Nonlinear Magneto-Optics of Vacuum: Second-Harmonic Generation

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The photon-photon scattering of intense laser radiation predicted by QED can give rise to second-harmonic generation in a dc magnetic field due to broken symmetry of the interaction. The laser energy required to observe this effect can be achieved by using available laser facilities and the state-of-the-art photon-counting technology.

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Photon-photon scattering in a vacuum[1] is perhaps one of the most fundamental mechanisms which can give rise to nonlinear optical effects. From the classical point of view, the expected nonlinear interaction [see below, Eqs. (1) and (2)] essentially corresponds to the third-order nonlinearity.[2] This interaction may contribute to the birefringence of the refractive index seen by a probe field under the action of either a dc magnetic (or electric) field[3] or intense laser pumping,[4] as well as to multiwave mixing processes.[5] To the best of our knowledge, no experimental work on this phenomenon has been done.

In this paper, we demonstrate the feasibility of new nonlinear magneto-optical effects in a vacuum that give rise to optical *second-harmonic generation* (SHG) of the fundamental wave under the action of both strong dc magnetic field and high-intensity optical laser radiation. We also propose an experiment for the observation of this effect. The advantage of using the SHG effect is twofold. Since only a second-order effect for the optical field is involved, the laser power required to observe SHG is much lower than in previously proposed effects.[3-5] The SHG can also be measured at a frequency different from the fundamental frequency injected into the system, which may result in higher sensitivity.

From the Heisenberg-Euler Lagrangian[1] for photon-photon scattering in quantum electrodynamics (QED) theory, one obtains the following expressions for the electric displacement $\mathbf{D}$ and magnetic induction $\mathbf{B}$:

$$\mathbf{D}=\mathbf{E}+\mathbf{D}^{\mathrm{NL}},\quad \mathbf{D}^{\mathrm{NL}}=\xi[2(E^2-H^2)\mathbf{E}+7\mathbf{H}(\mathbf{E}\cdot\mathbf{H})]\,; \tag{1}$$

$$\mathbf{B}=\mathbf{H}+\mathbf{B}^{\mathrm{NL}},\quad \mathbf{B}^{\mathrm{NL}}=\xi[2(E^2-H^2)\mathbf{H}-7\mathbf{E}(\mathbf{E}\cdot\mathbf{H})]\,; \tag{2}$$

where $\xi=\alpha/45\pi H_{\mathrm{cr}}^2=2.6\times10^{-32}\ \mathrm{G}^{-2}$ is a nonlinear interaction constant in the vacuum, with $\alpha=e^2/\hbar c=1/137$ the fine-structure constant and $H_{\mathrm{cr}}\equiv m_0^2c^3/e\hbar=4.4\times10^{13}$ G the QED critical field. These equations are valid only if the nonlinear corrections $\mathbf{D}^{\mathrm{NL}}$ and $\mathbf{B}^{\mathrm{NL}}$ are small, which holds if $|E|=|H|\ll H_{\mathrm{cr}}$. It is obvious that a single monochromatic plane wave of infinite extent does not exhibit any nonlinear effects, because it has the properties $E^2=H^2$, $\mathbf{E}\cdot\mathbf{H}=0$, and the nonlinear terms in Eqs. (1) and (2) vanish. This "degeneracy" of the nonlinearity is broken if either (i) the wave is nonplanar or nonmonochromatic or (ii) a strong static field (e.g., a dc magnetic field) is present. Both cases can result in birefringence of the refractive index for a probe field.[3,4] We show here that a dc magnetic field can also give rise to second-order nonlinear optical effects similar to those found in nonlinear materials.[2] In general, the optical second-order nonlinearity can give rise to the generation of a third wave (at frequency $\omega_2=\omega_a\pm\omega_b$) from two intense laser beams at frequencies ω_a and ω_b (i.e., the sum- and difference-frequency generation). Here, we consider only SHG in which $\omega_a=\omega_b$, $\omega_2=2\omega_1$. However, our calculations can easily be generalized for the case $\omega_a\neq\omega_b$.

Assuming that a single unperturbed fundamental wave, described by the fields $\mathbf{E}_1$ and $\mathbf{H}_1=(\mathbf{k}_1/|\mathbf{k}_1|)\times\mathbf{E}_1$ (where $\mathbf{k}_1$ is the wave vector of the field), propagates in vacuum in the presence of a dc magnetic field, $\mathbf{H}_0$, the nonlinear components in Eqs. (1) and (2) can be rewritten as

$$\mathbf{D}^{\mathrm{NL}}=\xi[-2H_0^2\mathbf{E}_1+7\mathbf{H}_0(\mathbf{E}_1\cdot\mathbf{H}_0)]+\mathbf{D}^{(2)}\,, \tag{3}$$

$$\mathbf{B}^{\mathrm{NL}}=-2\xi H_0^2\mathbf{H}_0-2\xi[H_0^2\mathbf{H}_1+2\mathbf{H}_0(\mathbf{H}_0\cdot\mathbf{H}_1)]+\mathbf{B}^{(2)}\,, \tag{4}$$

where

$$\mathbf{D}^{(2)}=\xi[-4\mathbf{E}_1(\mathbf{H}_1\cdot\mathbf{H}_0)+7\mathbf{H}_1(\mathbf{E}_1\cdot\mathbf{H}_0)]\,, \tag{5}$$

$$\mathbf{B}^{(2)}=\xi[-4\mathbf{H}_1(\mathbf{H}_1\cdot\mathbf{H}_0)-7\mathbf{E}_1(\mathbf{E}_1\cdot\mathbf{H}_0)]\,. \tag{6}$$

Here, we neglect the third-order nonlinearity caused by the finite size of the laser beam which may result in self-action effects. It can be shown that in the case of a quasiplane wave, e.g., a Gaussian beam with sufficiently large beam waist d, $d\gg\lambda=2\pi/k$, this effect is negligible when $E_1\ll H_0(d/\lambda)^2$. Nonlinear effects due to gradients in the field distribution will be discussed by us elsewhere. The terms in square brackets in Eqs. (3) and (4) are linear in optical-field strengths. They result in vacuum birefringence of the refractive index for a weak probe field ($|\mathbf{E}_1|=|\mathbf{H}_1|\ll H_0$) under the action of a dc magnetic field.[3] The first term on the right-hand side of Eq. (4) corresponds to dc corrections to the dc magnetic field induced by the magnetic field itself. Therefore, only the terms $\mathbf{D}^{(2)}$ and $\mathbf{B}^{(2)}$, Eqs. (5) and (6), give rise to the optical second-order nonlinearity. Our estimates also show that for the field intensities available now and in the foreseeable future, the phase mismatch between fundamental frequency and second harmonics can be neglected.

Because of the spatial anisotropy imposed by the magnetic field, SHG depends upon the propagation direction and the polarization of the fundamental optical wave with respect to $\mathbf{H}_0$. If the fundamental wave propagates along the direction of the dc magnetic field $\mathbf{H}_0$, then $\mathbf{D}^{(2)}=0$, $\mathbf{B}^{(2)}=0$, and the nonlinear effects are suppressed. The strongest interaction occurs when the laser radiation propagates in a direction normal to the dc magnetic field. Consider the general case of an elliptically polarized wave propagating in the plane normal to the dc magnetic field $\mathbf{H}_0=H_0\hat{\mathbf{e}}_z$, with its polarization lying in the plane normal to the propagation direction, e.g., $\mathbf{k}_1=k_1\hat{\mathbf{e}}_y$. Its electric field can be decomposed into two linearly polarized components along the $\hat{\mathbf{e}}_z$ and $\hat{\mathbf{e}}_x$ axes, respectively:

$$\mathbf{E}_1=E_1(\sin\theta_1\hat{\mathbf{e}}_x+\cos\theta_1 e^{i\phi}\hat{\mathbf{e}}_z)\exp[i(k_1y-\omega_1t)]\,, \quad (7)$$

where E_1 and ω_1 are the amplitude of the electric field and the angular frequency of the wave, respectively, ϕ is the phase between the linearly polarized components, and the angle θ_1 designates the relative amplitude between these two waves ($\phi=0$ would correspond then to a linearly polarized wave with θ_1 the angle of the linear polarization, whereas $\phi=\pi/2$ and $\theta_1=\pi/4$ correspond to a circularly polarized wave).

The generated second-harmonic field at frequency $\omega_2=2\omega_1$ propagates in the same direction as that of the fundamental wave. Its electric field after passing through the interaction length $y=L$ can be calculated using Maxwell's equations with nonlinear terms, Eqs. (5) and (6):

$$\mathbf{E}_2=iE_2(\sin\theta_2\hat{\mathbf{e}}_x+\cos\theta_2 e^{i\phi_2}\hat{\mathbf{e}}_z)\times\exp\{i[2(k_1L-\omega_1t)+\phi-\phi_2]\}\,, \quad (8)$$

where the magnitude E_2 is given by

$$E_2=k_1L\xi H_0E_1^2[56(1-\cos2\phi)\cos^4\theta_1+(56\cos2\phi-23)\cos^2\theta_1+16]^{1/2}\,, \quad (9)$$

the angle θ_2 is determined by

$$\tan\theta_2=-2[(65-56\cos2\phi)\cos^4\theta_1+(56\cos2\phi-32)\cos^2\theta_1+16]^{1/2}/(3\sin2\theta_1)\,, \quad (10)$$

and the phase ϕ_2 for the second harmonic is computed as

$$\tan\phi_2=-[(7-4\tan^2\theta_1)/(7+4\tan^2\theta_1)]\tan\phi\,. \quad (11)$$

[When $\theta_1=\theta_{1_0}\equiv\tan^{-1}(\sqrt{7}/2)\simeq53°$, the second harmonic is *linearly* polarized regardless of the polarization state of the fundamental wave.] Consider now the particular case of a linearly polarized fundamental wave. In such a case, $\phi=0$ in Eq. (7), and the second harmonic is also linearly polarized; the angles θ_1 and θ_2 in Eqs. (7) and (8) are then the polarization angles of the fundamental wave and the second harmonic, respectively [see inset, Fig. 1(b)]. Equations (9) and (10) then reduce to

$$E_2=k_1L\xi H_0E_1^2(33\cos^2\theta_1+16)^{1/2}\,,\qquad \theta_2=-\tan^{-1}[(3\cos2\theta_1+11)/(3\sin2\theta_1)] \quad (12)$$

(see Fig. 1). Therefore, the ratio of the maximum intensity of the second harmonic (which occurs at $\theta_1=0$) to minimum intensity (at $\theta_1=\pi/2$) is $(7/4)^2\simeq3$ [see Fig. 1(a)], which can be directly measured in an experiment. Note that the generated second harmonic can never be polarized along the direction of the dc magnetic field; i.e., the polarization angle $\theta_2\neq0$ in Eq. (12). A minimal angle between the polarization of the second harmonic and the dc magnetic field is $(\theta_2)_{\min}\simeq74°$, and the corresponding polarization angle of the fundamental wave is $\theta_1=\theta_{1_0}\simeq53°$; i.e., there is a prohibited sector for the polarization of the second harmonic, $-(\theta_2)_{\min}<\theta_2<(\theta_2)_{\min}$ [see Fig. 1(b)]. All these polarization properties can be used in a future experiment to rule out all other (i.e., nonvacuum) nonlinear mechanisms. In the case of a circularly polarized fundamental wave ($\phi=\pi/2$, $\theta_1=\pi/4$) propagating normal to the dc magnetic field, the ratio of the maximum intensity of second harmonic for linear polarization to that for circular polarization is $(7\sqrt{2}/3)^2\simeq10.9$ [see Fig. 1(a)].

The state-of-the-art photon-counting systems provide a dark photon count rate $r_{\text{dark}}\sim10$ photons/s and a typical quantum efficiency $\eta\sim0.25$.[6] For ideal spectral filtering and provided that, by using gating, the detector is open only during the laser pulse,[6] the signal-to-noise ratio (SNR) is $\langle n_{\text{det}}\rangle/\langle n_{\text{dark}}\rangle$, where $\langle n_{\text{det}}\rangle=\eta\langle n_{\text{SHG}}\rangle$ is the averaged number of detected photons per pulse, n_{SHG} is the number of SHG photons generated per pulse, $\langle n_{\text{dark}}\rangle=r_{\text{dark}}\tau_p$, and τ_p is the duration of a laser pulse. Stipulating now that SNR is sufficiently high, e.g., $\geq10^2$, and considering the case of normal polarization ($\theta_1=\pi/2$, which would correspond to the minimal SH photon output), we obtain the lowest intensity I_{cr} and energy J_{cr} of the laser beam required for such SNR:

$$I_1\geq I_{\text{cr}}(\text{W/cm}^2)=(10^{19}/H_0L)(\lambda_1/A)^{1/2}\,;\qquad J_{\text{cr}}(\text{J})=\tau_pAI_{\text{cr}}\,, \quad (13)$$

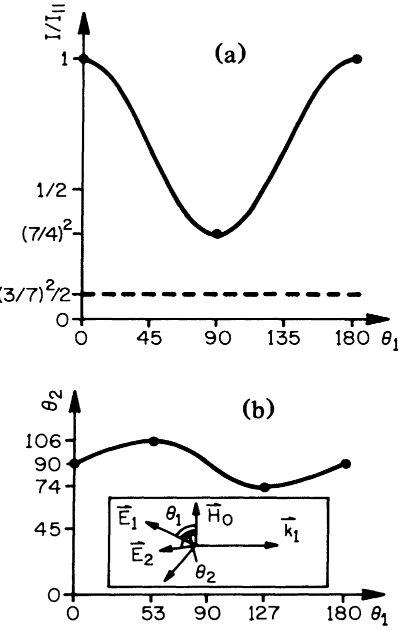

FIG. 1. (a) The normalized intensity $I/I_{\parallel}$ for the second harmonic with polarization angle θ_2 with respect to the dc magnetic field [inset in (b)] vs the polarization angle θ_1 (in degrees) of a linearly polarized fundamental wave (solid line); broken line corresponds to a circularly polarized wave; (b) the polarization angle θ_2 (in degrees) of the second harmonic vs the polarization angle θ_1 (in degrees) of a linearly polarized fundamental wave; the second harmonic is never polarized in sectors $|\theta_2 - 90°| > 16°$. Inset: The wave propagation configurations for both the fundamental wave and second harmonic with respect to the dc magnetic field $\mathbf{H}_0$.

where λ_1 is in μm, A is the laser focal area in cm^2, τ_p is in sec, H_0 is in gauss, and L is in cm. The minimal A should be chosen as $A \sim \lambda_1 L/2$ such that the diffraction of the beam is small within the distance L. With pulsed-magnet technology,[7] the best parameter values are $H_0 \sim 8\times10^6$ G, bore diameter ~ 2.8 cm, and pulse duration $\sim 10^{-6}$ sec. Therefore, in order to satisfy the condition, Eq. (13), a laser intensity of $I_1 \simeq 10^{14}$ W/cm^2 is required, which can readily be achieved even by using commercial laser systems. In fact, in the laser systems discussed below the intensity reaches $10^{15}-10^{18}$ W/cm^2, so this condition is satisfied with great margin.

Since in most high-power laser systems available, $\langle n_{\rm det}\rangle$ is of the same order as or less than unity, one has to use averaging of photon counts over a few (usually incoherent) laser beam lines for a single pulse and/or over many laser pulses. Assuming a Poisson distribution of SHG photons,[6] the probability of *not* seeing any SHG photons within N laser pulses is $p = \exp(-N\langle n_{\rm det}\rangle)$. Stipulating again that p should be sufficiently small, e.g., $\le 10^{-2}$, we obtain a condition for the required number of pulses (or number of beam lines for one laser shot):

$$N \ge 2(\ln 10)/\langle n_{\rm det}\rangle \simeq 5\times10^{36}\lambda_1 A\tau_p/J_1^2 H_0^2 L^2 . \quad (14)$$

The development of new powerful lasers is proceeding at a rapid pace, and a pulse energy of 1–10 MJ, pulse width of 10–20 ns, and repetition rate of 10 Hz,[8] as well as possible generation of a magnetic field $\sim 10^8$ G with a pulse duration of 10^{-9} sec and bore diameter of 0.1 cm using high-power lasers,[9] may be only a few years away. However, the intensity (or energy) required for the proposed experiment even with small N can be achieved using existing systems. A high-power pulsed Nd:glass laser with either $\lambda_1 \sim 0.35$ μm (NOVA[10]) or $\lambda_1 \sim 0.53$ μm (GEKKO XII[10]), $\tau_p \sim 10^{-9}$ sec, can provide a laser energy of 6–10 kJ/pulse in each of 10–12 beam lines. For this energy $\langle n_{\rm det}\rangle \simeq 1$ in each beam line, and the number of beam lines required to observe the effect for the normal polarization ($\theta_1 = \pi/2$, the worst case) even within a single laser shot is 4–5, which is therefore attainable. If all the beam lines are used, the probability p can be greatly reduced, i.e., $p \sim 10^{-6}-10^{-4}$. Most recently, great efforts have been made to increase both the output power (intensity) of very-short-pulse lasers and their repetition rate (which can be as high as 1–10 Hz). For XeCl,[11] Nd:glass,[12] and KrF (Ref. 13) lasers, $\langle n_{\rm det}\rangle \ll 1$ and the required number of pulses to observe the effect is $\sim 10^6-10^7$.

The generation of second-harmonic radiation caused by residual atoms and/or molecules in the laboratory vacuum system can mask the photon-photon scattering effect. In order to make an order-of-magnitude estimate of the contribution of these residual particles and to evaluate the vacuum pressure necessary to rule out nonvacuum components in the SHG, we presume that nonvacuum nonlinearity is mainly attributed to the plasma of magnetized free electrons, since at the required laser intensities, the residual gas is expected to be highly ionized. The nonmagnetized free electrons under the action of intense laser radiation can generate only odd-order higher harmonics; the second-order harmonics can only originate in the presence of the dc magnetic field. The numbers of second-harmonic photons per laser pulse, $N_{\rm SHG}$, for normal polarization can be obtained using[14]

$$N_{\rm SHG} = 2\pi e^4 a\omega_0^2 \tau_p E_1^4 n_e L/\omega_1^7 m_0^4 c^2 , \quad (15)$$

where n_e is the number of free electrons per unit volume and $\omega_0 = eH_0/m_0c$ is a cyclotron frequency of the electron. (It is worthwhile to note that for parallel polarization $N_{\rm SHG} = 0$.) Assuming that, on average, each molecule is singly ionized, and that the GEKKO XII laser ($\lambda_1 \sim 0.53$ μm) and the highest pulsed magnetic field ($\sim 8\times10^6$ G) are used, we estimate that vacuum and free-electron contributions to the SHG become equal at a pressure $\sim 2\times10^{-5}$ Torr. Since the vacuum provided by state-of-the-art vacuum technology is better than $\sim 10^{11}$ Torr, the free-electron nonlinear mechanism can be neglected. SHG may also be attributed to two-photon (and multiphoton, in general) excitations in ions of the residual gas; by selecting appropriate gas and frequency, this mechanism may also be made negligible. Since for the optical glass components (such as lenses and vacuum-chamber windows) the third-order nonlinearity is the lowest-order nonlinearity[15] in the absence of dc fields, SHG from those components can be eliminated by shielding them from the dc magnetic field. One of the ways to eliminate masking effects associated with possibility of SHG in laser amplifiers is to use sum-frequency effects (instead of the second harmonic) by employing two lasers with different frequencies. A more detailed evaluation of all these processes could be made only at the design stage of a particular experiment.

A large product H_0L could exist in may astronomical objects (e.g., in white dwarfs, where the spectral lines of elements still exist in the optical range[16]); the possibility exists that a second-harmonic signal generated by some characteristic spectral lines may be observed and used to study the nonlinearity of the vacuum and intrinsic properties of stars.

In conclusion, we have demonstrated the feasibility of second-harmonic generation by intense laser radiation in a vacuum which is due to photon-photon scattering in a dc magnetic field. The laser energy required to observe this effect can be achieved by using available high-power laser systems.

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