

## Limits on Tunneling Theories of Strong-Field Ionization

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(Received 6 March 2008; published 22 July 2008)

It is shown that tunneling theories of ionization by lasers are subject to upper and lower bounds on the Keldysh parameter  $\gamma$ . The *tunneling limit*,  $\gamma \rightarrow 0$ , applies to ionization by quasistatic electric fields, but not by laser fields. For lasers, the  $\gamma \rightarrow 0$  limit requires a relativistic treatment. Bounds on the applicability of tunneling theories depend on parameters other than  $\gamma$ , confirming the rule that strong-field phenomena require more than one dimensionless parameter for scaling.

DOI: 10.1103/PhysRevLett.101.043002

PACS numbers: 32.80.Rm, 33.20.Xx, 33.60.+q, 33.80.-b

Tunneling theories of strong-field ionization were introduced [1–4] early in the development of strong-field physics, preceded only by the use of Volkov solutions [5,6] as a basic method for the nonperturbative treatment of strong-field phenomena. The terminology of tunneling is part of the lexicon of strong-field physics: *tunneling domain*, *multiphoton domain*, *over-the-barrier ionization*, *tunneling limit*, *tunneling time*, etc. Despite its long history, there has been inadequate exploration of the limitations of tunneling methods. That task is undertaken here, with the outcome that previously unexamined constraints exist.

This investigation begins with an inquiry into the origins of a flawed prediction of a tunneling approach. Tunneling theories [1–4] of strong-field ionization seem to be extendable to the limit [7]  $\gamma \rightarrow 0$ , where  $\gamma$  is the Keldysh parameter

$$\gamma \equiv \sqrt{E_B/2U_p} = (\omega/\sqrt{I})\sqrt{2E_B}; \quad U_p = I/(2\omega)^2. \quad (1)$$

Here,  $E_B$  is the field-free ionization energy of a bound electron, and  $U_p$  is the ponderomotive energy of the interaction of a free electron with the field. The second form in Eq. (1) comes from the expression of  $U_p$  in atomic units, as shown to the right. From Eq. (1), it would appear that the inappropriate limit  $\gamma \rightarrow 0$  can be reached in two ways:

$$\left. \begin{array}{l} \omega \rightarrow 0 \\ I \rightarrow \infty \end{array} \right\} \Rightarrow \gamma \rightarrow 0. \quad (2)$$

The limit  $\gamma \rightarrow 0$  is called the *tunneling limit*. It has become widely (but incorrectly) accepted that one can assess the validity of nontunneling theories by examining their behavior in the tunneling limit. There is now voluminous literature where this assumption is made.

It is shown here that none of the limits expressed in Eq. (2) is accessible to a tunneling theory as applied to laser fields, and that both upper and nonzero lower limits on  $\gamma$  must be observed when describing laser-caused ionization. The practice of appraising a theory by comparing its behavior as  $\gamma \rightarrow 0$  with the behavior of a tunneling

theory in that limit is an invalid procedure for ionization by laser fields. Furthermore, the concepts of *multiphoton domain* and *tunneling domain*, usually viewed as determined by the value of  $\gamma$ , are not well defined. Both extreme limits  $\gamma \ll 1$  and  $\gamma \gg 1$  are outside the domain of applicability of tunneling concepts to laser-induced processes.

The first reason for these restrictions is that tunneling theories are dipole-approximation theories, and these are gauge equivalent to transverse fields (such as that produced by a laser) only when the dipole approximation is valid. In turn, the dipole approximation is valid only when magnetic field effects can be neglected. It is shown below that neither the  $\omega \rightarrow 0$  nor the  $I \rightarrow \infty$  limits in Eq. (2) can be reached without entering a domain where magnetic field effects due to laser radiation become important. Furthermore, these two limits are different, not equivalent as indicated by Eq. (2). The onset of magnetic effects sets a lower limit on  $\gamma$  where tunneling theories can be applied, and an upper limit on  $\gamma$  comes from the fact that a tunneling point of view is applicable only when the number of photons that enter into a process is very large. The lower limit is examined first.

Consider the limit  $I \rightarrow \infty$ . As the intensity increases indefinitely, an electron in a laser field would exhibit relativistic behavior, where electric and magnetic fields become equally important. (The reader is reminded that conservation conditions must be applied at the moment of ionization, and cannot be deferred until subsequent acceleration in the laser field. The influence of the magnetic field is thus immediately consequential. See Ref. [8] for the demonstration that quantum conservation conditions must be applied before subsequent classical motion in the external field.) That cannot occur in a tunneling theory, where only the electric field appears no matter how high the intensity gets. The consequences can be shown in the context of the behavior of a free electron in a laser field. In terms of the intensity parameter

$$z_f \equiv 2U_p/mc^2, \quad (3)$$

an electron will behave relativistically when the pondero-

motive energy due to its interaction with the laser field becomes so large that  $z_f = 1$ . [The electron mass  $m$  appears in (3) to preserve the recognizability of  $mc^2$ , otherwise lost in atomic units.] From the form of the ponderomotive energy in the last element of Eq. (1), it follows that

$$z_f = 1 \Rightarrow I = 2c^2\omega^2. \quad (4)$$

On a log-log plot of intensity as a function of frequency, this is a straight line of slope 2. Figure 1 shows such a plot, where intensity is in atomic units (left vertical axis) and  $\text{W}/\text{cm}^2$  (right vertical axis), and the frequency  $\omega$  on the lower horizontal axis is paired with the oppositely directed wavelength on the upper horizontal axis. The straight line given by Eq. (4) distinguishes the relativistic domain in the upper left of the figure from the nonrelativistic domain in the lower right. If the vertical and horizontal arrows, starting from an arbitrary position in the nonrelativistic region, are followed in the directions  $\omega \rightarrow 0$  (with  $I$  constant) or  $I \rightarrow \infty$  (with  $\omega$  constant), both limits lead to the relativistic domain, and they end in qualitatively distinct regions of relativistic behavior. The limit  $\gamma \rightarrow 0$  does not lie within the scope of a tunneling theory.

One can go further. As the intensity increases from a nonrelativistic domain towards relativistic behavior, the  $v/c$  effects of the magnetic field will manifest themselves significantly before the  $(v/c)^2$  effects of true relativistic

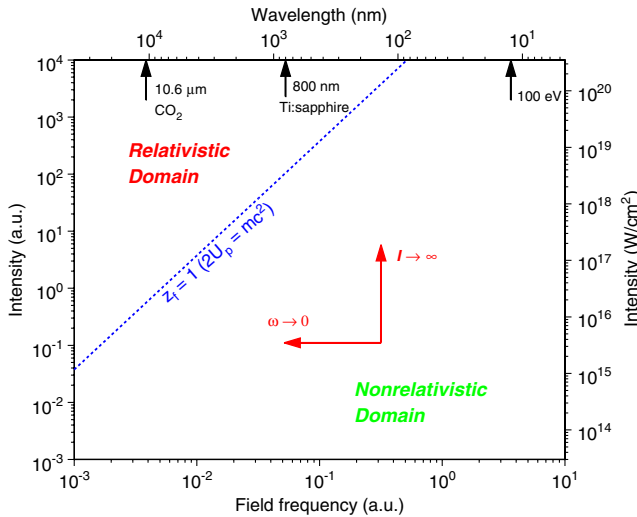


FIG. 1 (color online). The slanted line indicates the intensity (as a function of frequency) at which an electron in a plane-wave field acquires so much ponderomotive energy that the  $z_f$  intensity parameter of Eq. (3) is equal to one. This is a boundary between relativistic and nonrelativistic conditions. Starting at an arbitrary point in the nonrelativistic domain, a path showing the limit  $I \rightarrow \infty$  must enter the relativistic domain, as must a path showing the limit  $\omega \rightarrow 0$ . The two limits will be very different. Since both limits correspond to the Keldysh  $\gamma$  approaching zero, this means that the limit of very small  $\gamma$  in a tunneling theory is not applicable to plane-wave (i.e., laser) fields.

properties [9,10]. In the simplest frame of reference, the effect of a strong plane-wave field on a free electron is to produce a figure-8 pattern [11,12], where the long axis of the figure-8 is along the electric field  $\mathbf{E}$ , and the short axis is along the direction of propagation  $\mathbf{k}$  of the field. The vectors  $\mathbf{E}$ ,  $\mathbf{B}$ , and  $\mathbf{k}$  are mutually perpendicular. A direct measure of the effect of the magnetic field is given by the amplitude  $\beta_0$  parallel to the propagation direction:

$$\beta_0 = \frac{cz}{4(1+z_f)} \approx \frac{cz}{4}. \quad (5)$$

In Eq. (5),  $c$  is the velocity of light in atomic units ( $c = 1/\alpha \approx 137$ ) and  $z$  is the intensity parameter [13]

$$z \equiv U_p/\omega. \quad (6)$$

The last expression in Eq. (5) follows from the fact that  $z_f$  is normally small when  $\beta_0 \approx 1$ . To avoid the onset of magnetic field effects, and thus to maintain the validity of the dipole approximation, it must be true that

$$\beta_0 \ll 1, \quad \text{or} \quad z \ll 4/c. \quad (7)$$

With the expression for the ponderomotive energy from Eq. (1) and the definition of  $z$  in Eq. (6), the magnetic-field analog of Eq. (4) is

$$\beta_0 = 1 \Rightarrow I = 8c\omega^3. \quad (8)$$

On a log-log plot of intensity as a function of frequency, this is a straight line of slope 3; see Fig. 2.

It is possible to state precisely why the  $\gamma \rightarrow 0$  limit obtained from a tunneling theory is so different from the  $I \rightarrow \infty$  or  $\omega \rightarrow 0$  limits for a laser field. The defining

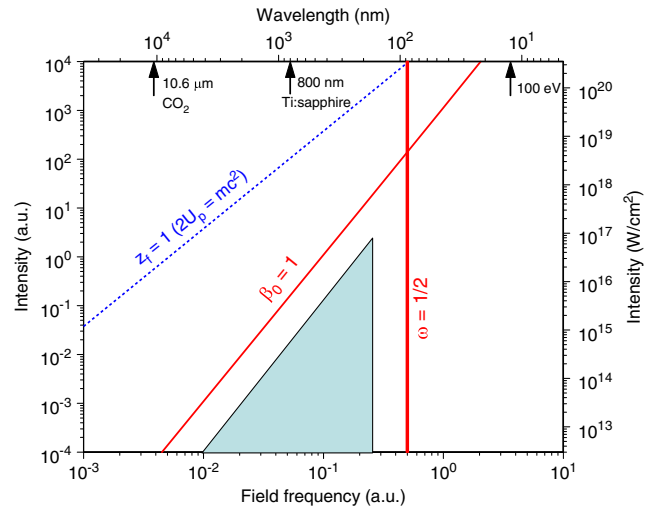


FIG. 2 (color online). The line  $\beta_0 = 1$  marks the low-frequency limit of dipole-approximation behavior, and  $\omega = E_B$  marks the upper limit on the frequency for a tunneling description of ionization. (The value of  $E_B$  is set to 0.5 as an illustration.) The shaded triangular area in the figure marks the region within which a tunneling theory can be applied to atomic ionization caused by a laser field.

properties for a plane-wave field like the laser field are that it is described by the fundamental relativistic invariants

$$\mathbf{E} \cdot \mathbf{B} = 0, \quad \mathbf{E}^2 - \mathbf{B}^2 = 0, \quad (9)$$

with the direct implications

$$|\mathbf{E}| = |\mathbf{B}|, \quad \mathbf{E} \perp \mathbf{B}. \quad (10)$$

By contrast, tunneling methods (and all length-gauge methods) describe the electromagnetic field solely in terms of the electric field by using the scalar potential

$$\phi = -\mathbf{r} \cdot \mathbf{E}(t). \quad (11)$$

If the electric field  $\mathbf{E}$  is independent of time, the potential (11) is exactly descriptive of a static electric field. When the field has the time dependence shown in Eq. (11), it is then a quasistatic field that has the relativistic invariants

$$\mathbf{E} \cdot \mathbf{B} \approx 0, \quad \mathbf{E}^2 - \mathbf{B}^2 \approx \mathbf{E}^2. \quad (12)$$

The ‘‘approximately equal’’ symbol is used in Eq. (12) because Maxwell’s equations demand that, if some time dependence exists for the electric field, the magnetic field cannot be exactly zero. It can, however, be very small:

$$|\mathbf{B}| \approx 0. \quad (13)$$

Equations (9) and (10) are very different from Eqs. (12) and (13). The quasistatic electric field is gauge equivalent to the plane wave *only* when  $v/c$  is sufficiently small that  $\mathbf{B}$  can be neglected. In other words, there can be gauge equivalence between the length-gauge expression of Eq. (11) and the usual velocity-gauge expression for a plane-wave field *only* when the dipole approximation is valid.

Equation (8) and the requirement that  $\beta_0 < 1$  demands that

$$\omega^3 > I/8c, \quad (14)$$

which constitutes a lower bound on the allowable frequency for the application of the dipole approximation. This is a lower bound on the frequency for which tunneling methods are applicable to laser-caused strong-field phenomena.

When combined with Eq. (1), expressing the Keldysh parameter in intensity and frequency terms, the result is that

$$\gamma > \frac{1}{(c^2 I)^{1/6}} \sqrt{\frac{E_B}{2}} \quad \text{or} \quad \gamma > \frac{1}{2} \sqrt{\frac{E_B}{\omega c}}, \quad (15)$$

as a function of the intensity, or as a function of the frequency. Equation (15) gives a lower limit beyond which a tunneling theory is no longer applicable to the description of laser-caused phenomena. This conclusion excludes the  $\gamma \rightarrow 0$  limit of a tunneling theory from having any significance for laser ionization phenomena.

The limits stated in Eqs. (14) and (15) pertain to *all* dipole-approximation theories, including the nonrelativistic strong-field approximation and numerical solution of the time-dependent Schrödinger equation. Matters are different when one seeks an upper limit on the frequency for which a tunneling theory is applicable. This limit will now be explored.

The graphical depiction of ionization occurring through a tunneling mechanism involves a static Coulomb potential well modified by a quasistatic electric field. One side of the Coulomb well is depressed sufficiently that a bound electron can leave the parent atom by tunneling through the finite potential barrier on the side of the well that is depressed by the electric field. Such a potential-energy diagram is applicable only when the number of photons required to achieve this result is so large that there is no distinction between an ionization caused by  $n$  photons as compared to ionization caused by  $n + 1$  photons. This can be true only when the energy of any single photon is small as compared to the energy required to ionize the electron:

$$\omega \ll E_B. \quad (16)$$

For definiteness,  $E_B$  is taken to be 0.5 in order to illustrate this limit in Fig. 2, which shows both the lower limit on frequency that comes from Eq. (14) and the upper limit from Eq. (16).

The domain of validity of a tunneling approximation is indicated by the shaded triangular area in Fig. 2.

The usual upper limit on  $\omega$  for a dipole-approximation theory is to require that the phase of a plane wave can be regarded as approximately constant over the size of the atom. That is, when  $r \leq 1$  a.u., the phase of the plane wave can be approximated by the purely time-dependent part of the phase:

$$\omega t - \mathbf{k} \cdot \mathbf{r} \approx \omega t. \quad (17)$$

This places the constraint on the propagation vector  $\mathbf{k}$  that

$$|\mathbf{k}| = \frac{\omega}{c} \ll 1, \quad \text{or} \quad \omega \ll c \approx 137. \quad (18)$$

As shown by a diagram like Fig. 2, the limit set by Eq. (18) lies off the figure towards the high-frequency end at the right. It is not at all as limiting as Eq. (16) for tunneling.

In terms of the Keldysh  $\gamma$  parameter, the condition for  $\beta_0 \ll 1$  in Eq. (14) combines with Eq. (1) to give

$$\gamma \gg \frac{1}{(c^2 I)^{1/6}} \sqrt{\frac{E_B}{2}}, \quad (19)$$

whereas Eq. (16) leads to

$$\gamma \ll \sqrt{\frac{2E_B^3}{I}}. \quad (20)$$

Figure 3 shows how the limits on  $\gamma$  appear as a function of the intensity  $I$  for two different choices for  $E_B$ . It is

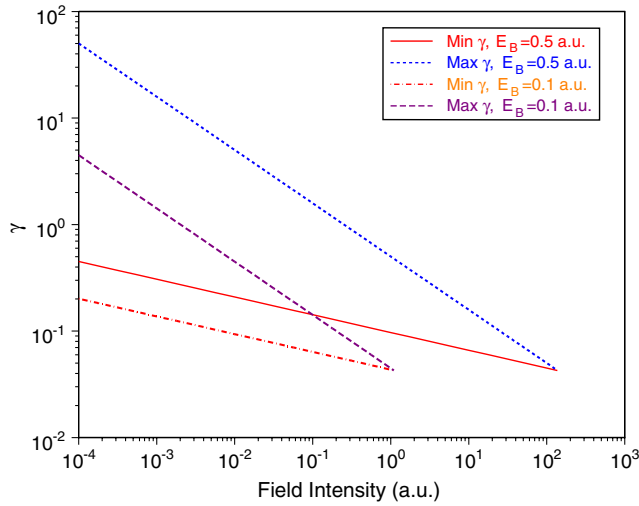


FIG. 3 (color online). The region of applicability exhibited in Fig. 2 is here indicated on a diagram of  $\gamma$  as a function of field intensity. The boundaries of this region depend on both the binding energy  $E_B$  and the intensity parameter  $z$  of Eq. (6), so the domain of applicability of tunneling cannot be described in terms of  $\gamma$  alone.

important that there is no universal rule for  $\gamma$ . This is a direct consequence of the fact that the lower limit of frequency in Fig. 3, as given by Eq. (5), depends upon an intensity parameter  $z$  that is distinct from  $\gamma$ . As discussed in Refs. [9,13], there are three energies that enter into nonrelativistic ionization problems: ponderomotive energy  $U_p$ , binding energy  $E_B$ , and photon energy  $\omega$ . There are thus two independent dimensionless intensity parameters that can be formed from ratios of these energies:

$$z_1 \equiv \frac{2U_p}{E_B} = \frac{1}{\gamma^2} \quad \text{and} \quad z \equiv \frac{U_p}{\omega}. \quad (21)$$

Only one of these ( $\gamma = 1/z_1^{1/2}$ ) seems to occur in tunneling, although the need for two parameters was emphasized in Ref. [13]. It now seen that the second parameter is actually present in a fundamental way since the validity domain for a tunneling theory has a lower bound that depends on the second parameter  $z$ , as shown by Eq. (5). If relativistic conditions exist, there is a fourth energy, the rest energy of the electron  $mc^2$  that introduces a third dimensionless parameter  $z_f$ , defined in Eq. (3).

Figure 3 shows another important matter: there does not appear to be much special significance attached to  $\gamma = 1$ , commonly regarded as the boundary between the tunneling domain and the multiphoton domain. It is usually considered that  $\gamma > 1$  defines the multiphoton domain, whereas Fig. 3 shows that values  $\gamma = O(10)$  can still be regarded as

allowing tunneling concepts to be applied. On the other hand,  $\gamma < 1$  is usually spoken of as tunneling, whereas Fig. 3 shows that  $\gamma$  as small as  $O(10^{-1})$  may be out of the reach of a tunneling theory.

This raises the subject of the interpretation [1] of  $\gamma$  as the ratio of the time required to tunnel through the barrier (the tunneling time) to the period of the applied field. That, however, is a concept that has meaning only within the context of a Coulomb potential well (represented by a scalar potential) being modified by the imposition of a quasistatic electric field, also represented by a scalar potential. Such a physical picture has meaning only within the length gauge where everything is represented by scalar potentials. The laser field is, however, a transverse field, requiring a vector potential for its full description. Even though there is a domain of intensity within which the length and velocity gauges are gauge equivalent, that equivalence extends only to physical measurables. The tunneling time is not a physical measurable, and there is no concept within the velocity gauge that corresponds to the notion of tunneling through a potential barrier. The impression given in a tunneling theory is that there does not exist an obstacle to seeking the limit  $\gamma \rightarrow 0$ . When the laser field is represented by the dipole-approximation vector potential  $\mathbf{A}(t)$ , it is obvious that when conditions no longer justify the use of the dipole approximation, then  $\mathbf{A}(t)$  must be replaced by  $\mathbf{A}(t, \mathbf{r})$ . The existence of a magnetic field is then inevitable.

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