Mixed quantum and classical processes in strong fields

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When a quantum transition is followed by a classical process, as in strong-field rescattering events, it is essential to recognize the different time scales appropriate to each process. This establishes the sequential nature of the phenomena, and the necessity of satisfying quantum and classical conservation conditions independently. The term "rescattering" has been used in both quantum and classical senses, and the distinction is important. An example is examined where proper care has not been taken in distinguishing between quantum and classical conditions. The result is a major violation of angular momentum conservation that has apparently been unrecognized. An explanation for how this could happen is provided. Application of the principles presented here is important for current proposals to alter the parameters of laser focusing in efforts to overcome relativistic effects that degrade higher harmonic generation. In the (extreme ultraviolet) XUV and x-ray environments, the quantum-classical time distinction is blurred, leading to a loss of the simplifying features of classical behavior.

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I. INTRODUCTION

The time scales involved in strong-field-induced atomic and molecular problems can be categorized as quantum vs classical or virtual vs real. The distinction is at the heart of the useful technique in strong-field physics, wherein a quantum process is envisaged as being followed by a classical interaction between, for example, a photoelectron and the field that produced it. Despite the widespread use of this dual procedure, the rules that govern its application are not always observed. The purpose of the present work is to clarify the rules, and to illustrate the negative consequences of ignoring them.

The first step is to distinguish the time scales that characterize quantum and classical processes. The quantum time interval has the characteristic that it is not directly visible to laboratory instruments. Quantum processes occur on a time scale whose magnitude is governed by the Heisenberg uncertainty principle. In contrast, processes occurring on a classical time scale can be measured explicitly. For example, strong fields employed in atomic and molecular physics are usually produced by lasers, and the quantum result of an interaction between the laser and the atom may be one or more photoelectrons. In turn, a photoelectron can, itself, be accelerated by the laser field. The period of a laser field can be measured classically, and thus this second interaction can be regarded as classical. The two time scales differ, typically, by a factor of 100, and sometimes by much larger factors.

A phenomenon called "rescattering" can occur in both quantum and classical senses. It is important to be clear about the distinction: rescattering in the quantum sense is virtual, and in the classical sense it is real.

When both quantum and classical behaviors are included in a problem, the need for caution is illustrated by examining a practical example from the strong-field literature. The example is based on the very strong-field ionization of an atom by an infrared-frequency, circularly polarized field. The example is instructive expressly because all photons in a circularly polarized field have their quantum angular momenta of $\hbar$ aligned and hence additive. The amount of angular momentum transferred in the quantum process can be very large. This very large exchange of angular momentum in the circular polarization environment has long been known and utilized in physical problems. Nevertheless, the presently employed mode of application, of following the classical path of a photoelectron after ionization by a strong field, is such as to omit this angular momentum transferred from the field in the quantum process. This failure to assure that quantum conservation conditions are met prior to the onset of the classical process leads to a major violation of angular momentum conservation.

Finally, we note that the convenience of using classical path analysis will not be available in the XUV and soft x-ray domains.

II. TIME SCALES

It is instructive to start with an examination of time scales. What is said in the following is well known and elementary. This makes it surprising that the distinction between quantum and classical time scales is so commonly ignored.

The time scale within which an atomic ionization event occurs is usually measured by the atomic unit of time, which is

$$\tau_{\text{atomic}} = \frac{\hbar^3}{me^4} = 2.4 \times 10^{-17}\ \text{s}. \tag{1}$$

That this is the appropriate time scale follows simply from the Heisenberg uncertainty principle in the form

$$\Delta E \Delta t = (E_B)(\tau_{\text{atomic}}) = O(1), \tag{2}$$

where E_B is the binding energy of the electron in the atom. Here and subsequently, atomic units are used, which means that

$$\tau_{\text{atomic}} = 1 \text{ a.u.} \tag{3}$$

The inference that Δt in Eq. (2) is the time scale for ionization comes from the assignment of $\Delta E = E_B$ as the energy uncertainty. When the energy is uncertain to the extent of the binding energy, then it is uncertain about whether the atom has had time to ionize.

There is a second time scale in atomic ionization following from the period of the laser field of frequency ω, or

$$\tau_{\text{field}} = \frac{2\pi}{\omega}. \tag{4}$$

For the commonplace example of a Ti:sapphire laser, $\tau_{\text{field}} = 2.7 \times 10^{-15}$ s. A comparison of this result to the atomic unit of time in Eq. (1) gives

$$\tau_{\text{field}} \approx 10^2 \tau_{\text{atomic}}. \tag{5}$$

In an intense laser field, a free electron must possess at least the ponderomotive energy

$$U_p = \frac{I}{(2\omega)^2}, \tag{6}$$

where I is the field intensity. When $U_p \gg E_B$, it is the transfer of the ponderomotive energy that determines whether ionization can occur. The distinction between quantum and classical time scales will then be much greater than the factor 10^2 shown in Eq. (5).

III. RESCATTERING

A. Quantum rescattering

Consider a state Ψ that describes an unbound electron exposed to a combination of a Coulomb field and a laser field. In general, there is no known exact solution of the Schrödinger equation for this physical problem. Two immediately apparent methods of approximate solution exist, based on the fact that there are two independent influences on the electron. If we regard the laser field as a perturbation on the state of the electron dominated by the effects of the Coulomb field, we have what is conventionally regarded as a perturbative approach. If, on the other hand, the laser field is so strong that its effect on the photoelectron can be regarded as the dominant effect, then it is possible to carry out a perturbative expansion of Ψ on the grounds of the Coulomb interaction as the perturbing influence. A general treatment of this two-interaction scenario is presented in (Ref. [1], Appendix A).

A specific example of such a two-interaction physical problem is given by the ionization of an atom as described by the "time-reversed" exact transition amplitude

$$M_{fi} = -i \int_{-\infty}^{\infty} dt (\Psi_f(t), H_I(t) \Phi_i(t)), \tag{7}$$

where atomic units are used, the fully interacting state Ψ satisfies the Schrödinger equation

$$i\partial_t \Psi(t) = [H_0 + H_I(t)]\Psi(t), \tag{8}$$

and the noninteracting state Φ satisfies the Schrödinger equation with the Hamiltonian H_0 that excludes the influence of the laser,

$$i\partial_t \Phi(t) = H_0 \Phi(t). \tag{9}$$

If the second of the two approaches described above should be followed, and Ψ is expanded in powers of the Coulomb interaction, then a Feynman diagram representation of the process would show repeated vertices representing individual recurrences of a Coulomb interaction. This can be termed rescattering, but it is important to note that all time orderings of the Coulomb interactions must be considered, and the time intervals between successive interactions are not real times that can be measured in the laboratory.

Whether Ψ is expanded in powers of the Coulomb interaction as just described, or in terms of powers of the laser interaction, these prescriptions are simply mathematical devices that are aids to an adequate statement of $\Psi(t)$. The successive interactions with the Coulomb field in one case or with the laser field in the other are often regarded as actual physical transitions between physically realized states. This is misleading. There are no real intermediate states unless some measurement exists that can identify such an intermediate state, as in the case of a resonance. Lacking such a measurable event, the expansions are mathematical schema to represent the state $\Psi(t)$ for each time t that is integrated over in Eq. (7). Other times that might be introduced into the perturbation expansion are not physical times. They have a quantum uncertainty as described in Eq. (2).

B. Classical rescattering

Quantum rescattering is a virtual process, and the details will depend on the approximation scheme adopted to calculate the fully interacting wave function. In contrast, a classical rescattering event has a direct physical meaning. An electron ionized by the oscillatory field will initially move away from the remnant ion. When the field reverses direction, so will the electromagnetic force on the photoelectron, driving the electron back toward the atom. If the approach is close enough that the electron experiences a scattering event, this event will be separated in time from the initial ionization by a macroscopic time of the order of Eq. (4).

The notion of classical rescattering has proven to be qualitatively quite fruitful for such phenomena as higher harmonic generation [2]. In application, the so-called simpleman's point of view [3] is generally adopted. It is supposed that the initial quantum stage of the process proceeds by ionization achieved by tunneling of an initially bound electron through a potential barrier [4–7]. The resulting spectrum (for linear polarization) will peak at zero momentum, and at a displacement from the center of the atom at a distance of the order of the atomic radius. Unlike quantum mechanics, classical mechanics requires the prescription of initial conditions. It is possible to select sophisticated initial conditions to broaden the scope of the possible solutions, but the usual application of the simpleman's method employs the elementary classical initial conditions [3]

$$\mathbf{r}_0 = 0, \quad \mathbf{p}_0 = 0. \tag{10}$$

In quantum mechanics, it is not possible to set simultaneous explicit values for the canonically conjugate variables **r** and **p**. Nevertheless, only slight error is introduced by the conditions (10), as long as linear polarization is being treated. For the opposite case of circular polarization, matters are quite different, as will be shown in what follows.

C. Limitations

An essential facet of the quantum-followed-by-classical sequence described above is that the interface between the two events should not involve unacceptable approximations. Specifically, the classical initial conditions expressed in Eq. (10) must be a reasonable estimate for the outcome of a quantum transition, within limits set by the Heisenberg uncertainty principle. An instance of a neglect of this constraint is exemplified in ionization by a circularly polarized laser field.

Consider an experiment in 1989 by Corkum *et al.* [8], in which the ionization of ground-state xenon by a CO_2 laser was studied. Despite the early date, this was a strongly intense-field experiment. For the reported field intensity of 2×10^{14} W/cm^2 at a wavelength of 10.6 μm, the ponderomotive potential of close to 2 keV was equivalent to more than 100 times the binding energy of 12.1 eV. Just to supply the binding energy requires the absorption of about 100 photons; thus, more than 10^4 photons are necessary to provide the ponderomotive energy to the photoelectron.

The significance of these numbers for circularly polarized light is that each photon carries one quantum unit of angular momentum, and these angular momenta are aligned. That is, they are directly additive. To transfer the energy of 10^4 photons to a photoelectron requires the concurrent transfer of 10^4 quantum units of angular momentum. Conservation of energy and of angular momentum must be satisfied on the time scale of the quantum process; it cannot be transferred at the later occurrence of classical motion of the photoelectron in the laser field. In this example, the quantum time scale is determined by U_p, as pointed out below Eq. (6); thus, it is of the order of 10^{-2} of an atomic time unit. The classical time scale, from Eq. (4), is more than ten times greater for a CO_2 laser than for a Ti:sapphire laser. The relative classical and quantum time scales thus differ by a factor of more than

$$\tau_{\text{field}} \approx 10^5 \tau_{\text{atomic}} \tag{11}$$

in this case, rather than the more modest 10^2 estimated in Eq. (5).

The angular momentum of the order of 10^4 quantum units is with respect to an axis parallel to the propagation direction of the laser beam, passing through the nucleus of the atom undergoing ionization. The generally accepted initial conditions for classical motion, shown in Eq. (10), cannot possibly be appropriate for the circular polarization problem just examined. Classical motion subsequent to the quantum ionization can still be specified, but the initial conditions must be such as to represent a classical orbit encircling the original atomic location, and possessing the classical energy and angular momentum. (An initial classical *angular coordinate*, however, cannot be specified.)

The example just cited represents relatively extreme conditions in order to emphasize the distinction between quantum and classical time scales. It stresses the need to set the classical initial conditions by the conservation rules demanded by the preceding quantum process. The same rules apply even in more mundane circumstances. The distinction between ionization by circularly as opposed to linearly polarized light is always characterized by the demands of angular momentum conservation in the case of circular polarization.

A commonly adopted point of view is that ionization by circularly polarized light is nothing more than ionization with linear polarization, but where the direction of the electric field vector is in constant rotation. This cannot be correct. The spectrum of photoelectrons created by linearly polarized light peaks at low energies (though not at zero energy as is assumed in a simple tunneling model). It is an observed experimental fact [8] that the photoelectron spectrum in strong-field ionization by circularly polarized light peaks near the ponderomotive energy U_p. This major distinction in photoelectron kinetic energy is related to the fact that linear polarization makes no demands on a minimum angular momentum, whereas circular polarization imposes the quantum condition that n units of angular momentum must be carried off by a photoelectron ionized by n photons. In direct consequence of these quantum conservation conditions, the initial classical conditions specified in Eq. (10) are *never* appropriate for circular polarization.

Classically, the motion of an electron in a circularly polarized field is in a circular path as described in the frame of reference in which the center of the circular motion is at rest. The classical angular momentum of the electron in this frame of reference is just what would be associated with the quantum angular momentum of the number of photons provided to acquire the ponderomotive and kinetic energies. The correspondence between the quantum conditions that must exist at the end of the quantum process in order to provide the classical initial conditions for subsequent motion of the photoelectron on a classical time scale thus corresponds to motion in a circular orbit in a plane perpendicular to the direction of laser propagation. This is *not* compatible with Eq. (10).

D. Magnetic field effects

An increase in field intensity, in itself, can lead to the onset of "v/c effects" that nullify the rationale for using the dipole approximation [9,10] in strong-field interactions. This is a far more limiting criterion than simply the comparison of wave length to atomic size that is often cited as the justification for the dipole approximation. This loss of the dipole approximation is often ascribed to relativistic effects, which are "$(v/c)^2$ effects" that require a higher intensity to be significant than do magnetic field effects. For a laser with a wave length of 800 nm, the onset of magnetic field effects can occur at $\sim10^{15}$ W/cm^2, whereas true relativistic effects do not set in before $\sim10^{17}$ W/cm^2 [9,10].

The Lorentz force **F** on a particle of charge q due to electric and magnetic fields **E** and **B**,

$$\mathbf{F} = q\left(\mathbf{E} + \frac{\mathbf{v}}{c} \times \mathbf{B}\right), \tag{12}$$

serves to identify the reason for the v/c onset of magnetic field effects. With a linearly polarized laser field, it has long been recognized that the propagation direction of the magnetic component of the Lorentz force will deflect an electron from an oscillation solely in the electric field direction. This can prevent a return of a photoelectron to the atom from which it came, and thus prevent rescattering. As intensity increases into the magnetic-effects and relativistic domains, and the peak in photoelectron spectra departs substantially from the $\mathbf{p}_0=0$ condition in Eq. (10), the conclusion is also applicable in circular polarization that the Lorentz force causes the classical trajectory of a photoelectron away from a path that can return to the parent atom. However, the nature of the modified path differs greatly for the two polarizations.

Ionization with circularly polarized light, as seen above, leads to a classical path that circulates around the parent atom because of the requirement that angular momentum must be conserved. There will be no rescattering because of the need to retain cylindrical symmetry. That is, even without any magnetic effects, rescattering will not occur.

When the magnetic force of Eq. (12) becomes significant, the effect is to shift the classical circular orbit forward so that the parent atom no longer lies in the plane defined by the orbit (See Fig. 3 in Ref. [11]).

IV. REMARKS

The ability of a strong, circularly polarized laser to transfer large amounts of angular momentum to a target is well known in other applications. For example, Rydberg wave packets within an atom have been made to circulate in circular orbits using this effect [12]. Another quite striking application of the same effect has been shown in a macroscopically observable way by using the angular momentum of a circularly polarized laser to couple to certain types of biological cells, causing them to rotate [13].

Apparently, the reason that the inapplicability of the conditions (10) has not attracted comment previously is that Eq. (10) predicts a continuous movement away from the atom of the electron, with no return to the atom when the radiation is circularly polarized. There is no classical rescattering. When angular momentum is included properly, the electron simply moves out to a circular orbit of large radius. This scenario also predicts no rescattering. The no-rescattering result is in accord with experiments, and thus, there has been no perceived need to examine the circular polarization case in more detail.

The need to distinguish quantum and classical time scales is not merely an academic matter in view of recent proposals (see, for example, Ref. [14]) to enhance the efficacy of higher harmonic generation despite the defocusing effects of relativity. Relativistic photoelectron spectra produced by linearly polarized lasers peak at energies that differ significantly from the constraints of Eq. (10).

A final remark concerns the XUV and x-ray regions of the electromagnetic spectrum, which are of increasing interest as the availability of strong, coherent sources of this part of the spectrum become available in the laboratory. For such short wavelengths, the inequalities between atomic and electromagnetic time scales, as expressed in Eq. (5) or (11), no longer apply. In that case, one cannot disengage a quantum part of the problem from a classical part, and the convenient classical rescattering concept no longer exists.

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