

CHARACTERIZATION OF THIN ZnO FILM BY OPTICAL SECOND HARMONIC GENERATION: EXPERIMENT AND THEORY

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Abstract—Thin polycrystalline zinc oxide film deposited on glass substrate have been investigated by optical second harmonic generation (SHG). The intensity of SHG in dependence of rotation angle of fundamental beam polarization for several incident angles has been measured. Developed theoretical model taking into account multiple reflections of light into the sample allows us to reveal a set of effective characteristics of film.

1. INTRODUCTION

The SHG by thin films is frequently used to study their nonlinear optical properties which are conditioned by the characteristics of corresponding textures. The preference of this method is caused by high sensitivity of nonlinear response to local symmetry properties when anisotropy of linear response is negligible. The dependence of SHG intensity measured in different experimental geometries provide a few integral responses containing the information about the film structure (for example, about common properties of ensemble of micro-crystals disordered with respect to one another). In principle, it allows one to extract statistically averaged (effective) characteristics of textured films.

We represent the results of experimental and theoretical investigations of SHG by thin polycrystalline ZnO film deposited on glass substrate. The SHG intensity has been measured as a function of polarization angle rotation φ_i at several fixed incident angle Θ_i . The sufficient peculiarity of our theoretical consideration is that we take into account the effect of multiple reflections of both the basic beam and SH one in the film and substrate. In our knowledge such approach is proposed for the first time.

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2. EXPERIMENT

Transparent ZnO film has been prepared on glass substrate by chemical vapour deposition method [1]. Electronic microscopy of sample shows that it has a roughness surface and consists of seeds with different sizes. The thickness of film varies within $0.1\mu\text{m}$ – $0.3\mu\text{m}$. It is sufficiently less than the magnitude of the coherence length ($3\mu\text{m}$) of bulk ZnO [2]. The investigation of SHG in transmission has been carried out on a standard experimental setup. The pump radiation for SH measurements was supplied by a pulsed YAG:Nd³⁺ laser ($\lambda_0 = 1064\text{ nm}$, pulse duration 20 ns, repetition rate 14 Hz). To avoid optical damage of the film the energy of pulse has been maintained below 10 mJ. At fixed incident angle we have measured the intensity of P-component of transmitted SH in dependence of rotation of fundamental wave polarization.

3. THE MODEL

In our calculations we consider a real polycrystalline film as fictitious thin single plate which is characterized by a set of efficient constants which should be determined by fit. By such a way we imitate the integral optical properties of mixture averaged over the ensemble of structural seeds with unknown distribution function. The disordering of micro-crystals leads to lowering of effective symmetry of medium, i.e. to increasing of the number of independent components of linear and nonlinear optical tensors. The main simplification involved in our model is that we propose to arise the symmetry of the effective medium up to the initial one of the bulk single crystal ZnO (C_{6v}) to avoid over-parameterization of our problem on this first step. So, we suppose that anisotropy caused by