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Enhanced green and red upconversion emissions in Er^{3+}-doped boro-tellurite glass containing gold nanoparticles

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HIGHLIGHTS

- Er^{3+}-doped boro-tellurite glasses containing gold NPs were prepared by melt-quench method.
- Au NPs with an average size of ~5.74 nm were observed by TEM.
- Red emission is enhanced up to 30 times.
- Finally, the mechanisms involving in such enhancements are discussed.

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ABSTRACT

Increasing the cross-section of upconversion emissions from the rare earth ions doped materials is a challenging issue. In this work, we report on the enhancement of the up-converted emissions of Er^{3+}-doped boro-tellurite glasses containing gold nanoparticles which have been prepared by a conventional melt-quench technique. Seven absorption bands and three emission lines are observed using the UV–Vis–IR and photoluminescence spectroscopic techniques, respectively. Red emission is enhanced up to 30 times in a sample having 1 wt% of Au nanoparticles. The presence of the gold nanoparticles with average size of ~5.74 nm is confirmed by transmission electron microscopy and corresponding surface plasmon band is observed at 630 nm in a singly-doped Au-nanoparticles embedded glass sample. A model to determine the enhancement factor of the emissions is suggested which could not describe the phenomenon for high concentrations of nanoparticles. Enhancement is attributed to the increased local field around the metal, and the results are discussed in details.

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Introduction

In the new millennium, the optical materials have attracted a large attention due to their versatile applications in science and technology. The novel challenge is to provide sources of energy from environment (such as solar cells) and generation of high energy resources through the nonlinear phenomenon. Although many efforts in these topics are reported, still the efficiency of proposed optical materials is not sufficient to be fabricated commercially, except for few materials used in the telecommunications, solar cells, diode-lasers, and lasing devices, such as Nd^{3+}–YAG crystals, GaAs silicon-based semiconductor lasers, GaAs solar cells and so on [1–3].

Rare earth (RE) doped glasses, glass–ceramics and crystals are promising materials since they show significant optical properties. One of the considerable prospective of the RE-doped glasses is to achieve higher energy emissions under lower-energy excitations, so-called "upconversion" phenomenon [4,5]. Upconversion occurs due to excited state absorption and/or energy transfer mechanism between neighboring ions majorly, while cross-relaxation processes may takes place as the minor factor. However, the intensity of those upconversion emissions from RE-doped optical materials is several times lower than normal fluorescence by the same emitters because of the inclusion of two/three-photon absorption mechanism to provide one up-converted photon and contribution of nonradiative multiphonon decays (especially in room and higher temperatures) and detrimental energy transfers among RE ions.

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Table 1
Nominal compositions of glass samples and their corresponding labels.

Glass	B_2O_3	TeO_2	ZnO	Er_2O_3	Au (wt%)
BT	30	59	20	1	0
BTA1	30	59	20	1	0.3
BTA2	30	59	20	1	0.5
BTA3	30	59	20	1	1
BTA	30	60	20	0	0.5

Here, the authors would like to refer to precious works by Auzel [4] and the review paper by Dong et al. [5] for more details on upconversion and energy transfer mechanisms.

In order to overcome the aforementioned problems, versatile methods have been proposed to increase the efficiency of the emissions of RE ions doped optical materials. Several approaches to avoid quenching and to enhance the intensity of the lanthanide doped materials are summarized by Zheng et al. [6]. One of the most motivating techniques is the introduction of the noble metallic nanoparticles (NPs) in the host matrix and generation of highly confined local field in vicinity of the metal which results in significant modifications in optical properties of the RE-doped materials, especially in the glassy systems. Such enhancements in different glasses doped RE ions have been reported by various authors. For instance, Amjad et al. [7] reported significant enhancement of upconversion emissions of the Er^{3+} ions in phosphate glasses containing silver NPs which is attributed to the local field effect by the metal with a surface plasmon band centered at 528 nm. Previously, we reported such enhancements in infrared to visible upconversion emissions of Nd^{3+} and Er^{3+}-doped tellurite glass [8,9]. It is also concluded that the presence of the silver NPs increases the thermal stability of the tellurite glass [10]. River et al. [11] reported on the enhancement of the emission at 800–840 nm region in Er^{3+}-doped tellurite glass containing gold NPs after annealing up to 7.5 h. However, they showed that only quenched observed in the same region of spectra while adding the silver NPs as an alternative for gold. They, moreover, reported that the lifetime of the $^4I_{13/2}$ decreased after introduction of metallic NPs. Kassab et al. [12,13] observed enhancements in upconversion emissions of the Er^{3+}–Yb^{3+} and Tm^{3+}–Yb^{3+} co-doped glasses containing silver NPs after 48 h annealing and this trend was attributed to the contribution of the intensified local field by SPR. The effect of gold NPs on the spectroscopic and structural properties of Er^{3+}–doped zinc tellurite glass [14] and the role of silver NPs on the chemical durability of similar composition are reported recently by our group [15].

The improvement of the optical properties in the RE-doped glasses containing metallic NPs strongly depends on the type of the metal, dielectric function of the host, size and shape of the NPs and the distance between NPs and RE ions (concentrations of the dopants). Therefore, it is interesting to investigate the effect of the metallic NPs in the different hosts. Up to date, there is no report on the influence of the gold NPs on the luminescence of Er^{3+}-doped boro-tellurite glasses. The momentous properties of the boro-tellurite glasses motivated us to carry out this study. The boro-tellurite glass consists of the both properties of the tellurite-based and borate-based glasses. Tellurite glasses show high linear and nonlinear refractive indices, wide range of transparency, high density and thermal stability [16]. On the other hand, borate glasses possess also high refractive index and transparency region, while they are eco-friendly [17].

Experimental procedure

Tellurite-based glasses are friendly in terms of melting process and preparation. Tellurium dioxide alone can be melted only around 750 °C in few minutes. However, the addition of the other constituents increases the required melting temperature and time interval for the preparation of the homogenous and viscous molten of the glass. The glass composition $59TeO_2$–$30B_2O_3$–10ZnO–$1Er_2O_3$–xAu NPs (where x = 0, 0.3, 0.5 and 1 wt%) have been prepared in several batches of 20 g (see Table 1 for the corresponding labels). Gold nano-powders 99.99% having size smaller than 100 nm were provided by Sigma–Aldrich. High quality starting agent materials were mixed completely and the batches were melted at 950 °C in an alumina crucible for 20 min. The melt were stirred frequently during the process. Then, the transparent melt were poured between two pre-heated stainless steel moulds, and kept for 3 h at 300 °C. Next, the annealing process was terminated by a slow cooling of furnace to ambient temperature.

X-ray diffraction (XRD) patterns were obtained by a Burker D8 Advanced diffractometer using Cu Kα radiation (λ = 1.54 Å) which works at 40 kV and 100 mA. Transmission electron microscope (TEM Philips CM12) was employed to record the TEM image of nanostructures. Very fine powder of each sample was dispersed in acetone, exposed to ultrasonic for 15 min, and dropped to dry before examining the specimen. The UV–Vis–IR absorption spectra of the samples were recorded using a Shimadzu UV-PC103 spectrophotometer with a resolution of 1 nm. Photoluminescence (PL) measurements were performed by a Perkin–Elmer LS55 PL spectrometer with a resolution of 1 nm. All the measurements were carried out at room temperature.

Results

Fig. 1 shows the XRD pattern of the glass BT. Occurrence of a broad hump in the range of 20–40 degree and the absence of any sharp peak confirm the glassy nature of the sample. The UV–Vis–IR absorption spectra of the samples BT, BTA1 and BTA are depicted in Fig. 2. Seven absorption bands are observed in the visible to near-infrared region of the samples which are attributed to the transitions from the ground state of the Er^{3+} ion ($^4I_{15/2}$) to different excited states. Bands centered at 1526 (FWHM ~85 nm), 976, 798, 654, 546, 522 and 488 nm correspond to electronic transitions from ground state to $^4I_{13/2}$, $^4I_{11/2}$, $^4I_{9/2}$, $^4F_{9/2}$, $^4S_{3/2}$, $^2H_{11/2}$ and $^4F_{7/2}$ respectively. The presence of the gold NPs samples BTA1-BTA3 did not alter the absorption peak positions and intensities. It is likely due to the small concentration of the gold NPs. In order to track the surface plasmon band of the metal, an alternative sample was prepared which contains Au NPs, but Er^{3+} ions. The small absorption hump was observed (as can be seen in inset of Fig. 2) at 630 nm which can be attributed to the surface plasmon band of the gold NPs. The observation of this peak in frequencies near

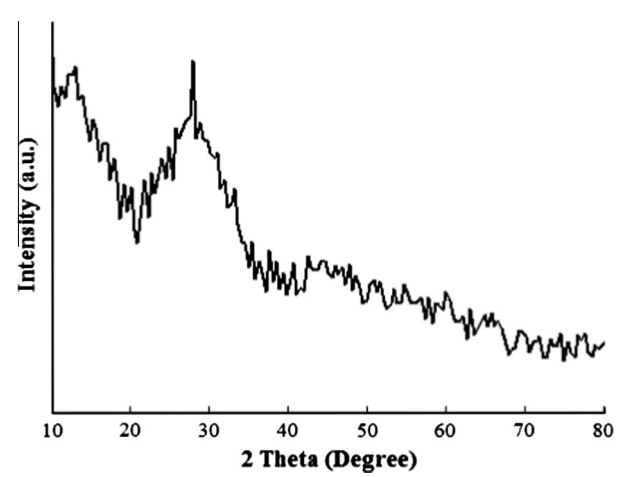

Fig. 1. XRD pattern of the BT glass sample.

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