Generalized gauge-invariant formulations of the strong-field approximation

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The gauge problem in the so-called strong-field approximation (SFA) describing atomic or molecular systems exposed to intense laser fields is investigated. By introducing a generalized gauge and partitioning of the Hamiltonian, it is demonstrated that the *S*-matrix expansion obtained in the SFA depends on both gauge and partitioning in such a way that two gauges always yield the same *S*-matrix expansion, if the partitioning is properly chosen.

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I. INTRODUCTION

Gauge invariance is one of the fundamental concepts of electrodynamics. As a consequence it is, e.g., possible to formulate the interaction of charged particles with electromagnetic fields in different gauges. Although the choice of the gauge clearly influences parameters like the scalar or the vector potential, all physical quantities (observables) are independent of the gauge, if a complete treatment is performed. On the other hand, an approximate treatment often leads to gauge-dependent predictions for physical observables. One prominent example is given by the so-called strong-field approximation (SFA) for describing atomic or molecular systems exposed to intense laser fields, which is also known as Keldysh-Faisal-Reiss (KFR) theory [1–3]. It is based on an (infinite) series expansion of the exact *S* matrix describing the interaction of an atomic system with a laser pulse. Besides a phase factor, the *S* matrix obtained from a calculation of the complete expansion should thus be independent of the chosen gauge, provided the series converge. Consequently, the corresponding observable transition probabilities (obtained from the squared absolute values of the *S*-matrix elements) should be gauge independent.

This gauge independence of physical observables is usually lost, if only a truncated series is considered. This is the case for the SFA which is defined as the first term of the *S*-matrix expansion. In a number of investigations it has been shown that transition probabilities or rates predicted by either the length- or the velocity-gauge formulation of the SFA differ easily by one or two orders of magnitude for experimentally relevant laser parameters [4]. Furthermore, it was shown that the velocity-gauge SFA rate does not converge to the tunneling limit for weak fields, if long-range Coulomb interactions are present [5]. Recently, it was also demonstrated that there are pronounced qualitative differences between the energy distributions of the electrons ejected from, e.g., the $2p$ state of hydrogen atoms exposed to intense laser fields, if they are calculated within the SFA in either the length or the velocity gauge [6]. The recent extensions of the SFA to molecular systems in velocity gauge [7], length gauge [8], or dressed length gauge [9] indicate similar or even more pronounced gauge dependencies for molecules. These findings have intensified the discussions whether the formulation of the SFA in one of the two gauges may be "superior" to the other. One approach to answer this question is rather pragmatic and based on a direct comparison of the SFA predictions in both gauges to either exact (numerical) solutions of the full time-dependent Schrödinger equation [6,10] or to experimental results [11,12]. Clearly, if there is no *a priori* reason that one of the two SFA formulations is superior to the other, the conclusions may vary depending on the considered atomic or molecular system (even its quantum state) as well as on laser-pulse parameters.

A second line of argumentation in favor of one of the gauges is based on the question of "universality." For example, it has been argued that an evident limitation of the length-gauge formulation is the fact that in this case the predicted observables depend only on the scalar potential and thus a one-dimensional parameter, while the full description of an electromagnetic field requires in principle more than one dimension [13]. Very recently, Faisal proposed a "gauge-invariant" intense-field *S*-matrix theory that yields equal transition probabilities in the length or velocity gauge, independent of the order of expansion [14]. Consequently, Faisal claims that his theory overcomes the above-mentioned long-standing discrepancy between the SFA in the two gauges. According to the findings in [14] the "gauge-invariant" and thus universal *S*-matrix theory appears to be equivalent to the traditional length-gauge formulation. Specifically, the first-order term reproduces exactly the Keldysh result [1] which was obtained in the length gauge. In view of the popularity of the SFA for describing atomic and molecular ionization in intense laser fields (see, e.g., [15] or [16] and references therein), this is an important result.

Besides the evident appeal of a universal *S*-matrix formulation which would provide an end to the long-lasting debates on the choice of the appropriate gauge (for a very recent example, see [17–19]), it appears, however, quite surprising that such a formulation should exist. In fact, Faisal derives in [20] an alternative *S*-matrix expansion that reproduces in length and velocity gauge the traditional velocity-gauge result. Inspired by the results in [14,20], it is shown in the present work that it is possible to achieve an *S*-matrix expansion in agreement to any traditional SFA formulation in either length, velocity, or radiation gauge for an arbitrary choice of the gauge, if the Hamiltonian is correspondingly partitioned. In fact, introducing a generalized gauge transformation that includes the mentioned particular gauges as special cases, an in principle infinite set of different *S*-matrix expansions can be formulated. All of them can be shown to be achievable within any gauge as long as a proper partition-

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ing of the Hamiltonian is performed. Furthermore, the introduction of the generalized gauge allows us to clearly demonstrate how the choice of the gauge and the partitioning of the Hamiltonian describing the atomic or molecular system exposed to a laser field are connected with each other. This provides a much deeper insight into the gauge problem of SFA that in fact turns out to be more properly described as an expansion problem.

In order to provide a clear definition of terms and notations, the following section gives a brief discussion on (local) gauge invariance. Most importantly, a generalized gauge is introduced and the wave function of a free electron in an electromagnetic field in this gauge is given. Equipped with these prerequisites, Sec. III discusses the S-matrix theory in different gauges. It represents thus the main results of this work, in which it is demonstrated how various results (S-matrix expansions) can be obtained using different combinations of gauge and partitioning. Atomic units $\hbar = m_e = |e| = 1$ are used throughout. In particular, -1 is used for the charge of an electron.

II. GENERALIZED GAUGE

In the validity regime of the SFA, the laser intensities are so high that the photon density is also very high and thus *semiclassical* theory can be used in which the radiation field is treated classically, but the atomic or molecular system is described using quantum mechanics. The influence of the quantum system on the external field is also neglected.

A classical electromagnetic field is described by electric and magnetic field vectors $\mathbf{F}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$ or, alternatively, by the scalar and vector potentials $\Phi(\mathbf{r},t)$ and $\mathbf{A}(\mathbf{r},t)$. Consider a system consisting of an electron in a electrostatic potential $U(\mathbf{r})$ created by a nucleus (or some nuclei) which interacts with an external electromagnetic field. In semiclassical theory, the evolution of the system is governed by the time-dependent Schrödinger equation that satisfies *local gauge invariance* and is given in the coordinate representation with the minimal-coupling Hamiltonian of the system, $\hat{H}_\chi$, by

$$i\frac{\partial}{\partial t}\Psi_\chi(\mathbf{r},t) = \hat{H}_\chi\Psi_\chi(\mathbf{r},t) = \left(\frac{1}{2}[\mathbf{p}_c + \mathbf{A}_\chi(\mathbf{r},t)]^2 - \Phi_\chi(\mathbf{r},t) + U(\mathbf{r})\right)\Psi_\chi(\mathbf{r},t), \tag{1}$$

where the subscript denotes the used gauge χ and the operator of canonical momentum is given independently of the gauge as $\mathbf{p}_c \equiv -i\nabla$, a consequence of the definition of the minimal-coupling Hamiltonian. The word *invariance* means that if the wave function Ψ_χ and both potentials $\mathbf{A}_\chi$ and Φ_χ are simultaneously transformed into a new gauge χ' using the transformation recipes

$$\Psi_{\chi'}(\mathbf{r},t) = \Psi_\chi(\mathbf{r},t)e^{[iT_{\chi\to\chi'}(\mathbf{r},t)]}, \tag{2}$$

$$\Psi_\chi(\mathbf{r},t) = \Psi_{\chi'}(\mathbf{r},t)e^{[iT_{\chi'\to\chi}(\mathbf{r},t)]},$$

$$\mathbf{A}_{\chi'}(\mathbf{r},t) = \mathbf{A}_\chi(\mathbf{r},t) - \nabla T_{\chi\to\chi'}(\mathbf{r},t), \tag{3}$$

$$\mathbf{A}_\chi(\mathbf{r},t) = \mathbf{A}_{\chi'}(\mathbf{r},t) - \nabla T_{\chi'\to\chi}(\mathbf{r},t),$$

$$\Phi_{\chi'}(\mathbf{r},t) = \Phi_\chi(\mathbf{r},t) + \frac{\partial}{\partial t}T_{\chi\to\chi'}(\mathbf{r},t), \tag{4}$$

$$\Phi_\chi(\mathbf{r},t) = \Phi_{\chi'}(\mathbf{r},t) + \frac{\partial}{\partial t}T_{\chi'\to\chi}(\mathbf{r},t),$$

Eq. (1) is transformed into itself, but with $\Psi_\chi \to \Psi_{\chi'}$, $\mathbf{A}_\chi \to \mathbf{A}_{\chi'}$, and $\Phi_\chi \to \Phi_{\chi'}$. All physical quantities like the probability $P(\mathbf{r},t)$ or the electric and magnetic fields $\mathbf{F}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$ are gauge independent, i.e.,

$$P(\mathbf{r},t) = |\Psi_\chi(\mathbf{r},t)|^2 = |\Psi_{\chi'}(\mathbf{r},t)|^2, \tag{5}$$

$$\mathbf{F}(\mathbf{r},t) = -\nabla\Phi_\chi(\mathbf{r},t) - \frac{\partial}{\partial t}\mathbf{A}_\chi(\mathbf{r},t) = -\nabla\Phi_{\chi'}(\mathbf{r},t) - \frac{\partial}{\partial t}\mathbf{A}_{\chi'}(\mathbf{r},t), \tag{6}$$

$$\mathbf{B}(\mathbf{r},t) = \nabla\times\mathbf{A}_\chi(\mathbf{r},t) = \nabla\times\mathbf{A}_{\chi'}(\mathbf{r},t). \tag{7}$$

If no sources are present, the radiation gauge, labeled in the following by subscript R and defined by the relations

$$\nabla\cdot\mathbf{A}_R = 0, \quad \Phi_R = 0, \tag{8}$$

is convenient. If the wavelength of the considered radiation is sufficiently long, the spatial variation of the radiation field across the system can be neglected, i.e., $\mathbf{A}_R(\mathbf{r},t) \approx \mathbf{A}_R(t)$. In the following, the vector function $\mathbf{A}(t)$ specifies the vector potential in radiation gauge, $\mathbf{A}_R(t)$. Thus, the relation $\mathbf{F}(t) = -d\mathbf{A}(t)/dt$ holds in any gauge.

There exist two further gauges, the length and velocity gauges (labeled in the following by subscripts L and V), which are extensively used in the context of the dipole approximation. However, all three gauges can be considered as particular cases of a generalized gauge defined by an arbitrary set of parameters, $X = \{x_1, x_2\}$. This gauge, which will be referred to as the X gauge, is obtained via the transformation

$$T_{R\to X}(\mathbf{r},t) = x_1\mathbf{A}(t)\cdot\mathbf{r} + x_2\beta(t), \quad \beta(t) \equiv \frac{1}{2}\int^t \mathbf{A}^2(t')dt', \tag{9}$$

so that $X = \{0,0\}$, $X = \{0,1\}$, and $X = \{1,0\}$ correspond to the radiation, velocity, and length gauges, respectively.

The vector and scalar potentials in the X gauge are given as

$$\mathbf{A}_X(t) = (1-x_1)\mathbf{A}(t), \quad \Phi_X = -x_1\mathbf{F}(t)\cdot\mathbf{r} + x_2\mathbf{A}^2(t)/2. \tag{10}$$

This leads to

$$\hat{H}_X = \mathbf{p}_c^2/2 + U(\mathbf{r}) + (1-x_1)\mathbf{A}(t)\cdot\mathbf{p}_c + x_1\mathbf{F}(t)\cdot\mathbf{r} + [(1-x_1)^2 - x_2]\mathbf{A}^2(t)/2 \tag{11}$$

as a definition of the total Hamiltonian in the X gauge.

If the electrostatic potential $U(\mathbf{r})$ is absent, the solution of Eq. (1) in the X gauge is given by the generalized (nonrelativistic) *Volkov wave function*

$$\Psi_{X,\mathbf{k}}(\mathbf{r},t) = e^{-i\Theta_X(t)}e^{i\mathbf{k}\cdot\mathbf{r}} \tag{12}$$

with real phase function

$$\Theta_X(t) = E_k t + \mathbf{k}\cdot\boldsymbol{\alpha}(t) - x_1\mathbf{A}(t)\cdot\mathbf{r} - (x_2-1)\beta(t), \tag{13}$$

where the vector parameter $\mathbf{k}$ specifies the so-called drift momentum, $\boldsymbol{\alpha}(t)=\int^t \mathbf{A}(t')dt'$, $E_k=k^2/2$, and δ-function normalization is used.

III. FORMAL S-MATRIX FORMULATION OF THE SFA

The following formulation of the S-matrix theory describing atomic and molecular systems in intense laser fields considers the case of a one-electron system for the sake of simplicity. The generalization to an arbitrary number of electrons is, however, straightforward. As a starting point the time-dependent Schrödinger equation (TDSE) formulated in the X gauge is considered,

$$\left(i\frac{\partial}{\partial t} - \hat{H}_X(t)\right)|\Psi_X(t)\rangle = 0. \tag{14}$$

The electromagnetic field is absent before and after the pulse, i.e.,

$$\mathbf{A}(t) = \mathbf{A}_0, \quad \mathbf{F}(t) = 0, \quad \hat{H}_X(t) = \hat{H}^0 \quad \text{for } t < t_i \text{ and } t > t_f, \tag{15}$$

where the constant $\mathbf{A}_0$ has no physical meaning (since both the electric and magnetic fields are obtained as derivatives of the vector potential) and will be set to zero for the sake of simplicity. (Note that using $\mathbf{A}_0 \neq \mathbf{0}$ requires a considerable modification of the following formulations.) The operator $\hat{H}^0$ is the field-free Hamiltonian with eigenvalues E_α and eigenvectors $|\psi_\alpha\rangle$,

$$\hat{H}^0 = \mathbf{p}_c^2/2 + U, \quad \hat{H}^0|\psi_\alpha\rangle = E_\alpha|\psi_\alpha\rangle. \tag{16}$$

(The index α denotes discrete as well as continuum states and is thus itself either discrete or continuous.)

To describe the action of the pulse on the system, complete and orthonormal initial- and final-state basis sets are introduced. The initial-state basis set is given by $|\psi_\alpha(t_i)\rangle = e^{-iE_\alpha t_i}|\psi_\alpha\rangle$ where the phase factor is introduced for convenience. The final-state basis set is given by plane waves with momentum $\mathbf{k}$, again for convenience multiplied by a phase factor, and depends both on the adopted gauge and on $\mathbf{k}$,

$$|\Psi_{X,\mathbf{k}}(t_f)\rangle = e^{-i\Theta_X(t_f)}|\mathbf{k}\rangle. \tag{17}$$

The phase [see Eqs. (13) and (15)]

$$\Theta_X(t_f) = E_k t_f + \mathbf{k}\cdot\boldsymbol{\alpha}(t_f) - (x_2-1)\beta(t_f) \tag{18}$$

is $\mathbf{r}$ independent but depends on the used gauge, $\mathbf{k}$, and the pulse. Note that the phase factors introduced for reasons of convenience add only constant phases in the transition amplitudes and do not alter transition probabilities.

The probability amplitude of a transition from an initial state $|\psi_\alpha(t_i)\rangle$ to a final state $|\Psi_{X,\mathbf{k}}(t_f)\rangle$ is given by

$$S_{\mathbf{k}\alpha} = i\langle\Psi_{X,\mathbf{k}}(t_f)|\hat{G}_X(t_f,t_i)|\psi_\alpha(t_i)\rangle, \tag{19}$$

where the propagator $\hat{G}_X(t,t')$ is associated with $\hat{H}_X(t)$ by the inhomogeneous equation

$$\left(i\frac{\partial}{\partial t} - \hat{H}_X(t)\right)\hat{G}_X(t,t') = \delta(t-t'). \tag{20}$$

To obtain a systematic expansion of the transition amplitudes of interest it is convenient to express the total propagator $\hat{G}_X$ of the system in terms of a partial propagator, defined by a *partitioning* of the total Hamiltonian. The choice of the partitioning is made in such a way that the partial propagator can be expressed analytically, i.e., the Schrödinger equation with the corresponding partial Hamiltonian is solvable.

A first class of Hamiltonians that leads to analytical solutions is the one describing a free electron in the field. As was discussed in Sec. II, such Hamiltonians are gauge dependent and their solutions are given by Volkov states. The partitioning of $\hat{H}_X$ using the free-electron Hamiltonian in the X gauge, $\hat{H}_X^f$, is given by

$$\hat{H}_X = \hat{H}_X^f + U. \tag{21}$$

The corresponding propagator can be written analytically using the solutions $|\Psi_{X,\mathbf{k}}(t)\rangle$,

$$\hat{G}_X^f(t,t') = -i\theta(t-t')\sum_{\mathbf{k}}|\Psi_{X,\mathbf{k}}(t)\rangle\langle\Psi_{X,\mathbf{k}}(t')|, \tag{22}$$

where $\theta(x)$ is the step function. From Eq. (22) follows

$$-i\langle\Psi_{X,\mathbf{k}}(t_f)|\hat{G}_X^f(t_f,t) = \langle\Psi_{X,\mathbf{k}}(t)| \quad \text{for } t < t_f. \tag{23}$$

Another Hamiltonian that can be used for the partitioning is the field-free Hamiltonian $\hat{H}^0$. It is, however, only a special case of the class of Hamiltonians which will be referred to as generalized field-free Hamiltonians and will be considered in the following section.

A. Generalized field-free Hamiltonian

The generalized field-free Hamiltonian

$$\hat{H}_\gamma^0 = e^{i\gamma(\mathbf{r},t)}\hat{H}^0 e^{-i\gamma(\mathbf{r},t)} - \frac{\partial\gamma(\mathbf{r},t)}{\partial t}$$

$$= \hat{H}^0 + \frac{i}{2}\Delta\gamma - (\boldsymbol{\nabla}\gamma)\cdot\mathbf{p}_c + \frac{1}{2}(\nabla\gamma)^2 - \frac{\partial\gamma}{\partial t} \tag{24}$$

is defined with the aid of an arbitrary function $\gamma(\mathbf{r},t)$ in such a way that it reduces for $\gamma=0$ to the field-free Hamiltonian $\hat{H}^0$. Solutions of the TDSE with $\hat{H}_\gamma^0$ can be expanded in terms of the solutions of the TDSE with $\hat{H}^0$ as

$$|\Psi_{\gamma,\alpha}(t)\rangle = e^{i\gamma(\mathbf{r},t)}e^{-iE_\alpha t}|\psi_\alpha\rangle. \tag{25}$$

In general, the function $\gamma(\mathbf{r},t)$ can be chosen independent of the gauge that is used to formulate the TDSE. Consider a

particular choice of $\gamma(\mathbf{r},t)$ parametrized by a set of parameters $\lambda=\{\lambda_1,\lambda_2\}$,

$$\gamma_\lambda(\mathbf{r},t)=\lambda_1\mathbf{A}(t)\cdot\mathbf{r}+\lambda_2\beta(t). \tag{26}$$

The corresponding generalized field-free Hamiltonian (the subscript λ is adopted instead of γ_λ for the sake of notational simplicity) is then given by

$$\hat{H}_\lambda^0=\hat{H}^0-\lambda_1\mathbf{A}(t)\cdot\mathbf{p}_c+\lambda_1\mathbf{F}(t)\cdot\mathbf{r}+(\lambda_1^2-\lambda_2)\mathbf{A}^2(t)/2. \tag{27}$$

Note that for all choices of λ the Hamiltonian $\hat{H}_\lambda^0$ gives an equivalent description of the evolution before and after the pulse, since for those times both $A(t)$ and $F(t)$ are equal to zero. Different choices of λ yield, however, different partial propagators $\hat{H}_\lambda^0$ during the pulse that can be written analytically as

$$\hat{G}_\lambda^0(t,t')=-i\theta(t-t')\sum_\alpha|\Psi_{\lambda,\alpha}(t)\rangle\langle\Psi_{\lambda,\alpha}(t')|. \tag{28}$$

In order to express the total propagator $\hat{G}_X$ in terms of $\hat{G}_\lambda^0$, the total Hamiltonian is partitioned in two parts,

$$\hat{H}_X=\hat{H}_\lambda^0+V_{X,\lambda}^0, \tag{29}$$

where the interaction operator $V_{X,\lambda}^0$ is given by

$$V_{X,\lambda}^0(t)=(1-x_1+\lambda_1)\mathbf{A}(t)\cdot\mathbf{p}_c+(x_1-\lambda_1)\mathbf{F}(t)\cdot\mathbf{r}+[(1-x_1)^2-x_2-\lambda_1^2+\lambda_2]\mathbf{A}^2(t)/2. \tag{30}$$

It is worth recalling that the two sets of parameters $X=\{x_1,x_2\}$ and $\lambda=\{\lambda_1,\lambda_2\}$ are independent of each other. Therefore, the same interaction operator can be obtained for different X gauges, if the λ parameters are appropriately chosen. It can be shown, for example, that

$$V_{R,\{-1,0\}}^0=V_{V,\{-1,1\}}^0=V_{L,\{0,0\}}^0=\mathbf{F}(t)\cdot\mathbf{r}, \tag{31}$$

$$V_{R,\{0,-1\}}^0=V_{V,\{0,0\}}^0=V_{L,\{1,1\}}^0=\mathbf{A}(t)\cdot\mathbf{p}_c, \tag{32}$$

$$V_{R,\{0,0\}}^0=V_{V,\{0,1\}}^0=V_{L,\{1,2\}}^0=\mathbf{A}(t)\cdot\mathbf{p}_c+\mathbf{A}^2(t)/2. \tag{33}$$

Since $|\Psi_{\lambda,\alpha}(t_i)\rangle=|\psi_\alpha(t_i)\rangle$, Eq. (28) yields

$$i\hat{G}_\lambda^0(t,t_i)|\psi_\alpha(t_i)\rangle=|\Psi_{\lambda,\alpha}(t)\rangle\quad\text{for } t>t_i. \tag{34}$$

B. Matrix elements

It will now be shown that most of the matrix elements of interest depend at most on the two parameters $v=\{v_1,v_2\}$ with $v_1=1+\lambda_1-x_1$ and $v_2=\lambda_2-x_2$. Indeed, one finds for different matrix elements the relations

$$\langle\Psi_{X,\mathbf{k}}(t)|\Psi_{\lambda,\alpha}(t)\rangle=\langle\mathbf{k}|e^{i\Omega_{v\mathbf{k}\alpha}(t)}|\psi_\alpha\rangle, \tag{35}$$

$$\langle\Psi_{X,\mathbf{k}}(t)|U|\Psi_{\lambda,\alpha}(t)\rangle=\langle\mathbf{k}|e^{i\Omega_{v\mathbf{k}\alpha}(t)}U|\psi_\alpha\rangle, \tag{36}$$

$$\langle\Psi_{\lambda,\alpha'}(t)|V_{X,\lambda}^0(t)|\Psi_{\lambda,\alpha}(t)\rangle=e^{i(E_{\alpha'}-E_\alpha)t}\langle\psi_{\alpha'}|\bar{V}_v^0(t)|\psi_\alpha\rangle, \tag{37}$$

and

$$\langle\Psi_{X,\mathbf{k}}(t)|V_{X,\lambda}^0(t)|\Psi_{\lambda,\alpha}(t)\rangle=\langle\mathbf{k}|e^{i\Omega_{v\mathbf{k}\alpha}(t)}\bar{V}_v^0(t)|\psi_\alpha\rangle, \tag{38}$$

where

$$\bar{V}_v^0(t)=V_{X,\lambda}^0(t)+\lambda_1(1-x_1+\lambda_1)\mathbf{A}^2(t)=v_1\mathbf{A}(t)\cdot\mathbf{p}_c+(1-v_1)\mathbf{F}(t)\cdot\mathbf{r}+(v_1^2+v_2)\mathbf{A}^2(t)/2 \tag{39}$$

and

$$\Omega_{v\mathbf{k}\alpha}(t)=\Theta_X(t)+\gamma_\lambda(\mathbf{r},t)-E_\alpha t=(E_k-E_\alpha)t+\mathbf{k}\cdot\boldsymbol{\alpha}(t)+(v_1-1)\times\mathbf{A}(t)\cdot\mathbf{r}+(v_2+1)\beta(t). \tag{40}$$

Finally, the matrix element

$$\langle\Psi_{X,\mathbf{k}'}(t)|U|\Psi_{X,\mathbf{k}}(t)\rangle=e^{i(E_{k'}-E_k)t+i(\mathbf{k}'-\mathbf{k})\cdot\boldsymbol{\alpha}(t)}\langle\mathbf{k}'|U|\mathbf{k}\rangle \tag{41}$$

is independent of both gauge and partitioning.

As a consequence of these properties of the matrix elements the transition amplitude depends only on v, as is shown below.

C. *S*-matrix series

The operator $\hat{G}_X(t,t')$ can be expanded either in terms of the operator $\hat{G}_\lambda^0(t,t')$,

$$\hat{G}_X(t,t')=\hat{G}_\lambda^0(t,t')+\int dt_1\hat{G}_X(t,t_1)V_{X,\lambda}^0(t_1)\hat{G}_\lambda^0(t_1,t'), \tag{42}$$

or in terms of the operator $\hat{G}_X^f(t,t')$,

$$\hat{G}_X(t,t')=\hat{G}_X^f(t,t')+\int dt_1\hat{G}_X^f(t,t_1)U\hat{G}_X(t_1,t'). \tag{43}$$

Substitution of Eq. (43) in (42) yields

$$\hat{G}_X(t,t')=\hat{G}_\lambda^0(t,t')+\int dt_1\hat{G}_X^f(t,t_1)V_{X,\lambda}^0(t_1)\hat{G}_\lambda^0(t_1,t')+\int\int dt_2dt_1\hat{G}_X^f(t,t_2)U\hat{G}_X(t_2,t_1)V_{X,\lambda}^0(t_1)\hat{G}_\lambda^0(t_1,t'). \tag{44}$$

A further substitution of either (42) or (43) in Eq. (44) results in a series expansion of $\hat{G}_X(t,t')$. Inserting this expansion in Eq. (19) generates the S-matrix series for the transition amplitude between the initial state and the final state to any desired order,

$$S_{\mathbf{k}\alpha}=\sum_{n=0}^{\infty}S_{\mathbf{k}\alpha}^{(n)}, \tag{45}$$

with

$$S^{(0)}_{\mathbf{k}\alpha} = i\langle \Psi_{X,\mathbf{k}}(t_f)|\hat{G}^0_\lambda(t_f,t_i)|\psi_\alpha(t_i)\rangle, \tag{46}$$

$$S^{(1)}_{\mathbf{k}\alpha} = i\int dt_1 \langle \Psi_{X,\mathbf{k}}(t_f)|\hat{G}^f_X(t_f,t_1)V^0_{X,\lambda}(t_1)\hat{G}^0_\lambda(t_1,t_i)|\psi_\alpha(t_i)\rangle. \tag{47}$$

Depending on whether (42) or (43) is substituted in (44) one obtains either

$$S^{(2)}_{\mathbf{k}\alpha} = i\int\int dt_1 dt_2 \langle \Psi_{X,\mathbf{k}}(t_f)|\hat{G}^f_X(t_f,t_2)U \times \hat{G}^0_\lambda(t_2,t_1)V^0_{X,\lambda}(t_1)\hat{G}^0_\lambda(t_1,t_i)|\psi_\alpha(t_i)\rangle \tag{48}$$

or

$$S^{(2)}_{\mathbf{k}\alpha} = i\int\int dt_1 dt_2 \langle \Psi_{X,\mathbf{k}}(t_f)|\hat{G}^f_X(t_f,t_2)U \times \hat{G}^f_X(t_2,t_1)V^0_{X,\lambda}(t_1)\hat{G}^0_\lambda(t_1,t_i)|\psi_\alpha(t_i)\rangle \tag{49}$$

and so on, where the integration is performed in the range t_i to t_f.

From Eqs. (34) and (35) it follows that

$$S^{(0)}_{\mathbf{k}\alpha} = \langle \Psi_{X,\mathbf{k}}(t_f)|\Psi_{\lambda,\alpha}(t_f)\rangle = e^{i\Omega_{v\mathbf{k}\alpha}(t_f)}\tilde{\psi}_\alpha(\mathbf{k}) \tag{50}$$

where $\tilde{\psi}_\alpha(\mathbf{k}) = \langle \mathbf{k}|\psi_\alpha\rangle$ is the Fourier transform of ψ_α. From Eqs. (34) and (23) it follows on the other hand that

$$S^{(1)}_{\mathbf{k}\alpha} = i\int_{t_i}^{t_f} dt \langle \Psi_{X,\mathbf{k}}(t)|V^0_{X,\lambda}(t)|\Psi_{\lambda,\alpha}(t)\rangle, \tag{51}$$

which—using the identity (38)—can be reduced to

$$S^{(1)}_{\mathbf{k}\alpha} = i\int_{t_i}^{t_f} dt \langle \mathbf{k}|e^{i\Omega_{v\mathbf{k}\alpha}(t)}\bar{V}^0_v(t)|\psi_\alpha\rangle. \tag{52}$$

In an analogous way, Eq. (48) can be transformed using (28) as

$$\begin{aligned} S^{(2)}_{\mathbf{k}\alpha} &= i\int_{t_i}^{t_f} dt_2 \int_{t_i}^{t_f} dt_1 \langle \Psi_{X,\mathbf{k}}(t_2)|U\hat{G}^0_\lambda(t_2,t_1)V^0_{X,\lambda}(t_1)|\psi_{\lambda,\alpha}(t_1)\rangle \\ &= \int_{t_i}^{t_f} dt_2 \sum_{\alpha'} \langle \Psi_{X,\mathbf{k}}(t_2)|U|\Psi_{\lambda,\alpha'}(t_2)\rangle \int_{t_i}^{t_2} dt_1 \langle \Psi_{\lambda,\alpha'}(t_1)|V^0_{X,\lambda}(t_1)|\Psi_{\lambda,\alpha}(t_1)\rangle \\ &= \int_{t_i}^{t_f} dt_2 \sum_{\alpha'} \langle \mathbf{k}|e^{i\Omega_{v\mathbf{k}\alpha'}(t_2)}U|\psi_{\alpha'}\rangle \int_{t_i}^{t_2} dt_1 e^{i(E_{\alpha'}-E_\alpha)t_1}\langle \psi_{\alpha'}|\bar{V}^0_v(t_1)|\psi_\alpha\rangle, \end{aligned} \tag{53}$$

or Eq. (49) can be transformed using (22) as

$$\begin{aligned} S^{(2)}_{\mathbf{k}\alpha} &= i\int_{t_i}^{t_f} dt_2 \int_{t_i}^{t_f} dt_1 \langle \Psi_{X,\mathbf{k}}(t_2)|U\hat{G}^f_X(t_2,t_1)V^0_{X,\lambda}(t_1)|\psi_{\lambda,\alpha}(t_1)\rangle \\ &= \int_{t_i}^{t_f} dt_2 \sum_{\mathbf{k}'} \langle \Psi_{X,\mathbf{k}}(t_2)|U|\Psi_{X,\mathbf{k}'}(t_2)\rangle \int_{t_i}^{t_2} dt_1 \langle \Psi_{X,\mathbf{k}'}(t_1)|V^0_{X,\lambda}(t_1)|\Psi_{\lambda,\alpha}(t_1)\rangle \\ &= \int_{t_i}^{t_f} dt_2 \sum_{\mathbf{k}'} e^{i(E_k-E_{k'})t_2+i(\mathbf{k}-\mathbf{k}')\cdot\boldsymbol{\alpha}(t_2)}\langle \mathbf{k}|U|\mathbf{k}'\rangle \int_{t_i}^{t_2} dt_1 \langle \mathbf{k}'|e^{i\Omega_{v\mathbf{k}'\alpha}(t_1)}\bar{V}^0_v(t_1)|\psi_\alpha\rangle. \end{aligned} \tag{54}$$

Continuing in an analogous manner, it can be shown that $S^{(n)}_{\mathbf{k}\alpha}$ for any order n depends only on v. Therefore, $S_{\mathbf{k}\alpha}$ itself depends only on v. In the next section some particular cases will be considered explicitly.

D. Particular cases

As a first example, consider the case studied in [14]. It is obtained using $v=\{0,0\}$ where one has

$$\bar{V}^0_v(t) = \mathbf{F}(t)\cdot\mathbf{r}, \tag{55}$$

$$\Omega_{v\mathbf{k}\alpha}(t) = (E_k - E_\alpha)t + \mathbf{k}\cdot\boldsymbol{\alpha}(t) - \mathbf{A}(t)\cdot\mathbf{r} + \beta(t). \tag{56}$$

This formulation is achieved using the following partitionings for different gauges:

$$\lambda = \{-1,0\} \quad \text{in } R \text{ gauge}, \quad \lambda = \{-1,1\} \quad \text{in } V \text{ gauge}, \tag{57}$$

$$\lambda = \{0,0\} \quad \text{in } L \text{ gauge}.$$

Since in the L gauge the relation $\hat{H}^0_\lambda = \hat{H}^0$ holds, the gauge-invariant formulation with $v=\{0,0\}$ reproduces the traditional SFA in the L gauge.

However, in an analogous way, the traditional V-gauge SFA is obtained with $v=\{1,-1\}$ (cf. [20]), where

$$\bar{V}^0_v(t) = \mathbf{A}(t)\cdot\mathbf{p}_c, \tag{58}$$

$$\Omega_{v\mathbf{k}\alpha}(t) = (E_k - E_\alpha)t + \mathbf{k}\cdot\boldsymbol{\alpha}(t). \tag{59}$$

It can be achieved using the following partitionings for different gauges:

$$\lambda = \{0,-1\} \quad \text{in } R \text{ gauge}, \quad \lambda = \{0,0\} \quad \text{in } V \text{ gauge}, \quad \lambda = \{1,-1\} \quad \text{in } L \text{ gauge}. \tag{60}$$

In a similar way, the traditional R-gauge SFA is obtained with $v=\{1,0\}$, where

$$\bar{V}^0_v(t) = \mathbf{A}(t)\cdot\mathbf{p}_c + \mathbf{A}^2(t)/2, \tag{61}$$

$$\Omega_{v\mathbf{k}\alpha}(t) = (E_k - E_\alpha)t + \mathbf{k}\cdot\boldsymbol{\alpha}(t) + \beta(t). \tag{62}$$

It can be achieved using the following partitionings for different gauges:

$$\lambda = \{0,0\} \quad \text{in } R \text{ gauge}, \quad \lambda = \{0,1\} \quad \text{in } V \text{ gauge}, \quad \lambda = \{1,0\} \quad \text{in } L \text{ gauge}. \tag{63}$$

Clearly, every S-matrix expansion (SFA formulation) in one of the "conventional" (length, velocity, or radiation) gauges can be obtained by adopting any of the possible gauges, if the partitioning of the Hamiltonian is chosen accordingly.

IV. CONCLUSION

In this work it is shown how the (infinite-order) S-matrix expansion describing atomic or molecular systems in intense laser fields depends on the choice of both the gauge *and* the partitioning of the Hamiltonian. For this purpose a generalized gauge as well as a generalized partitioning scheme is introduced. They are defined by four independent parameters (x_1 and x_2 for the gauge as well as λ_1 and λ_2 for the partitioning). However, the S-matrix expansion is then shown to depend on only two parameters, $v_1 = 1+\lambda_1 - x_1$ and $v_2 = \lambda_2 - x_2$. Clearly, every possible combination of the parameters defining the gauge and the partitioning that conserves the values $\lambda_1 - x_1$ and $\lambda_2 - x_2$ leads to an identical S-matrix expansion (up to an overall phase factor that cancels when calculating physical observables).

The present analysis thus shows that one has to be very careful not to consider solely the gauge, since a suitable choice of the partitioning may lead to an S-matrix expansion that is identical to the one obtained in some other gauge, as was also demonstrated in [14] where the velocity-gauge expansion appeared to agree with what is usually known as the length-gauge S-matrix expansion. In fact, it is much more appropriate to discuss different expansions (defined by a specific combination of gauge and partitioning) than different gauges. This allows introduction of some terminology like "equivalent expansions in different gauges" instead of the perhaps confusing "gauge-invariant (first-order) KFR approximation in the velocity gauge" [14].

Furthermore, the present result demonstrates that there remains an in principle infinite set of S-matrix expansions (characterized by different values of v_1, v_2) that are shown to provide the same transition probabilities only in the limit of an infinite series expansion, if the latter converges. Truncated series like, e.g., the zeroth-, first-, or second-order expansions will, however, in general disagree. Thus the question of a "proper" choice of the expansion in the case of truncation remains and can be clarified only by a comparison to either experiment or gauge-independent theory (like full solutions of the time-dependent Schrödinger equation).

In the authors' opinion, there is no *a priori* reason to believe that one expansion is necessarily advantageous over others for all atomic or molecular systems as well as all possible laser parameters. Similarly to various physical applications, where the choice of a suitable expansion is optimized in order to achieve faster convergence, there may exist an optimal parameter set v for a given problem. This set may, however, not necessarily agree with one of the traditional SFA expansions. A deeper understanding certainly requires further studies.

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