Thermal diffusivity of BaLiF$_3$ crystals at low temperature

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Abstract

BaLiF$_3$ crystals doped with transition metals ions are a promising laser active medium when cooled to low temperatures, near 77K. In this work, the thermal diffusivity of pure BaLiF$_3$ single crystals was obtained at 80K using the photoacoustic two-beam phase lag method, which consists in measuring the relative phase-lag for the photoacoustic signal between rear and front surface illumination at a single modulation frequency.

Introduction

When a certain region of a sample is heated, the thermal energy due to this heating trends to distribute uniformly throughout this material until the thermal equilibrium is reached. Considering that the main mechanism of heat propagation is the conduction mechanism, the thermal parameter called thermal diffusivity (α) provides the rate at which the heat flows through a medium. The thermal diffusivity is also related to the thermal conductivity by:

$$\alpha = k/\rho.c$$

where c is the specific heat at constant pressure and ρ is the density of the medium.

The knowledge of the thermal diffusivity value is very important for the laser engineering prototypes development and for the crystal growth process.

BaLiF$_3$ crystals are important materials since, when doped with transition metal ions, they are a potential new laser host.

BaLiF$_3$ crystals are an "inverse" perovskite with cubic structure (space group O$_h^1$ - Pm3m) where the monovalent Li$^+$ ion is at the center of six F$^-$ ions octahedron, whereas the Ba^{2+} divalent ions are in the 12-fold environment site. The dopants (transitions metals ions) can be incorporated to the BaLiF$_3$ matrix by substituting the Li$^+$ ions.

Previous studies on transition metal ions doped BaLiF$_3$ crystals such as BaLiF$_3$:Ni^{2+} [1] and BaLiF$_3$:Co^{2+} [2] show that they can emit in the near infrared region in a broad band centered at 1.5 μm and 1.6 μm respectively. Once the possibility of laser action in theses crystals at low temperature [3] was observed, the main subject of this work is to determine the thermal diffusivity of BaLiF$_3$ crystals at liquid nitrogen temperature. For this purpose, the photoacoustic two-beam phase lag method [4], that consists in illuminating the sample on the front and rear surfaces, at the same light modulation frequency, was applied.

This method is based on the theory developed by Rosencwaig and Gersho (RG-Model) [5] in 1976, where the exciting light produces a local heating due to the non-radiative decay process. This local heating will flow through the sample by conduction and will reach the interface with the medium. If the sample is placed inside a gas closed chamber (photoacoustic cell), the surrounding gas will be heated too, starting a piston motion inside the cell, producing then the photoacoustic signal. This photoacoustic signal is detected by a microphone and amplified by a lock-in amplifier.

In the photoacoustic two-beam phase lag method, the photoacoustic signals phase is measured when the sample is illuminated in the front (Ψ_F) and rear surface(Ψ_R). Once the phase-lag ($\Delta\Psi=\Psi_F-\Psi_R$) is obtained, the thermal diffusivity is determined by:

$$\tan(\Delta\Psi) = \tan\left(\sqrt{\frac{l^2\pi f}{\alpha}}\right)\tanh\left(\sqrt{\frac{l^2\pi f}{\alpha}}\right)$$

where l is the sample thickness and f is the excitation light modulation frequency.

The restrictions of the applied methodology are to consider that all the incident light is absorbed at the surface, and to assume that the heat flux into the surrounding air is negligible. For this reason, optically opaque absorbing layers had to be provided, by thin film deposition, for example.

Experimental

The experimental setup is shown in fig. 1. An Argon laser was used to excite the sample, in the multiline mode whith laser lines between 457.9 nm and 514.5 nm. The most intense laser lines are at 514.5 nm and 488.0 nm. This laser beam is modulated by a mechanical chopper and directed through an objective lens before reaching the optical fiber. The photoacoustic cell is placed inside the nitrogen bath. Each fiber guides the exciting light to the frontal and rear surfaces. To prevent the solidification of water drops from the air and to rise the thermal coupling, a previous vacuum is done and Helium gas is inserted inside the chamber. The microphone is placed outside the chamber to prevent its damage; the photoacoustic wave passes through a resonance tube to reach the microphone. A rubber ball with Helium gas inside keeps the pressure constant inside the cell while the temperature is lowered. A digital multimeter gives the temperature inside the photoacoustic cell.

The BaLiF$_3$ crystal used in this experiment was grown by the Czochralski [6] method with fixed growth direction [111]. The sample area was 1cm^2 and the thickness was 480 μm. The value of thermal diffusivity determined for this sample at 80K is 0.048(4) cm^2/s.
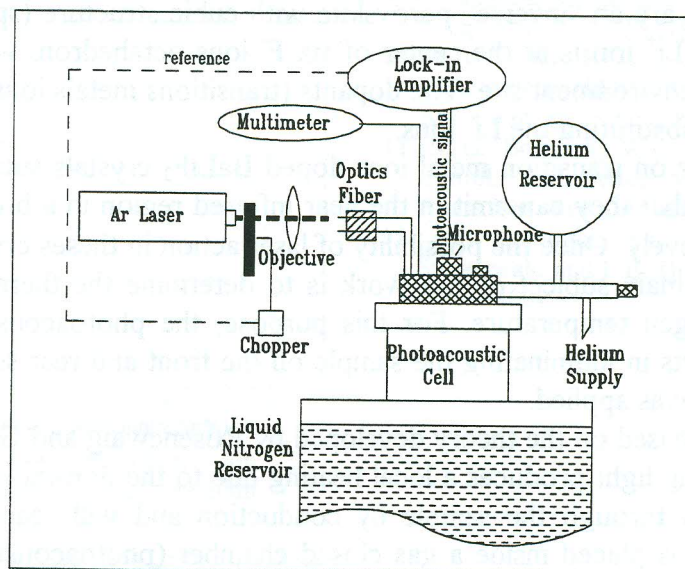

Fig 1. Experimental set-up.

Discussion

When cooling the crystals, the trend is the reduction of the thermal agitation amplitude of the atoms. This fact results in an increase of the phonons average free path. So, it is expected the value of the thermal diffusivity to be higher for low temperatures than at room temperature. The experimental values obtained agree with the theoretical prevision.

The determination of the thermal diffusivity of the doped samples, at low temperature, is on the way. This results will help to understand the mechanisms involved in the low temperatures process.

Table 1. Experimental results

	$\alpha(cm^2/s)$ T=300K	$\alpha(cm^2/s)$ T=80K
$BaLiF_3^a$	0,037(5)	-
$BaLiF_3$	0,039(1)	0,048(3)

ª Ref.[7]

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