Direct observation of below-diffraction-limited optical spot induced by nonlinear saturable absorption of Ag-doped Si nanofilms

Jingsong Wei* and Jing Liu
Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, No. 390, Qinghe Road, Jiading District, Shanghai 201800, China
*Corresponding author: weijingsong@siom.ac.cn

Received May 24, 2010; revised August 27, 2010; accepted August 27, 2010; posted August 30, 2010 (Doc. ID 128834); published September 15, 2010

In this work, using the scanning near-field optical fiber probe method, we carry out a direct observation of below-diffraction-limited optical spot induced by nonlinear saturable absorption characteristic of Ag-doped Si nanofilms. The experimental results indicate that the squeezed spot size decreases with laser power increase, and the smallest spot can be squeezed to 68% of the diffraction-limited focusing spot size, which is very useful and can be applied to superresolution optical recording, optical lithography, and optical imaging. © 2010 Optical Society of America

OCIS codes: 190.4360, 260.5950.

To break through the limit, in past years, researchers proposed all kinds of techniques and methods, such as a near-field optical probe [1,2], a solid immersion lens [3], a negative refraction lens [4], a surface plasmon superlens [5], binary optics [6], a grating substrate method [7], and selective excitation [8]. Recently, optical nonlinearity-induced superresolution has become a hot subject owing to its excellent technical applicability in high-density optical memory [9], subwavelength optical lithography [10], and superresolution scanning laser microscopy [11]. Song et al., Choi et al., and Helseth gave a direct observation and description of self-focusing super resolution [12–14]. We proposed a self-focusing thermal lens model [15] and an internal multi-interference super resolution [16]. In this work, using the scanning near-field optical fiber probe method, we carry out a direct observation of below-diffraction-limited optical spot induced by nonlinear saturable absorption characteristic Ag-doped Si nanofilms. The results indicate that the spot can be squeezed to about 68% of the original focusing spot size, which is very useful for superresolution optical recording, optical lithography, and optical imaging.

The Ag-doped Si thin films were prepared by magnetron-controlled cosputtering of an Si target and an Ag target. The backscattering image of scanning electronic microscopy of Ag-doped Si thin film (not shown here) indicates that the Ag particles are homogeneously distributed in the Si matrix, and the particle size ranges from several nanometers to tens of nanometers. The nonlinear properties were measured by a z-scan method, where a He–Ne laser with a wavelength (\(\lambda\)) of 632.8 nm was used.

In this work, the laser pulse width was fixed at 50 ns, and a converging lens with an NA of 0.09 was used to conduct the z-scan measurement. The laser power \(P\) was fixed at 2.5 mW. The laser intensity can be calculated by \(I_0 = 2P/\pi w_0^2 = 8.625 \times 10^7 \text{W/m}^2\), accordingly, where \(w_0 = (1.22\lambda/2 \text{NA})\) is the beam waist radius. The measured results are shown in Fig. 1. One can see that the thin film presents an obvious nonlinear saturable absorption characteristic; in other words, the larger the transmittance, the higher the laser intensity. The theoretical fitting indicates that the nonlinear saturable absorption coefficient \(\beta\) is about −0.024, which is a very large value compared with general nonlinear absorption materials. The internal physical mechanism may be from the combination of both the localized surface plasmons resonance and scattering enhancement effects of Ag particles and excited-state absorption of the semiconductor Si matrix. Here, it needs to be pointed out that no nonlinear refraction effect occurs in the measurement, which may be because the nonlinear refraction signal is too weak to be detected.

To understand the nonlinear saturable absorption-induced superresolution effect, we first prepared the sample structure as “Glass substrate with a thickness of 1.2 mm/Ag-doped Si thin film with a thickness of 20 nm.” The diffraction-limited focusing spot with a Gaussian intensity profile passes through the glass substrate and irradiates the nonlinear saturable absorption layer surface, as shown in Fig. 2. For nonlinear saturable absorption materials, the higher the laser beam intensity, the larger the transmittance. Thus, there is the largest transmittance in the central part of spot, which causes...
a small channel to be formed in the center of the spot. The laser beam passes through the channel, and a below-diffraction-limited optical spot will be generated at the surface of the Ag-doped Si thin film. The reduction degree of the spot can be tuned by changing the laser intensity. A near-field optical spot scanning setup was established to directly observe the spot, where a laser diode at a wavelength of 635 nm was applied as the light source, and the converging lens with an NA of about 0.55 was used to focus the spot (as shown in Fig. 2). In this setup, a higher magnification converging lens is not adopted, because the working distance of the higher converging lens is shorter than the thickness of the glass substrate. The theoretical FWHM of the diffraction-limited spot can be calculated by \( D = \frac{0.61\lambda}{NA} \). For our setup, the FWHM is about 0.704 \( \mu \)m. The laser beam passed through the glass substrate and was focused on the Ag-doped Si thin film. After the thin film, a metal-coated fiber tip affiliated onto the tuning fork was used to scan the laser beam spot intensity distribution. The tuning fork was attached on a three-dimensional piezoelectric transducer (PZT). The lateral maximum movement distance of the PZT was 60 \( \mu \)m, and the lateral scanning resolution was about 100 nm, limited by the aperture of the fiber probe. The distance between the sample surface and the fiber tip was modulated by an atomic force microscopy controller; when the fiber tip got close to the sample surface, the resonance frequency of the tuning fork would shift owing to the generated shear force, and the controller received this shift signal as error and regulated the \( z \)-axis voltage onto the PZT. The spot intensity profile information was collected by the scanning fiber tip, was transferred to the photodiode, and produced the signal in the form of voltage to the computer.

We first scanned the diffraction-limited focusing spot through the glass substrate with a thickness of 1.2 mm, and the result is shown in Fig. 3(a). It can be seen that the focusing spot is an elliptical shape, and the FWHM of the short axis is about 1.48 \( \mu \)m. This size is larger than the theoretical spot of the setup itself, which may result from three aspects. The first is that the glass substrate (1.2 mm thickness) affects the light path due to the refraction; the second is that the semiconductor diode laser beam is not collimated and has a large divergence angle; the third is that the setup may be not very perfect [for example, the
real NA of the converging lens is a little smaller than theoretical NA (0.55). However, we focus mainly on the squeezed spot size ratio; thus, the theoretical spot size of the setup has little effect on our experimental analysis. In addition, the formation of the elliptical spot could be experienced due to the laser diode’s junction. It is also possible to avoid this elliptical spot by overfilling the back aperture of the converging lens. However, the effective laser power will be decreased greatly, which will have an ill effect on the direct observation of spot reduction. Then, we replaced the glass substrate with “Glass substrate with a thickness of 1.2 mm/Ag-doped Si thin film with a thickness of 20 nm” and scanned the spot along the Ag-doped Si thin film surface; the laser power was fixed at about 11.5 mW. The spot intensity profile is presented in Fig. 3(b), and one can find that the spot shape is still elliptical. Whether at the short-axis direction or at the long-axis direction, the spot size is obviously squeezed, i.e., the below-diffraction-limited spot is generated by the Ag-doped Si thin film due to the nonlinear saturable absorption effect. To compare effectively, the cross-section distribution curves of Figs. 3(a) and 3(b) at the short-axis direction (marked in a green color line) are plotted in Fig. 3(c). It can be seen that the FWHM of the short-axis size of the squeezed spot is reduced to about 1.12 μm, which is about 75% FWHM of the real diffraction-limited spot size. The squeezed spot size ratio is about 0.75, where the squeezed spot size ratio is defined as the ratio of squeezed spot size to the real diffraction-limited focusing spot size.

By changing the laser power, we can tune the squeezed spot size as well; the results are given in Fig. 4. The results indicate that the squeezed spot ratio decays exponentially with increasing laser power. The exponential decay seems to accord with the Beer–Lambert law $I_t = I_0 \exp(-\alpha d)$, where $I_t$ is the transmitted spot intensity, $\alpha = (\alpha_0 + \beta I_0)$ is the absorption coefficient, $\alpha_0$ is the linear absorption coefficient, and $I_0$ is proportional to laser power. That is, the reduction degree of the squeezed spot size increases with laser power; and the smaller the spot size, the higher the laser power. However, we see from Fig. 4 that the spot is not squeezed until the laser power increases to about 11 mW. In other words, there is an abrupt reduction of spot size at a laser power of 11 mW, which may be due to the nonlinearity excitation with a threshold effect. The detailed reason is not clear so far and will be further discussed in the near future. In addition the spot cannot be squeezed infinitely, because it is impossible to infinitely increase the laser power due to strong laser intensity, leading to damage of the materials. In our experiment, the smallest spot can be squeezed to 68% of the real diffraction-limited focusing spot size.

In summary, Ag-doped Si nanofilms with a nonlinear saturable absorption coefficient of $\beta = -0.024$ are obtained. The nonlinear saturable absorption induced below-diffraction-limited spot is directly observed by the near-field scanning optical fiber probe method. The results indicate that the squeezed spot size decreases with increasing laser power, and the smallest spot can be squeezed to 68% of the real diffraction-limited focusing spot size.

The work is partially supported by the National Natural Science Foundation of China (NSFC) (50772120, 60977004), the Shanghai Rising Star Tracking Program (10QH1402700), and the National Basic Research Program of China (2007CB935400).

References