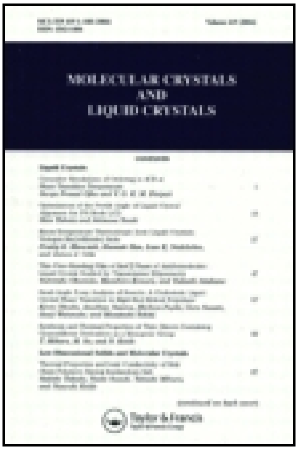

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Response Times and Their Temperature Dependence Measured for a Series of Liquid Crystals Using Gain Spectra of Stimulated Light Scattering

S. Nersisyan [a] , S. Serak [a] & N. Tabiryan [a]

[a] Beam Engineering for Advanced Measurements Co., Winter Park, Florida

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S. Nersisyan
S. Serak
N. Tabiryan
Beam Engineering for Advanced Measurements Co.,
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In the present work we have shown the opportunity of using the process of stimulated light scattering due to recording of dynamic orientation gratings for directly measuring the characteristic orientation relaxation time of liquid crystals (LCs) and related material parameters, the elastic constants and the coefficients of orientational viscosity. The measurement consists in obtaining the gain spectra of LCs and determining the magnitude of the gain coefficient and the optimum frequency shift between interfering laser beams. We have designed a setup where the gain spectra of stimulated light scattering process in LCs are obtained within seconds. These spectra are highly specific to LCs serving as a precise and fast tool for studying LC materials and their mixtures with the purpose of optimizing their linear and nonlinear optical properties. Using this technique we have found that certain azo LC materials are faster compared to several well-known Merck's formulations for LC displays.

Keywords: liquid crystals; material characterization; response time; stimulated light scattering

INTRODUCTION

Speeding up the process of reorientation of liquid crystals (LCs) under the influence of electric fields or optical radiation is one of the most

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Address correspondence to S. Nersisyan, Beam Engineering for Advanced Measurements Co., 809 S. Orlando Ave., Suite I, Winter Park, FL 32789. E-mail: nelson@beamco.com

important tasks for material development for LC displays, spatial light modulators, etc. The most common technique for evaluation of the response time of LCs in electro-optical devices consists in measuring the time it takes to reach certain, typically 90%, modulation depth of a light beam due to LC reorientation induced by an electric (or optical) field. The information obtained in such measurements is highly technical in nature. While it can be related with material properties, anchoring conditions and backflow effects affect the results. The very determination of threshold field strength is a time-consuming process.

Due to LC display industry, hundreds of LC materials have been synthesized, optimized with respect to their electro-optical and nonlinear optical properties, and tested with respect to their stability against chemical and physical influences. Numerous blends of optical and mechanical properties of LC materials are available for reaching specific application goals. Testing a large number of LC materials is required for identifying the most promising materials for any given application, linear as well as nonlinear optical.

In the present work we have shown the opportunity of using the process of stimulated light scattering due to recording of dynamic orientation gratings [1] for directly measuring the characteristic response time of LCs and related material parameters, the elastic constants and the coefficients of orientational viscosity. The measurement includes determination of the optimum frequency shift between the interfering laser beams and the gain coefficient. The inverse magnitude of the optimum frequency shift relates to the orientation relaxation time of LCs while the maximal gain coefficient allows directly measuring the elastic constants of the LC. We have designed a setup where the gain spectrum $G(\Omega)$ of energy exchange process in LCs can be obtained within seconds. We show that these spectra are highly specific to LCs serving as a precise and fast tool for studying LC materials and their mixtures with the purpose of optimization of their linear and nonlinear optical properties.

GAIN COEFFICIENT FOR STIMULATED LIGHT SCATTERING

The test setup is shown in Figure 1. The beam of a solid-state laser ($\lambda = 1064\,\mathrm{nm}$) is split into two beams by a polarizing beam splitter $2b$. A $\lambda/2$ wave plate 1 and the polarizing beam splitter $2a$ are introduced into the main beam at the output of the laser to control its power and the power ratio between the two beams obtained as a result of splitting.

These beams are reflected from the mirrors 4 and 6 and are merged together with the aid of the non-polarizing beam splitter 5. The mirror 4

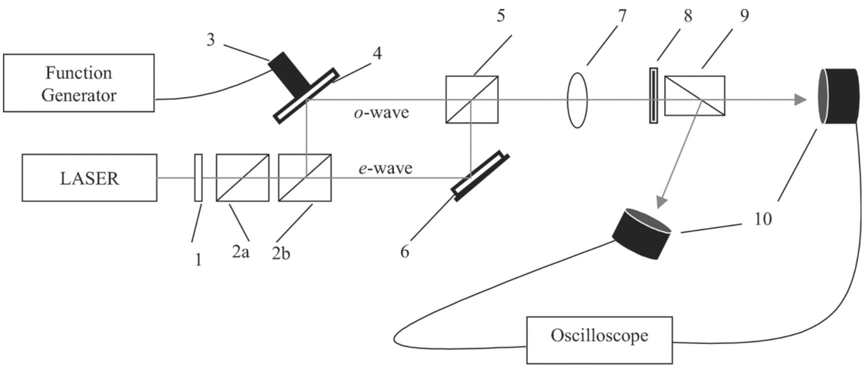

FIGURE 1 Schematic of the experimental setup for realization and study of the process of generation of orientation gratings in LC: 1 – $\lambda/2$ wave plate, 2 – polarizing beam splitters, 3 – piezo-transducer; 4 – IR reflecting mirror; 5 – non polarizing beam splitter; 6 – IR reflecting mirror; 7 – lens; 8 – NLC cell being tested; 9 – Glan prism; 10 – power meters.

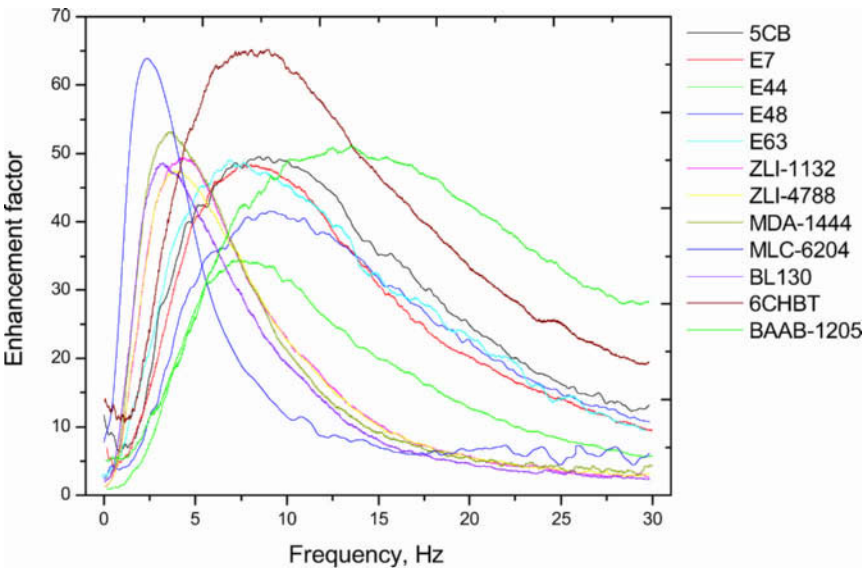

FIGURE 2 Gain spectra (enhancement factor as a function of frequency shift) for several LC materials. The thickness of all cells is 300 μm. The data are taken at fixed power density of the pump beam $6.8\,\mathrm{kW/cm^2}$.

TABLE 1 Material Parameters of Several LCs: T_{cl} – Clearing Temperature; Δn – Optical Anisotropy; $\Delta\varepsilon$ – Dielectic Anisotropy; η – Hydrodynamic Viscosity Coefficient; Ω_{opt} – Optimum Frequency Shift; and $G_{max}I_PL$ – the Gain Factor

LC	T_{cl} [°C]	Δn	$\Delta\varepsilon$	η [mm^{2}s^{-1}]	Ω [Hz]	$G_{max}I_PL$
5CB	35	0.212	+12.8	67	8.2	3.899
E7	58	0.2253	+13.8	39	7.9	3.892
E44	100	0.2627	–	–	7.5	3.533
E48	84	0.2306	+15.1	44	9.0	3.724
E63	82	0.2272	+14.6	39	7.2	3.886
ZLI-1132	72.4	0.137	–	27	4.4	3.896
ZLI-4788-000	83	0.1647	– 5.7	35	4.0	3.859
MDA-1444	98.5	0.1774	+31.1	–	3.6	3.977
MLC-6204-000	63	0.1478	+35.3	65	2.4	4.166
BL130	102	–	–	–	3.2	3.886
6CHBT	43.2	0.152	+12	–	8.0	4.178
BAAB1205	59	0.21	+0.8	–	12.7	3.942

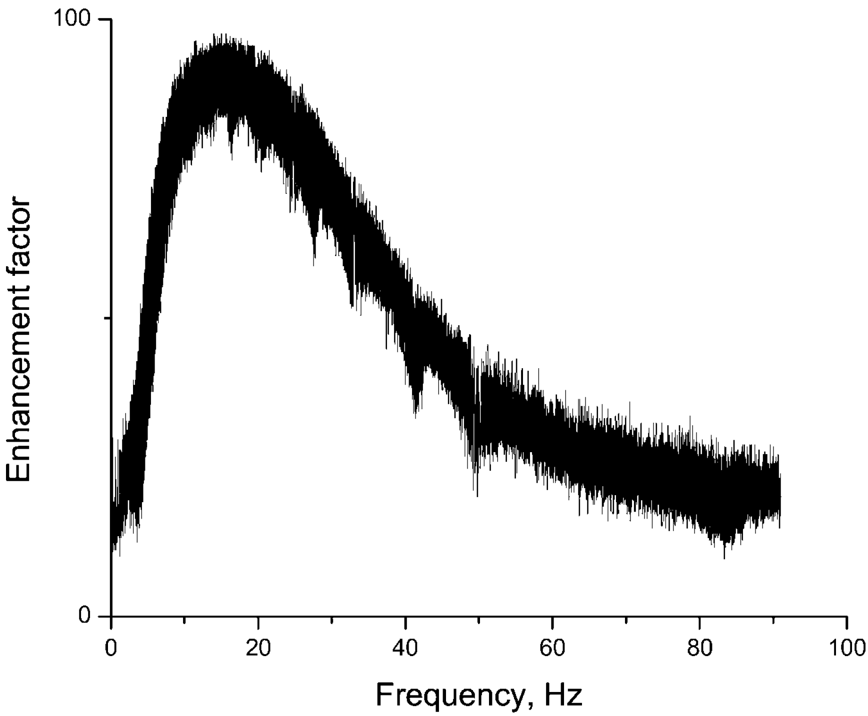

FIGURE 3 Gain spectrum in the large range of frequency shifts for BAAB 1205.

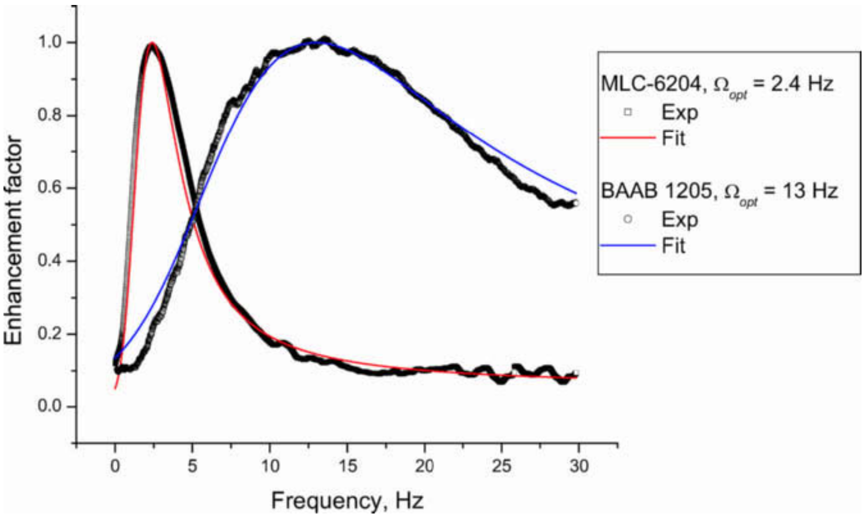

FIGURE 4 Normalized gain spectra and their theoretical fit for materials with the smallest and the largest optimum frequency shifts.

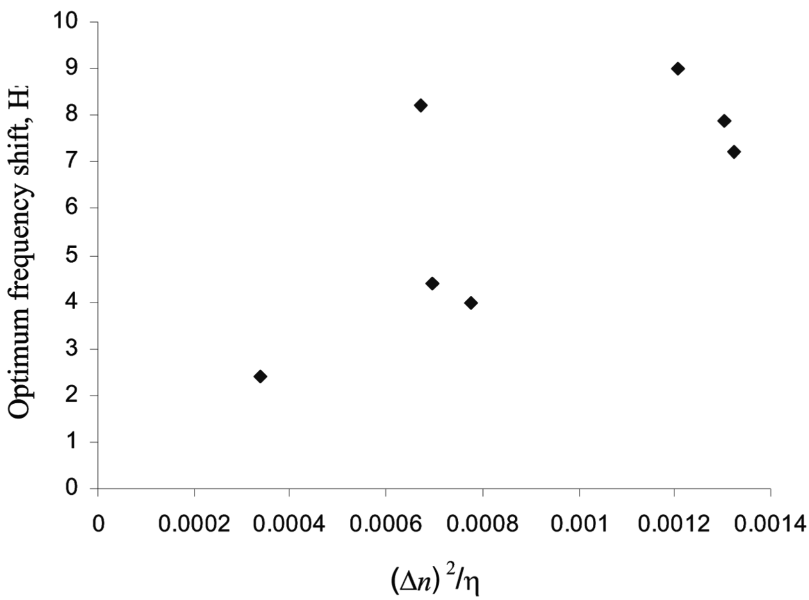

FIGURE 5 The dependence of the optimum frequency shift on the ratio of the optical anisotropy to the coefficient of hydrodynamic viscosity.

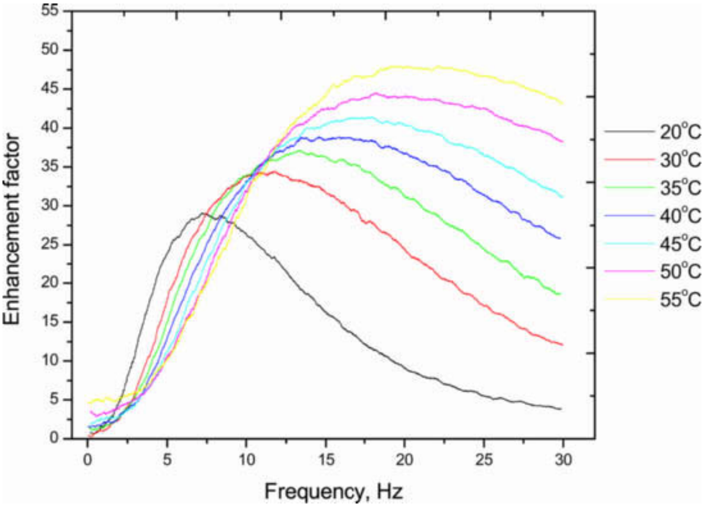

FIGURE 6 Variation of the gain spectrum with temperature for NLC E48 far from the critical region of nematic-isotropic phase transition.

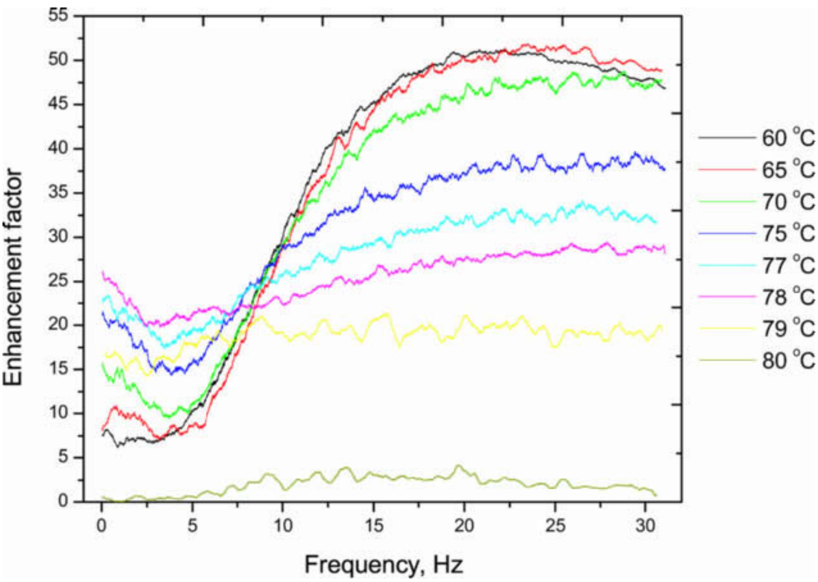

FIGURE 7 Variation of the gain spectrum with temperature for NLC E48 near the critical region of nematic-isotropic phase transition.

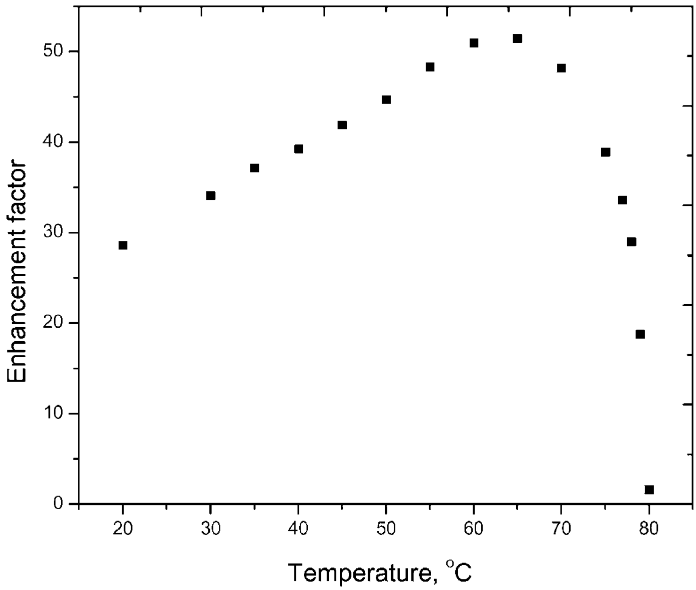

FIGURE 8 Temperature dependence of the enhancement factor for LC E48.

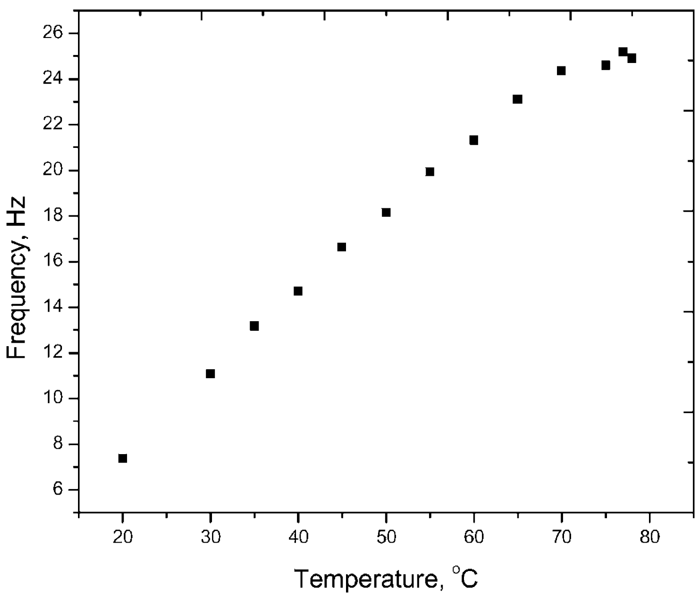

FIGURE 9 Temperature dependence of the optimum frequency shift for LC E48.

is attached to a piezo-transducer 3 which, controlled by a function generator, allows introducing frequency shift in the beam reflected from the mirror 4. The system for introducing a constant frequency shift Ω is one of the most important components of the setup. Both the form of the voltage provided by the function generator and its amplitude shall be chosen properly in order to drive the piezo-transducer in such a way that the coherence of the beam is not reduced.

The beams propagate along the same path after the non-polarizing beam splitter 5 and are focused into the test NLC cell 8 by a lens 7 of 75 mm focal length. Thus, the polarization of the frequency-shifted beam is perpendicular to the optical axis of the NLC (*o*-wave) and the polarization of the second beam (with no frequency shift) is along the orientation of the optical axis of the NLC (*e*-wave). A Glan prism 9 separates the beams of different polarization components the power of which was measured by power meters 10.

The dependence of the signal beam enhancement factor on the frequency shift between the signal and the pump beams at fixed power

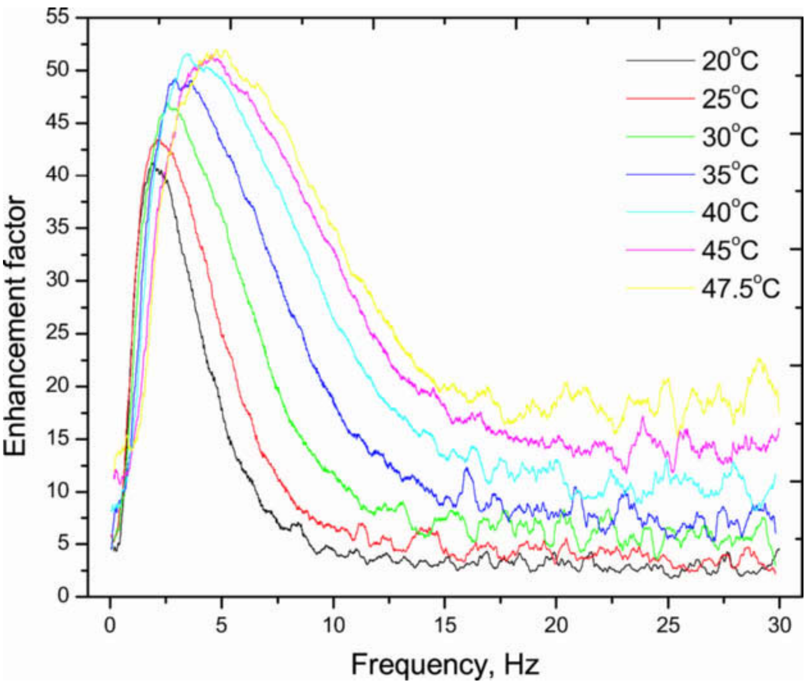

FIGURE 10 Variation of the gain spectrum with temperature for LC MLC 6204-000 far from the critical region of nematic-isotropic phase transition.

density of the pump beam provides the gain spectrum. The enhancement factor is defined as the ratio of the signal beam power in the regime of energy transfer (in the presence of frequency shift between the beams Ω) to the signal beam power at the input of the LC cell [1]:

$$\frac{P_S(\Omega)}{P_S} = \exp\left(\frac{2\Omega\Omega_{opt}}{\Omega^2 + \Omega_{opt}^2} G_{\max} I_P L\right) \tag{1}$$

where $G_{\max}$ is the maximal gain coefficient,

$$G_{\max} = \frac{(n_{\parallel} + n_{\perp})^2 \lambda}{8\pi c n_e K_2} \approx \frac{n_{\parallel}\lambda}{2\pi c K_2}, \tag{2}$$

and Ω_{opt} is the frequency shift for the optimum energy transfer from the pump to the signal beam,

$$\Omega_{opt} = \left(\frac{2\pi(n_{\parallel} - n_{\perp})}{\lambda}\right)^2 \frac{K_2}{\gamma} \tag{3}$$

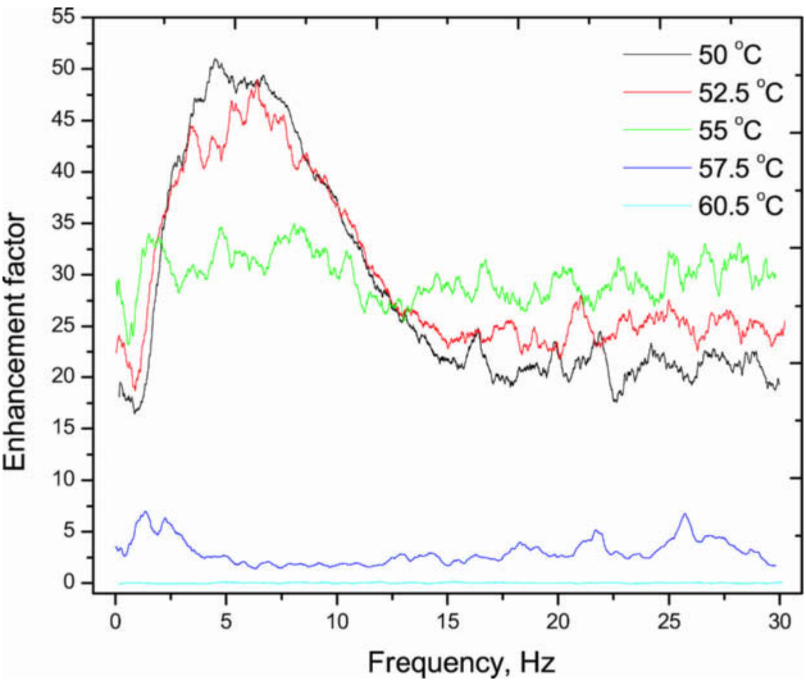

FIGURE 11 Variation of the gain spectrum with temperature for LC MLC 6204-000 near the critical region of nematic-isotropic phase transition.

In Eqs. (1)–(3), L is thickness of the LC layer and I_P is the power density of the pump laser beam, γ is the constant of orientational viscosity, and K_2 is the elastic constant. In these series of experiments, the energy transfer rates are kept at relatively low levels in order to be able to use analytical formulae for treatment of the results.

Since the dependence of the enhancement factor on the frequency-shift allows determination of both Ω_{opt} and G_{max}, this technique becomes a powerful tool for fast and high precision determination of the material parameters of LCs important both for nonlinear optical as well as electro-optical applications.

Figure 2 shows the enhancement factor for a number of LC materials of the same thickness ($L = 300\,\mu m$) obtained at the fixed power density of the pump beam $I_P = 6.8\,kW/cm^2$. The results obtained for the optimum frequency shift and the value of the maximal gain constant are summarized in the Table 1 along with other available material parameters relevant to electro-optical and nonlinear optical applications of LCs.

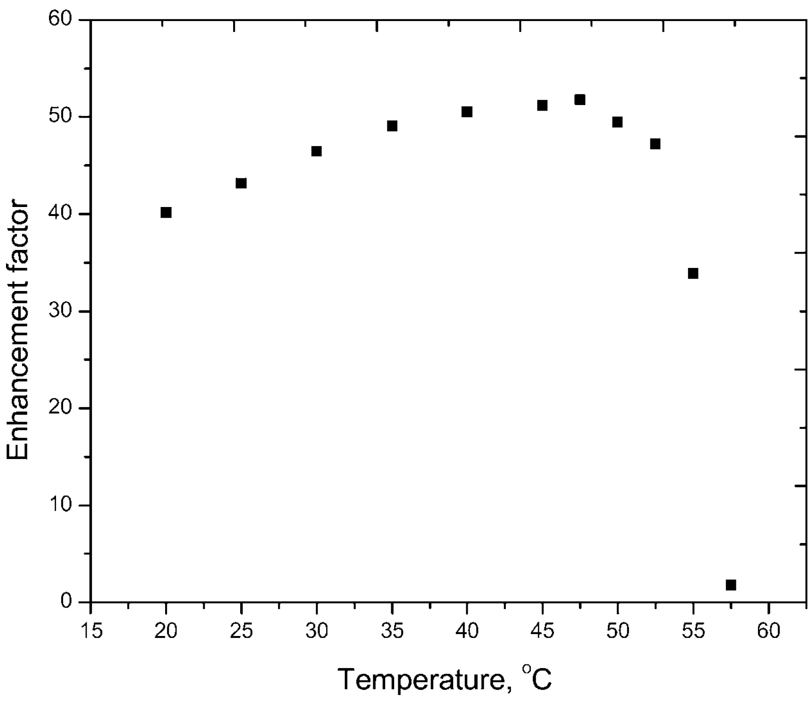

FIGURE 12 Temperature dependence of the enhancement factor for LC MLC 6204-000.

The LCs 6CHBT and MLC-6204-000 (Merck) stand out for their high gain coefficients. The latter has larger temperature range of the nematic phase. BEAM Co.'s LC BAAB 1205 proved to be among the materials with the largest gain while possessing with the highest frequency shift $\Omega_{opt} = 13\,\text{Hz}$. Note that the width of the frequency shift spectrum of the enhancement factor increases with increasing optimum frequency shift, and that $\Delta\Omega_{opt} = 3.5\ \Omega_{opt}$ if defined by the e^{-1} level of the peak enhancement. This is seen clearly in the dependence of the enhancement factor of the LC BAAB 1205 on frequency shift plotted for up to 90 Hz in Figure 3.

One of the interesting observations is that the optimum frequency for the material MLC-6204-000 (Merck) is only 2.4 Hz. Actually, substantial enhancement for this material is observed at a frequency shift as small as $\Omega_{opt} = 0.5\,\text{Hz}$! The theoretical fit of the normalized gain spectra corresponding to the smallest and the largest optimum frequency shifts are shown in Figure 4.

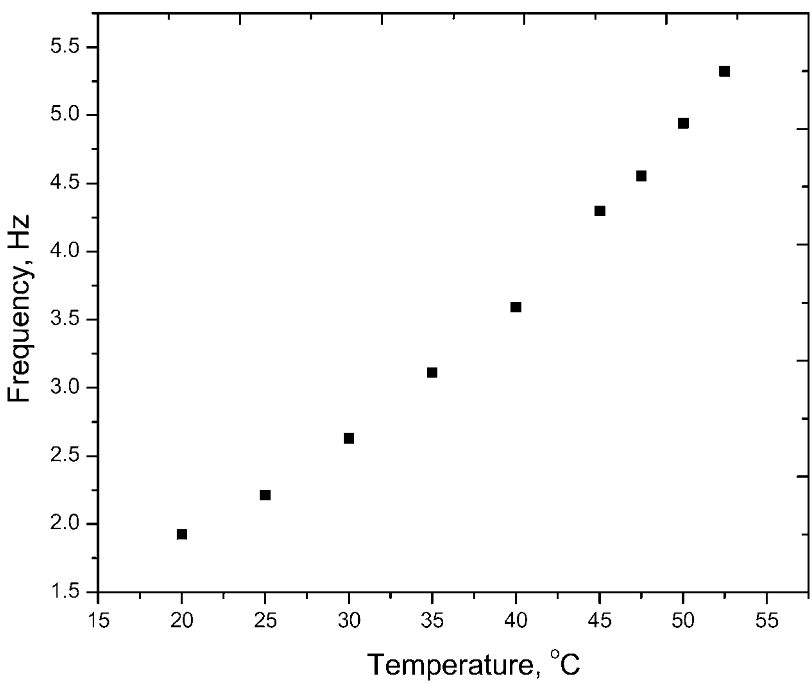

FIGURE 13 Temperature dependence of the optimum frequency shift for LC MLC 6204-000.

For many of tested materials, only the constants of hydrodynamic viscosity η and the optical anisotropy $\Delta n = n_{\parallel} - n_{\perp}$ can be found in literature. Figure 5 shows that the tendency of the optimum frequency to increase with increasing ratio $(\Delta n)^2/\eta$ holds well even though the parameters that determine the magnitude of the optimum frequency shift is the orientational viscosity γ and the elastic constant K_2.

THE EFFECT OF TEMPERATURE

Temperature dependence of material parameters of LCs is clearly reflected on the gain spectra of the materials. Below these dependences are shown for several LCs. In all cases, the LCs were planarly oriented in a $L = 300\,\mu$m-thick layer and the power density was fixed at $5.4\,\mathrm{kW/cm^2}$.

NLC E48 (Merck)

The temperature of phase transition from nematic to the isotropic phase is equal to 84°C for LC E48. Namely near this critical range of

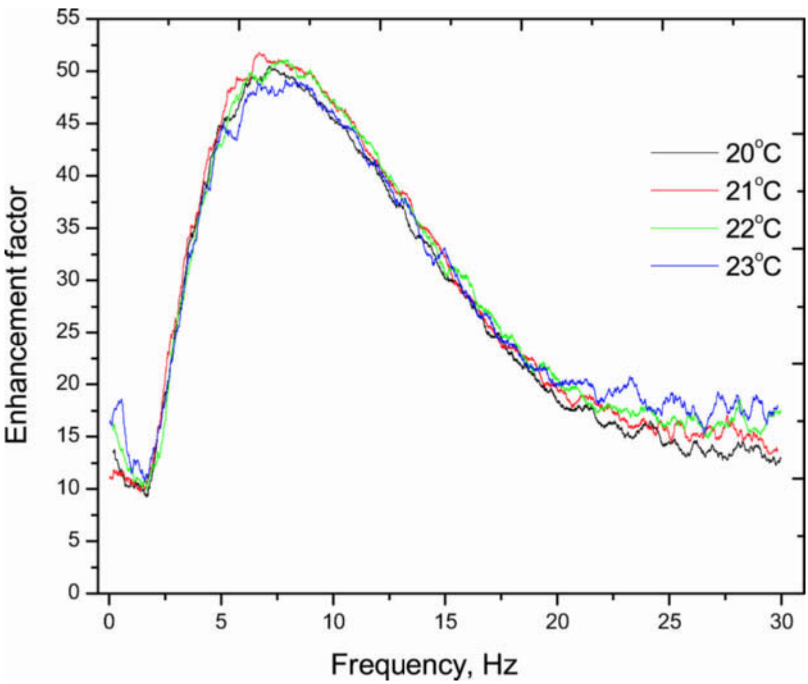

FIGURE 14 Variation of the gain spectrum with temperature for LC 5CB far from the critical region of nematic-isotropic phase transition.

phase transition the temperature dependence of LC material parameters are well pronounced. Figures 6 and 7 correspond to two well-defined regions of temperature dependence of the signal beam power. Far from the critical temperature, the optimum frequency and the amplitude of the signal beam power increase with increasing temperature, Figure 6. The temperature dependence of material parameters of LCs is determined by the temperature dependence of their order parameter $S(T)$. Typically, the elastic constant K decreases with increasing temperature T as $K \sim S^2$ whereas $\Delta n \sim S$. Thus the gain coefficient $G_{\max}$ is expected to grow with increasing temperature as $G_{\max} \sim K^{-1} \sim S^{-2}(T)$.

Decreasing elastic constant and the optical anisotropy of LCs tend to decrease the optimum frequency shift with increasing temperature. However, increasing temperature decreases viscosity which, as we will see below, has the major effect resulting in higher optimum frequency and shorter response times at higher temperatures.

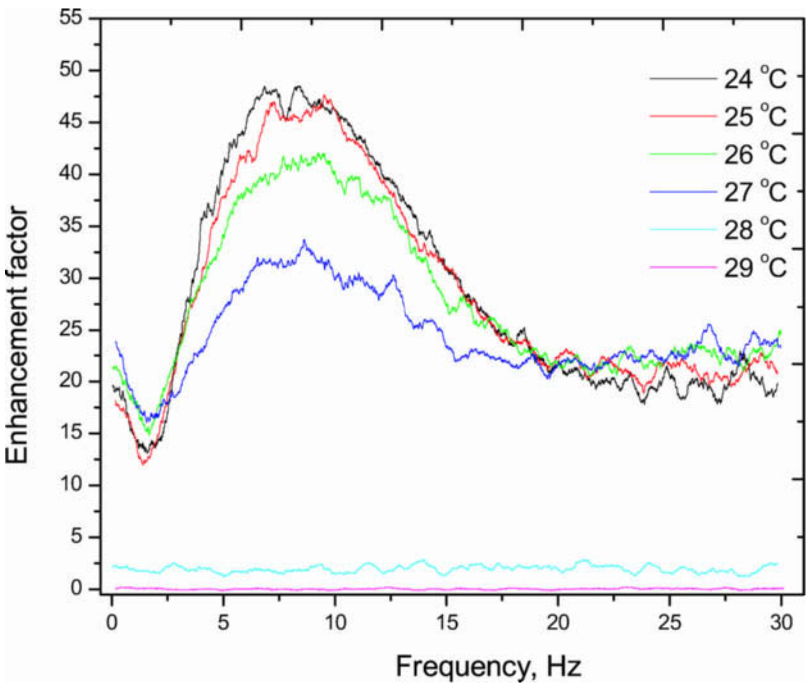

FIGURE 15 Variation of the gain spectrum with temperature for LC 5CB near the critical region of nematic-isotropic phase transition.

Near the critical region, enhanced fluctuations decrease the efficiency of energy transfer up to its complete disappearance in the isotropic phase as it can be seen in Figures 7 and 8. The optimum frequency shift continues to increase up to the phase transition point, Figure 9.

Figures 7 and 8 suggest that the actual phase transition temperature for the LC E48 is 80°C which is by 4°C smaller than reported in its datasheet. This difference can be attributed to an increase in the LC temperature due to absorption of the laser beam by impurities; no special filtering procedures or clean room environment were used when making the LC cells under tests.

NLC MLC 6204-000

Figures 10–13 describe the effect of temperature on the gain spectra for NLC 6204. The phase transition temperature for MLC 6204 is 63°C, however, the signal beam amplification was lost at 60.5°C in our experiments. This difference, as we noted above, may be attributed to the laser heating of the LC.

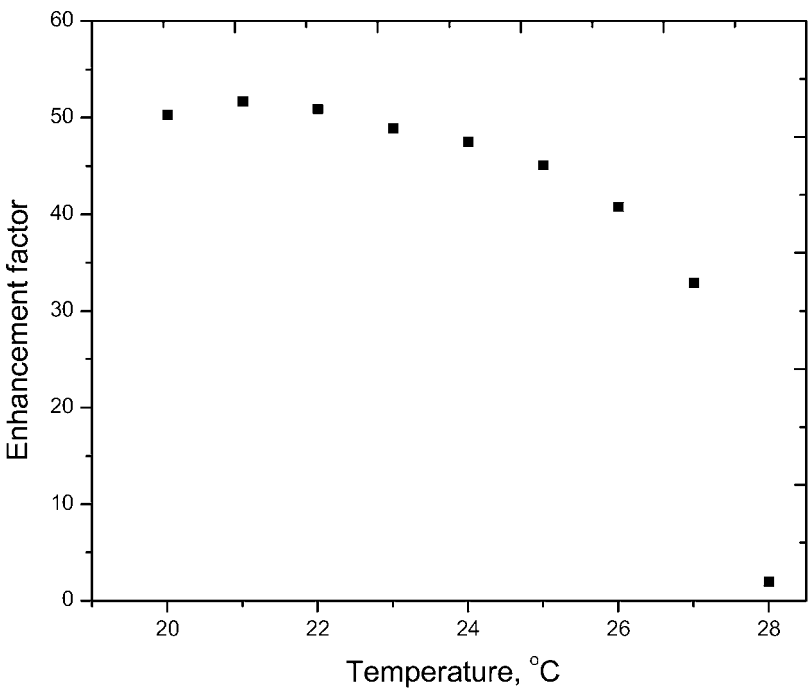

FIGURE 16 Temperature dependence of the enhancement factor for LC 5CB.

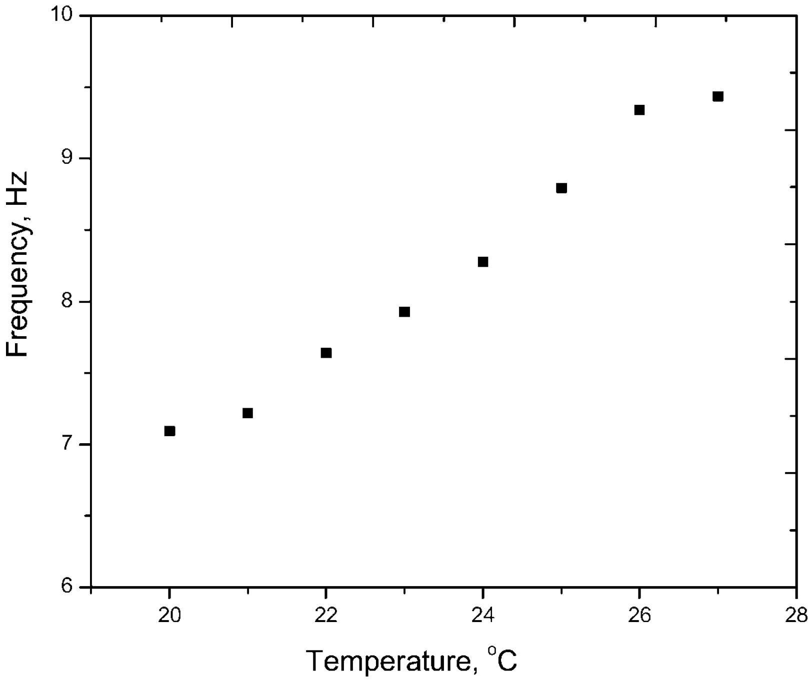

FIGURE 17 Temperature dependence of the optimum frequency shift for LC 5CB.

NLC 5CB (Merck)

Figures 14–17 describe the effect of temperature on gain spectra for NLC 5CB. The phase transition temperature for LC 5CB is 33°C. The signal beam amplification was lost at 29°C in our experiments.

CONCLUSIONS

Thus, gain spectra of stimulated light scattering due to recording of dynamic orientation gratings in LCs prove to provide important information about the electro-optical and nonlinear optical properties of LCs. The experimental geometry used in this paper provides information only about one of the three elastic constants that characterize nematic LCs. It can be modified to obtain information about other constants as well.

It is important that the gain spectrum can be obtained within seconds since the frequency sweep of the generator that controls with the motion of the piezo-transducer is easily integrated into an automized data acquisition system. Studies directed towards developing new materials are related with empirical formulations and tests of numerous mixtures, and the proposed technique can considerably enhance their efficiency. Due to the capability of the technique for characterizing large number of LC materials with small time and resources, we found certain azo LC materials that are over 30% faster compared to well-known Merck's formulations for LC displays.

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