Highly efficient frequency tripling of laser radiation in a low-temperature laser-produced gaseous plasma

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Relatively efficient (up to 3%) third-harmonic generation of picosecond Nd:YAG laser pulses for wavelength $\lambda = 1.06$ μm in a low-density laser plasma is reported for a coherent (i.e., collinear) geometry. The third-harmonic beam produced is nearly diffraction limited and coherent and can be used in other nonlinear-optical experiments. A simple theoretical model is proposed in order to explain this result. In the model, third-harmonic generation is considered to be a process that arises from the scattering of electrons by ions in the presence of the strong laser field.

INTRODUCTION

Optical harmonic generation in plasmas has been studied for a long time. Usually this phenomenon is considered in relation to laser fusion,[1] where it leads to parasitic laser energy loss and hence is undesirable. Previous experiments were performed in order to investigate harmonic generation in a low-temperature low-density gaseous plasma, with the aim of achieving efficient nonresonant frequency upconversion. It was shown that moderately efficient frequency conversion is possible.[2–7] In these studies we developed a new approach to harmonic generation in a plasma. This approach involves (a) a plasma with a density no higher than 5×10^{19} cm^{-3}, which is transparent for visible and near-IR radiation; (b) Nd:YAG laser pulses of nanosecond and picosecond duration with moderate energies no higher than 200 mJ; (c) preparation of the plasma and harmonic generation, carried out with separate lasers and a variable time delay between the laser pulse used for spark initiation and the pulse used for harmonic generation. It was shown in Refs. 2–7 that the harmonics are coherent, i.e., diffraction limited, spectrally narrow band, and collinear with the incident beam.

In the present paper we report the relatively efficient (up to 3%) nonresonant third-harmonic generation (THG) of a picosecond Nd:YAG laser pulse with wavelength of 1.06 μm in a low-density laser plasma in a coherent (i.e., collinear) geometry. Thus conditions of our experiments differ significantly from those usually used for optical harmonic-generation experiments in dense, hot, thermonuclear laser plasmas. Our experiments produced relatively high THG efficiency and also produced a nearly diffraction-limited, coherent THG beam.

In these experiments the spark arose from the optical breakdown of an atmospheric-pressure gas near a metal surface; the breakdown was produced by a nanosecond Nd:YAG laser source with 250-mJ energy and a pulse duration of 15 nsec. The laser that was used for harmonic generation had a pulse duration of 40 psec and an energy not exceeding 50 mJ. The density of the plasma produced under these conditions is difficult to estimate accurately in this experiment, so we use published results[8,9] to obtain an approximate value. The plasma density in our experiments never exceeded 5×10^{19} cm^{-3} for the time delay used. This means that all radiation frequencies were much higher than the plasma oscillation frequency and that the medium was almost totally transparent for the optical pulses. A THG efficiency as high as 3% was achieved, determined as the ratio of the third-harmonic pulse energy to the incident pulse energy.

EXPERIMENT

The experimental configuration is shown in Fig. 1. The probing system that is used for THG consists of a passively mode-locked Nd:YAG oscillator, low-voltage single-pulse selection circuitry,[10] a variable optical delay line, and two Nd:YAG amplifiers. The pulse source had an energy of 50 mJ and a repetition rate of 1–2 Hz.

The laser, which produced the laser plasma in air, was synchronized to the picosecond laser source. The synchronization of these two lasers was achieved by triggering the Q switch of the nanosecond source by one of the first pulses of the picosecond train. One of the last pulses of that train was selected in the manner described in Ref. 11. The pulse was amplified by the two Nd:YAG stages and was used as a pump for THG. The nanosecond laser beam was focused by a cylindrical lens ($f = 10$ cm) on the surface of a metal target (Fig. 1), so the laser spark had a length of 8 mm along the direction of the probing picosecond beam. A significant difficulty was encountered in working with solid samples. An intense laser pulse produced a crater on the surface of the sample, so the conditions for plasma creation by subsequent pulses were essentially changed. To overcome this difficulty, we decided to use molten-metal targets. The best results were obtained with molten Sn and Pb, since these metals have low melting temperatures. However, it appears that THG efficiency in a plasma does not depend strongly on the target material. Third-harmonic light was generated in a transverse direction along the surface of the target. It was well collimated, with an almost diffraction-limited divergence, and was initially recorded by an optical multichannel analyzer that was placed after a double monochromator in order to ensure that the light was really

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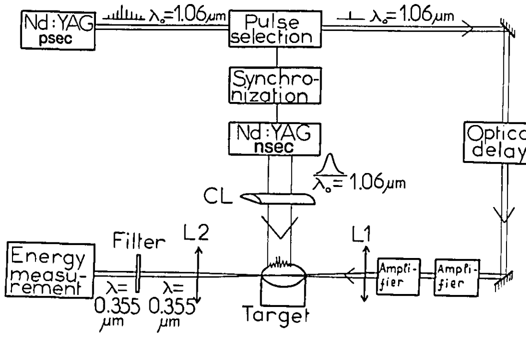

Fig. 1. Experimental setup for THG study.

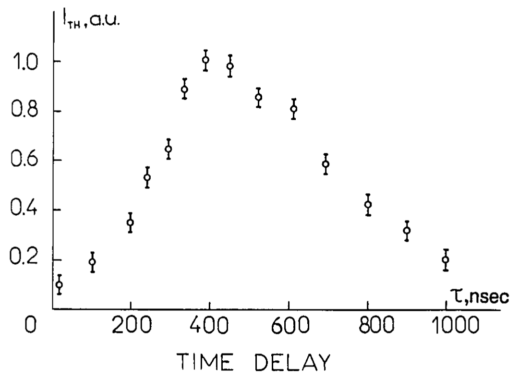

Fig. 2. Dependence of third-harmonic intensity on time delay τ between plasma formation and THG probing.

narrow band and had the expected frequency. In measurements of conversion efficiency the double monochromator was removed and the radiation was filtered by a set of colored-glass bandpass filters. We determined that these filters fully absorbed the pump radiation when the laser that produced breakdown was switched off. The optimization of the THG efficiency was achieved by varying the time delay τ and the displacement of the target in a direction perpendicular to the THG beam. The maximum THG efficiency achieved was 3%, at 400-nsec delay, for both Sn and Pb targets at a distance of the probing beam from the target of $\sim$500 μm. The value of the THG efficiency was measured by an absolute-energy meter and was corrected for the filter absorption. The energy of the third-harmonic pulse was measured to be $\sim$1 mJ after the filters.

It is not fully understood why the maximum efficiency occurred after such a long delay. This result could be attributed to an optimum population of high-lying ionic or atomic states of the plasma[12] or to a maximum recombination rate (see the theory below). Another explanation, which is being tested, predicts that at some time delay the plasma dispersion [governed by the dielectric function $\varepsilon(\omega) = 1 - \omega^2/\omega_P{}^2$] and the dispersion that is due to the populations of excited discrete atomic or ionic states compensate for each other. This result should lead to optimum phase matching for THG and to maximum third-harmonic intensity at that time delay.

The time dependence of the third-harmonic intensity is shown in Fig. 2. The shape of the implied curve and the maximum position strongly depend on the experimental conditions, especially on the energy of the pulse that produces the spark. This dependence could be explained by the hydrodynamical effects that govern plasma disintegration. We have also found that pump-beam absorption does not exceed 10% at time delays greater than 100 nsec.

We note that the THG in a plasma under self-breakdown of an atmospheric-pressure gas with picosecond laser pulses is 1–2 orders of magnitude less efficient than that reported above. In Table 1 the results of recent experiments are summarized. The experiments demonstrated that THG efficiency could be greatly improved by using laser pulses of shorter duration and by selecting the optimum time delay between excitation of the plasma and harmonic generation.

THEORETICAL MODELS

It was shown in Ref. 12 that the enhancement of THG and other four-photon processes in a low-density plasma produced by laser-induced breakdown could be attributed to the quasi-resonant enhancement of the third-order nonlinear susceptibility of the medium caused by population of highly excited atomic and ionic states (for details see Ref. 13). An alternative theoretical calculation of harmonic generation in a low-temperature collisional gaseous plasma involves using the simple classical model of a plasma.[14] In this model the optical nonlinearity is due to the dependence of the electron–ion collision frequency on their relative velocity and on the light-field strength (see also Ref. 15). In Ref. 14 it was predicted that in a nonstationary regime one could generate third-harmonic radiation with an efficiency as high as 1%. However, this prediction does not apply to the non-steady-state regime, where the basic equations of the theory are not valid. In addition, this theory cannot be applied to the description of harmonic generation of picosecond and femtosecond pulses in a low-density plasma when the total number of electron collisions during the pulse duration is equal to or smaller than one.

We do not discuss the specific nonlinearities of the hot, dense plasmas that are important in laser fusion (see, for example, Refs. 1 and 16–18) but are definitely not important in the case of a transparent, cold gaseous plasma. Further experimental and theoretical studies are necessary for us to determine which model of optical nonlinearity is appropriate for this work.

Another nonlinear mechanism is introduced here, which we believe to be a more appropriate description

Table 1. Experimental Data on Frequency-Tripling Conversion Efficiency in Plasmas

Ref. and Year	Plasma Preparation[a]	Pulse Energy (mJ)	Pulse Duration (psec)	Frequency-Tripling Efficiency ($W_{TH}/W_{1.06}$)
4, 1986	S	100	30000	10^{-10}
4, 1986	T	100	30000	10^{-8}
6, 1988	S	200	700	10^{-5}
1988[b]	S	40	40	10^{-3}
1988[b]	T	40	40	3×10^{-2}

[a] S, self-breakdown of atmospheric-pressure gas by the same laser pulse used for THG; T, breakdown of atmospheric-pressure gas near a metal target, initiated by an independent pulsed laser source.
[b] Our results.

of the nonlinear optical processes in a relatively low-temperature, low-density, gaseous plasma with picosecond and femtosecond laser pulses. In our approach the THG process is treated as a result of the scattering of electrons by a strongly nonlinear ionic potential in the presence of a strong laser field. We assume that the interaction of an optical electron with the light beam is stronger than that with the ion; thus the influence of the latter should be considered to be a perturbation.[19] The proposed method significantly differs from the one usually used for calculation of nonlinear susceptibilities of neutral atomic and condensed media. In the usual case the interaction of an optical electron with an ion or atom is considered to be much stronger than that with the laser field (see, for example, Ref. 20).

It should be emphasized that this theory is applicable to the important case of harmonic generation for extremely short picosecond and femtosecond pulses, since the theory is valid for the case of a single electron–ion interaction during the laser pulse duration.

The Schrödinger equation for an electron in an intense laser field $E(t) = E\sin(\omega t)$ and in an ionic potential can be written as follows:

$$i\hbar\frac{\partial\Psi}{\partial t} = (H_0 + V)\Psi, \tag{1}$$

where

$$H_0 = \frac{1}{2m}[\hbar k - \frac{e}{\omega}E\cos(\omega t)]^2$$

is the Hamiltonian of a free electron in a strong ac electromagnetic field, and $V = -e^2/r$ in the case of Coulomb potential and $V = -e^2/r\exp(-\alpha r)$ in the case of Debye potential, both being considered small perturbations.

The wave function of a free electron with the wave vector $\mathbf{k}$, in the field $E(t) = E\sin(\omega t)$ (Volkov solution), can be written in the following closed form[21]:

$$\varphi_k(\mathbf{r}) = \frac{1}{(2\pi)^{3/2}}\exp\left\{i\left[kr - \frac{\hbar\mathbf{k}^2}{2m}t + \frac{ekE}{m\omega^2}\sin(\omega t) - \frac{1}{4m\hbar}\left(\frac{eE}{\omega}\right)^2\left(\frac{\sin(2\omega t)}{2\omega} + t\right)\right]\right\}. \tag{2}$$

We assume that before the laser field is applied the electron's nonperturbed wave function can be described as a wave packet,

$$\psi_k{}^0 = \frac{(2\pi\alpha)^{3/2}}{\pi^{1/2}}\varphi_k\exp(-\alpha r), \tag{3}$$

which satisfies the following condition:

$$\int(\psi_k{}^0)^*\psi_k{}^0\mathrm{d}\mathbf{r} = 1, \tag{4}$$

where $l = \alpha^{-1}$ is a characteristic linear scale for localization. In the case of Debye screening, we take $\alpha = r_d{}^{-1}$, where $r_d{}^2 = kT_e/8\pi e^2 N_e$ (N_e is the electron number density of the plasma). The expectation value of the dipole moment is

$$\langle\mathbf{d}\rangle = \langle\psi_k{}^0 + \psi_k|e\mathbf{r}|\psi_k{}^0 + \psi_k\rangle. \tag{5}$$

Here ψ_k is the first-order term in the perturbation-theory series for the wave function.

This result for first-order perturbation theory gives the following expression for the component of the dipole moment, oscillating at the frequency $n\omega$, along the light-field vector $\mathbf{E}$:

$$|d^{n\omega}| = -\frac{2^7\alpha^4e^3}{\pi\hbar}\left|\int_0^\infty\sum_{s=1}^\infty(V_s^{(-)} + V_s^{(+)})\times\left(\frac{1}{\omega_{k'k} + s\omega} - \frac{1}{\omega_{k'k} - s\omega}\right)\frac{q^3\mathrm{d}q}{(\alpha^2 + q^2)^3(4\alpha^2 + q^2)}\right|, \tag{6}$$

where

$$q = k - k', \qquad \omega_{k'k} = \frac{\hbar}{2m}(k'^2 - k^2),$$

$$V_s^{(\pm)} = \frac{\left(\frac{eqE}{m\omega^2}\right)^{2s+n}}{2^{2s+n}(2s + n + 2)!(s + n)!}\,{}_3F_4\left[\frac{2s + n + 1}{2}, \frac{2s + n + 2}{2}, \frac{2s + n + 2}{2}; s + 1, s + n + 1, 2s + n + 1, \frac{2s + n + 4}{2}; -\left(\frac{eqE}{m\omega^2}\right)^2\right] \tag{7}$$

for $s > n$. For $s < n$ the expression for $V^{(+)}$ is still correct, but for $V^{(-)}$ it must be written as

$$V_s^{(-)} = \frac{(-1)^{n-s}\left(\frac{eqE}{m\omega^2}\right)^n}{2^n(n + 2)!(n - s)!}\,{}_3F_4\left[\frac{n + 1}{2}, \frac{n + 2}{2}, \frac{n + 2}{2}; s + 1, n - s + 1, n + 1, \frac{n + 4}{2}; -\left(\frac{eqE}{m\omega^2}\right)^2\right], \tag{8}$$

where ${}_3F_4(\alpha_3; \beta_4; z)$ is a hypergeometric function.

For an even n the component of the dipole moment along E is equal to zero.

Given that the dipole moment of the system oscillates at the third-harmonic frequency, we obtain the following expression for the intensity (compare with Ref. 22):

$$I_3 \sim N_e{}^2[d(3\omega)]^2|I|^2, \tag{9}$$

where the integral I accounts for phase matching. The phase-matching term can be written in the following form (see Ref. 22):

$$I(\Delta k, \xi, \zeta) = \int_\zeta^\xi \mathrm{d}\xi'\frac{\exp[\frac{1}{2}ib\Delta k(\xi - \xi')]}{(1 + i\xi')^2},$$
$$\Delta k = k_{3\omega} - 3k_\omega. \tag{10}$$

Here $b = 2\pi w_0{}^2/\lambda$, $\xi = 2(L - f)/b$, $\zeta = 2f/b$, w_0 is the pump beam radius, L is the length of the nonlinear medium, and f is the relative location of the focus in the nonlinear medium.

The refractive index of the plasma is given by the expression

$$n = 1 - \frac{\omega_p{}^2}{2\omega^2} \tag{11}$$

and is used to calculate Δk; $\omega_p^2 = 4\pi e^2 N_e/m$ is the plasma oscillation frequency. To estimate the dielectric function of the dynamic laser plasma, we should also take into account the effect of excited atoms and ions, which is important when time delays enable the free electrons to begin to recombine with the ions.

The above expressions enable us to estimate the third-harmonic intensity some tens of nanoseconds after the breakdown. For a 1.06-μm pump pulse with an intensity of 3×10^{13} W/cm^2 (corresponding to the focus of a single picosecond pulse with an energy of ~40 mJ) and an electron density of 10^{18} cm^{-3}, when we assume that the pump-beam focus is located in the middle of the nonlinear medium (i.e., $f = L/2$, $\xi = \zeta$), the THG efficiency is estimated to be ~0.5%. Optimum phase matching can occur at greater time delays, which can lead to even higher efficiencies, in agreement with the experiment. The tail in the THG time dependence (Fig. 2) is probably due to an enhanced third-order nonlinear optical susceptibility of excited atoms and is possibly due to ions with populated metastable states.[13]

CONCLUSIONS

It has been demonstrated that through the use of independent synchronized light sources, one for optical breakdown and the other for nonresonant THG, THG has been obtained in a laser-produced plasma with an unusually high efficiency (up to 3%). We hope that increasing the intensity of the pump wave will lead to still higher conversion efficiencies. However, the threshold and mechanism for saturation of this process are of great importance.

The observed THG efficiency is satisfactorily explained by a theory in which harmonic generation is considered to be the result of scattering of optical electrons by ions in the presence of a strong laser field.

The new approach to harmonic generation developed here could be of use in vacuum-UV spectroscopy, especially when UV (excimer, harmonics of Nd:YAG, etc.) picosecond or femtosecond laser pulses are used. With this technique relatively strong and broadly tunable pulsed vacuum UV radiation may be produced.

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