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Light scattering in gadolinium molybdate due to domain structure

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Abstract. Integral light scattering from $Gd_2(MoO_4)_3$ crystals due to domain structure in the phase transition region has been investigated. The size and anisotropy of the scattering centres are estimated.

1. Introduction

Gadolinium molybdate $Gd_2(MoO_4)_3$ (GMO) is an improper ferroelectric with a phase transition (PT) temperature T_c of 432 K. It is a representative of the family of rare-earth molybdates [1–6]. Unusual physical properties have been revealed in this class of compounds [4–9] which makes it necessary to subject them to further studies.

Of particular interest both for practical applications and for the analysis of the mechanism of the ferroelectric phase and polarisation switching are the investigations of specific features of the domain structure in the PT region.

In the last few years, light scattering (LS) has found increasing use in studying the crystal structure heterogeneities, including the domain structure of ferroelectrics. This non-contact method offers significant advantages over the previous methods [10–13], as it allows one to study crystals without damaging their surfaces.

It has been shown in a number of reports that the angular dependence of scattering in a single crystal can provide information on the size, shape and crystallographic orientation of impurity centres in a matrix, whereas the temperature dependence of the scattered-light intensity may be used to obtain information on their origin [14–18].

The present paper deals with the domain structure of ferroelectric GMO crystals by the LS method. GMO is the most typical representative of the family of rare-earth oxides.

2. Experimental procedure and treatment of crystals

The GMO crystal used in this investigation has been grown by the Czochralski method and had a PT temperature T_c of 432 K determined on the basis of dielectric measurements. The GMO specimens for LS measurements were cut from one boule in the desired orientation with geometrical axes parallel to the [100], [001] and [110] directions. Samples of cylindrical shape 8 mm in diameter and 15 mm long were used to eliminate

refraction of scattered light in the plane of scattering. They were annealed to relieve elastic stresses.

The integral LS was measured with the help of a Gaertner L119x ellipsometer. The optical diagram of the instrument is shown in figure 1. It consists of a photodetector, an amplification and accumulation system, and a plotter. We could measure LS amounting to 10^{-8} of the incident radiation. Measurements were carried out in a vacuum of 10^{-7} Torr. Glan-Thompson prisms were used as polarisers. Laser light at $\lambda = 6328 \text{ \AA}$ was used as the radiation source. It passed through a collimator and a polariser before impinging on the crystal. The scattered light passed through an analyser and the telescope and was focused on a photomultiplier. The specimen chamber allowed us to make measurements over a wide temperature range ($T = 77\text{--}600 \text{ K}$) with stabilisation better than $0.01 \text{ }^\circ\text{C}$. The specimen could be rotated through 360° about its geometrical axis.

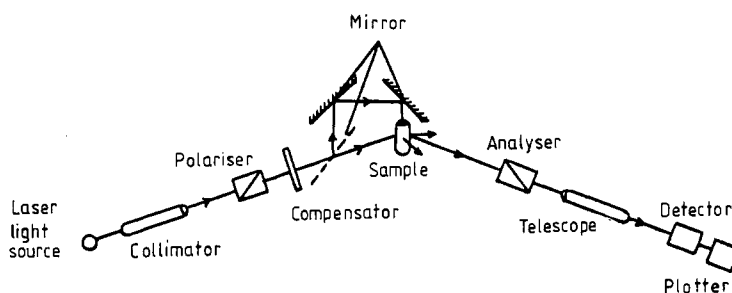


Figure 1. The components of the ellipsometer.

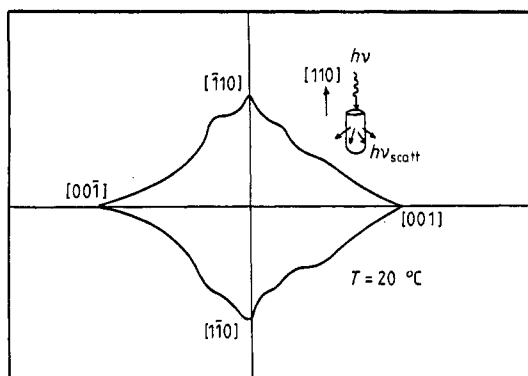


Figure 2. LS diagram for a multiple-domain GMO crystal.

3. Experimental results

Figure 2 gives the diagram of the LS of multiple-domain GMO specimens in the case of fundamental-wave (FW) propagation in the $[110]$ direction. Measurements have shown that in this case the LS intensity is one order of magnitude higher than that in the case of FW propagation along $[100]$ and $[001]$. An anisotropy of the scattering intensity is

observed which depends on temperature. Figure 3 shows the temperature dependence of the LS intensity for multiple-domain GMO (curve B). It is seen from this figure that the LS intensity in multiple-domain GMO increases drastically as the PT point is approached. In the ferroelectric phase a tendency for slow growth of the LS intensity remains.

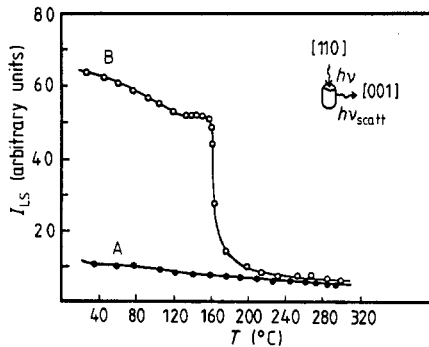


Figure 3. Temperature dependence of integral LS in GMO crystals: curve A, single-domain crystal; curve B, multiple-domain crystal.

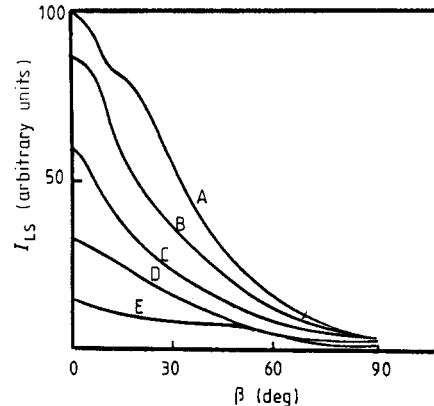


Figure 4. Angular dependence of integral LS in multiple-domain GMO for light propagation along [110] at different temperatures T : curve A, 300 K; curve B, 350 K; curve C, 380 K; curve D, 400 K; curve E, 460 K.

In single-domain GMO specimens, the LS intensity does not vary substantially with temperature. We have also obtained angular diagrams of multiple-domain GMO crystals, presented in figure 4. As shown from the figure, the anisotropy of LS intensity decreases as the temperature increases and disappears completely above T_c .

4. Discussion of results

The results demonstrate that the scattering centres are very anisotropic. Our observations of the integral LS, and its temperature and angular dependence, allows us to assume that the scattering centres in GMO are rod like with a length larger than the optical wavelength of the incident light. The high scattering intensity for [110] FW propagation and its temperature dependence indicate preferred orientation of the rods along the polar axis of the crystal.

Assuming that this LS in GMO can be described by the Rayleigh–Hans formula for long rods ($L \gg \lambda$) [19], we calculated the size of the scattering centres from the angular distribution of the temperature-dependent component of the scattering (figures 2–4). The integral LS in GMO in the PT region and in the ferroelectric phase is much more complex than the phenomena described by the Rayleigh–Hans theory. Nevertheless, the calculation shows satisfactory agreement between the sizes of the scattering rods (8–10 μm) and the domain sizes determined by other experiments [12, 13]. We established that the LS intensity is defined to a great extent by the unipolarity of the crystals. All the above special features are inherent only in multiple-domain crystals. In single-domain

crystals the LS intensity shows no anomaly. The intensities are similar for all propagation directions. The anisotropic nature of the scattering is, however, maintained.

These investigations show that the integral LS in GMO and the anomalous behaviour of its temperature dependence in the PT region arise because the crystal breaks up into domains and because of the gradient of optical constants at the domain boundaries.

Acknowledgments

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