Theory of Relativistic Optical Harmonic Generation

Q-Han Park, Robert W. Boyd, J. E. Sipe, and Alexander L. Gaeta

Invited Paper

***Abstract*—We present a theoretical model of optical harmonic generation excited by laser beams sufficiently intense that relativistic effects are important. This model shows that, under relativistic conditions, third-harmonic generation can be excited with comparable efficiency by either linear or circularly polarized light. This result is to be contrasted with experience from traditional (nonrelativistic) nonlinear optics, where group-theoretical arguments show that third-harmonic emission cannot occur under circularly polarized excitation. These results are in good agreement with the observed polarization dependence of the third-harmonic emission reported recently in an experiment conducted under conditions such that relativistic effects are important. Our theoretical model also predicts that all even and odd harmonics of the fundamental laser frequency are emitted in the near-forward direction with an intensity that increases with that of the incident laser field.**

***Index Terms*—Optical harmonic generation, relativistic effects.**

RECENT ADVANCES in laser technology [1] have led to the ability to construct lasers with intensities exceeding 10^{20} W/cm^2. Such large intensities have led to the observation of new forms of light-matter interaction, including above threshold ionization [2], high-harmonic generation [3]–[8], and relativistic self-focusing [9]–[11]. An additional form of intense-field nonlinear optics is relativistic harmonic generation. In a recent publication, Chen *et al.* [12] describe their observation of phase-matched third-harmonic generation (THG) in a highly ionized gas under laser intensities sufficiently large (2×10^{17} W/cm^2) that relativistic effects are expected to be important. The harmonic radiation is found to be emitted in the near-forward direction, with a reasonably large ($\sim 2 \times 10^{-5}$) conversion efficiency [13].[1] Perhaps most surprisingly, they found that the intensity of the THG is comparable for linear and circular polarization of their incident laser beam. In traditional nonlinear optics, the process of THG is known to vanish identically for an isotropic medium for a circularly polarized fundamental laser beam [14].

Let us recall why traditional models [15] predict the vanishing of THG under excitation with circularly polarized radiation and see why this argument breaks down under relativistic conditions. Nonlinear optical interactions are often described in terms of a nonlinear optical susceptibility, which for the case of third-harmonic generation relates the polarization of the medium to the third power of the electric field amplitude of the incident laser beam. Harmonic light is then radiated as a consequence of the time-varying polarization of the medium. The standard calculational procedure presupposes the validity of the electric-dipole approximation, both in the calculation of the nonlinear susceptibility and in terms of calculating how the response of the medium leads to the generation of new frequency components. Detailed examination of the tensor nature of the nonlinear susceptibility for an isotropic nonlinear medium shows that the nonlinear response vanishes for a circularly polarized input beam [14]. Physically, this result can be understood from the perspective that under nonrelativistic conditions, each electron (whether bound or free) is induced to rotate in a circular orbit at the fundamental frequency of the incident laser light. For a bound electron, the radius of this orbit can depend nonlinearly on the amplitude of the laser field, but there can still be no harmonic generation because the motion is purely sinusoidal.

Let us next see how the analysis of harmonic generation is different for excitation by an ultra-intense laser field [16], [17]. Such a laser field is capable of ionizing some fraction of the atoms, producing a collection of free electrons. We consider the motion of a free electron in an intense laser field of wavelength λ. For field strengths of the order of $mc^2/\lambda e$, which for $\lambda = 1\ \mu$m corresponds to an intensity of the order of 2×10^{17} W/cm^2, the electron is accelerated to relativistic velocities in a half optical period, and thus the excursion of the electron from its equilibrium position occurs over a distance of the order of λ. Under such conditions, the radiation emitted by the electron cannot be described within the electric-dipole approximation, and in fact cannot be well described by a multipole expansion for a finite number of terms [18]. We instead calculate the intensity of the emitted radiation through use of the Lienard–Wiechert potentials, as described below.

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[1]Ref. 5 reported that the third-harmonic radiation was emitted in a cone in the forward direction, but the authors have subsequently determined that the cone-like nature of the emission was an experimental artifact.

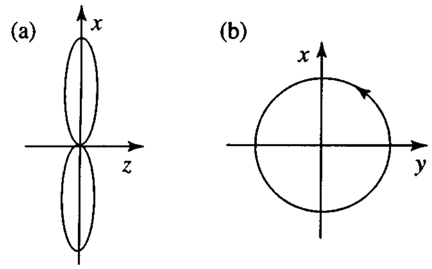

Fig. 1. Motion of a free electron in (a) a linearly polarized laser field and (b) a circularly polarized field. Note that for linearly polarized light the motion is in the xz plane and that for circularly polarized light is in the xy plane, where z is the propagation direction and x is the direction of the electric field in the linearly polarized case.

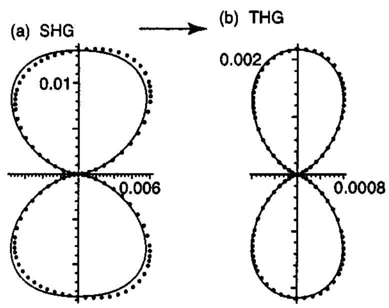

Fig. 2. Radiation patterns for (a) SHG and (b) third-harmonic generation for a circularly polarized fundamental laser beam. The arrow denotes the propagation direction of the laser beam. In each of the figures, we assume a laser intensity of 2×10^{17} W/cm^2. The solid curves give the predictions in the electron drift frame and the dotted curves give the results in the lab frame.

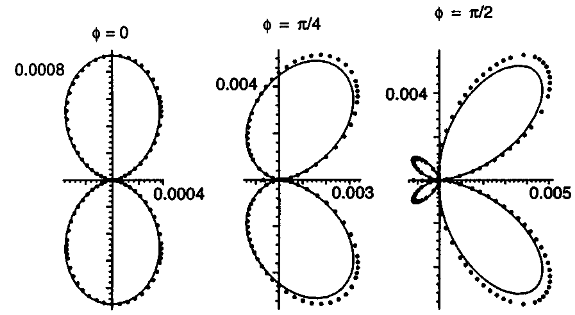

Fig. 3. Radiation patterns for SHG for a linearly polarized fundamental laser beam; ϕ is the angle between the electric field vector of the laser beam and the plane of the figure. In each of the figures, we assume a laser intensity of 2×10^{17} W/cm^2. The solid curves gives the predictions in the electron drift frame and the dotted curves give the results in the lab frame.

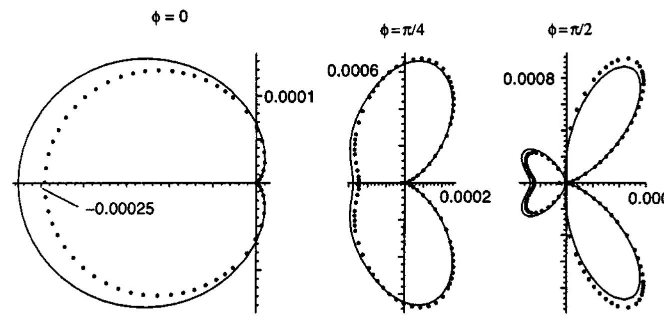

Fig. 4. Radiation patterns for third-harmonic generation for a linearly polarized fundamental laser beam; ϕ is the angle between the electric field vector of the laser beam and the plane of the figure. In each of the figures, we assume a laser intensity of 2×10^{17} W/cm^2. The solid curves give the predictions in the electron drift frame and the dotted curves give the results in the lab frame.

The motion of a free electron in an intense electromagnetic field of frequency ω is well known [19] (see Fig. 1). For circularly polarized light, the electron orbits at frequency ω in a circle of radius $r_0 = \beta c/\omega$ where $\beta = eE_0/\gamma\omega$ and $\gamma^2 = m^2c^2 + e^2E_0^2/\omega^2$. For linearly polarized light, the electron moves in a figure-eight pattern of comparable dimensions. This motion can be described in cartesian coordinates by the equations $x = 0, y = (\beta c/\omega)\cos\eta$ and $z = (\beta^2 c/8\omega)\sin 2\eta$, where $\eta = \omega(t - z/c)$. Even though the motion is strictly harmonic for the circularly polarized situation, and (when expressed in terms of the retarded time variable $\eta = \omega(t - z/c)$) contains only frequencies ω and 2ω in the linearly polarized case, the radiation field contains all harmonics of the fundamental frequency in both cases because of retardation effects. In particular, the vector potential of the n-th harmonic component of the radiated wave at distance R_0 from the center of the electron orbit is given by [19]

$$\mathbf{A}_n = \frac{c\omega e^{ik_0 R_0}}{\pi c R_0} \oint e^{i(n\omega t - \mathbf{k}\cdot\mathbf{r})} d\mathbf{r} \tag{1}$$

where $k_0 = \omega/c, k = n\omega/c, \mathbf{k}$ has magnitude k and points in the direction of the observation point, and $\mathbf{r}$ gives the *instantaneous* position of the electron. The power per unit solid angle emitted by this harmonic component is then given by

$$J(\theta,\phi) \equiv \frac{dP_n}{d\Omega} = \frac{c}{8\pi}|\mathbf{k}\times\mathbf{A}_n|^2 R_0^2. \tag{2}$$

Some of the predictions of this model are shown in Figs. 2–4. In all cases, we show the radiation pattern $J(\theta,\phi)$ (power per unit solid angle) of the emitted radiation in cgs units of erg s^{-1} rad^{-2}. Fig. 2 shows the predicted radiation patterns for second- and third-harmonic generation for a circularly polarized fundamental laser beam. These patterns are of course symmetric about the propagation direction. Note that both radiation patterns vanish in the exact forward direction. Radiation is thus, expected only in the off-forward direction, at an angle determined by phase matching considerations. Under actual laboratory conditions, it is likely that the emitted radiation would be observed even on axis, after the emission pattern in averaged over the angular spread of the exciting laser beam. Fig. 3 shows the predicted radiation pattern for second-harmonic generation (SHG) for a linearly polarized laser beam; Fig. 4 shows the analogous predictions for third-harmonic generation. The radiation patterns are not azimuthally symmetric in this case, and cuts through the pattern are shown for angles of 0°, 45°, and 90° between the plane of the pattern and the electric field direction of the incident laser beam. Again, harmonic generation is not emitted in the exact forward direction. Note that linearly and circularly polarized light are roughly equally efficient at exciting harmonic generation for both second- and third-harmonic generation. We have evaluated (2) analytically for the circularly polarized case and numerically for the linearly polarized case. The analytic expression for the circularly polarized case is

$$\frac{dP_n}{d\Omega} = \frac{n^2 w^2 e^2}{2\pi c}\left[\beta^2 J_n'^2(n\beta\sin\theta) + \cot^2\theta\, J_n^2(n\beta\sin\theta)\right] \tag{3}$$

where the normalized velocity is given by $\beta = v/c = q/\sqrt{2+q^2}$ for $q = \sqrt{2}eE_0/(mcw)$.

We note further that the predicted conversion efficiency is in good order-of-magnitude agreement with the measured value. Chen *et al.* state that the electron density under their experimental conditions is 3×10^{19} cm^{-3}. We take the interaction region to be the focal volume of a laser beam focused to a waist of radius 7 μm. We thus, find that $N = 1.4 \times 10^{12}$ electrons participate in the interaction. We take the power of the input beam to be the quoted intensity of 2×10^{17} W/cm^2 times the effective cross-sectional area of a beam of 7-μm radius or as $P_1 = 3 \times 10^{11}$ W. We estimate the power emitted in the third harmonic in a nearly phase-matched interaction as

$$P_3 = \eta N^2 \int J(\theta, \phi)\, d\Omega \tag{4}$$

where η is an efficiency factor that accounts for the possibility of imperfect phase matching. The details of the refractive index distribution in and around the interaction region are not known to sufficient accuracy to allow for a precise determination of the influence of phase matching on the emission process. To provide a first estimate of the expected efficiency of the harmonic generation process, we assume that phase-matching considerations restrict the range of solid angles in the integration of (4) to those in a filled-in cone of angular extent θ_1 equal to that of the fundamental laser beam. We take θ_1 to be $\lambda/\pi w_0$, the standard result for a gaussian beam. For the case of third-harmonic generation, we then find that for both linear and circular polarization the predicted emitted power is given by $P_3 = 1 \times 10^7 \eta$ W. The measured conversion efficiency was $f = 10^{-5}$, which implies that the phase-mismatch efficiency factor η has the reasonable value of 0.3.

The present model predicts that second-harmonic generation (SHG) should also be emitted, with an efficiency no smaller than that of the third-harmonic. Although, Chen *et al.* [12] make no mention of the observation of SHG in their initial publication, SHG has been observed in subsequent work [20]. The efficiency of the emission of the SHG is likely to depend upon the subtleties of the phase-matching process and can perhaps be explained in terms of the model presented in [12] or [21]. The precise measurements of the angular distribution of the emitted radiation for various harmonics for both linear and circularly polarized input beams could provide valuable data to allow a more complete testing of the understanding of nonlinear optical processes in relativistic plasmas.

The model described above can also be used to make predictions of the intensity dependence of the generation of the higher harmonic orders. These predictions could prove useful in performing the next generation of experiments in relativistic harmonic generation. These predictions are shown in Fig. 5 for both linearly and circularly polarized laser light. As above, we calculate the total emitted power in each order through use of (3) with $\eta = 1$ by integrating the radiated power per unit solid angle $J(\theta, \phi)$ over a cone of maximum angular extent equal to θ_1. Several general features appear from examination of these figures. We note that linearly and circularly polarized light are roughly equally effective at exciting relativistic harmonic generation. We also see that higher harmonics initially increase more rapidly with laser intensity, but that each harmonic eventually saturates to a constant value. Detailed examination of Fig. 5 (or

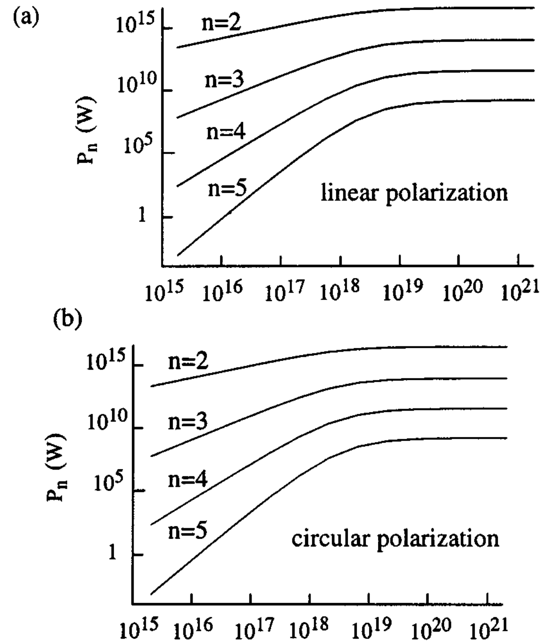

Fig. 5. Predicted power (in the electron drift frame) radiated in the first four harmonics of the fundamental laser frequency plotted as functions of the intensity of the incident laser field for (a) linearly and (b) circularly polarized laser radiation.

of(1) through (3)) reveals that the power in the n-th harmonic increases initially as I^n for both linearly and circular polarization of the fundamental wave. In the limit of very high intensity of the fundamental laser, the power in each harmonic is given through use of (1) through (3) for a circularly polarized input by the simple, intensity-independent expression

$$P_n = \frac{n^2\omega^2 e^2}{2\pi c} \times \int_0^{\theta_1} \left[\cot^2\theta J_n^2(n\sin\theta) + J_n'^2(n\sin\theta)\right] 2\pi\sin\theta\, d\theta. \tag{5}$$

This expression accurately predicts the asymptotic values of P_n shown in Fig. 5. Similar conclusions hold for the high-intensity limit for a linearly polarized fundamental beam, but we have not found a closed-form expression analogous to (4). For either type of polarization, the saturation of the harmonic power can be understood from the perspective that even at very high intensities the electron excursion from its equilibrium position is limited to a distance of the order of the wavelength of the incident radiation.

In the results given above, we have not taken into account of the drift motion of the free electron and our results are in fact valid only in the "drift (average rest) frame" [16]. The drift velocity, $v_D = q^2/(4+q^2), q = \sqrt{2}eE_0/(mcw)$, however becomes large for intense incident laser fields. In order to make a realistic comparison with experimental detections, we need to transform our result into the lab frame. The Lorentz transformation relates the angle θ_D in the drift frame to the angle θ_L in the lab frame through

$$\cos\theta_D = \frac{\cos\theta_L - (q^2/2)\sin^2(\theta_L/2)}{1 + (q^2/2)\sin^2(\theta_L/2)}$$
$$\sin\theta_D = \frac{\sqrt{1+q^2/2}\sin\theta_L}{1 + (q^2/2)\sin^2(\theta_L/2)}. \tag{6}$$

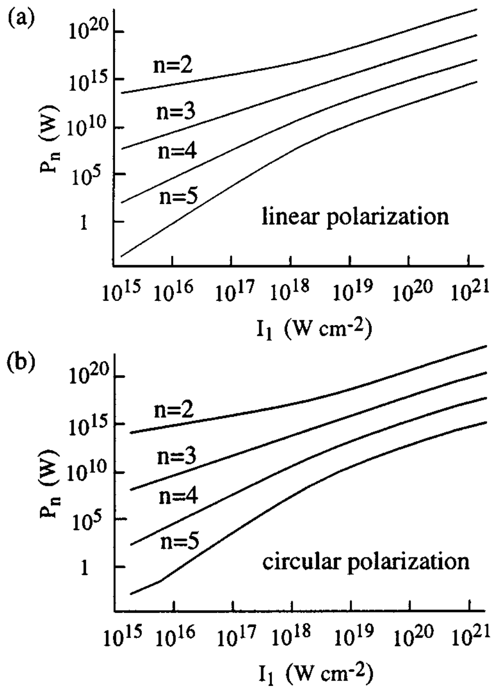

Fig. 6. Predicted power (in the lab frame) radiated in the first four harmonics of the fundamental laser frequency plotted as functions of the intensity of the incident laser field for (a) linearly and (b) circularly polarized laser radiation.

The emitted power per unit solid angle is related by

$$\frac{dP_L}{d\Omega_L} = \frac{(1+q^2/2)^2}{[1+(q^2/2)\sin^2(\theta_L/2)]^4}\frac{dP_D}{d\Omega_D}. \tag{7}$$

In particular, in contrast to (3), the harmonic component of the circularly polarized case in the lab frame becomes

$$\frac{dP_L^{(n)}}{d\Omega_L} = \frac{e^2w^2q^2}{4\pi c}\frac{n^2}{[1+(q^2/2)\sin^2(\theta/2)]^4}\left[J_n'^2(n\Theta) + \frac{2(\cos\theta-(q^2/2)\sin^2(\theta/2))^2}{q^2\sin^2\theta}J_n^2(n\Theta)\right] \tag{8}$$

where

$$\Theta = \frac{q\sin\theta}{\sqrt{2}(1+(q^2/2)\sin^2(\theta/2))}. \tag{9}$$

Lab frame results are also drawn in Figs. 2–4 using dotted curves and show small but noticeable deviations from the drift frame results. However, in the high intensity limit, Fig. 6 shows a significant deviation of the radiated power in the lab frame.

In summary, we have presented a theoretical model that explains the polarization dependence of the relativisitic third-harmonic generation observed by Chen *et al.* [12]. This model also predicts that all even and odd harmonics of the laser frequency are emitted with an efficiency that decreases with harmonic order. We have also found that the power emitted in each order saturates with increasing laser intensity and eventually reaches an asymptotic value. These predictions could prove useful in future research into relativistic nonlinear optics.

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J. E. Sipe, photograph and biography not available at the time of publication.

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