Time ordering of two-step processes in energetic ion-atom collisions: Basic formalism

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The semiclassical approximation is applied in second order to describe time ordering of two-step processes in energetic ion-atom collisions. Emphasis is given to the conditions for interferences between first- and second-order terms. In systems with two active electrons, time ordering gives rise to a pair of associated paths involving a second-order process and its time-inverted process. Combining these paths within the independent-particle frozen orbital model, time ordering is lost. It is shown that the loss of time ordering modifies the second-order amplitude so that its ability to interfere with the first-order amplitude is essentially reduced. Time ordering and the capability for interference is regained, as one path is blocked by means of the Pauli exclusion principle. The time-ordering formalism is prepared for papers dealing with collision experiments of single excitation [Stolterfoht *et al.*, following paper, Phys. Rev. A **48**, 2986 (1993)] and double excitation [Stolterfoht *et al.* (unpublished)].

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I. INTRODUCTION

Time ordering is a well-known phenomenon in time-dependent perturbation theory, which describes a multistep processes by means of higher-order terms of the Dyson series [1,2]. The time ordering of a multistep process is provided by the time-ordering operator whose action is responsible for the occurrence of a certain step before the next one can take place. Multistep processes are important in ion-atom collisions where electronic excitation proceeds via appropriate intermediate states. A convenient tool, describing energetic ion-atom collisions, is the semiclassical approximation (SCA) [3,4], which is based on time-dependent perturbation theory. Therefore, multistep processes in ion-atom collisions are expected to be well represented by higher-order terms of the SCA. Nevertheless, the theoretical work, treating higher-order terms of the SCA, is rather limited in the field of ion-atom collisions.

Conceptually, it is useful to distinguish two categories of time-ordering phenomena. First, in a one-electron system or in a multielectron system with one active electron, time ordering is essential. For instance, the hydrogen atom in the ground state cannot undergo the transition $2s \rightarrow 2p$ before the transition $1s \rightarrow 2s$ has occurred. Second, in a multielectron system with two active electrons, time-ordering effects are less important as the independent-particle model (IPM) is valid. In fact, time ordering is expected to be lost, as the interplay between the electrons is completely disregarded. Then, the action of one electron may take place before or after the action of the other electron. However, it should be kept in mind that the loss of time ordering occurs only under restrictive conditions. The action of one electron may be influenced by the other electron due to dynamic screening and correlation effects. Hence it is expected that time-ordering effects are probing in detail the electron-electron interaction.

The dynamics of the electron-electron interaction has recently received particular attention in the field of atomic collisions [5–7]. The major part of the electron-electron interaction can be treated as a mean field which is incorporated in the IPM [1]. This mean field is associated with one-body operators and hence it may be considered as the monoelectronic aspect of the electron-electron interaction. The residual part, not incorporated in the IPM, is responsible for phenomena of electron correlation. Since this part is represented by two-body operators, it is referred to as dielectronic interaction [8]. It should be noted that, in ion-atom collisions, the concept of electron correlation is still a matter of controversy [5–7], as the borderline between the monoelectronic and dielectronic aspects of the electron-electron interaction is not easy to draw.

Recently, McGuire and Straton [9] have studied for the first time the relationship between electron-correlation and time-ordering phenomena. They have shown that electron correlation and/or time ordering are necessary conditions for interferences between first- and second-order processes. Also, using different arguments based on time reversal, Briggs and Macek [10] have shown that interferences between first- and second-order terms cancel in rather general cases. In the past few years, particular interest have been focused on interference effects between first- and second-order mechanisms in the process of double ionization of He [5]. In this case interferences between the two-step process TS1 involving *one* nucleus-electron interaction followed by an electron-electron interaction and the two-step process TS2 involving *two* nucleus-electron interactions has been studied in detail [11,12]. Similar interference effects have been considered in the process of double excitation [13] and they have been searched for experimentally [14,15]. However, for double excitation, the interference term could not conclusively be verified. Hence it appears that more detailed information is needed about the process of double excitation. In particular, phenomena of time ordering is felt to deserve further consideration in ion-atom col-

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lisions. It should be noted that the two step processes TS1 and TS 2 provide illuminating examples for time ordering. In TS1, time ordering is dominant, whereas time ordering is less relevant for TS2.

In this work, the semiclassical approximation up to second order is used to describe the principles of time-ordered two-step processes. A two-step process is considered to proceed via a path involving a certain intermediate state. For a system with two active electrons, double excitation involves two associated paths whose time ordering is opposite. Within the framework of the independent-particle frozen-orbital model, it is formally shown that the associated paths can be combined so that the time ordering is lost. Furthermore, it is revealed that the loss of time ordering is accompanied by the cancellation of important parts of the corresponding transition amplitudes [16]. The reduction of the transition amplitudes is found to diminish strongly the interference between first- and second-order processes. Finally, conditions are discussed to inhibit the loss of time ordering. It is pointed out that the Pauli blocking mechanism [17,18] can be used to avoid the cancellation of interference effects. Experimental evidence for the present analysis will be given in papers that are devoted to the study of single excitation [19] and double excitation [20] in energetic ion-atom collisions.

II. TIME ORDERING IN THE SEMICLASSICAL APPROXIMATION

In the present analysis, the electronic process of excitation is described using the semiclassical approximation [3,4]. Emphasis is given to phenomena associated with second-order terms. The SCA is based on the partition of the electronic Hamiltonian $H(t)=H^a+V(t)$ where $H^a=H(t\rightarrow\infty)$ is the asymptotic (or atomic) Hamiltonian and $V(t)$ is the time-dependent perturbation relevant during the scattering. The asymptotic states Φ_j^a of the collision problem are obtained from the stationary Schrödinger equation

$$H^a|\Phi_j^a\rangle=E_j^a|\Phi_j^a\rangle \tag{1}$$

where E_j^a are the eigenenergies of the atomic Hamiltonian H^a. The amplitude for transitions from the initial to the final state is given by [1,2]

$$A_{if}=\left\langle\Phi_f^a\left|\mathcal{T}\exp\left[-i\int V(\tau)d\tau\right]\right|\Phi_i^a\right\rangle \tag{2}$$

where $\mathcal{T}$ is the time-ordering operator whose effects shall be studied in this work. In an approximation, we consider the SCA transition amplitude up to second order:

$$A_{if}\approx A_{if}^{(1)}+A_{if}^{(2)} \tag{3}$$

where $A_{if}^{(1)}$ and $A_{if}^{(2)}$ are the first- and second-order terms of the time-dependent perturbation theory expansion. The truncation of the SCA series after the second term is justified, if the second-order term is noticeable and the third and higher-order terms are negligible. Hence the present analysis implies the following conditions:

$$|A_{if}^{(1)}|\gtrsim|A_{if}^{(2)}|\ ,\quad |A_{if}^{(1)}|\gg|A_{if}^{(n>2)}|\ . \tag{4}$$

Also, it is assumed that the *squared* second-order amplitude is sufficiently small so that it can be neglected. In general, these conditions can be fulfilled in collision experiments by means of high incident energies for which the perturbation becomes small.

In the semiclassical approximation, the first-order amplitude is given by [1]

$$A_{if}^{(1)}=-i\int_{-\infty}^{\infty}V_{if}(\tau)e^{-i\omega_{if}\tau}d\tau \tag{5}$$

where $\omega_{jj'}=E_{j'}^a-E_j^a$ are transition energies obtained by means of Eq. (1) and $V_{jj'}$ are coupling matrix elements obtained as

$$V_{jj'}(t)=\langle\Phi_{j'}^a|V(t)|\Phi_j^a\rangle\ . \tag{6}$$

These coupling matrix elements have specific symmetries. It is assumed that the initial, intermediate, and final states have parity as a good quantum number and they are located at one center as it is characteristic for the process of excitation treated in this work. Then, the matrix elements are *even* or *odd* with respect to time variation, depending on the change of parity and the magnetic quantum number. The symmetries of the matrix elements are shown in Table I.

The second-order amplitude is obtained as an expansion $A_{if}^{(2)}=\sum_k A_{if}^k$ over individual second-order terms

$$A_{if}^k=-\int_{-\infty}^{\infty}V_{kf}(\tau)e^{-i\omega_{kf}\tau}d\tau\int_{-\infty}^{\tau}V_{ik}(\tau')e^{-i\omega_{ik}\tau'}d\tau'\ . \tag{7}$$

The label k runs over the eigenfunction spectrum of H^a. It should be emphasized that the amplitude A_{if}^k describes a two-step process involving the time-ordered transitions, i.e., $i\rightarrow k$ is followed by $k\rightarrow f$. This time ordering is provided by the τ dependence of the second integral (via the upper integration limit). Hence the term A_{if}^k is also referred to as *time-ordered amplitude.*

Interference effects are governed by the phases of the matrix elements. To obtain information about these phases it is useful to consider separately their real and imaginary parts [16]. It is assumed that the initial and final wave functions are real as is the case for bound states involved in the case of an excitation process. From the relation $e^{i\varphi}=\cos\varphi+i\sin\varphi$ one obtains the real and

TABLE I. Even or odd symmetry of the matrix elements V_{if} produced by the interaction V. In parentheses are also given the real or imaginary value of the corresponding transition amplitude $A_{if}^{(1)}$. The quantity $\Delta\Pi$ denotes the change of parity and ΔM denotes the change of magnetic quantum number in the transition from the initial to the final state.

Transition	$\Delta\Pi$	$\Delta M=0$	$\Delta M=1$	$\Delta M=2$
Monopole	No	even (imaginary)		
Dipole	Yes	odd (real)	even (imaginary)	
Quadrupole	No	even (imaginary)	odd (real)	even (imaginary)

imaginary part of the first-order transition amplitude:

$$\mathrm{Re}(A_{if}^{(1)}) = \int_{-\infty}^{\infty} V_{if}(\tau)\sin(\omega_{if}\tau)d\tau \tag{8}$$

and

$$\mathrm{Im}(A_{if}^{(1)}) = -\int_{-\infty}^{\infty} V_{if}(\tau)\cos(\omega_{if}\tau)d\tau\ . \tag{9}$$

It can readily be shown that either the expression (8) or (9) cancels, depending on whether the coupling matrix element V_{if} is *even* or *odd*, respectively. Therefore, due to the symmetry of the matrix elements V_{if} (Table I), the amplitude $A_{if}^{(1)}$ is either real or imaginary. The complex properties of the first-order amplitudes are summarized in Table I.

For the second-order terms, the real and imaginary part is obtained as

$$\begin{aligned}\mathrm{Re}(A_{if}^{k}) = &-\int_{-\infty}^{\infty} V_{kf}(\tau)\cos(\omega_{kf}\tau)d\tau \\ &\times \int_{-\infty}^{\tau} V_{kf}(\tau')\cos(\omega_{ik}\tau')d\tau' \\ &+ \int_{-\infty}^{\infty} V_{ik}(\tau)\sin(\omega_{kf}\tau)d\tau \\ &\times \int_{-\infty}^{\tau} V_{ik}(\tau')\sin(\omega_{ik}\tau')d\tau'\end{aligned} \tag{10}$$

and

$$\begin{aligned}\mathrm{Im}(A_{if}^{k}) = &-\int_{-\infty}^{\infty} V_{kf}(\tau)\cos(\omega_{kf}\tau)d\tau \\ &\times \int_{-\infty}^{\tau} V_{ik}(\tau')\sin(\omega_{ik}\tau')d\tau' \\ &- \int_{-\infty}^{\infty} V_{kf}(\tau)\sin(\omega_{kf}\tau)d\tau \\ &\times \int_{-\infty}^{\tau} V_{ik}(\tau')\cos(\omega_{ik}\tau')d\tau'\ .\end{aligned} \tag{11}$$

No simple rules exist for the complex properties of the second-order transition amplitudes A_{if}^{k}. In this case it is important to note that both a real and an imaginary part contribute to the transition amplitude. *A priori* it is difficult to predict whether the real or the imaginary part dominates. However, we are primarily interested in fast collisions. For high incident energies it follows that the terms including the cosine function are expected to be significantly larger than those including the sine function. This is due to the fact that $\omega_{if}\tau \ll 1$ and thus $\sin(\omega_{if}\tau) \ll 1$ for small collision times. Therefore, the terms in Eqs. (10) and (11) containing sine functions are generally small. Consequently, the first term in Eq. (10) is most important, as it contains only cosine functions. This term contributes to the *real* part $\mathrm{Re}(A_{if}^{k})$ of the transition amplitude. Hence it follows for high velocities that the second-order amplitude is essentially real regardless of whether the magnetic quantum number $M=0$ or 1 are excited.

As an example, we consider the excitation process in a one-electron system. Figure 1(a) shows a two-step process attributed to a second-order amplitude. It is recalled that the second-order amplitude is essentially real in fast collisions. Furthermore, at high energies, dipole transitions are dominant in the first-order process, also shown in Fig. 1(a). It follows from Table I that the related first-order amplitude is real for $\Delta M=0$. Therefore, for high energies and $\Delta M=0$, favorable conditions are created for interference effects between first- and second-order amplitudes in the one-electron system. On the other hand, interference effects are small for $\Delta M=1$ since the first-order amplitude is imaginary (Table I) whereas the second-order amplitude remains essentially real. In the following we shall focus on the $\Delta M=0$ transition, which is relevant for the 0° Auger method used experimentally in Refs. [19] and [20]. In this case it is important to note that significant interferences can be expected for the one-electron system.

III. LOSS OF TIME ORDERING WITHIN THE IPM

The conclusion of significant interferences in one-electron systems does not necessarily hold for many-electron systems. In fact, the situation changes completely since two active electrons participate in the process of single excitation. When the electrons are treated within the framework of the independent-particle model, the transition probability for a two-electron transition is given by the product of the related single-particle transition probabilities [9]. In the following it will be shown that this product is based on a pair of time-ordered transitions involving significant cancellation of real or imaginary parts in the related amplitudes. In particular, it is shown that these cancellations results in a loss of the time ordering in the corresponding transition amplitudes.

The following analysis shall be performed in terms of

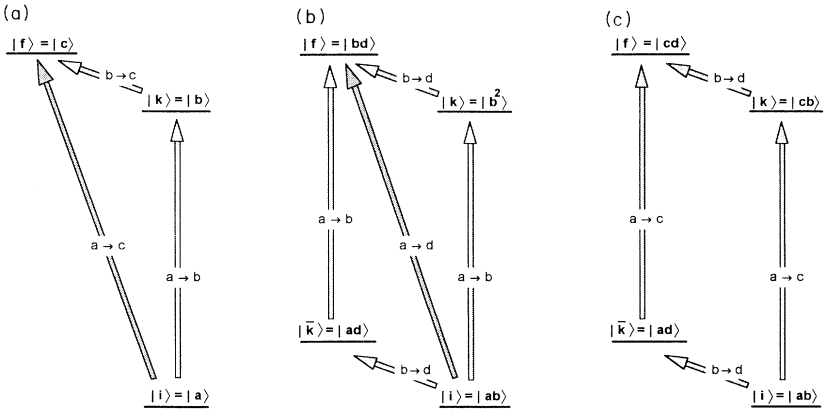

FIG. 1. Associated paths leading from the initial state $|i\rangle$ to the final state $|f\rangle$ via the intermediate states $|k\rangle$ and $|\bar{k}\rangle$ for which the order of the one-electron transitions is interchanged. In (a) a single-electron system is treated. In (b) and (c), respectively, single excitation and double excitation is exhibited for a system of two active electrons.

the diagram in Figs. 1(b) and 1(c), which refer to the processes of single and double excitation, respectively. The attention is focused on the diagram for double excitation [Fig. 1(c)] which represents the more general case and which exhibits all characteristic features of second-order transitions. In fact, the diagram in Fig. 1(b) is obtained from that in Fig. 1(c) by specifying the electron in orbital φ_c, i.e., by setting $c=b$. Within the independent-particle model, the initial, intermediate, and final states are obtained as single configuration states. Hence, as shown in Fig. 1(c), the initial and final state are given by

$$\Phi_i^0=\det(\varphi_a,\varphi_b,\ldots)\ ,\quad \Phi_f^0=\det(\varphi_c,\varphi_d,\ldots) \tag{12}$$

where the symbol det stands for the (normalized) Slater determinant and φ_a, φ_b, φ_d, and φ_d are single-electron orbitals associated with the active electrons in the initial and final state. Figure 1(c) indicates that two intermediate states are important where one electron did undergo a transition and the other one did not:

$$\Phi_k^0=\det(\varphi_c,\varphi_b,\ldots)\ ,\quad \Phi_{\bar{k}}^0=\det(\varphi_a,\varphi_d,\ldots)\ . \tag{13}$$

The paths attributed to these intermediate states labeled k and $\bar{k}$ will be referred to as *associated paths* [Fig. 1(b)]. The related time-ordered amplitudes are given by

$$A_{if}^{k}=-\int_{-\infty}^{\infty}V_{kf}(\tau)e^{-i\omega_{kf}\tau}d\tau\int_{-\infty}^{\tau}V_{ik}(\tau')e^{-i\omega_{ik}\tau'}d\tau' \tag{14}$$

and

$$A_{if}^{\bar{k}}=-\int_{-\infty}^{\infty}V_{\bar{k}f}(\tau)e^{-i\omega_{\bar{k}f}\tau}d\tau\int_{-\infty}^{\tau}V_{i\bar{k}}(\tau')e^{-i\omega_{i\bar{k}}\tau'}d\tau' \tag{15}$$

In the following, we shall consider the sum of amplitudes

$$A_{if}^{k\bar{k}}=A_{if}^{k}+A_{if}^{\bar{k}}\ . \tag{16}$$

This *double-path amplitude* has particular properties as the electron orbitals are frozen, i.e., that they are not influenced by changes of the mean field postulated within the framework of the independent particle model. From the "frozen" energies it follows that

$$\omega_{ik}=\omega_{\bar{k}f}=\omega_{ac}\ ,\quad \omega_{i\bar{k}}=\omega_{kf}=\omega_{bd} \tag{17}$$

where ω_{ac} and ω_{bd} are the energy differences for the single-electron orbitals involved. Moreover, from the frozen wave functions it follows that

$$V_{ik}=V_{\bar{k}f}=V_{ac}\ ,\quad V_{i\bar{k}}=V_{kf}=V_{bd} \tag{18}$$

where $V_{ac}=\langle\varphi_c|v|\varphi_a\rangle$ and $V_{bd}=\langle\varphi_d|v|\varphi_b\rangle$ are single-electron matrix elements. The single-particle operator v is the part of V which is relevant for the active electron. As expected, within the frozen-orbital approach, the multielectron matrix elements reduce to single-electron matrix elements.

Using the identities (17) and (18) and the well-known rule for partial integration $\int f\,dx\int g\,dy=\int(f\int g\,dy)dx+\int(g\int f\,dx)dy$ it follows for the double-path amplitude that

$$A_{if}^{k\bar{k}}=-\int_{-\infty}^{\infty}V_{bd}(\tau)e^{-i\omega_{bd}\tau}d\tau\int_{-\infty}^{\infty}V_{ac}(\tau)e^{-i\omega_{ac}\tau}d\tau\ . \tag{19}$$

It is seen that the upper integration limit is extended to infinity and thus the τ dependence of the second integral has vanished. Consequently, the time ordering is lost in the transition amplitude. Accordingly, by introducing the single-electron amplitudes

$$\begin{aligned}a_{ac}&=-i\int_{-\infty}^{\infty}V_{ac}(\tau)e^{-i\omega_{ac}\tau}d\tau\ ,\\ a_{bd}&=-i\int_{-\infty}^{\infty}V_{bd}(\tau)e^{-i\omega_{bd}\tau}d\tau\end{aligned} \tag{20}$$

one obtains a product form for the sum of associated amplitudes

$$A_{if}^{k\bar{k}}=a_{ac}a_{bd}\ . \tag{21}$$

In fact, this product amplitude is expected within the framework of the independent-particle model. Here, it is shown that the essential feature of the independent-particle approach involving frozen orbitals is the loss of time ordering. Hence, if the double-path amplitude $A_{if}^{k\bar{k}}$ can be factored, it is also referred to as *non-time-ordered amplitude.*

The important point to be made here is that the *time-ordered amplitudes* A_{if}^{k} and $A_{if}^{\bar{k}}$ are generally complex, whereas the *non-time-ordered amplitude* $A_{if}^{k\bar{k}}$ is either real or imaginary. This, in turn, follows from Eq. (21) and the fact that the first-order amplitudes a_{ac} and a_{bd} are either real or imaginary (Table I). When the non-time-ordered amplitude is formed, significant interference effects occur. The fact that the coherent sum $A_{if}^{k\bar{k}}=A_{if}^{k}+A_{if}^{\bar{k}}$ is real or imaginary implies that either the sum $\mathrm{Im}(A_{if}^{k})+\mathrm{Im}(A_{if}^{\bar{k}})$ or $\mathrm{Re}(A_{if}^{k})+\mathrm{Re}(A_{if}^{\bar{k}})$ is completely canceled.

These cancellations may affect the dominant part of the transition amplitude. As noted before, in fast collisions, the real parts $\mathrm{Re}(A_{if}^{k})$ and $\mathrm{Re}(A_{if}^{\bar{k}})$ are most important. These real parts, however, are canceled if the related two-electron process involves, e.g., a dipole and a monopole transition as can readily be shown by means of Eq. (21) and Table I. The cancellation of dominant parts of the second-order amplitudes may strongly affect their capability to interfere with the corresponding first-order amplitude. In the process of single excitation [Fig. 1(b)] involving a second-order monopole plus dipole transition, the final state may also be reached by a first-order transition of dipole type which involves a real amplitude (Table I). As a consequence the first-order amplitude cannot interfere with the second-order amplitude due to the fact that it is imaginary. This inability of interference is a direct consequence of losing the time ordering in the second-order amplitude.

It should be emphasized that the loss of time ordering is achieved under rather restrictive conditions. This loss is based on the validity of the independent-particle model involving frozen orbitals (IPM-FO). Hence the observation of time-ordering effects may be an indication for the breakdown of the IPM-FO due to orbital-relaxation and/or electron-correlation effects. In addition, the Pauli

exclusion principle may create time ordering. It is evident that the associated path becomes closed as the corresponding intermediate state cannot be formed due to the Pauli exclusion principle. In this case the cancellation of amplitude pairs does not occur and the related second-order amplitude remains time ordered. The closing of an associated path is referred to as *Pauli blocking* [17,18]. As the Pauli blocking mechanism retains the time ordering, it may become responsible for an interference between first- and second-order processes.

More information about Pauli blocking will be given in the following paper [19], which is devoted to experimental studies of time-ordering effects in ion-atom collisions at high energies.

IV. CONCLUDING REMARKS

In this work, basic properties of time ordering in ion-atom collisions are analyzed in second order. Time ordering of multistep processes is provided by the time-ordering operator used in perturbation theory. Hence the formal aspect of time ordering is well known. Moreover, a rich variety of time-ordering phenomena are expected to occur in ion-atom collisions. However, in the field of ion-atom collisions, the theoretical work concerning time-ordering effects is limited. This may partially be due to the fact that a higher-order term of the time-dependent perturbation theory are scarcely used in atomic collisions. Indeed, experimentally, it is difficult to elicit cases where the second-order term is significant without having contributions from all the other higher-order terms. In this case, a nonperturbative theory is favorable, such as the coupled-channel method [3], which has extensively been used in the past. It should be noted that time ordering is incorporated in the coupled-channel method. However, to observe specific low-order terms and their possible interferences, particular effort is required.

An important case, attributed uniquely to second order, refers to two active electrons which undergo, successively or simultaneously, first-order transitions. However, in this case, time ordering is less relevant. In fact, under restrictive conditions involving the independent-particle frozen-orbital model, time ordering is completely lost. At this point, the question may arise as to how independent the electrons in the independent-particle model are. This model does not neglect all interactions between the electrons. Rather it includes effects of an electronic mean field [1]. For instance, within the independent-particle model it is evident for two-electron transitions that the removal of the second electron implies a binding energy which is increased due to the removal of the first one; see, e.g., [21]. These changes in energy are due to mean-field (or screening) effects which may create time ordering in the two-step processes. Furthermore, apart from mean-field effects, the Pauli exclusion principle and electron correlation phenomena induce time-ordering. Hence it is expected that the verification of time-ordering phenomena contributes to a better understanding of the independent-particle model and electron correlation.

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