Visible Optical Limiting Property of Germano-Silicate Glass Fibers with Au Nanoparticles Incorporated

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In investigating with a continuous-wave 488-nm argon-ion laser, we firstly report on the visible optical limiting properties of germano-silicate optical fibers with gold nanoparticles incorporated. The ground-state conduction, the surface plasmon band and the free carrier band were considered as a three-level model to theoretically explain the obtained optical limiting behaviors. In the visible wavelength region around the surface plasmon resonance absorption peak of the gold nanoparticles, the effective nonlinear absorption coefficient was estimated to be 22.4 cm/GW.

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I. INTRODUCTION

There is an increasing interest in synthesizing new materials possessing nonlinear optical properties. In particular, optical limiters in the visible wavelength domain with a nonlinear spectrum and with fast response time are required for eye or optical sensor protection and switching applications. An ideal optical limiter exhibits a linear transmission below a threshold and clamps the output to a constant above it [1, 2]. Therefore, candidates for optical limiting materials should have high transmittance for weak incident light, low transmittance for strong incident light and instantaneous response over a broadband spectral range [3].

Among all the candidates, metal nanoparticles (NPs), such as silver, gold, cobalt, platinum and copper NPs dispersed in various insulator matrices exhibiting a wide variety of nonlinear optical properties, have attracted attention as potential nonlinear optical devices because of their unique linear and nonlinear optical characteristics [4-6], distinguishing optical limiting properties that arise from the quantum size effects and surface plasmon resonance (SPR) effects [7-9], high polarizability and ultrafast nonlinear response [10, 11]. The most conspicuous manifestation of confinement in the optical properties of metal NPs is the appearance of a morphological resonance, the SPR effects that strongly enhance their linear and nonlinear responses around specific wavelengths [11].

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In this paper, we firstly report on the optical limiting behaviors of Au NPs incorporated in germano-silicate glass fibers having SPR absorption band positions in the visible wavelength region. The results demonstrated that the effective nonlinear absorption coefficient around the SPR absorption peak of the gold-NP incorporated fiber was almost ten times that of a reference fiber without any dopant and that at the same time, the optical limiting threshold remarkably decreased. The ground-state conduction, the surface plasmon band and the free carrier band were considered as a three-level model to theoretically explain the obtained optical limiting behaviors of the fibers.

II. RESULTS AND DISCUSSION

A germano-silicate glass fiber incorporated with Au NPs (Au fiber) and a reference fiber without any dopant, but with the same recipes, were fabricated using the modified chemical vapor deposition (MCVD) technique and solution doping process, which is described in detail in Refs. 4-6. Since the structure of the fiber (8 - 10-μm-diameter core surrounded by a 123-μm-diameter outside cladding) and an extremely low dopant concentration of ppm level make the Au NPs difficult to detect by direct measurement methods, such as transmission electron microscopy (TEM) and X-ray diffraction (XRD), the cut-back method was used to measure the absorption spectrum of the optical fiber to confirm the
formation and existence of the Au NPs in the core of the fiber [5]. The optical limiting measurement was carried out by measuring the output power of the argon-ion laser (Spectra-Physics, Stabilite 2017) at 488 nm coupled into the fiber with a 3-dB coupler, as shown in Figure 1 of Ref. 5. The continuous wave quasi-collimated light from the 488-nm argon-ion laser was used in this measurement. At the same time, a power controller was used to measure the feedback power from the other arm of the given coupler to determine the actual input power into the fiber under test. For accurate comparison, the optical limiting properties were measured with 30-m-long fibers and the fibers were twisted with an 8-cm radius to remove possible bending loss.

The linear absorption spectrum of the Au fiber is compared with that of the reference fiber in Figure 1. A SPR absorption peak centered at 498.4 nm appeared in the Au fiber and this clearly indicates the existence of the Au NPs inside the fiber core [5, 6]. Compared with the other reported Au-NP incorporated samples [9, 12–14], the SPR peaks were in the range of 495 ~ 500 nm and the average radius of the doped Au NPs was estimated to be 2.6 ~ 5 nm.

The optical limiting properties of the Au fiber and the reference fiber were measured with the fundamental output of an argon-ion laser as the light source. The transmitted intensity increased nonlinearly with the incident intensity and the experimental results are shown in Figure 2. Taking the wavelength of 488 nm as an example, we show the experimental results in Figures 2(a) and (b) for the reference fiber and the Au fiber, respectively. As shown in Figure 2, both fibers exhibit a deviation of the output intensity from linearity and the differences between the ideal linear output intensity and the actual output intensity indicate the corresponding optical limiting properties.

It is important and interesting to note that the reference fiber without Au-NP incorporation also showed an observable optical limiting behavior when it was pumped with the 488-nm argon-ion laser. The two-photon absorption (TPA) with a maximum at around 480 nm, which corresponds to twice the wavelength of the intense absorption band due to the diamagnetic germanium-related defects, is responsible for the observed optical limiting properties of the reference fiber [15, 16].

Since the effective medium theories can be applied to the case of individual clusters or NP incorporated matter with extremely low filling factors and to the case of $d/\lambda < 10^{-2}$, where $d$ is the average diameter of the doped NPs and $\lambda$ is the input signal wavelength [10], we simplified the optical limiting process by assuming the Au fiber to be a homogeneous cluster. Thus, saturation and nonlinear absorption [17], nonlinear refraction and nonlinear scattering effects from the doped-Au NPs were neglected. Note that the average diameter of the Au NPs at a ppm-level concentration inside the core of the Au fiber was about 2.6 ~ 5 nm.

Several mechanisms may contribute to the third-order nonlinearities in small metal NPs, including SPR enhancement, intraband transitions in the quantum confined conduction band (quantum size effect), interband
Table 1. Optical limiting parameters of the reference fiber and the Au fiber measured at 488 nm.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>α</th>
<th>a</th>
<th>λc</th>
<th>A_{eff}</th>
<th>I_t</th>
<th>T_L</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>reference fiber</td>
<td>cm^{-1}</td>
<td>0.0008</td>
<td>9.04</td>
<td>1180</td>
<td>68.0</td>
<td>1.18</td>
<td>1.62</td>
<td>2.50</td>
</tr>
<tr>
<td>Au fiber</td>
<td>μm^2</td>
<td>0.0022</td>
<td>8.94</td>
<td>1159</td>
<td>63.2</td>
<td>0.68</td>
<td>0.59</td>
<td>22.4</td>
</tr>
</tbody>
</table>

a: core radius; λc: cut-off wavelength.

Fig. 3. Energy-level diagram of Au NPs explaining the process leading to the observed optical limiting processes of the Au fiber.

In summary, we have presented an applicable optical limiting germano-silicate glass fiber incorporated with Au nanoparticles. The strong effective nonlinear absorption coefficient of the Au-nanoparticle-incorporated germano-silicate glass fiber was demonstrated and was estimated to be 22.4 cm/GW, and a low optical limiting threshold of 0.68 GW/m^2 was obtained, showing the obvious advantages over the reference fiber without any dopant. Because of the obtained strong nonlinear absorption coefficient, we expect Au-nanoparticle-incorporated germano-silicate glass fibers to exhibit better optical limiting properties over the full range of visible wavelengths covering from 400 nm to 700 nm if the size distribution and diverse shapes of the gold nanoparticles inside the fiber core can be optimized, leading to a good optical limiter.

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REFERENCES