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Nonresonant above-threshold ionization by circularly polarized subpicosecond pulses

P. H. Bucksbaum, L. D. Van Woerkom, R. R. Freeman, and D. W. Schumacher
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
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Recent experiments show that above-threshold ionization (ATI) with intense linearly polarized light occurs via Stark-shifted bound-state resonances. Under identical conditions, we find that ATI with subpicosecond *circularly polarized* light is nonresonant. Unlike longer-pulse experiments, these spectra directly show the ionization rate versus intensity, and can therefore test ATI theories without deconvolving the inhomogeneous laser focus. Keldysh-Faisal-Reiss calculations do not agree with these new experimental results.

Above-threshold ionization (ATI) is the production of photoelectrons with more energy than the electromagnetic quantum $\hbar\omega$.[1,2] This is a general feature of multiphoton ionization of atoms and simple molecules at intensities of 10 TW/cm^2 or higher, where the electromagnetic fields approach the strength of the atomic field. Recently, several experiments have shown that ATI by linearly polarized subpicosecond laser pulses is assisted by intermediate bound-state resonances.[3-8] Although no bound states may be in resonance with the laser frequency (or integer multiples) in the unperturbed atom, large ac Stark shifts induced by the enormous light fields in these experiments cause states to shift into multiphoton resonance during the laser pulse. Resonances are observed in nearly all subpicosecond ATI experiments, leading to speculation that purely nonresonant direct ionization may never occur in the high-intensity, high-order multiphoton ATI regime: resonant enhancements inevitably intervene at intensities below those necessary for direct ionization.[9,10]

In this Rapid Communication, we report new evidence for direct *nonresonant* photoionization from the ground state to the continuum for ATI by *circularly polarized* light. The experiment was performed on xenon, photoionized by 140-fsec pulses of 616-nm light. Photoelectron spectra from these new experiments yield direct measurements of the relative photoionization rates into each ATI peak as a function of laser intensity.

The ATI spectrum is known to change with the laser polarization,[11,12] and this dependence provides an important test of ATI theories.[13-16] Some simple approaches to this problem, such as Keldysh-Faisal-Reiss (KFR) theories,[17-19] have reasonable success in calculating effects observed in circular polarization.[13,14] However, existing studies of polarization dependence have employed pulses of 50 psec or longer.[11,13,20-22] With these rather long pulses, much information about the ATI process, including the presence of resonances, is lost through the ponderomotive forces on the free electrons as they escape the laser focus.[23] Ponderomotive forces, which are due to the stimulated Thomson scattering of laser photons,[24] can steer and accelerate the electrons, thereby corrupting both the observed energy spectra and angular distributions.[25]

Ponderomotive contamination of the energy spectra can be avoided by using subpicosecond laser pulses, which turn off before there is appreciable momentum transferred between the laser pulses and the electrons.[3] The subpicosecond pulses measure the dependence of the ionization rate on laser intensity in a model-independent way.[7] Subpicosecond ATI experiments with circular polarization are therefore critical new tests of ATI theories.

These experiments used 616-nm pulses ($\hbar\omega = 2.01$ eV) from an amplified colliding-pulse mode-locked dye laser,[26] with a pulse duration of 140 fsec, operating at a repetition rate of 10 Hz. The laser pulses were circularly polarized, with $I_{\text{right}}/I_{\text{left}} \approx 1000$ or better.

A focus with a known and reproducible intensity distribution was formed in a field-free high-vacuum region containing a target gas of xenon with a density of 10^{10} to 10^{13} cm^{-3}. The xenon density could be adjusted to avoid space-charge effects in the photoionization spectra. Photoelectrons from ATI were energy-analyzed by field-free time of flight.

Figure 1 is a comparison between a typical ATI photoelectron spectrum using linearly polarized light, and a spectrum with the same laser intensity and pulsewidth, but circular polarization. The linear-polarization ATI spectrum is dominated by a series of closely spaced peaks, which have been identified as xenon bound states, Stark shifted into resonance by the intense laser field.[3]

In the limit of very short laser pulses, the energy of any ATI electron leaving the laser focus is given by

$$E_{\text{final}} = n\hbar\omega + E_0 - U_P \quad \text{(short pulse limit)}, \tag{1}$$

where E_0 is the (negative) difference between the energy of the ground-state ion and the ground state of the initial atom, and U_P is the ponderomotive potential[24] $U_P = (2\pi e^2 I)/(m_e c\omega^2)$. Here e is the electron charge, m_e its mass, and I, ω, and c are the intensity, angular frequency, and speed of the light, respectively. Both U_P and E_0 are evaluated at the point of ionization. For tightly bound atoms such as the rare gases in these laser fields, experiments carried out with long laser pulses[27] and calculations[28] show that E_0 is shifted very little from its field free value ($E_0 = 12.13$ eV for xenon). Thus we arrive at the important conclusion that *the measured energy of the electron in subpicosecond ATI experiments is directly related to the intensity of the field where the atom was ionized.*[3,4,7]

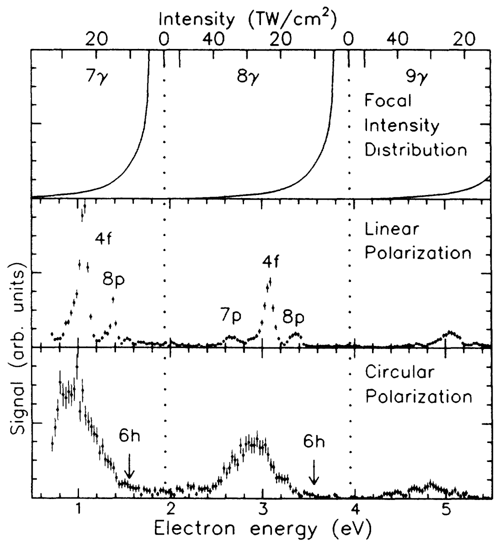

FIG. 1. Linear polarization: ATI spectrum for xenon ionized by 140-fsec, 616-nm linearly polarized laser pulses. Spectrum was obtained for electrons emitted along the laser-polarization direction. $I_{\text{peak}} \approx 40$ TW/cm^2. Bound-state resonances are indicated. Circular polarization: ATI spectrum taken under the same conditions as the top spectrum, but with circular polarization. The vertical scale for the circular spectrum is magnified 40 times the vertical scale for the linear spectrum. Arrows point to the lowest-allowed resonance (six photons to the $6h$ state). The peak of the distribution occurs in a region where there are no allowed resonances. Top: Different electron energies correspond to ionization at different times and in different parts of the laser focus, as described in the text. Solid lines show the relative laser intensity distribution for these spectra. The distributions have been smeared by 10% to simulate the effect of shot-to-shot energy fluctuations in the laser.

The positions of the peaks in Fig. 1 (linear polarization), therefore, indicate the intensity when particular bound states of xenon Stark shifted into multiphoton resonance. In fact, each peak matches a p or f state, Stark shifted into six-photon resonance at an intensity where the shift approximately equals the ponderomotive potential.[3]

The lower spectrum in Fig. 1 repeats the same experiment as the top figure, but with circularly polarized light. (The vertical scales are not equal in the upper and lower spectra; measured rates are lower for circular polarization, due to a reduced cross section, and the small collection solid angle of the detector.) For circular polarization, angular momentum restricts the allowed intermediate states. Since the valence electrons in xenon are in ground-state p orbitals, the lowest-lying state that can shift into multiphoton resonance for circularly polarized light is the $5p^5(^2P_{3/2})6h$ state, 0.38 eV below the ionization limit. This level must shift by 0.31 eV in order to come into six-photon resonance. Assuming the total-energy shift of the $6h$ level is approximately equal to the ponderomotive potential, the resonance would occur at an intensity of 8.4 TW/cm^2, producing ATI electrons of 1.63, 3.64, 5.65, . . . eV. Arrows point to these energies in the spectrum in Fig. 1; however, there is no sign of any resonance. Instead, the spectrum is continuous for each ATI peak. The essential difference between the linear and circular spectra is that *for circular polarization, electron production is maximum at intensities where there are no allowed intermediate resonances.* We take this as a strong indication that the process observed is direct multiphoton ionization into the continuum.

The electron distribution is the product of the multiphoton ionization rate versus intensity, and the distribution of different intensities in the laser focus.[7] At low intensities, according to Eq. (1) with $\hbar\omega = 2.01$ eV and $E_0 = 12.13$ eV, photoelectrons may appear at energies slightly below 2, 4, 6, . . . eV. From the spectra in Fig. 1, the ATI rate is evidently too small to cause ionization at these intensities. In more intense parts of the focus, where U_P is larger, the energies of photoelectrons decrease as shown in Eq. (1), and Fig. 1 shows the appearance of ATI electrons at around 10 TW/cm^2. However, the distribution of intensities in the focused laser pulse drops sharply for higher intensities, and as a result, the ionization yield does not continue to increase, but falls to zero at an energy corresponding to electrons ionized at the peak intensity of the laser pulse.

Figure 2 shows ATI spectra for circular polarization in xenon at several peak laser intensities. The lower three spectra have ATI maxima that shift linearly with intensity; the top two spectra show peaks merging, as shifts exceed the fundamental ATI separation of $\hbar\omega$. At these highest intensities, additional effects clearly influence the spectrum. For example, ground-state atoms must deplete during the pulse, producing distortion in high-intensity (low-electron energy) parts of each ATI peak. In addition, multiple ionization becomes important at higher laser intensities, producing electrons that have a different relationship between energy and intensity at the point of ionization. We confine our detailed analysis to the lower

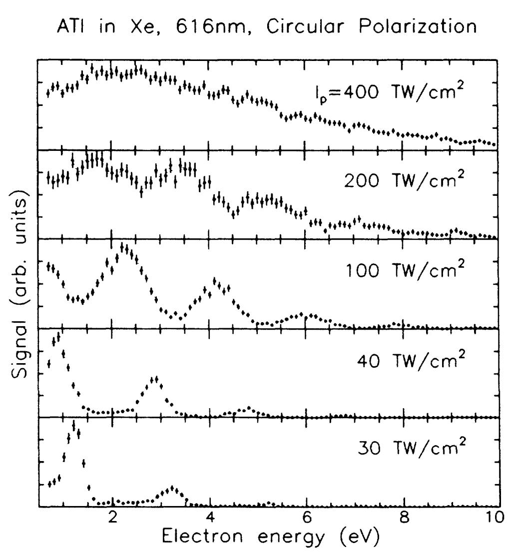

FIG. 2. Circular-polarization ATI spectra for five different peak laser intensities, for xenon ionized by 616-nm 140-fsec laser pulses.

three spectra, where depletion and multiple ionization are unimportant. As an aside, however, we note that strong merging of the ATI peaks in the top two spectra of Fig. 2 suggests that a significant fraction of neutral atoms survive the rising laser pulse to intensities of 100 TW/cm^2.

Spectra such as Fig. 2 can be converted to show relative ionization probability versus laser intensity, for a known laser focal shape.[7] For subpicosecond pulses, since the emerging electron's energy tags the intensity where it was produced, *no averaging over the inhomogeneous focus is required.*

The temporal distribution for the amplified colliding-pulse mode-locked laser is a squared hyperbolic secant function, with a full width at half maximum (determined by standard autocorrelation techniques) of 140 fsec. The spatial distribution is more complex. The final amplifier produces a multimode spatial laser distribution that resembles a nearly uniform illuminated disk. In this experiment, the central (10–20)% of the beam was apertured by a variable round iris, and focused with an achromatic lens into the xenon. The measured focal intensity distribution was that of a focused, uniformly illuminated aperture.[29] This information was combined with the temporal pulse distribution, to form the laser-intensity distribution function shown in Fig. 1. The relative rate for photoionization at a particular intensity is equal to the observed rate for electrons emitted at that intensity, divided by this function. Relative rate curves for the first three ATI peaks in xenon, taken from the lower curves in Fig. 2, are shown in Fig. 3.

These rate curves may now be compared to calculated rates for ATI in xenon by 616-nm circularly polarized light. As an example, the curves in Fig. 3 are calculations using the KFR theory, proposed by Reiss and Faisal, as an extension of a theory by Keldysh.[17–19] This comparison is particularly apt, since Reiss showed that data from a circularly polarized ATI experiment using 1064-nm-long (100 psec) laser pulses could be fitted by a similar calculation.[11,14] That comparison, however, could not use the detailed intensity information available in these subpicosecond experiments. Instead, previous KFR calculations in xenon[14] and helium[13] have relied on simulating experimental data by fitting the laser intensity in the calculations to model laser-beam parameters.

The KFR theory treats ionization as a scattering process between the atomic ground state and the laser-light-dressed continuum. This approximation neglects intermediate states, and also neglects the effects of the ion on the continuum. The clear disagreement between KFR predictions and rates derived from this experiment, shown in Fig. 3, may be partly due to the Coulomb force on the outgoing electron. In circularly polarized ATI, the elec-

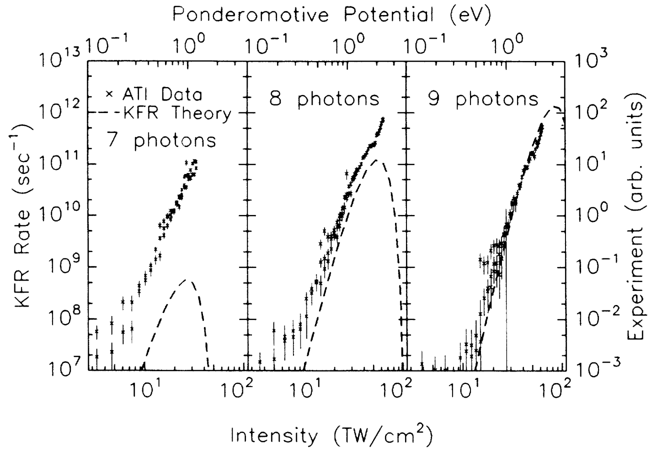

FIG. 3. Relative photoionization rates vs intensity. Experimental points come from dividing the signal at each energy by the relative focal volume × time at the corresponding intensity (e.g., the solid curve in Fig. 1). Calculated curves: Predictions of the KFR model for ATI in xenon by 616-nm laser pulses (Refs. 14 and 19).

tron carries away most of the angular momentum associated with the absorbed photons. The resulting centrifugal repulsion effectively reduces the ATI rate for low-energy electrons.[11] Although the KFR calculation includes the full effect of the centrifugal barrier suppression, the neglected Coulomb potential of the ion partly counteracts this effect, raising the relative rates for seven- and eight-photon ATI in xenon.

In summary, we have observed ATI induced by intense subpicosecond 616-nm circularly polarized laser pulses. In contrast to experiments with linearly polarized light, we find that ATI with circular polarization in xenon appears to be a nonresonant process in which the ground state is directly coupled to the continuum via seven or more photons. For 140-fsec laser pulses, depletion of the initial state does not occur even for intensities exceeding 100 TW/cm^2. Finally, we show that the data may be reduced in a model-independent way to yield experimental rates versus laser intensity, due to the close relationship between the electron energies and the laser intensity in subpicosecond ATI experiments. This new technique allows direct comparison of data to calculations, thus providing critical tests of theories of above-threshold ionization.

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