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New material with strong electro-optic effect: Rubidium hydrogen selenate ($RbHSeO_4$)

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A very large electro-optic Pockels coefficient is reported in a crystal of rubidium hydrogen selenate. This new material is found to be promising for modulation applications because of its very low half-wave voltage (~27 V) which is determined.

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 wideband electro-optic modulators using the Pockels effect need high optical quality materials with large electro-optical coefficients and other requirements such as low dielectric losses and permittivity.

Especially interesting for electro-optic and nonlinear optic applications,[1] the group of the hydrogen-bonded compounds has been largely studied and well known for a long time; especially, the KDP (KH_2PO_4) and ADP ($NH_4H_2PO_4$) crystals are widely used in different commercially available systems. This letter concerns the first investigation of the electro-optical properties in a single crystal of rubidium hydrogen selenate which is less studied and belongs to the same family as KDP and ADP.

Very large electro-optic coefficients are obtained for the first time in this new material which seems to be promising and may be very interesting for opto-electronic devices, particularly for modulation applications. In addition, the low dielectric permittivity and the weak thermo-optic coefficient, are other advantages of this material in regard to requirements for modulators.

Rubidium hydrogen selenate $RbHSeO_4$ is a crystal which presents the triclinic symmetry at room temperature, with pseudo-orthorhombic lattice parameters.[2] It possesses ferroelastic, pyroelectric, and ferroelectric properties, which have been widely studied.[3,4] Most investigations in this crystal concern the study of phase transitions in relation with these properties.

The investigation of the electro-optic properties in triclinic crystals is not simple since, owing to the symmetry, all the components of the electro-optic tensor are allowed.

In the configuration used in our experiments, where **E** is applied along the y axis and the light propagation is along the z axis the index ellipsoid of the crystal in the presence of an applied electric field is given by

$$(1/n_1^2+r_{12}E)x^2+(1/n_2^2+r_{22}E)y^2+2r_{62}Exy=1, \quad (1)$$

where n_1, n_2 are the refractive indices in the principal axis system of the crystal without applied field and r_{12}, r_{22}, and r_{62} the electro-optic coefficients. According to the crossed term which appears in Eq. (1), the indicatrix of the crystal under the electric field undergoes a rotation with an angle θ around the z axis given by

$$tg2\theta=(2r_{62}E)/[(1/n_1^2-1/n_2^2)+(r_{12}-r_{22})E]. \quad (2)$$

In such a case one generally finds a new axis system where the indicatrix is diagonalized.[5,6] Two situations can be encountered according to the magnitude of the angle θ. In the general case, the change of the birefringence due to the applied electric field is expressed as a complicated combination of the various electro-optic effects involving different powers of the electric field E and the coefficients r_{12}, r_{22}, and r_{62}. In our case, the angle θ is very small $[(1/n_1^2-1/n_2^2)\gg r_{62}E]$, then the variation of the birefringence depends only on the effective coefficient $r_b=r_{22}-(n_1^3/n_2^3)r_{12}$, and is simply linear with the electric field E. So the electric field-induced phase shift between x and y light polarization components is expected to depend on the dc voltage amplitude $V=Exd$ as

$$\Gamma_E=(\pi L)/(\lambda d)(n_2^3r_{22}-n_1^3r_{12})V=(\pi L/\lambda d)n_2^3r_bV$$

or

$$\Gamma_e=(\pi L/\lambda d)FV, \quad (3)$$

where d is the interelectrode spacing, L is the crystal length along the laser beam propagation direction, λ is the wavelength of the laser beam, and F is the figure of merit of materials used for modulation applications.

A good optical quality crystal used in our measurements was obtained by isothermal evaporation from aqueous solution and is of the shape of a parallelepiped with the dimensions $7.49\times3.02\times1.88$ mm^3 with respect to the pseudo-orthorhombic axes. The faces have been polished and electroded with silver paste.

Electro-optic measurements were performed with an original and very sensitive method[7] which is based upon the Sénarmont compensating setup. The optical transmission of such an optical system follows the general law: $\mathbf{I}=\mathbf{I}_0\times\sin^2(\Gamma/2-\beta+\pi/4)$ if the optical absorption is neglected. In this equation I_0 and I are, respectively, the input and output laser intensity and Γ the total phase shift introduced by the sample due to the natural birefringence and/or its variation as a function of the temperature, the electric field or the strain applied to the sample. Here, we are only

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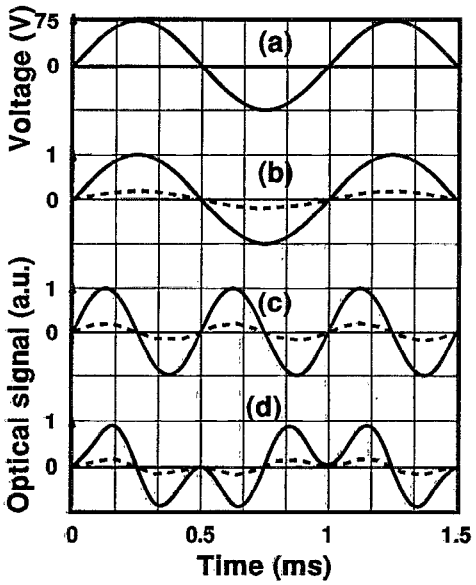

FIG. 1. Performances of the electro-optic modulation obtained with a rubidium hydrogen selenate crystal: (a) applied ac voltage with $f=1$ kHz; (b) modulated optical signal obtained at 1 kHz for $RbHSeO_4$ (solid line) and ADP (dashed line); (c) optical signal obtained at the double frequency, corresponding to a minimum transmission; (d) optical signal obtained when a dc voltage is applied to the crystal. It corresponds to a shift in comparison with double frequency position induced by a dc value of 1 V for $RbHSeO_4$ and 100 V for ADP or a phase shift Γ of 14°.

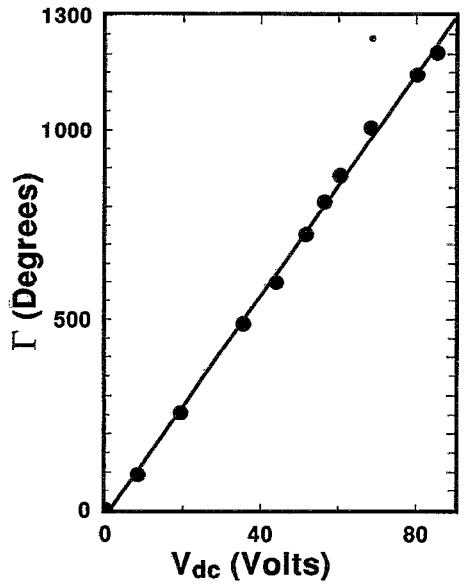

FIG. 2. Electric field-induced phase shift Γ_E obtained at 23.8 °C for an ac applied field of 150 V peak-to-peak with a modulation frequency $f=1$ kHz.

concerned about the dependence on the electric field. In this method an ac voltage with a frequency f is superimposed on a dc voltage and we measure the angle β_{2f} of the analyzer corresponding to the detection of the output signal with the frequency $2f$.

Measurements are carried out with an electric field applied along the y axis and a laser beam of 633-nm wavelength propagating along the z axis. They are performed around room temperature with an ac modulating voltage of 150 V peak-to-peak and frequency varying between 100 Hz to 1 MHz, and a dc voltage amplitude between -150 and 300 V.

Without applying electric field the crystal presents a spontaneous birefringence and thus a natural phase shift which needs to be compensated by an appropriate rotation of the analyzer. A clear modulation of the laser beam is achieved with the $RbHSeO_4$ crystal for all available frequencies of the modulation field, from 100 Hz up to 1 MHz. An example of the modulated signal which is recorded for a modulation frequency $f=1$ kHz is given in Fig. 1, and compared with the corresponding signal obtained with an ADP crystal. The high sensitivity of our method and the large efficiency of the rubidium hydrogen selenate crystal are clearly pointed out in this figure.

The phase shift $\Gamma_E=2\times\beta_{2f}$ obtained at $T=23.8$ °C for a modulation frequency $f=1$ kHz is plotted as a function of the dc voltage in Fig. 2. It exhibits a remarkably linear behavior with the applied voltage up to 90 V but deviates from this law for amplitude larger than 100 V and finally saturates. The result is checked to be completely reproducible for various samples. On the other hand, if we compare the values which are obtained for a zero voltage reached with increasing or decreasing amplitude the shift is very large and is equal to 428°. This large memory effect[8] is also obtained after switching off the electric field applied to the crystal. Both these phenomena are probably attributable to the domain reversal and require special investigation of the change of birefringence in relation with the domain structure.

Here, we focus our attention on the linear dependence of the phase shift which is consistent with Fig. 2, revealing the electro-optic coefficient r_b. We can, therefore, deduce from the experimental data and Eq. (3) the figure of merit $F=n_2^3 r_b$ of the crystal. We obtain $F=13.5\times10^{-9}$ mV^{-1}. The corresponding half-wave voltage inducing a phase shift $\Gamma_E=\pi$ is also derived from Eq. (3) and found to be equal to $V_\pi=7$ V for the particular sample used in our experiments. The reduced half-wave voltage characterizing the rubidium hydrogen selenate is obtained when $L/d=1$: $V_\pi^*=26.5$ V. The values of F and V_π^* obtained for $RbHSeO_4$ are given in Table I and compared with corresponding values reported for the most known electro-optic materials.[9] This comparison shows that rubidium hydrogen selenate is a very promising material for modulation applications owing to its very large F and its very low half-wave voltage. Deviations from the linear dependence of Γ vs V which occur for larger voltage can be indeed easily avoided in applications. In addition, to a rela-

TABLE I. Values of the figure of merit $F=n^3\times r_b$ and the reduced half-wave voltage V_π^* in various materials at room temperature and for $\lambda=633$ nm.

Compounds	$F=n^3\times r$ (pm/V)	$V_\pi^*(V)$
$BaTiO_3(r_{42})$	24 859	332
KDP (r_{63})	40	15 540
$LiNbO_3$ (r_c)	211	2800
KTP (r_c)	184	3417
$RbHSeO_4$	13 540	26.5

tively low dielectric permittivity,[3,4] $RbHSeO_4$ possesses also the advantage of a thermo-optic coefficient which is very weak compared to other materials.[10]

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