Observation of Laser Assisted Photoelectric Effect and Femtosecond High Order Harmonic Radiation

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We report the first observation of laser-induced free-free transitions in the primary photoelectron spectra of gaseous helium ionized by ultrashort soft x-ray pulses. Measured transition amplitudes are well described by projecting the initial electronic state onto a Volkov wavefunction. Additionally, we report the first direct measurements on the temporal duration of femtosecond high order harmonic radiation. The harmonic pulse duration is observed to exhibit strong dependencies upon both laser intensity and position of the generating medium relative to the laser focus.

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Laser-induced transitions of an electron from one continuum state to another (free-free transitions) are of fundamental interest because processes involving differing numbers of photons can occur with comparable probability. Accordingly, models of laser interaction with matter can be assessed in a nonperturbative regime. Free-free transitions also have considerable practical significance since they are of central importance in the heating of plasmas by electromagnetic waves and can, in principle, be used to measure ultrashort x-ray pulses with significantly higher temporal resolution than is currently obtained using x-ray streak cameras ($\sim$ 1 ps). Direct observation of multiple absorptions and emissions of photons by electrons in a laser field was first reported by Weingartshofer *et al.* in experiments on laser assisted charged particle scattering [1]. More recently, continuum transitions have been studied using single and two-color above-threshold ionization [2] and using laser assisted Auger decay [3].

In the present work free-free transitions are studied through observation of the laser assisted photoelectric effect (LAPE). The primary photoelectron (PE) spectrum resulting from soft x-ray ionization is observed to exhibit two distinct modifications due to the presence of a high intensity laser pulse during the ionization process. First, absorption and emission of laser photons during ionization gives rise to sidebands in the PE spectrum. Second, the PE spectrum is observed to shift to lower energy as a result of a laser-induced increase in the binding energy of the ionized (gas) medium. Measured free-free scattering amplitudes are well described by approximating the final electronic wave function as that of free particle oscillating in the laser field, i.e., as a Volkov wave function [2]. Finally, we demonstrate the first measurement of soft x-ray pulses shorter than 100 fs with the use of a laser and x-ray cross correlation technique based on LAPE. The measurements allow for the first direct investigation of parameters critical to determining the temporal duration of femtosecond high order harmonic radiation [4]. We observe a dramatic variation in the harmonic pulse duration (ranging from 50 to 120 fs) with variation of the laser intensity and position of the harmonic generating medium.

The laser system used in these experiments is based on titanium:sapphire and delivers 70 fs, 800 nm pulses at 10 Hz with energies up to 65 mJ. The laser intersects a beam splitter; one beam is used to generate harmonic radiation, while the other beam is directed out of a vacuum chamber, through an optical delay arm, and then propagated back into vacuum for subsequent spatial and temporal overlap with the harmonic radiation. High order harmonics are generated using a 75 cm focal length lens which focuses the laser at $f/50$ (200 μm focal spot diameter, 8 cm confocal parameter) to the exit of a pulsed gas valve. The valve is backed with argon and delivers a density of $\sim 10^{18}$ atoms/cm^3. The harmonics pass through a 1500 Å thick aluminum filter (which eliminates 800 nm radiation) and propagate $\sim$2 m to a Mo:Si multilayer-coated curved mirror. The multilayer (peak reflectivity of 22% at 38 eV and 4 eV FWHM bandpass) focuses soft x-ray pulses to the exit of a second pulsed gas valve backed with helium. We estimate that the focused x-ray spot diameter is 20 μm. The output pressure of the He gas jet (between 0.1 and 1 Torr) is carefully adjusted to obtain the maximum signal level without creating significant space charge broadening of the PE peaks. Photoelectrons generated by soft x-ray ionization of He are collected by a pair of flight-time-preserving reflecting parabolic grids. The grids collect electrons over $\sim 4\pi$ and direct them along a 1 m flight tube to a microchannel plate detector. Electron energies are recorded via time-of-flight spectroscopy using a 2 Gs, 500 MHz digitizing oscilloscope. Modifications to the PE spectra are induced by focusing the delayed laser pulses to the exit of the He gas jet at $f/67$ (450 μm focal spot diameter) through a 4 mm diameter hole in the multilayer mirror.

A PE spectrum obtained at a peak laser intensity of 1×10^{15} W/cm^2 is shown by the dashed curve of Fig. 1(a). Harmonics 21 (33 eV) through 27 (42 eV) are evident and result in four PE peaks ranging from 8 to 17 eV. The

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presence of the delayed laser induces two distinct modifications to the PE spectrum. First, sidebands appear at integral units of the photon energy (1.55 eV) as evidenced by the solid curve of Fig. 1(a). Second, the PE peaks appear at positions ~200 meV lower in energy than expected based on the zero field peak positions. The dashed vertical lines of Fig. 1(a) (inset) illustrate this effect for the peak near 17 eV. We make two comments concerning this second effect. First, the solid and dashed curves of Fig. 1(a) represent two different relative time positions of the x-ray and laser pulses. This indicates that space charge is not the cause of the observed shift. Second, the shift varies linearly with delayed laser pulse intensity and its magnitude is consistent with the ponderomotive potential (U_p) of the delayed laser pulse [2]. The shift can therefore be used to calibrate the delayed laser intensity.

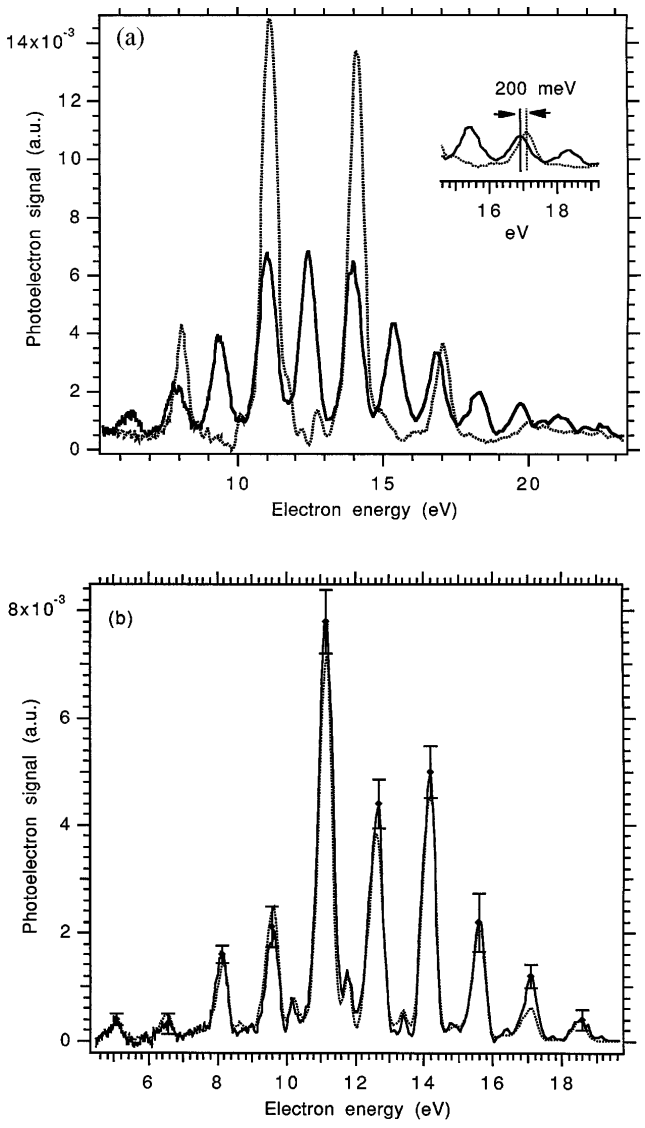

FIG. 1. (a) Photoelectron spectra obtained in the presence (solid line) and absence (dashed line) of the laser pulse. The inset shows an expanded view indicating a ponderomotive shift in the photoelectron peak positions. (b) Measured photoelectron spectrum obtained in the presence of the laser field (solid line). The dashed curve is a theoretical prediction discussed in the text.

The observed shift results from an increase in binding energy (E_b) an atom experiences in a high intensity laser field [2]. The effect can be understood in terms of an ac Stark shift (by an amount equal to U_p) of continuum states or, equivalently, by the fact that upon ionization a photoelectron must acquire sufficient energy to exist as a positive energy electron oscillating in the laser field. The observed shift in the PE peak position indicates that this extra energy is not supplied by the laser field but comes at the expense of whatever excess energy the electron has from the (x-ray) photoelectric effect. This suggests the interesting possibility of using a high intensity laser (with $U_p > h\nu_{\mathrm{x\ ray}} - E_b$) to suppress the photoelectric effect. While x rays typically dominate optical radiation in interaction with core electrons, the strong modification of continuum states by a high intensity laser can, in principle, inhibit x-ray interaction with core electrons.

We next discuss the sideband structure. The LAPE spectrum is modeled under the assumption that the initial electronic state is, under action of the x-ray field, projected onto a Volkov state. The Volkov wave has been useful in describing certain aspects of high intensity laser-atom interaction. The S matrix is calculated in the radiation gauge with the initial electronic state taken as hydrogenic. We obtain the following expression for the probability that the photoelectric effect is accompanied by absorption ($n > 0$) or emission ($n < 0$) of n laser photons:

$$A_n = (3/2)\,(p/p_0)^3[\{1 + (a/\hbar)^2 p_0^2\}/\{1 + (a/\hbar)^2 p^2\}]^4 \times \int_0^{\pi} \sin(\Theta)\cos^2(\Theta) J_n^2(\alpha,\beta)\, d\Theta\,. \quad (1)$$

In (1), a is the Bohr radius divided by the nuclear charge; the momenta, p_0 and p, are $p = [2m(h\nu_{\mathrm{x}} + nh\nu - E_b - U_p)]^{0.5}$ and $p_0 = [2m(h\nu_{\mathrm{x}} - E_b)]^{0.5}$ where m is the electron mass, $h\nu_{\mathrm{x}}$ is the x-ray photon energy, $h\nu$ is the laser photon energy, E_b is the field free binding energy, and U_p is the ponderomotive potential of the laser. The generalized Bessel function, $J_n(\alpha,\beta)$, is discussed in [5], and its arguments are $\alpha = eA_0 p\cos(\Theta)/mch\nu$, $\beta = -U_p/2h\nu$ where e is the electron charge and A_0 is the vector potential of the laser.

Scattering probabilities for each field-free peak in the PE spectrum are calculated using Eq. (1). A series of delta functions spaced by the laser photon energy, with amplitudes determined by the scattering probabilities, is then convolved with the zero field PE spectrum. Calculated scattering probabilities for the 23rd harmonic (at 7.4×10^{11} W/cm^2) are: $A_0 = 0.593$, $A_1 = 0.197$, $A_{-1} = 0.174$, $A_2 = 0.020$, and $A_{-2} = 0.010$. The model calculation is compared to a measured (laser modified) spectrum in Fig. 1(b). Also shown in Fig. 1(b) are standard deviations for the measured peak height amplitudes as determined from multiple data acquisitions.

Figure 1(b) indicates that the model describes the measured spectrum quite well. We emphasize that there are no free parameters in the comparison between data and theory as the laser intensity is calibrated from observed ponderomotive shifts. The most significant deficiency in the present theory is the neglect of modifications to the final electronic wave function resulting from the presence of the Coulomb potential. The agreement observed between data and theory indicates that these effects are not severe, a result consistent with the expectation that atomic "dressing" of the Volkov wave is most significant near the (x-ray) photoionization threshold [5]. The success of this relatively simple approach to LAPE is encouraging in light of the fact that an exact analytic expression for the final state of the electron, which incorporates both the laser and Coulomb potentials, remains an unsolved problem.

We next turn the discussion to measurements of femtosecond high order harmonic radiation. While high order harmonic generation has been the subject of intense theoretical and experimental investigation for more than 10 years [4], experimental investigation into the temporal duration of high order harmonic radiation has been confined to lower orders (7th and 9th) and picosecond time scales [6]. X-ray pulses can be measured via cross correlation with laser pulses by observing either the ponderomotive shift or the change in PE peak height as a function of relative pulse delay. While ponderomotive shifts provide a certain advantage in that they do not saturate as can (at high intensity) changes in LAPE peak height, correlations are obtained by measuring peak height changes in the LAPE spectra since the lower laser intensity required for this effect results in a lower level of laser-induced background.

Cross correlation measurements were performed with the laser beam propagating colinearly with the x-ray beam. The crossing angle between the two pulses was 10 mrad so that broadening of the correlation widths due to geometrical factors is negligible (<1 fs). The laser pulse duration was carefully measured using a single shot autocorrelator and is determined to be 69 ± 3 fs. While measurements have been made over a span of several months, all of the temporal data presented in this work was measured in a continuous 30 h period during which the laser pulse duration was checked several times and never varied beyond the 3 fs margin mentioned above.

Cross correlations are shown in Fig. 2. The data of Fig. 2 (diamonds) is obtained by measuring the height of the 11 eV PE peak (23rd harmonic) as a function of delay. The argon gas jet was positioned 3 cm beyond the laser focus, and the intensity of the harmonic generating beam at the focal point was 1.4×10^{15} W/cm^2. The delayed laser intensity was sufficiently low that the ponderomotive shift in the spectrum is negligible and the change in peak height was determined to vary linearly with laser intensity. A Gaussian least squares fit to the data indicates a correlation FWHM of 85 fs. Gaussian curves at ± 15 fs are also shown in Fig. 2. The 85 fs correlation width

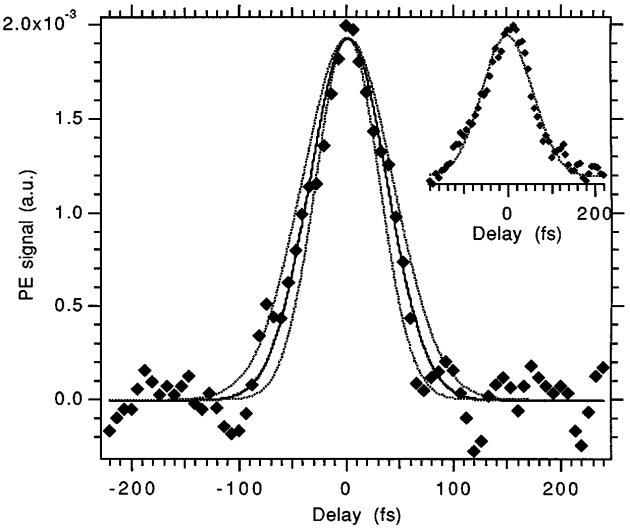

FIG. 2. A cross correlation obtained at $z = 0.6b$ and 1.4×10^{15} W/cm^2. The data points are shown as diamonds and the least squares Gaussian fit (FWHM = 85 fs) to the data is shown as a solid curve. The deconvolved soft x-ray pulse duration is 50 fs. Also shown are Gaussians at ± 15 fs (dashed curves). The inset shows a cross correlation obtained at $z = 0.6b$ and 1.9×10^{15} W/cm^2. The least squares Gaussian fit (FWHM = 138 fs) to the data is shown as a solid curve and indicates a soft x-ray pulse duration of 120 fs.

indicates that the harmonic pulse duration is 50 ± 15 fs (FWHM). The error bars are determined by taking the error margin of the correlation (dashed lines of Fig. 2) in quadrature with the error margin of the laser (3 fs).

In previous experiments on lower order harmonics of picosecond duration, substantial shortening of the harmonics relative to the laser pulse was observed [6]. Perturbation theory indicates that the yield of the qth harmonic scales with laser intensity as I^q, thus predicting a factor of $q^{-0.5}$ shortening for the approximately Gaussian temporal profile of our laser pulse. The measured pulse of Fig. 2 is only 30% shorter than the laser pulse, significantly longer than the factor of 5 shortening expected from $q^{-0.5}$ scaling. It is well established, however, that high harmonic generation is nonperturbative and that the yield of the qth harmonic scales with intensity as I^p, where p is an experimentally determined parameter which is less than q [4]. In order to determine whether the harmonic pulse duration obeys a $p^{-0.5}$ scaling, the value of p was measured.

The laser energy was increased to produce a peak intensity of 1.9×10^{15} W/cm^2. The 36% increase in laser intensity increased the yield of the 23rd harmonic by a factor of 6 which indicates an intensity scaling of $I^{6.6}$. This value of p implies a pulse duration (27 fs) significantly shorter than the measured value (50 fs); the harmonic duration is therefore not determined by a simple scaling of the effective nonlinearity p. This conclusion is substantiated by the dramatic increase in the harmonic pulse duration obtained at the higher laser intensity. Figure 2 (inset) shows the correlation obtained

with the higher laser intensity along with the least squares Gaussian fit to the data. The correlation FWHM is 138 fs which implies a factor of 2.4 increase in x-ray pulse duration to 120 fs. We therefore observe the counterintuitive yet significant result that high harmonic radiation can be significantly *longer* than the generating laser pulse duration.

The strong dependence of harmonic pulse duration on laser intensity indicates that phase matching may play a dominant role in determining the pulse duration; phase matching is known to be highly dependent on laser intensity [7]. Phase matching is also expected to vary from one side of the laser focus to the other since a focused laser beam undergoes a phase shift upon propagation through the focal spot [7]. The pulse duration was therefore measured by positioning the gas jet on the other side of the laser focus. We adopt a convention where z is the distance between the gas jet and the laser focus and is positive for gas jet positions beyond the laser focus. The above-mentioned measurements were made at $z = 0.4b$ (b is the confocal parameter). The gas jet was moved to $z = -0.2b$ and the harmonic pulse duration, measured at 5.5×10^{14} and 9.4×10^{14} W/cm^2, was 95 and 100 fs, respectively. While the harmonic pulse duration does change from $z < 0$ to $z > 0$, the strong intensity dependence observed for $z > 0$ is (to within error bars of ± 15 fs) not reproduced for $z < 0$.

The duration of femtosecond high order harmonic radiation has been modeled by Salieres *et al.* [7]. The results indicate pulse durations ranging from half the laser pulse duration to nearly the full laser pulse duration. The variation of pulse duration is principally a result of the strong intensity dependence of the phase of the atomic dipole response. Since focusing causes the laser intensity to vary (both longitudinally and transversely) throughout the generating medium, phase matching can occur at different points along the temporal profile of the laser pulse and can dramatically affect the harmonic pulse duration. While our results seem to substantiate some aspects of [7] (namely, the importance of phase matching) there are discrepancies. For instance, Ref. [7] indicates that the pulse duration changes by only 10% between $z = 0.6b$ and $-0.2b$; we observe a factor of 2 change in pulse duration over a similar range in z. While there are differences in the experimental parameters of the present work and those chosen in [7], a potentially significant mechanism which is not studied in [7] is ionization of the harmonic generating medium. We calculate, using tunneling ionization rates [8], that at all laser intensities used in this work Ar is fully ionized (to $z = 1$) over the temporal duration of our laser pulse. We identify three ways in which ionization is expected to play a significant role in harmonic generation. First, depletion of the ground state by ionization acts to limit the generated polarization as predicted by the atomic dipole response. Second, ionized electrons are expected to dramatically affect the phase matching [4] and hence the harmonic pulse duration. Third, harmonics measured in this work may result not only from the neutral species but also from ions. Harmonic generation from ions has been suggested by recent observations [9]. Experiments to elucidate the relative importance of the above-mentioned effects are in progress.

In summary, we observe LAPE in the dual interaction of high intensity laser and high order harmonic radiation with gaseous helium. Photoelectron peaks are observed to shift to lower energy due to ponderomotive increase in the atomic binding energy. Measured free-free scattering amplitudes are well described by projecting the initial electronic state onto a Volkov wave function. Ultrashort visible/x-ray cross correlations are obtained and the resulting 50 fs pulses are the shortest measured x-ray pulses by a factor of 20. The duration of high order harmonic radiation is observed to vary with both laser intensity and position of the generating medium relative to the laser focus. These initial measurements of femtosecond high order harmonic radiation elucidate the complex nature of the harmonic generation process and suggest the importance of incorporating ionization effects into the modeling.

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Note added.—Subsequent to submission of this manuscript, we learned that cross correlation measurements of high harmonic radiation have recently been performed by Schins *et al.* [10] using a technique similar to that described in this Letter.

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