Laser Action in Emerald

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Abstract—Stimulated emission has been achieved in emerald, a chromium doped beryllium aluminium silicate ($Be_3Al_2Si_6O_{18}:Cr^{3+}$) crystal over a wavelength band peaked at 684.8 nm.

THERE is considerable interest in the development of high-power tunable solid-state lasers based on phonon terminated transition of metal ions and color centers. The theory of the basic concept was proposed in the mid-sixties by McCumber [1] and experimentally observed first by Johnson *et al.* [2] in Ni^{2+} doped MgF_2. The successful operation of alexandrite (Cr^{3+} in $BeAl_2O_4$) by Walling *et al.* [3] at room temperature as a tunable laser has led to the search for new crystals and glasses with broad-band emission as potential laser materials [4]. This has led us to renew research [5]–[9] into chromium doped beryllium aluminium silicate (Cr^{3+} in $Be_3Al_2Si_6O_{18}$), commonly called emerald.

Emerald is similar to ruby and alexandrite in electronic structure [3], [5], [7]–[9]. Contrary to the emission from ruby and alexandrite, the R line is smaller and is only indicated by a small peak in the broad emission spectra. This is caused by the weaker crystal field which alters the energy states of Cr^{3+} ions in emerald. As a result, the two $4T$ levels ($^4T_2$ and $^4T_1$) lie at slightly lower energies but the position of the $2E$ band is essentially unaltered. The energy difference at room temperature between $^4T_2$ (responsible for broad-band emission) and $2E$ (R-line radiation) is about 400 cm^{-1} [7]. This is in contrast to the energy differences for ruby and alexandrite of 2300 [7] and 800 cm^{-1} [3], respectively. This energy difference allows the repopulation of the $^4T_2$ level due to thermalization from the long-lived $2E$ level. Therefore, emerald can have a relatively larger inversion of population of vibronic modes at room and higher temperatures.

Emerald exhibits a wide fluorescence spectra capable of laser action for all wavelengths from 670 to 850 nm. The broad absorption spectra of emerald is from the visible region up to 700 nm, which makes it suitable for pumping by readily available flashlamps or by a suitable laser source.

This letter reports on the first laser action in emerald. In the experiment, a hydrothermally grown crystal (0.3 At percent of Cr^{3+} ions) of length 14.7 mm was placed in the short cavity of two 30 cm radius dielectric coated mirrors to pass 630 nm and maximum reflectivity for wavelengths greater than 680 nm.
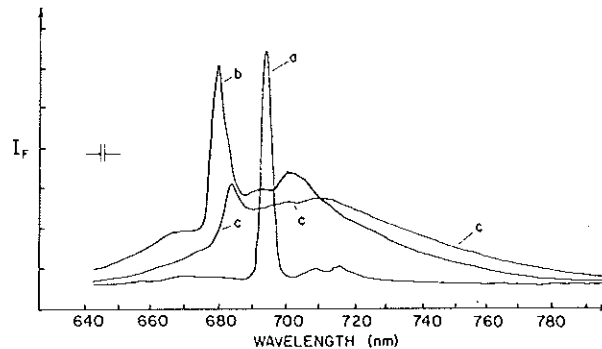
Fig. 1. Fluorescence spectra from a) ruby, b) alexandrite, and c) emerald at room temperature under similar conditions.

The output mirror had $T = 55$ percent for 680 nm. The crystal was longitudinally pumped along the C-axis by a 630 nm 8 ns pulse produced by SRS in ethanol by a SHG pulse emitted by a Q-switched YAG laser. A pumping beam was focused in the center of the crystal by a 12 cm lens to about 60 μm spot size. The output from the laser cavity was analyzed using a Jarrell-Ash Mark X 275 mm focal length spectrometer with 300 grooves/mm grating and 50 μm slit coupled to either a fast photomultiplier or a Hamamatsu C-1000 SIT video camera with a Hamamatsu temporal analyzer minicomputer system.

The relative fluorescence spectra measured from ruby, alexandrite, and emerald under similar steady-state excitation conditions are displayed in Fig. 1. One notices that emerald emits wider spectra than ruby and alexandrite in the 700-850 nm region, and the emission intensities are comparable. Fig. 2(a) shows the spectrum of laser emission from emerald, and Fig. 2(b) displays the relative output versus input power characteristics. The slope of this curve is 1.7, indicating large losses in the cavity due to the poor grade of the crystal. The emission band is centered at 684.8 nm with a bandwidth of about 30 Å which varies from shot to shot. These emission peaks are linked to the transition from $2E$ to $^4A_2$ level as well as from $^4T_2$ level [10]. The laser action observed by a fast photomultiplier follows the excitation pulse, sometimes showing oscillations commonly observed in the multimode regime of anisotropic lasers [11]. At this time, one can only speculate why we did not observe the broad emission from the phonon terminated $^4T_2$ level. This may be attributed to a short excitation pulse and the thermalization time effect in addition to a higher gain at the R-line. Another line often appears at 670 nm in the laser emission spectra. The source of this line is not known at this time but could arise from the $^4T_1$ level of Cr^{3+} ions or optical phonon enhanced anti-Stokes emission.

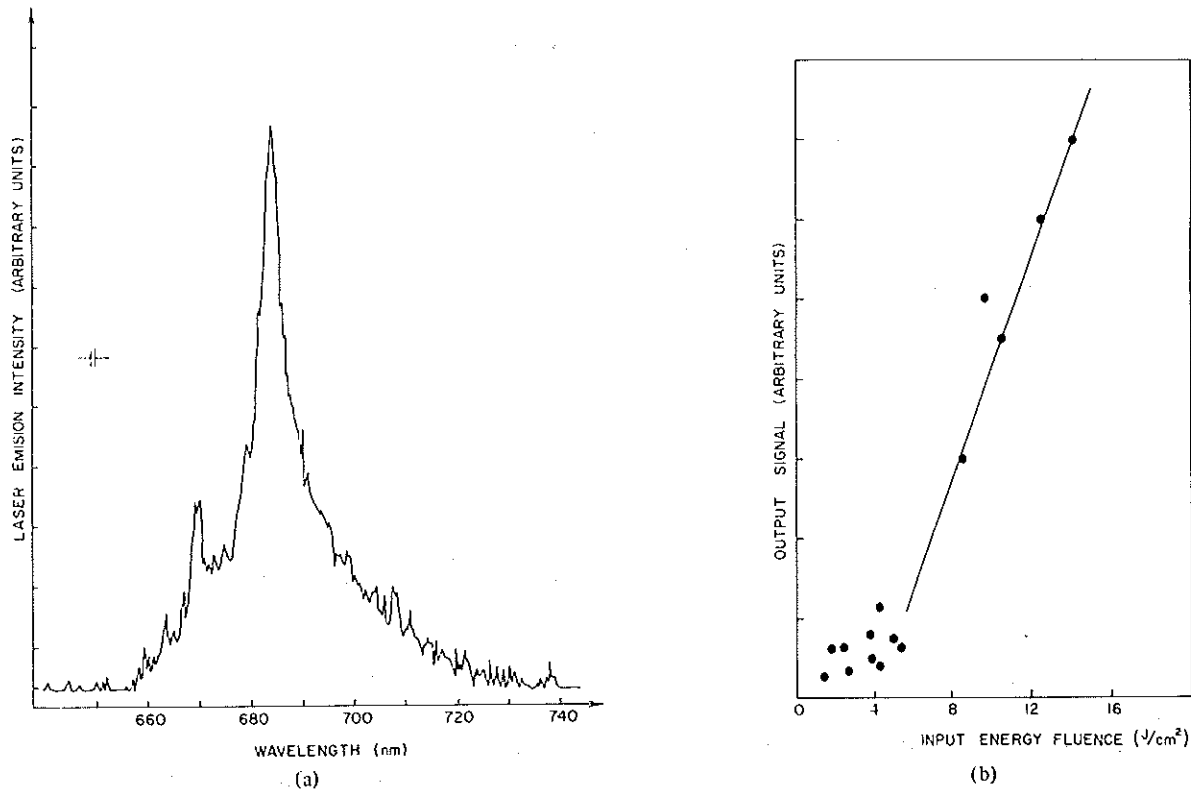Longer excitation pulses with better grade crystals will be

Fig. 2. (a) Laser emission spectra from emerald. (b) Relative output
signal versus input energy for emerald laser.

used to interpret the phenomena observed and reported here.

Note added in proof: After submission of our paper, [12] was published. In addition, in 1981 a patent application was filed by R. R. Alfano and J. Buchert entitled "Chromium-doped aluminum silicate laser system."

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