01 Jan 2005

Interference Effects for Low-Energy Electron-Impact Ionization of Nitrogen Molecules

Junfang Gao

Don H. Madison
Missouri University of Science and Technology, madison@mst.edu

Jerry Peacher
Missouri University of Science and Technology, peacher@mst.edu

Interference effects for low-energy electron-impact ionization of nitrogen molecules

Junfang Gao,* D. H. Madison, and J. L. Peacher
Department of Physics, University of Missouri–Rolla, 1870 Miner Circle, Rolla, Missouri 065409, USA
(Received 26 April 2005; published 21 September 2005)

Young's double slit interference effects for low incident energy 75.6 eV electron impact ionization of N_2 are investigated using the distorted-wave impulse approximation (DWIA) for both coplanar symmetric and asymmetric scattering. Although the DWIA does not predict observable interference effects for the coplanar symmetric case, it predicts some strong Young's double-slit-type interference effects for the highly asymmetric scattering case. These effects are strong enough that they should be experimentally observable if one can make measurements in the backscattering region.

DOI: 10.1103/PhysRevA.72.032721 PACS number(s): 34.80.Gs

INTRODUCTION

The measurement of fully differential cross sections (FDCS) for molecular ionization by electron impact, normally referred to as $(e,2e)$, represents a powerful tool to study the electronic structure of molecules as well as to examine the fundamental interactions between continuum electrons and molecules. Over the last few decades, there have been many theoretical and experimental studies performed for the $(e,2e)$ process with molecular targets. However, most of these studies have been either for high incident-energy electron-impact ionization or small molecules [1–10]. At high enough energies, where all the continuum electrons can be expressed as plane waves, the FDCS becomes proportional to the momentum space wave function of the ejected electron so that measuring the cross section translates into a direct measurement of the active electron's wave function. A very successful theoretical approach for interpreting these high energy data is the plane-wave impulse approximation (PWIA) of McCarthy and co-workers [2–4] and much valuable information about molecular wave functions was obtained from these studies. However, it has been known for several years that the PWIA fails as the energy of the incident electron is decreased. We recently proposed [11] a distorted-wave impulse approximation (DWIA) which gave reasonably good agreement with experimental FDCS data for coplanar symmetric ionization of N_2 down to incident energies of about 50 eV.

There has recently been a renewed interest in FDCS for molecules which stems from two developments: (1) the possibility of seeing interference effects for diatomic molecules comparable to a double slit pattern for light and (2) the very recent possibility of experimentally determining the orientation of the molecule at the time of ionization [12]. The idea of seeing double slit-type interference patterns for diatomic molecules was first proposed by Cohen and Fano [13] for photon ionization of H_2 (Martin [14] presented a review of the photoionization work). Very recently, indications of interference effects have also been observed by fast multicharged ion impact by Stolterfoht *et al.* [15,16] and Misra *et al.* [17] and by proton impact by Hossain *et al.* [18]. Stia *et al.* [19] have predicted that interference effects should also be seen for electron impact ionization. The purpose of this paper is to examine the possibility of observing double-slit-type interference effects for electron-impact ionization of molecular nitrogen within the framework of the DWIA. We will present FDCS for different orientations of the molecular axis (X orientation, Y orientation, Z orientation, and average orientation) and we will demonstrate that the DWIA predicts an interference pattern which should be experimentally observable. Atomic units are used unless noted otherwise.

THEORY

The DWIA approach was presented by Gao *et al.* [11] so we will only present a brief outline here. The main idea of the DWIA is to use the PWIA as a starting point, take advantage of the elementary factorization features of the PWIA, and then replace the plane waves with distorted waves. In the PWIA [4], the Born-Oppenheimer approximation is used to treat the rotational, vibrational, and electronic parts of the wave function, the initial vibrational state is assumed to be the lowest one, the initial rotational states are normalized to unity by Maxwellian weight factors, and the final rotational and vibrational states are also assumed to be degenerate and obey the closure relations. With these assumptions, the PWIA FDCS is given by

$$\frac{d^5\sigma}{d\Omega_a d\Omega_b dE_b d\Omega_R} = \frac{4}{(2\pi)^5}\frac{k_a k_b}{k_i} F(\vec{k}_i,\vec{k}_a,\vec{k}_b)\sigma^{\mathrm{PWIA}}(\mathbf{R}), \quad (1)$$

where

$$F(\vec{k}_i,\vec{k}_a,\vec{k}_b) = \frac{1}{|\vec{k}-\vec{k}'|^4} + \frac{1}{|\vec{k}+\vec{k}'|^4} - \frac{1}{|\vec{k}-\vec{k}'|^2}\frac{1}{|\vec{k}+\vec{k}'|^2} \times \cos\left(\eta \ln\frac{|\vec{k}+\vec{k}'|^2}{|\vec{k}-\vec{k}'|^2}\right), \quad (2)$$

$$\sigma^{\mathrm{PWIA}}(\mathbf{R}) = \left|\int d\mathbf{r}\beta_a^*(\vec{k}_a,\mathbf{r})\beta_b^*(\vec{k}_b,\mathbf{r})\beta_i(\vec{k}_i,\mathbf{r})\psi_j(\mathbf{r},\mathbf{R})\right|^2, \quad (3)$$

*E-mail: jgzm6@umr.edu

where

$$\vec{k}' = \frac{1}{2}(\vec{k}_i - \vec{k}_a), \quad \vec{k} = \frac{1}{2}(\vec{k}_i + \vec{q}), \tag{4}$$

$$\vec{q} = \vec{k}_i - \vec{k}_a - \vec{k}_b. \tag{5}$$

In Eqs. (1)–(3), $F(\vec{k}_i,\vec{k}_a,\vec{k}_b)$ is an elementary function of the momenta of the incident ($\vec{k}_i$), scattered ($\vec{k}_a$), and ejected electrons ($\vec{k}_b$), and $\eta = 1/2k'$. The functions $\beta_i(\vec{k}_i,\mathbf{r})$, $\beta_a(\vec{k}_a,\mathbf{r})$, and $\beta_b(\vec{k}_b,\mathbf{r})$ are plane waves for the incident, scattered, and ejected electrons, and $\psi_j(\mathbf{r},\mathbf{R})$ is the oriented molecular orbital for the active electron with $\mathbf{R}$ the internuclear vector. In Eqs. (4) and (5), $\vec{q}$ is the momentum transferred to the residual ion. In the DWIA of Gao *et al.* [11], the plane waves of Eq. (3) are replaced with molecular distorted waves:

$$\sigma^{\mathrm{DWIA}}(\mathbf{R}) = \left| \int d\mathbf{r} \chi_a^{-*}(\vec{k}_a,\mathbf{r}) \chi_b^{-*}(\vec{k}_b,\mathbf{r}) \chi_i^{+}(\vec{k}_i,\mathbf{r}) \psi_j(\mathbf{r},\mathbf{R}) \right|^2, \tag{6}$$

where $\chi_i(\vec{k}_i,\mathbf{r})$, $\chi_a(\vec{k}_a,\mathbf{r})$, and $\chi_b(\vec{k}_b,\mathbf{r})$ are molecular distorted waves for the incident, fast-final, and slow-ejected electrons. The molecular distorted waves are calculated using a spherically averaged static potential for the molecule U_S, a polarization potential U_P, and a local exchange potential U_E. Consequently, the Schrödinger equation for the incident channel distorted wave is given by

$$\left(T + U_S + U_P + U_E - \frac{k_i^2}{2} \right) \chi_i^{-}(\vec{k}_i,\mathbf{r}) = 0, \tag{7}$$

where T is the kinetic energy operator. The details of how these potentials are calculated is contained in Gao *et al.* [11]. The Hartree-Fock molecular orbital $\psi_j(\mathbf{r},\mathbf{R})$ is obtained from GAMESS [20] and the spherically symmetric distorting potential is obtained by taking a spherical average over all possible orientations using the Hartree-Fock charge density for the molecule. The two final channel distorted waves are obtained from a Schrödinger equation similar to Eq. (7):

$$\left(T + U_I + U_P + U_E - \frac{k_{a(b)}^2}{2} \right) \chi_{a(b)}^{+}(\vec{k}_{a(b)},\mathbf{r}) = 0. \tag{8}$$

Here U_I is the spherically averaged static distorting potential for the ion obtained the same way as U_S except that the active electron is removed from the charge distribution. Following the procedure used for atomic ionization [21–23], we use the same U_P and U_E with appropriate energies for both the initial and final channels.

The DWIA cross sections of Eq. (6) depend upon the orientation of the molecule. To date most experiments that have been performed represent an average over all possible orientations. Consequently we also need theoretical orientation averaged (OA) cross sections. Gao *et al.* [11] showed that a good approximation for taking the orientation average for ionization of σ_g states is to replace the oriented molecular wave function in Eq. (6) with an OA molecular wave function:

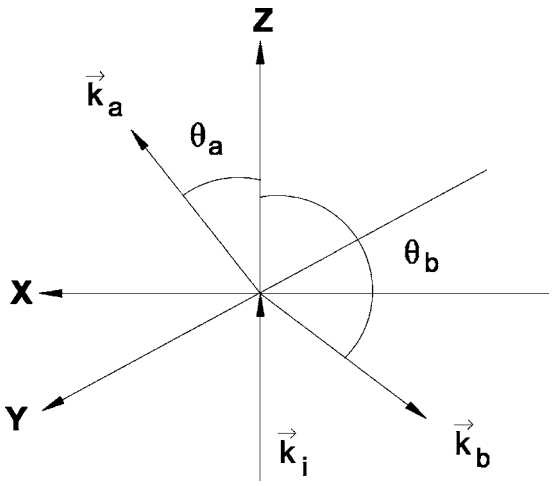

FIG. 1. Coordinate system used in this work. The scattering angle of the fast outgoing electron is θ_a and the scattering angle of ejected electron is the θ_b. The wave vectors of the incident, scattered and ejected electrons are $\mathbf{k}_i$, $\mathbf{k}_a$, and $\mathbf{k}_b$, respectively, and for this work, they are all in the scattering plane.

$$\sigma^{\mathrm{DWIA\ OA}} = \left| \int d\mathbf{r} \chi_a^{-*}(\vec{k}_a,\mathbf{r}) \chi_b^{-*}(\vec{k}_b,\mathbf{r}) \chi_i(\vec{k}_i,\mathbf{r}) \phi_j^{\mathrm{OA}}(\mathbf{r}) \right|^2 \tag{9}$$

where

$$\phi_j^{\mathrm{OA}}(\mathbf{r}) = \frac{1}{4\pi} \int \psi_j(\mathbf{r},\mathbf{R}) d\hat{\mathbf{R}}. \tag{10}$$

We would point out that the approach of Stia *et al.* [10] predicts an interference pattern which is an oscillatory function times an atomic cross section. As a result, an interference pattern is guaranteed by the theory. The same is true for the original approach of Cohen and Fano [13] and it results from using the simple LCAO (linear combination of atomic orbitals) wave function for H_2. However, our approach has no guaranteed interference effects. If we get interference effects, it has to be in the physics contained in the DWIA and, as will be shown below, under most conditions we do not see any double slit effects. In this context, the present work is similar to the recent study of H_2 photoionization by Fojón *et al.* [25] who used nearly exact molecular wavefunctions instead of the standard simple LCAO wave function.

RESULTS

The reaction of interest is $e + N_2(3\sigma_g)^2 \rightarrow 2e + N^+{}_2(3\sigma_g)^1$. We have performed calculations in the coplanar geometry which is depicted in Fig. 1. The scattered, ejected, and incident electrons are all in the XZ plane. The incident electron comes from the bottom in the $+Z$ direction, the faster electron scatters to the left in the $+X$ direction at an angle θ_a, and the slower electron is ejected at an angle θ_b which is measured clockwise relative to the Z axis. If $\theta_a = \theta_b$, the process is called coplanar symmetric scattering, and if $\theta_a \neq \theta_b$, the

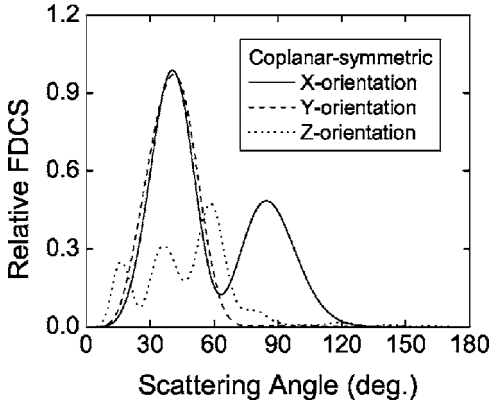

FIG. 2. Relative FDCS for electron impact ionization of the $3\sigma_g$ state of N_2 for coplanar symmetric scattering. The incident electron energy is 75.6 eV and both final state electrons have an energy of 30 eV. The theoretical DWIA results are for different orientations of the molecule as indicated in the legend. The results for the x orientation are normalized to unity at $\theta_b=40°$. The same normalization factor is used for all molecular results.

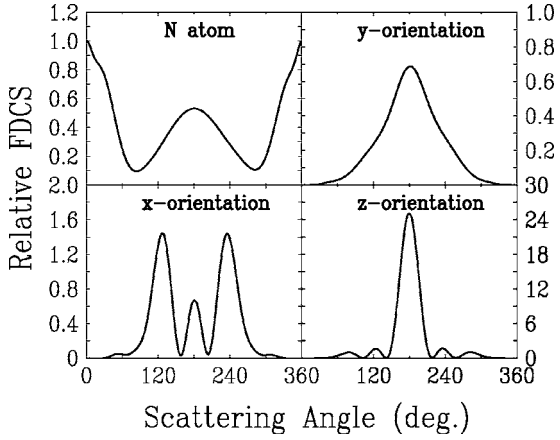

FIG. 3. Relative FDCS for electron impact ionization of the $3\sigma_g$ state of N_2 for coplanar asymmetric scattering. The incident electron energy is 75.6 eV, both final state electrons have an energy of 30 eV and $\theta_a=1°$. The horizontal axis corresponds to θ_b. The upper left hand corner of the figure contains the cross sections for an isolated N atom. The other parts of the figure are DWIA results for ionizing a N_2 molecule oriented as indicated. The normalization is the same as Fig. 2.

process is called coplanar asymmetric scattering. We will consider both coplanar symmetric scattering and coplanar asymmetric scattering.

As mentioned in the introduction, many years ago Cohen and Fano [13] suggested that interference effects similar to that of a Young's double slit should be observable for photodouble ionization and more recently it has been suggested that similar effects should be observable for charged particle ionization [15–19]. However, the first important question concerns how to identify possible Young's interference effects since wiggles are common to all cross sections and essentially all wiggles can ultimately be traced to some kind of quantum-mechanical interference. The question then is how one can look at a cross section and tell if there might be some effects similar to those of a double slit. As mentioned in the theory section, Stia *et al.* [10] predicted that if one wants to look for double slit interference patterns for H_2, one should look for an oscillating structure on top of hydrogen atom cross sections. Consequently, we will look for double slit interference patterns by comparing atomic and molecular cross sections.

Let us first examine coplanar symmetric scattering. Gao *et al.* [11] found that the FDCS for N_2 were largest for incident electrons having energies in the 75–100 eV range. Consequently, we will restrict our study to these energies so that the theoretical predictions would have the best chance of being observable experimentally. If there is going to be an observable interference effect, it should be most pronounced for oriented molecules. Figure 2 shows FDCS for electron impact ionization of the $3\sigma_g$ state of N_2 in the coplanar symmetric geometry. The incident electron energy E_i is 75.6 eV and each outgoing electron's energy is $(E_i-15.6\ \text{eV})/2=30$ eV. (This corresponds to the experimental setup of Hussey and Murray [24]). In Fig. 2, results are shown for the N_2 molecule being oriented along each of the axes. From the above discussion, we would expect that the double slit effects should appear as single atom cross sections with side bands of decreasing amplitude and none of the orientations yield cross sections which resemble this expectation. The x orientation has a binary peak and a strong secondary peak, the y orientation has only a binary peak, and in the z orientation, the binary peak is decomposed into several wiggles undoubtedly resulting from some other type of interference. Consequently, we conclude that the coplanar symmetric FDCS's that have been measured would not exhibit any Young's double slit interference effects even if measurements could be made with oriented molecules.

We then looked for evidence of double slit interference effects for the case of coplanar asymmetric scattering but keeping the electron energies the same as coplanar symmetric (i.e., both final state electrons have an energy of 30 eV but different observation angles). Figures 3 and 4 show the atomic and oriented molecular cross sections for 75.6 eV incident electrons in the coplanar asymmetric geometry. Figure 3 is for $\theta_a=1°$ and Fig. 4 is for $\theta_a=10°$. The atomic cross section shown in the figures was for the process $e+N(2p)^2 \rightarrow 2e+N(2p)^1$. The atomic cross section contains two peaks—the binary peak near the forward direction and a secondary peak near 180°. The secondary peak is called the recoil peak and it is attributed to a double scattering process in which the projectile electron collides with the atomic electron and then the atomic electron, which is headed in the binary direction, elastically scatters at 180° from the atomic nucleus. As seen from the figures, the binary peak is completely missing in the molecular cross sections for some reason. On the other hand, the recoil peak structure is observable in all three molecular orientations. For the y orientation, all we see in the molecular cross section is the recoil peak. For the z orientation, we see a cross section that looks very much like a Young's interference pattern centered on the recoil peak and for the x orientation we see what looks like two superimposed interference patterns—one at about 120° and a symmetric one at about 240°.

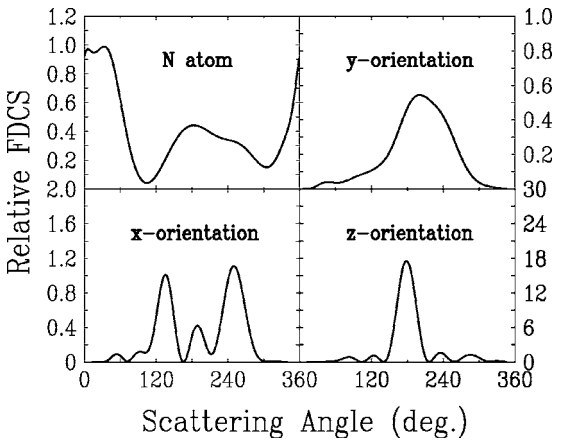

FIG. 4. Relative FDCS for electron impact ionization of the $3\sigma_g$ state of N_2 for coplanar asymmetric scattering. The incident electron energy is 75.6 eV, both final state electrons have an energy of 30 eV and $\theta_a = 10°$. The horizontal axis corresponds to θ_b. The upper left hand corner of the figure contains the cross sections for an isolated N atom. The other parts of the figure are DWIA results for ionizing a N_2 molecule oriented as indicated. The normalization is the same as Fig. 2.

The present results are consistent with the electron impact ionization of H_2 results of Stia *et al.* [19] who found interference effects only for the highly asymmetric geometries. For our case, the geometry of Fig. 4 is less asymmetric than that of Fig. 3 and the interference effects are correspondingly weaker. We find that the z orientation has by far the strongest interference effects (about a factor of 15 larger). A similar conclusion was reached by Fojón *et al.* [25] for the photoionization of H_2 where the photon polarization direction plays the same role as the momentum transfer transversal to the incident electron direction. Fojón *et al.* [25] showed that, if the photon polarization is parallel to the internuclear direction ($^1\Sigma_u^+$ symmetry), visible oscillations are seen as a result of the coherent superposition of the charge cloud from the two nuclei. Fojón *et al.* [25] also showed that, if the photon polarization is perpendicular to the internuclear direction ($^1\Pi_u$ symmetry), no oscillations are seen. We find no interference effects for the perpendicular orientation if the internuclear vector is also perpendicular to the scattering plane. However, if the internuclear vector is in the scattering plane, our results do suggest interference effects. Of course, there is no scattering plane for photoionization.

The interference pattern seen here is particularly interesting since it results completely from the atomic recoil peak. Figure 5 shows a classical cartoon illustrating the observed interference pattern resulting from two different paths to the same point. In this cartoon model, it would be expected that the y orientation should not produce any interference since any electron backscattered by a nucleus in the y orientation would be scattered out of the scattering plane and not detected in a coplanar experiment (an out-of-plane measurement should see some interference, however). As noted above, the z orientation produces much larger cross sections than the x orientation. Evidently the backscattering from the molecule is much more likely if the molecular axis is oriented parallel to the beam axis.

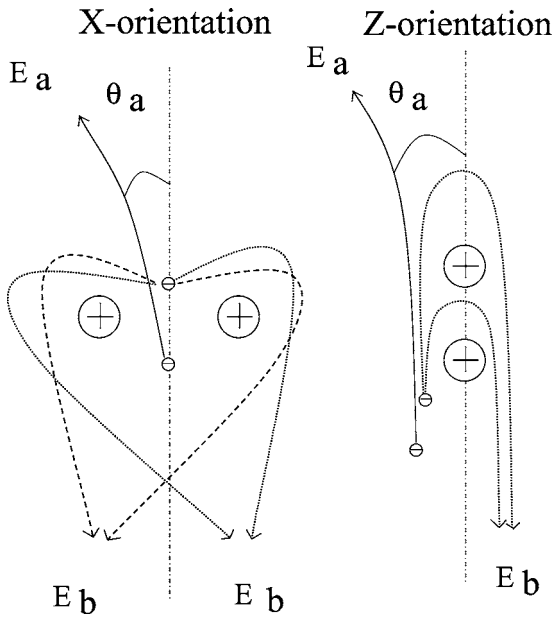

FIG. 5. Cartoon illustrating the two different classical paths which would produce the interference pattern seen in Figs. 3 and 4.

The strong interference effects from the z-axis orientation translates into a prediction that interference effects should also be observable in cross sections averaged over all orientations. Figure 6 shows the DWIAOA coplanar asymmetric cross sections averaged over all orientations and it is seen that strong back scattering interference effects are predicted here as well which should be experimentally measurable.

CONCLUSION

In summary, we have looked for Young double slit interference effects for electron-impact ionization of N_2. Our theory contains no built in interference effects so anything that we find is contained in the physics of the DWIA. For

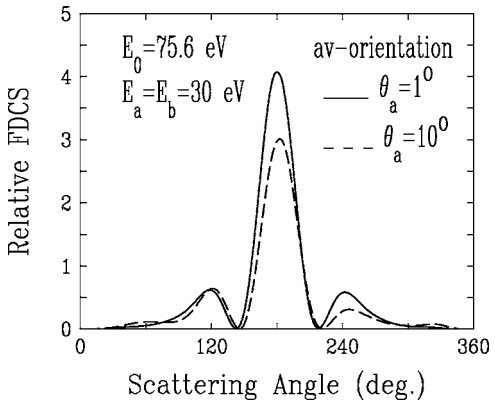

FIG. 6. Relative FDCS for electron impact ionization of the $3\sigma_g$ state of N_2 for coplanar asymmetric scattering averaged over all molecular orientations. The incident electron energy is 75.6 eV and both final state electrons have an energy of 30 eV. The scattering angle θ_a is noted in the legend, the horizontal axis corresponds to θ_b and the theoretical results are those of the DWIAOA. The normalization is the same as Fig. 2.

oriented molecules, the DWIA predicted no Young-like interference effects for coplanar symmetric scattering. For equal energy coplanar asymmetric scattering, strong interference effects were observed if the molecule is oriented either parallel to the beam axis (strongest) or perpendicular to the beam axis in the scattering plane. No Young's type interference was seen for a molecule oriented perpendicular to the beam axis and perpendicular to the scattering plane. The cross section for the z-axis orientation was sufficiently dominant that the orientation averaged DWIA OA results exhibit a measurable interference effect as well.

ACKNOWLEDGMENTS

The authors would like to acknowledge helpful discussions with Roberto D. Rivarola, Pablo Fainstein and Michael Schulz. The support of the NSF under Grant No. PHY-0070872 is also gratefully acknowledged.

[1] E. Weigold and I. E. McCarthy, *Electron Momentum Spectroscopy* (Kluwer Academic/Plenum Publishers, New York, 1999).
[2] I. E. McCarthy and E. Weigold, Phys. Rep., Phys. Lett. **27**, 275 (1976).
[3] I. E. McCarthy and A. M. Rossi, Phys. Rev. A **49**, 4645 (1994).
[4] I. E. McCarthy and E. Weigold, Rep. Prog. Phys. **51**, 299 (1988).
[5] F. Robicheaux, J. Phys. B **29**, 779 (1996).
[6] H. Deutch and K. Becker, J. Phys. Chem. A **102**, 8819 (1998).
[7] C. Champion, J. Hanssen, and P. A. Hervieux, Phys. Rev. A **63**, 052720 (2001).
[8] A. L. Monzani, L. E. Machado, M. T. Lee, and A. M. Machado, Phys. Rev. A **60**, R21 (1999).
[9] P. Weck, O. A. Fojon, J. Hanssen, B. Joulakian, and R. D. Rivarola, Phys. Rev. A **63**, 042709 (2001).
[10] C. R. Stia, O. A. Fojón, P. F. Weck, J. Hanssen, B. Joulakian, and R. D. Rivarola, Phys. Rev. A **66**, 052709 (2002).
[11] J. F. Gao, D. H. Madison, and J. L. Peacher, Phys. Rev. A **72**, 020701(R) (2005).
[12] C. Dimopoulou, R. Moshammer, D. Fischer, P. D. Fainstein, C. Höhr, A. Dorn, J. R. Crespo, López Urrutia, C. D. Schröter, H. Kollmus, R. Mann, S. Hagmann, and J. Ullrich, J. Phys. B **38**, 593 (2005).
[13] H. D. Cohen and U. Fano, Phys. Rev. **150**, 30 (1966).
[14] F. Martin, J. Phys. B **32**, R197 (1999).
[15] N. Stolterfoht, B. Sulik, V. Hoffmann, B. Skogvall, J. Y. Chesnel, J. Rangama, F. Frémont, D. Hennecart, A. Cassimi, X. Husson, A. L. Landers, J. A. Tanis, M. E. Galassi, and R. D. Rivarola, Phys. Rev. Lett. **87**, 023201 (2001).
[16] N. Stolterfoht, B. Sulik, L. Gulyás, B. Skogvall, J. Y. Chesnel, F. Frémont, D. Hennecart, A. Cassimi, L. Adoui, S. Hossain, and J. A. Tanis., Phys. Rev. A **67**, 030702(R) (2003).
[17] Deepankar Misra, U. Kadhane, Y. P. Singh, L. C. Tribedi, P. D. Fainstein, and P. Richard, Phys. Rev. Lett. **92**, 153201 (2004).
[18] S. Hossain, A. S. Alnaser, A. L. Landers, D. J. Pole, H. Knutson, A. Robison, B. Stamper, N. Stolterfoht, and J. A. Tanis, Nucl. Instrum. Methods Phys. Res. B **205**, 484 (2003).
[19] C. R. Stia, O. A. Fojón, P. F. Weck, J. Hanssen, and R. D. Rivarola, J. Phys. B **36**, L257 (2003).
[20] M. W. Schmidt, K. K. Baldridge, J. A. Boats, S. T. Elbert, M. S. Gordon, J. H. Jensen, S. Koseki, N. Matsunaga, K. A. Nguyen, S. J. Su, T. L. Windus, M. Dupuis, and J. A. Montgomery, Jr., J. Comput. Chem. **14**, 1347 (1993).
[21] D. A. Biava, H. P. Saha, E. Engel, R. M. Dreizler, R. P. McEachran, M. A. Haynes, B. Lohmann, C. T. Whelan, and D. H. Madison, J. Phys. B **35**, 293 (2002).
[22] D. A. Biava, K. Bartschat, H. P. Saha, and D. H. Madison, J. Phys. B **35**, 5121 (2002).
[23] A. Prideaux, D. H. Madison, and K. Bartschat, Phys. Rev. A **72**, 032702 (2005).
[24] M. J. Hussey and A. Murray, J. Phys. B **35**, 3399 (2002).
[25] O. A. Fojón, J. Fernández, A. Palacios, R. D. Rivarola, and F. Martín, J. Phys. B **37**, 3035 (2004).