Photo-refractive effect of double doped Ce:Co:KNSBN crystal

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ABSTRACT

The materials ratio is compounded followed by the molecular formula $Ba_{0.5}Sr_{1.5}K_{0.5}Na_{0.5}Nb_5O_{15}$. Doping 0.05 wt% CeO_2 and 0.03 wt% Co_2O_3 in KNSBN, Ce:Co:KNSBN crystal was grown by Czochralski technique adopting SiMo heater furnace. The exponential gain coefficient, diffraction efficiency and response time of Ce:Co:KNSBN crystal were measured through two-beam couple light path. The phase conjugation reflect efficiency and response time of Ce:Co:KNSBN and Cu:SBN crystals were measured by four-wave mixing light path. The holographic associative storage were established by using Ce:Co:KNSBN crystal as recording apparatus and Cu:SBN crystal as phase conjugation lens. Addressing was operated by 75% and 50% images and the associative storage reappearing images are clear, complete and noise was small. The photo-refractive effects of Ce:Co:KNSBN crystal are more excellent than KNSBN crystal.

Keywords: Ce:Co:KNSBN crystal, Cu:SBN crystal, photo-refractive effect

1. INTRODUCTION

($Ba_{0.5}Sr_{1.5}K_{0.5}Na_{0.5}Nb_5O_{15}$, KNSBN) crystals belong to tungsten bronze structure and 4mm point group. They have excellent electro-optic, photo-refractive and thermal-electro effects[1]. They were widely used in holographic storage, optic wave-guide and millimeter apparatus. KNSBN crystals have strong lengthways effect and optic properties. There are so many lattice vacancies in KNSBN crystals that a lot of metal ions can be doped in the lattices. The photo-refractive properties of KNSBN can be increased by doping photo-refractive sensitive impurities in KNSBN crystals. The exponential gain coefficient, diffraction efficiency and phase conjugate reflectivity of KNSBN increase when doping Cu ions. The response time of the KNSBN shorten when doping Co ions. The growth of large-size, high optical quality $BaTiO_3$ crystals is very difficult. The photo-refractive effect and optical properties of doped KNSBN crystals can or almost reach the level of $BaTiO_3$ so KNSBN are the best substitute material.

2. GROWTH CE:CO:KNSBN CRYSTAL

The ratio of KNSBN crystals is compounded followed by $Ba_{0.5}Sr_{1.5}K_{0.5}Na_{0.5}Nb_5O_{15}$. The raw materials are Ba_2CO_3, $SrCO_3$, K_2CO_3 Na_2Co_3 and Nb_2O_5 with purity of 99.99%. And the doped materials CeO_2 and Co_2O_3 are spectral purity.

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The ratio of the raw materials is Ba_2CO_3:$SrCO_3$:K_2CO_3:Na_2CO_3:Nb_2O_5=1:3:1/2:1/2:2(mol ratio). The doped concentration of CeO_2 is 0.05 wt% and that of Co_2O_3 is 0.03 wt%.

The raw materials are mixed and sintered and polycrystalline material is produced. The sintering program is as below:

$$\text{Room temperature} \underset{\text{increasing}}{\longrightarrow} 800℃ \underset{\text{holding}}{\overset{\text{4h}}{\longrightarrow}} 1350℃ \underset{\text{increasing}}{\longrightarrow} 1150℃ \underset{\text{holding}}{\overset{\text{4h}}{\longrightarrow}} 1350℃ \underset{\text{decreasing}}{\longrightarrow} \text{Room temperature}$$

The temperature kept at 800^{o}C is to well decompose Ba_2CO_3, $SrCO_3$, K_2CO_3 Na_2Co_3 and Nb_2O_5 and the temperature kept at 1350^{o}C is to make the raw materials react complete and form Ce:Co: KNSBN polycrystalline material. The melting point of KNSBN crystal is 1510^{o}C so we select SiMo heater furnace. Ce:Co:KNSBN crystals are grown by Czochralski technique and the growth direction is c-axis(001). The axial direction temperature gradient in the growth furnace is 40~50^{o}C/cm.and the rotating rate is 20rpm~35rpm. The crystals are annealed in air and the annealing temperature is 1350^{o}C, the annealing time holds 24h. The Curie temperature of Ce:Co: KNSBN is 180^{o}C. The size of the crystal is Φ20×30mm^3. The crystal is polarized along the c-face (001) under the impressed electric field 200V/mm. The crystal is cut into samples of 6×6×3mm^3. The samples are polished to optical standard.

3. MEASUREMENT OF EXPONENTIAL GAIN COEFFICIENT, DIFFRACTION EFFICIENCY AND RESPONSE TIME

3.1 Measurement of exponential gain coefficient

The exponential gain coefficient expresses how much the energy of pump light can transfer to signal light. It reflects the amplification radio of the light in photo-refractive crystal. The exponential gain coefficient is an important index of the photo-refractive properties of the crystal. The photo-refractive effect of the crystal is better when the exponential gain coefficient is larger. Larger exponential gain coefficient can make the signal light be clearer when reappearing. The coupling-wave equation is approximate solute and the results are below:

$$I_2(d) = I_2(0)\frac{I_1(0)+I_2(0)}{I_1(0)+I_2(0)\exp(\Gamma d)}\exp[(\Gamma - a)d] \quad (1)$$

$$I_1(d) = I_1(0)\frac{I_1(0)+I_2(0)}{I_1(0)+I_2(0)\exp(\Gamma d)}\exp(-ad) \quad (2)$$

among which, d is the thickness of the crystal and a is the absorption coefficient.

From (1), we can see that when Γ> a , the signal has an exponential gain. Γ expresses the per length gain induced by the grating component of the π/2 phase shift. It mainly determined by the degree of modulation of two light and the amplitude of the photo-induced refractivity change. From (1) and (2), it can be seen two beam can produce energy exchange when coupling. One beam gets energy and amplifies and the other loses energy and attenuates. When the optical absorption and the reflection of the front and back faces are ignored, it can be got from (1) and (2):

$$\Gamma = \frac{1}{d}\ln\left(\frac{I_2^{'}I_1}{I_2 I_1^{'}}\right) \quad (3)$$

Among which, $I_2^{'}(I_2)$ is the transmission signal beam intensity when coupling exists(not exists). $I_1^{'}(I_1)$ is the reference beam intensity. When $I_1 >> I_2$, the pump damage can be ignored, so $I_1^{'} \approx I_1$, and the gain co efficiency is:

$$\Gamma = \frac{1}{d} \ln[\frac{I_2(d)\ \text{(reference beam exists)}}{I_2(d)\ \text{(reference beam not exists)}}] \tag{4}$$

Ar^+ laser acts as the light source. The light length λ =514.5nm and the polarization direction in the incident plane (e light). The beam splitter splits the laser to two beams and they meet at Ce:Co:KNSBN crystal in the angle of 2θ, the thickness of the crystal is 3mm. The beams transit through y-face. I_{10} =1.82w/cm^2, $\beta = I_{10}/I_{20}$=1100. The set-up of two-beam coupling is shown in figure 1.

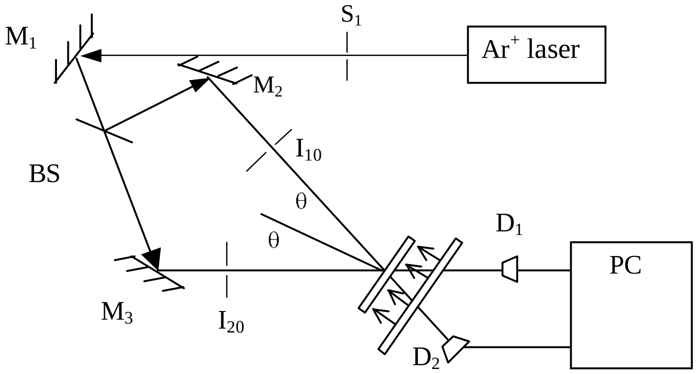


Figure 1. Light path scheme of two-beam coupling

M_1, M_2, M_3,-- reflective mirrors; BS— beam splitter; D_1,D_2—detectors

When 2θ=25°, the maximal exponential gain coefficient Γ_{max} were measured. The results are given in table 1.

3.2 Measurement of diffraction efficiency and response time

As holographic recording materials, photo-refractive crystals are required to have high diffraction efficiency. And the images recorded in the crystal can reappear clearly. The diffraction efficient is defined as the ratio of the diffraction light intensity and the transmission beam intensity before the establishment of the grating. It is $I_2^{'}/I_2\times100\%$, in which, I_2 is the transmission beam intensity of I_{20} before the establishment of the grating and $I_2^{'}$ is diffraction light intensity of I_1 in the direction of I_2 after the establishment of the grating. Adopting two-beam coupling set-up, the diffraction efficient of Ce:Co:KNSBN was measured. I_{20} was cut off and the diffraction light $I_2^{'}$ that I_1 in the direction of I_2 was measured. When the incident beams intensity $I_{10}= I_{20}$=1.1w/cm^2, 2θ=25°, the maximal diffraction efficiency(η_{max})of Ce:Co:KNSBN was measured. The results were shown in table 1.

The response time τ_0 is defined as the time from the recording beginning to the diffraction efficiency gets to (1-e^{-1}) of the maximal. The results are shown in table 1, too.

Table 1 The measurement results of the holographic properties of Ce:Co:KNSBN crystal

Crystals	Γ_{max}(cm^{-1})	η_{max}(%)	τ_0(ms)
Ce:Co:KNSBN	15	68	700
KNSBN	13	62	1200

4. MEASUREMENT OF PHASE CONJUGATION PROPERTIES OF CE:CO:KNSBN AND CU:SBN CRYSTAL

SBN and KNSBN series crystals belonging to tungsten bronze structure have important application in photo-refractive nonlinear optics. The phase conjugation waves are derived from the degenerate four-wave mixing in photo-refractive crystals. The set-up is shown in figure 2.

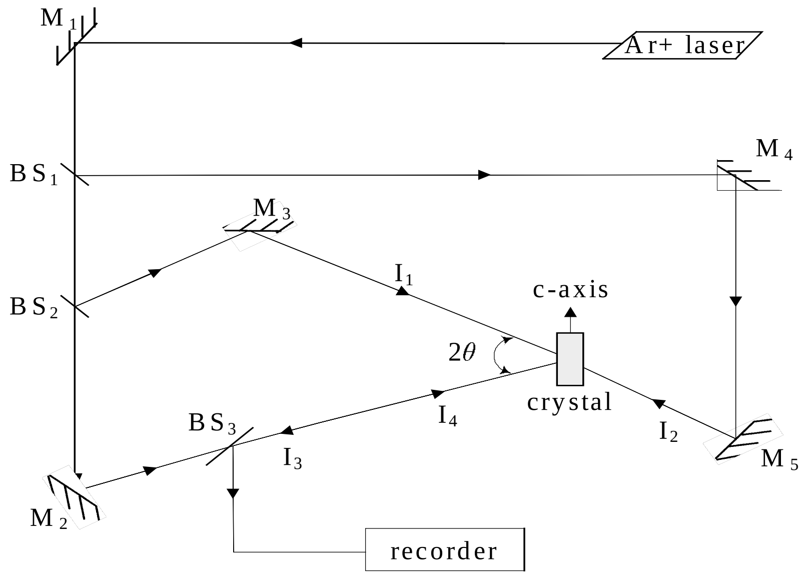


Figure 2. Four-wave mixing experimental set-up for photo-refractive crystal

M_1, M_2, M_3, M_4, M_5: reflect mirrors; BS_1, BS_2, BS_3:beam splitter

In the experiments, the light source is Ar^+ laser. The wave length λ =514.5nm and the polarization direction in the incident plane (e light). The beam splitters BS_1 and BS_2 split the light from the laser to two reverse pump beams I_1, I_2, and one detect beam I_4. A shutter S controls pump beam I_2 so that the transient phase conjugation beam can be gotten. The intensity attenuator L.A. is to change the beam intensity of incident beam I_4. The measuring part of phase conjugation beam I_3 is separated by a splitter and received by a silicon photocell, then it changes to electrical signal and records by X-Y function recording apparatus. In the process of degenerate four-wave mixing, the shutter S makes the incident crystal time of pump beam I_2 delay several seconds and a transient peak phase conjugation reflection efficient can be observed. The reason is the erasing effect of the anti-pump beam I_2 to the transmission grating recorded by I_1 and I_4. The intensities of pump beams I_1 and I_2 are 605 mW/cm^2 and 375 mW/cm^2, respectively. The measurement results of phase conjugation reflect efficient and response time in Ce:Co:KNSBN and Cu:SBN are shown in table 2.

Table 2 Results of phase conjugation properties

crystals	Ce:Co:KNSBN	Cu:SBN
transient R (%)	115	125
steady R(%)	82	81
τ(s)	1.2	3.2

5. ASSOCIATIVE STORAGE ADOPTING FOUR-WAVE MIXING PHASE CONJUGATION LENS

The experimental set-up is shown in figure 3.

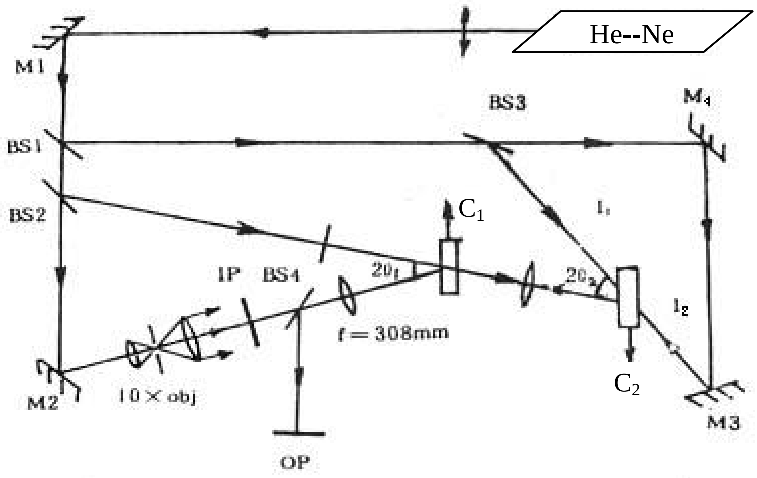
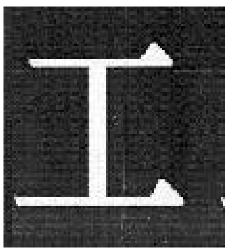


Figure 2. Optical Set-up for holographic associative storoge experiment

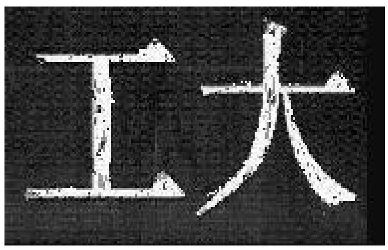
IP:input flat; OP:output flat; M_1~M_4:mirrors; BS_1~BS_4:beam splitters;C_1 :record crystal ;C_2:phase conjugate crystal

The light source is He-Ne laser and the output wavelength is 632.8nm, the output power is 30mW. The storage material is Ce:Co:KNSBN crystal and the Cu:SBN crystal acts as phase conjugation lens. The beam incident from He-Ne laser splits to reference beam I_3 and object beam I_4 in holographic storage. The angle between them is 16.5^o. Object beam I_4 loads the image information (a_1 a_2 in figure 4) and records the image information in Ce:Co:KNSBN. The recording time is 12 seconds. The other two reverse coherent beams I_1 and I_2 split from He-Ne laser form the pump beams of four-wave mixing phase conjugation lens (Cu:SBN). I_1 = 432.0 mW/cm^2 and I_2 = 216.0 mW/cm^2. The diffraction beam that records object beam I_4 act as the signal light of four-wave mixing and the angle between it and I_1 is 21^o. The phase conjugation beam returns along the original road and oppositely reads out the holographs in the medium. At this time, addressing (b_1 b_2 in figure 4) is from 75% and 50% information of original object in the input face and the associative output complete images (c_1 c_2 in figure 4) can appear in the output face.

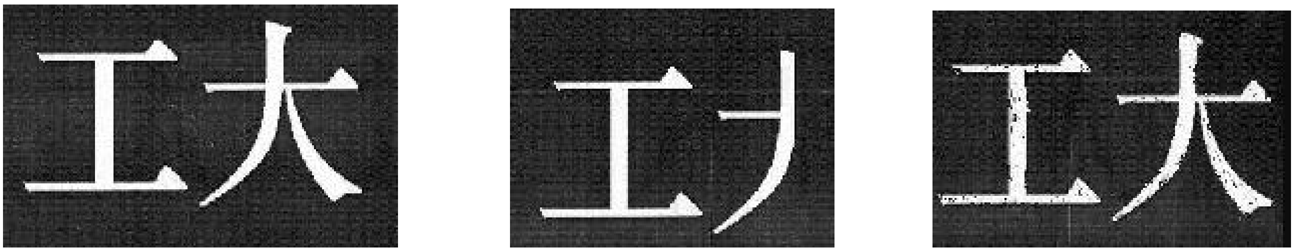
(a_1)



(b_1)



(c_1)



(a$_2$) (b$_2$) (c$_2$)

Figure 4. The results of associative storage experiments

(a$_1$,a$_2$)input images; (b$_1$,b$_2$)input addressing images; (c$_1$,c$_2$)read-out images

Adopting four-wave mixing method to form phase conjugation reflect, the apparatuses are more and the light path is more complex, but the introduction of I_1 and I_2 can make the beam transiting recording crystal and incident to phase conjugation lens not only get feed-back effect, but also receive gain effect.

6. CONCLUSION

Ce:Co:KNSBN crystal was grown by Czochralski technique adopting SiMo heater furnace. The exponential gain coefficient, diffraction efficiency and response time of Ce:Co:KNSBN crystal were measured through two-beam couple light path. Ce:Co:KNSBN crystal possesses high diffraction efficiency and short response time (ms level). The phase conjugation reflect efficiency and response time of Ce:Co:KNSBN and Cu:SBN crystals were measured by four-wave mixing light path. The holographic associative storage were established by using Ce:Co:KNSBN crystal as recording apparatus and Cu:SBN crystal as phase conjugation lens. The holographic storage apparatus and phase conjugation lens are all photo-refractive crystals, so it has the merits of real time processing, repetitive using, good quality image and high signal-to-noise ratio.

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REFERENCES

[1] Li Minghua, Xu Xuewu, Qiu Dongxiao, J. Crystal Growth, **21**, 225-229, 2000.

[2] Xu Jingjun, Liu Simin, Wu Yuanqing, Chin Phys. Lett., **9(7)**, 352-354, 1992.

[3] Chen Huanchu, J. Cryst. Growth, **96**, 357-368, 1989.

[4] Gimter P. Phys. Rev. **93(4)**, 246-253, 1982.

[5] D. Psaktis, N. H. Farhat, Opt. Lett. **10**, 98, 1985.

[6] B. H. Soffer, G. J. Dunning, Y. Owechko, Opt. Lett. **11**, 118, 1986.