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Ellipticity and polarization effects in harmonic generation in ionizing neon

N. H. Burnett, C. Kan,* and P. B. Corkum
National Research Council of Canada, Ottawa, Canada K1A 0R6
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We report observations of the ellipticity dependence of harmonic generation in a thin layer of neon gas at an intensity of 10^{15} W/cm^2 and a wavelength of 7540 Å. We observe a markedly different ellipticity dependence and polarization for harmonic energies below and above the ionization potential of neon (21.56 eV). The ellipticity dependence of the higher harmonics can be understood in terms of free-bound transitions of ionizing electrons within the first cycle of the fundamental subsequent to tunnel ionization, while other mechanisms must be invoked to explain the characteristics of the lower harmonics.

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There is much current interest in the generation of harmonic radiation when atomic gases are irradiated at intensities near threshold for multiphoton ionization [1,2]. The single-atom dipole moment responsible for such emission has been investigated by Kulander and Shore [3] through numerical solutions of the three-dimensional time-dependent Schrödinger equation and the propagation equations with these source terms have been numerically integrated [4] to yield good agreement with experimental observations of high harmonic emission. A quantum-analytical model for the single-atom nonlinear polarizability in ionizing systems consisting of a single bound state has been developed by Becker, Long, and McIver [5]. A conceptual understanding of the origin of harmonics with energies much in excess of the ionization potential is facilitated by a semi-classical model based on ionization, acceleration in the continuum, and recombination (i.e., a spontaneous free-bound transition) which has been proposed by Corkum [6] and by Schafer *et al.* [7]. A quantum treatment, which follows this phenomenological picture, has been developed by Lewenstein *et al.* [8]. A classical model based on the high-frequency time dependence of the tunnel ionization current has been proposed for the lowest harmonics by Brunel [9].

Clarification of the basis in atomic physics for harmonic generation can be facilitated by studies of the ellipticity dependence of harmonic yield. To a first approximation, propagation and phase matching in the atomic medium are not affected by small variations in ellipticity of the fundamental, while the individual atom nonlinear response can vary greatly. Ellipticity can thus be regarded as a tool, allowing one to isolate the single-atom response to the fundamental field. Previously reported studies [10,11] have shown that harmonics with energies much in excess of the ionization potential (plateau harmonics) generated with 800-nm lasers in neon have a near universal ellipticity dependence that can be accounted for quite well by the semiclassical picture. These harmonics are very sensitive to ellipticity, with a value of $\epsilon=E_y/E_x\sim 0.1$ sufficient to significantly quench harmonic yield. The variation of yield with ϵ was observed to be Gaussian to a good approximation. It has been demonstrated that the ellipticity dependence of the plateau harmonics is the same as that of simultaneous two-electron ionization in neon, suggesting a common phenomenological basis for these effects [11].

In the present work, we report studies of the ellipticity dependence of harmonic yield from neon at energies above and below the ionization potential. Our observations support the conclusion that the lower-energy harmonics are generated by different mechanisms than that responsible for the highest harmonics.

We use a compact all Ti:sapphire laser based on a cw mode-locked oscillator, and chirped pulse amplification in a regenerative amplifier followed by a two-pass final amplifier to produce nominal 5-mJ, 150-fs pulses at a repetition rate of 2 Hz and wavelength of 754 nm. The laser radiation is focused by an $f/30$ thin lens into a target in the form of a 3.1-mm-diam stainless-steel tube with 150-μm-thick walls, which has been squeezed down to an internal thickness of 750 μm. A 150-μm-diam hole is drilled by slightly defocusing the laser and irradiating the target for a few hundred shots. The laser is then focused to a spot 75 μm in diameter for harmonic production. The target tube is transiently filled with 100 Torr of Ne with a fast pulsed valve. The laser intensity averaged over its 90% energy circle is estimated to be 1×10^{15} W/cm^2 with a 5-mJ, 150-fs incident pulse. The laser output after switchout from the regenerative amplifier and recompression with a double-pass two-grating compressor is linearly polarized to better than 500:1 in intensity.

Ellipticity is introduced into the incident laser pulse by rotating a zero-order quarter waveplate placed just before the focusing lens and target chamber window. The harmonic radiation is dispersed with a 1200-g/mm gold coated Hitachi variable-space concave grating used at an incident angle of 87°. A movable slit is placed in the focal plane of the grating to isolate one harmonic. High harmonic yield is monitored on a channeltron or venetian blind type electron multiplier placed behind the slit. Zero-order (specular reflection) radiation from the grating is collected and transmitted through a MgF_2 window to monitor the third and fifth harmonic yield (by means of a Czerny-Turner 0.5-m vuv spectrograph and photomultiplier with uv converter).

The major axis of the incident polarization rotates as ellipticity is varied and assuming that harmonic emission is mainly polarized along the major axis, the angle of its polar-

*Present address: Department of Electrical Engineering, University of Alberta, Edmonton, Canada T6G 2E1.

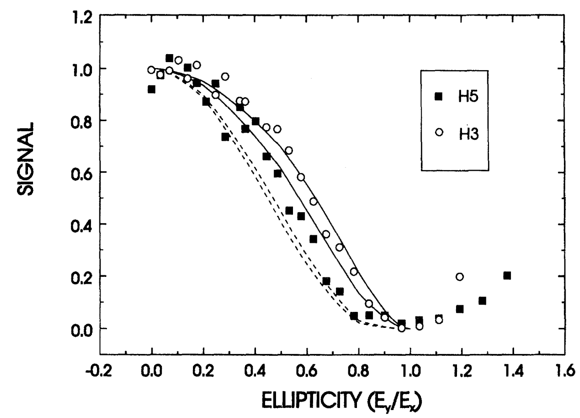

FIG. 1. Ellipticity dependence of third (3H) and fifth (5H) harmonics from neon. The solid and dashed curves represent a calculation for the third (solid) and fifth (dashed) harmonic yield based on the Brunel [9] model for intensities of 8×10^{14} W/cm^2 (lower) and 1×10^{15} W/cm^2 (upper).

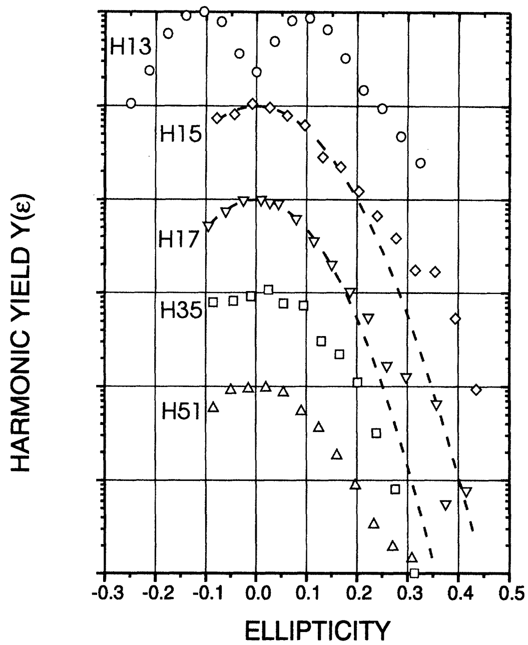

FIG. 2. Ellipticity dependence for the 13th and several of the higher harmonics. The dashed curves with the 15th and 17th harmonics represent computer generated "best-fit" Gaussians. The data for each harmonic have been normalized in an arbitrary fashion and vertically offset for display purposes.

ization with respect to the grating changes. For this reason, one needs to consider the possible effect on harmonic yield of a change in grating reflectivity as ellipticity is added. This effect is small however for the present measurements with both spectrographs described above. The grazing incidence spectrograph was measured to be 50% more sensitive to vertically polarized light (the incident fundamental is horizontally polarized) than to the horizontal polarization at a wavelength of 580 nm (the 13th harmonic). This preferential transmission decreased at smaller angles of diffraction [12] to less than 20% (still favoring vertical polarization) at wavelengths around the 21st harmonic. The near normal incidence spectrograph used to monitor the third and fifth harmonics was measured to be slightly (<20% for both the third and fifth harmonics) more sensitive to vertically polarized light than to the horizontal polarization ($\epsilon=0$). No attempt has been made to correct for any for of these effects in the results to be shown below.

Using the apparatus just described and a 150-fs, 5-mJ, 754-nm driver pulse, odd harmonics extending up to about $N=65$ are observed in neon at 100 Torr. Harmonic yield, $Y^N(\epsilon)$ was defined as the average harmonic signal observed over 15 shots, with numerical discrimination used to ensure that the incident laser energy was constant to within $\pm2\%$. The dynamic range over which it was possible to follow the yield of an individual harmonic varied with N. For most harmonics it was limited by fluorescence from the stainless-steel target tube.

Yields $Y^N(\epsilon)$ for the harmonics $N=3$ and 5 are plotted in Fig. 1 and yields for $N=13$, 15, 35, and 51 are plotted in Fig. 2. It is apparent that the yield of the lowest harmonics (third and fifth) is quite tolerant to ellipticity, while harmonics from $N=15$ to 51 (the highest measured) exhibit a nearly Gaussian ϵ dependence of approximately constant width. The 15th and 17th harmonics often showed a slight but consistent departure from Gaussian at large ellipticity, as seen in Fig. 2. Particularly striking, however, is the large difference in ellipticity dependence between $N=13$ and 15. The 13th harmonic corresponds to an energy $h\nu=21.39$ eV, just below the ionization potential of neon, 21.56 eV. The 13th harmonic yield at zero ellipticity was lower than that of the 15th by approximately an order of magnitude. $Y^{13}(\epsilon)$ has a pronounced minimum for zero ellipticity and peaks near $\epsilon=0.1$ after increasing by about a factor of 5. This increase in yield is much too large to be accounted for by the polarization sensitivity of our spectrograph, even allowing for a rotation of the polarization ellipse of the 13th harmonic with increasing ellipticity of the fundamental (see discussion below). It is interesting to note that $\epsilon=0.1$ corresponds closely to the maximum of the derivative in ϵ of the ellipticity dependence of the higher harmonics. Qualitatively similar behavior for the 13th harmonic yield was also observed in helium whose ionization potential 24.59 eV also lies between $N=13$ and 15. For argon (ionization potential 15.76 eV) both the 13th and 15th harmonics had a Gaussian ellipticity dependence characteristic of the plateau harmonics. Unfortunately it was not practical to examine the ellipticity dependence from $N=7$ to 11 with either of the spectrographs used in the present study.

The polarization of the 13th and 15th harmonics from neon was studied by inserting a polarizer consisting of three gold mirrors [13], with incident angles of 14°, 28°, and 14° between the slit and electron multiplier. This device has a transmission of about 0.15 and calculated polarization ratio of 20:1. The 13th harmonic was observed to have a significant component polarized transversely to the major polarization axis of the incident fundamental which increased rapidly with ellipticity. Yield profiles for the 13th harmonic with the auxiliary polarizer oriented parallel and perpendicular to the fundamental (at $\epsilon=0$) are shown in Fig. 3. The absolute yield in the transverse polarization was approximately three

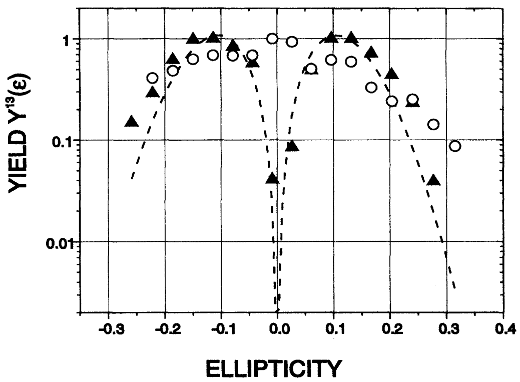

FIG. 3. Yield of the 13th harmonic with auxiliary polarizer inserted between xuv spectrograph and electron multiplier. Open circles, polarizer is oriented to transmit polarization parallel to that of fundamental; closed triangles, polarizer transmits perpendicular polarization. The two data sets have been normalized in an arbitrary manner. As discussed in the text, the absolute yield at $\epsilon=0.1$ is estimated to be a factor of 3 greater for the perpendicular polarization than for the parallel polarization. The dashed line is the square of the derivative of the best-fit Gaussian to the 15th harmonic as plotted in Fig. 2.

times greater than that in the parallel polarization at $\epsilon=0.1$. In contrast to the 13th harmonic, the 15th harmonic was largely polarized parallel to the major polarization axis of the fundamental for ϵ in the range 0–0.1; although a weak component (reduced by about a factor of 10) was observed in the transverse polarization, this transversely polarized component was observed to have a minimum at $\epsilon=0$ as expected.

The ellipticity and polarization dependence of harmonic generation observed in the present experiment can be largely understood by considering transitions involving free electrons during either the one-half collision associated with the ionization event itself (third and fifth harmonics) or the full collision associated with the reencounter of ionizing electrons with their parent ion within the first period of the fundamental after ionization (15th and higher harmonics). During this reencounter, the free electron, having acquired energy from the fundamental field, can spontaneously recombine to the ground state emitting a photon with $h\nu>I_p$. The 13th harmonic evidently has an origin that is distinct from either the lowest or highest harmonic energies. We will discuss the ellipticity dependence expected from the first two mechanisms and briefly speculate about possible origins for the 13th harmonic with its unique ellipticity dependence and polarization.

The ellipticity dependence expected for harmonics generated by spontaneous recombination subsequent to tunnel ionization follows from applying Newton's law to electrons released at rest in an ac electric field. We will assume $E_x=E_0\sin\omega t$, $E_y=\epsilon E_0\cos\omega t$, where the ellipticity ϵ is assumed to be much less than 1. Because of the nonlinear field dependence of the tunnel ionization rate, electrons are created (released) mainly around the crests of the x field. Electron trajectories in the x and y directions are entirely determined by the phase at which ionization occurs. There exists a maximum velocity (energy) with which electrons can reencounter their parent [6,11] corresponding to about $3.17U_{\text{osc}}$ (where U_{osc} denotes the ponderomotive potential). This corresponds to electrons released at a phase near the crests of the field that return about $\frac{3}{4}$ of a cycle later. Lower-energy reencounters are possible at two phases on either side of that corresponding to maximum energy. When a small y field 90° out of phase is added, the y trajectory is such as to displace high-energy reencounters in the y direction [11] a distance $\sim 5.7\epsilon eE_0/m\omega^2$. If one assumes a saturation intensity of 5×10^{14} W/cm^2 for the present experiment, the spatial scale $eE_0/m\omega^2$ is approximately 17 Å, so that an ellipticity ϵ of about 0.01 would be sufficient to displace high-energy electrons by more than an atomic dimension at the time corresponding to their reencounter, thus precluding recombination (harmonic generation). Classically, this displacement can be overcome, of course, by giving the ionizing electron a suitable transverse momentum at the time of its creation. The transverse momentum (or energy) distribution of the tunnelling electron is determined by quantum effects. The finite width of this distribution places a second somewhat less restrictive limit on the maximum ellipticity compatible with harmonic generation. The experimental results shown in Figs. 1 and 2 represent a direct measurement of the transverse momentum distribution of tunneling electrons, or alternatively, the harmonic yield $Y_{fb}(\epsilon)$ is proportional to the squared modulus of the returning (free) electron wavepacket, $\psi^2(r)$ at radius $r(\epsilon)\approx 5.7\epsilon eE_0/m\omega^2$. In the present experiment the plateau harmonics suggest a full width at half maximum for $\psi^2(r)$ of $11.4\epsilon_{1/2}eE_0/m\omega^2\approx 20$ Å, where $\epsilon_{1/2}$ is the ellipticity required to quench harmonic yield by a factor of 2.

Recollision is not the only possible source of harmonic emission in ionizing gases. The contribution to harmonic generation from the high-frequency variation in the tunnel ionization rate is ignored in the semiclassical picture and has also been neglected in the calculations of Becker [5] and Lewenstein *et al.* [8]. This neglect is most serious for the first few harmonics. A classical model for harmonic generation based on the high-frequency variation of the tunnel ionization current has been proposed by Brunel [9]. A similar model has been used by Rae and Burnett [14]. Although this model does not predict a plateau in the harmonic spectrum, as observed experimentally, it does appear capable of producing about the right magnitude for the third and fifth harmonics observed in the present experiment (the experimentally observed third harmonic yield is about 10^{-6} of the fundamental; and the fifth, about 50 times weaker). In this case the dipole acceleration or $\partial J/\partial t$ responsible for harmonics can be written as

$$\partial J/\partial t=n_e(t)\left(\frac{e^2}{m_e}E(t)\sin(\omega t)\right),$$

where $n_e(t)$ is the electron density (or ionization probability for a single atom). The harmonic spectrum from a single atom is given by the squared modulus of the Fourier transform of the dipole acceleration. Harmonics produced according to this model will be sensitive to ellipticity through its effect on the time-dependent ionization probability (circularly polarized light produces no high-frequency variation of n_e). The theoretical fits in Fig. 1 for the third and fifth harmonic yield were obtained by taking the Fourier transform of the dipole acceleration for a pulse corresponding to the experimentally used duration and intensity and assuming an ionization rate given by Ammosov-Delone-Krainov theory [15]. The observed fits are not unreasonable in view of the fact that propagation and phase-matching effects are ignored completely while the degree of ionization reached during the pulse depends slightly on ellipticity.

Neither of the above models appears capable of predicting the polarization and ellipticity dependence of the 13th harmonic observed here, namely that the yield of this harmonic is enhanced by an elliptically polarized fundamental and that this harmonic can be significantly polarized in the direction orthogonal to the major polarization axis of the fundamental. Although there is not a great deal of published data on harmonic generation just below the ionization potential in gases undergoing tunnel ionization, it is noteworthy that strong quenching of the first harmonic below the unshifted ionization potential has been reported by Li *et al.* [16] for argon irradiated with a linearly polarized 1-μm laser. This may be a further indication of a change in mechanism for harmonic production below the ionization potential. Candidate mechanisms for harmonic production in this intermediate-energy regime include the nonlinear mixing of bound states in the neutral atom and free-free transitions associated with electron recollision subsequent to tunnel ionization. Of these candidates, the free-free mechanism seems likely to provide, in the most straightforward way, for the observed ellipticity dependence and polarization characteristics. Free-free transitions have been considered as a source of harmonic emission from a linearly polarized fundamental [17] but have not to our knowledge been analyzed in the case of elliptically polarized light. Some of the essential elements of coherent bremsstrahlung emission in this geometry can be inferred form the theory of classical bremsstrahlung [18]. The cutoff frequency for classical bremsstrahlung is given by $\omega < v/b$, where b is the impact parameter associated with the collision at velocity v. In order to emit a harmonic photon of angular frequency $13\omega_0$, an electron with energy $3.17U_{\rm osc}$ ($U_{\rm osc}=53$ eV at 10^{15} W/cm^2) must pass within about 2 Å of its parent. In the case of a coherent returning electron wavepacket with a linearly polarized fundamental, equal and opposite accelerations will result from the two portions of the wavepacket situated to the"right" and "left' of the parent ion. It is only by breaking this symmetry that a net transverse acceleration will result and orthogonally polarized coherent bremsstrahlung radiation can be emitted. Since the impact parameters of interest are small compared to the radial extent of the returning wavepacket, as inferred from the ellipticity dependence of harmonics above 13, one might expect a harmonic yield proportional to the square of the derivative of $\Psi^2(r)$, i.e., $Y_{ff}(\epsilon)\propto(\partial Y_{fb}/\partial\epsilon)^2$. In fact, the data for the ellipticity dependence of the orthogonally polarized component of the 13th harmonic, as shown in Fig. 3, is reasonably fit by taking the square of the derivative of $Y^N(\epsilon)$, where N is greater than 13. This agreement must be regarded as suggestive rather than conclusive since no quantitative theory for coherent bremsstrahlung has yet been formulated.

In summary, the data presented here indicate that several different mechanisms are necessary to account for harmonic generation in neon. Under the conditions of the present experiment, the lowest harmonics may be reasonably accounted for with a tunnel current model, but it does not appear that this mechanism makes a smooth transition to harmonic generation based on the ionization-acceleration-recombination model. At least in the cases of neon and helium there appears to be an intermediate region in the vicinity of the ionization potential ($N=13$) where free-free or bound-bound transitions can contribute to the nonlinear atomic susceptibility.

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