

**Robust coherent control of two-body collisions beyond the ultracold regime**Adrien Devolder,<sup>1</sup> Paul Brumer,<sup>1</sup> and Timur V. Tscherbul<sup>2</sup><sup>1</sup>*Chemical Physics Theory Group, Department of Chemistry, and Center for Quantum Information and Quantum Control, University of Toronto, Toronto, Ontario, Canada M5S 3H6*<sup>2</sup>*Department of Physics, University of Nevada, Reno, Nevada 89557, USA*

(Received 23 June 2023; accepted 14 September 2023; published 14 November 2023)

Quantum coherent control of collisions beyond the ultracold regime can face a major challenge due to the incoherent addition of different partial-wave contributions to the total scattering cross section. These contributions become increasingly numerous as the collision energy increases, leading to a loss of overall control. Here, we overcome this limitation by leveraging the recently discovered partial-wave phase locking (PWPL) effect, which synchronizes the oscillations of all partial-wave contributions. By using rigorous quantum scattering calculations, we demonstrate that PWPL enables coherent control of spin exchange in ion-atom collisions, far outside the ultracold regime. The predicted extent of control is sufficient to be measurable in cold atom-ion hybrid experiments.

DOI: [10.1103/PhysRevResearch.5.L042025](https://doi.org/10.1103/PhysRevResearch.5.L042025)

*Introduction.* Two-body collisions and chemical reactions of atoms and molecules are responsible for a wide range of phenomena in physics and chemistry [1–3], such as energy transfer, thermalization, relaxation, decoherence, and spectral line broadening to name a few. As these phenomena determine the properties of dilute gases, they play a central role in atomic and molecular spectroscopy [4], atmospheric science [5], astrochemistry [6], and ultracold chemistry [7,8]. For this reason, controlling the quantum dynamics of two-body collisions has long been a major thrust of physics and chemistry, and led to the development of several vast fields of research, including coherent control [9], laser control of chemical reactions [10], mode-selective chemistry [1], and stereochemistry [11,12].

A key challenge in controlling two-body collisions and chemical reactions lies in the random nature of scattering events. Specifically, under ambient conditions, the integral collision cross section (or reaction rate) is determined by many partial-wave contributions, which are essentially random functions of  $\ell$ , the orbital angular momentum for the collision [1,13]. Quantum control protocols target a given partial-wave contribution and rely in the phase of the underlying scattering amplitude (or  $S$  matrix). Thus, the optimal values of the control parameter (be it the value of an external field in field-based control schemes, laser pulse parameters in optimal control, or superposition parameters in coherent control) are necessarily  $\ell$  dependent, and thus cannot be optimized for all values of  $\ell$  contributing to the integral scattering cross section. This fundamental issue, which we will refer to

as “partial-wave scrambling,” has generally prevented the application of quantum control techniques to collisions [14] and the observation of scattering resonances [15] in the multiple partial-wave regime.

A common way to combat partial-wave scrambling is to cool the colliding species down to the ultracold regime, where collisions are dominated by a single initial partial wave. This eliminates the scrambling in the incident collision channel and reduces its severity in the outgoing channels, allowing for a high degree of control. The current state-of-the-art control techniques rely on cold molecules/atoms prepared in a well-defined internal states interacting with electric [16–18], microwave [19–23], optical [24], or magnetic fields [25,26]. However, the use of external fields may not be suitable for some applications, especially when the collision partners lack electric and/or magnetic dipole moments [27].

Coherent control is an attractive method that does not rely on external fields. This technique involves preparing superpositions of the internal states of colliding particles to create interference effects that can be manipulated by changing the relative phase between the states [9,28,29]. Complete coherent control is possible over ultracold resonant exchange processes, such as spin, charge, or excitation exchange, where only a single partial wave is involved in both the incident and final collision channels [27]. These processes can be completely suppressed (or activated), via destructive (constructive) interference. By contrast, coherent control in the multiple partial-wave regime can face a major challenge due to the partial-wave scrambling.

Here, we show that efficient coherent control in the multiple partial-wave regime can be achieved using the partial-wave phase locking (PWPL) effect [30–34], which manifests in a coherent addition of different partial-wave contributions. The physical origin of the PWPL effect, as elucidated in Refs. [30,33], can be attributed to the short-range nature of the spin-exchange interaction and the small

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magnitude of the centrifugal kinetic energy compared to the well depth of the interaction potential [35], enhancing quantum interference in the multiple partial-wave regime. Here, we show this robust PWPL-assisted coherent control of spin exchange in ion-atom collisions (Sr<sup>+</sup>-Rb). Cold atom-ion hybrid systems have been realized experimentally as a promising platform for quantum science [32]. Hence our results can be readily verified in the laboratory. This approach to control collisions beyond the ultracold regime can be applied to a wide range of quasideviant processes [33].

*Initial superposition and coherent control of cross section.* Consider a binary collision  $A + B$ , where  $A$  and  $B$  denote atoms or molecules initially prepared in a coherent superposition of internal angular momentum states,

$$|\psi_A\rangle = N(\sqrt{\cos\eta}|j_A, m_{1A}\rangle + \sqrt{\sin\eta}e^{i\beta/2}|j_A, m_{2A}\rangle), \quad (1)$$

$$|\psi_B\rangle = N(\sqrt{\sin\eta}e^{i\beta/2}|j_B, m_{1B}\rangle + \sqrt{\cos\eta}|j_B, m_{2B}\rangle), \quad (2)$$

where  $N = \frac{1}{\sqrt{\sin\eta + \cos\eta}}$  is a normalization factor,  $\eta \in [0, \pi/2]$  and  $\beta \in [0, 2\pi]$  are the parameters that determine the relative population and phase of the superposition,  $j_A$  and  $j_B$  are the internal angular momenta of the colliding partners  $A$  and  $B$ , and  $m_{1A}$ ,  $m_{2A}$ ,  $m_{1B}$ , and  $m_{2B}$  are the corresponding projections on the space-fixed quantization axis  $Z$ , subject to the constraint  $m_{1A} + m_{2B} = m_{1B} + m_{2A}$  imposed by rotational symmetry, which is required to obtain interference [36]. The initial state for the collision is given by the product  $|\psi_A\rangle|\psi_B\rangle$ ,

$$|\Psi_{\text{sup}}\rangle = N^2[\cos\eta|m_{1A}; m_{2B}\rangle + \sin\eta e^{i\beta}|m_{2A}; m_{1B}\rangle + \sqrt{\cos\eta \sin\eta}e^{i\beta/2}(|m_{1A}; m_{1B}\rangle + |m_{2A}; m_{2B}\rangle)], \quad (3)$$

where we have defined  $|m_{1A}; m_{2B}\rangle \equiv |j_A, m_{1A}\rangle|j_B, m_{2A}\rangle$ , etc., for brevity. Due to rotational symmetry, the states  $|m_{1A}; m_{2B}\rangle$  and  $|m_{2A}; m_{1B}\rangle$  interfere with each other, while the states  $|m_{1A}; m_{1B}\rangle$  and  $|m_{2A}; m_{2B}\rangle$  instead give rise to satellite terms [9,27]. For this reason, the superposition

$$|\Psi_{\text{ent}}\rangle = \cos\eta|m_{1A}; m_{2B}\rangle + \sin\eta e^{i\beta}|m_{2A}; m_{1B}\rangle \quad (4)$$

provides better control, as previously demonstrated [37]. While this superposition is harder to prepare experimentally than that in Eq. (3), it also provides a useful reference point for comparing different control schemes, as shown below. The creation of an entangled superposition, that given by Eq. (4), has been demonstrated for two <sup>85</sup>Rb atoms in optical tweezers [38]. The same method could also be explored for Rb-Sr<sup>+</sup> collisions.

The cross section from the nonentangled superposition (3) to a final state  $|f\rangle$  can be split into two parts: one related to the cross section from the entangled superposition (4), and one from the satellite terms,

$$\sigma_{\text{sup}\rightarrow f}(\eta, \beta) = N^4(\sigma_{\text{ent}\rightarrow f} + \sigma_{\text{sat}\rightarrow f}), \quad (5)$$

where

$$\begin{aligned} \sigma_{\text{ent}\rightarrow f}(\eta, \beta) &= \frac{\pi}{k^2} \sum_{\ell, m_\ell} \sum_{\ell', m'_\ell} |\cos\eta T_{m_{1A}m_{2B}\ell m_\ell \rightarrow f\ell' m'_\ell} \\ &+ \sin\eta e^{i\beta} T_{m_{2A}m_{1B}\ell m_\ell \rightarrow f\ell' m'_\ell}|^2, \end{aligned} \quad (6)$$

and

$$\sigma_{\text{sat}\rightarrow f}(\eta) = \cos\eta \sin\eta(\sigma_{m_{1A}; m_{1B}\rightarrow f} + \sigma_{m_{2A}; m_{2B}\rightarrow f}). \quad (7)$$

Here,  $\ell$  and  $\ell'$  are the initial and final orbital angular momenta for the collision, while  $m_\ell$  and  $m'_\ell$  are the projections of  $\ell$  and  $\ell'$  on a space-fixed quantization axis  $Z$ ,  $T_{m_{1A}m_{2B}\ell m_\ell \rightarrow f\ell' m'_\ell}$  and  $T_{m_{2A}m_{1B}\ell m_\ell \rightarrow f\ell' m'_\ell}$  are the  $T$ -matrix elements associated with the initial states  $|m_{1A}; m_{2B}\rangle$  and  $|m_{2A}; m_{1B}\rangle$ , and  $k$  is the relative wave vector.

To achieve good control of  $\sigma_{\text{sup}\rightarrow f}$ , it is necessary to exert efficient control over  $\sigma_{\text{ent}\rightarrow f}$ . However, even with good control over the latter, the inclusion of large satellite terms can significantly reduce overall control. This illustrates two important requirements for the efficient coherent control of  $\sigma_{\text{sup}\rightarrow f}$ : (i) achieving the best possible control over  $\sigma_{\text{ent}\rightarrow f}$  and (ii) minimizing the value of  $\sigma_{\text{sat}\rightarrow f}$ . To quantify these requirements, we define two control indices. First, the extent of control over the cross section from the entangled initial superposition  $\sigma_{\text{ent}\rightarrow f}$  can be defined as

$$R_{c,\text{ent}} = \frac{|\sigma_{\text{int}}|}{\sqrt{\sigma_{m_{1A}; m_{2B}\rightarrow f} \sigma_{m_{2A}; m_{1B}\rightarrow f}}}, \quad (8)$$

where

$$\sigma_{\text{int}} = \frac{\pi}{k^2} \sum_{\ell, m_\ell} \sum_{\ell', m'_\ell} T_{m_{1A}m_{2B}\ell m_\ell \rightarrow f\ell' m'_\ell} T_{m_{2A}m_{1B}\ell m_\ell \rightarrow f\ell' m'_\ell}^* \quad (9)$$

is the interference contribution to the integral cross section, and  $\sigma_{m_{1A}; m_{2B}\rightarrow f}$  and  $\sigma_{m_{2A}; m_{1B}\rightarrow f}$  are the cross sections from the states  $|m_{1A}; m_{1B}\rangle$  and  $|m_{2A}; m_{2B}\rangle$ , respectively, to the final state  $|f\rangle$ . The definition relies on the Schwartz inequality, so that  $R_{c,\text{ent}}$  lies between zero and one. Second, the effect of the satellite terms is quantified by

$$R_{\text{sat}} = \frac{\min(\sigma_{m_{1A}; m_{2B}\rightarrow f}, \sigma_{m_{2A}; m_{1B}\rightarrow f})}{\max(\sigma_{m_{1A}; m_{1B}\rightarrow f}, \sigma_{m_{2A}; m_{2B}\rightarrow f})}. \quad (10)$$

Here,  $\min(\sigma_{m_{1A}; m_{2B}\rightarrow f}, \sigma_{m_{2A}; m_{1B}\rightarrow f})$  [ $\max(\sigma_{m_{1A}; m_{1B}\rightarrow f}, \sigma_{m_{2A}; m_{2B}\rightarrow f})$ ] is the smallest (largest) of the two values. Small satellite terms correspond to a small denominator in Eq. (10) and then  $R_{\text{sat}} \gg 1$ , a favorable condition for coherent control.

Finally, we define a global control index as  $R_{c,\text{sup}} = \max_\eta(V)$ , where  $V$  represents the visibility that measures the oscillation of  $\sigma_{\text{sup}\rightarrow f}(\eta, \beta)$  when only the relative phase  $\beta$  is varied,

$$V(\tilde{\eta}) = \frac{\sigma_{\text{sup}\rightarrow f}(\tilde{\eta}, \beta_{\tilde{\eta}}^{\text{min}}) - \sigma_{\text{sup}\rightarrow f}(\tilde{\eta}, \beta_{\tilde{\eta}}^{\text{max}})}{\sigma_{\text{sup}\rightarrow f}(\tilde{\eta}, \beta_{\tilde{\eta}}^{\text{max}}) + \sigma_{\text{sup}\rightarrow f}(\tilde{\eta}, \beta_{\tilde{\eta}}^{\text{min}})}, \quad (11)$$

where  $\beta_{\tilde{\eta}}^{\text{min}}$  ( $\beta_{\tilde{\eta}}^{\text{max}}$ ) is the value of  $\beta$  for which the cross section is minimal (maximal) when  $\eta = \tilde{\eta}$ . As  $R_{c,\text{ent}}$ , the values of  $R_{c,\text{sup}}$  are bound between 0 and 1.

*Partial-wave scrambling in coherent control.* The control of the entangled cross section  $\sigma_{\text{ent}\rightarrow f}$  is limited by incoherent addition of the initial and final partial waves ( $\ell m_\ell, \ell' m'_\ell$ ) in Eq. (6). For a given superposition determined by the parameters ( $\eta$  and  $\beta$ ), some partial-wave contributions ( $\ell m_\ell, \ell' m'_\ell$ ) may experience constructive interference while others may exhibit destructive interference, suppressing the interference term in Eq. (9) [39]. This partial-wave scrambling issue becomes more significant as the collision energy increases, along with the number of ( $\ell m_\ell, \ell' m'_\ell$ ) contributions, and can result in complete loss of control.

To quantify partial-wave scrambling, we examine the distribution of the superposition parameters ( $\eta$ ,  $\beta$ ) for which each partial-wave contribution reaches a minimum value,  $\eta_{\min}^{\ell m_\ell, \ell' m'_\ell} = \arctan\left(\frac{|T_{m_{1A} m_{2B} \ell m_\ell \rightarrow f \ell' m'_\ell}|}{|T_{m_{2A} m_{1B} \ell m_\ell \rightarrow f \ell' m'_\ell}|}\right)$  and  $\beta_{\min}^{\ell m_\ell, \ell' m'_\ell} = \arg(T_{m_{1A} m_{2B} \ell m_\ell \rightarrow f \ell' m'_\ell}) - \arg(T_{m_{2A} m_{1B} \ell m_\ell \rightarrow f \ell' m'_\ell})$  depending on the ratio of the magnitudes, and on the difference of phases (arguments) of the  $T$ -matrix elements, respectively. Note that the maximum parameters are simply obtained from  $\eta_{\min}^{\ell m_\ell, \ell' m'_\ell} + \eta_{\max}^{\ell m_\ell, \ell' m'_\ell} = \pi/2$  and  $\beta_{\max}^{\ell m_\ell, \ell' m'_\ell} - \beta_{\min}^{\ell m_\ell, \ell' m'_\ell} = \pi$ . The distribution of the optimal parameters  $\eta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  and  $\beta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  determines the degree of partial-wave scrambling. A random distribution leads to a rapid decrease in control as the number  $N_{\ell m_\ell, \ell' m'_\ell}$  of significant partial-wave contributions ( $\ell m_\ell, \ell' m'_\ell$ ) increases, because  $R_{c, \text{ent}}$  scales as  $1/\sqrt{N_{\ell m_\ell, \ell' m'_\ell}}$  [39]. It is worth noting that the number of ( $\ell m_\ell, \ell' m'_\ell$ ) contributions is equal to  $(\sum_{\ell=0}^{\ell_{\max}} 2\ell + 1) \times (\sum_{\ell'=0}^{\ell'_{\max}} 2\ell' + 1)$ , where  $\ell_{\max}$  is the maximal partial wave involved in the process. While only a fraction of these contributions usually will have an important value due to symmetries, this number stays significantly larger than  $\ell_{\max}$ , as illustrated below. The solution of the partial-wave scrambling problem lies in the clustering of the optimal parameters  $\eta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  and  $\beta_{\min}^{\ell m_\ell, \ell' m'_\ell}$ . As shown below, the clustering of  $\beta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  is manifest in the PWPL mechanism, which dramatically reduces the partial-wave scrambling and thereby paves the way to coherent control in the multiple partial-wave regime.

*Application: Coherent control of spin relaxation in Sr<sup>+</sup>-Rb collisions.* The PWPL phenomenon was first predicted to occur in spin relaxation in ion-atom collisions [30,33], making hybrid ion-atom systems [32] ideal for investigating PWPL-assisted coherent control. Consider a rubidium atom (<sup>87</sup>Rb) prepared in a superposition of hyperfine states  $|2, -1\rangle_B$  and  $|2, 0\rangle_B$ , colliding with a trapped strontium ion <sup>88</sup>Sr<sup>+</sup> prepared in a superposition of Zeeman states  $|1/2, -1/2\rangle_A$  and  $|1/2, 1/2\rangle_A$ . We present the results for relaxation to the final states  $|1/2, -1/2\rangle_A |1, 0\rangle_B \equiv |\downarrow\rangle$  and  $|1/2, +1/2\rangle_A |1, 0\rangle_B \equiv |\uparrow\rangle$ , as these states have larger cross sections than the final states  $|1/2, \pm 1/2\rangle_A |1, -1\rangle_B$  and  $|1/2, \pm 1/2\rangle_A |1, 1\rangle_B$ .

To motivate experimental studies we carried out rigorous coupled-channel (CC) calculations of Sr<sup>+</sup>-Rb collisions using state-of-the-art *ab initio* interaction potentials and second-order spin-orbit interactions, as described in Ref. [30]. To ensure numerical convergence of the results for collision energies ranging from 1  $\mu$ K to 50 mK, we used extended CC basis sets including up to 80 partial waves.

The control indices  $R_{c, \text{ent}}$  and  $R_{c, \text{sup}}$  for Sr<sup>+</sup>-Rb calculated from exact CC results, are shown in Fig. 1(a). The high value of the entangled control index,  $R_{c, \text{ent}}$ , demonstrates that efficient control is possible in the multiple partial-wave regime. For the final state  $|\uparrow\rangle$ ,  $R_{c, \text{ent}}$  is close to 1, indicating complete control, whereas for the final state  $|\downarrow\rangle$ ,  $R_{c, \text{ent}}$  is around 0.5–0.6. Remarkably, the high entangled control index remains mostly independent of collision energy, indicating robust control over a wide energy range. Variation of the control can be caused by the presence of resonances, for example, at  $E = 100 \mu$ K, and 5 mK.

As expected, the control index for the nonentangled superposition,  $R_{c, \text{sup}}$ , is affected by satellite terms, which can

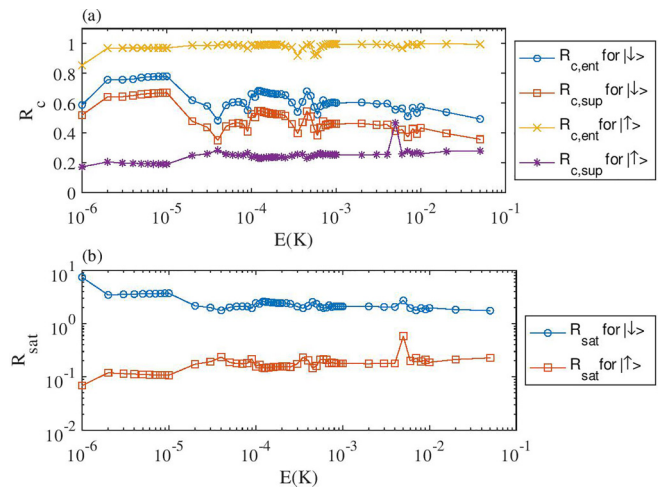


FIG. 1. (a) Control indices  $R_{c, \text{ent}}$  and  $R_{c, \text{sup}}$  for the transition from the entangled and nonentangled superpositions to the final states  $|\downarrow\rangle$  and  $|\uparrow\rangle$ . (b) Satellite term index  $R_{\text{sat}}$  for the final states  $|\downarrow\rangle$  and  $|\uparrow\rangle$ .

be quantified using the parameters  $R_{\text{sat}}$  [Eq. (10)] shown in Fig. 1(b). For the final state  $|\uparrow\rangle$ , the satellite terms are significant, resulting in a large difference between  $R_{c, \text{ent}}$  and  $R_{c, \text{sup}}$ , with  $R_{c, \text{sup}}$  being around 0.2–0.3. In contrast, the satellite terms are small for the final state  $|\downarrow\rangle$ , resulting in a small difference between  $R_{c, \text{ent}}$  and  $R_{c, \text{sup}}$ , with  $R_{c, \text{sup}} \approx 0.4$ –0.5. The reason for the smaller impact of the satellite terms on the final state  $|\downarrow\rangle$  is that for this state, the interfering transitions conserve the total internal angular momentum projection,  $m_A^i + m_B^i = m_A^f + m_B^f$ , whereas the transitions in the satellite terms do not. The situation is the opposite for the final state  $|\uparrow\rangle$ . These results highlight a complex trade-off between the partial wave scrambling and satellite terms. Even though partial-wave scrambling is less significant for the final state  $|\uparrow\rangle$ , the overall control is better for the final state  $|\downarrow\rangle$  due to the smaller effect of the satellite terms.

To illustrate coherent control in the multiple partial-wave regime, we consider the experimentally realistic case of Sr<sup>+</sup>-Rb collisions at 50 mK [30]. At this collision energy, convergence was obtained for  $\ell_{\max} = 80$ , for which  $(\sum_{\ell=0}^{\ell_{\max}} 2\ell + 1) \times (\sum_{\ell'=0}^{\ell'_{\max}} 2\ell' + 1) = 4.3 \times 10^6$ , but only 5000 terms make a significant contribution to the cross sections. Figures 2(a)–2(d) illustrate coherent control of the cross section by varying the phase angle  $\beta$  of the initial superposition (4). To obtain the best visibility  $V$ , the value of  $\eta$  is fixed at  $\eta = \pi/2$  and  $21\pi/32$  for the final states  $|\downarrow\rangle$  and  $|\uparrow\rangle$ , respectively (see Supplemental Material for the discussion of the dependence of the visibility with  $\eta$  [40]). We observe a remarkable instance of complete control of scattering from the entangled superposition to the final state  $|\uparrow\rangle$ , showing near vanishing of the cross section due to destructive interference [see Fig. 2(c)]. The minimum value of the cross section is 6.2 a.u., which is three orders of magnitude smaller than the maximum value of 2517.3 a.u. This complete control is made possible by the clustering of the optimal control parameters  $\eta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  and  $\beta_{\min}^{\ell m_\ell, \ell' m'_\ell}$  [see Figs. 4(c) and 4(d)], caused by PWPL. Note that if these parameters were randomly distributed, the entangled control index  $R_{c, \text{ent}}$

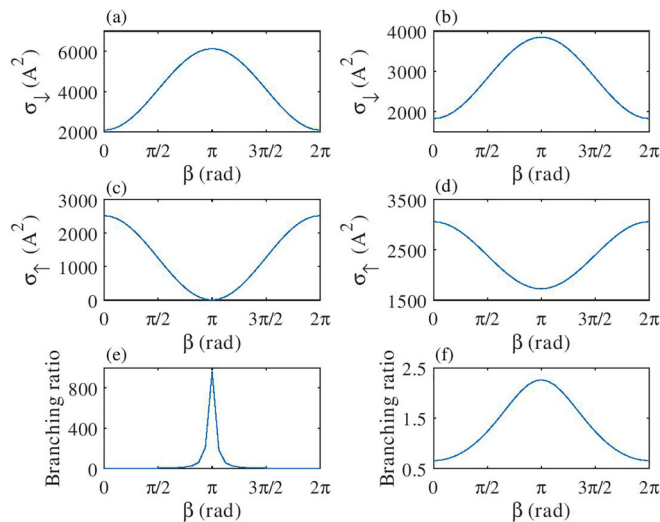


FIG. 2. Coherent control of scattering cross sections and branching ratios for  $\text{Sr}^+$ -Rb collisions with 5000 partial-wave contributions ( $\ell m_\ell, \ell' m'_\ell$ ). Cross sections (a) from  $|\Psi_{\text{ent}}\rangle$  to  $|\downarrow\rangle$ , (b) from  $|\Psi_{\text{sup}}\rangle$  to  $|\downarrow\rangle$ , (c) from  $|\Psi_{\text{ent}}\rangle$  to  $|\uparrow\rangle$  and (d) from  $|\Psi_{\text{sup}}\rangle$  to  $|\uparrow\rangle$ . Branching ratios  $\sigma_\downarrow/\sigma_\uparrow$  (e) from  $|\Psi_{\text{ent}}\rangle$  and (f) from  $|\Psi_{\text{sup}}\rangle$ . The parameter  $\eta$  is kept constant,  $\eta = \pi/2$  for  $\sigma_\downarrow$ , while  $\eta = 21\pi/32$  for  $\sigma_\uparrow$  and the branching ratios.

would be equal to  $\sim 1/\sqrt{5000} = 0.01$ , significantly smaller than the calculated value. When the initial superposition is nonentangled, complete destructive interference is countered by the presence of satellite terms, resulting in a variation from 1926.6 to 3350.2 a.u., which remains significant and certainly large enough to be detected in modern hybrid trapped ion-atom collision experiments [30–33].

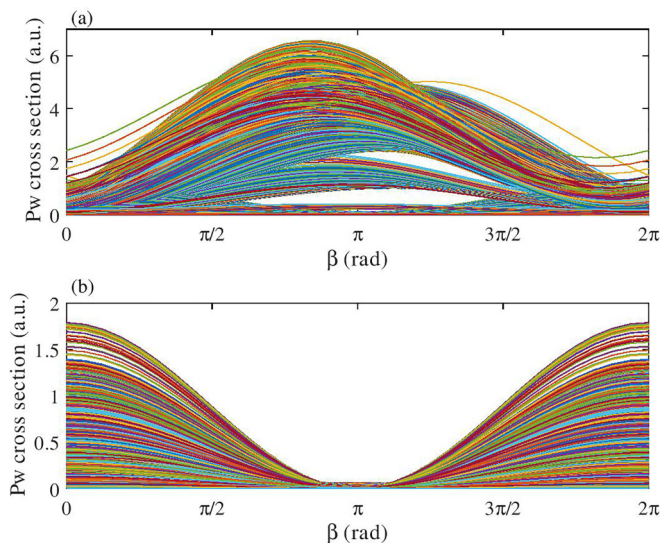


FIG. 3. Phase variation of the 5000 significant partial-wave contributions ( $\ell m_\ell, \ell' m'_\ell$ ) to the cross sections for the final states (a)  $|\downarrow\rangle$  and (b)  $|\uparrow\rangle$ . The value of  $\eta$  is fixed at  $\eta = \pi/2$  and  $21\pi/32$  for the final states  $|\downarrow\rangle$  and  $|\uparrow\rangle$ , respectively. All partial-wave contributions are plotted to illustrate the phase locking, with less efficiency observed for the final state  $|\downarrow\rangle$ .

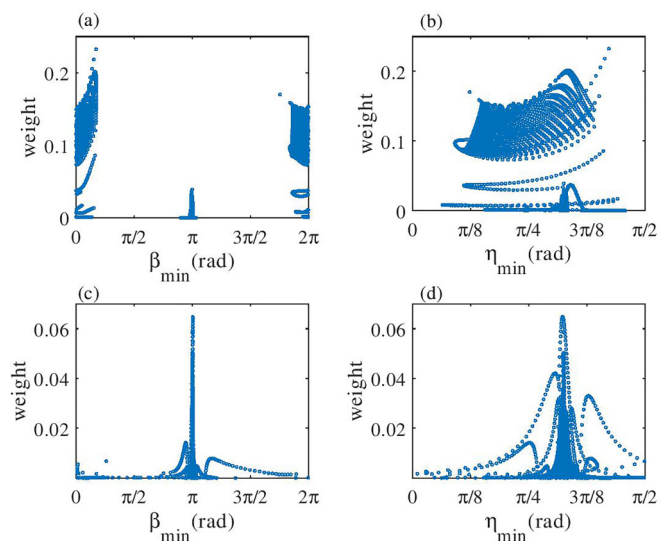


FIG. 4. Distribution of the optimal parameters  $\eta_{\text{min}}^{\ell m_\ell, \ell' m'_\ell}$  and  $\beta_{\text{min}}^{\ell, m_\ell, \ell', m'_\ell}$  for the final states (a), (b)  $|\downarrow\rangle$  and (c), (d)  $|\uparrow\rangle$ . Each point corresponds to a pair  $(\ell m_\ell, \ell' m'_\ell)$ . The weight  $w$  is calculated by the sum of squared magnitudes of  $T$ -matrix elements:  $w = |T_{m_1 A m_2 B \ell m_\ell \rightarrow f \ell' m'_\ell}|^2 + |T_{m_2 A m_1 B \ell m_\ell \rightarrow f \ell' m'_\ell}|^2$ .

For the final state  $|\downarrow\rangle$ , the clustering of  $\beta_{\text{min}}^{\ell, m_\ell, \ell', m'_\ell}$  is less effective than for  $|\uparrow\rangle$ , as shown in Figs. 3(a) and 4(a). More detrimental for the control, the distribution of the other optimal parameter,  $\eta_{\text{min}}^{\ell m_\ell, \ell' m'_\ell}$  [see Fig. 4(b)], is broad, highlighting the importance of the distribution of  $\eta_{\text{min}}^{\ell m_\ell, \ell' m'_\ell}$ . The PWPL effect locks the difference of  $T$ -matrix phases but not the ratio of their magnitudes, and thus does not guarantee the clustering of  $\eta_{\text{min}}^{\ell m_\ell, \ell' m'_\ell}$ . While this limitation can prevent PWPL from completely solving the problem of partial-wave scrambling, we observe that despite the relatively broad distribution of ratios, a good degree of control is still achievable in the multiple partial-wave regime, with  $\sigma_{\text{sup} \rightarrow f}(\eta, \beta)$  ranging from 2085.5 to 6131.7 a.u. The inclusion of the satellite terms has a slight impact on the control, resulting in a variation from 1827.7 to 3850.7 a.u.

As noted above, the control of the scattering to the final states  $|\downarrow\rangle$  and  $|\uparrow\rangle$  is optimized in different regions of parameter space allowing for efficient control of the corresponding branching ratio  $\sigma_\downarrow/\sigma_\uparrow$ , as shown in Figs. 2(e) and 2(f) at  $\eta = 21\pi/32$ . With the entangled superposition Eq. (4), extremely robust control is achieved, with  $\sigma_\downarrow/\sigma_\uparrow$  varying by three orders of magnitude (from 0.949 41 to 960.66), owing to the complete control of  $\sigma_\uparrow$ . When using the nonentangled superposition (3), the presence of satellite terms limits the extent of control over the branching ratio to 0.66–2.26, which is large enough to be experimentally measurable.

**Conclusion.** In summary, we have shown that partial-wave phase locking enables coherent control in the multiple partial-wave regime via a dramatic reduction of partial-wave scrambling. The clustering of the optimal superposition parameters  $\beta_{\text{min}}^{\ell, m_\ell, \ell', m'_\ell}$  enabled by PWPL allows for the synchronized control of different partial-wave contributions to the total scattering cross section. In cases where the distribution

of the optimal control parameters  $\eta_{\min}^{\ell, m_\ell, \ell', m'_\ell}$  is broad, such as for the final state  $|\downarrow\rangle$ , partial-wave scrambling is only partially eliminated by PWPL. Even though the satellite terms reduce the control with nonentangled superpositions, our rigorous CC calculations show that coherent control over state-to-state integral cross sections and of the branching ratios is significant and measurable. Therefore, collisions between  $^{87}\text{Rb}$  and  $^{88}\text{Sr}^+$ , observed in a series of recent experiments [30–32], appear to be ideal for an experimental observation of coherent control of two-body scattering outside of the ultracold domain. As the PWPL phenomenon was shown to apply to any quasisonant scattering process [33], a wide range of these processes could soon become amenable to robust coherent control. Furthermore, the extreme sensitivity of coherent control to the PWPL effect implies that the study of collisions

of atoms and molecules prepared in a coherent superposition of internal states provides an ideal approach for investigating the PWPL effect.

*Note added in proof.* External magnetic fields typically used in trapped ion-atom collision experiments can cause decoherence due to field inhomogeneities and lift the degeneracy of atomic or molecular levels, reducing the efficiency of coherent control. The latter issue can be resolved by tuning the magnetic field to a “magic point”, where the degeneracy lifting is small compared to collision energy, which is possible for Rb-Sr<sup>+</sup> collisions at 50 mK in magnetic fields below 500 G.

*Acknowledgment.* We thank Roe Ozeri and Nitzan Akerman for valuable discussions. This work was supported by the U.S. Air Force Office of Scientific Research (AFOSR) under Contract No. FA9550-22-1-0361.

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