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# Elastic scattering and impact ionization by returning electrons induced in a strong laser field

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Abstract. We study elastic scattering and impact ionization by returning electrons induced in a strong laser field based on the recently developed quantitative rescattering theory. The high-energy angle-resolved photoelectron spectra of molecules in strong fields are attributed to elastic scattering of the returning electrons with the parent ion. We demonstrate that the high-energy photoelectron spectra can be used to retrieve the position of atoms in a molecule. We also investigate the nonsequential double ionization of an atom in intense laser fields which is partly due to the impact ionization of the parent ion by the returning electron.

# 1. Introduction

The rescattering model proposed by Corkum [1] and Krause *et al* [2] is one of the most significant advances in strong laser physics. According to this model, an electron that was released earlier in the laser pulse may return to recollide with the target ion. From the recollision, many possible processes can occur. The elastic scattering experienced by the returning electron accounts for the high energy photoelectron spectra, while the nonsequential double ionization (NSDI) is due to inelastic scattering of the returning electron by the parent ion.

High energy photoelectrons for atomic targets have been well studied based on a recently developed quantitative rescattering (QRS) model [3, 4, 5]. It has been demonstrated that the two-dimensional (2D) photoelectron momentum distributions can be separated into a returning wave packet multiplied by laser-free electron elastic differential cross section (DCS). This separation makes the evaluation of high energy photoelectron spectra much easier than the traditional theoretical method, say, the solution of time-dependent Schrödinger equation (TDSE), and much more accurate than the strong field approximation (SFA). More significantly, it also allows one to retrieve atomic structure information [6, 7, 8, 9]. It is well known that the highest returning electron energy is  $3.2U_p$ , where  $U_p$  is the ponderomotive energy. For a typical laser pulse at an intensity of  $10^{14}$  W/cm<sup>2</sup> and wavelength of 800 nm, the returning electron energy is less than 20 eV. The elastic differential cross sections at such low energies are sensitive to the specific ion, and hence carries target structure information.

In this paper, we apply the QRS model to molecular targets. However, the retrieval of molecular structure is not straightforward since accurate evaluation of elastic differential cross sections for laser-free electrons with the molecular ion is extremely difficult, especially for low incident energies. Fortunately, we will show that the independent atom model (IAM) is valid for DCS at large angles for molecular scattering at energies as high as 100 eV, corresponding to

 $U_p \sim 30$  eV. In experiments, this high ponderomotive energy can be achieved by using a laser pulse with long wavelength while keeping the intensity as low as possible to avoid saturation.

In another rescattering process, NSDI can occur if the returning electron knocks out another electron in the parent ion directly, similar to the (e, 2e) process in field-free collisions. This process represents a unique example where electron-electron interaction plays an essential role in the presence of a strong laser field. While these phenomena have been understood qualitatively based on the rescattering model, a full quantum mechanical theory for the complete quantitative description of the NSDI, even for helium atom target, is still desirable. In this paper, we apply the QRS model to the NSDI of helium in an intense laser field. The experimental measurements on NSDI which provide the most detailed information are the correlated two-electron momentum spectra. To carry out numerical simulation on the correlated momentum spectra, we first calculate the returning electron wave packet by using QRS. Then by multiplying this wave packet with standard field-free (e, 2e) differential cross sections, we obtain the correlated twoelectron momentum spectra. Actually, besides the (e, 2e) process, another mechanism involved in the NSDI is excitation-tunneling, in which the ground state electron in the parent ion is excited to a higher energy bound state, followed by subsequent tunnel ionization. The investigation of excitation-tunneling in NSDI will be reported elsewhere.

Atomic units are used in this paper unless otherwise noted.

# 2. Theory

## 2.1. Strong field rescattering

Single ionization of an atom or molecule in a strong laser field can be obtained by solving the TDSE numerically. However, this is not practical due to the high computational demands. An alternative method is to base on the SFA. Although it has been well recognized that the SFA does not treat the target structure accurately, the role of lasers is adequately described by the SFA.

We use linearly polarized laser pulse in which the electric field is written as

$$\mathbf{F}(t) = F_0 a(t) \cos(\omega t + \phi) \hat{z},\tag{1}$$

where  $\omega$  is the frequency of the carrier wave and  $\phi$  the carrier envelope phase, with the envelope function a(t) chosen to be

$$a(t) = \cos^2\left(\frac{\pi t}{\tau}\right) \tag{2}$$

for the time interval  $(-\tau/2, \tau/2)$  and zero elsewhere. It should be mentioned that in all the calculations the carrier envelope phase is set to zero in this paper.

In SFA, by treating the electron-target interaction as a perturbation, the amplitude of detecting a photoelectron with momentum p at an outgoing angle  $\theta$  with respect to the polarization is given by

$$f_{\text{SFA}}(\mathbf{p}) = f_1(\mathbf{p}) + f_2(\mathbf{p}),\tag{3}$$

where only the first two terms of the perturbation series are included.

In Eq. (3), the first term  $f_1(\mathbf{p})$ , called SFA1, is the traditional strong field approximation,

$$f_1(\mathbf{p}) = -i \int_{-\infty}^{\infty} dt \left\langle \chi_{\mathbf{p}}(t) \left| H_i(t) \right| \Psi_0(t) \right\rangle, \tag{4}$$

where  $\Psi_0$  is the ground state wave function,  $\chi_{\mathbf{p}}$  the Volkov state, and  $H_i(t) = \mathbf{r} \cdot \mathbf{F}(t)$  the atom-field interaction in length gauge.

The second term in Eq. (3), called SFA2, is expressed as

$$f_{2}(\mathbf{p}) = -\int_{-\infty}^{\infty} dt \int_{t}^{\infty} dt' \int d\mathbf{k} \langle \chi_{\mathbf{p}}(t') | V | \chi_{\mathbf{k}}(t') \rangle \\ \times \langle \chi_{\mathbf{k}}(t) | H_{i}(t) | \Psi_{0}(t) \rangle.$$
(5)

It consists of three time-ordered steps by the electron: the electron is first tunnel ionized at time t, propagating in the laser field, and rescattered elastically with the parent ion at time t'. The potential V is the electron-ion interaction. The integration over  $\mathbf{k}$  is performed by means of saddle point approximation.

The momentum distribution of emission of an electron with energy  $E = p^2/2$  in the direction of  $\theta$  is given by

$$D_{\text{SFA}}(p,\theta) = |f_{\text{SFA}}(\mathbf{p})|^2.$$
(6)

The form of  $f_{\text{SFA}}$  in Eq. (3) allows us to identify the contribution from each individual term. The standard SFA results can be obtained if  $f_2$  is ignored in Eq. (3).

While the SFA1 contributes to the direct ionization, the indirect ionization related to rescattering is attributed to the SFA2. The fatal shortage of SFA lies in the fact that both the long range Coulomb potential and the short range potential from the target are completely ignored when an electron is described by the Volkov state. As a result, the target structure information is missed in SFA2.

The QRS model is set up in the light of improving SFA2 and based on the assumption that photoelectron momentum distribution in Eq. (6) can be written as

$$D_{\rm SFA}(p,\theta) = W(k_r)\sigma_{\rm B}(k_r,\theta_r),\tag{7}$$

where  $\sigma_{\rm B}(k_r, \theta_r)$  is the elastic differential cross section in the first-order Born approximation for electron with momentum  $k_r$  scattered at an angle  $\theta_r$  with respect to the incident direction, which is given by

$$\sigma_{\rm B}(k_r, \theta_r) = |V(\mathbf{q})|^2,\tag{8}$$

where  $V(\mathbf{q})$  is the Fourier transform of the potential

$$V(\mathbf{q}) = \frac{1}{2\pi} \int d\mathbf{r} \exp(i\mathbf{q} \cdot \mathbf{r}) V(r), \qquad (9)$$

and **q** is the momentum transfer and its magnitude is  $q = 2k_r \sin(\theta_r/2)$ . In Eq. (7),  $W(k_r)$  is called the returning wave packet which can be interpreted as the momentum distribution of the returning electron beam.

The photoelectron momentum  $\mathbf{p}$  and the momentum  $\mathbf{k}_r$  of the electron after scattering with the parent ion are related by

$$\mathbf{p} = \mathbf{k}_r - \mathbf{A}_r,\tag{10}$$

where  $\mathbf{A}_r$  is the vector potential at the time when electrons return to the origin. In addition, based on the classical simulation [5], the QRS relates

$$k_r = 1.26|A_r|.$$
 (11)

Figure 1 illustrates the relationship between the photoelectron momentum  $\mathbf{p}$  and the momentum  $\mathbf{k}_r$  of the rescattered electron for the case in which the returning electron revisits the parent ion along  $-\hat{z}$ .



**Figure 1.** Relationship between the photoelectron momentum  $\mathbf{p}$  and the momentum  $\mathbf{k}_r$  of the rescattered electron.

The projection of photoelectron momentum in the parallel and perpendicular directions are

$$p^{||} = p \cos \theta = -A_r - k_r \cos \theta_r = -A_r + k_r^{||},$$
  

$$p^{\perp} = p \sin \theta = k_r \sin \theta_r = k_r^{\perp}.$$
(12)

...

The good property of the wave packet  $W(k_r)$  is that it does not depend on the rescattering angle. This property has been well tested. Recently, Cerkic *et al* [10] also presented a derivation of the factorization formula for the back-rescattering ridge, i.e. electrons returning with maximum kinetic energy of about  $3U_p$ , and showed that the cross sections in the first-order Born approximation can be factored out.

The great advantage of Eq. (7) is that in the photoelectron spectra, the wave packet depends only on the laser pulse and the DCS only on the target. This allows one to insert the structure information by replacing  $\sigma_{\rm B}(k_r, \theta_r)$ , calculated within the first-order Born approximation in Eq. (7), by the accurate DCS,  $\sigma(k_r, \theta_r)$ , i.e.,

$$D_{\text{QRS}}(p,\theta) = W(k_r)\sigma(k_r,\theta_r).$$
(13)

The key idea of the QRS model is that, one first evaluates the wave packet  $W(k_r)$  from Eq. (7) using

$$W(k_r) = D_{\text{SFA}}(p,\theta) / \sigma_{\text{B}}(k_r,\theta_r)$$
(14)

at an arbitrary angle, say  $\theta = 170^{\circ}$ , then uses Eq. (13) to obtain the 2D momentum spectra in the whole space. It should be noted that the wave packet only needs to be evaluated at one angle.

# 2.2. Field-free elastic scattering for atoms

The standard potential scattering theory has been well documented in textbooks, see Ref. [11, 12], for example. Here we address elastic scattering of an electron by a spherical potential

$$V(r) = -\frac{Z}{r} + V_{\rm s}(r), \tag{15}$$

where  $V_{\rm s}(r)$  is a short-range potential. For elastic scattering of the returning electron with parent ion, Z = 1. For a modified Coulomb potential in Eq. (15), the scattering amplitude for detecting an electron with energy  $E = k^2/2$  at an outgoing angle  $\theta$  with respect to the incident direction is given by

$$f(\theta) = f_{\rm c}(\theta) + \hat{f}(\theta). \tag{16}$$

The first term in Eq. (16) is the scattering amplitude by a pure Coulomb potential

$$f_{\rm c}(\theta) = -\eta \exp(2i\sigma_0) \frac{\exp\{-i\eta \ln[\sin^2(\theta/2)]\}}{2k \sin^2(\theta/2)},\tag{17}$$

and the second term is given by

$$\hat{f}(\theta) = \sum_{l=0}^{\infty} \frac{2l+1}{k} \exp(2i\sigma_l) \exp(i\delta_l) \sin \delta_l P_l(\cos \theta),$$
(18)

where the  $P_l(\cos \theta)$  are Legendre polynomials,  $\delta_l$  is the phase shift from the short-range potential, and  $\sigma_l$  is the Coulomb phase shift which is given by

$$\sigma_l = \arg[\Gamma(l+1+i\eta)] \tag{19}$$

with  $\eta = -Z/k$ . The elastic scattering DCS is then given by

$$\sigma(k,\theta) = |f_{\rm c}(\theta) + f(\theta)|^2.$$
<sup>(20)</sup>

## 2.3. Field-free elastic scattering for molecules

Electron-molecule interaction potential does not have spherical symmetry. Rigorous theory for electron-molecule scattering is quite complicated. Instead, we examine the independent atom model [13, 14, 15] used in the standard diffraction theory. Within IAM, a molecule is modeled as a collection of its component atoms fixed in space, and the electron-molecule interaction potential is assumed to be the sum of the potentials from each atom, which is centered at each atomic site  $\mathbf{R}_i$  with *i* the index of atom.

According to IAM, the total scattering amplitude for a molecule fixed in space is expressed as a superposition of atomic scattering amplitudes  $f_i$ , as given in Eq. (16),

$$F(k,\theta,\varphi;\Omega_L) = \sum_i f_i e^{i\mathbf{q}\cdot\mathbf{R}_i},\tag{21}$$

where  $\mathbf{q} = \mathbf{k} - \mathbf{k}_0$  is the momentum transfer vector, and  $\Omega_L$  is the orientation/alignment angles of the molecule. The incident electron moves along z-axis with momentum  $\mathbf{k}_0$ , and is scattered into the direction  $\mathbf{k} = (k, \theta, \varphi)$ . The condition  $|\mathbf{k}| = |\mathbf{k}_0|$  tells this is elastic scattering.

Taking the modulus squared of Eq. (21), we have the molecular differential cross section to be the form of

$$I_{\text{tot}}(\theta,\varphi;\Omega_L) = I_A + \sum_{i \neq j} f_i f_j^* e^{i\mathbf{q}\cdot\mathbf{R}_{ij}},\tag{22}$$

which reveals important features of electron diffraction from molecules. The first term on the right, called atomic term, is the incoherent sum of all atomic scattering cross sections in the molecule

$$I_A = \sum_{i} |f_i|^2.$$
 (23)

The second term in Eq. (22) includes all of the interatomic interferences, and is defined as the molecular interference term (MIT). It depends on the molecular geometry, and embeds full molecular structural information, i.e., positions of all atoms in the molecule, in the form of position vector displacement  $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$  between any two atoms *i* and *j*.

For gas-phase electron diffraction (GED), molecule samples are in random orientations such that Eq. (22) should be averaged over all orientations. The recorded electron scattering intensity is then given by

$$\langle I_{\text{tot}} \rangle(\theta) = I_A + \sum_{i \neq j} f_i f_j^* \frac{\sin(qR_{ij})}{qR_{ij}},\tag{24}$$

in which q and  $R_{ij}$  are the moduli of  $\mathbf{q}$  and  $\mathbf{R}_{ij}$ , respectively. It is interesting to note that molecular characteristic interference features are still preserved after the angular average, which was pointed out early by Cohen and Fano in 1966 [16]. This is essential to GED experimentalists, allowing them to pull out interatomic distances.

The IAM has been widely applied in stereochemistry to reconstruct molecules. As the molecular structure parameters are determined from the molecular interference term, we can define a molecular contrast factor (MCF) as the ratio of molecular interference term to atomic interference-free term

$$\gamma = \frac{1}{I_A} \sum_{i \neq j} f_i f_j^* \frac{\sin(qR_{ij})}{qR_{ij}}.$$
(25)

#### 2.4. Field-free (e, 2e)

We consider electron impact ionization of He<sup>+</sup>. Using the prior form, the direct transition amplitude for this (e, 2e) process is expressed by

$$f_{e2e}(\mathbf{k}_1, \mathbf{k}_2) = \langle \Psi_{\mathbf{k}_1, \mathbf{k}_2}^- | V_i | \Psi_{\mathbf{k}_i} \rangle, \qquad (26)$$

where  $V_i$  is the perturbation interaction,

$$V_{i} = \frac{1}{|\mathbf{r}_{1} - \mathbf{r}_{2}|} - \frac{Z_{N}}{r_{1}} - U_{i}(r_{1}), \qquad (27)$$

where  $\mathbf{r}_1$  and  $\mathbf{r}_2$  are the position vectors for the projectile and the bound state electron, respectively. Here, we assume that the He<sup>+</sup> ion is initially in the ground state, and the charge of the nucleus is  $Z_N = 2$ . In Eq. (27),  $U_i(r_1)$  is the initial state distorting potential which is used to calculate the initial state wave function for the projectile.

In (26),  $\Psi_{\mathbf{k}_1,\mathbf{k}_2}^-$  is the final state wave function describing the two outgoing electrons with momenta  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , respectively. In this work, we use the BBK model [19] in which the final continuum state is expressed as

$$\Psi_{\mathbf{k}_1,\mathbf{k}_2}^-(\mathbf{r}_1,\mathbf{r}_2) = (2\pi)^{-3} \exp(i\mathbf{k}_1 \cdot \mathbf{r}_1) \exp(i\mathbf{k}_2 \cdot \mathbf{r}_2)$$
$$\times C(\alpha_1,\mathbf{k}_1,\mathbf{r}_1)C(\alpha_1,\mathbf{k}_2,\mathbf{r}_2)C(\alpha_{12},\mathbf{k}_{12},\mathbf{r}_{12}),$$
(28)

where

$$\mathbf{k}_{12} = \frac{1}{2}(\mathbf{k}_1 - \mathbf{k}_2), \quad \mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2,$$
  
$$\alpha_1 = -\frac{Z_N}{k_1}, \quad \alpha_2 = -\frac{Z_N}{k_2}, \quad \alpha_{12} = \frac{1}{2k_{12}}.$$
 (29)

The Coulomb part of the wave function in Eq. (28) is defined as

$$C(\alpha, \mathbf{k}, \mathbf{r}) = \exp(-\pi\alpha/2)\Gamma(1 - i\alpha)_1 F_1[i\alpha; 1; -i(kr + \mathbf{k} \cdot \mathbf{r})], \qquad (30)$$

where  $\Gamma$  is the gamma function and  ${}_{1}F_{1}$  the confluent hypergeometric function. The Coulomb interaction between the two outgoing electrons included in Eq. (28) can be turned off by setting  $\alpha_{12} = 0$ .

The initial state  $\Psi_{\mathbf{k}_i}$  in (26) is a product of the wave function  $\varphi_{\mathbf{k}_i}(\mathbf{r}_1)$  describing the incident electron with momentum  $\mathbf{k}_i$  and the ground state wave function of He<sup>+</sup>

$$\Psi_{\mathbf{k}_i}(\mathbf{r}_1, \mathbf{r}_2) = \varphi_{\mathbf{k}_i}(\mathbf{r}_1)\phi_{\mathrm{He}^+}(\mathbf{r}_2).$$
(31)

In principle,  $\varphi_{\mathbf{k}_i}(\mathbf{r}_1)$  can be obtained by solving the differential equation

$$\left[-\frac{1}{2}\nabla_1^2 + U_i(r_1) - \frac{1}{2}k_i^2\right]\varphi_{\mathbf{k}_i}(\mathbf{r}_1) = 0,$$
(32)

where the distorting potential  $U_i(r_1)$  is not determined by the formalism. For the current purpose, we simply choose  $U_i(r_1) = 0$  such that a plane wave is obtained to describe the incident electron, i.e.,

$$\varphi_{\mathbf{k}_i}(\mathbf{r}_1) = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{k}_i \cdot \mathbf{r}_1).$$
(33)

Finally, the triple differential cross section (TDCS) for the electron impact ionization process is given by

$$\frac{d^{3}\sigma_{e2e}}{d\Omega_{1}d\Omega_{2}dE} = (2\pi)^{4} \frac{k_{1}k_{2}}{k_{i}} \Big[\frac{3}{4} |f_{e2e}(\mathbf{k}_{1}, \mathbf{k}_{2}) - g_{e2e}(\mathbf{k}_{1}, \mathbf{k}_{2})|^{2} \\ + \frac{1}{4} |f_{e2e}(\mathbf{k}_{1}, \mathbf{k}_{2}) + g_{e2e}(\mathbf{k}_{1}, \mathbf{k}_{2})|^{2}\Big],$$
(34)

where  $\Omega_1(\theta_1, \phi_1)$  and  $\Omega_2(\theta_2, \phi_2)$  are the solid angles of detection of the two electrons leaving the collision with momenta  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , and  $g_{e2e}(\mathbf{k}_1, \mathbf{k}_2)$  is the exchange amplitude with  $g_{e2e}(\mathbf{k}_1, \mathbf{k}_2) = f_{e2e}(\mathbf{k}_2, \mathbf{k}_1)$ .

The TDCS in Eq. (34) is spin-averaged for the usual (e, 2e) collision. We have used Eq. (34) in our previous QRS calculations for NSDI on helium [20]. However, for ionization of He<sup>+</sup> by the impact of a returning electron induced in laser field, the two electrons in the process started in the singlet ground state, and their singlet coupling is preserved during ionization [21]. Consequently, for laser induced (e, 2e) process, the TDCS should be given by

$$\frac{d^3 \sigma_{\text{e2e}}^{\text{Laser}}}{d\Omega_1 d\Omega_2 dE} = (2\pi)^4 \frac{k_1 k_2}{k_i} |f_{\text{e2e}}(\mathbf{k}_1, \mathbf{k}_2) + g_{\text{e2e}}(\mathbf{k}_1, \mathbf{k}_2)|^2.$$
(35)

#### 3. Applications

#### 3.1. Dynamic imaging of molecules with infrared lasers

To image microscopic objects, x-ray diffraction and electron diffraction are the two principal experimental tools for the determination of structure of molecules over many decades. These methods can achieve sub-angstrom spatial resolution, but it is unable to perform time-resolved measurements of molecular dynamics on femtosecond time scale. Although ultrafast electron diffraction (UED) [22, 23] and free-electron x-ray lasers are aiming at improving the temporal resolution, many technical challenges still remain [24].

Alternatively, infared (IR) laser pulse offers the potential for dynamic imaging since the temporal resolution is already in femtoseconds. Moreover, two recent theoretical developments in strong field rescattering physics [4, 5, 25] have made it quite promising for using lasers to image the structure of a molecule. First, using high energy above-threshold-ionization (HATI) spectra, it has been shown that accurate field-free electron-target-ion scattering differential cross sections can be extracted [5, 25]. Second, these HATI electrons are backscattered from the atomic centers in the molecule. The IAM introduced above is adequate to give a good description of those collisions for energies at or above hundreds eV. Such returning energies are much lower than those used in GED, but can be easily obtained with mid-infared (MIR) lasers. At such energies, the corresponding de Broglie wavelength of electrons is less than 1 angstrom, which is short enough to resolve different atoms in a molecule.



Figure 2. (Color online) (a,b,c,e,g) Elastic differential cross sections for  $e^-$ -N<sub>2</sub> collisions at incident energies of 20, 50, 100, 150 and 200 eV, respectively. Solid red curves are IAM simulations, asterisk symbols are experimental data from DuBois *et al* (a,b) and Herrmann *et al* (c,e,g). For energies 100 eV and above, atomic terms are also displayed by dashed green lines, around which the total DCS oscillates. (d,f,h) Molecular contrast factor of IAM (solid red curves) compared with experimental data (asterisk symbols) for energies of 100, 150 and 200 eV, respectively.

3.1.1. The validity of IAM In this section, we examine the validity of the simple independent atom model for elastic electron-molecule collisions since no such data are available for molecular ions as target. The molecular DCS's are taken directly from experimental measurements using conventional collision techniques with electron guns.

In Fig. 2, we compare the DCS's from IAM for elastic collision of N<sub>2</sub> with experimental measurements at energies of 20, 50, 100, 150, and 200 eV, respectively. The experimental data are taken from DuBois *et al* [26] and Herrmann *et al* [27]. The IAM calculations are carried out by adopting the known N-N bond length. The nitrogen atomic potential takes the Yukawa form with a damping factor of 1.695 a.u. We only present the data for scattering angles  $\theta > 60^{\circ}$ . It can be seen from Fig. 2(a) that, at 20 eV, although the IAM reproduces the similar parabolic

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angular dependence as in experiment, the dip position in DCS from IAM is shifted at least  $20^{\circ}$  toward small angles. As shown in Fig. 2(b), much improved agreement between theory and experiment is achieved at 50 eV. Clearly when energy is above 100 eV, the IAM predicts quite reliable DCS's in comparison with the experimental data, as shown in Figs. 2(c,e,g). In the covered angular region, all the DCS's show a broad minimum. This broad minimum is due to the diffraction from individual atoms, i.e., the atomic term. For energies above 100 eV, the atomic terms are also displayed in Figs. 2(c,e,g).

As indicated in Sec.2.3, the broad minimum, the main feature of interference-free atomic term, is of no use for the retrieval of molecular structure information. The molecular interference, i.e., the small oscillations in the total DCS against the smooth atomic DCS, can be clearly seen in Figs. 2(c,e,g). For the purpose of molecular imaging, we are more interested in the molecular interference. To check its accuracy, we subtract the atomic DCS from experimental data and show the difference as the molecular interference term. We renormalize the experimental data to the atomic DCS, such that the mean square of their difference is minimum. In Figs. 2(d,f,h), we display the molecular contrast factors from both experiment and the IAM. Clearly, theoretical MCF's at all energies shows similar behaviors as those from experiment. As the energy increases, the theoretical MCF is almost on top of the experimental one.

The accuracy of DCS predicted with IAM is generally determined by the momentum transfer q. More detailed discussion about this issue can be found in our recent work [18].

Table 1.	Retrieved	N-N bond	length of $N_2$	from experime	ental DCS's at	100  eV, 150  eV,  and
200  eV,  re	espectively,	$\operatorname{compared}$	to the know	a experimental v	values. Errors a	are also indicated.

	N-N bond length (Å)
experiment	1.09 1 161(+6 5%)
150  eV 150  eV	1.039(-4.7%)
200  eV	1.091(+0.1%)

3.1.2. Retrieval of molecular structure Once the validity of IAM has been established, we are ready to retrieve molecular geometry information from the experimental data, assuming that the bond lengths and bond angles are not known. Genetic algorithm (GA) is applied here to search for the global structure parameters in the parametric space. Simple molecule N<sub>2</sub> is taken in the first attempt. The experimental data shown in Fig. 2(c,e,g) for energies at 100, 150, and 200 eV are used as the fitting objects, from which the retrieved N-N bond lengths are 1.161, 1.039, and 1.091, respectively, see Table 1. Compared to the experimental bond length of 1.09 Å, the errors are consistent with our analysis in Sec.3.1.1 on both DCS's and MCF's: 6.5% for DCS at 100 eV is the worst, -4.7% for 150 eV shows IAM works better, and 0.1% for 200 eV is the best where the retrieved N-N bond length is almost on top of the known value.

**Table 2.** Retrieved structural information of  $C_2H_4$  from experimental DCS's at 100 eV and 150 eV, respectively, compared to the known experimental values. Errors are also indicated.

	C-C length (Å)	C-H length (Å)	∠HCH
experiment	1.339	1.087	$117.4^{\circ}$
100 eV 150 eV	1.436(7%) 1.324(-1%)	1.281(18%) 1.075(-1%)	144.4° 105.8°



Figure 3. (Color online) (a) Molecular structure of  $C_2H_4$  in ball-and-stick model, with structure information shown at the bottom. (b,c) Comparison between measured DCS's (red solid squares) of  $C_2H_4$  and corresponding IAM simulations (green full lines) for electron energies at 100 and 150 eV. (d,e) Same as (b,c) but for comparison of molecular contrast factors.

We take ethylene molecule as another example, whose structure is shown in Fig. 3(a).  $C_2H_4$  is a planar molecule and has  $D_{2h}$  symmetry. The structural information, the lengths of the double C-C bond and the C-H bond, and the angle  $\angle$ HCH, are shown at the bottom. In Figs. 3(b,c) the DCS's from IAM simulation are presented in comparison with experimental data at 100 and 150 eV, which are taken from Brescansin *et al* [28]. In both cases, theory matches with experiment quite well. Similar to N<sub>2</sub>, we also show the molecular contrast factors, see Figs. 3(e,f). The overall agreement looks very good, except that the last 3 data points appear to be too high. Fitting to experimental DCS's with GA, the retrieved results are listed in Table 2. At 100 eV, the bond lengths and angles are not well recovered, and the error for C-H bond length is quite big; nevertheless, the C-C bond length is still acceptable. However, the three parameters are all accurately retrieved from DCS at 150 eV. Considering that the structure determination of  $C_2H_4$  is a challenge for GED, as the light atom H has a very small scattering cross section, such accuracy is quite encouraging.

3.1.3. HATI spectrum The checking we have done in the above section shows that the IAM is an adequate theory in the description of electron-molecule collisions at large scattering angles for energies on the order of 100 eV and more, and that it is feasible to apply IAM for molecular structure determination from the DCS of electron-molecule collisions. Based on such tests, we believe that our proposal to image transient molecules with intense infared laser pulses are quite promising. In considering the DCS extracted from the HATI spectra, Eq. (24) has been modified into

$$\langle I_{\text{tot}} \rangle(\theta) = \left(\sum_{i} |f_{i}(\theta)|^{2}\right) \int N(\Omega_{L}) d\Omega_{L} + \sum_{i \neq j} f_{i}(\theta) f_{j}^{*}(\theta) \int e^{i\mathbf{q}\cdot\mathbf{R}_{ij}} N(\Omega_{L}) d\Omega_{L},$$
(36)

where  $N(\Omega_L)$  is the ionization rate for each orientation angle  $\Omega_L$  calculated using MO-ADK theory [29, 30].

Whether we have to refine our proposal, i.e., the application of IAM to molecular ions, we have to evaluate the situation until HATI spectra from MIR lasers become available. For energies of interest here, however, we did observe that the DCS's at large angles for atomic target and atomic ion target are quite close to each other. So far, experiments [31, 32, 33, 34] have reported HATI spectra for  $N_2$ ,  $O_2$ , and  $CO_2$  using 800 nm Ti-Sapphire lasers where the returning electron energies are below 50 eV. At such low energies, the IAM is not expected to work well. Therefore, HATI spectra for molecules measured in laser pulses at longer wavelengths are highly desirable.

3.2. Nonsequential double ionization of helium due to the (e, 2e) processes in strong laser fields Nonsequential double ionization of atoms by linearly polarized laser pulses has drawn considerable interest for over two decades. Experimentally, measurements on the full momentum vectors of the two outgoing electrons have been carried out since 2000's [35, 36, 37, 38, 39, 40, 41, 42]. So far, such experiments have been performed only for 800 nm lasers, and often only the longitudinal momentum distributions, with respect to laser's polarization axis, of the two electrons have been presented. These data have attracted intensive theoretical research including the "S-matrix theory" [43, 44, 45, 46] based on the strong field approximation, and the pure numerical approaches by solving classical equation of motion [47] or the TDSE [40, 48].

In this section, we apply the QRS model to NSDI due to (e, 2e) collisions by the returning electrons induced in a strong laser field.

3.2.1. Quantitative rescattering theory for laser-induced (e, 2e) processes The QRS model has already been successfully applied to obtain the total nonsequential double ionization of Ar atoms by intense laser pulses [49]. The purpose of the present work is to simulate the correlated momentum spectra measured by Staudte *et al* [40].

For the laser-induced (e, 2e) process, the incident electron is actually the recollision electron, which is first tunnel ionized in the laser field and then driven back to recollide with the parent ion. Therefore the momentum  $\mathbf{k}_i$  of the incident electron is

$$k_i = k_r. aga{37}$$

In the correlated two-electron momentum spectra induced in a strong laser field, the experimental measurements were taken for the momentum components along the laser polarization of the two electrons against each other such that the data were integrated over the other components. Correspondingly, to compare with experiment, we integrate the TDCS, for laser-free (e, 2e) process at a given incident energy  $E_i = k_i^2/2$ , over  $\phi_2$  and  $E_2$ 

$$Y_{E_i}^{e,2e}(k_1^{||},k_2^{||}) = \frac{4\pi}{k_1k_2} \int_0^{E_{\max}} dE_2 \int_0^{\pi} d\phi_2 \frac{d^3 \sigma_{e2e}^{\text{Laser}}}{d\Omega_1 d\Omega_2 dE_2} \Big|_{\phi_1=0},$$
(38)

where  $E_{\text{max}} = E_i - I_p$ . Due to the cylindrical symmetry, we set  $\phi_1 = 0$  in Eq. (38). In the actual calculations, the integration over  $\phi_2$  is performed only from 0 to  $\pi$  since the TDCS is symmetric about the plane containing  $\mathbf{k}_1$  and  $\mathbf{k}_i$ .

By shifting the momenta of the two outgoing electrons using the relation in Eq. (10), we obtain the correlated momentum spectra at an incident energy of  $E_i$  for NSDI,

$$D_{E_i}^{e,2e}(p_1^{||}, p_2^{||}) \equiv D_{E_i}^{e,2e}(k_1^{||} - A_r, k_2^{||} - A_r) = Y_{E_i}^{e,2e}(k_1^{||}, k_2^{||}).$$
(39)

Similar to Eq. (13), the final distribution can be evaluated by

$$D_{e,2e}(p_1^{||}, p_2^{||}) = \int_{I_p}^{\infty} dE_i D_{E_i}^{e,2e}(p_1^{||}, p_2^{||}) \bar{W}(E_i),$$
(40)



Figure 4. (Color online) (a) Experimental correlated momentum spectra along the polarization direction for double ionization of helium at 800 nm,  $4.5 \times 10^{14}$  W/cm<sup>2</sup>. The indicated lines are H<sub>2</sub><sup>+</sup> background from cold H<sub>2</sub> in the gas jet. (b) Same as (a), only the (e, 2e) portion is retained. The original data are from Staudte *et al* [40].

where  $\overline{W}(E_i)$  is the volume-integrated wave packets [5], i.e., the wave packet integrated over the volume distribution of the laser pulse at peak intensity  $I_0$ .

3.2.2. Correlated momentum spectra due to (e, 2e) processes in strong laser field Now we apply the QRS model to simulate the correlated two-electron momentum spectra of NSDI on helium in a 800 nm,  $4.5 \times 10^{14}$  W/cm<sup>2</sup> laser field measured by Staudte *et al* [40], in which a fingerlike structure in the correlated electron momentum distribution was observed. The measured correlated momentum spectra from Staudte *et al* are shown in Fig. 4(a).

It has been well recognized that NSDI consists of both (e, 2e) and excitation-tunneling. To compare with the experimental data due to (e, 2e) process only, one needs to separate these two mechanisms. The separation of (e, 2e) and excitation-tunneling can be carried out according to the analysis of the recollision mechanisms in NSDI by Feuerstein *et al* [36]. Based on the analysis, the (e, 2e) events are located in the first and third quadrants only, while the excitation-tunneling events are distributed symmetrically in all four quadrants. Following Feuerstein *et al* [36], by subtracting the corresponding number of excitation-tunneling events from the measured number of all events in the first and third quadrants, we obtain the correlated momentum distributions for the "pure" (e, 2e) process, which are displayed in Fig. 4(b). It should be noted that the original experiment spectra include contamination from the  $H_2^+$  background as indicted by the lines in Fig. 4(a). This background should be removed to ensure that the spectra in the second quadrant are from excitation-tunneling only.

The simplest theoretical model used in the (e, 2e) TDCS calculations in this work is called P-CC in which a plane wave (P) is used to describe the incident electron and two Coulomb waves (C) are used for the outgoing electrons. The calculated P-CC correlated momentum spectra for two outgoing electrons in the recollisional (e, 2e) process on He<sup>+</sup> at an incident energy of 85 eV is displayed in Fig. 5(a). The momentum spectra shows no minimum along the diagonal of equal momenta.

The primary weakness of the P-CC model lies in the fact that the final state electron-electron correlation is contained only in the first order. This interaction is taken into account in the well-known BBK model [19] in which two Coulomb waves are used to describe the two outgoing electrons and an additional Coulomb interaction is employed for the interaction between the two outgoing electrons. In this paper, this model is referred to as P-CCC. The P-CCC result for the



**Figure 5.** (Color online) Theoretical normalized correlated momentum spectra for two outgoing electrons in the recollisional (e, 2e) process on He<sup>+</sup> at an incident electron energy of 85 eV. Shown are the momentum components along the polarization direction. The theoretical models used in the simulation are (a) P-CC and (b) P-CCC. See text for detail.



Figure 6. Volume integrated wave packet against the momentum of the recollision electron (the bottom horizontal axis) for 45 fs (FWHM) laser pulses at peak intensity of  $4.5 \times 10^{14}$  W/cm<sup>2</sup> with wavelength of 800 nm. The arrow indicates the minimum momentum of the recollision electron to ionize the parent ion He<sup>+</sup>. The corresponding energy of the returning electron is also used as the top horizontal axis.

same collision parameters is shown in Fig. 5(b). One can see that the fingerlike structure, which was observed in experiment [40], is clearly reproduced by the P-CCC model. The significant discrepancy in the correlated momentum spectra between P-CC and P-CCC indicates that the Coulomb interaction plays a crucial role in the (e, 2e) process at least for the energy regime considered here.

To obtain correlated electron momentum spectra that can be compared to experimental data, the integration over all possible incident energies weighted by the returning wave packet should be performed using Eq. (40). For the 800 nm laser at peak intensity of  $4.5 \times 10^{14}$  W/cm<sup>2</sup> used by Staudte *et al*, the maximum energy of the returning electron is  $E_i^{\text{max}} = 3.17U_p = 85.6$  eV. The returning electron wave packet is shown in Fig. 6.



Figure 7. (Color online) Momentum spectra for two out-going electrons in the recollisional (e, 2e) process on He<sup>+</sup> at 800 nm,  $4.5 \times 10^{14}$  W/cm<sup>2</sup>. Shown are the momentum components along the polarization direction. The theoretical models used are: (a) P-CC, (b) P-CCC, and (c) DS3C, respectively. (d) Experiment.

The volume-integrated correlated momentum spectra of P-CC and P-CCC are plotted in Figs. 7(a) and (b). Since the spectra in the third quadrant is symmetric with respect to the first quadrant, only the first quadrant spectra are shown. One can see that the final correlated momentum spectra of P-CC and P-CCC are slightly different from the results for fixed incident returning energy shown in Fig. 5 while the main features remain. Again, different from the P-CC results, the P-CCC predicts a fingerlike structure which is in better agreement with the experimental measurements shown in Fig. 7(d).

It has already been recognized that the BBK model is not in agreement with the measurements at low energies because the interaction between each pair of charges is not screened by the presence of the third charge. This deficiency was first corrected by Berakdar and Briggs [50], by introducing an effective Sommerfeld parameter for the case in which the two outgoing electrons have equal energies. Such a modification represents a dynamic screening (DS) of the three-body Coulomb interactions and hence the improved BBK model is called DS3C. The DS3C model was later generalized by Chen *et al* [51] for any geometry and energy sharing. In this work, we also performed the DS3C calculations and the final correlated momentum spectra are shown in Fig. 7(c). A comparison of P-DS3C with P-CCC shows that the two separated peaks in the spectra of P-CCC is connected by an almost evenly distributed bridge. This indicates the effect of screening included in the P-DS3C model, which suppresses the strong electron-electron Coulomb repulsion.

# 4. Conclusion

In conclusion, we have studied the elastic scattering of the laser-induced returning electrons with the parent molecular ion. From high energy photoelectron spectra, laser-free scattering DCS can be extracted. Using traditional electron-molecule collision data, we show that IAM gives reasonably good predictions on the scattering DCS for large scattering angles when electron energy is above 100 eV, and the bond lengths of  $N_2$  and  $C_2H_4$  are successfully retrieved. Based on the QRS and IAM, the feasibility of self-imaging of transient molecules with intense infrared laser pulses has been established.

The NSDI on helium in an intense laser field due to the (e, 2e) process has also been studied. We obtain the correlated momentum spectra for the two outgoing electrons by multiplying the returning electron wave packet with the differential cross sections for laser-free (e, 2e) reactions. It turns out that the post-collision interaction between the two outgoing electrons is crucial in reproducing the finger-like structure in the correlated momentum space observed in the experiment.

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