Raman Gain in Transparent Nanostructured Glass-Ceramic

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Abstract. Stimulated Raman scattering in transparent glass-ceramics (TGCs) based on bulk nucleating phase Ba2NaNb5O15 were investigated with the aim to explore the influence of micro- and nanoscale structural transformations on Raman gain. TGCs are composed of nanocrystals that are 10–15 nm in size, uniformly distributed in the residual glass matrix. A significant Raman gain improvement for both BaNaNS glass and TGCs with respect to SiO2 glass is demonstrated, which can be clearly related to the nanostructuring process.

1 Introduction

The SRS phenomenon occurs in the presence of high energy transfer from a high power pump beam to a probe beam (copropagating or counterpropagating) [1]. SRS depends on the pump intensity and on the gain coefficient, which is proportional to the spontaneous Raman scattering cross section and inversely proportional to the linewidth of the corresponding Raman line. Because of its coherent nature, the molecular bonds oscillate in the phase and interfere constructively inside the focus area of the laser beam. As a consequence, the SRS signal, which is orders of magnitude bigger than that of spontaneous Raman scattering, is generated. Due to its Raman-shifted output, SRS is a workable method for generating coherent radiation at new frequencies. SRS permits, in principle, the amplification of wavelengths in a wide interval, from the ultraviolet to the infrared ones [2]. Since the Raman frequency of a medium is usually fixed, the tunability can be achieved by using tunable pump laser.
Important accomplishments related to integrated laser sources based on SRS have been achieved in the last two decades in the fields of photonics [3], microphotonics [4], and nanophotonics [5]. In the investigation of SRS at the nanoscale, how it often happens for nonlinear optics phenomena and fundamental and applicative issues are two faces of the same coin. Concerning the fundamental ones, there have been several investigations, both experimental [6] and theoretical [7] ones, but the "question is still open," while from an applicative point of view, there are some important perspectives to realizing micro-/nano-sources with improved performances. Although a general theory on the relationship between nanostructuring and Raman gain has not been established, we expect that when the particle dimensions are of a few nanometers, the phonon confinement effect plays a significant role; therefore, SRS enhancement should be attributed to the quantum confinement effect, and the gain of nanomaterials should be different from bulk and related to the intrinsic properties of materials [8].
Among the nonlinear nanostructured materials, which are generally formed by an active material (rare-earth ions, quantum dots, metal clusters or nanoparticles, and organic and carbon-related materials) uniformly dispersed in a host matrix, inorganic glasses and transparent glass-ceramics (TGCs) play a key role due to their chemical stability and their linear and nonlinear optical features, which can be finely tuned by changing the chemical composition and applying suitable heat treatments [9]. In a glassy matrix, structural transformation at the nanoscale can occur by phase separation when it takes place as a binodal mechanism. In this case, the development of the new liquid phase occurs by nucleation and the subsequent growth of nuclei into droplets, whose size may be controlled by a proper heat treatment [10]. Depending on the composition of the initial glass and the heat treatment temperature, this process produces either two amorphous phases (amorphous nanostructuring) or the growth of a crystalline phase uniformly dispersed in
the amorphous phase (crystalline nanostructuring), thus producing TGCs [11, 12].
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2. Raman Characterization

Concerning the Raman gain, it is worth noting that for several materials, for example silicon, a direct measurement is possible. However, when the Raman gain is not so high and the length of the sample is small, an indirect measurement must be utilized.

Usually, this happens, for example for glasses; in this case, the Raman gain is estimated by a numerical procedure starting from experimental data provided by spontaneous Raman scattering [13].

The Raman gain spectra of investigated samples normalized to the one of silica were calculated and are reported in Figure 1. For the initial glass, the value of Raman gain peak was evaluated to be 7.5 times that of silica. It increases up to 15 and 40 for the samples obtained from heating program (3) and the (1) and (2), respectively. Noticeable modifications in the Raman spectra and a significant enhancement of Raman gain (up to 40 times higher) of both the BaNaNS glasses and annealed samples with respect to initial one and to SiO2 glass due to nanostructural transformation that occurred during the thermal treatment are demonstrated.

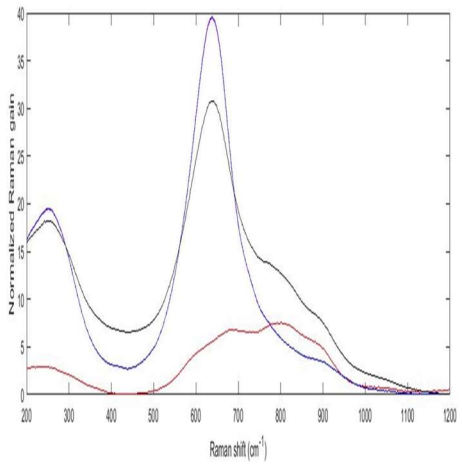

Figure 1. Raman gain of the initial BaNaNS glass and nucleated samples subjected to different thermal treatments.

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