Quantum Electronics **30** (12) 1080 – 1082 (2000)

PACS numbers: 52.50.Jm; 52.70.Kz; 42.65.Ky
DOI: 10.1070/QE2000v030n12ABEH001868

Three-dimensional microscopy of laser-produced plasmas using third-harmonic generation

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Abstract. A three-dimensional microscopy of an optical-breakdown plasma based on third-harmonic generation (THG) is experimentally implemented. THG is shown to offer a universal method for three-dimensional microscopy of inhomogeneities of linear and nonlinear optical parameters of various media. Due to the nonlinear-optical character of this process, THG microscopy allows the transverse spatial resolution to be considerably improved as compared with conventional methods of microscopy. A high spatial resolution along the direction of probing is achieved due to the fact that the prohibition on THG in the regime of tight focusing is removed whenever inhomogeneities of linear and/or nonlinear optical parameters are probed by THG.

1. Introduction

The possibility of applying third-harmonic generation (THG) for a microscopy of biological objects has been actively discussed in several recent papers [1 – 4]. This approach is essentially based on the fact that the THG signal produced in a homogeneous infinite nonabsorbing medium with normal dispersion vanishes in the regime of tight focusing because of the destructive interference of third-harmonic waves generated in front of and behind the focal plane. Whenever pump radiation is focused on an interface between two media, the spatial symmetry of wave mixing is perturbed, which gives rise to a nonzero THG signal. This circumstance opens the way for a high-resolution microimaging of biological objects [3, 5].

The purpose of this paper is to demonstrate that THG not only allows imaging of interfaces between media with different refractive indices, but also offers a universal method for a microscopy of inhomogeneities in spatial distributions of linear optical characteristics (such as the absorption coefficient and the refractive index) and nonlinear optical parameters (nonlinear-optical susceptibility) of a medium. The results of our experiments demonstrate that THG provides an opportunity of three-dimensional plasma microscopy, permitting the imaging of plasma boundaries and visualisation of inhomogeneities in the spatial distribution of plasma species with a spatial resolution determined by the confocal parameter of the pump beam.

2. The main idea of the method

The expression for the power of the third harmonic generated in the field of a Gaussian pump beam is well known from textbooks on nonlinear optics [6]:

$$P_3 = \frac{768\pi^2 \left|\chi^{(3)}\right|^2}{n_1 n_3 \lambda_1^4 c^2} P_1^3 G, \tag{1}$$

where P_1, and λ_1 are the power and the wavelength of the pump wave in vacuum, respectively; n_1 and n_3 are the refractive indices at the frequencies of the pump wave and the third harmonic, respectively; $\chi^{(3)}$ is the third-order nonlinear-optical susceptibility;

$$G = \left| \int_{-2f/b}^{2(L-f)/b} \frac{\exp[-\mathrm{i}b\Delta k\xi/2]}{(1+\mathrm{i}\xi)^2} \mathrm{d}\xi \right|^2 \tag{2}$$

is the phase-matching integral; Δk is the phase mismatch for the THG process; L is the length of the nonlinear medium; f is the waist coordinate; and b is the confocal parameter of the pump beam.

Since the phase of a Gaussian beam reverses its sign at the focal point, the third-harmonic fields produced in front of and behind the focal point destructively interfere with each other. As a result, the THG signal produced in media with a normal dispersion vanishes in the regime of tight focusing ($L-f, f \gg b$). Inhomogeneities of the refractive index, absorption coefficient, or nonlinear-optical susceptibility around the focal area may change the regime of destructive interference, removing the prohibition on THG. The influence of phase-matching and absorption effects on harmonic generation in optical-breakdown plasmas has been previously studied both theoretically and experimentally (e.g., see [7]).

Let us examine in greater detail the case when a pump beam is tightly focused into a two-layer nonlinear-optical medium where the nonlinear-optical susceptibility changes in a stepwise way from zero up to some value χ on the interface between the layers, while all the other parameters remain constant within the entire volume of the medium. One can see from Eqns (1) and (2) that the power of the third

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Received 5 June 2000
Kvantovaya Elektronika **30** (11) 1080 – 1082 (2000)
Translated by A M Zheltikov

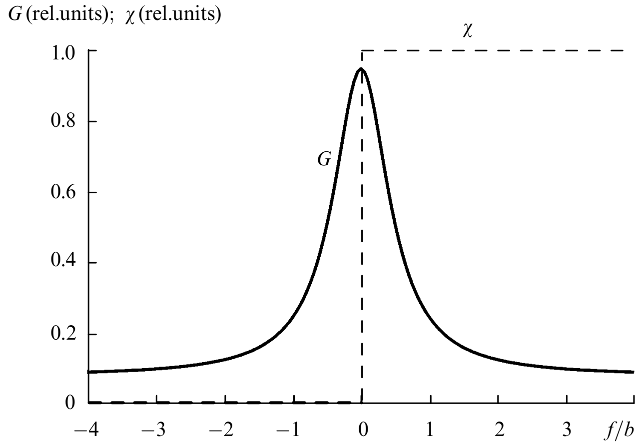

Figure 1. The phase-matching integral G and the nonlinear-optical susceptibility χ around the focal plane of the pump beam as functions of the focus position in THG with a confocal parameter equal to 0.2 cm and $\Delta k = 2$ cm^{-1} in atmospheric air under normal conditions for the wavelength of the pump beam equal to 1.064 μm.

harmonic is proportional to χ^2 in this case. A high amplitude of the THG signal can be achieved in such a situation by focusing the pump beam within an area adjacent to the interface between the layers. The size of this area, as can be seen from Fig. 1, is of the order of the confocal parameter of the pump beam.

Thus, in agreement with our intuitive expectations, THG microscopy allows the magnitude of an inhomogeneity in the nonlinear-optical susceptibility to be assessed and the position of such an inhomogeneity to be determined with a spatial resolution of the order of the confocal parameter of the pump beam. Note that, by virtue of the superposition of third-harmonic fields, the power of the THG signal remains the same as in the case considered above if we deal with a jump χ in the nonlinear-optical susceptibility of the medium from some nonzero level.

3. Experimental

We employed THG microscopy to image the plasma of optical breakdown induced by 15-ns pulses of the Nd:YAG-laser radiation with an energy of 150 mJ and a wavelength of 1.06 μm. To generate the third harmonic, we used 15-ns pulses of the Nd:YAG-laser radiation with an energy no higher than 10 mJ. Third-harmonic-generating laser pulses were synchronised in time with laser pulses used to produce the optical-breakdown plasma (Fig. 2). The delay time between these pulses in our experiments was equal to 3.5 μs. The diameter of the pump beam in the focal plane was equal to 12 μm, and the confocal parameter of the pump beam was about 300 μm.

The laser-produced plasma was probed along the x-axis (Fig. 2). To image the plasma, we scanned the laser plume with respect to a tightly focused pump beam in three coordinates with a 100-μm step in the vertical coordinate z, a 500-μm step in the longitudinal coordinate x, and a 100-μm step in the transverse coordinate y. The third-harmonic energy measured in this way was averaged over 50 laser pulses. A standard three-point smoothing procedure was then employed to plot grey-scale maps of the third-harmonic energy as a function of spatial coordinates.

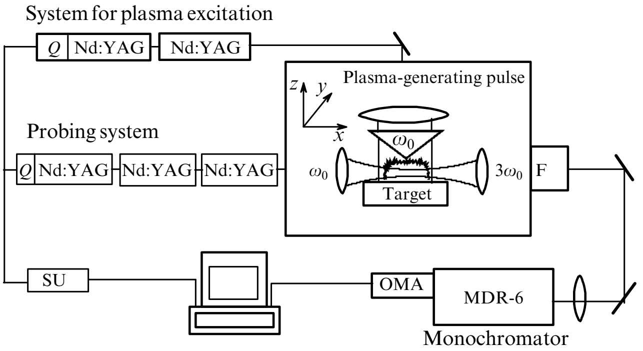

Figure 2. Diagram of the experimental setup for THG microscopy of a laser-produced plasma: OMA, optical multichannel analyser; F, filters; and SU, synchronisation unit.

4. Results and discussion

Fig. 3 presents two-dimensional maps of the spatial distribution of the third-harmonic energy for different plasma regions obtained by scanning the plasma plume with respect to a tightly focused pump beam in the x- and y-coordinates in a horizontal plane. The levels of grey scale represent the energy of the third harmonic in arbitrary units. The plasma of optical breakdown was produced on the surfaces of lead (Fig. 3a) and copper (Fig. 3b) targets. The maps of the third-harmonic energy presented in Fig. 3 were measured in a horizontal plane 1.1 mm (Fig. 3a) and 0.5 mm (Fig. 3b) above the target surface.

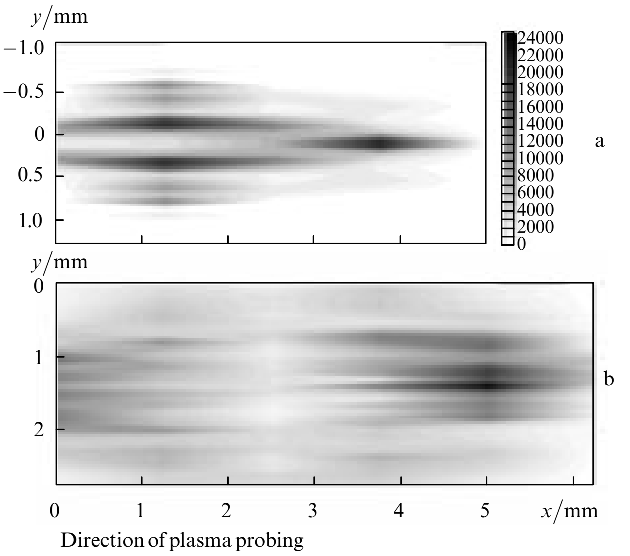

Figure 3. Two-dimensional maps of the spatial distribution of the third-harmonic energy in the plasma of optical breakdown produced on the surfaces of (a) lead 1.1 mm above the target surface and (b) copper 0.5 mm above the target surface.

The three-dimensional distribution of the third-harmonic energy measured with the use of the above-described procedure provides an information regarding the inhomogeneities in the density of plasma species inside the plume and the positions of plasma boundaries. In particular, darker areas in Fig. 3a distinctly visualise the boundaries of an excited gas in the case of a plasma produced on the surface of a lead target. As can be seen from the comparison of Figs. 3a and 3b, the boundaries of an excited gas in the case of a plasma produced on the surface of a lead target are much sharper than in the case of a copper target.

Even a brief look at Eqns (1) and (2) shows that it is rather difficult to extract the information concerning the spatial distribution of each of the linear and nonlinear optical parameters of a plasma, and it is unlikely that this problem can be solved in the general case. However, in several practically important situations, including the imaging of excited gas media and biological objects (see [1 – 5]), the ability of THG to visualise the spatial distribution of some combination of parameters of a medium seems to be extremely useful, as it gives a general idea of the spatial arrangement of objects inside a medium and reveals important general tendencies in spatial variations of parameters of such media. As can be seen from the results of our measurements, this approach even allows characteristic spatial scales of objects inside such media to be estimated.

Due to the nonlinear-optical character of THG, the spatial resolution of THG microscopy in the directions transverse to the direction of the pump beam may be made very high. In our experiments, this resolution was estimated as 12 μm. As demonstrated above, the spatial resolution of THG microscopy in the direction of probing is determined by the confocal parameter. This resolution was about 300 μm in our experiments. The results of measurements performed by means of THG microscopy agree well, with an accuracy of experimental errors, with the data extracted from emission spectroscopy, coherent four-wave mixing [8], and photoimages of a laser plume.

5. Conclusions

Thus, third-harmonic generation can be considered as a universal means for a three-dimensional microscopy of inhomogeneities of linear and nonlinear optical parameters. Due to the nonlinear-optical character of this process, THG microscopy allows the transverse spatial resolution to be considerably improved as compared with conventional methods of microscopy. A high spatial resolution of THG microscopy along the direction of probing is achieved due to the fact that inhomogeneities of linear and/or nonlinear optical parameters remove the prohibition on THG in the regime of tight focusing.

Acknowledgements. This study was supported in part by the President of Russian Federation Grant No. 00-15-99304, the Russian Foundation for Basic Research (Grant No. 00-02-17567), INTAS Grant No. 97-0369, and the 'Fundamental Metrology' and 'Fundamental Spectroscopy' Federal Research Programs of the Russian Federation.

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