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ELECTROOPTIC INVESTIGATIONS IN AMMONIUM HYDROGEN SELENATE (NH_4HSeO_4)

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Abstract We present optical and electrooptical characterization of ammonium hydrogen selenate (NH_4HSeO_4). The properties of this new electrooptic crystal are compared with those of rubidium hydrogen selenate (RbHSeO_4).

INTRODUCTION

The use of highly efficient devices for the treatment of the optical signal continuously drives the research of new optical materials for various specific applications. Thus, demands for electrooptic (EO) modulators using the Pockels effect need high optical quality material with large EO coefficients and other requirements such as a large transmission spectrum. Especially interesting for EO and non linear optic applications¹, the group of the hydrogen-bonded compounds, like KDP or ADP, has been largely studied and widely used in different commercially available systems for a long time. We have shown² that rubidium hydrogen selenate (RbHSeO_4), which belongs to the same family as KDP or ADP, exhibits very large EO coefficients as well as a small thermo-optic coefficient and therefore seems to be promising for opto-electronic devices. In this paper we present the results obtained from the optical and EO characterization of a second selenate crystal : the ammonium hydrogen selenate (NH_4HSeO_4).

SYMMETRY CONSIDERATIONS

Ammonium hydrogen selenate (AHSe) is a crystal which is paraelectric and presents the monoclinic symmetry³ ($I2$) at room temperature. It possess ferroelectric properties below 261 K. In the configurations used in our EO experiments, where the electric field

E is applied along the z axis, the index ellipsoid of the crystal under application of an electric field is given by :

$$\left(\frac{1}{n_x^2} + r_{13} E\right) x^2 + \left(\frac{1}{n_y^2} + r_{23} E\right) y^2 + \left(\frac{1}{n_z^2} + r_{33} E\right) z^2 + 2r_{63} E xy = 1 \quad (1)$$

where n_x , n_y and n_z are the refractive indices in the principal axis system of the crystal without applied field and r_{13} , r_{23} , r_{33} and r_{63} are the EO coefficients. For a light beam propagating along the x axis, equation (1) becomes :

$$\left(\frac{1}{n_y^2} + r_{23} E\right) y^2 + \left(\frac{1}{n_z^2} + r_{33} E\right) z^2 = 1 \quad (2)$$

We can rewrite equation (2) as :

$$\frac{y^2}{n_y'^2} + \frac{z^2}{n_z'^2} = 1 \quad (3)$$

where $n_y' = n_y - (n_y^3 r_{23} E)/2$ and $n_z' = n_z - (n_z^3 r_{33} E)/2$

Finally, the electric field induced birefringence $\delta\Delta n_{yz}$ is given by :

$$\delta\Delta n_{yz}(E) = -\frac{1}{2} n_y^3 r_c' E \quad \text{with} \quad r_c' = r_{23} - \frac{n_z^3}{n_y^3} r_{33} \quad (4)$$

In the same way, we can deduce the electric field induced birefringence $\delta\Delta n_{xz}$ obtained for a light propagating along the y axis :

$$\delta\Delta n_{xz}(E) = -\frac{1}{2} n_x^3 r_c' E \quad \text{with} \quad r_c' = r_{13} - \frac{n_z^3}{n_x^3} r_{33} \quad (5)$$

Considering that the electric field induced phase shift between the two light polarization components is $\Gamma = \frac{2\pi L}{\lambda} \delta\Delta n$ and that $V = E d$, we can rewrite equation (4) and (5) as :

$$\Gamma_{yz} = \frac{\pi L}{\lambda d} n_y^3 r_c' V \quad , \quad \Gamma_{xz} = \frac{\pi L}{\lambda d} n_x^3 r_c' V \quad (6)$$

where d is the inter electrodes spacing, L is the crystal length along the laser beam propagation direction, λ is the wavelength of the laser beam and V the voltage amplitude.

EXPERIMENTAL RESULTS

We use a good optical quality crystal, with $7.11 \times 2.44 \times 1.5 \text{ mm}^3$ in size. The crystal is placed into an experimental setup described previously⁴, which yields the transmission factor between output I and input I_0 laser intensities :

$$T = \frac{I}{I_0} = \frac{1}{2} [1 - \sin(\Gamma - 2\beta)] \quad (7)$$

where β is the analyser angle. If an ac electric field $E = E_m \sin \omega_m t$ is applied on the crystal, the phase shift Γ can be expressed as :

$$\Gamma = \Gamma(0) + \Gamma_m \sin \omega_m t \quad (8)$$

where $\Gamma(0)$ is the phase shift corresponding to the spontaneous birefringence of the crystal and Γ_m is induced by the ac field at the ω_m frequency. The phase shift Γ_m due to the EO effect is assumed to be very small. Moreover, if the azimuthal position of the analyser is adjusted at a particular point where $\beta = \Gamma(0)/2 + k\pi/2$, equation (7) becomes, after expansion in Bessel functions limited to the first order :

$$T = \frac{1}{2} \pm \frac{\Gamma_m}{2} \sin \omega_m t \quad (9)$$

The second term of the equation (9) corresponds to the output modulated signal defined by the modulation coefficient m as :

$$m = \frac{2J_\omega}{I_0} = \Gamma_m \quad (10)$$

where J_ω is the amplitude of the output modulated signal. Using equations (6) and (8), we can derive the value of r_c and r'_c as :

$$r_c = -\frac{\lambda d m}{\pi L n_y^3 V} \quad , \quad r'_c = \frac{\lambda d m}{\pi L n_x^3 V} \quad (11)$$

Measurements are performed at room temperature ($T = 23.1 \text{ }^\circ\text{C}$) using the 633 nm laser wavelength of a HeNe laser, and an ac voltage of 50 volts amplitude with the frequency varying between 500 Hz and 200 kHz. Results are plotted in figure 1. We have measured the refractive indices, for wavelengths corresponding to visible spectrum : $n_x \sim 1.56$, $n_y \sim 1.54$, $n_z \sim 1.58$. Instead of the EO coefficient r we plot the figure of merit $n^3 r$, deduced from equation (11). The value of the figure of merit $n_x^3 r'_c$ is larger than the value of $n_y^3 r_c$ but exhibits the same behavior as a function of the modulation frequency. The values of the two figures of merit present a large decrease with increasing frequency. Thus, for a modulation frequency varying from 200 Hz up to 10 kHz, $n_x^3 r'_c = 115 \text{ pm/V}$ and $n_y^3 r_c = 5.9 \text{ pm/V}$ whereas they become respectively 40 pm/V and 2 pm/V for a frequency of 100 kHz. These last values are comparable to those of ADP. Even if they are large at lower frequencies (200 Hz), the values of $n_x^3 r'_c$ and $n_y^3 r_c$ are much lower than this of RbHSeO_4^2 which is 13240 pm/V.

In order to complete the characterization of AHSe we have measured its transmission spectrum by using the CARY 17 setup. Figure 2 shows this spectrum obtained, for a non polarized light and for a crystal length of 1.5 mm in the z direction (corresponding to the light propagation direction). We can notice that the transmission spectrum is wider in the infrared compared to ADP^5 . Indeed, for a wavelength of 2000

nm, the value of the transmission for ADP is only a few percents compared to at least 20 percents for AHSe. Nevertheless, the maximal transmission value of AHSe in our measurements is only 60 percents. It may be attributed to surface quality since the high hygroscopy of AHSe damages the faces of the crystal.

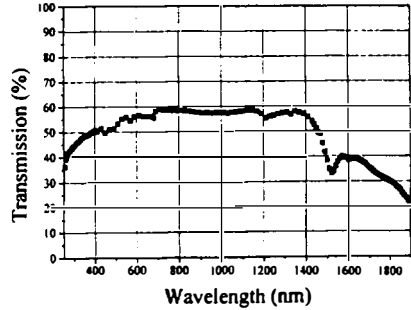
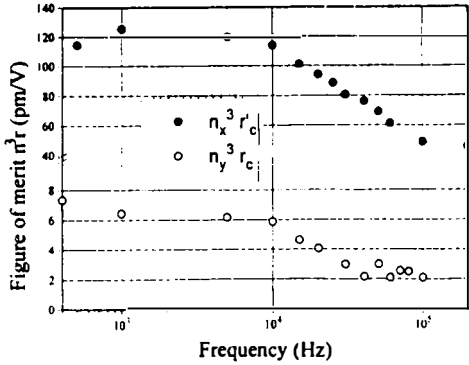


FIGURE 1 Modulation frequency behaviour of $n_x^3 r'_c$ and $n_y^3 r_c$ in NH_4HSeO_4

FIGURE 2 Transmission spectrum of NH_4HSeO_4

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