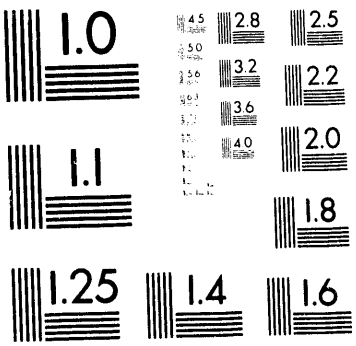
1.0
4.5
5.0
5.6
2.8
3.2
3.6
4.0
2.5
2.2
2.0
1.1
1.8
1.25
1.4
1.6

1 of 1

CONF-930612--4

UCRL- JC-114857
PREPRINT

# LIMITS ON HIGH-ORDER HARMONIC GENERATION FROM SINGLE-ATOM CALCULATIONS

K. C. KULANDER and K. J. SCHAFER
Physical Sciences
V-Division
Lawrence Livermore National Laboratory
Livermore, CA

This paper was prepared for submittal to:

**"ICOMP VI: International Conference on Multiphoton Processes"**

Quebec City, Quebec, Ontario, Canada
June 26-30, 1993

August 18, 1993

SEP 03 1993
OSTI

Lawrence Livermore National Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

# LIMITS ON HIGH-ORDER HARMONIC GENERATION FROM SINGLE-ATOM CALCULATIONS

K. C. KULANDER and K. J. SCHAFER
*Physics Department, Lawrence Livermore National Laboratory*
*Livermore, CA 94551 USA*

## 1. Introduction

In recent years it has been observed that when a linearly polarized, short pulse, high intensity laser excites a dense atomic medium, a surprisingly copius amount of very high-order harmonic radiation is generated. In the direction of propagation of the incident beam, strong coherent emission at odd multiples of the driving frequency is produced. For example, odd harmonics up to the 109th order of a 140 fs Ti-sapphire (806nm) laser[1] and 133rd harmonic of a 1053 nm, 1 ps Nd-YLF laser[2] have been obtained in neon. Even harmonics are forbidden by symmetry. In this regime the distribution of harmonic emission strengths exhibits a characteristic pattern. The spectrum is found to decrease quickly for the first few orders, then flatten out with a broad plateau up to a very high order beyond which there is an abrupt cutoff. We have shown that the time-dependent wave function for the atomic electrons in the pulsed field contains the information that reproduces this highly nonlinear, non-perturbative behavior.[3] The Fourier transform of the dipole induced in the electronic charge distribution gives the spectrum of emitted radiation. The cut-off of the single-atom plateau has been found[4] to be given by $\approx I_p + 3U_p$ where $I_p$ is the field free ionization potential of the atom and $U_p = I/4\omega^2$ is the ponderomotive energy of a free electron in a laser of frequency, $\omega$, and intensity $I$. The single-atom spectral distribution very closely resembles that emitted from the macroscopic medium.

In the quantum mechanical calculations of electron and photon emission from atoms in strong laser fields we have employed a single-active-electron (SAE) model.[5] We determine the effect of the time varying electric field of the laser on each of the valence electrons separately. The active electron in each calculation moves in the time-independent mean field of the remaining, unexcited electrons and the nucleus. This approach works well for the rare gas atoms, at least partially because the neglected double or higher excitations involve states well above the ionization threshold. The photoelectron and photon emission spectra calculated using this technique agree quantitatively with observed emission rates.[6,7]

In this paper we will present a simple semiclassical model for high intensity ionization which reproduces the observed harmonic emission spectra obtained in this regime and which provides considerable insight into the dynamics of this process. The basic model[8] has been used in the past to predict electron energy distributions in the tunneling regime and we will use it here for harmonics.[7,9]

## 2. Harmonic emission following tunneling ionization

As the laser intensities increase, the route to the continuum for an electron in an atom changes character. When the laser is strong enough its oscillating electric field will,

when combined with the long-range Coulomb attraction, form a barrier through which the electron can tunnel. This barrier suppression is illustrated for the case of atomic hydrogen in Fig. 1. As the barrier is suppressed (twice) during each optical cycle, electrons are born into the continuum at the turning point on the outer side of the barrier ($z_{init}$ in Fig. 1.) The subsequent evolution of the emitted electrons is then dominated by the oscillating laser field. Thus we can invoke a simple two step semiclassical model for ionization in this regime. An initiation step occurs at each instant during the cycle when the electron can tunnel through the instantaneous barrier with a probability given by the appropriate dc tunneling rate. We can use the dc rate because the laser field changes very slowly on the time scale for the electronic response. Second the wave packet of free electrons created during each half cycle evolves mainly in response to the dominant laser electric field, oscillating back and forth along the direction of polarization. Because of the high density of states in the continuum, the dynamics of this wave packet can be described using classical mechanics. This tunneling ionization picture becomes valid when the intensity is large enough that $2U_p > I_p$ which holds in the experiments mentioned above.

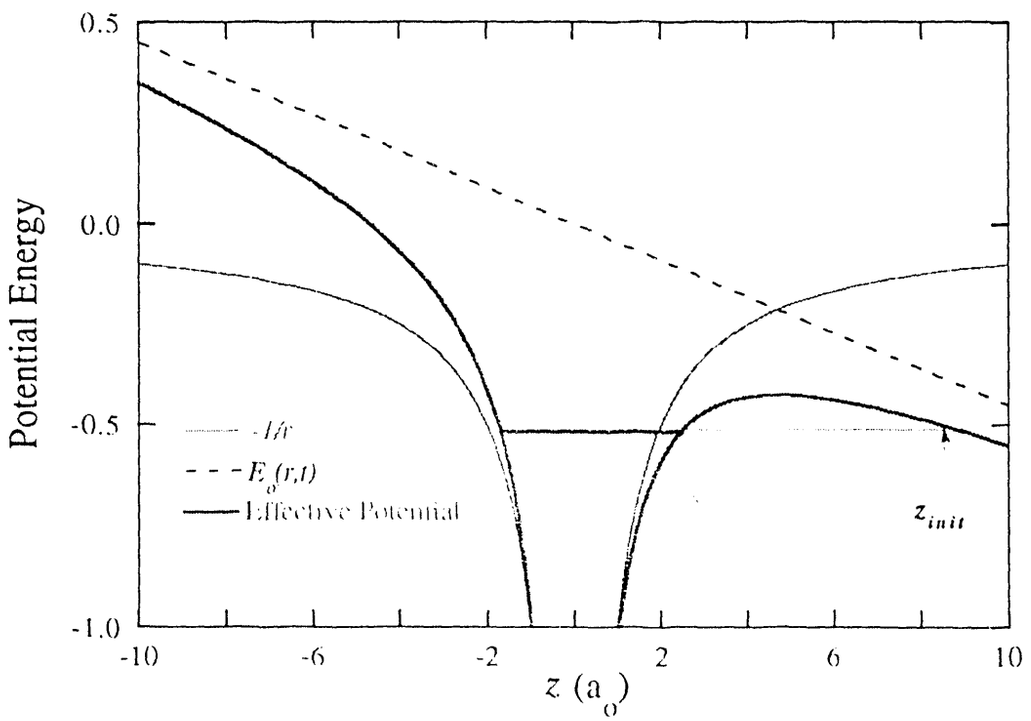


Fig. 1 Instantaneous effective potential for hydrogen in a field polarized along the $z$-axis. The ground state wave function (indicated by bold line within well) ionizes by tunnelling through the barrier. Electrons are released at $z_{init}$.

Tunnelling is most probable when the field is at its maximum ($\omega t=(n+1/2)\pi$ in Eq. (1)) which occurs twice during each optical cycle. Because the amplitude of the field varies slowly with the phase, there will be a finite interval around these maxima during which electrons will be freed. In Fig. 2 we show the wave packet created by a half cycle of a

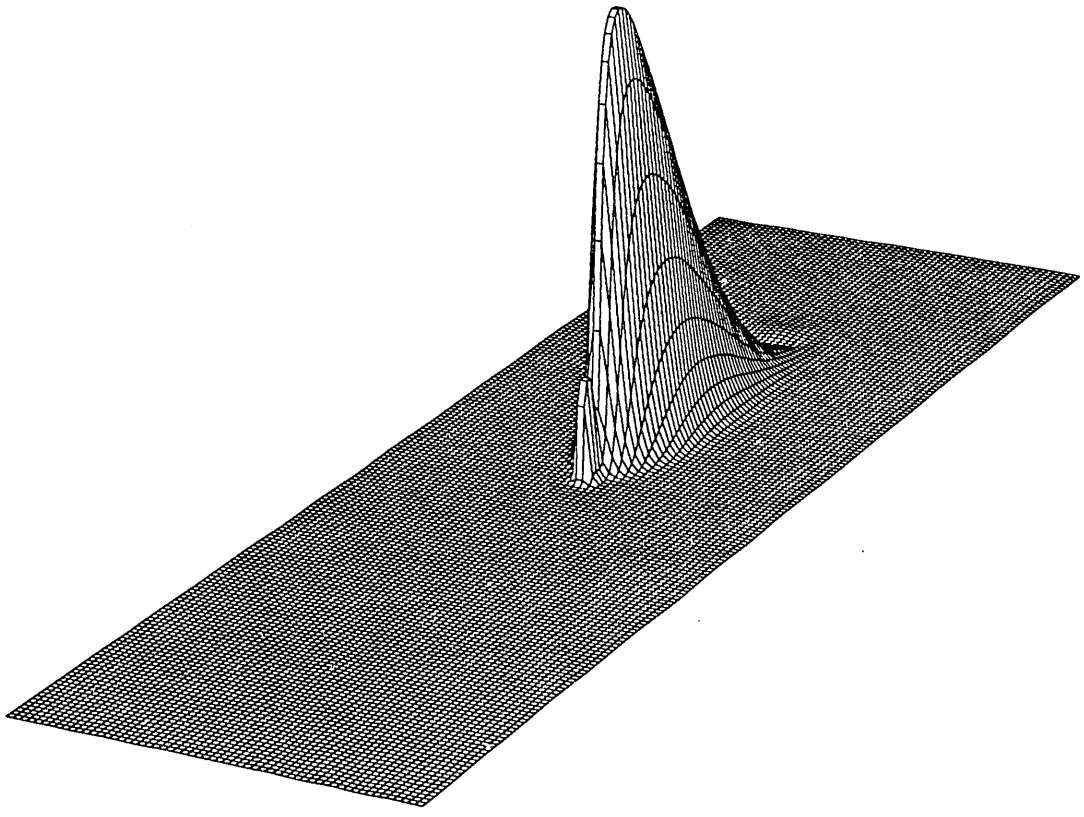

Fig. 2 The wave packet created by tunnel ionization from Ne in one-half cycle of a $6 \times 10^{14}$ W/cm$^2$, 806 nm laser. The grid extends to $\pm 200 a_0$ in z and $\pm 30$ in the radial direction and the nucleus is at the center.

Ti:sapphire (806 nm) laser at an intensity of $6 \times 10^{14}$ W/cm$^2$. We obtain this wave packet by integrating the time-dependent Schrödinger equation for the most weakly bound valence electron through one half of a cycle at a fixed laser intensity using the interaction

$$V(t) = - ez\varepsilon_0 \sin(\omega t). \tag{1}$$

At the end of this interval, the electric field of the laser is zero so we can project the ground state component out of the wave function leaving the excited wave packet shown. Since the field, during this half cycle, has been driving the electron away from the atom, the wave packet has moved well away from the ionic core along the direction of polarization. During the next half cycle the field will turn around and drive a substantial fraction of this wave packet back across the nucleus. When an electron returns and rescatters from the core, it can undergo a transition back to the ground state, emitting a high energy photon. The energy of the resulting photon corresponds to the energy the electron has when it returns. This return energy, which depends strongly on the particular phase during the optical cycle at which the electron escaped through the barrier, can be well above the ionization limit.

We can approximate the evolution of this wave packet by following a series of classical trajectories originating at slightly different times during the cycle with initial

conditions defined by the shape of the instantaneous barrier at the time of the transition to the continuum and weighted by the instantaneous tunneling rate. In Fig. 3 we show a phase space plot of a representative trajectory which was released with a phase which results in multiple returns to the nucleus. The initial conditions of the trajectory are defined by zero velocity and $z(t=0) = z_{init}$. The figure shows that the electron mostly moves as a free electron in the laser field, occasionally experiencing abrupt collisions with the nucleus. Only when the electron is close to the nucleus can it release its energy by emitting a photon. The energies at the return points are identified as possible harmonic photons.

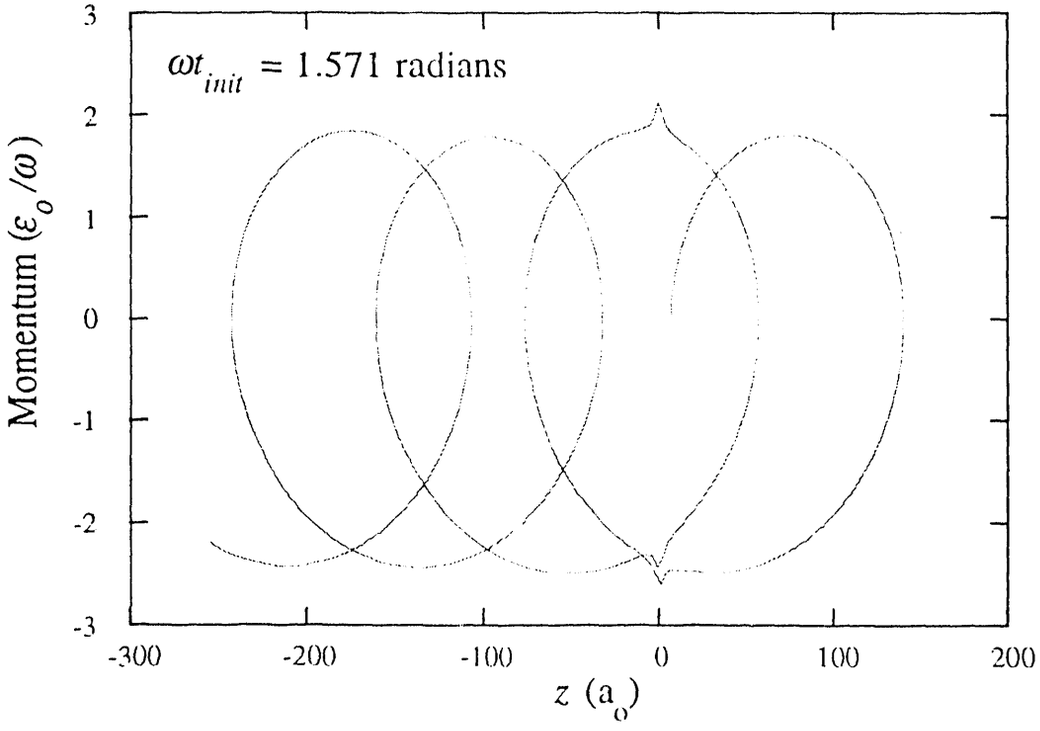


Fig. 3 Trajectory of an electron which tunnelled to freedom just before the maximum in the oscillating electric field. ($I = 10^{14}$ W/cm$^2$ and $\lambda = 1.8$ μm.)

We repeat the above procedure for different phases of the optical cycle and record the return energies of the electrons. We show in Fig. 4 the distribution of return energies obtained for 180 trajectories equally spaced in time over the first half of the cycle. The results are identical to those obtained during the second half by symmetry. The trajectories are followed for four cycles of the field. We plot the return energy in excess of the field-free binding energy in units of $U_p$. We have also plotted the tunnelling rate for hydrogen in this field as a function of the phase. A small fraction of the emitted electrons (about 25 percent in this case) initiated in the interval just after the phase $\pi/4$ drift away from the nucleus without a collision. Phases earlier than $\pi/4$ produce trajectories which do not recross the nucleus, but the tunnelling barrier is too broad for any significant release to occur. The rest of the trajectories return at least once. For initial conditions near the

maximum of the field the electron undergoes multiple collisions with the nucleus. This is because these phases induce small initial drift velocities. We note it is the *drift* velocity (a cycle averaged quantity) which defines the electron energy measured in an ATI experiment. By rescattering electrons can obtain large drift energies, well above the harmonic cutoff.[7]

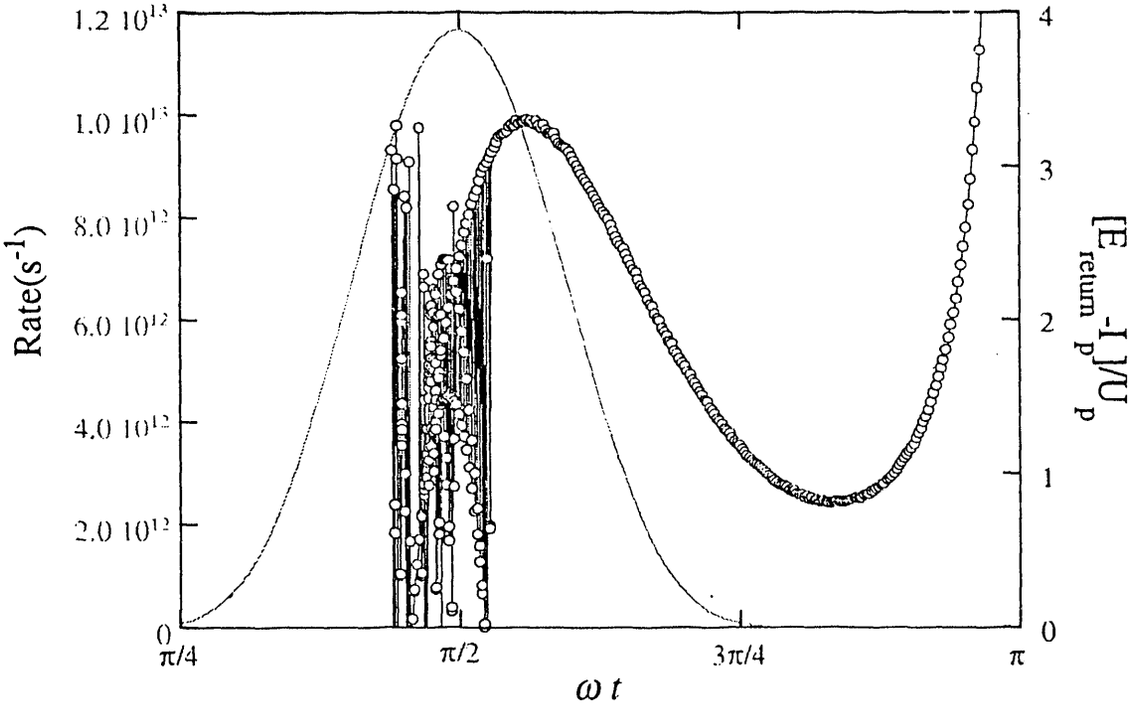


Fig. 4 Hydrogen tunnelling rate (solid line-left axis) and energies of returning trajectories at the nucleus (symbols-right axis) as a functions of the phase of the oscillating field ($I = 10^{14}$ W/cm$^2$ and $\lambda = 1.8$ μm.)

In Fig. 5 we show a histogram of return energies weighted by the appropriate initial tunnelling rate. The result is a broad, flat distribution out to ~ $3.2U_p$ followed by an abrupt cutoff. Higher return energies occur for phases near $\pi$, but their contributions are negligible due to low tunneling rates through thick barriers to large initial displacements. For probable initial conditions the return energies do not exceed $I_p + 3.2U_p$. Electrons returning to the nucleus, will have a probability for emitting a photon which is proportional to the oscillator strength of transition back to the ground state at the return energy. *Therefore the tunneling wave packet provides the source of the plateau in the harmonic spectrum and explains the origin of the observed sharp cutoff.* The strong lower order harmonics come from a different source and can be shown to be due to short range charge distortions.[10]

## 3. Acknowledgment

This work has been carried out under the auspices of the U. S. Department of Energy at the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

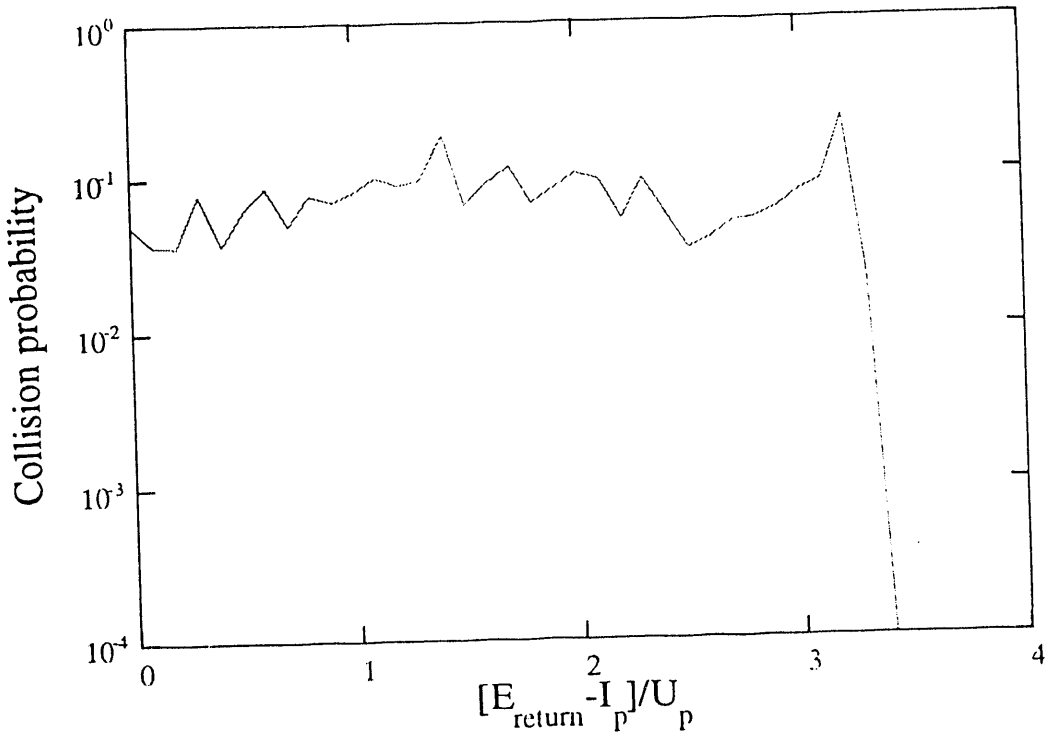


Fig. 5 Energy histogram for electron trajectories returning to the nucleus for hydrogenic, soft-Coulomb potential. ($I = 10^{14}$ W/cm$^2$ and $\lambda = 1.8$ μm.)

## 4. References


1. J. Macklin, J. D. Kmetec and C. L. Gordon III, *Phys. Rev. Lett.* **70** (1993) 766.
2. A. L'Huillier and P. Balcou, *Phys. Rev. Lett.* **70** (1989) 524.
3. K. C. Kulander and B. W. Shore, *Phys. Rev. Lett.* **62** (1993) 774.
4. J. L. Krause, K. J. Schafer and K. C. Kulander, *Phys. Rev. Lett.* **68** (1992) 3535.
5. K. C. Kulander, K. J. Schafer and J. L. Krause, *Intl. J. Quant. Chem.: Quant. Chem. Symp.* **25** (1991) 415.
6. A. L'Huillier, P. Balcou, C. Candel, K. J. Schafer and K. C. Kulander, *Phys. Rev. A* **46** (1992) 2778.
7. K. J. Schafer, B. Yang, L. F. DiMauro and K. C. Kulander, *Phys. Rev. Lett.* **70** (1993) 1599.
8. H. B. van Linden van den Heuvell and H. G. Muller, in *Multiphoton Processes*, eds. S. J. Smith and P. L. Knight (Cambridge University Press, Cambridge, 1988); T. F. Gallagher, *Phys. Rev. Lett.* **61** (1988) 2304; P. B. Corkum, N. H. Burnett and F. Brunel, *Phys. Rev. Lett.* **62** (1989) 1259.
9. Similar results have been recently reported by P. B. Corkum, *Phys. Rev. Lett.* (submitted) and A. l'Huillier et al., *Phys. Rev. A.* (submitted).
10. K. C. Kulander and K. J. Schafer, in *Super-Intense Laser-Atom Physics*, eds. B. Piraux, A. l'Huillier and K. Rzazewski (Plenum, New York, 1993).

DATE
FILMED
11/30/93

END