pairs, including trapped entangled pairs of reactants, individual pairs of products in a chemical reaction, and measuring each of their scattering phase shifts that could envision "complete chemical experiments" at ultracold temperatures.

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The advent of ultracold controlled dipolar molecules has opened many exciting perspectives for the field of ultracold matter. Their extremely controllable properties have inspired many theoretical proposals for promising quantum applications, such as quantum simulation and quantum information processes, quantum-controlled chemistry, and tests of fundamental laws [1,2]. The molecules can be well prepared in individual quantum states [3], their long-range interactions can be controlled [4], they can be long lived and protected from their environment [5-22], enabling the formation of quantum degenerate gases [20,23-25], they can be manipulated in optical lattices [26,27] or in optical tweezers [28–30], they can be used to explore many-body effects [31–40], they can be electroassociated to form long-range tetramer molecules [41–44], and they can be entangled [45,46]. Ultracold molecules can also be used to probe chemical reactions with an unprecedented control at the quantum level, as was done with the chemical reaction $KRb + KRb \rightarrow K_2 + Rb_2$ at ultracold temperatures [47,48], including the control of the rotational parity of the products [49,50] and the creation of entangled product pairs [51].

In this Letter, we propose to apply the ideas of coherent control [52–59] to current experiments of ultracold chemical reactions [17,18,48]. The condition of coherent control between colliding particles relies on the energy degeneracy of at least two colliding states [54]. For ultracold molecules such as KRb, it was proposed in Ref. [59] that the projection m_i of the rotational quantum number *j* being used to obtain a superposition of energy-degenerate colliding states as $m_i = 0, m_i = 0$ and $m_j = +1$, $m_j = -1$ (for j = 1). However, strong hyperfine couplings are always present in such ultracold molecules [60-63] and can lift the energy degeneracy of these states. To overcome this, we propose to make two collisional states degenerate, using their rotational quantum numbers instead of their projection ones, namely j = 1, j = 1 and j = 0, j = 2(with all $m_i = 0$) for a same fixed nuclear spin projection of the molecules. This is done by using a static electric field at a Förster resonance and by using a microwave to prepare ultracold dipolar molecules in an appropriate quantum superposition of three stationary states (qutrit). The energy

difference between these two states is not affected by the hyperfine couplings nor by the magnetic fields employed in current experiments. We predict that interferences in the rate coefficients of colliding ultracold molecules can be quantitatively observed. This Letter provides a realistic and concrete experimental setup for current experiments to achieve coherent control in ultracold molecular collisions.

We first consider an ultracold dipolar gas of fermionic ⁴⁰K⁸⁷Rb molecules, taken as an example, in their ground electronic and vibrational state, and in their first excited rotational state j = 1. Their rotational states will be denoted by the kets $|i, m_i\rangle$. We do not consider the nuclear spins of the atoms as they are considered to be fixed and to remain spectators at the magnetic fields considered in current experiments. This hypothesis is confirmed by previous very good agreements between theoretical predictions and experimental results [17, 18]. In a static dc electric field E, the molecules are dressed into new states $|\tilde{j}, m_j\rangle$ (also noted $|j\rangle$ for simplicity), preserving the value of m_i . These are stationary states with well-defined energies ε_i , as illustrated in Fig. 1(a) as a function of E. Following the experiments in Refs. [17, 18], we assume that all the molecules are prepared in a dressed state $|\tilde{1}, 0\rangle$, noted $|1\rangle$, with energy ε_1 and we will assume a collision energy of $E_c/k_B = 500$ nK. Two other states, namely $|\tilde{0},0\rangle$ (noted $|0\rangle$) with energy ε_0 and $|\tilde{2},0\rangle$ (noted $|2\rangle$) with energy ε_2 , will be of interest in the following. We impose the electric field to be $E = E^* = 12.506 \text{ kV/cm}$, reachable in those experiments and for which $\varepsilon_1 - \varepsilon_0 = \varepsilon_2 - \varepsilon_1$. This characterizes the position of a Förster resonance [64].

Then, by applying a linearly polarized microwave of defined frequency and intensity during a time τ , we couple the states $|0\rangle$ and $|2\rangle$ to the state $|1\rangle$, and create Rabi oscillations between those three stationary states, consequently forming a qutrit. This is illustrated in Fig. 1(b). The frequency of the microwave defines an equal detuning $\hbar\Delta$ as the energy spacings are the same, while its intensity defines an ac electric field $E_{\rm ac}$, corresponding to two Rabi frequencies $\Omega_{10} = \tilde{d}^{1\leftrightarrow0} E_{\rm ac}/\hbar \equiv \Omega$ and $\Omega_{12} = \tilde{d}^{1\leftrightarrow2} E_{\rm ac}/\hbar \equiv \chi \Omega$, depending on the electric dipole moments of the transitions. The quantities $\tilde{d}^{1\leftrightarrow2} = 0.293$ D and $\tilde{d}^{1\leftrightarrow0} = 0.198$ D are generalized induced



FIG. 1. (a) Energies ε_0 , ε_1 , ε_2 of the dressed states $|0\rangle$, $|1\rangle$, $|2\rangle$ as a function of the electric field *E* (with all $m_j = 0$). (b) Sketch of the same energies at the Förster resonance located at $E = E^* =$ 12.506 kV/cm representing a ladder configuration in the presence of a microwave. As the levels are equally spaced, the microwave creates a coupling between $|0\rangle$, $|1\rangle$ and $|1\rangle$, $|2\rangle$, with a detuning $\hbar\Delta$ and Rabi frequencies $\Omega_{01} \equiv \Omega$ and $\Omega_{12} \equiv \chi \ \Omega \approx 1.48 \ \Omega$. (c) Evolution of the coefficient $|C_{0,1,2}|^2$ as a function of η when $\Delta = 0$.

dipole moments at E^* [65], the ratio of which gives $\chi \approx 1.48$. This prepares a quantum superposition defined by the wave function at any time $t \ge \tau$,

$$|\Psi_{\rm mol}\rangle = \sum_{i=0}^{2} c_i |i\rangle \, e^{i(\vec{k}_i \cdot \vec{\rho} - \omega_i t)} \equiv \sum_{i=0}^{2} C_i |i\rangle \, e^{i[\vec{k}_i \cdot \vec{\rho} - \omega_i (t-\tau)]}, \quad (1)$$

with $c_i = C_i e^{i\omega_i \tau}$, \vec{k}_i , and $\hbar\omega_i = \hbar^2 k_i^2 / 2m + \varepsilon_i$ are respectively the initial wave vector and energy of the individual molecule of mass *m*, described by a position vector $\vec{\rho}$, formed in states *i*. Due to negligible recoil energy and Doppler effect for a microwave transition [66], the wave vector of the molecule when excited in state $|2\rangle$ or deexcited in state $|0\rangle$ is the same as the initial one in state $|1\rangle$, namely $\vec{k}_2 \simeq \vec{k}_0 \simeq \vec{k}_1$.

Control of the interferences is reached through the control of this quantum superposition and the C_i factors. This is achieved by monitoring the parameters Δ , Ω , and τ . At the Förster resonance, the dynamics of the superposition is dictated by the resulting light-matter Hamiltonian. To keep this work general, we will focus on the condition $\Delta = 0$ for which the superposition factors are analytical. At time $t = \tau$, the microwave is turned off and the C_i factors are well defined [66] given by $C_1 = \cos \eta$, $C_0 = -i \sin \Theta \sin \eta$, $C_2 = -i \cos \Theta \sin \eta$, with $\eta = \sqrt{1 + \chi^2} \Omega \tau/2$ and $\tan \Theta = \Omega_{10}/\Omega_{12} = 1/\chi \approx 0.68$. The modulus square of these factors are represented in Fig. 1(c). The preparation time τ should be shorter than a typical collisional time ($\tau \ll \tau_{col} \simeq 1$ ms for KRb molecules at E^*).

Then after a time $t \ge \tau$, the molecules are free to collide. The overall incident collisional wave function between two



FIG. 2. (a) Energies ε_{α} with $\alpha = 00, 01+, 11, 02+, 12+, 22$ as a function of E (with all $m_j = 0$). At E^* , the energy $\varepsilon_{11} = \varepsilon_{02+} \equiv \varepsilon^*$. Not shown are energies for combined molecular states including nonzero values of m_j . (b) Sketch of the same energies at the Förster resonance. (c) Evolution of the coefficient $|C_{\alpha}|^2$ as a function of η when $\Delta = 0$.

molecules [53,54] prepared as in Eq. (1) is

$$|\Psi_{\rm col}^{\rm inc}\rangle = |\Psi_{\rm mol}\rangle \otimes |\Psi_{\rm mol}\rangle = \sum_{\alpha}^{*} C_{\alpha} |\alpha\rangle e^{i\vec{k}_{\alpha}\cdot\vec{r}} e^{i\vec{K}_{\alpha}\cdot\vec{R}} e^{-i\omega_{\alpha}(t-\tau)},$$
(2)

where \vec{k}_{α} , \vec{K}_{α} , and $\hbar\omega_{\alpha} = \hbar^2 k_{\alpha}^2 / 2\mu + \hbar^2 K_{\alpha}^2 / 2M + \varepsilon_{\alpha}$ are respectively the initial wave vector of the relative motion of reduced mass $\mu = m/2$ described by a position vector \vec{r} , the initial wave vector of the center-of-mass motion of total mass M = 2m described by a position vector \vec{R} , and the energy of the two molecules initially prepared in one of the six possible combined molecular states $|\alpha\rangle \equiv |00\rangle, |11\rangle, |22\rangle, |01+\rangle, |12+\rangle, |02+\rangle$, arising from the possible combinations of states $|0\rangle$, $|1\rangle$, $|2\rangle$ in Eq. (1). The internal energy ε_{α} of these states is plotted in Fig. 2(a) as a function of E. These combined molecular states are properly symmetrized under exchange of identical particles [66]. The asterisk over the sum in Eq. (2) means that the states α are restricted to the six ones mentioned above. The C_{α} factors are given by $C_{11} = \cos^2 \eta$, $C_{00} = -\sin^2 \Theta \sin^2 \eta, \qquad C_{22} = -\cos^2 \Theta \sin^2 \eta, \qquad C_{01+} = -i\sqrt{2}\sin\Theta \sin\eta \cos\eta, \qquad C_{12+} = -i\sqrt{2}\cos\Theta \sin\eta \cos\eta,$ $C_{02+} = -\sqrt{2}\sin\Theta\cos\Theta\sin^2\eta$ [66], the modulus squares of which are represented in Fig. 2(c).

Equation (2) is then a quantum superposition of six possible incident wave functions. Each of them have a well-defined energy $\hbar\omega_{\alpha}$ and produces a scattered wave function that we can compute [12] as the result of the collision. Note that the goal of the microwave preparation is not to keep the coherence of the individual molecules, as these scattered terms will definitively destroy the qutrits, but rather to populate different molecular collisional states with the controlled C_{α} factors. The asymptotic form of the general wave function of the

system is then given by the quantum superposition of different collisional wave functions

$$\begin{split} |\Psi_{\rm col}\rangle &= \sum_{\alpha}^{*} \left[C_{\alpha} |\alpha\rangle \; e^{i\vec{k}_{\alpha}\cdot\vec{r}} \right. \\ &+ \sum_{\alpha'} C_{\alpha} \; f_{\alpha \to \alpha'}^{\rm scat}(\vec{k}_{\alpha},\hat{r}) \frac{e^{ik_{\alpha'}^{\alpha}r}}{r} |\alpha'\rangle \left] e^{i\vec{k}_{\alpha}\cdot\vec{R}} \; e^{-i\omega_{\alpha}(t-\tau)}, \end{split}$$
(3)

with the scattering amplitude given by

$$f_{\alpha \to \alpha'}^{\text{scat}}(\vec{k}_{\alpha}, \hat{r}) = \frac{2\pi}{ik_{\alpha}^{1/2}} \sum_{l=0}^{\infty} \sum_{l=0}^{l} \sum_{m_{l}=-l}^{\infty} \sum_{l'=0}^{l'} \sum_{m'_{l}=-l'}^{l'} i^{l-l'} [Y_{l}^{m_{l}}(\hat{k}_{\alpha})]^{*} Y_{l'}^{m'_{l}}(\hat{r}) T_{\alpha'\,l'\,m'_{l},\alpha\,l\,m_{l}}(k_{\alpha}), \quad (4)$$

in term of elements of the transition matrix *T* that we can compute, employing an usual partial-wave expansion over l, m_l, l', m'_l , for a given total angular momentum projection quantum number *M* [12]. The quantities $k^{\alpha}_{\alpha'}$ are found using the conservation of energy after a collision $\hbar^2 (k^{\alpha}_{\alpha'})^2/2\mu + \varepsilon_{\alpha'} = E_c + \varepsilon_{\alpha}$. Note that the states α' are all the possible combined molecular states to which the system can end up after a collision and are not restricted to the six ones prepared in Eq. (2). For example, molecules can end up in states with values of $m_j \neq 0$ such as $|\tilde{1}, \pm 1\rangle$, $|\tilde{2}, \pm 1\rangle$, $|\tilde{2}, \pm 2\rangle$, and this is included in our study [12].

Because $\vec{k}_2 \simeq \vec{k}_0 \simeq \vec{k}_1$ as mentioned earlier in Eq. (1), all the vectors \vec{k}_{α} are also equal [66] (we will denote them as \vec{k}) and the related collision energies $\hbar^2 k_{\alpha}^2/2\mu$ are the same as the initial molecules in the gas, namely $E_c/k_B = 500$ nK. Similarly, all the vectors \vec{K}_{α} are equal (we will denote them as \vec{K}) so that the six possible centers of mass, where a collision can take place, move in the same way. This is in fact a minimal requirement needed to expect for interferences between the six collisional waves in Eq. (3) [53,54]. However, if the angular frequencies ω_{α} appearing in the time-dependent terms are different, the six collisional waves are not synchronous, that is, they will not occur at the same time. They will do so if and only if the combined molecular states $|\alpha\rangle$ have the same internal energies ε_{α} [53,54]. This is in general not an obvious requirement to fulfill. But this can be done precisely for the states $|11\rangle$ and $|02+\rangle$ at $E = E^*$, by definition of the Förster resonance, where $\varepsilon_{11} = \varepsilon_{02+} = \varepsilon^*$, as illustrated in Fig. 2(b). Then $\hbar\omega_{11} = \hbar\omega_{02+}$ (we note them $\hbar\omega$) and $k_{\alpha'}^{11} = k_{\alpha'}^{02+}$ (we note them $k_{\alpha'}^{11/02+}$). As the states $|11\rangle$ and $|02+\rangle$ collide in the same center of mass and become synchronous, they can interfere. This is the key point of the Letter. We can factorize those two terms in Eq. (3) to get the interference part of the collisional wave function,

$$\begin{split} \left| \Psi_{\text{col}}^{\text{int}} \right\rangle &= \left\{ [C_{11} \left| 11 \right\rangle + C_{02+} \left| 02+ \right\rangle] e^{i\vec{k}\cdot\vec{r}} \right. \\ &+ \sum_{\alpha'} \left[C_{11} f_{11\to\alpha'}^{\text{scat}} + C_{02+} f_{02+\to\alpha'}^{\text{scat}} \right] \left| \alpha' \right\rangle \frac{e^{ik_{\alpha'}^{11/02+}r}}{r} \right\} \\ &\times e^{i\vec{K}\cdot\vec{R}} e^{-i\omega(t-\tau)}. \end{split}$$
(5)

(

For the four remaining terms $\alpha = 00, 22, 01+, 12+$, also called "satellite" terms [53–55], they do not interfere in Eq. (3) and will provide each of them an independent result. For a starting quantum superposition state, one can then define in Eq. (5),

$$f_{11/02+\to\alpha'}^{qs} = C_{11}f_{11\to\alpha'}^{scat} + C_{02+}f_{02+\to\alpha'}^{scat},$$

$$T_{\alpha'l'm'_{l},11/02+lm_{l}}^{qs} = C_{11}T_{\alpha'l'm'_{l},11\,lml} + C_{02+}T_{\alpha'l'm'_{l},02+lm_{l}},$$
(6)

where the notation 11/02+ is used now to illustrate that 11 and 02+ are interfering and cannot be considered separately, and

$$f^{\rm qs}_{\alpha \to \alpha'} = C_{\alpha} f^{\rm scat}_{\alpha \to \alpha'}, \quad T^{\rm qs}_{\alpha' l' m'_l, \alpha l m_l} = C_{\alpha} T_{\alpha' l' m'_l, \alpha l m l}$$
(7)

for the satellite terms. The rate coefficient ending in any combined molecular state α' is given by

$$\beta_{\alpha'}^{qs}(E_c) = \sum_{\alpha}^* \beta_{\alpha \to \alpha'}^{qs}(E_c), \qquad (8)$$

where the sum runs over $\alpha = 11/02+, 00, 01+, 12+, 22$, with

$$\beta_{\alpha \to \alpha'}^{\rm qs}(E_c) = 2 \frac{\pi \hbar}{\mu k} \sum_l \sum_{m_l} \sum_{l'} \sum_{m_l'} \sum_{m_l'} \left| T_{\alpha' l' m_l', \alpha l m_l}^{\rm qs} \right|^2.$$
(9)

The rate coefficients depend parametrically on η via the C_{α} coefficients in Eqs. (6) and (7). The factor of 2 takes into account that the initial molecules are indistinguishable. The overall reactive rate coefficient is given by [66]

$$\beta_{\rm re}^{\rm qs}(E_c) = \sum_{\alpha}^* 2\frac{\hbar\pi}{\mu k} |C_{\alpha}|^2 \sum_l \sum_{m_l} P_{\alpha l m_l}^{\rm re}, \qquad (10)$$

where $P_{\alpha lm_l}^{\text{re}}$ represents the reactive probability of an initial state in αlm_l and where the sum runs over $\alpha = 11, 02+, 00, 01+, 12+, 22.$

We computed all the *T* and *P*^{re} elements appearing in Eqs. (9) and (10) at $E_c/k_B = 500$ nK, using the same basis sets as in Ref. [12] that showed very good agreement with experimental observations in a free-space three-dimensional (3D) geometry ($M = 0, \pm 1, l = 1, 3, 5$; see conditions in Ref. [18]) and in a confined quasi-2D geometry ($M = \pm 1, l = 1, 3, 5$; see conditions in Ref. [17]). We plot the corresponding rate coefficients in Fig. 3 for the free-space case and in Fig. 4 for the confined case. In all cases, the rate to a state α' in Eq. (8) comes mainly from the contribution of the elastic term $\alpha = \alpha'$ in Eq. (9), those for which the kinetic energy of the relative motion of the two molecules does not change [67]. It means that all the final reactants in a state α' have mainly the same final kinetic energies than their initial ones, so that they remain ultracold and still trapped.

First, we can see in both figures that the rates present variations as a function of η , though, for $\alpha' = 00$ (solid brown curves) and 12+ (solid pink curves), they do not correspond to interferences as can be seen from Eq. (7). The rates are just respectively proportional to $|C_{00}|^2$ and $|C_{12+}|^2$ in Eq. (9). They just exhibit the same η behavior as those preparation coefficients, as can be seen in Fig. 2(c). The rates to $\alpha' = 01+$, 22, as well as to the other combined molecular states involving $m_j \neq 0$ are much smaller and do not appear in the figure. For $\alpha' = 11$ (solid red curves)



FIG. 3. Rate coefficients $\beta_{\alpha'}^{qs}$ of two qutrits at a Förster resonance as a function of the control parameter η , in a free-space 3D geometry at $E_c/k_B = 500$ nK. The solid red (respectively green) curve corresponds to a measurement in the final state $\alpha' = 11$ (respectively $\alpha' = 02+$). The dashed curves correspond to the same curves but if there were no interferences. The solid black curve corresponds to the overall reactive rate coefficient. The solid brown (respectively pink) curve corresponds to a measurement in the final state $\alpha' = 00$ (respectively $\alpha' = 12+$).

and 02+ (solid green curves) for the free-space case, the variations correspond to destructive and constructive interferences, respectively, when compared to the same rates (dashed curves) without the crossed interference term taken into account in Eqs. (8) and (6). By controlling individually C_{11} and C_{02+} with η , one coherently controls the scattering amplitude and the *T* matrix elements, hence the observables β_{11}^{qs} and β_{02+}^{qs} , with changes of magnitude up to a factor of 5 here. For the confined case, similar conclusions hold but now both states $\alpha' = 11$ and 02+ correspond to constructive interferences.

Second, the rich physics at this Förster resonance is a remarkable source for ultracold entangled pairs remaining in the trap. As can be seen for $\eta = 0$ (conditions already fulfilled in Refs. [17,18]), we predict a large production in $\alpha' = 02+$, namely trapped pairs in the entangled states $|02+\rangle = \{|02\rangle + |20\rangle\}/\sqrt{2}$, even larger than the production in the nonentangled trapped pairs $\alpha' = 11$, or in the nontrapped reactive pairs.



FIG. 4. Same as Fig. 3 but in a confined quasi-2D geometry, using the same conditions as in Ref. [17].

When η increases from 0 to $\pi/2$, it turns out that β_{02+}^{qs} decreases in a monotonic way. Even though interferences are constructive, the fact that the coefficients C_{11} and C_{02+} are purely real numbers for $\Delta = 0$ (without additional phases to monitor) decreases somewhat the flexibility to control the amount of interferences in Eq. (6). However, if we consider the case $\Delta \neq 0$, the C_{11} and C_{02+} coefficients become arbitrary complex numbers (including two phases to monitor) that can be controlled in many ways by the three independent values Δ , Ω , and τ . This will be considered in future studies. This can be used to control the amount of interferences, for example, to increase even more the production of entangled states. Coherent control could then become a tool to enhance the production of trapped entangled pairs. Note that at E^* , these trapped entangled pairs are not long lived as they will suffer from losses. Future studies will also consider a ramp of an electric field to a place $E < E^*$ where the entangled states become shielded, using the same mechanism as in Refs. [12,17,18].

Finally, the overall reactive rate coefficients, while showing variations with the control parameter η , do not exhibit signatures of interferences, as it results in the sum of many state-to-state contributions of the possible reactive product pairs, included in a phenomenological way through $P_{\alpha lm}^{\rm re}$ in Eq. (10) [12,66]. This is due to the fact that we cannot theoretically predict the individual state-to-state reactive rates, because we cannot predict the complex-valued T matrix elements of each of the products pairs with our formalism. Only the norms of those elements are known and observed [48], seemingly consistent with a statistical model [68–70], but the individual phases remain still unknown. However, an experimental setup as in Ref. [48] combined with the one in Refs. [17,18], would be able to directly measure those state-to-state interfering reactive probabilities as a function of η . By fitting the experimental results, one could imagine to get back to all the state-to-state complexvalued T matrix elements (including the phases) of the $\alpha = 11$ and $\alpha = 02 +$ columns in Eq. (6) [54,56,59]. This is reminiscent of a "complete chemical experiment" [1] at ultracold temperature, here from an interferometry-type experiment between two collisional waves of matter, with the prospect of measuring all the scattering phase shifts of a reaction [54,56,59].

In conclusion, we showed that a quantum-controlled microwave preparation of a dipolar molecule at a Förster resonance in an electric field, set the conditions for observing interferences between collisional waves and coherent control of their dynamics, in current ultracold molecular experiments. Evidences of constructive or destructive interferences are predicted to appear in the rate coefficients of two dipolar molecules measured in their first excited rotational state $(\alpha' = 11)$ or in their ground and second excited rotational state ($\alpha' = 02+$), for different cases of confinements. The overall reactive rate coefficients do not exhibit signatures of interferences, but the state-to-state reactive rates are expected to do so. Future experimental observations of such individual rates in the distribution of the product pairs could be done combining the setups of Ref. [48] and Refs. [17,18]. This would be a signature of coherent control of ultracold chemistry at the state-to-state level. This study opens many

interesting perspectives as coherent control could be used to enhance entanglement production of ultracold trapped pairs needed for many quantum physics applications, and to measure each individual state-to-state scattering phase shifts of a reaction envisioning "complete chemical experiments" at ultracold temperatures.

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