Phase Dependence of Intense Field Ionization: A Study Using Two Colors

D. W. Schumacher,[1] F. Weihe,[1] H. G. Muller,[2] and P. H. Bucksbaum[1]
[1] *Physics Department, Randall Laboratory, University of Michigan, Ann Arbor, Michigan 48109-1120*
[2] *FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
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We present the first measurement of the ionization rate from above-threshold ionization (ATI) in an optical field whose phase dependence is known and controllable. Our method uses a harmonic two-color light field with a calibrated phase difference between the 1ω and 2ω fields. Our measurements confirm many of the predictions of recent semiclassical theories, but show an important discrepancy: An asymmetry in the ATI rates relative to the sign of the two-color phase difference supports recently suggested modifications to the models to include electron-core rescattering during ionization.

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Above-threshold ionization (ATI) can occur in two regimes: a "multiphoton" regime, and "tunneling" [1]. The multiphoton picture holds for relatively low intensities and short wavelengths. Here the ionization per optical cycle is small so the process may be described by time-averaged quantities such as the energy levels of the atom and the photon energy. In the tunneling regime of high intensities and long wavelengths, the ionization rate per optical cycle is significant, so one may consider the modulation of the instantaneous ionization rate as the laser electric field distorts the atomic electrostatic potential over the course of each cycle of the light. Under these conditions an atom is far removed from a "golden rule" ionization rate; rather, ionization is phase dependent, with the highest rates occurring when the field is largest and the barrier most depressed. We describe an experiment that takes place in an *intermediate* regime where multiphoton and tunneling characters are both present. We find that experiments employing two commensurate laser frequencies—a fundamental and its harmonic—can interrogate the phase dependence of ionization in this intermediate regime. There is a third regime, called "over-the-barrier" field ionization, where the ground state is significantly depleted in a single optical cycle. This will not be considered here.

There is no definite boundary between multiphoton and tunneling ionization [2], but the Keldysh parameter $\gamma = \sqrt{2\omega^2 I_p/I}$ is commonly invoked [1] as a dimensionless figure of merit. Here ω and I are the field frequency and intensity, and I_p is the ionization potential. The Keldysh parameter can be thought of as the ratio of the time it takes an electron to tunnel through the atom-laser potential barrier and one-half of an optical period. When $\gamma < 1$, ionization is expected to be dominated by tunneling, and when $\gamma < 1$, by multiphoton ionization. Our experiments take place in the regime $\gamma = 1$–2. Nonetheless, we shall show that most features of phase-dependent tunneling ionization are present.

We use a two-color laser field $\xi(t)$ consisting of a fundamental frequency ω and its second harmonic:

$$\xi(t) = F_1(t)\cos\omega t + F_2(t)\cos(2\omega t + \phi). \qquad (1)$$

Here $F_{1,2}$ are the envelope functions for the fundamental and second harmonic fields, ω the fundamental frequency, and ϕ the relative phase. Such a field has the useful property that although the intensity and ponderomotive potential are ϕ independent, the peak electric field and its evolution depend on ϕ. We were motivated to use this field as a probe by a recent paper by Schafer and Kulander [3] which points out that these characteristics make phase-dependent two-color field excitation an excellent tool to study these regimes *if the absolute phase between the two colors can be determined.* We report the first experiment to our knowledge to do this. In addition, we analyze our results in the light of recent work by Corkum [4,5], who has introduced relatively simple methods to model the results of experiments performed in the tunneling regime.

Two-color laser fields have already been used to probe and control the dynamics of light-induced processes [6–8]. There have been numerous theoretical studies examining these effects [3,9,10], as well as the predictions that two-color fields will allow the coherent control of a system [11,12].

Our experiment used 100 ps pulses of 1.06 μm light from a Nd:YAG laser system. This light passed through a type-II second harmonic generation (SHG) KD*P crystal, followed by a dielectric polarizer. The orientation of the crystal and polarizer were arranged so that the two laser fields were polarized parallel, and the intensities would be equal at the waist of a focus. The relative phase ϕ was controlled using the dispersion in a 1 cm thick fused silica plate. We could select any arbitrary ϕ by tilting the angle of incidence near Brewster's angle, thereby changing the relative optical path length between the two colors.

The light entered a vacuum chamber through a fused quartz window. The chamber (base pressure 4×10^{-9} Torr) was backfilled with up to 10^{-5} Torr of krypton gas. The light was focused into a field-free region with a 150 mm focal length spherical mirror. The polarization vector pointed toward a microchannel plate time-of-flight detector in the field free region $\approx$38 cm from

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the focus. The detector subtended a small solid angle ($\approx 2 \times 10^{-4}$ sr) so that we only detected electrons leaving the region in one direction.

We measured the phase ϕ between the two fields in the vacuum chamber by inserting a type-I phase-matched KD*P crystal $\approx$15 cm from the focus, oriented so that the polarization of the incident light had both ordinary and extraordinary components. Sum- and difference-frequency radiation at ω and 2ω interfered with the incident light depending on the following: the phase between the two colors at the interaction region; the relative phase shift of $\pi/2$ that developed between them as they moved from the focus into the far field where the second crystal was located (assuming Gaussian spatial profiles); and the $\pi/2$ phase shift between the 1.06 μm light and the newly generated second harmonic in the second crystal. Thus, we related the amount of 0.532 μm light leaving the chamber to the optical phase difference between the ω and 2ω components in the interaction [13]. A remaining sign ambiguity in the phase was removed by measuring optical rectification of the field in the same crystal.

Figure 1 shows a typical krypton spectrum using the combined field with $\phi = 0$, and $F_1 = F_2 = 1.2$ V/Å. (Total intensity of 4×10^{13} W/cm^2.) The 100 ps pulse width places this experiment in the "long-pulse regime" of ponderomotive scattering, where the photoelectron spectrum reflects the *unshifted* ionization potential of the atom [14]. The large and small peaks correspond to ATI where the ion is left in the $^2P_{3/2}$ and $^2P_{1/2}$ ground states, respectively. The multiphoton character of the experiment is clearly evident in this spectrum. The tunneling character, on the other hand, becomes dramatically evident when we examine the phase dependence. We did this by recording the number of electrons in each of the large ($^2P_{3/2}$) peaks as a function of ϕ over 5 periods (10π in ϕ), and then folded the results into a single period with the phase varying between $-\pi$ and π.

Figure 2 shows the relative ϕ dependence for four different ATI peaks (the third through sixth $^2P_{3/2}$ peaks) together with the results of a model calculation, described later. The lower energy electrons clearly peak at $\phi = 0$ and $\phi = \pi$, where ω and 2ω constructively interfere to produce the largest field. The higher energy electrons peak at other phases. Evidently, the phases best suited for producing the largest rates do not produce the highest energy electrons.

The higher energy electrons in Fig. 2 show a strong forward (backward) [towards (away) from the detector] asymmetry. Although the detector only measured forward rates, it is still possible to determine how many electrons went backward. The electric field (1) is symmetric with respect to $\phi \rightarrow \phi + \pi$, $z \rightarrow -z$, and $\omega t \rightarrow \omega t + \pi$. Therefore, the backward rate at phase ϕ is the forward rate at phase $\phi + \pi$. There is also an interesting bimodal structure with lobes π rad in phase apart for the low energy electrons, but moving considerably closer in phase for the higher energy electrons.

Figure 3 shows data for a total intensity of 8×10^{13} W/cm^2. The behavior is now quite different. Although higher energy electrons are still preferentially produced away from $\phi = 0$, the difference in phase from the lower energy electrons is much smaller. The forward/backward asymmetry is also less dramatic. The bimodal structure, however, is still present.

These effects were predicted by Schafer and Kulander [3], who numerically integrated Schrodinger's equation for hydrogen under similar conditions. We have also measured the phase dependence of ATI in xenon and seen similar results, which we will report in a future publication. These effects are general *for any* experiment performed in the tunneling regime because the most important parameters determining the behavior of the system are simply the ionization potential, the electric

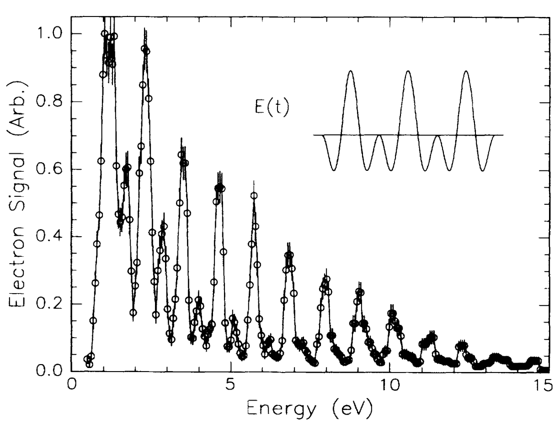

FIG. 1. A typical krypton photoelectron spectrum for the combined field with $\phi = 0$. The electric field for this case is shown in the inset.

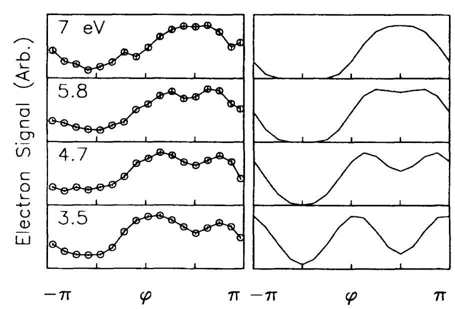

FIG. 2. Phase dependence of the third through sixth ATI peaks (lowest to highest graphs with the ATI peak energy labeled) for a total intensity of 4×10^{13} W/cm^2. The absolute intensity is only known to within a factor of 1.5. Also shown to the right are the results of a model calculation described in the text which assumed a total intensity of 6×10^{13} W/cm^2.

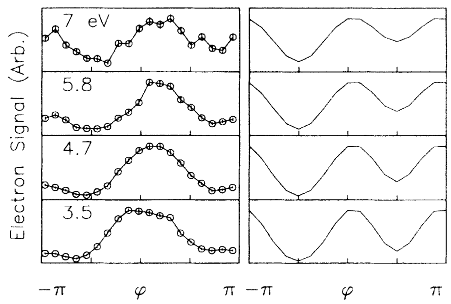

FIG. 3. Phase dependence of the third through sixth ATI peaks for a total intensity of 8×10^{13} W/cm^2. The absolute intensity is only known to within a factor of 1.5, but the relative intensity between Figs. 2 and 3 is known to 10%. The total intensity used for the model results was 12×10^{13} W/cm^2.

field, and the frequency. Any underlying structure is less important.

We now proceed to analyze these results after Corkum [5] and show that most of them can be described in a simple and intuitive fashion. The interaction is divided into two steps. In the first step, we employ a standard tunneling formula relating the ionization rate to the instantaneous electric field [4]:

$$\Gamma(t) = 4I_P^{5/2}\frac{1}{\xi(t)}\exp\left[-\frac{2}{3}I_P^{3/2}\frac{1}{\xi(t)}\right] \qquad (2)$$

with all quantities in atomic units. Our results are not very sensitive to the form of Eq. (3), as long as Γ is a strongly increasing function of ξ. In the second step, the kinetic energy E_k of the electron ionized in step 1 is evaluated by tracing its classical trajectory in the time-varying laser field. Since we are in the long pulse regime, the time-averaged energy of this drifting electron is the kinetic energy observed in our apparatus. The direction of the emitted electron is just given by the direction of its drift momentum in the field—either towards the detector or away from it, for linear polarization.

The kinetic energy depends on the overall phase of the two-color field at the instant of ionization t_0:

$$E_k = U_{p1} + 2U_{p1}\sin^2\omega t_0 + U_{p2} + 2U_{p2}\sin^2(2\omega t_0 + \phi) + 4\sqrt{U_{p1}U_{p2}}\sin\omega t_0\sin(2\omega t_0 + \phi), \qquad (3)$$

where U_i is the ponderomotive potential or electron "wiggle energy" for field i given by

$$U_{pi} = e^2F_i^2/4m\omega_i^2. \qquad (4)$$

Note that if $U_{p2} = 0$ we recover the well-known result for a monochromatic field that the electron energy must range between 1 and 3 times its ponderomotive potential.

Our calculation assumes a 100 ps, 1.06 μm pulse coincident with a 71 ps (as would be expected after doubling) 0.532 μm pulse, with the same peak intensity and with some selected phase difference ϕ between them. The results have not been averaged over the different intensity profiles in the spatially inhomogeneous laser focus; although the results are sensitive to intensity, the general trends that match the experiment are clearly present without this additional complication.

The results are displayed in the right sides of Figs. 2 and 3, for model total intensities of 6×10^{13} and 12×10^{13} W/cm^2, respectively. All the phase-dependent trends mentioned above show good agreement with the model. The primary failing lies in the relative strengths of the various ATI peaks: As previously mentioned, the experimental spectra in this mixed regime have higher energy electrons than predicted by tunneling. The normalized, phase-dependent ionization rate shows the following trends: (1) In the low intensity data, more high energy electrons are produced at phases away from $\phi = 0$; (2) there is a strong forward/backward asymmetry in the emitted electrons and a bimodal structure in the ϕ dependence. This model also provides an intuitive picture for why this should be so.

The calculated phase-dependent spectrum is dominated by electrons emitted near the peak field. For the case $\phi = 0$, the electric field peaks when $\omega t_0 = 0$ (see inset in Fig. 1), an initial phase where the electron gains minimum drift energy in the field [Eq. (3)]. As ϕ increases from 0 the resulting electric field is less symmetric with respect to the peak field. Therefore electrons produced at the peak have larger subsequent drift velocity, and *contribute to the high energy part of the spectrum.* The highest energy electrons the peak field can produce occur for $\phi = \pm\pi/2$, and this is clearly seen at low intensities in the data and the model. Finally, the increasing lack of symmetry as ϕ increases from zero favors electrons drifting more in one direction than another—hence the forward/backward asymmetry. The multiphoton regime can also show phase dependence, of course, but is not tied to the peak field in this manner.

The agreement between the model and experiment for the high intensity data is not as good. In this case, ionization can occur over a significantly larger range of ωt_0 and so produce higher energy electrons, resulting in the broader distributions in phase and the filling in of the distribution at $\phi = \pi/2$ in the model. The experiment, however, clearly demonstrates a stronger tendency for the high energy electrons to *peak* at phases away from $\phi = 0$ for high ATI peaks.

The two-step model predicts that the *total* rates for each ATI channel are symmetric about $\phi = 0$, independent of intensity. We can measure forward and backward rates combined, which should also be symmetric under this model. For the low intensity data, in fact, they are. For the high intensity data, however, there is a clear asymmetry in the total rates for the high energy electrons as shown in Fig. 4, which plots the phase (closest to zero) for which each ATI peak was largest. The two-step model predicts this phase will be zero for

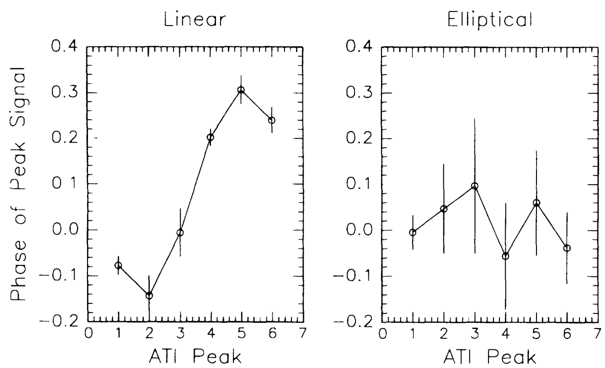

FIG. 4. Phase (shown as a multiple of π) which yielded the largest total (summed forward and backward) rates for linear and slightly elliptically polarized light as a function of ATI order. The strong dependence on ellipticity suggests that rescattering may be responsible for the fore-aft asymmetry.

all peaks. Clearly, the two-step model is able to describe the essential physics involved at low intensities and/or for low energy electrons; however, the high energy electrons produced at high intensities involve some new process not present in the model. Corkum [5] has argued that core scattering after the ionization step can be important, especially in understanding high harmonic generation and second ionization. We tested to see if scattering was important in this experiment by introducing a small amount of retardation to the two optical fields. The retardation was small enough that the electron spectrum was not significantly altered, but large enough that it would be unlikely for the electron to see the core after ionization. The symmetry has clearly been restored. (We were unable to calibrate the relative phase for the data collected with elliptical polarization. The data were plotted based on the reasonable assumption that the low energy electrons peak at phase $\phi = 0$ and π.)

In conclusion, we have measured atomic ionization rates in an intense two-color field which has an absolutely calibrated phase difference between the two colors. This permits us to infer the phase dependence for the ionization rate. At low intensities, a simple semiclassical model seems to embody most of the important physics. At high intensities, however, the model becomes insufficient. Studies using elliptical polarization suggest electron-core scattering may be significant.

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