

Magnetic-field-induced interference of scattering states in ultracold collisions

Felipe Herrera*

Department of Chemistry, University of British Columbia, Vancouver, British Columbia V6T 1Z1, Canada

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We propose a general method to induce interference between scattering wave functions in atomic and molecular collisions at ultracold temperatures by using a static magnetic field. The scheme is based on the preparation of collision partners in coherent superpositions of Zeeman states. The magnetic field is used to induce interference between two incoming channels. As an illustrative example, we consider ultracold collisions between ${}^7\text{Li}$ and ${}^{133}\text{Cs}$ atoms, and show that ratios of state resolved collision cross sections can be enhanced by engineering the preparation of the initial coherent superpositions.

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The production of cold and ultracold atoms and molecules has led to many discoveries in atomic, molecular, and optical physics [1–3], and has opened the possibility to study interactions between particles in a temperature regime where their translational energy is smaller than perturbations due to external fields [4]. External fields shift the atomic or molecular energy levels by up to a few K, so control of gas-phase dynamics can be most easily achieved for translational energies near or less than 1 K. External control of collisions and chemical reactions has been a long sought-after goal in the fields of ultracold physics and chemistry [4]. Schemes for control using laser fields [5,6] or static fields [7–10] have been developed. An alternative method is coherent control of collisions based on interference between scattering wave functions. In general, the purpose of coherent control is the manipulation of the probability that a desired outcome occurs. In order to achieve this, the system is initially prepared in a coherent superposition of states by laser fields. Interference between two or more indistinguishable pathways to the desired outcome is then controlled by tuning laser parameters. Although coherent control of unimolecular processes has been successfully achieved [11], coherent control of bimolecular processes is yet to be realized.

Shapiro and Brumer developed a theory of coherent control of collisions [12,13]. The method is based on the preparation of colliding particles in coherent superpositions of internal states with well-defined momenta. The normalized wave functions of the colliding particles A and B in the laboratory frame can then be expressed as

$$|\psi\rangle_A = a_1|\phi(1)\rangle_A e^{i\mathbf{k}_1^A \cdot \mathbf{r}_A} + a_2|\phi(2)\rangle_A e^{i\mathbf{k}_2^A \cdot \mathbf{r}_A}, \quad (1)$$

$$|\psi\rangle_B = b_1|\phi(1)\rangle_B e^{i\mathbf{k}_1^B \cdot \mathbf{r}_B} + b_2|\phi(2)\rangle_B e^{i\mathbf{k}_2^B \cdot \mathbf{r}_B}, \quad (2)$$

where a_i and b_i are complex coefficients. The wave functions $|\phi(i)\rangle_A$ and $|\phi(i)\rangle_B$ correspond to the internal states of the particles, where the index i labels the corresponding nondegenerate eigenvalues. The vectors \mathbf{k}_i^A , \mathbf{k}_i^B , \mathbf{r}_A , and \mathbf{r}_B , are the wave vectors and position vectors in the laboratory frame associated with the i th internal state $|\phi(i)\rangle$ for particles A and B , respectively.

At interatomic distances where the interaction potential is negligible compared with the collision energy, the scattering wave function of the incoming state is given by

$$\begin{aligned} |\psi\rangle_{\text{inc}} = & a_1 b_1 |\Gamma(11)\rangle e^{i\mathbf{k}_{11} \cdot \mathbf{r} + i\mathbf{K}_{11} \cdot \mathbf{R}_{CM}} \\ & + a_1 b_2 |\Gamma(12)\rangle e^{i\mathbf{k}_{12} \cdot \mathbf{r} + i\mathbf{K}_{12} \cdot \mathbf{R}_{CM}} \\ & + a_2 b_1 |\Gamma(21)\rangle e^{i\mathbf{k}_{21} \cdot \mathbf{r} + i\mathbf{K}_{21} \cdot \mathbf{R}_{CM}} \\ & + a_2 b_2 |\Gamma(22)\rangle e^{i\mathbf{k}_{22} \cdot \mathbf{r} + i\mathbf{K}_{22} \cdot \mathbf{R}_{CM}}, \end{aligned} \quad (3)$$

where

$$|\Gamma(ij)\rangle = |\phi(i)\rangle_A |\phi(j)\rangle_B, \quad \mathbf{K}_{ij} = \mathbf{k}_i^A + \mathbf{k}_j^B,$$

and

$$\mathbf{k}_{ij} = (m_B \mathbf{k}_i^A - m_A \mathbf{k}_j^B) / (m_A + m_B).$$

The relative position vector is given by \mathbf{r} , the center of mass position vector by \mathbf{R}_{CM} , and the masses of A and B by m_A and m_B . The incident scattering superposition thus consists of four incoming wave functions. In general, a collision pathway is defined by an incoming wave function coupled to an outgoing wave function by the scattering T operator, which conserves the center of mass momentum and total energy. Two collision pathways thus interfere only when their corresponding incoming wave functions have the same total energy and center of mass momentum [12]. In order for the four terms of the scattering superposition (3) to have the same total energy, it is necessary that the internal state $|\phi(1)\rangle_A$ have a well-defined wave number (k_1^A), which must be in general different from the well-defined wave number (k_2^A) of state $|\phi(2)\rangle_B$. In addition, since the four terms of the scattering superposition (3) have different center of mass momentum ($\hbar \mathbf{K}_{ij}$), further conditions must be imposed on the momenta of at least two terms in Eq. (3) in order for them to interfere. For example, the second and third terms in Eq. (3) will interfere when

$$\frac{\hbar^2 k_{12}^2}{2\mu} + e_B(2) - e_B(1) = \frac{\hbar^2 k_{21}^2}{2\mu} + e_A(2) - e_A(1), \quad (4)$$

$$\mathbf{K}_{12} = \mathbf{K}_{21}, \quad (5)$$

where μ is the reduced mass of the collision pair. $e_A(i)$ and $e_B(i)$ are the energies of the eigenstates $|\phi(i)\rangle_A$ and $|\phi(i)\rangle_B$,

*fherrera@chem.ubc.ca

with $i=1,2$. If A and B are identical particles, we have that

$$e_A(2) - e_A(1) = e_B(2) - e_B(1), \quad (6)$$

$k_{12}^2 = k_{21}^2$, and $\mathbf{K}_{12} = \mathbf{K}_{21}$ by definition. The second and third terms in Eq. (3) therefore can interfere if the states $|\phi(1)\rangle$ and $|\phi(2)\rangle$ have either the same momentum or different momenta. On the contrary, to obtain interference in collisions of nonidentical particles, it is necessary to prepare the superpositions (1) and (2) in which different momenta $\hbar\mathbf{k}_1$ and $\hbar\mathbf{k}_2$ are associated with the states $|\phi(1)\rangle$ and $|\phi(2)\rangle$ so that conditions (4) and (5) are satisfied. The preparation of such superpositions is experimentally challenging. It is much easier to create coherent superpositions of different internal states with the same translational energy. Therefore it would be desirable to design a scheme for coherent control of nonidentical particle collisions using these kind of superpositions.

In order to do this, we propose to use coherent superpositions of atomic or molecular Zeeman states to generate a scattering superposition (3). This scheme can be most easily realized with a mixture of atoms or molecules at ultracold temperatures. In such a mixture it is easier to create superpositions (1) and (2) in which a particle in different internal states has the same translational energy, because the Maxwell-Boltzmann distribution of speeds is very narrow [14]. If the wave vector of a particle in different internal states is the same, i.e.,

$$\mathbf{k}_1^A = \mathbf{k}_2^A = \mathbf{k}_A \quad \text{and} \quad \mathbf{k}_1^B = \mathbf{k}_2^B = \mathbf{k}_B, \quad (7)$$

then the incoming wave function (3) in the limit of large interatomic distance can be rewritten as

$$\begin{aligned} |\psi\rangle_{\text{inc}} = & a_1 b_1 |\Gamma(11)\rangle e^{i\mathbf{k}\cdot\mathbf{r} + i\mathbf{K}_{CM}\cdot\mathbf{R}_{CM}} \\ & + a_1 b_2 |\Gamma(12)\rangle e^{i\mathbf{k}\cdot\mathbf{r} + i\mathbf{K}_{CM}\cdot\mathbf{R}_{CM}} \\ & + a_2 b_1 |\Gamma(21)\rangle e^{i\mathbf{k}\cdot\mathbf{r} + i\mathbf{K}_{CM}\cdot\mathbf{R}_{CM}} \\ & + a_2 b_2 |\Gamma(22)\rangle e^{i\mathbf{k}\cdot\mathbf{r} + i\mathbf{K}_{CM}\cdot\mathbf{R}_{CM}}. \end{aligned} \quad (8)$$

The different terms in Eq. (8) are eigenstates of the asymptotic Hamiltonian with different total energy, but the same relative momentum and center of mass momentum. If condition (6) is satisfied, then the second and third terms of the superposition may interfere.

Arango *et al.* [15,16] proposed to use a scattering superposition with a single translational wave function similar to Eq. (8) to control ionization processes. In their scheme, one collision partner is prepared in a coherent superposition of degenerate states, and the other particle is in its ground state. Unlike superposition (8), the resulting scattering superposition in their scheme is degenerate.

To illustrate our scheme, we consider an optically trapped mixture of ultracold ^7Li and ^{133}Cs atoms, which have been produced and studied experimentally by Mudrich *et al.* [17]. For alkali-metal atoms, we choose the internal states of the superpositions (1) and (2) to be Zeeman states with the same value of M_F . We label the states as $|\phi(1), M_F\rangle$ and $|\phi(2), M_F\rangle$ depending on the hyperfine state $|F, M_F\rangle$. F is the total angular momentum quantum number, with $F = I \pm \frac{1}{2}$. I is the nuclear spin quantum number, and M_F is the projection of the total angular momentum on the quantization axis. A

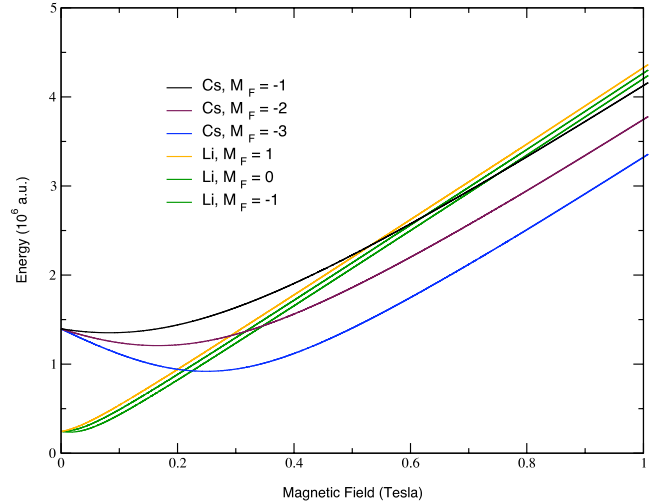


FIG. 1. (Color online) Energy difference between atomic Zeeman states $|\phi(1), M_F\rangle$ and $|\phi(2), M_F\rangle$ as a function of the magnetic field. The curves correspond to different values of M_F for Li and Cs states. At the intersection points, the magnetic field induces degeneracy between the incoming channels $|\phi(1), M_F\rangle_{\text{Li}}|\phi(2), M'_F\rangle_{\text{Cs}}$ and $|\phi(2), M_F\rangle_{\text{Li}}|\phi(1), M'_F\rangle_{\text{Cs}}$.

given superposition of hyperfine states with the same value of M_F can be prepared using optical pumping of the population from the $F = I - \frac{1}{2}$ and the $F = I + \frac{1}{2}$ manifolds to the stretched state $|I + \frac{1}{2}, I + \frac{1}{2}\rangle$ [18]. Then a small magnetic field is applied to split the energy levels corresponding to the different M_F values of each value of F . It is possible to transfer all the population from the stretched state to a state $|I - \frac{1}{2}, M_F\rangle$ with a defined projection M_F , by rapid adiabatic passage [19]. Finally, a coherent superposition of states with the same projections $|I - \frac{1}{2}, M_F\rangle$ and $|I + \frac{1}{2}, M_F\rangle$ can be produced using linearly polarized light, and the evolution of the Bloch vector can be controlled by changing laser parameters. After the preparation process, the magnitude of the net momentum transfer to the atoms in the mixture is given by their hyperfine splitting, and is approximately a factor of 10^{-7} smaller than the magnitudes of their atomic momenta, for a Li-Cs equilibrium temperature of $33 \mu\text{K}$ [17]. We can thus assume that the equality (7) is satisfied.

We use the magnetic field to further shift the Zeeman levels of the states $|\phi(1), M_F\rangle$ and $|\phi(2), M_F\rangle$, so that condition (6) is satisfied. In Fig. 1 we plot the energy difference between the Zeeman states $|\phi(1), M_F\rangle$ and $|\phi(2), M_F\rangle$ of Li and Cs atoms, for several values of M_F . For particular values of M_F for Li and Cs, condition (6) is satisfied at the intersection point between the corresponding curves. In general, the range of magnetic field values that induce interference is related to the range of energies at which condition (6) is satisfied. This energy range is centered at the intersection point between two curves in Fig. 1, and is given by the sum of the natural widths of the excited states $|\phi(2)\rangle_A$ and $|\phi(2)\rangle_B$. The corresponding range in magnetic field can be obtained from Fig. 1. For the case of Li and Cs, the widths of the excited hyperfine levels are infinitesimal, so the range of magnetic field that induces interference is very narrow.

The total cross section for inelastic scattering in the ultra-

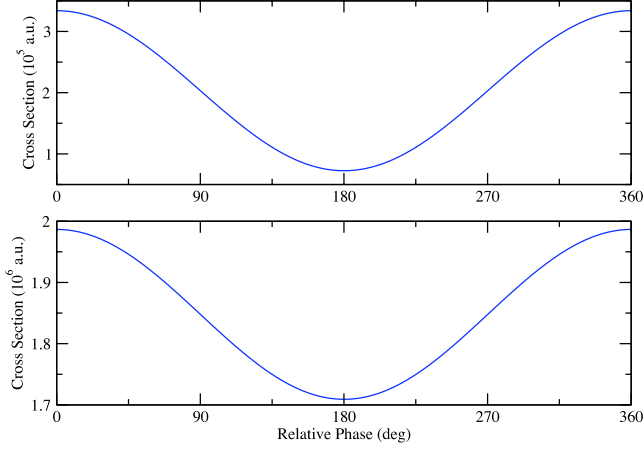


FIG. 2. (Color online) Total elastic (upper panel) and inelastic (lower panel) cross sections for incoming channels with $M_F=1$ for Li and $M_F=-3$ for Cs. The curves correspond to the magnetic field that induces interference between the incoming channels $|\phi(1), M_F\rangle_{\text{Li}}|\phi(2), M_F'\rangle_{\text{Cs}}$ and $|\phi(2), M_F\rangle_{\text{Li}}|\phi(1), M_F'\rangle_{\text{Cs}}$. The collision energy is 10^{-7} cm^{-1} .

cold s -wave regime is generally given by the expression

$$\sigma = \frac{\pi}{k^2} \sum_{n'} |\langle \Gamma(n') | T | \psi_{\text{inc}} \rangle|^2, \quad (9)$$

where k is the wave number of the incident channel $|\psi_{\text{inc}}\rangle$, and n' represents the set of quantum numbers that define the outgoing channel $|\Gamma\rangle$. The incident wave function can be a superposition of states given by Eq. (8). When the value of the magnetic field is such that condition (6) is *not* satisfied, there is no interference between the second and third terms in Eq. (8), and Eq. (9) gives

$$\begin{aligned} \sigma = & \frac{\pi}{k_{(11)}^2} |a_1 b_1|^2 \sum_{n'} \sigma_{n' \leftarrow 11} + \frac{\pi}{k_{(22)}^2} |a_2 b_2|^2 \sum_{n'} \sigma_{n' \leftarrow 22} \\ & + \frac{\pi}{k_{(12)}^2} |a_1 b_2|^2 \sum_{n'} \sigma_{n' \leftarrow 12} + \frac{\pi}{k_{(21)}^2} |a_2 b_1|^2 \sum_{n'} \sigma_{n' \leftarrow 21}, \end{aligned} \quad (10)$$

where $k_{(ij)}$ is the number corresponding to the incoming state $|\Gamma(ij)\rangle$,

$$\sigma_{n' \leftarrow 11} = |\langle \Gamma(n') | T | \Gamma(11) \rangle|^2,$$

$$\sigma_{n' \leftarrow 22} = |\langle \Gamma(n') | T | \Gamma(22) \rangle|^2,$$

$$\sigma_{n' \leftarrow 21} = |\langle \Gamma(n') | T | \Gamma(21) \rangle|^2,$$

$$\sigma_{n' \leftarrow 12} = |\langle \Gamma(n') | T | \Gamma(12) \rangle|^2.$$

The state $|\Gamma(n')\rangle$ represents an outgoing channel different from the incoming channel $|\Gamma(ij)\rangle$, with $i, j=1, 2$. The total inelastic cross section defined by Eq. (10) can be referred to as the background cross section. When the magnetic field is such that condition (6) is satisfied, the incoming channels $|\Gamma(12)\rangle$ and $|\Gamma(21)\rangle$ are degenerate. In this case, the cross

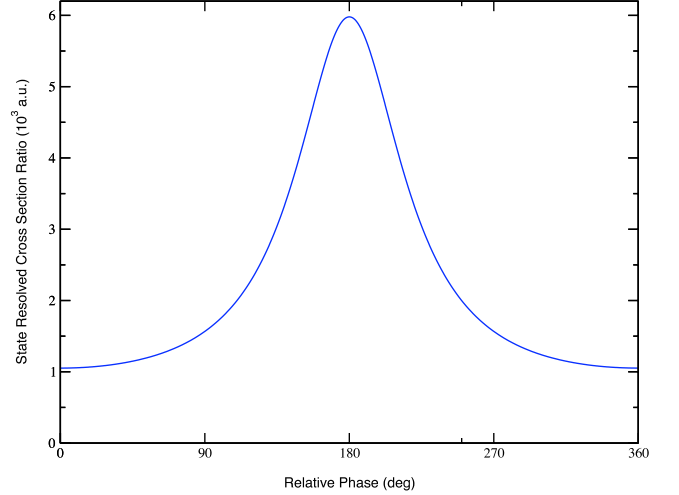


FIG. 3. (Color online) Ratio of the state resolved cross sections $\sigma_{i \leftarrow n} / \sigma_{j \leftarrow n}$. State $|\Gamma(i)\rangle$ corresponds to the outgoing channel with the projections $M_F=0$ for Li and $M_F=-2$ for Cs. State $|\Gamma(j)\rangle$ corresponds to the outgoing channel with the projections $M_F=1$ for Li and $M_F=-3$ for Cs. The curve corresponds to the magnetic field that induces interference between the incoming channels $|\phi(1), M_F\rangle_{\text{Li}}|\phi(2), M_F'\rangle_{\text{Cs}}$ and $|\phi(2), M_F\rangle_{\text{Li}}|\phi(1), M_F'\rangle_{\text{Cs}}$. The incoming asymptotic states have projections $M_F=1$ for Li states and $M_F=-3$ for Cs states. The collision energy is 10^{-7} cm^{-1} .

section obtained by inserting Eq. (8) into Eq. (9) is

$$\begin{aligned} \sigma = & \frac{\pi}{k_{(11)}^2} |a_1 b_1|^2 \sum_{n'} \sigma_{n' \leftarrow 11} + \frac{\pi}{k_{(22)}^2} |a_2 b_2|^2 \sum_{n'} \sigma_{n' \leftarrow 22} \\ & + \frac{\pi}{k^2} \left(|a_1 b_2|^2 \sum_{n'} \sigma_{n' \leftarrow 12} + |a_2 b_1|^2 \sum_{n'} \sigma_{n' \leftarrow 21} + 2|a_1 a_2 b_1 b_2| \right. \\ & \left. \times \left| \sum_{n'} \sigma_{n' \leftarrow 12|21} \right| \cos[\Phi + \delta_i(12|21)] \right), \end{aligned} \quad (11)$$

where $\sigma_{n' \leftarrow 12|21} = \langle \Gamma(12) | T | \Gamma(n') \rangle \langle \Gamma(n') | T | \Gamma(21) \rangle$. The relative phase Φ depends on the coefficients of the initial atomic superpositions, and in general can be varied by tuning laser parameters. It is given by

$$\tan \Phi = \frac{\text{Im}(a_2 b_1) / \text{Re}(a_2 b_1) - \text{Im}(a_1 b_2) / \text{Re}(a_1 b_2)}{1 + [\text{Im}(a_2 b_1) / \text{Re}(a_2 b_1)] [\text{Im}(a_1 b_2) / \text{Re}(a_1 b_2)]}.$$

The phase $\delta(12|21)$ is defined by $\tan \delta(12|21) = \text{Im}(\sum_{n'} \sigma_{n' \leftarrow 12|21}) / \text{Re}(\sum_{n'} \sigma_{n' \leftarrow 12|21})$. If the summations in Eqs. (10) and (11) are restricted to only one term so that $|\Gamma(n')\rangle = |\Gamma(ij)\rangle$, the cross section is elastic. If we choose a particular outgoing channel as the target of a measurement, i.e., $|\Gamma(n')\rangle = |\Gamma(i)\rangle$, the total cross section is state resolved. At the magnetic field that induces degeneracy between the incoming channels $|\Gamma(12)\rangle$ and $|\Gamma(21)\rangle$, the branching ratio between two state resolved inelastic cross sections can be written as $R = \sigma_{i \leftarrow n} / \sigma_{j \leftarrow n}$, where

$$\begin{aligned} \sigma_{i \leftarrow n} = & \frac{\pi}{k_{(11)}^2} |a_1 b_1|^2 \sigma_{i \leftarrow 11} + \frac{\pi}{k_{(22)}^2} |a_2 b_2|^2 \sigma_{i \leftarrow 22} \\ & + \frac{\pi}{k^2} \{ |a_1 b_2|^2 \sigma_{i \leftarrow 12} + |a_2 b_1|^2 \sigma_{i \leftarrow 21} + 2 |a_1 a_2 b_1 b_2| \\ & \times |\sigma_{i \leftarrow 12|21}| \cos[\Phi + \delta_i(12|21)] \}. \end{aligned} \quad (12)$$

In order to estimate the effect of interference on elastic and inelastic scattering at ultracold temperatures, we performed numerical calculations for collisions between ^7Li and ^{133}Cs atoms. The Hamiltonian and the interaction potentials that describe the dynamics of Li-Cs collisions in the presence of a magnetic field are described in Ref. [8]. The T matrix elements were obtained by solving the corresponding coupled differential equations, as described in [8]. For calculations of cross sections defined in Eqs. (10)–(12), the magnitudes of the coefficients in the coherent superpositions (1) and (2) were $|a_1| = |a_2| = |b_1| = |b_2| = 1/\sqrt{2}$, and the collision energy was 10^{-7} cm^{-1} . Figure 2 shows the phase dependence of the total elastic and inelastic cross sections for incoming channels with the projection $M_F=1$ for Li and $M_F=-3$ for Cs. The mean values of the oscillations in Fig. 2 correspond to the background cross sections. The amplitudes of the oscillations show the extent of enhancement or suppression of the cross sections. Elastic and inelastic cross sections have the same oscillation pattern, but the magnitudes are in general different.

Figure 3 shows the ratio of state resolved inelastic cross sections, given by Eq. (12), as a function of the relative phase Φ . State $|\Gamma(i)\rangle$ corresponds to the outgoing channel with the projection $M_F=0$ for Li and $M_F=-2$ for Cs. State $|\Gamma(j)\rangle$ corresponds to the outgoing channel with the projec-

tion $M_F=1$ for Li and $M_F=-3$ for Cs. The ratio is calculated at the value of the magnetic field that induces interference between the incoming channels $|\phi(1), M_F\rangle_{\text{Li}}, |\phi(2), M'_F\rangle_{\text{Cs}}$ and $|\phi(2), M_F\rangle_{\text{Li}}, |\phi(1), M'_F\rangle_{\text{Cs}}$, for the incoming states with the projection $M_F=1$ for Li and $M'_F=-3$ for Cs.

Although for some pairs of outgoing states, the ratio has the same oscillation pattern as the elastic and inelastic cross sections (see Fig. 2), there are cases in which there is an average enhancement of the ratio. In the case shown in Fig. 3, the average enhancement is approximately 3.5 times the background value.

In summary, we present a scheme to induce interference between scattering states in ultracold collisions with static magnetic fields. The scheme is based on the preparation of coherent superpositions of Zeeman states for each collision partner. The energy difference between Zeeman states forming the superpositions is tuned by the magnetic field. At a certain value of the magnetic field, two incoming channels become degenerate, and the collision cross section includes an interference term that depends on the magnitudes and phases of the coefficients of the initial superpositions. The scheme we propose is general, and can be applied to collisions of ultracold molecules. The energy levels of dipolar molecules can be shifted by dc electric fields, so electric fields can also be used to induce interference in collisions of polar molecules. The possibility of using electric or magnetic fields makes the scheme more flexible and independent of the choice of the trapping method. This may be an advantage for future experimental realizations of the scheme.

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