Anisotropy in excitation-ionization double-differential cross sections

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Simultaneous ionization and excitation of helium by electron impact is studied through ejected electron angle integrated double-differential cross sections (DDCSs) as a function of ionized electron energy using the four-body distorted wave model. Results are presented for different alignments of the $2p0$ magnetic substate of the $He⁺$ ion, and alignment effects observed in fully differential cross sections are shown to persist in the DDCS. Examination of the DDCS for different orientations leads to a determination of anisotropy parameters and phase angles between substate amplitudes as a function of ejected electron energy.

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I. INTRODUCTION

The study of ionizing collisions involving oriented targets has become more prevalent in recent years with improvements in experimental technology and computational power $[1-8]$. These studies can provide valuable information about charge cloud structure and angular momentum transfer during the collision. Most of the studies focusing on alignment and orientation effects fall into one of two groups: excitation collisions or molecular collisions. In the case of excitation collisions, a typically neutral ground-state atom is excited by electron, photon, or heavy particle impact, leaving the finalstate atom in a nonisotropic excited state. Electronic decay of the atom to the ground state can result in photon emission, and measurement of the angular distribution or polarization of these photons results in the determination of alignment and orientation parameters that can then be related back to atomic charge cloud information. Excitation collisions have been well studied for several decades, and reviews can be found in [\[9–13\]](#page-5-0). In a few instances, the target atom has been prepared in an oriented excited state, and ionization cross sections are then measured [\[1\]](#page-5-0). This type of oriented initial-state collision is less studied, but recent results have produced new information regarding alignment effects.

For molecular collisions, alignment is typically achieved through the nuclear arrangement of the molecule, which is naturally anisotropic. Although some symmetry may exist, the electronic structure of the target is also anisotropic. The complexity of targets other than the simplest diatomic molecules makes molecular collisions difficult to study, both experimentally and theoretically. However, both ionization and excitation collisions have been used to examine alignment and orientation effects in molecular collisions, and some examples can be found in [\[4–7\]](#page-5-0) and references therein.

In this paper, we choose a simple atomic system in which simultaneous excitation and ionization (EI) leads to an oriented final-state ion, allowing for the study of alignment effects. Electron impact simultaneous EI of helium is a well-studied collision process, but continues to provide challenges due to effects such as electron correlation, postcollision interaction, and the four-body nature of the collision. Ground breaking coincidence measurements of all final-state particles, including the emitted photon [called $(e, \gamma 2e)$], were presented in [\[14\]](#page-5-0), but no additional such measurements are available due to their inherent difficulty. Aside from (*e,γ* 2*e*) studies, a much broader body of work is available for EI collisions in which the momenta of both final-state electrons are measured, but no information about the photon is known. These fully differential cross sections (FDCSs) typically provide energy resolved data that indicate the principle quantum number of the final-state bound electron, but not its angular momentum or magnetic quantum number [\[15–18\]](#page-5-0).

A series of results were published in the 1990s and 2000s that provided differential cross sections for EI leading to determination of various alignment parameters, electron-photon correlations, and Stokes parameters [\[19](#page-5-0)[–26\]](#page-6-0). These joint experimental and theoretical studies typically examined scattering angle integrated cross sections, total cross sections, or photon polarization or angular distributions. Here we present both FD-CSs and ionized electron angle integrated double-differential cross sections (DDCSs) for electron impact simultaneous EI of helium for the 2*p*0 magnetic sublevel of the final-state ion. The EI process provides a challenging system in which to study alignment effects. Target electron correlation effects are known to be important, and sophisticated atomic wave functions are required for adequate theoretical treatment. Experimentally, the EI process is very challenging due to small cross sections. Prior work using our four-body distorted wave (4DW) model has shown clear orientation effects in FDCSs for the EI process, with some of the structures being traced to specific angular momentum components of the ejected electron and the target atom wave function [\[27–28\]](#page-6-0).

By focusing on the $m_l = 0$ magnetic substate, and using the 4DW model to calculate both FDCS and DDCS, we identify specific orientation effects that persist, despite integration over ionized electron angle. Unlike prior work, which presented results as a function of scattering angle or electron energy with integration over scattering angle, we present results for fixed scattering angle as a function of electron energy. The

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ejected electron angle integrated DDCSs are then used to calculate anisotropy and phase angle parameters. We calculate these parameters as a function of ionized electron energy, and we show their relationship to the magnetic sublevel DDCS. Our calculations are also compared to the experimental data of Hayes and Williams [\[20\]](#page-5-0) and Dogan *et al.* [\[21\]](#page-6-0). Atomic units are used throughout unless otherwise noted.

II. THEORY

The details of the 4DW model have been presented in [\[29\]](#page-6-0), and only the necessary details are described here. In the 4DW model, the target helium atom is treated using a 20-parameter Hylleraas wave function [\[30\]](#page-6-0). Also, all continuum electrons are treated as distorted waves, and the postcollision Coulomb interaction between the final-state continuum electrons is included. The FDCS can be written in terms of the transition matrix T_{fi} :

$$
\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE_2} = \mu_{pa}^2 \mu_{ie} \frac{k_f k_e}{k_i} |T_{fi}|^2 \tag{1}
$$

where

$$
T_{fi} = \langle \Psi_f | V | \Psi_i \rangle. \tag{2}
$$

The momenta of the scattered projectile, ionized electron, and incident projectile are \vec{k}_f , \vec{k}_e , and \vec{k}_i , respectively, and μ_{pa} and μ_{ie} are the reduced masses of the projectile and target atom and the final-state ion and the ionized electron, respectively.

The initial and final-state wave functions are written as products of the bound electron and continuum particle wave functions. Specifically, the initial-state wave function is given by

$$
\Psi_i = \chi_{\overrightarrow{k_i}}(\overrightarrow{r_1}) \Phi_i(\overrightarrow{r_2}, \overrightarrow{r_3}), \qquad (3)
$$

and the final-state wave function is given by

$$
\Psi_f = \chi_{\overrightarrow{k_f}}(\overrightarrow{r_1}) \chi_{\overrightarrow{k_e}}(\overrightarrow{r_2}) \varphi_{nlm}(\overrightarrow{r_3}) C(\overrightarrow{r_{12}}). \tag{4}
$$

In Eqs. (3) and (4), $\chi_{\overrightarrow{k_i}}(\overrightarrow{r_1})$ and $\chi_{\overrightarrow{k_i}}(\overrightarrow{r_1})$ are the incident and scattered projectile wave functions, $\Phi_i(\vec{r}_2, \vec{r}_3)$ is the target helium atom wave function, $\chi^2_{\vec{k}_e}(\vec{r}_2)$ is the ionized electron wave function, $\varphi_{nlm}(\overrightarrow{r_3})$ is the He⁺ ion wave function, and $C(\overline{r_{12}})$ is the postcollision interaction. The perturbation is *V* = $(V_i - U_i)$, with V_i being the Coulomb interaction between the projectile electron and target atom and U_i a spherically symmetric distorting potential for the target atom.

The coordinate system used here has the incident projectile momentum along the positive *z* axis, with the projectile scattering at an angle θ_s toward the positive *x* axis. The quantization axis of the final-state ion's angular momentum is located at an angle *γ* measured counterclockwise from the *z* axis. This is the ion orientation direction for the 2*p*0 substate. Figure 1 depicts the coordinate system with the quantization axis and momentum transfer directions shown.

The results presented below are for an entirely coplanar geometry in which the incident and scattered projectile

FIG. 1. Coordinate system for the collision scattering plane. The incident projectile momentum (not shown) is along the *z* axis and the scattered projectile momentum (not shown) is oriented at an angle θ_s counterclockwise from the *z* axis. The solid blue arrow indicates the quantization axis of the $He^{+}(2p)$ ion and the dashed arrow indicates the momentum transfer direction.

momenta, as well as the ionized electron momentum, lie in a plane. In addition, the orientation of the final-state ion is restricted to this same plane (the *x-z* plane of Fig. 1). DDCSs can be obtained from the FDCS of Eq. (1) by integrating over the ionized electron angle for fixed scattering angle and incident projectile energy. The transition matrix for coplanar geometry and different orientations of the residual ion in the scattering plane for the 2*p*0 magnetic substate can be found from [\[28\]](#page-6-0)

$$
T_0^R = \frac{\sin \gamma}{\sqrt{2}} (T_{-1} - T_1) + \cos \gamma T_0.
$$
 (5)

This can be rewritten in terms of the real orbitals 2*px* and 2*pz* as

$$
T_0^R = T_z^R = \sin \gamma T_x + \cos \gamma T_z \tag{6}
$$

where T_0 , T_1 , and T_{-1} are the 2*p*0, 2*p*1, and 2*p*−1 transition matrices with quantization along the *z* axis ($\gamma = 0$). The transition matrices T_x and T_z are for excitation to the $2px$ and $2pz$ states. Note that $\gamma = 0$ does not imply that any of the magnetic sublevel *T* matrices are zero; rather it results in the *T* matrix for excitation to a sublevel with the quantization axis along the incident beam direction. In coplanar geometry, $T_{-1} = -T_1$ and Eq. (5) becomes

$$
T_0^R = -\sqrt{2}\sin\gamma T_1 + \cos\gamma T_0. \tag{7}
$$

From Eqs. (6) and (7), it is evident that the distribution of ion orientations is determined by the magnitude and relative phase of the amplitudes. Thus, examining the cross sections as a function of orientation angle *γ* provides information about the relative distribution of ions.

III. RESULTS

A. FDCS

Using our 4DW model, we have calculated FDCS and DDCS for coplanar simultaneous excitation and ionization of

FIG. 2. FDCS for excitation and ionization of helium by electron impact as a function of ionized electron momentum components. Four different alignments of the He⁺(2*p*0) ion (labeled columns in figure) and three different sets of kinematic conditions (rows) are shown. Specifics for the kinematics are described in Sec. [III A.](#page-1-0) The white arrow indicates the momentum transfer direction. The magnitude of the FDCS is represented by the color bar in atomic units.

helium. In Fig. 2, we present the 2*p*0 FDCS as a function of ionized electron momentum components for several different ion orientations. The parallel and perpendicular momentum components are chosen relative to the beam direction and in the scattering plane. The radial distance from the center is equal to the magnitude of the ejected electron momentum, and higher-energy ejected electrons are shown in the outer rings of the plots. Calculations were performed for ejected electron energies between 1 and 30 eV, and the hole in the center of each plot is for ejected electron energies below 1 eV. Due to having data for only discrete ejected electron energies, the color plots of Fig. 2 show sharp changes at the boundary of each energy ring. This boundary is an artifact of the plots and has no physical significance.

Three sets of kinematic conditions are shown. In the top row of Fig. 2, the scattered electron energy is fixed at 60 eV with a scattering angle of 8◦. In the middle row, the scattered electron energy is fixed at 130 eV with a scattering angle of 8[○]. In the bottom row, the incident electron energy is fixed at 200 eV and the scattering angle is 5◦. These results show a clear dependence of FDCS on orientation of the ion, consistent with previous work $[27,28]$. In the case of orientation along the beam direction or along the momentum transfer direction, almost no forward ejection is observed, but a strong backward recoil peak is seen. This feature has previously been traced to the postcollision interaction between the outgoing electron and scattered projectile, as well as projectile-ion interactions [\[31\]](#page-6-0). For orientation perpendicular to the beam direction or perpendicular to the momentum transfer direction, a broader distribution of ejected electrons is observed, although the magnitude of the FDCS for these orientations is five to ten times smaller than orientations along the beam or momentum transfer directions.

Figure 2 also shows that, as the ionized electron energy increases, the magnitude of the FDCS decreases, indicating that the collision process produces more low-energy ejected electrons. These features hold, regardless of whether the incident projectile energy or scattered projectile energy is held fixed. One notable difference between the results for the different kinematical conditions can be seen in the FDCS for all alignments. In this case, the FDCS is larger by up to a factor of 2 for faster projectiles $(E_i = 200 \text{ eV}$ and $E_f = 130 \text{ eV}$) compared to the slower projectile case ($E_f = 60 \text{ eV}$). This is consistent with total cross-section values for the excitationionization process, which have a maximum around $E_i =$ 200 eV [\[20\]](#page-5-0).

B. DDCS

By integrating the FDCS over ionized electron angles in the scattering plane, a DDCS can be obtained. We present the DDCS for fixed scattering angle as a function of ejected electron energy, but undetermined ejected electron angle in Fig. [3.](#page-3-0) In column 1 of Fig. [3,](#page-3-0) the DDCS as a function of ejected electron energy is compared to the experimental data of [\[20,](#page-5-0)[21\]](#page-6-0). In order to compare with experiment, the substate cross sections must be summed $(2p0 + 2p1 + 2p-1)$ because the magnetic substate of the ion is unknown. The experimental

FIG. 3. DDCS for EI of helium to the 2*p*0 state by electron impact as a function of ejected electron energy (left-hand column) and orientation angle (right-hand column). The kinematics are the same as in Fig [2.](#page-2-0) In column 1, the 4DW results are shown for $He⁺$ orientations along the beam direction (dashed red line), the momentum transfer direction (dash-dotted purple line), perpendicular to the beam direction (dash-dotted blue line), and perpendicular to the momentum transfer (dotted green line). The experimental data of [\[20](#page-5-0)[,21\]](#page-6-0) are depicted as crosses. The sum of the substate DDCS is the solid black line in column 1, but is not shown in column 2 because it is constant with respect to γ . The dashed curves in column 2 decrease in magnitude with ejected electron energy. The largest DDCS corresponds to an ejected electron energy of 1 eV (dashed black line), and those below are in descending order of 5, 10, 15, 20, 25, and 30 eV.

data are cross-normalized to the 4DW 2*p* sum for $E_f = 60$ and 130 eV by normalizing the experimental cross section for $E_e = 1$ eV and $E_f = 130$ eV to the 4DW 2*p* sum for this case. This value was then used to normalize the $E_f =$ 60 eV data. Because the $E_i = 200 \text{ eV}$ data are from a different experiment, they were normalized independently by scaling the experimental cross section to the 4DW DDCS at $E_e = 1$ eV.

In general, the experimental data and 4DW theory show the same overall trend of nearly exponential decrease in the cross section as ionized electron energy increases. However, experiment shows a double peak structure at lower ionized electron energies, while no evidence of a peak structure is observed in any of the 4DW calculations. This peak structure is most likely due to interference with the doubly excited $3/31'$ states [\[24\]](#page-6-0). Because the 4DW calculations include only the amplitudes for the excitation-ionization process, no interference between the two processes can occur and the peak structure is not present in the 4DW results.

In addition to the DDCS sum, which is independent of ion orientation, we also show in column 1 of Fig. 3 the DDCS for the magnetic substates of the ion for the four orientations of Fig. [2.](#page-2-0) The orientation dependence of the FDCS is carried through to the DDCS, although integration over the ejected electron angle naturally washes out any structure that would show binary to recoil peak ratios. Consistent with the FDCS results of Fig. [2,](#page-2-0) the DDCSs as a function of ejected electron energy for orientation along the beam or momentum transfer direction are very similar and nearly an order of magnitude

larger than the DDCSs for orientations perpendicular to the beam or momentum transfer directions. Column 2 of Fig. 3 shows a clear oscillatory structure to the DDCS as a function of orientation direction. The 2*p*0 DDCS is largest for orientations near the beam direction ($\gamma = 0$ or 180[°]) and smallest for orientations perpendicular to the beam direction ($\gamma = 90$ or 270[°]). This is easily predicted from Eq. [\(7\)](#page-1-0), where if $|T_1| \ll$ $|T_0|$ the ion is oriented near the beam direction. However, when $|T_0|$ and $|T_1|$ are of similar magnitude, the relative phase between the two amplitudes becomes important.

The relative magnitudes and phase between the 2*p*0 and 2*p*1 DDCS can be studied by fitting the data in column 2 of Fig. 3 with a function of the form

$$
DDCS = A\{1 - B\cos[2(\gamma - \kappa)]\}\tag{8}
$$

where *A* is a normalization constant, *B* is defined as the anisotropy parameter, and κ is the DDCS phase shift. These parameters can be written in terms of the $2p0(\sigma_0)$ and $2p1$ (σ_1) DDCS for the quantization axis along the beam direction by

$$
A = \frac{1}{2}(2\sigma_1 + \sigma_0),\tag{9}
$$

$$
B = \frac{\sqrt{\sigma_1^2 + \frac{\sigma_0^2}{4} + \sigma_0 \sigma_1 \cos(2\delta)}}{A},
$$
 (10)

$$
\tan(2\kappa) = 2\sqrt{2} \frac{\sqrt{\sigma_0 \sigma_1} \cos \delta}{2\sigma_1 - \sigma_0},
$$
 (11)

where δ is the relative phase between the substate amplitudes T_0 and T_1 . A detailed derivation of these expressions is contained in the Appendix.

The anisotropy parameter*B* is a measure of the sensitivity of the DDCS to the orientation of the ion and has a value between zero and one. *B* is zero when the amplitudes are exactly out of phase ($\delta = \pi/2$ or $3\pi/2$) and $\sigma_0 = 2\sigma_1$. In this case, as *B* approaches zero, the DDCS as a function of alignment angle becomes more uniform, and is perfectly isotropic when the 2*p*0 and $2p1$ amplitudes are exactly out of phase and $\sigma_0 = 2\sigma_1$. *B* is 1 when the amplitudes are exactly in phase ($\delta = 0$ or π). Larger values of *B* result in greater variation of the DDCS as a function of alignment angle.

Because the relative phase between substate amplitudes is related to the DDCS phase shift κ by Eq. (11), we can find its value as a function of ejected electron energy. Recall that to calculate the DDCS the FDCS is integrated over ejected electron angle. Because the FDCS is calculated from the square of the transition matrix amplitude, phase information for the DDCS amplitudes cannot be found directly from the 4DW cross sections. Figure [4](#page-4-0) shows *κ*, *δ*, *A*, and *B* as functions of ejected electron energy, along with plots of the momentum transfer magnitude and momentum transfer angle.

Because parameter *A* is simply half of the 2*p* DDCS sum, it exhibits the same nearly exponential decay as the results shown in Fig. 3. The relative phase *δ* between the 2*p*0 and 2*p*1 amplitudes increases with increasing ejected electron energy. The anisotropy parameter *B* is greatest for the lowestenergy projectile. This indicates that these DDCSs are less uniform than those for a higher-energy projectile, consistent with the results of Fig. 3. None of the parameters *A*, *B*, or *δ*

FIG. 4. Normalization (A), anisotropy (B), phase angle (*δ*), and DDCS phase shift (κ) as a function of ejected electron energy for EI of helium. Also shown are the momentum transfer magnitude (*q*) and angle (θ_a) as a function of ejected electron energy.

appear to be correlated with the momentum transfer vector magnitude or direction. The DDCS phase shift *κ* exhibits interesting behavior at larger ionized electron energy. For the two largest projectile energies, *κ* decreases with increasing ejected electron energy, while for the lowest projectile energy it increases. This is likely indicative of the relationship between the 2*p*0 and 2*p*1 DDCS since all projectile energies show similar behavior of the relative phase with increasing ejected electron energy and have nearly identical values of the phase at $E_e = 30 \text{ eV}$.

In addition to the parameters described above, the anisotropy parameter *B* can be used to define a relative length and width of the DDCS:

$$
l = \frac{1}{2}(1 + B),\tag{12}
$$

$$
w = \frac{1}{2}(1 - B). \tag{13}
$$

Figure 5 shows the ratio *l/w* as a function of ejected electron energy. As*B* goes to zero, the relative length and width become equal and the DDCS is perfectly isotropic. This is exactly what is observed in Figs. [3–](#page-3-0)5; as ionized electron energy increases, *B* tends to zero and the DDCS as a function of alignment angle becomes more isotropic. The change in shape happens quite rapidly with increasing ionized electron energy, as seen by the rapid decay in the *l/w* ratio in Fig. 5. Based on the trend shown in Fig. 5, we might expect that DDCS for ejected electron energies greater than 30 eV will be nearly isotropic. However, further study is needed to confirm this. It is tempting to assume that the DDCS become more isotropic for larger ejected electron energy because faster outgoing electrons are less influenced by the $He⁺$ ion charge distribution. However, this cannot be the case for the 4DW model because the ejected electron distorted wave is calculated using a spherically symmetric distorting potential that does not change with ion orientation. The quantization axis of the ion is only included in the calculation in the $He⁺$ wave function. Therefore, the change in anisotropy with increasing ejected electron energy

FIG. 5. Ratio of relative length to width of the DDCS as a function of ejected electron energy. Results are derived from Eqs. (12) and (13) for the kinematics of Fig. [2.](#page-2-0)

is likely due to the relative magnitude and/or phase between the $2p0$ and $2p1$ amplitudes.

IV. CONCLUSION

We have presented fully differential cross sections and ionized electron angle integrated double-differential cross sections for simultaneous excitation and ionization of helium by electron impact using the 4DW model. In particular, we focused on alignment effects and showed that the shape of the FDCS varies significantly with ion alignment. By examining the $2p0$ magnetic substate of the He⁺ ion, we were able to show that alignment effects persist from the FDCS through to the DDCS. Comparison of the DDCS with experiment showed a similar behavior between experiment and theory as a function of ejected electron energy, although no resonance peaks were observed in the 4DW results, as was expected. The DDCS as a function of alignment angle was shown to fit a function that yielded normalization, anisotropy, and relative phase information. These values showed that the DDCS becomes more isotropic with increasing ionized electron energy. From the anisotropy parameter, relative lengths and widths of the DDCS were calculated. Experiments corresponding to these types of calculations are quite difficult, but some limited results are available and we hope this paper encourages further study of alignment effects in the excitation-ionization process.

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APPENDIX

We show here the derivation of the parameters used in Eq. [\(8\)](#page-3-0). We begin by taking the magnitude squared of Eq. [\(7\)](#page-1-0):

$$
|T_0^R|^2 = |-\sqrt{2}\sin\gamma T_1 + \cos\gamma T_0|^2. \tag{A1}
$$

Expanding this equation yields

$$
|T_0^R|^2 = 2\sin^2 \gamma |T_1|^2 + \cos^2 \gamma |T_0|^2
$$

- 2\sqrt{2} \sin \gamma \cos \gamma \text{Re}(T_0^* T_1). (A2)

Grouping terms and simplifying leads to

$$
|T_0^R|^2 = \cos(2\gamma) \left[\frac{|T_0|^2}{2} - |T_1|^2 \right] + \left[\frac{|T_0|^2}{2} + |T_1|^2 \right] - \sqrt{2} \sin(2\gamma) \operatorname{Re}(T_0^* T_1). \tag{A3}
$$

Let $A = \frac{1}{2} [|T_0|^2 + 2|T_1|^2] = \frac{1}{2} [\sigma_0 + 2\sigma_1]$, as in Eq. [\(9\)](#page-3-0). Note that we have used the definition $\sigma_0 = |T_0|^2$ and $\sigma_1 =$ $|T_1|^2$. Then, Eq. (A3) becomes

$$
\left|T_0^R\right|^2 = A \left\{ 1 + \frac{\cos 2\gamma [\sigma_0 - 2\sigma_1]}{2A} - \frac{\sqrt{2}\sin 2\gamma \text{ Re}(T_0^* T_1)}{A} \right\}.
$$
\n(A4)

Next, let $C = 2\sigma_1 - \sigma_0$ and factor this term out from the last two terms to get

$$
\left|T_0^R\right|^2 = A \left\{ 1 - \frac{C}{2A} \left[\cos(2\gamma) + \frac{2\sqrt{2}\sin(2\gamma)\text{Re}(T_0^*T_1)}{C} \right] \right\}.
$$
\n(A5)

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Define $tan(2\kappa) = 2\sqrt{2} \frac{Re(T_0^* T_1)}{C}$. Then, using some trigonometric identities

$$
\left|T_0^R\right|^2 = A \left\{ 1 - \frac{C \cos[2(\gamma - \kappa)]}{2A \cos(2\kappa)} \right\}.
$$
 (A6)

We can then identify $B = \frac{C}{2A \cos(2\kappa)}$. The parameter *B* can be rewritten using the trigonometric identity $cos(tan^{-1}x) =$ $\frac{1}{\sqrt{x^2+1}}$ such that

$$
B = \frac{C\sqrt{\frac{8[\text{Re}(T_0^*T_1)]^2}{C^2} + 1}}{2A}
$$

= $\frac{C}{2A} \sqrt{\frac{[8\sigma_0\sigma_1\cos^2(\theta_0 - \theta_1) + C^2]}{C^2}}$ (A7)

or

$$
B = \frac{1}{A} \sqrt{\frac{\sigma_0^2}{4} + \sigma_1^2 + \sigma_0 \sigma_1 \cos\left[2(\theta_0 - \theta_1)\right]},
$$
 (A8)

which is Eq. [\(10\)](#page-3-0). Note that θ_0 and θ_1 come from writing T_0 and *T*₁ in polar form, i.e., $T_0 = |T_0|e^{i\theta_0}$ and $T_1 = |T_1|e^{i\theta_1}$. Then, we define the relative phase between the 2*p*0 and 2*p*1 amplitudes as $\delta = \theta_0 - \theta_1$. Finally, we arrive at Eq. [\(8\)](#page-3-0) with the parameters *A*, *B*, and κ defined as in Eqs. [\(9\)](#page-3-0)–[\(11\)](#page-3-0).

DDCS
$$
\propto |T_0^R|^2 = A\{1 - B\cos[2(\gamma - \kappa)]\}.
$$
 (A9)

Note that both *B* and the DDCS phase shift *κ* depend on the relative phase *δ*, and therefore can be used to calculate *δ*:

$$
\cos \delta = \frac{(2\sigma_1 - \sigma_0) \tan 2\kappa}{2\sqrt{2\sigma_0 \sigma_1}} \,. \tag{A10}
$$

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