Velocity-gauge theory for the treatment of strong-field photodetachment

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A recent experimental study of the photodetachment of negative fluorine ions by a strong, circularly polarized laser field has been used as a test case for comparison of different gauges in a strong-field theory. Earlier studies concluded that the length gauge was the more suitable theory, but it was necessary to view the experimental peak intensity as a fitting parameter, and this had to be altered to 145% of the measured peak intensity. That amount of variation is not possible. There were two independent experimental measurements of the peak intensity: one direct and the other through a measurement of the momentum distribution of the photodetached electron. These two measurements give the same result for the peak intensity, and so the 45% increase required to have a fit to the length gauge is not an acceptable theoretical explanation for the experimental results. In contrast, the velocity gauge calculation provides a good match to the spectrum peak with no alteration necessary for the measured peak intensity. The theoretical spectrum is somewhat narrower than the measured spectrum, but that is consistent with the fact that the intensity parameters are not quite large enough to ensure detailed accuracy of the strong-field approximation.

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I. INTRODUCTION

It has long been known that photodetachment of a negative ion is well suited to treatment by a Volkov-solution-based theory that neglects the effect of the binding potential on the outgoing electron. That point of view was a premise of the strong-field approximation (SFA) theory of 1980 [1]. It also a matter of long-standing recognition [1–4] that this type of analytical approximation will be gauge dependent. Based on a theory by Yang [5–8], the proposition has been made that the length gauge is the fundamentally correct gauge for the treatment of charged systems in interaction with an electromagnetic field. Several recent papers [9–11] have taken this point of view. To be definite about what is meant by the length gauge and the widely used alternative—the velocity gauge—the respective interaction Hamiltonians for a single electron are given here in atomic units (a.u.):

$$H_I^L = \mathbf{r} \cdot \mathbf{E}, \tag{1}$$

$$H_I^V = \frac{\mathbf{A}}{c} \cdot \hat{\mathbf{p}} + \frac{1}{2}\left(\frac{\mathbf{A}}{c}\right)^2, \tag{2}$$

where $\mathbf{E}$ is the electric field vector, $\mathbf{A}$ is the vector potential of the field, $\hat{\mathbf{p}}$ is the momentum operator, and the superscript on H_I indicates the gauge.

The notion that the length gauge is fundamentally superior to the velocity gauge was posited by Lamb in 1952 [12]. A formal basis for this concept was provided by Yang [5]. The recent revival of interest in this issue [9–11] follows from the possibility of experimentally testing the hypothesis by a comparison of the predictions of the length-gauge version of the velocity-gauge SFA in application to photodetachment of an electron from a negative ion.

It is difficult to accept the notion that the length gauge has universally superior validity. The experiments that form the basis for comparison are done with lasers, and a laser produces a propagating, transverse, plane-wave field. A scalar potential such as that employed in the length gauge is not capable of representing a transverse field. The length gauge is therefore necessarily an approximation of limited applicability that must inevitably prove inadequate [13] as the intensity rises. Nevertheless, it is possible that the length gauge might provide a more accurate result in some particular cases as long as the intensity is not very high. The experimental results being analyzed are nonperturbative, but not of extremely high intensity.

The important question of the formal foundations of the length-gauge-favoring Yang theory will be analyzed elsewhere. The present paper will be confined to the examination of a specific set of experimental data.

The following section will discuss the reasons why photodetachment of negative ions by circularly polarized light represents a particularly uncomplicated and informative situation. In an intense-field environment, one expects the detached electron to acquire many of the properties of a classical electron moving in the laser field [14–16]. In an intense field with circular polarization, the detached electron should move in a circular orbit around the remnant ion or atom with essentially classical parameters [14,17].

Momentum distribution measurements of fundamental significance were made in the experiments of Bergues, Helm, and Kiyan [10] (BHK) on photodetachment of an electron from the negative fluorine ion. These measurements are completely consistent with the concept of the detached electron circulating around the remnant atom in a toroidal probability distribution that possesses many of the properties of a free electron in the laser field. This is a necessity based on quantum conservation conditions that must be satisfied at the moment of photodetachment [16]. One of the reasons this is especially interesting is that it contradicts the premise of the "simpleman's model" [15], which conceives of an electron being detached or ionized with zero initial velocity and then acquiring linear and angular momentum from the laser in a subsequent classical interaction with the laser field. It was shown in Ref. [16] that this is not possible since it violates quantum conservation requirements, and the BHK experiment gives a clear confirmation of this assertion.

Section III exhibits the comparison between theory and experiment. An advantage of using modern data is that the intensity of the laser beam can now be measured far more accurately than was the case only a few years ago. The point of view adopted here is that the published intensity, stated to be accurate to within 15%, is correct. This is confirmed in BHK by their measurement of the momentum distribution of the detached electron. The confirmation of the measured intensity by the momentum distribution means that the intensity is well known and is not available as a fitting parameter. Other comparisons [10,11] use the intensity as a fitting parameter and find the need to hypothesize an intensity that is 145% of the measured value. That precludes the use of the length gauge for this problem. On the other hand, the peak of the spectrum, an especially important quantity with circular polarization, is matched very well by the velocity-gauge theory used here. The width of the peak is narrower in the theory than in the experiment, but not egregiously so. Furthermore, the values of the intensity parameters that obtain in the experiment are such that one should not expect precise agreement of a SFA calculation with the experiment.

Section IV summarizes the results. The fact that two independent experimental measurements coincide in predicting a peak ponderomotive energy U_p of 5.5 eV means that the length gauge fails the test of matching the experiment, since a fit to the length gauge would require a peak U_p=8 eV. It is concluded that those features of the velocity-gauge prediction that coincide with the experimental data are more fundamental than what is attainable with a length-gauge theory. The length gauge is suspect because it does not account for the transverse property of laser fields, it requires an adjustment of the laser intensity that is far outside the experimental bounds of accuracy, and it contradicts the measured momentum distribution.

II. STRONG-FIELD PHENOMENA IN A CIRCULARLY POLARIZED FIELD

Strong fields generate phenomena that have many classical characteristics [14,15]. The particularly useful property imparted to an electron that is photodetached or ionized is that the energy that must be invested in overcoming the binding energy and providing the ponderomotive energy to the unbound electron requires the contributions of many photons, each of which carries one quantum unit of angular momentum. These angular momenta are aligned, and they are manifested in a circular orbit centered on the atom that is followed by the detached electron. This transfer of angular momentum has to occur at the instant of the photodetachment and cannot be accomplished through some later transfer of angular momentum to the electron from the field [16]. The amount of angular momentum acquired by the electron in this process, as well as the kinetic energy imparted to the electron, is consistent with the classical properties of an electron in a circularly polarized field [14,16,17]. The kinetic energy of the electron in its circular orbit around the parent atom is just U_p, the ponderomotive energy of the electron in interaction with the field [18].

One of the most significant results of the BHK paper is their Fig. 1 [10]. The figure shows the momentum components of the detached electron measured parallel and perpendicular to the plane of polarization of the circularly polarized laser. The figure shows what is essentially a slice through the toroid of the momentum distribution of the detached electron, exhibiting a "doughnut" cross section. The momentum corresponding to the most intense part of the distribution is on the azimuthal axis of the toroid. At this location, $p_\parallel = 0.62$ a.u. and $p_\perp = 0$. This gives a kinetic energy of 0.192 a.u.=5.23 eV. This is within 5% of the value of the ponderomotive energy U_p associated with the measured intensity of the laser field of 2.6×10^{13} W/cm^2.

The authors of the BHK paper assert that their intensity measurement is the 2.6×10^{13} W/cm^2 just cited, accurate to within 15%. Since this is consistent with the momentum distribution shown in Fig. 1 of BHK, that intensity measurement will be regarded hereafter as a well-established quantity. In an attempt to achieve a fit with a length-gauge theory, the BHK paper makes conjectures about how their intensity measurement might have been enough in error to match the 1.38×10^{13} W/cm^2 required for consideration of the length gauge, but those conjectures do nothing to explain how the momentum distribution could be that much in error as well.

Two independent intensity parameters are necessary [1,13,19] to characterize a nonrelativistic strong field. A qualitative physical significance can be assigned to the z and z_1 dimensionless parameters to be employed here. (See Sec. 1.3 of Ref. [19].) They are defined as

$$z \equiv U_p/\omega, \tag{3}$$

$$z_1 \equiv 2U_p/E_B, \tag{4}$$

where atomic units are used, ω is the energy of a single photon, and E_B is the no-field binding energy of the electron before detachment. For the BHK experiment on photodetachment of an electron from the negative fluorine ion by an infrared laser with a wavelength of 1510 nm, these parameters have the peak values

$$z = 6.7, \tag{5}$$

$$z_1 = 3.2. \tag{6}$$

Since z can be regarded as a measure of the validity of perturbation theory, with $z=1$ as an absolute upper limit for the convergence of perturbation theory, it is seen that the BHK experiment was done in an indisputably strong-field environment. However, the value of z_1 in Eq. (6) is not especially large, in view of the fact that $z_1 \geqslant O$ (10) can be viewed as necessary for accurate application of the SFA.

Useful guidance about the use of the SFA to describe ionization by a circularly polarized field can be found in an earlier application of the SFA to the description of a strong-field ionization experiment with helium. The experiments were done by Mohideen *et al.* [20], and the SFA description of the experimental spectrum was published in Ref. [21]. At that time, measurement of the focused intensity of the laser was very difficult. The estimate by Mohideen *et al.* was that the peak intensity was 6×10^{15} W/cm^2. This was plainly inaccurate, since the peak of the circular polarization spectrum would then be at only 20% of the maximum U_p. It was

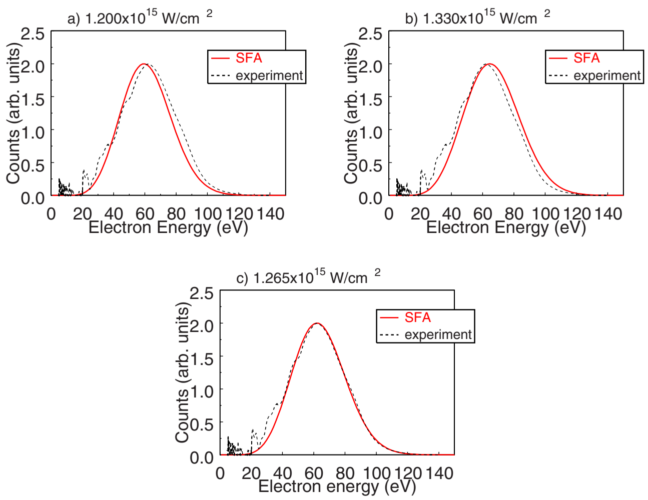

FIG. 1. (Color online) Measured spectra from the ionization of helium by circularly polarized light from a laser of 815 nm wavelength [20] as compared to theoretical predictions of the velocity-gauge SFA [21] for several assumed peak intensities. (a) Intensity of 1.200×10^{15} W/cm^2, corresponding to 95% of the best-fit intensity. (b) Intensity of 1.330×10^{15} W/cm^2, corresponding to 105% of the best-fit intensity. (c) Intensity of 1.265×10^{15} W/cm^2, found to be the best fit to the experiments. Notice how a change of only 5% in intensity produces significant shifts in the peak of the spectrum and how well the best-fit intensity matches the experimental spectrum in peak location and in overall shape.

proposed [21] that the fit of the experiment by the SFA could be used as a far more accurate determination of the actual focal intensity than that measured in the laboratory. The laser field was clearly very intense, and the SFA is applicable since the intensity parameters found for the experiment were

$$z = 51.6, \tag{7}$$

$$z_1 = 6.38. \tag{8}$$

A simple Coulomb correction [22] valid for atomic ionization by a circularly polarized field was employed. Figures 1(a)–1(c) show, respectively, the SFA spectra obtained for 95%, 105%, and 100% of the best-fit intensity of 1.265×10^{15} W/cm^2. (The figure reported in Ref. [21] was 1.275×10^{15} W/cm^2. A more efficient calculational algorithm and a more capable computer make possible the more accurate result now attainable.) The best-fit intensity corresponded to the peak of the circular-polarization spectrum occurring at 80% of the peak U_p. This is far more acceptable than the 20% of peak U_p reported by the experimenters. The reason the number is not 100% of U_p is that, with such strong fields, an important fraction of the total ionization yield comes from regions of the spatiotemporal distribution of intensity in the focal region that are at significantly less than the maximum intensity. The calculation modeled the intensity distribution in a Gaussian focus, with the time distribution also given by a Gaussian distribution with the full width at half maximum (FWHM) stated by the experimenters. In that experiment, the target gas was present as a gas fill in an evacuated chamber, so the calculation was done by integrating over isointensity

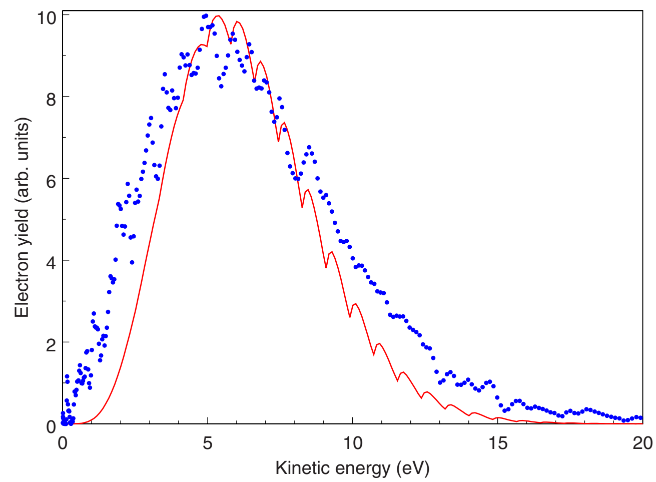

FIG. 2. (Color online) The velocity-gauge SFA produces the spectrum shown when the peak laser intensity measured in Ref. [10] is employed without change. The peak of the spectrum is at about 5.4×10^{13} W/cm^2, very close to the 5.5×10^{13} W/cm^2 reported for the peak ponderomotive energy U_p reported in Ref. [10].

shells in the focus. Depletion effects were also included. The theory underlying how this is done is presented in Sec. 4.3.6 of Ref. [19]. Hartree-Fock analytical wave functions were used in Ref. [21].

The purpose of showing fits at three different intensities in Fig. 1, including values at 5% more and less than the best-fit value, is to show how accurately the best-fit intensity can be determined. A deviation of only 5% from the best-fit intensity produced obviously inappropriate estimates.

Another important purpose in showing Fig. 1 is to demonstrate how near perfect a match to the data can be provided by the velocity-gauge SFA. See Fig. 1(c).

III. COMPARISON BETWEEN THE VELOCITY-GAUGE SFA AND THE BHK EXPERIMENTS

The computational algorithm used to compare the SFA with the data of BHK [10] differs from that used with the data of Mohideen *et al.* because the experiment was done by intersecting the laser focus and an ion beam, rather than using the gas-fill method as was done in the helium experiments [20]. The constant-intensity profiles in the Gaussian focus are assumed to be circular cylinders in the waist region of the Gaussian focus intersected by the ion beam. A comparison of the calculated result with the data of BHK is shown in Fig. 2.

The SFA result in Fig. 2 represents a computation done with the field-free initial state of the negative fluorine ion given by an analytical Hartree-Fock wave function. The standard SFA velocity-gauge S-matrix form

$$(S-1)_{fi} = \frac{1}{(2\pi)^2}\sum_{n=n_0}^{\infty} p\left(\frac{p^2}{2}+E_B\right)^2 |\phi_i(\mathbf{p})|^2[J_n(\alpha_0 p \sin\theta)]^2, \tag{9}$$

$$\frac{p^2}{2} = n\omega - E_B - U_p, \tag{10}$$

$$n_0 \geqslant \frac{1}{\omega}(E_B + U_p), \tag{11}$$

$$\alpha_0 = \left(\frac{2z}{\omega}\right)^{1/2} \tag{12}$$

is used. The momentum-space wave function $\phi_i(\mathbf{p})$ of the initial state is defined with the normalization

$$\phi_i(\mathbf{p}) = \int d^3x \exp(i\mathbf{p}\cdot\mathbf{r})\phi_i(\mathbf{r}); \tag{13}$$

J_n in Eq. (9) is the ordinary Bessel function; α_0 in Eq. (12) is the radius of rotation of a free electron in the presence of a circularly polarized field of frequency ω and intensity parameter z, in the coordinate system in which the electron rotates about a fixed center; and θ is the polar angle of spherical coordinates measured from the propagation direction of the laser field as polar axis. As mentioned, $\phi_i(\mathbf{r})$ is the space part of the initial-state wave function expressed in terms of the Hartree-Fock wave-function parameters that are tabulated in the book of Radzig and Smirnov [23]. The Hartree-Fock wave function for F^- is a linear combination of $2p$ states, and the result employed was found by averaging the $l=1$ spherical harmonic over initial m substates.

An alternative procedure that was also investigated is to replace the Hartree-Fock wave function by the analytical solution found for a zero-range potential, with an initial p-state spherical harmonic. The results thus obtained differ only slightly from what is shown in Fig. 2.

The SFA result in Fig. 2 represents a spectrum obtained by summing all electron emissions over a 4π solid angle. A spectrum representing emission strictly in the plane of polarization was found to differ only slightly.

Depletion effects were included in the computation, but they are of negligible consequence for this experiment.

Comparison between the predictions of the analytical computation and the experimental data is complicated by the somewhat erratic appearance of the data. This is a consequence of the rather marginal value of the z_1 parameter given in Eq. (6). The computations provide some valuable insight in that they show evidence of remnants of the multipeaked behavior to be expected with values of z and z_1 that are nonperturbative but not unambiguously of a strong-field character [24]. Some of the irregularities of the data appear to follow the outlines of the multipeaked features of the calculated spectrum.

[Parenthetically, it is to be noted that the descriptive terms "multiphoton" and "tunneling" are avoided here in view of the inherent contradictions attendant on the use of that terminology, as described in Ref. [13]. The related inadequacy of reliance on a single intensity parameter is avoided by employing the necessary two intensity parameters, here selected to be z and z_1, as given in the pairs of expressions (5) and (6) for the F^- spectrum treated here and the expressions (7) and (8) for the helium ionization spectrum employed for comparative purposes. The Keldysh parameter γ is given by the inverse square root of z_1: $\gamma=1/z_1^{1/2}$.]

Considering that the intensity circumstances of the experiment were somewhat marginal for application of the SFA, the fit between the calculated and measured results exhibited in Fig. 2 can be regarded as quite satisfactory. It was shown in Sec. II that the location of the peak is an essential feature of strong-field ionization or photodetachment spectra with circularly polarized light. The test case examined in Sec. II, unambiguously a strong-field situation, found that the peak value was at 80% of the maximum ponderomotive potential U_p. The reason that the result was not 100% of U_p is that much of the intensity distribution within the time- and space-delimited focal region possessed sufficient intensity to contribute a significant part of the total ionization. In the case being examined here—photodetachment of F^-—the most intense part of the spatiotemporal focal region would be expected to be dominant. The zero-field minimum photon order necessary to overcome the binding energy of the outermost electron in F^- is 5. The minimum photon order required to supply both the binding energy and the ponderomotive energy of the detached electron is 11. This rather high required photon order, in combination with the modest value of z_1, means that the maximum-intensity part of the spatiotemporal intensity distribution in the laser focus should be dominant.

The maximum ponderomotive energy measured in the experiment is $U_p=5.5$ eV [10] and the momentum distribution gives 5.2 eV. Using the measured maximum intensity of 2.6×10^{13} W/cm^2, the calculated spectrum has a peak at about 5.4 eV, subject to some uncertainty due to the irregular shape of the peak. This is in excellent agreement with the laboratory measurements of the properties of the photoelectron spectrum. The calculations thus describe very well the photoelectron properties at and near the all-important peak in the spectrum.

The spectral distribution from the velocity-gauge SFA is somewhat narrower than the measured spectrum. This can be ascribed to the marginal value of z_1 in Eq. (6). It is important that with the much more satisfactory (from an intense-field point of view) z_1 value in Eq. (8), there is no such problem with the width of the spectrum.

For all of these reasons, including the all-important fidelity of the calculated spectrum to the measurements near the peak, the fact that the accurately measured maximum intensity was used without adjustment, and the somewhat marginal value of z_1, it is concluded that the velocity-gauge SFA performs very well in reproducing the experimental results.

IV. SUMMARY AND DISCUSSION

As just concluded, the velocity-gauge SFA performs well in comparison with the experimental results [10] on the photodetachment of F^- by circularly polarized laser radiation at a wavelength of 1510 nm. Both Refs. [10,11] conclude, in contrast, that a length-gauge-based theory works better. Some comments on the length-gauge results are therefore appropriate.

First, there is the general matter that a laser field is a transverse field, and a single scalar component of the fourcomponent electromagnetic potential vector cannot properly describe a transverse field. As mentioned, however, this is not decisive because of the limited intensities reached in the experiment.

Second is the matter that, to obtain an apparent fit of the length-gauge theory to the experimental spectrum, a peak intensity of 3.8×10^{13} W/cm^2 had to be assumed. (Reference [11] also considers 3.4×10^{13} W/cm^2.) This intensity differs from the measured value by 46%, far beyond the 15% uncertainty in the intensity measurement.

Third, the momentum distribution results presented in Fig. 1 of Ref. [10], leading to an estimated kinetic energy of 5.2 eV, are completely consistent with the measured peak ponderomotive energy of 5.5 eV, but far distant from the maximum $U_p=8$ eV that would be demanded by an assumed peak intensity of 3.8×10^{13} W/cm^2 necessary for the length-gauge calculation.

The claim that the length gauge theory provides a fit to the data superior to that of the velocity gauge is strongly contested here.

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