Strong-field approximation for intense-laser atom processes: the choice of gauge

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The strong-field approximation can be and has been applied in both length gauge and velocity gauge with quantitatively conflicting answers. For ionization of negative ions with a ground state of odd parity, the predictions of the two gauges differ qualitatively: in the envelope of the angular–resolved energy spectrum, dips in one gauge correspond to humps in the other. We show that the length-gauge SFA matches the exact numerical solution of the time-dependent Schrödinger equation.

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Quantum mechanics is gauge invariant: it is easily proven that a given physical quantity can be evaluated in any gauge with the same result [1]. In nonrelativistic quantum mechanics, when the dipole approximation is adopted, the interaction of an atom with a time-dependent field such as a laser field is usually described in either one of two gauges: the length gauge (L gauge) or the velocity gauge (V gauge). In numerical solutions of the time-dependent Schrödinger equation (TDSE), gauge invariance has been confirmed many times. In analytical work, however, some approximations almost always have to be adopted. There is no formal reason of why after such approximations the resulting theory should still be gauge invariant. Indeed, the lack of gauge invariance after what seems like very well justified approximations has driven many a researcher to despair [2].

In this paper, we will address one of the most glaring manifestations of this “gauge problem”: the lack of gauge invariance of the strong-field approximation (SFA) in intense-laser–atom physics [3]. The SFA underlies almost any analytical approach to total ionization rates, above-threshold ionization, high-order harmonic generation, and nonsequential double ionization, both of atoms and of molecules. Briefly, it assumes that the initial bound state of the atom or molecule is unaffected by the laser field while the final state, which is in the continuum, does not feel the presence of the binding potential. The lack of gauge invariance of the SFA has been noted many times; see, e.g., Ref. [4]. Comparisons that have been carried out indeed have exhibited significant disagreements between the results obtained from L gauge and V gauge [5]. Different authors have preferred different gauges. The question of which gauge is superior for which problem has often been raised, but never led to any consensus about its answer. Below, we will give an answer for the case of a short-range binding potential, where the SFA is expected to be most accurate [6], by comparing the SFA in L gauge and V gauge with the numerical solution of the TDSE.

For a fixed nucleus and in the single-active-electron approximation, where the effects of all electrons but one are absorbed into an effective binding potential, the complete Hamiltonian in the presence of an external electromag-
Gauge invariance then implies that \( M_p \) be gauge invariant, and indeed this can easily be verified explicitly. The SFA is obtained if we insert the Dyson equation \([3]\) into the ionization amplitude \([6]\). The first term, which comes from \( U_0(t, t') \), cancels since the initial and the final state are orthogonal, and we are left with \([7]\)

\[
M_p = -i \lim_{t \to \infty} \int_{-\infty}^t d\tau \langle \psi_p(t) | U_x(t, \tau) H_{1z}(\tau) | \psi_0(\tau) \rangle, \tag{7}
\]

which is still exact.

In the argument that follows we restrict ourselves for the sake of transparency and simplicity to “direct” electrons, i.e., those that after the initial ionization never feel the binding potential. In order to obtain the transition amplitude for the direct electrons, we replace in Eq. \([7]\) the exact state at time \( \tau \), which is \( \langle \psi_p(t) | U_x(t, \tau) \rangle \), by the Volkov state \( \langle \psi_p^{(\text{V})}(\tau) | \rangle \) [given below in Eq. \([8]\)] where the interaction with the binding potential is neglected. This yields the well-known SFA amplitude \([8]\)

\[
M_p = -i \int_{-\infty}^{\infty} d\tau \langle \psi_p^{(\text{V})}(\tau) | H_{1z}(\tau) | \psi_0(\tau) \rangle. \tag{8}
\]

Here, for times \( t < \tau \) the state of the electron is governed by the Hamiltonian \( H_0 \), while for \( t > \tau \) its time evolution follows the Hamiltonian \( H_{1z} \).

The matrix element \([8]\) conveys the following physical picture: for times \( t < \tau \) the electron is sufficiently deeply bound that to a good approximation its interaction with the laser field can be ignored. At time \( \tau \), it is ionized, and the laser intensity is high enough to move the electron so rapidly out of the range of the binding potential that now the latter can be neglected.

However, this physical picture is in agreement with the formal description only within \( L \) gauge. In \( L \) gauge, the interaction with the laser field is accomplished by the scalar potential \( e\Phi(t) = H_{1z}(t) \). There is no vector potential, so that the operator of the velocity is \( V = p/m \). Hence, \( p^2/(2m) \) is the operator of the kinetic energy, and \( H_0 \) describes an atom that does not interact with the field, even if a field is present. In \( V \) gauge, the operator of the velocity is \( V = [p - eA(t)]/m \) where \( p \) is the operator of the canonical momentum. This is a conserved quantity under the dipole approximation, but not a physical quantity \([1]\), since \( p = m\vec{v} + eA(t) \), and \( \vec{v} \) is a physical quantity while \( A(t) \) is not. In consequence, in the presence of a laser field, the operator \( H_0 \) is not the field-free Hamiltonian, and its eigenstate \( \langle \psi_0(t_0) | \rangle \) does incorporate some interaction with the field \([8]\). Hence, in \( V \) gauge, the physical picture formulated above is not supported by the matrix element \([8]\).

It is instructive to evaluate the matrix element \([8]\) by the method of steepest descent, which is known to work very well for sufficiently high intensity. We first recall the explicit form of the Volkov wave function

\[
\langle r | \psi_p^{(\text{V})}(t) \rangle = \frac{e^{-i S_p(t)}}{(2\pi)^{3/2}} \left\{ e^{i p \cdot r}, \quad (x = V) \right. \nonumber \\
\left. e^{i(p - eA(t)) \cdot r}, \quad (x = L) \right. \tag{9}
\]

with the action

\[
S_p(t) = \frac{1}{2m} \int_{t}^{t'} d\tau (p - eA(\tau))^2. \tag{10}
\]

which has the same form in either gauge \([9]\).

Via an integration by parts, the transition amplitude \([8]\) can be recast in the form \([7]\)

\[
M_p = -i \int_{-\infty}^{\infty} d\tau \langle \psi_p^{(\text{V})}(\tau) | H_{1z}(\tau) | \psi_0(\tau) \rangle, \tag{11}
\]

which depends on the gauge only via the Volkov wave function \([9]\). Collecting the exponential time dependence of the integrand in Eq. \([11]\) we find that the stationary points with respect to \( \tau \) are determined as the solutions of the saddle-point equation

\[
[p - eA(\tau)]^2 = -2m I_p. \tag{12}
\]

The transition amplitude then can be represented as the superposition of the contributions of all those solutions \( \tau_s \) of Eq. \([12]\) for which \( \text{Im} \tau_s > 0 \), with the result

\[
M_p = \sum_s V_{pxs} \sqrt{\frac{2\pi}{E(t_s) \cdot |p - eA(t_s)|}} e^{i \left[ S_p(t_s) + I_p \tau_s \right]} e^{i \left[ S_p(t_s) + I_p \tau_s \right]}, \tag{13}
\]

Only the form factor

\[
V_{pxs} = \left\{ \begin{array}{ll} \langle p | V(r) | 0 \rangle, & (x = V), \\ \langle p - eA(t_s) | V(r) | 0 \rangle, & (x = L) \end{array} \right. \tag{14}
\]

depends on the gauge. In \( V \) gauge, it is evaluated at the momentum \( p \) at the detector, which is the same for all saddle-point solutions. In \( L \) gauge, it is evaluated at the instantaneous velocity at the ionization time \( t_s \), whose component parallel to the laser field according to Eq. \([12]\) is purely imaginary and can have either sign. For a monochromatic linearly polarized laser field, there are two solutions \( t_s \) per cycle of the saddle-point equation \([12]\) with \( \text{Im} t_s > 0 \), one on either side of the pertinent extremum of the vector potential.

To find out the signs of \( p - eA(t_s) \) that correspond to the solutions with \( \text{Im} t_s > 0 \), let us consider the vector potential \( A(t) = eA \cos \omega t \). We let \( t = t_R + it_t \), where \( t_R \) and \( t_t \) denote the real and the imaginary part of \( t \), respectively. The real and imaginary parts of the saddle-point equation \([12]\) are

\[
p_{\parallel} - eA \cos \omega t_R \cosh \omega t_t = 0, \tag{15a}
\]

\[
eA \sin \omega t_R \sinh \omega t_t = \sqrt{2 \text{Im} I_p + p_{\perp}^2}, \tag{15b}
\]

where \( p_{\parallel} \) and \( p_{\perp} \) are the components of \( p \) parallel and perpendicular to the laser field and the square root may have either sign. From Eq. \([15a]\), the two solutions per cycle are such that \( \cos \omega t_R \) has the same sign. Then, from Eq. \([15b]\) and the fact that \( t_t > 0 \) for the physical saddle-point solutions, we have that \( \sin \omega t_R \) has the opposite sign for the two solutions. Hence, the two instantaneous
The electric-field vector is $E$ with the sine-square envelope $s_0$. In contrast, in V gauge dips and humps are for amplitude $s_0$ in L gauge and V gauge, respectively, taking positions of the interference dips for a pure state. Consequently, for an odd-parity initial state, constructive interference in L gauge implies destructive interference in V gauge, and vice versa. In contrast, for an even-parity ground state, both gauges predict interference maxima and minima at the same positions. As soon as $p_\perp \neq 0$, there is no complete destructive or constructive interference anymore.

In Figs. 1, 2, and 3 we compare the results of the SFA in L gauge and V gauge with a numerical solution of the TDSE. All calculations have been carried out for a 4-cycle linearly polarized laser pulse having the intensity $2.4 \times 10^{14}$ W/cm$^2$ (field strength $E_0 = 0.0834$ a.u.), and wavelength 800 nm (photon energy $\omega = 0.056$ a.u.). The electric-field vector is $E(t) = (\pm i \sqrt{2mI_p} + p_\perp \hat{e}_\perp)\cos(\omega t + \phi)\hat{e}$, with the sine-square envelope $E(t) = E_0 \sin^2(2\pi t/T_p)$ for $0 \leq t \leq T_p = n_p T$, $T = 2\pi/\omega$, and $E(t) = 0$ outside this interval. The carrier-envelope phase is $\phi = 0$. Figure 1 exhibits the results of a numerical computation (not using the saddle-point approximation) of the SFA amplitude in L gauge and V gauge, respectively, taking for $|\psi(t)|^2$ the bound state of a zero-range potential $Z/r$. They illustrate the above statements. In other words, in L gauge, everything else being equal, the positions of the interference dips for a $p$ ground state coincide with those of the interference humps for an $s$ ground state. In contrast, in V gauge dips and humps occur at the same positions regardless of the parity of the ground state. Figure 2 presents the corresponding TDSE spectrum calculated by methods introduced elsewhere [12]. In order to mimic a short-range potential in the TDSE calculations, the Coulomb potential $-Ze/r$ has been cut at $r_c = 2$ a.u. The nuclear charge $Z_{eff}$ was adjusted in such a way as to keep the ionization potential $I_p$ at 0.5 a.u. for both the $1s$ and the $2p$ state. It has been shown recently [14] that the agreement between SFA and TDSE low-energy electron spectra improves with decreasing potential range $r_c$. A direct comparison of the TDSE and SFA (L gauge) results is presented in Fig. 3. The agreement with respect to the energetic positions of the various peaks is excellent. Residual discrepancies are observed in the shape of the spectrum for low energies, especially for the $p$ ground state, and are likely due to the different large-distance behavior of the wave functions (zero range for the SFA vs. cut Coulomb for the TDSE).

The exact solution for ionization of negatively charged ions that is available in the context of effective-range theory exhibits the interference dips in complementary positions for $s$ and $p$ ground states [12], in agreement with the L-gauge SFA and the exact TDSE solution. The L-gauge SFA also appears to be supported by the experimental data: the above-threshold-detachment energy spectrum for the negative $F^-$ ion [15], which has a $p$ ground state, displays a pronounced change of its slope at the energy where the L-gauge SFA predicts an interference dip [10].

For elliptical polarization, for ellipticities higher than a certain critical value the saddle-point equation [12] only has one solution per cycle rather than two, so that the interference ceases to exist [17]. This is so, in particular, for circular polarization. Recently, the latter case was considered in detail [18]. Even in the absence of interference, the form factor [14] is still different in L gauge and in V gauge. For an $s$ ground state $|0\rangle$, the form factor $\langle p|V|0\rangle$ has a maximum for $p = 0$ and decreases.

**FIG. 1:** (color online) SFA electron energy spectrum for emission in the (positive) direction of the laser field (four-cycle $\sin^2$-pulse with $\phi = 0$, $\omega = 0.056$ a.u., $E_0 = 0.0834$ a.u.) in L gauge, starting from an initial 1s (solid) or 2p (dashed) state. The corresponding V gauge result is shown in the inset.

**FIG. 2:** (color online) Same as Fig. 1 but computed from the numerical solution of the TDSE.
with increasing $|p|$, while for a $p$ state, it has a zero at $p = 0$ and extrema away from $p = 0$. In Ref. [18], for ionization of $F^-$ by a circularly polarized laser field, the energy spectrum was calculated in either gauge. The $V$-gauge spectrum peaks at a higher energy than the $L$-gauge spectrum, which conforms with the considerations given above. Moreover, Wigner’s threshold law is only reproduced in $L$ gauge [18].

Before concluding, we recall that in a numerical solution of the TDSE the choice of gauge is “merely” a question of convenience. Generally, convergence is faster in $V$ gauge where fewer angular momenta contribute, much faster indeed for high intensity and low frequency. In contrast, in approximations such as the SFA, the choice of gauge is a contributing factor for the quality of the approximation. In fact, making formally the same approximation in two gauges may correspond to different approximations physically. A general argument in favor of the $L$ gauge for use in the SFA has been put forward in Ref. [4]: the $L$-gauge interaction Hamiltonian puts the emphasis on large distances from the atom, where the Volkov wave function is a good approximation to the final state. In a similar vein, we add that it appears to make more sense to evaluate the form factor at the instantaneous velocity at the time of ionization (as in $L$ gauge) rather than at the drift velocity (as in $V$ gauge), which for low frequencies the electron does not assume before it is far away from the ion.

On the basis of a comparison with the solution of the time-dependent Schrödinger equation, we conclude that the strong-field approximation applied to above-threshold detachment of negative ions affords a better description in length gauge than in velocity gauge. In view of the fundamental significance of the SFA for strong-field physics, it is of great importance to find out which gauge is better suited for above-threshold ionization of atoms and molecules as well as nonsequential double ionization. In all of these cases, the two gauges are known to yield different answers as well.

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