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SCIENTIFIC CORRESPONDENCE

Random laser?

SIR— Lawandy *et al.*[1] describe optical experiments on colloidal suspensions of TiO_2 microparticles in rhodamine dye solutions. These suspensions were pumped with 532-nm pulses and the spectral and temporal behaviour of light emitted from the pumped surface was recorded. Above a certain pump power threshold a dramatic narrowing of the emission linewidth and a shortening of the emitted pulses were observed. These are known characteristics of laser action. As the same effects were not observed for a clear dye solution, the laser-like behaviour was ascribed to the presence of the microparticles. The authors suggested a possible interpretation in terms of a feedback mechanism supplied by photon diffusion, which would make their system a fascinating 'random laser', but they also point out that part of their data remains puzzling. In view of the large reported values for the (photon diffusion) mean free path, we expected the proposed feedback mechanism to be very unlikely. We therefore decided to look into the reported phenomena.

On the basis of our experimental findings, we propose an alternative explanation in terms of amplified spontaneous emission, which is a common phenomenon for pumped laser dyes [2,3] and does not require optical feedback. Laser-dye molecules absorb light in a frequency range known as the pump band. Once excited they become non-absorbing for pump light (the dye is 'bleached'). The excited population decays spontaneously with some characteristic lifetime, under emission of broadbanded light at lower frequencies. This is called spontaneous emission. If the emitted light passes through a region containing excited dye molecules, it may de-excite these, being amplified in the process (stimulated emission). This accelerates the decay and therefore shortens the (pulsed) output. Because the gain is strongest at the wavelength where the cross-section for stimulated emission is highest, (exponential) amplification also leads to band narrowing. In a laser, cavity mirrors bring about a multiple passage of the light through the amplifying region. This feedback mechanism is used to obtain a high gain. In the absence of feedback, however, single-pass amplification may already produce significant band narrowing and pulse shortening, a phenomenon termed amplified spontaneous emission (ASE).

Within a pumped region, ASK will occur in the direction(s) of highest gain, that is, in general, in the direction(s) in which this region is most extended. If a spatially broad pump pulse is incident on a dye cell, its front edge will create a disk-shaped amplifying region, and ASK will develop in the plane of this disk, parallel to the cell window. Once ASK has started, subsequent pump light is efficiently converted into ASK light in a cyclic absorption-stimulated emission process, and will not penetrate beyond the active region. On the other hand, for a spatially narrow pump pulse, no significant ASK will develop across its diameter. The initial bleaching will live longer and pump light will penetrate deeper. A pencil-shaped amplifying region is formed which, once long enough, will yield ASK parallel to the pump beam. For the pump geometry used by Lawandy *et al.* one would expect ASK parallel to the front cell window.

We pumped a clear dye solution in this geometry and, like Lawandy *et al.,* observed broad-banded and long-pulsed light, emerging from the front window of the cell. This is spontaneously emitted light that has travelled through the amplifying region over (at most) its very limited thickness. However, we also observed from a clean dye solution, far more intense, narrow-banded (Fig. 1), and short-pulsed (Fig. 2) light emerging from the side windows. This is a ASK light which has the same 'laser-like' characteristics as the output that Lawandy *et al.* observed from colloidal suspensions (Figs 1 and 5 of ref. 1).

We think that the reason that Lawandy *et al.* did not observe ASK from clear dye solutions is that they studied emission through and not parallel to the front window. With TiO_2 added, they did observe narrow-banded emission, because the particles scatter some ASK light out of its plane. We think that the main role of the particles in Lawandy's experiments is not feedback, but post-processing: they direct ASK light to the detector.

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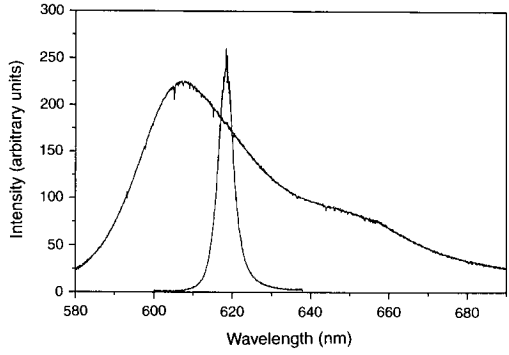

FIG. 1 Emission spectrum of a 2.5×10^{-3} M solution of rhodamine 640 perchlorate in methanol pumped by 3-mJ (10-ns) pulses at 532 nm. Pump beam diameter 1.5 mm. The broad spectrum corresponds to the (spontaneous) emission from the front window of the dye cell whereas the narrow spectrum corresponds to the emission (ASE) from the side windows.

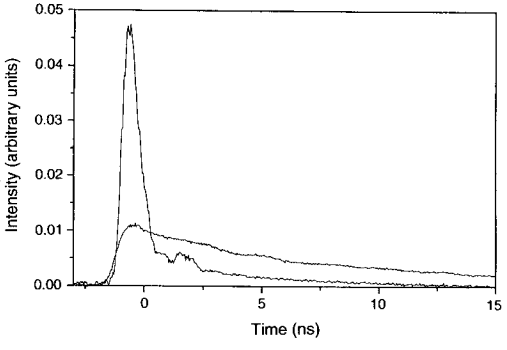

FIG. 2 Temporal emission of the same dye solution as in Fig. 1, pumped with 58-μJ (30-ps) pulses at 532 nm. The slowly decaying peak corresponds to the spontaneous emission from the front window whereas the short peak corresponds to the emission (ASE) from the side windows. The width of the short peak is determined by the time resolution of the detector, which is about 1 ns. The rapid structure following the short peak is a common artefact from ringing in the fast detection system.

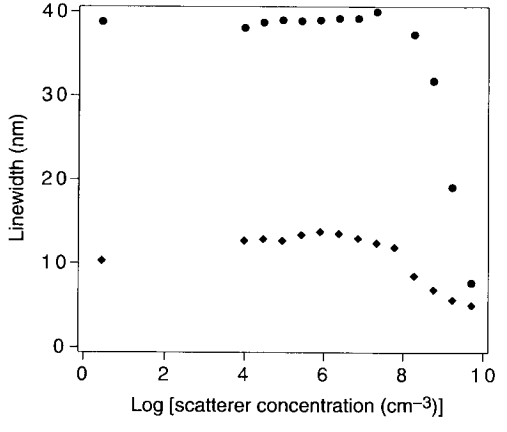

FIG. 3 Linewidth of the front (circles) and side (diamonds) emission as a function of scatterer concentration at a constant fluence of 8.5 mJ cm^{-2}. The sample consisted of TiO_2 scatterers (diameter ~250 nm) in a 2.5×10^{-3} M solution of rhodamine 640 perchlorate in methanol and was pumped using 80-ps pulses at 532 nm from a frequency doubled, Q-switched mode-locked Nd:YAG laser.

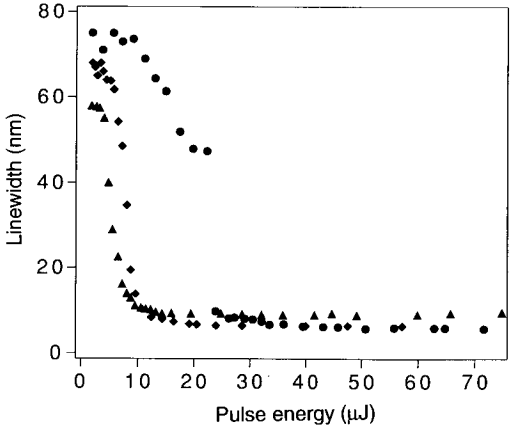

FIG. 4 Linewidth as a function of the pump pulse energy for transport lengths (circles, 690; diamonds, 69; triangles, 11 μm). The area of the pump beam at the cell face was 3×10^{-3} cm^2. The rest of the experimental arrangement was as for Fig. 3.