Intrinsic Bistability of Luminescence and Stimulated Emission in Yb- and Tm-Doped Glass

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Intrinsic bistability and hysteresis have been observed in the photoluminescence of bulk glass doped with Yb^{3+} and Tm^{3+} impurities at room temperature. Multiple instabilities also occur in channel waveguide lasers in this material. Consequently, local field effects exhibit more degrees of freedom than expected and have important implications for compact optical amplifiers and oscillators.

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Nearly a century ago, Lorentz developed an elegant theory for the optical polarization of matter [1] which introduced the concept of local fields. He calculated a correction for fields incident on a solid which yielded the internal field value using a simple, approximate procedure applicable to many materials. This theory was reformulated and extended for specific crystal structures subsequently by a number of authors [2,3]. With the advent of nonlinear optics [4] and the discovery of phenomena such as surface-enhanced Raman scattering [5], local field corrections were gradually recognized as being important for quantitative explanations of some optical interactions with matter because of the multiplicative dependence of output intensity on several fields, and, consequently, several local field factors. On the other hand, new nonlinear phenomena specifically *mediated* by local field corrections were not reported, although dramatic nonlinearities originating from dynamic local field modifications were predicted to occur in dense collections of radiating atoms driven by resonant electromagnetic fields [6]. It was shown theoretically—for example, by incorporation of the local field correction in the Maxwell-Bloch equations for two-level systems subject to resonant light—that intrinsic switching of optical emission could take place. Recently, these predictions were confirmed by the observation of intrinsically bistable emission of rare-earth impurities at low temperature in the compounds $Cs_3Y_2Br_9$:10% Yb^{3+}, $Cs_3Lu_2Br_9$:10% Yb^{3+}, and $CsCdBr_3$:1% Yb^{3+} [7–9]. Nonlinearities mediated by local field effects have also been reported in composite dielectrics [10].

In the present work, we have extended observations of intrinsic switching to a new rare-earth system—namely, a laser glass doped with Yb and Tm ions in which behavior that is qualitatively different from earlier observations is found. Intrinsically bistable emission is reported at room temperature and multiple hysteresis loops are observed for the first time to our knowledge. The influence of the intrinsic switching process on the stimulated emission from a channel waveguide fabricated in this material is also investigated and found to be strikingly different from conventional devices. We show that in this glass system optical switching involves an interplay in dynamics of two sensitizers Yb and Tm. We consider whether multiple hysteresis loops arise from the same local field mechanism identified in earlier work [7], or whether a new nonlinear mechanism is needed. Our experimental results are immediately relevant to a variety of compact laser systems utilizing concentrated Yb-doped gain media in which reliable, reproducible energy storage or constant output intensities are desired, such as Er, Yb-fiber amplifiers for communications [11] and tabletop terawatt laser systems. However, we also believe they are pertinent to laser materials utilizing high concentrations of many other rare-earth ions.

We examined the optical properties of glass specimens prepared by Corning. The batch melts consisted of 12.50% Na_2O, 3.53% K_2O, 4.08% BaO, 73.70% SiO_2, and 5.99% Yb_3O_2 by weight. In addition, the starting material for the rare-earth component contained 38 parts per million Tm_2O_3. The final composition thus included Yb^{3+} and Tm^{3+} ions in a concentration ratio of nearly 30 000 to 1. Other rare-earth oxides, including Er_2O_3 and Ho_2O_3, were present in such low concentrations (less than 10 ppm) as not to influence our measurements. Bulk samples were cut from the optical grade glass specimen into 1 mm $\times$ 10 mm $\times$ 10 mm slabs and polished to a mirror finish. Additionally, in one sample, waveguides were prepared by etching parallel strips 10 μm wide through an aluminum mask evaporated onto one surface of the glass slab. Silver was then diffused into the exposed glass surface by immersion in a sodium nitrate–silver nitrate (0.5% $AgNO_3$ by weight) melt at 618 K for 12 min. This created silver rich channels 10 μm wide and up to 5 μm deep in which the refractive index differed by 0.03 from that of the surrounding glass [12].

In all, seven channels with transmission losses less than 3 dB at 1020 nm were formed in the waveguide sample. Laser cavities were subsequently formed by placing a pair of plane dichroic mirrors perpendicular to the channel ends. Index matching fluid was used to minimize coupling losses. The mirrors had a transmission of 80% at 910 nm and a reflectance of 95% at 1020 nm. To observe bistability in bulk samples, we monitored the intensity of Tm^{3+} and Yb^{3+} emissions at 483 and 960 nm, respectively, while varying the power of the exciting beam. For

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waveguide measurements, the intensity of emission at 483 and 960 nm was monitored perpendicular to the channels, together with laser output power at 1020 nm in the forward direction. Luminescence was split into two portions with a mirror and simultaneously analyzed at 483 and 960 nm with a pair of Czerny-Turner spectrometers equipped with photon-counting electronics.

A continuous-wave Ti:sapphire laser was used as the exciting source. Its output power was stabilized and controlled with an electro-optic servo. Bulk samples were excited at a wavelength of 973 nm, corresponding to the strongest absorption peak of the Yb^{3+} ion in this glass host. In the case of the waveguide laser, however, the transmission of the high reflector was too low and the absorption in the channel too high at 973 nm to provide reasonable gain over the whole length of the waveguide channel. Hence the guide was instead pumped at 910 nm, a wavelength corresponding to a secondary absorption peak of Yb^{3+} (Fig. 1, inset). This provided reasonable pumping efficiency without any change of output coupling. Near-infrared measurements of laser output power were made with a pyroelectric detector.

Portions of the emission spectrum excited at room temperature by absorption on the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ transition of Yb^{3+} at 960 nm are shown in Fig. 1. Identical spectra were observed in the bulk and waveguide samples. The main figure shows lines in the blue band centered at 483 nm and attributed to $^1G_4 \rightarrow {}^3H_6$ emission of the Tm^{3+} ion, together with a red band near 650 nm assigned to $^1G_4 \rightarrow {}^3F_4$ transitions in the same ion. In the inset is the near-infrared emission spectrum from the $^2F_{5/2}$ state of Yb^{3+} in this sample. Both visible and near-infrared spectra have been normalized to the most intense feature in the corresponding wavelength range. The broad near-infrared band is much stronger than the visible bands. No known Tm^{3+} transitions involving single ions in the ground state occur in the wavelength range of Yb^{3+} absorption, and off-resonant single ion excitation involving photons in this energy range has not been reported for isolated Tm^{3+} ions. In samples co-doped with Yb^{3+} and Tm^{3+}, however, energy transfer processes evidently do populate the excited states of Tm^{3+} [13–15], as indicated by the visible emission trace in Fig. 1. Although the concentration of Tm^{3+} is quite low, and the transfer is off resonant, transfer efficiency is high enough to ensure significant excited-state populations of Tm^{3+}. At the present time, there is no detailed information available on the configuration of centers at which Yb^{3+} ions in this materials interact with Tm^{3+}, although there have been discussions of models of their interaction [13].

Figure 2 shows the observed intensities of 483 and 960 nm emission in the bulk sample as a function of incident power at 973 nm. The upper trace shows that the visible emission intensity drops abruptly at 370 mW, as the incident power is increased. As power is lowered, however, it undergoes an abrupt increase at 330 mW. A distinct hysteresis loop is thereby produced. The lower trace shows corresponding behavior in the near infrared. There too, emission intensity changes discontinuously at optical powers where the visible emission at 483 nm undergoes its sudden variations, but the sense of the hysteresis is opposite: The near-infrared emission intensity switches down when the visible switches up and vice versa. This is similar to an earlier observation of intrinsic switching in metal halides [7]. It is worth noting, however, that bistability and hysteresis was much more difficult to observe cleanly in *bulk* glass samples than in previous experiments. These effects were very weak in bulk material and were usually obscured by somewhat larger modulation due to a slow thermal nonlinearity ($\tau_{\text{relax}} \sim 12$ s). Results in Fig. 2 were obtained at a slow rate ($4\,\text{mW/s}$) in an irregularly shaped sample to minimize the effect of thermal nonequilibrium and avoid optical feedback.

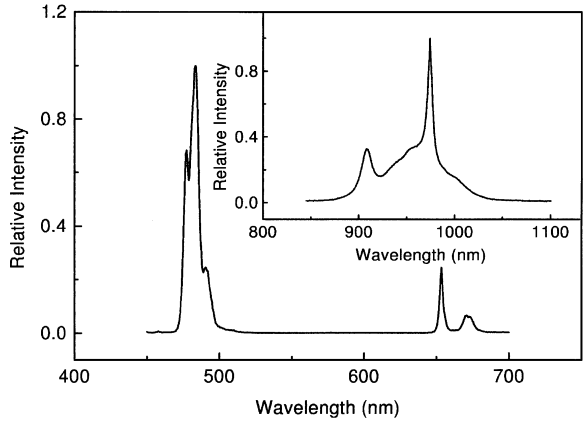

FIG. 1. Unpolarized emission spectra of Yb^{3+} and Tm^{3+} ions in glass excited at 960 nm. The main figure shows visible emission features. The inset shows near-infrared emission. Vertical axes have been normalized to the most intense spectral peak.

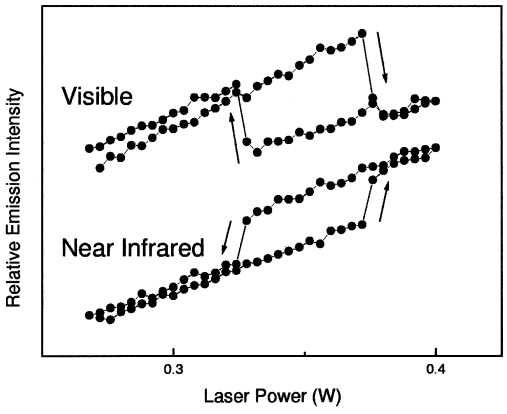

FIG. 2. Power dependence of visible and near-infrared luminescence intensity in a thin sample of Yb^{3+} and Tm^{3+} doped glass at room temperature. The arrows indicate the directions in which the intensities are changing.

Figure 3(a) shows the variation of the intensities of visible and near-infrared light emitted perpendicular to the optical channel when cavity mirrors were aligned for waveguide laser operation at 1020 nm. Fluorescence intensities at 483 and 960 nm corresponding to spontaneous emission transitions of Tm and Yb, respectively, saturate in unison above the laser threshold, confirming the presence of significant Tm-Yb coupling. Near 400 mW input power, the emission traces at 483 and 960 nm reveal switching behavior similar to that in the bulk sample (Fig. 2), producing near-infrared and visible hysteresis loops with opposite sense. At higher power, a second hysteretic region is encountered where high contrast is achieved and, curiously, the visible and near-infrared intensities switch in the same direction. The laser output itself, at 1020 nm, exhibits saturation and switching behavior as shown in Fig. 3(b). The laser output switching "polarity" around 400 mW follows that observed in the the near-infrared luminescence trace of Fig. 3(a). The onset of switching is closely followed by strong saturation of output, consistent with earlier predictions [8] for emission on the upper branch of the instability.

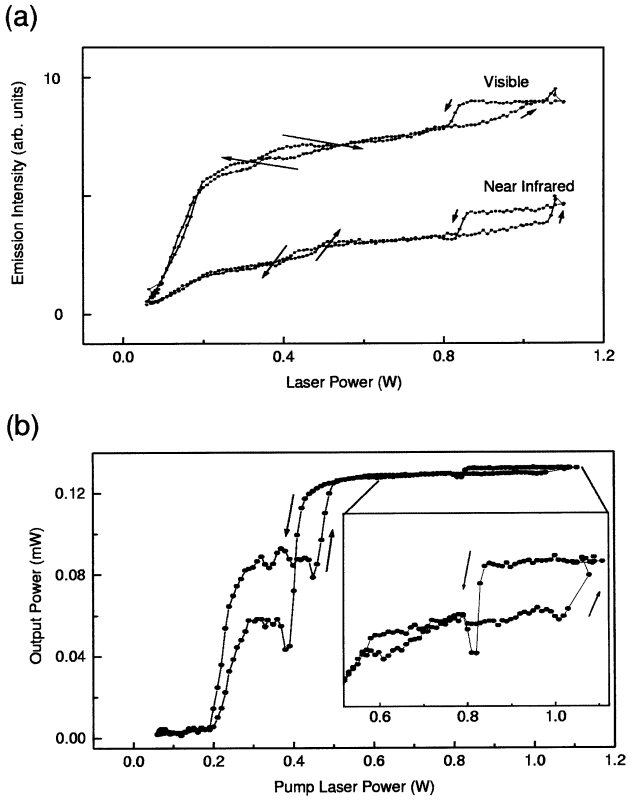

FIG. 3. (a) Intensity of visible (483 nm) and near-infrared (960 nm) spontaneous emission in waveguide laser cavity as the incident laser power is first increased and then decreased. Arrows show the directions of traversal of the hysteresis loops. The visible and near-infrared emissions switch with opposite polarity at the transition points in the low power region, and with like polarity at higher intensity. (b) Output power of the waveguide laser at 1020 nm versus incident laser power for increasing and decreasing pump power. Inset: an expanded view of the high power region.

Previous theoretical analysis by several groups [7,16] identified the Lorentz local field correction as the factor responsible for polarization-population coupling which leads to bistable luminescence. Pair interactions were shown to be capable of amplifying the hysteresis [7], and recent site-selective spectroscopy of intrinsic bistability in $CsCdBr_3$:Yb^{3+} has provided information on the roles of energy migration and ion-ion distances needed to observe intrinsic switching [8]. As presently formulated, however, the local field model predicts only a cubic dependence of incident light intensity on excited state populations, and hence only a single intensity region where bistability can occur, in contrast to the data shown in the foregoing figures. The combined results of Figs. 2 and 3 indicate that intrinsic switching occurs in Tm, Yb:glass at room temperature in two physically distinct ways. Consequently, the question of the origin of bistable luminescence in the present system acquires interest as a fundamental problem, and an explanation for observations of this phenomenon at high temperature is needed.

The observations presented here in which the relative polarities of visible and near-infrared hysteresis loops are *opposite* are consistent with the detailed balance argument of Hehlen *et al.* [8] for Yb^{3+}:$Cs_3Y_2Br_9$. On the other hand, hysteresis loops having the *same* polarity are consistent with recent observations of intrinsic bistability in Yb^{3+}:$CsCdBr_3$ [9]. The difference between these two cases is that upconversion and infrared luminescent processes draw on coupled ions which are either divisible into two distinguishable populations or are equivalent throughout the sample. When two or more Yb^{3+} sites are active, one site can account for visible upconversion while the other emits primarily infrared radiation. Fast migration of energy between them ensures that emission at an upconversion site lowers the excited-state density available at infrared emitting sites. Where identical switching polarities are observed, a single excited-state population must be responsible for both visible and near-infrared emission. In the present experiments we find examples not only of opposite polarities, but also of identical polarities in one and the same sample. Hence our present results are consist with the existence of two different switching sites.

In summary, we have demonstrated intrinsic switching and bistability of photoluminescence in glass doped with Yb^{3+} and Tm^{3+} impurities at room temperature. Laser emission in waveguide channels fabricated in this glass also underwent hysteretic switching at room temperature. The clamping of output power and its intrinsic instabilities seriously limited the extraction efficiency and predictability of waveguide laser output. In its present form the Lorentz local field model does not account for multiple instabilities in the luminescence of Yb, Tm:glass or reversal of relative switching polarity of near-infrared and visible hysteresis in a single sample. However, we believe

our results can be accounted for with a model based on multiple sites for Yb^{3+} ions undergoing switching. Since energy transfer mechanisms similar to those described here for Yb-Tm apply also to the dynamics of Yb-Er systems, the present results can be expected to extend to Er systems and other concentrated rare-earth media, introducing a new intrinsic limit to doping (and therefore compactness) of solid state oscillators and amplifiers.

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