Optical Materials 31 (2009) 1151–1154

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Optical constants and dispersion behavior of $PbMg_{1/3}Nb_{2/3}O_3$ single crystals

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ARTICLE INFO

Article history:
Received 5 September 2008
Received in revised form 11 December 2008
Accepted 12 December 2008
Available online 24 January 2009

PACS:
78.20.Ci
46.40.Cd
42.25.Lc

Keywords:
PMN single crystals
Refractive indices
Sellmeier optical coefficients
Transmission ratio

ABSTRACT

The optical properties of $PbMg_{1/3}Nb_{2/3}O_3$(PMN) single crystals in the wavelength range of 300–1700 nm have been investigated using Variable Angle Spectroscopic Ellipsometer (VASE). The refractive indices and extinction coefficients have been obtained. The optical band gap energy of PMN single crystal was derived to be about 3.41 eV. The Sellmeier optical coefficients E_0, λ_0, S_0, and E_d were calculated by fitting the single-term oscillator equation. The transmission ratio of PMN single crystal ranging from 200 to 2200 nm has also been investigated using UV–visible–NIR spectrometer. The transmission ratio is above 60% in near IR region and the reflection loss is calculated to be about 20%.

1. Introduction

Recently, relaxor ferroelectrics have been investigated for a variety of electro-optic properties; good electro-optic switching times and modest half-wave voltages (V_π), could make these materials viable alternatives to the (Pb,La)(Zr,Ti)O,(PLZT) compositions conventionally used in electro-optic devices [1–5]. $PbMg_{1/3}Nb_{2/3}O_3$(PMN) is a typical relax ferroelectric with perovskite type, pseudo cubic crystal structure and belongs to Pm3m space group in its paraelectric phase [6,7]. The dielectric properties of PMN have been widely investigated in both the single crystals, polycrystalline ceramic and films forms [8–13]. As a typical relax ferroelectric, most of the research has concentrated on the structure of PMN [14–18]. However, the optical properties of PMN single crystals are seldom reported.

In this article, the optical properties of PMN single crystals were intensively studied. The refractive indices and extinction coefficients have been measured by VASE. The optical band gap energy of the PMN was derived to be about 3.41 eV. The Sellmeier optical coefficients E_0, λ_0, S_0, and E_d were calculated by fitting the single-term oscillator equation. The transmission characterization of PMN single crystals was investigated by a JASCO's V-570 UV–visible–NIR spectrometer. The reflection loss is also discussed.

2. Experimental procedure

Large size and high quality PMN single crystals were grown using modified Bridgman method [19,20]. The single crystal samples were oriented using X-ray diffractometer. For VASE measurement, the sample was cut along <001> direction. The surface for light reflection was polished using alumina and diamond polishing compounds to achieve a final optical polish, with specular reflection. The ellipsometric measurement was carried out at room temperature in the wavelength range of 300–1700 nm, with 5 nm steps, by a VASE with synchronously rotating polarizer and analyzer, at incidence angle of 65°. For transmission measurement, the sample was also cut along <001> direction. The surfaces for light transmission were polished to achieve a final optical polish.

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doi:10.1016/j.optmat.2008.12.005

3. Results and discussion

3.1. Refractive indices and dispersion equations

VASE measures the ellipsometric angles, Ψ and Δ, defined by the ellipsometric ratio, ρ, as a function of photon energy, E, (or wavelength) and angle of incidence [21],

$$\rho = \frac{Rp}{Rs} = \tan\Psi \exp(i\Delta) \tag{1}$$

where Rp/Rs is the ratio of complex Fresnel reflection coefficients for p- and s- polarized light, respectively. Rp and Rs are complex functions of the refractive index n and extinction coefficient k. For bulk isotropic samples investigated here, their complex refractive index ($\tilde{n} = n + ik$) can be revealed automatically by comparing the computer modeling data with the experimental data. Fig. 1 and Fig. 2 show the spectral dependence of the refractive indices and extinction coefficients at room temperature of PMN single crystals, respectively. Similar to other ABO_3-type perovskite structure compounds, PMN single crystal has large refractive indices and obvious dispersion. The extinction coefficients decrease with increasing wavelength, which indicates that PMN single crystals are transparent in the near IR region. Because of the similar BO_6 octahedron unit structure, they have analogous energy band structure determining the refractive indices [22–24].

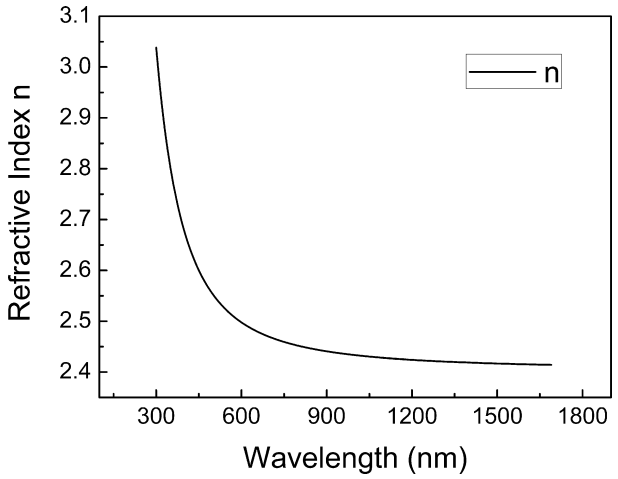

Fig. 1. The refractive indices of PMN single crystals measured at different wavelengths.

From the well known simple dispersion theory, the refractive index can be given by typical Sellmeier dispersion equation,

$$n^2 = A + \frac{B}{\lambda^2 - C} - D\lambda^2 \tag{2}$$

where A, B, C and D are all constants and λ is wavelength in microns. These constants can be obtained by the least squares fitting of Eq. (2) using the data obtained by VASE. Thus, the Sellmeier dispersion equation of n for PMN is

$$n^2 = 5.71717 + \frac{0.16661}{\lambda^2 - 0.0433} + 0.02215\lambda^2 \tag{3}$$

By the dispersion equation, the refractive indices of PMN crystal can be calculated. For instance, at the wavelength of 514.5 nm argon laser, $n = 2.520$.

To determine the band gap energy, the absorption coefficient α of the single crystals were derived from the extinction coefficients k using

$$\alpha = 4\pi k/\lambda \tag{4}$$

The band gap of the PMN single crystal was deduced from the absorption coefficient α and the energy of the incident light $h\nu$ using the Tauc equation,

$$(\alpha h\nu)^2 = Q(h\nu - E_g) \tag{5}$$

where Q is a constant and Eg is the band gap energy. Extrapolation of the $(\alpha h\nu)^2$ data (Fig. 3) gives the band gap energy of PMN 3.41 eV, which is a little larger than that of PMN-xPT single crystals [3].

3.2. The Sellmeier optical coefficient

Although Eq. (3) can precisely predict refractive indices at different wavelength, the coefficients in it do not have any special physical significance. By the single-oscillator approximation, Wimple and Didomenico developed a single-term Sellmeier relation [2,25,26],

$$n^2 - 1 = \frac{S_0\lambda_0^2}{1 - \lambda_0^2/\lambda^2} = \frac{E_d E_0}{E_0^2 - E^2} \tag{6}$$

where n is the refractive index at λ and E, which are the wavelength and energy of the incident light, respectively. λ_0 is the average oscillator position, S_0 is the average oscillator strength, E_0 is the single-

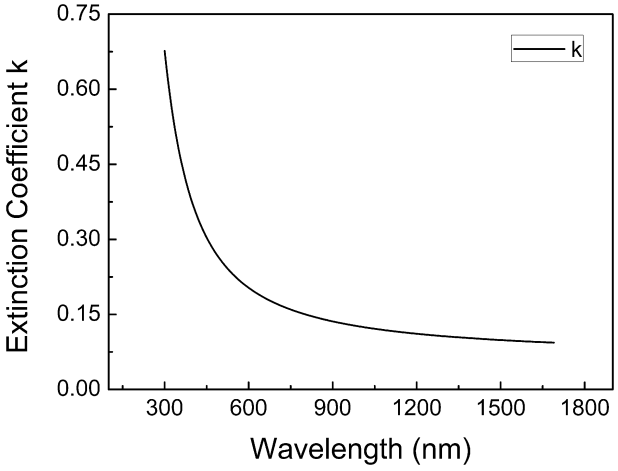

Fig. 2. Spectral dependence of extinction coefficients at room temperature of PMN single crystals.

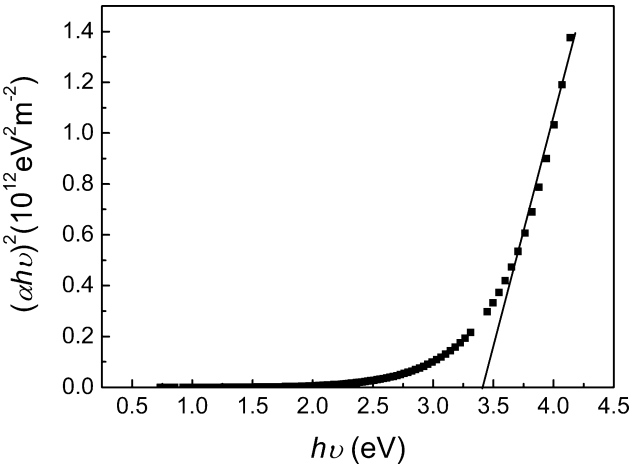

Fig. 3. The linear fitting curves of $(n^2-1)-1$ dependent on λ^{-2} for PMN single crystals.