Amplified Spontaneous Emission in Broad-band Confining Embedded Strips Produced By Electron-Beam Lithography in LiF Crystals and Films

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Amplified spontaneous emission of aggregate colour centres, laser active in a broad wavelength interval in the visible, has been measured in confining embedded strips produced by electron-beam lithography in lithium fluoride crystals and films.

Keywords: amplified spontaneous emission, colour centres, Lithium Fluoride, photoluminescence

Introduction

Nowadays a renewed interest has been increasing for broad-band emitting materials. The area of growth and characterisation of thin films has seen a considerable expansion due to the need for a substantial shrink of scale and cost of optical devices. Colour centres (CCs), consisting in lattice vacancies trapping electrons or holes, are among the most investigated electronic defects in insulating materials [1], and in particular in ionic crystals [2]. Several of them are used for tuneable solid-state lasers due to the inhomogeneous broadening of their emission bands characterised by large oscillator strength and high quantum efficiencies [3].

Recently very promising results have been obtained in the generation [4], confinement [5] and amplification [6] at room temperature (RT) of visible light in active channel waveguides produced by electron beam lithography (EBL) at the surface of Lithium Fluoride (LiF) crystals. The limited penetration (from about 0.1 to 3.7 μm) of low energy (2-20 keV) electrons allows the formation of stable primary and aggregate CCs in an irradiated layer of controlled depth located at the surface of the crystalline materials. A local modification of the complex refractive index in this irradiated layer is induced by CCs embedded in it [7]. In particular, the efficient formation of F_3^+ and F_2 laser-active defects (two electrons bound to three and two close anion vacancies, respectively), characterised by broad photoemission bands in the green and in the red spectral ranges and almost overlapped absorption bands located at around 450 nm [8], is accompanied by a simultaneous local increase of the real part of the refractive index in the same wavelength interval where their emission are located [7,9]. So through a direct-writing process it is possible to create an active channel waveguide.

Research activities about CCs in alkali halide films [10] started more recently. The use of EBL techniques allows the definition of coloured embedded strips at the surface of LiF films grown on different types of substrates. This approach shows high potentialities for the development of novel active waveguide devices, like miniaturised broad-band optical amplifiers and waveguide tuneable lasers, due to the peculiar optical characteristics of the irradiated material, to the lower cost and simpler fabrication processes as well as to the easier integration with optical fibres and other passive and active optical components.

Generation [11], confinement[12] and amplification [13] of visible light in active coloured embedded strips produced by low energy electrons at the surface of LiF films grown by thermal evaporation on few types of substrates have been demonstrated. However much efforts are required to improve their design, preparation and performances. In this work some peculiarities of the LiF films with respect to single crystals are discussed, with particular emphasis on the amplification properties of LiF confining active strips based on CCs.

Experimental results and discussion

Several coloured strips were produced by EBL on LiF single crystals and polycrystalline films, grown by thermal evaporation on glass substrates, with irradiation doses in the range $10^{-3} \div 10^{-2}$ C/cm^2 and 12 keV electrons, whose penetration depth in LiF is estimated around 1.5 μm [14].

Photoluminescence (PL) represents a simple and powerful tool to investigate CCs formation efficiency and their optical behaviour, particularly on samples characterized by restricted geometries, like channels, which preclude the use of conventional optical absorption measurements.

On coloured LiF strips, photoemission measurements are generally performed by exciting the samples with the 458 nm line of an argon laser impinging on the irradiated sample surface and collecting the PL signal, properly filtered, by a photomultiplier (S20 response) placed on the same side of the sample. In this frontal geometry, the unconfined PL laterally emitted by the coloured strip is measured along the direction normal to the sample surface. The same set-up, in a perpendicular geometry between pumping source and detector, has been used to determine the optical gain coefficients g by measuring the amplified spontaneous emission (ASE) signal [15]. In this perpendicular geometry, the guided PL that comes out of the strip end is collected.

Fig.1 shows the RT photoemission spectra, acquired in the frontal geometry, of 25 μm wide coloured strips, irradiated by 12 keV electrons with a dose of 6×10^{-3} C/cm^2 in the same experimental conditions, at the surface of a LiF single crystal and of a LiF film of thickness 1.8 μm grown on a glass substrate at a temperature of 250°C. They consist of two broad emission bands, peaking at around 540 nm and 680 nm, typical of F_3^+ and F_2 CCs [8]; their intensities are about four times higher in film than in bulk, although their spectral features are quite similar. The increase in the visible photoemission signal from coloured LiF films is attributed to larger concentrations of F_3^+ and F_2 active defects. As a matter of fact, it is accompanied by a higher intensity of their superimposed absorption bands, located at 450 nm, as it has been measured on the same samples of Fig.1, irradiated on large areas with a dose of 10^{-4} C/cm^2. Their absorption spectra are reported in Fig.2: in the case of LiF film, this main absorption is superimposed to the interference fringes due to the refractive index difference between the LiF film (<1.39) and glass substrate (~1.5).

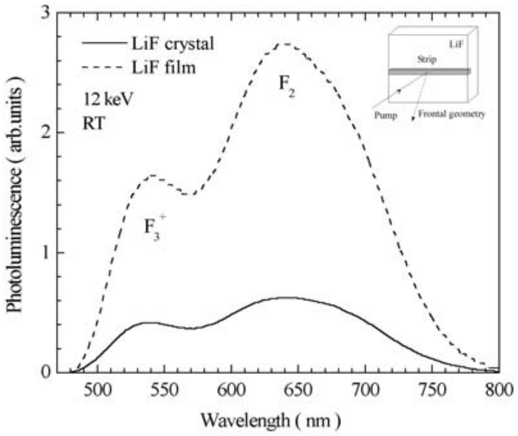

Fig.1. RT PL spectra of 25 μm wide strips coloured by 12 keV electrons in a LiF crystal (solid) and film (dashed) irradiated at RT in the same conditions. The frontal collecting scheme is reported in the inset.

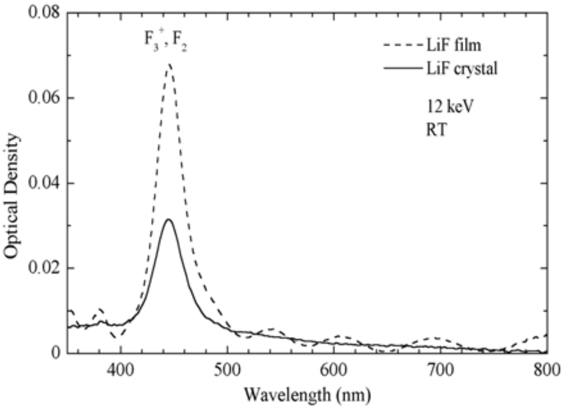

Fig.2. RT absorption spectra of a LiF crystal (solid) and the film of Fig.1 coloured by 12 keV electrons in the same conditions on a large area. They are roughly corrected for the absorption of the unirradiated part.

In terms of gain, ASE occurs when the spontaneous emission from a distribution of inverted systems is amplified by stimulated emission without the help of a resonant cavity. Sizeable optical gain coefficients, several cm^{-1}, have been measured at RT on the coloured strip induced by 12 keV electron beams on the LiF film thermally evaporated on glass substrates of Fig.1 [13] by means of the standard ASE technique [15]. At constant power density of the exciting beam, the photoemission spectra are collected in a perpendicular geometry between pumping source and detector as a function of the pumped length of the coloured strip, which is varied by moving a screen perpendicular to the exciting beam. Spontaneous luminescence was amplified (stimulated luminescence) as it passed through the excited volume to the edge of the sample. The excited medium can be treated as a simple one-dimensional optical amplifier. The optical gain has been determined by monitoring the ASE signal as function of the pumped length. The output signals, measured around the emission peaks of F_3^+ (540 nm) and F_2 (680 nm), show an exponential behaviour. The gain coefficients were computed in all the emission spectral range and amplification has been observed in the two intervals 510-590 nm and 620-710 nm, which are centred on the emission peaks of both the laser active defects.

On several strip waveguides, coloured by 12 keV electrons on LiF crystals at doses of 2.2×10^{-3}, 3.3×10^{-3} and 8.8×10^{-3} C/cm^2, emission spectra were acquired in the perpendicular geometry as a function of the pump power density in a quasi c.w. regime, keeping constant the length of the excited strip. Around the peak of the F_2 emission band, the signal supra-linearly grows by increasing the pump power [16]. Sizeable values of the gain coefficient, several cm^{-1}, have been obtained again. Their numerical values increase with increasing irradiation dose. The same supra-linear behaviour was observed in the wavelength range extending from 580 to 730 nm [17]. On these confining structures, a strong reduction of the F_3^+ emission band and a narrowing of the F_2 one are observed in the perpendicular geometry, as shown in Fig.3, where their ASE spectra are compared with the ASE spectrum of the coloured strip on LiF film in Fig.1. Both these effects are more pronounced for the coloured waveguides irradiated at higher doses. The typical F_3^+ and F_2 broad PL bands, due to the light isotropically emitted, are spectrally modified due to confinement effects at the interfaces with the lower refractive index surrounding media. The great differences they undergo upon waveguiding should be related to the wavelength-dependent behaviour of the complex refractive index of the irradiated layers [7,9].

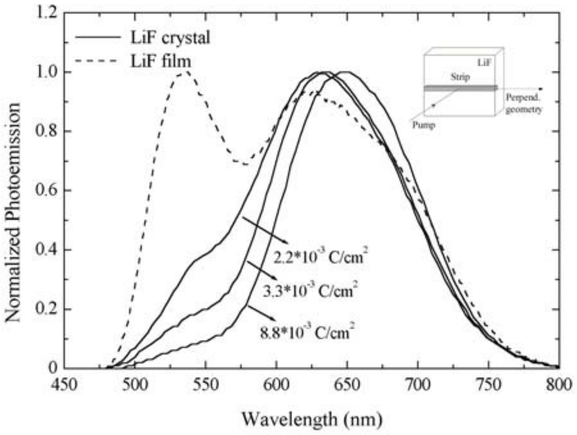

Fig.3. RT normalized ASE spectra of three strip waveguides coloured by 12 keV electrons at different doses in a LiF crystal (solid) and of the strip on film of Fig.1, measured for an excitation length of 2.5 mm at a power density of 0.2 W/cm^2. The perpendicular collecting scheme is reported in the inset.

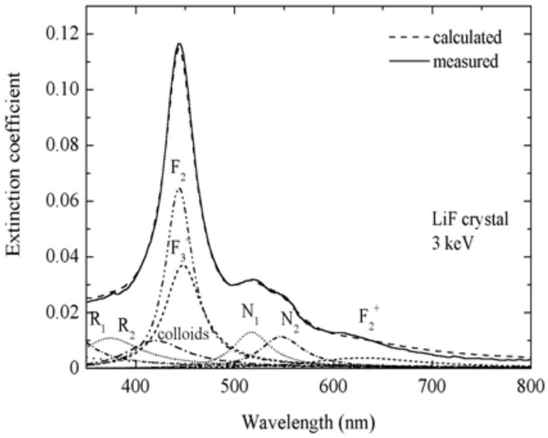

Fig.4. Extinction coefficient of a LiF crystal coloured by 3 keV electrons at RT on a large area. The measured curve is obtained through ellipsometry, the calculated one from spectrophotometry through a model [7], which allows computing the single contributions of different kinds on defects, is also plotted.

Higher defect densities imply the improvement of the confining properties of the waveguides; on the other hand, the presence of different aggregate CCs causes optical losses due to emitted light re-absorption. In the coloured LiF based amplifying strips under investigation, the net gain coefficient is independent from the position along the medium in the direction of amplification. It is expressed as the difference between the gain coefficient g, dependent on the luminescence process, and the losses α, due both to the optical quality of the active media as well as to other processes, like re-absorption. The absence of amplification in the green is probably due to parasitic centres, like the F_4 ones, which possess associated N_1 and N_2 absorption bands, peaked around 515 and 545 nm. Their presence is not distinguishable in the absorption spectra of Fig.2, but it is clearly appreciable in the extinction coefficient of the irradiated part of a LiF crystals heavily bombarded by low energy electrons with a dose of 10^{-1} C/cm^2 on a large area, shown in Fig.4. Its experimental values are in excellent agreement with the calculated ones.

Although the optical losses for light scattering, related to the quality of the hosting matrix, are higher in polycrystalline LiF films [12], they show less optical losses in the green spectral range than single crystals irradiated in similar conditions. This behaviour should be ascribed to the polycrystalline nature of LiF films, which increases the formation efficiency of visible active CCs, and also reduces the final densities of larger complex aggregate defects, like F_4. Further investigation could allow for a selective generation of the requested defects and therefore an improvement of light emission performance.

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