

LASER PHYSICS

Light in the darkness

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The term laser is an acronym that stands for Light Amplification by Stimulated Emission of Radiation¹. However, stimulated emission is inherently linked to spontaneous emission, and one would therefore expect the lasing emission to be limited to the spectral range of the spontaneous emission of the active material. Actually, it is possible to obtain laser emission beyond this range by exploiting the coupling of the electron transitions to lattice vibrations, but, so far, this has enabled to obtain laser emission outside the spontaneous emission range by only a few nanometers. Now, writing in *Nature Physics*², Fei Liang and colleagues have taken full advantage of this coupling, achieving laser emission from a Yb-doped $\text{YCa}_4\text{O}(\text{BO}_3)_3$ crystal that extended well beyond the spontaneous emission limits by almost 400 nm².

The theoretical basis of laser emission was laid by Albert Einstein in 1917, who introduced the concept of stimulated emission³. In practice, a laser is a device that amplifies the emission of the active material to obtain coherent light. An optical cavity, usually made of dielectric mirrors, provides the necessary feedback to sustain the amplified emission with the right coherence properties.

For many years, it was thought that direct laser emission could only be achieved within the spectral range of the gain medium's spontaneous emission, that is, that laser emission was possible only for purely electronic transitions. This imposes severe limitations on the obtainable laser wavelengths. As a result, despite the large number of active materials, each with its own spontaneous emission characteristics, part of the electromagnetic spectrum is still inaccessible by direct laser emission because no gain media exist in those regions.

Recent decades have seen several examples of laser emission slightly beyond the spontaneous emission band of the active material by coupling electron transitions with the mechanical vibrations of the atoms in the material^{4,5}. Because this electron-phonon coupling is weak, vibronic emission has been limited to a side emission that does not extend more than a few nanometres beyond the intrinsic spontaneous emission range of the gain medium — until now.

Liang and colleagues were able to achieve phonon-assisted laser emission as much as 400 nm beyond the spontaneous emission spectrum. They faced two key problems: electron-phonon coupling is weak and is usually overwhelmed by much stronger processes such as pure electronic emission. Moreover, the strength of the vibronic interaction decreases with the number of phonons involved and therefore with the spectral distance from the purely electronic emission band.

The team realized that, in order to observe the vibronic emission, one has to suppress the spontaneous emission of the material. They achieved this with a judicious choice of the dielectric coatings on the input and output faces of the Yb-doped $\text{YCa}_4\text{O}(\text{BO}_3)_3$ crystal and of the cavity mirrors to suppress the stronger electronic emission and enhance the electron-phonon coupling. In this way, they were able to obtain laser emission involving an increasing number of phonons (Fig. 1.) As expected, the

42 efficiency of the system decreases with the number of phonons involved from a maximum of 41%
43 with three phonons to 0.3% with a maximum of seven phonons involved and an emission wavelength
44 as long as 1436 nm.

45 This extended lasing range comes at a cost. In order to achieve tuneable laser emission, Liang and
46 colleagues had to modify the dielectric coating of the active material and of the cavity mirrors to
47 access a specific narrow spectral region, with each coating further away from the intrinsic emission
48 band.

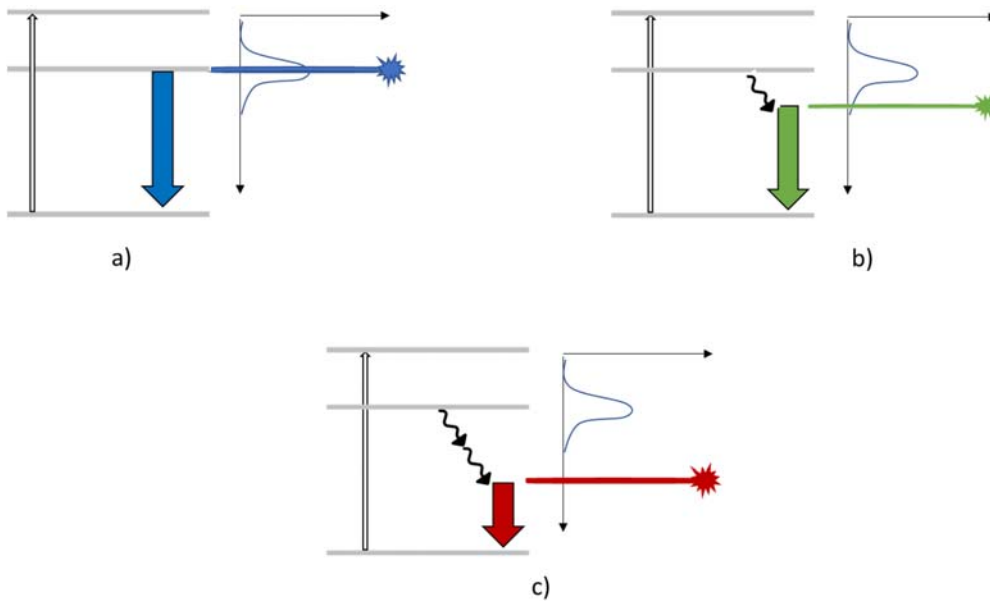
49 Although this cannot be considered an ultrabroadband tunable laser because each spectral region
50 requires a different coating, it is remarkable that such a huge tunability range can be obtained with
51 just one active material and a few sets of coatings. Indeed, it is one of the largest segmentally
52 tunable regions ever obtained. For example, it compares well with the performance of the well known
53 Ti:Sapphire system from which an overall tunability range of 445 nm has been obtained with four
54 coating sets of the cavity mirrors³. Moreover, the team's experiments accessed a spectral region where
55 not many other active laser materials exist. In fact, they partially bridged the gap between the typical
56 emission regions of ytterbium-based and erbium-based lasers.

57 It should be noted that coherent emission from materials far beyond their spontaneous emission range
58 can also be achieved with indirect conversion methods based on nonlinear optics, such as harmonic
59 generation⁶, stimulated Raman scattering⁷, supercontinuum generation⁸ and self-phase modulation⁹.
60 All these approaches can convert the radiation to a much longer wavelength but, as they are based on
61 high order processes, they usually require the injection of high-power coherent light (usually from
62 another laser), and in most cases they can only work in the pulsed regime. They also come with other
63 disadvantages, such as complicated configurations, alignment sensitivity and high costs. For these
64 reasons, direct laser emission far beyond the spontaneous emission band of a material from a simple
65 and robust setup can open new application perspectives.

66 The potential of Liang and colleagues' does not end there. The gain medium they used has self-
67 frequency-doubling capacity, therefore, they obtained both near infrared and second-harmonic-
68 generated visible light with just one setup. Moreover, they proposed a few innovative approaches to
69 implement new schemes for dielectric coatings that could achieve larger tunability with just one type
70 of coating. They also applied the same approach to another active material (Yb- doped La₂CaB₁₀O₁₉
71 crystal) with similar performances, demonstrating that the same principle can be used with other
72 materials to extend the laser emission of many other systems.

73 The possibility of extending the tuning region of laser materials opens up new perspectives also for
74 ultrafast lasers, since the light pulse duration of a mode-locked laser is related to the bandwidth of the
75 gain medium. This could pave the way to new frontiers in the field of chirped pulse amplification of
76 ultrashort pulse lasers and frequency comb generation.

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79 **Fig. 1: Laser emission inside and outside the spontaneous emission band**

80 **a.** Laser emission (blue line) inside the spontaneous emission band (blue curve). In this purely
 81 electronic process, a photon (blue arrow) is emitted without interaction with the lattice **b.** Laser
 82 emission (green line) beyond the spontaneous emission band. A photon (green arrow) is emitted
 83 with the involvement of one phonon (wavy arrow), **c.** Laser emission (red line) far beyond the spontaneous
 84 emission band. A photon (red arrow) is emitted with the involvement of two phonons (not to scale).
 85 [Note for editors: this is just a simple sketch of what can be put in the figure. Maybe we could add a
 86 picture of the laser cavity from which the laser beam exits. Moreover, in **c.** it would be better to show
 87 the involvement of 7 phonon instead of two]

88

89 **Competing interests.** The author declares no competing interests.

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91 **References**

92 [1] Maiman T. H. *Nature* **187**, 493-494 (1960).
 93 [2] Liang F. et al. *Nat. Phys.* (2022) [3] Einstein A. *Physikalische Zeitschrift* **18**, 121 (1917).
 94 [4] Moulton, P. F. *Proceedings of the IEEE* **80**, 348-364 (1992).
 95 [5]. Sennaroglu, A. *Prog. Quantum Electron.* **26**, 287-352 (2002).
 96 [6] Zhu, S., Zhu, Y., and Ming, N. *Science*, **278**, 843-846 (1997).
 97 [7] Ferreira, M. S. and Wetter, N. U. *Opt. Lett.* **46**, 508-511 (2021).
 98 [8] Zahra et al. *Nature Comm.* **13**, 2126 (2022).
 99 [9]. Khurgin, J. B., Clerici, M. and Kinsey, N. *Laser Photonics Rev.* **14**, 2000291 (2020).