Energetic electrons in strong-field ionization

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A strong-field theory is extended to the description of energetic photoelectron spectra. Experimental spectra are accurately described for both linear and circular polarizations, subject only to a single adjustment of reported peak laser intensities. Examined are single and (sequential) double ionization of helium, to energies of about 1 keV. An approximation of the spectrum as a Maxwellian distribution yields a temperature dependent on the spectrometer viewing angle. Final-state Coulomb effects in circular polarization are examined. [S1050-2947(96)50509-3]

Experiments by Mohideen *et al.* [1,2] and by Walker *et al.* [3] on the ionization of helium by short, intense laser pulses have yielded photoelectron spectra that are remarkably ''hot,'' with energies up to 1 keV, even though each laser photon possesses only about 1.5 eV. It had been concluded [1,2] that no analytical theory exists that can explain the high energies observed. Direct numerical solution of the Schrödinger equation is too computer-intensive to be feasible for the energetic spectra considered here. No theory of any kind appeared to be applicable.

It is shown here that the SFA (strong-field approximation) [4,5] gives excellent agreement with the experimental spectra [1–3] from modest photoelectron energies all the way to the most energetic photoelectrons observed. This is true for circular laser polarization, and, with qualifications, also for linear polarization. The agreement applies to both single ionization and sequential double ionization of helium. We are unaware of any other theoretical method that makes possible a calculation of such energetic spectra. We show results for double-ionization spectra that extend nearly to 1 keV. We conclude that it is possible to describe analytically even the most energetic of the photoelectrons. We also demonstrate the practical application of a Coulomb-corrected SFA (CSFA) for circular polarization [6].

It has been observed [1,2] that the photoelectron spectrum (for linear polarization) is not unlike that of a Maxwellian thermal distribution. We show that the apparent temperature of the electron spectrum depends upon the acceptance angle of the electron spectrometer, and is thus not a fundamental property of the ionized electrons.

This physical problem is not in the tunneling domain in the sense that overbarrier ionization can occur. About 1.5×10^{15} W/cm^2 of laser intensity is required to ionize He, by suppression of the combined Coulomb and electric-field potentials, to a point that lies below the ionization potential of helium. Peak experimental laser intensities employed extend beyond this value. Further, ''hot'' electrons in the spectra represent high-order ATI (above-threshold ionization) electrons with substantial amounts of kinetic energy. A tunneling description is thus inappropriate. It has become customary to label as ''tunneling'' any process in which ATI peaks in the spectrum cannot be resolved. This is an unfor-

tunate practice in that the actual physical process is most likely to be by barrier penetration in the so-called ''multiphoton'' regime, but can be over the barrier in the misnamed ''tunneling'' regime. Conventional terminology is thus backward in this sense. We note that the SFA is independent of any restriction to tunneling or otherwise.

The SFA transition amplitude is formed by noting that an electron state, in interaction with both atomic and strong laser fields, is analyzed in the laboratory in a region outside the field. Thus the transition amplitude is given by an overlap between an interacting state and a ''reference'' state free of the laser field. One can elect to employ the time-reversed amplitude, so that it is the initial state that is free of the field. This is a critical advantage, since the fully interacting initial bound state is difficult to approximate, whereas the interacting final detached state can be approximated easily when the magnitude of the laser interaction energy of the free electron (U_p) dominates the atomic binding energy (E_B). In the problem at hand, the ponderomotive potential U_p can extend to hundreds of eV. The only approximation of the SFA is that the final-state electron is presumed to be a Volkov state, with its motion governed by the laser field. Thus the SFA includes the oscillations of the detached electron in the laser field. It also includes what would appear in the numerical solution of the Schrödinger equation as wave-packet spreading of the ionized electron. In the present theory, this comes from a smearing in angular distributions due to the combined effects of the Volkov oscillations of the ionized electrons in the field and the spatial and temporal variations in intensity.

To neglect the attraction of the residual ion for the ionized electron, which is the approximation made by the SFA, requires $z_1 \gtrsim 10$, where $z_1 \equiv 2U_p/E_B$. For the linear polarization data of Mohideen *et al.* [1,2], we find that the peak value of z_1 is about 18. The calculations are compared to the data in Fig. 1. Linear polarization poses a difficulty for the SFA in that linear polarization spectra peak strongly at low photoelectron energies where the SFA understates the transition rate. We have consistently found that one must assume a peak laser intensity of 1.8–2.0 times the best laboratory estimates. This bias of the SFA is observed in total ion yields as well as in spectra. For the computations, we employ a peak intensity of 3.6×10^{15} W/cm^2, but conclude that the true
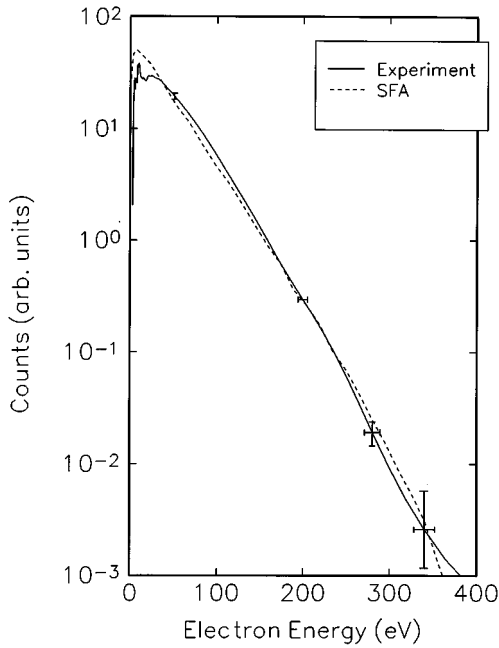
FIG. 1. Linear polarization spectrum for He$^+$ compared with Ref. [1]. Error bars are as given in [1]. The 7×10^{15} W/cm^2 cited in [1] is reduced to 3.6×10^{15} W/cm^2 in the theory.

peak laser intensity is most likely to be about 1.8×10^{15} W/cm^2, which is consistent with what we shall find for circular polarization. The agreement is excellent, in view of the fact that calculated results are within experimental error bars from about 100 eV to the highest energies shown at nearly 400 eV.

For the circular polarization spectrum given by Mohideen *et al.* [1,2], the peak z_1 is about 6.4. This is not adequate for confident application of the SFA, so we employ a Coulomb-corrected SFA [6] available for circular polarization when $\alpha_0 \geqslant 10$, where α_0 is the radius of motion of a free electron rotating under the influence of a circularly polarized field. For the experiments, the peak value is $\alpha_0 \approx 43$, so the Coulomb SFA should work well. This is the case, as seen in Fig. 2, which shows the comparison of the CSFA with the data, with a presumed peak value of the laser intensity of 1.275×10^{15} W/cm^2 in place of the 6×10^{15} W/cm^2 stated by Mohideen *et al.* [1,2]. From very general considerations, a strong-field photoelectron spectrum for circular polarization should peak at an energy near U_p. We find the peak to be at $0.8U_p$. If the experimentally stated peak intensity were correct, the peak would be at only $0.17U_p$, which is not plausible. The sensitivity to peak intensity of the spectral peak location, width of the peak, and shape of the high-energy tail are so acute as to lead us to regard the agreement in all these features shown in Fig. 2 as warranting the conclusion that the theory can be used to determine the actual peak intensity. The value of the peak laser intensity can be specified to within better than $\pm 5\%$. We conclude that Mohideen *et al.* overstated the peak laser intensity by a factor of about 4 with both linear and circular polarizations.

A proper comparison of theory and experiment requires an appropriate initial-state wave function (we employ an analytical Hartree-Fock wave function for helium [7]) and an accurate representation of the spatial and temporal intensity
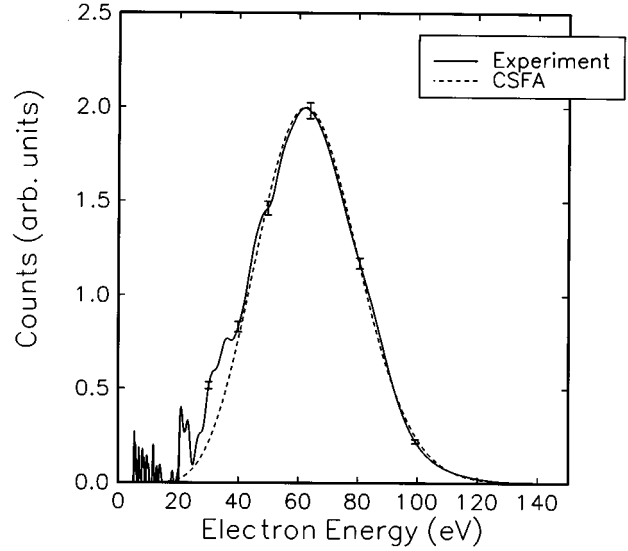

FIG. 2. Circular polarization spectrum for He$^+$ compared with Ref. [1]. Error bars are as given in [1]. The 6×10^{15} W/cm^2 cited in [1] is reduced to 1.275×10^{15} W/cm^2 in the theory.

profiles of the focused laser pulse. The negative conclusion of Mohideen *et al.* [1,2] about the possibility of explaining the hot electrons is an artifact of their procedure, which employed a monochromatic approximation of the theory.

To further test the ability of the SFA to predict the spectrum of energetic electrons in strong-field ionization, we turn to the experiments of Walker *et al.* [3], also with helium. The Walker experiments go to intensities high enough to cause extensive double ionization. Doubly ionized helium has contributions from both sequential and nonsequential processes [8–10]. The SFA, as employed here, applies to the sequential process, although this is not a fundamental restriction [11]. A spectrum for sequential double ionization of helium [3,12] is calculated as in Fig. 3. The peak intensity of 6×10^{15} W/cm^2 cited by the experimentalists was increased to 1.4×10^{16} W/cm^2 for the theory, and the peak intensity of 4.9×10^{15} W/cm^2 was increased to 1.0×10^{16} W/cm^2. (This is in contrast to the decrease found necessary in the Mohideen intensities.) In view of the systematic bias to overstated intensities of the linear polarization SFA, our final estimate for actual peak intensities is about 7×10^{15} W/cm^2 and 5×10^{15} W/cm^2, results close to those cited by the experimentalists. We see that the agreement is very good out to the end of the data at about 1 keV.

The procedure employed in calculating the spectrum from the theory and taking account of the spatiotemporal profile of the laser pulse and the depletion of the atoms initially present is summarized for single ionization. The density of ions formed in a local volume situated at $\mathbf{r},t$ is given by the solution of the rate equation as

$$\frac{N(\mathbf{r},t)}{N_0} = 1 - \exp\left\{ -\int_{-\infty}^{t} d\tau \, W[I(\mathbf{r},\tau)] \right\}, \qquad (1)$$

where N_0 is the initial density of neutral atoms, and W is the SFA transition rate as a function of the intensity $I(\mathbf{r},t)$. When a total ion yield is calculated, the $N(\mathbf{r},t)$ of Eq. (1) is integrated over the spatial and temporal profiles of the laser
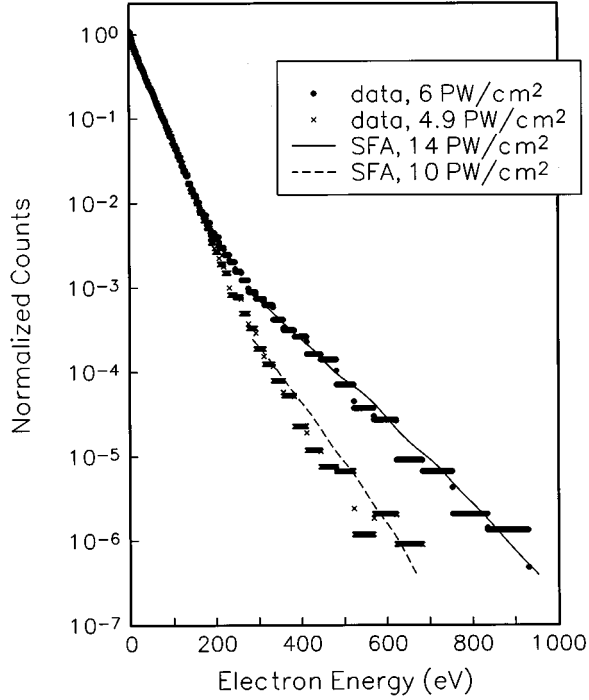
FIG. 3. Linear polarization spectrum for He^{2+} compared with Ref. [3]. The 6.0 and 4.9×10^{15} W/cm^2 cited in [3] are increased to 1.4 and 1.0×10^{16} W/cm^2 in the theory. The data beyond the "knee" in the spectrum at about 250 eV are ascribed [12] to sequential double ionization.

beam. Depletion of the atoms in the focus of the laser is automatically incorporated by using the rate equation. The total ion yield $\mathfrak{N}$ is the integral of Eq. (1) over the beam profile,

$$\mathfrak{N} = 2\pi N_0 \int_{-\infty}^{\infty} dz \int_0^{\infty} r\, dr \left(1 - \exp\left\{ -\int_{-\infty}^{t} d\tau\, W[I(\mathbf{r},\tau)] \right\} \right),$$

(2)

using cylindrical coordinates r,z. For a laser beam of wavelength λ with Gaussian time distribution of full width T_0 brought to a focus with waist radius w_0, the intensity profile employed in Eq. (2) is

$$I(r,z,t) = I_0 \exp[-(2t/T_0)^2] \frac{1}{[1+(\lambda z/\pi w_0^2)^2]}$$

$$\times \exp\left\{ \frac{-2r^2}{w_0^2[1+(\lambda z/\pi w_0^2)^2]} \right\}.$$

(3)

To calculate a spectrum, it is necessary to find that part of $N(\mathbf{r},t)$ due to a number s of photons above the threshold order n_0, given by $n_0 = \{(1/\omega)(E_B + U_p)\}$, where the symbol $\{\}$ signifies the smallest integer containing the quantity inside the braces. It is desired to find the contribution of the particular spectral peak of ATI order n_s, where $n_s = n_0 + s$. We write $W = \Sigma_{n_0}^{\infty} W_n = (\Sigma_{n \neq n_s} W_n) + W_s$. Since s is a single order among many, we assume $\int_{-\infty}^{\infty} d\tau\, W_s \ll 1$. We find that

$$\frac{N_s}{N} = \left(\int_{-\infty}^{t} d\tau\, W_s \right) \exp\left(-\int_{-\infty}^{t} d\tau\, W \right).$$
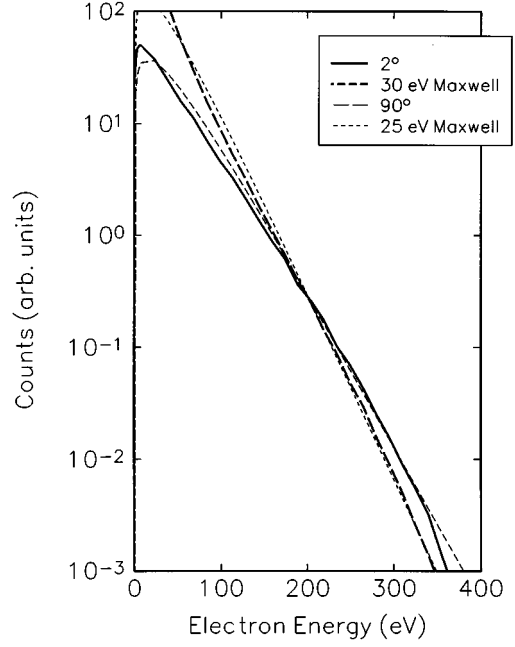
(4)

FIG. 4. Maxwell distribution fit to the linear He$^+$ data [1]. The spectrometer acceptance angle of 2° employed in the experiments is best fitted by a Maxwell distribution with a temperature of 30 eV. If the spectrometer could view all angles, the spectrum would best be described by a Maxwellian with a temperature of 25 eV. The figure pairs the spectrum calculated with the 2° acceptance with the 30-eV Maxwellian, and the all-angle acceptance with the 25-eV Maxwellian.

Here, W_s is the partial transition rate for only the order s and for only that part of the angular distribution contained within the acceptance angle of the spectrometer, whereas W is the total transition rate for all orders and all angles. In the evaluation of the spectrum, we let $t \to \infty$, and integrate over the spatial volume to obtain

$$\mathcal{N} = N_0 \int_V d^3r \left\{ \int_{-\infty}^{\infty} d\tau\, W_s[I(\mathbf{r},t)] \right\}$$

$$\times \exp\left\{ -\int_{-\infty}^{\infty} d\tau\, W[I(\mathbf{r},t)] \right\}.$$

(5)

A generalization of this procedure to the second-order rate equation is used for the calculation of the sequential double-ionization spectrum.

This procedure involves no arbitrary parameters. The only selection made is that of the peak laser intensity.

It was observed by Mohideen et al. [1,2] that the spectrum in linear polarization can be approximated reasonably well by a Maxwellian distribution. They find the best fit to be about 30 eV, with which we concur, as shown in Fig. 4. Their data were collected with a spectrometer acceptance angle of 2°. Since we now find that the SFA agrees well with the data, one can use the SFA to explore whether this temperature is a universal characteristic of the experiment or whether it is dependent on the acceptance of the spectrometer. The latter possibility obtains, as shown in Fig. 4. If one were to have a spectrometer that could accept a 4π solid
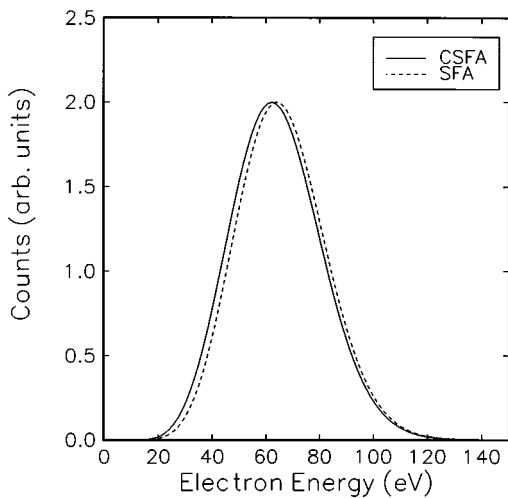
FIG. 5. Comparison of SFA and CSFA predictions for the same conditions as in Fig. 2.

angle, the closest Maxwellian temperature would be 25 eV, not 30 eV. The apparent electron temperature is thus not a fundamental feature of strong-field photoionization.

Experimental conditions are marginal for application of the SFA to the circular polarization results in Fig. 2, and so the CSFA was used. Figure 5 gives a comparison of the SFA and CSFA. The small displacement and narrowing of the peak in the SFA are not major, but noticeably degrade the excellent agreement seen in Fig. 2. Not visible in these figures with normalized amplitudes is the fact that the CSFA spectrum has about 1.8 times the absolute amplitude of the SFA spectrum. The explanation for these differences is that the Coulomb correction enhances rates at lower intensities, but makes no difference at higher intensities. The CSFA spectral peak is slightly shifted towards a lower energy, and the low-energy tail is slightly enlarged as compared to the SFA, with these modifications due to the increased weight acquired in the CSFA by the lower-intensity parts of the spatiotemporal distribution. This also accounts for the larger overall absolute amplitude of the CSFA.

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