Quantum Electronics **47** (3) 245–247 (2017)

https://doi.org/10.1070/QEL16308

A method for measuring the nonlinear refractive index from the contrast of an amplitude object image in the optical scheme with phase Zernike filters based on electronic Kerr nonlinearity

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Abstract. **We propose a method for determining the nonlinear refractive index from the image contrast of amplitude objects in the phase-contrast scheme with a sample acting as a phase Zernike filter based on the electronic Kerr nonlinearity. This method implies the measurement of the calibration dependence of the contrast of an amplitude object image on the laser pulse energy for a test sample with a known nonlinearity and makes it possible to find the nonlinear refractive index of samples under study using a single laser pulse.**

Keywords: *Kerr nonlinearity, nonlinear refractive index, phase contrast, nonlinear Zernike filter.*

1. Introduction

The change in the refractive index of optical materials exposed to high-power laser radiation is one of the most actively studied nonlinear optical effects, which is of fundamental importance for laser and fibre optics [1]. The modern methods for measuring nonlinear optical coefficients of laser materials are based on the change in the energy [2, 3], polarisation state [4], spectrum [5], or wavefront [6] of a probe laser beam that are caused by the nonlinear response of the medium. Four-wave mixing methods [2, 3] use the Bragg scattering of a probe laser beam from the nonlinear refractive index grating formed by two pump laser beams. In the polarisation methods, the nonlinearity of the refractive index of the medium causes self-rotation of the polarisation ellipse [4] or, in the presence of the second laser beam, optical sampling effect [7]. Spectral methods are based on measuring the spectral broadening [8] and the shifts of the centre frequency [5] and spectral phase [9–11] of laser radiation. The methods implying analysis of the nonlinear phase shift and wavefront distortion include measurements of the wavefront by a Shack–Hartmann wavefront sensor [12], self-focusing threshold [13], and the distribution of laser beam intensity in the far-field zone [6]. The methods of spatial interferometry [14–16] and various techniques of z-scan measurements [17–24] and nonlinear imaging [25–27] are also widely used.

In this study, we have developed a method for measuring the nonlinear refractive index of optical materials according to the scheme of spatial nonlinear Zernike-type filtering [25]. In contrast to the previous studies [24–26], an amplitude object was chosen for visualisation.

2. Experimental

The radiation source was a laser system based on Ti: sapphire crystals, generating pulses with a duration $\tau \approx 70$ fs and a centre wavelength $\lambda_0 \approx 800$ nm. The energy W of the laser pulses incident on the sample did not exceed 100 μJ in our experiments. It was controlled by a polarisation attenuator. The laser radiation (Fig. 1) was partially directed to a calibrated

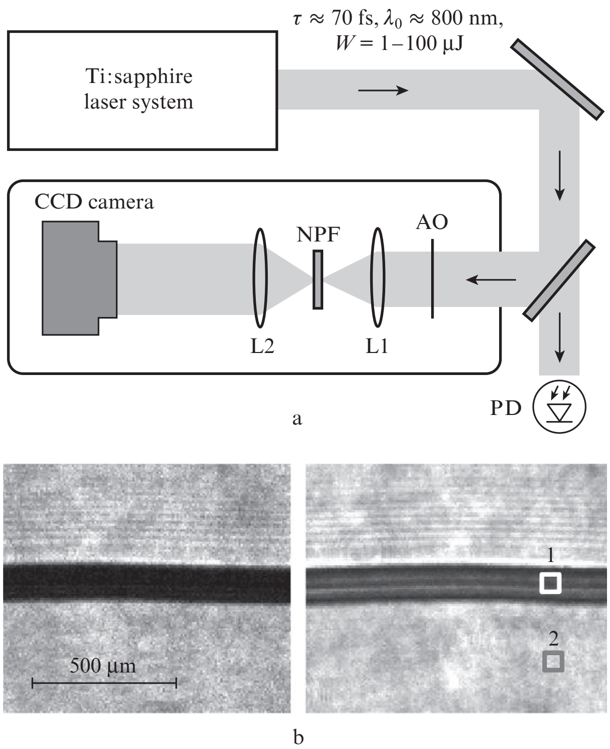

Figure 1. (a) Schematic of the experiment and (b) a wire image in the linear (low laser power, on the left) and nonlinear (on the right) regimes: (L1, L2) achromatic lenses with focal lengths of 15 and 25 cm, respectively; (AO) amplitude object (a wire 140 μm in diameter); (NPF) nonlinear phase filter; (PD) calibrated photodiode; (1, 2) areas from which signals were recorded to determine the image contrast as the signal ratio.

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Received 6 February 2017
Kvantovaya Elektronika **47** (3) 245–247 (2017)
Translated by Yu.P. Sin'kov

photodiode to monitor the pulse energy. The rest of the radiation arrived at a two-lens confocal imaging system, the object plane of which contained an amplitude object (a long metal wire 140 μm in diameter, passing through the entire beam cross section). The laser beam intensity profile was close to Gaussian, and the beam diameter was 13 mm (at a level of e^{-2}). The imaging system consisted of two achromatic lenses, L1 and L2, with a diameter of 50 mm and focal lengths of 15 and 25 cm, respectively; the lenses were spaced by 40 cm. The wire image was recorded using a CCD camera (Pixelfly, PCO). Measurements were performed in two regimes: linear (at a low laser power) and nonlinear (Fig. 1b).

To implement the method of adaptive phase contrast, either a thin (1-mm-thick) fused silica plate 50 mm in diameter (a test sample) or a 2-mm-thick chalcogenide glass plate (composition $Ge_{25}S_{65}I_{10}$, refractive index $n = 2.1$ [28]), whose nonlinear refractive index was intended to be measured, was installed in the focal plane of the first lens of the imaging system; these objects served as nonlinear Zernike filters. The laser-beam radius at the point of location of the samples acting as a nonlinear filter was measured by an auxiliary CCD camera to be 11 μm (at a level of e^{-2}), a value exceeding the size of the diffraction spot in the lens focus by a factor of 1.8.

3. Results and discussion

In the conventional phase-contrast method, a phase plate having sizes approximately corresponding to the beam size in the focus and introducing an additional phase shift of light waves on the order of $\sim\pi/2$, was installed in the focus of the first lens of the imaging system to visualise phase objects. The 'inverted' phase-contrast scheme implemented in [27] uses an object with a known phase, and its image yields information about the nonlinear phase gained by the sample placed in the focus of the first lens of the imaging system. In this study, we developed an inverted phase-contrast method with a nonlinear phase Zernike filter, using an amplitude object instead of a phase one to measure the unknown nonlinear refractive index of the chalcogenide glass sample under study. The basic concept of this method is as follows: the dependence of the contrast of an amplitude object image on the laser beam intensity is measured for two samples placed one by one in the focal plane of the imaging system: one sample with a known nonlinear refractive index (fused silica plate as a test sample) and the other with a nonlinear refractive index to be determined (chalcogenide glass). The object image contrast will be defined as the ratio of the image brightnesses in the object area (area 1 in Fig. 1b) and beyond it (area 2). The image contrast is determined by the nonlinear phase gained by the samples [25]. Therefore, the same image contrast for the two samples is obtained at the same nonlinear phase. Having equated the nonlinear phase shifts corresponding to identical image contrast in the dependences of the object image contrast on the laser beam intensity for the two samples, one obtains the expression for the unknown nonlinear refractive index $n_2^{\rm chg}$ of the chalcogenide sample:

$$n_2^{\rm chg} = n_2^{\rm q}\left(\frac{l_{\rm q}}{l_{\rm chg}}\right)\left(\frac{I_{\rm q}}{I_{\rm chg}}\right),$$

where $I_{\rm q}$ and $I_{\rm chg}$ are, respectively, the laser beam intensities for the fused silica plate and sample under study (chalcogenide glass) at which the same image contrast is obtained; $n_2^{\rm q}$ is the nonlinear refractive index of the test fused silica sample; and $l_{\rm q}$ and $l_{\rm chg}$ are the corresponding thicknesses of the two samples.

The experimental dependences of the image contrast of an opaque amplitude object (wire) on the laser beam intensity for the two samples, fused silica plate and chalcogenide glass, are shown in Fig. 2. The dependence of the image contrast for chalcogenide glass coincides with a high accuracy with that for the fused silica sample (when the latter dependence is shifted along the intensity axis in correspondence with the scaling factor $I_{\rm q}/I_{\rm chg} \approx 68$).

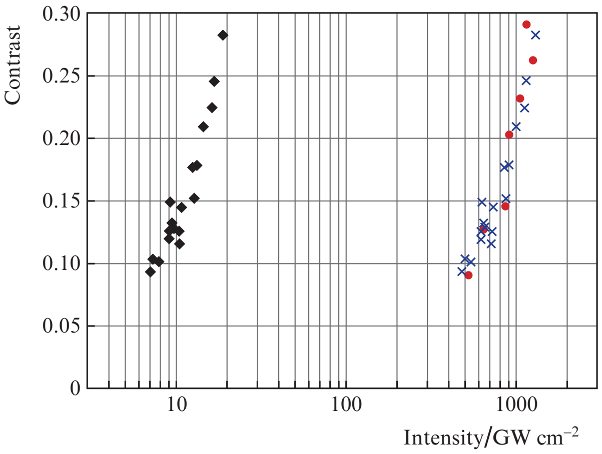

Figure 2. Dependences of the amplitude object image contrast on the laser beam intensity for (◆) chalcogenide glass, (●) fused silica, and (×) chalcogenide glass with the scaling factor $I_{\rm q}/I_{\rm chg} = 68$.

The scaling factor for the intensity is the same for the entire range of the image contrast values under study (Fig. 2). This fact suggests that the nonlinear refractive index of the sample in question can be determined with a single laser pulse if the calibration dependence of the contrast of the amplitude object image for the test sample is already obtained (circles in Fig. 2). Thus, being supplemented with preliminary calibration, the proposed method is single-pulse and makes it possible to measure promptly the nonlinear refractive index. Assuming the nonlinear refractive index of fused silica to be 3.2×10^{-16} cm^2 W^{-1} [1] and taking into account the thicknesses of the samples, we find the nonlinear refractive index of chalcogenide glass $Ge_{25}S_{65}I_{10}$: $\sim1.1\times10^{-14}$ cm^2 W^{-1}. This value is in agreement with the results of [28], where measurements were performed with allowance for the change in the laser beam intensity distribution in the far-field zone due to the nonlinear phase shift in the sample [6]. Note that the maximum shift of the nonlinear phase in the samples for the image contrast values under study does not exceed 2.5 rad and should not lead to any significant beam distortions in the samples as a result of self-focusing.

Thus, we proposed and implemented a method for measuring the nonlinear refractive index of optical samples, which is based on nonlinear modification of the phase-contrast Zernike method (using femtosecond laser pulses and electronic Kerr nonlinearity). Having compared the contrasts of amplitude object images obtained with a test sample and a sample under study, we measured the nonlinear refractive index of a chalcogenide glass ($Ge_{25}S_{65}I_{10}$) sample. This method provides quantitative information about the nonlinear refractive index using a single laser pulse.

Acknowledgements. This work was supported (in the part concerning the development of the method for measuring the nonlinear properties of chalcogenide samples) by the Russian Science Foundation (Grant No. 15-19-00147), the Russian Foundation for Basic Research (Grant No. 15-02-08212), and the Presidium of the Russian Academy of Sciences (Programme 'Extreme laser radiation: physics and fundamental applications').

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