

Photostimulated change of phase-transition temperature and "giant" optical nonlinearity of liquid crystals

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It is shown that the optical "giant" conformation nonlinearity of nematic liquid crystals is due to the change of the macroscopic order parameter of the crystal and to the high-frequency polarizability of the molecules that enter it during the conformation transition. The change of the order parameter leads to the experimentally observed shift of the phase-transition temperature of a nematic MBBA liquid crystal when acted upon by laser radiation of wavelength near the crystal absorption band. The magnitude and sign of the shift determine the experimentally obtained values and signs of the nonlinearity parameter, while the temperature dependence of the refractive index determines the observed growth of these parameters as the clearing point is approached from the mesophase side.

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"Giant" nonlinearity of liquid crystals is being intensively investigated of late.^{1–5} Among the mechanisms that cause the anomalously large values of ε_2 ($\varepsilon_2 = 2\Delta n \bar{n} / |\mathbf{E}|^2$, where Δn is the optically induced change of the refractive index of the nematic liquid crystal (NLC), $\bar{n}^2 = (1/3)(2n_o^2 + n_e^2)$ is the average refractive index, n_o and n_e are the refractive indices of the ordinary and extraordinary waves, and \mathbf{E} is the optical-wave field intensity) following action near the absorption edge of such media are optically induced conformation transformations of the molecules^{2,6} of NLC.

It was shown earlier² that the change of the refractive indices of these media is due to the change of the polarizability of the liquid-crystal molecules upon phototransformation. We establish in the present paper that the appearance of a conformation transition in the optical nonlinearity is due to specific properties of the liquid-crystal state. An investigation of the temperature characteristics of the conformation nonlinearity of the NLC methoxybenzylidenebutylaniline (MBBA) has shown that besides the change of the high-frequency polarizabilities γ of the molecules, phototransformations of the molecules lead to a change of the macroscopic order parameter S . Each of these mechanisms contributes to the optical nonlinearity, and the contribution of the change of the order parameter to the nonlinearity increases as the "nematic-isotropic liquid" phase-transition point T_c is approached. A change in the value of S shifts the temperature T_c ; this is interpreted as a manifestation of the dependence of the order parameter on the conformation state of the molecules.⁷

The model proposed explained the previously obtained^{2,8} relations between the values of the components of the linear cubic susceptibility tensor $\chi^{(3)}$, as well as the signs of these components.

EXPERIMENTAL RESULTS

We investigated the characteristics of the conformation nonlinearity of the NLC MBBA as a function of temperature. Nominally pure samples ($T_c \approx 40^\circ\text{C}$) were placed in glass cuvettes 40–50 μm thick with covers that ensured

planar orientation of the liquid-crystal director. The cuvette was placed in a thermostat whose temperature was monitored accurate to 0.05 $^\circ\text{C}$.

The nonlinearity characteristics were investigated by a standard dynamic holography technique.² Sinusoidal thin phase gratings with period $\Lambda = 10\text{--}20 \mu\text{m}$ were recorded with polarized radiation of a helium-cadmium laser ($\lambda = 0.44 \mu\text{m}$) having a power $P \lesssim 50 \text{ mW}$. The lattice vector $|\mathbf{q}| = 2\pi/\Lambda$ was perpendicular to the NLC director. The cuvette was perpendicular to the bisector of the light-beam convergence angle, and the radiation polarization was either extraordinary or ordinary. This experimental geometry prevented the onset of orientational "giant" nonlinearity.¹

The radiation intensity I_{d1} in first-order self-diffraction is uniquely connected with the optically induced change $\Delta n_{o,e}$ of the refractive index, viz.,

$$I_{d1} = (\pi z \Delta n_{o,e} / \lambda)^2 I_0, \quad (1)$$

where I_0 is the intensity of the incident radiation, z is the sample thickness, and the subscripts o and e label the ordinary and extraordinary light polarizations.

In the entire investigated temperature region below $T - T_c \approx -1^\circ\text{C}$ the measured basic nonlinearity characteristics at fixed temperature did not differ qualitatively from those previously investigated for conformation nonlinearity.^{2,8}

The experimental dependence of the self-diffraction intensity of the extraordinary polarized radiation on the normalized temperature $\tau = (T - T_c) / T_c$ is shown in Fig. 1. The same figure shows the temperature dependence of the characteristic hologram-erasure time t_H , which is equal to the writing time as well as to half the characteristic relaxation time of the nonlinearity.² The value of I_{d1} , which is proportional to $(\Delta n_{o,e})^2$, increases as the phase-transition point is approached, even though in this case the hologram-erasure time, which determines the stationary value of $\Delta n_{o,e}$ (Ref. 2), decreases. The dependence of the refraction intensity of the ordinary polarized radiation is similar in form.

In the phase transition region, which amounted to about 0.5 $^\circ\text{C}$, as well as at temperatures close to it

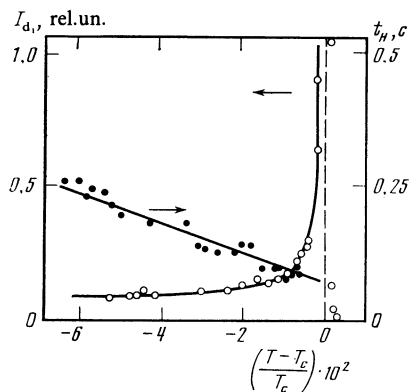


FIG. 1. Dependences of the self-diffraction intensity I_{d1} and of the effective time t_H of erasure of the holographic grating on the normalized temperature.

($-0.003 \lesssim \tau \lesssim 0.003$), nonlinear effects with various characteristics appeared. Strong self-action effects with prolonged buildup and relaxation times ($t_H \gtrsim 10$ sec) were observed near the transition into the nematic state. At higher temperatures there appeared sometimes directly in the phase-transition region additional self-diffraction orders corresponding to writing holographic gratings with vector \mathbf{q}_1 perpendicular to the basic vector \mathbf{q} , and equal to it in absolute value. We did not succeed in establishing the conditions necessary for the onset of such gratings, which indicate the appearance of two-dimensional spatial structures in the NLC. Above the clearing temperature T_c , the self-diffraction intensity decreased to zero, while near T_c ($\tau \lesssim 0.003$) (see Fig. 1.) an unusually strong nonlinearity was observed (the nonlinearity parameter was comparable with the parameter ε_2 in the mesophase). The nonlinearity lifetimes were longer than the characteristic times of the conformation nonlinearity.¹¹

DISCUSSION

1. We have shown earlier by the self-action procedure that extraordinary polarized radiation induces a diverging lens in an MBBA liquid crystal, on account of conformation nonlinearity, while ordinary polarized light induces a converging lens. This means that the change of the MBBA refractive index is negative for the e -wave and positive for the o -wave.⁸ In this case the increase of the nonlinearity on approaching the clearing point can be explained by assuming that the phase-transition temperature is lowered when photoconverted molecules appear in the NLC (see Fig. 2).

The shift of the clearing point can be qualitatively understood by recognizing that the photoconverted molecules act as impurities in the NLC volume. It is known that impurities can alter substantially the temperature T_c and the character of the phase transition, bringing the latter closer to a second-order transition.¹⁰⁻¹⁴ The change of the character of the phase transition leads to a change of the form of the temperature dependence of the NLC refractive index, while the change of T_c shifts the $n(\tau)$ curve as a whole along the τ axis by an amount $\Delta\tau$. If the density of the photoconverted molecules is small or if the molecules themselves are mesogenic, the decisive factor in the change of the refractive index

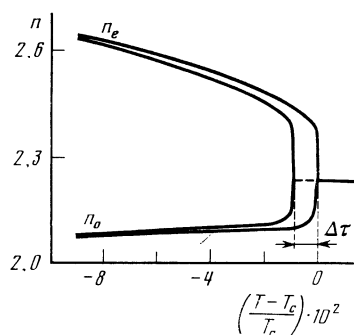


FIG. 2. Shift of temperature dependence of the refractive index of an NLC with changing temperature of the phase transition temperature T_c . The induced refractive index at a given temperature is determined by the shape of the $n(\tau)$ curve and the value of the shift of T_c , while the sign of $\Delta n_{o,e}$ is determined by the sign of the change of T_c .

will be the shift of its temperature dependence as a result of the change of T_c . Let us obtain in this approximation an expression for the increment Δn_c of the refractive index, due to the lowering of T_c . The refractive indices of an NLC, for the o - and e -waves, can be written in the form¹⁵

$$\frac{n_{o,e}^2 - 1}{n^2 + 2} = \frac{4\pi N_A \rho}{3M} [\bar{\gamma} + \beta(\gamma_e - \gamma_o)S], \quad (2)$$

where N_A is the Avogadro number, ρ is the density, M is the molecular weight, $\gamma_{e,o}$ the polarizabilities of the mesogenic molecule, $\bar{\gamma} = 1/3\gamma_e + 2/3\gamma_o$, $\bar{n}^2 = 1/3n_e^2 + 2/3n_o^2$, $\beta = 2/3$ for the o -polarization, and $\beta = 1/3$ for the e -polarization.

The main contribution to the $\Delta n_{o,e}(\tau)$ dependence is made by the temperature dependences of the order parameter and of the polarizability of the individual molecules. The refractive-index growth increment is therefore

$$\Delta n_{o,e} = \frac{2\pi N_A \rho}{3M} \frac{\bar{n}^2 + 2}{n_{o,e}} \beta \left[S \frac{\partial(\gamma_e - \gamma_o)}{\partial \tau} + (\gamma_e - \gamma_o) \frac{\partial S}{\partial \tau} \right] \Delta \tau. \quad (3)$$

The change of the temperature T_c can be treated as the result of an interaction of the macroscopic order parameter S with the microscopic order parameters Q that characterize the NLC intramolecular degrees of freedom.^{7,11} Such a degree of freedom can be, in particular, the angle between the planes of the benzene rings in the liquid-crystal molecule⁷ or the time-averaged deviation of end fragment of the mesogenic molecule.¹⁶ A change of the parameter Q alters the local electric fields, while the macroscopic order parameter S , meaning also T_c , also changes because of the short-range character of the forces that bind the NLC molecules. The interaction of Q and S makes the quantities S and $\gamma_e - \gamma_o$ in expression (3) for the induced refractive index functions of the intramolecular parameter Q .

If it is assumed that the phase-transition temperature shift is proportional to the density of the photoconverted molecules, then

$$\Delta \tau = \frac{N}{N_o} \frac{\partial \tau^*}{\partial Q} \Delta Q,$$

where $(\partial \tau^* / \partial Q) \Delta Q$ is the shift of the phase-transition tem-

perature when all the liquid-crystal molecules are converted into the new conformation state $\Delta\tau^*$.

Recognizing that in the stationary case $N = I_0\alpha_{o,e}t/h\nu$, where $\alpha_{o,e}$ is the absorption coefficient and $N_0 = N_A/\rho M$, we find ultimately that the induced change of the refractive index is proportional to the light intensity, i.e., is described by the cubic nonlinearity

$$\Delta n_{o,e} = \frac{2\pi\beta}{3} \frac{I_0\alpha_{o,e}t}{n_{o,e}h\nu} \left[S \frac{\partial(\gamma_e - \gamma_o)}{\partial\tau} + (\gamma_e - \gamma_o) \frac{\partial S}{\partial\tau} \right] \Delta\tau^*. \quad (4)$$

This coincides with the experimental results obtained for the conformational nonlinearity.² The dependence of the absorption coefficient $\alpha_{o,e}$ on the order parameter also leads to a change of the NLC refractive index. This change, however, is described by a sixth-order nonlinearity, and can be neglected at the intensities employed.

The temperature dependence of the square of the refractive index $(\Delta n_e)^2$ that determines the self-diffraction intensity in our experiments [see (1)] can be calculated by using for the employed wavelength ($\lambda = 0.44 \mu\text{m}$) a relation approximated from the experimental data:

$$n_e = 2.3 + 0.38|\tau|^{0.5} + 0.33|\tau|. \quad (5)$$

If it is assumed that the shift $\Delta\tau$ of the phase transition is small compared with the values of τ , the stationary value of the self-diffraction intensity will be proportional to the square of the first term of the expansion (5) in τ :

$$I_{d_1} \propto [(0.19|\tau|^{-0.5} + 0.33)\Delta\tau]^2, \quad (6)$$

where, in accord with (4), the shift of the phase-transition temperature is $\Delta\tau = 2\Delta\tau^*t_H$, while $t_H(\tau)$ is the experimentally obtained erasure time of the holographic gratings.

The calculation corresponding to (6) is represented by the solid line in Fig. 1. The dark points are the experimental $t_H(\tau)$ and their variation was approximated by a straight line sloped to yield best agreement between calculation and experiment. It can be seen that the calculated curve fits the experimental data within the error limit.

We can thus state that the refractive-index change due to the conformational nonlinearity is described, within the experimental accuracy, by a shift of the $n(\tau)$ dependence along the τ axis, without changing the form of the function $n(\tau)$ itself; this can be treated as a lowering of the phase-transition temperature T .

2. We have performed an independent experiment to observe the phase-transition temperature shift following illumination of an NLC by a helium-cadmium laser. The change of the temperature T_c was recorded in terms of the change of the birefringence of the liquid crystal by the action of this radiation.

The measurements were made with a nonlinear polarization interferometer (see Fig. 3). A thermostat with the liquid crystal was placed between two crossed polarizers p_1 and p_2 . The beam from a helium-neon laser ($\lambda = 0.638 \mu\text{m}$), whose ε_2 is vanishingly small, was broadened by a telescope T and passed through polarizers into a cell with the NLC. Polarized and intensity-modulated emission from a helium-cadmium laser was also guided into the cell with the crystal whose birefringence it altered on account of conformation

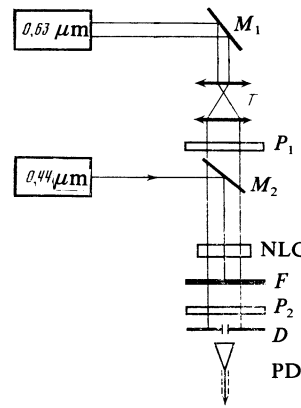


FIG. 3. Diagram of nonlinear polarization interferometer: M —mirrors, T —telescope, P —polarizers, F —filter absorbing the $\lambda = 0.44 \mu\text{m}$ radiation, D —diaphragm, PD —photodiode.

nonlinearity. The He-Cd laser emission was modulated in such a way that the induced birefringence reached alternately either its stationary value or a zero value. The shutter operating time was in this case substantially shorter than the characteristic nonlinearity-establishment times.

The diameter of the helium-cadmium laser beam was substantially less than the diameter of the helium-neon laser beam. The interferometer output was the interference pattern of the polarized beams of the helium-neon laser, against the background of which was observed a periodically time-varying intensity region corresponding to the action of the helium-cadmium laser.

With the shutter open, the emission intensity in the He-Cd laser illumination region varies as

$$I_1 \propto \cos^2 \left\{ \frac{\pi z}{\lambda} [\Delta n^0(\tau) + (\Delta n_e + \Delta n_o)(\tau)(1 - e^{-\tilde{t}/2t_H})] \right\}, \quad (7)$$

where Δn^0 is the MBBA intrinsic birefringence, $\Delta n_e + \Delta n_o$ is the stationary induced birefringence, and \tilde{t} is the time reckoned from the instant of shutter operation. At $\tilde{t} \gg 2t_H$ this expression yields the stationary value of the birefringence of the mixture of the ground state and of the modified temperature-dependent conformational state of the liquid crystal.

When the shutter is closed, the intensity change is given by the expression

$$I_2 \propto \cos^2 \left\{ \frac{\pi z}{\lambda} [\Delta n^0(\tau) + (\Delta n_e + \Delta n_o)(\tau)e^{-\tilde{t}/2t_H}] \right\}, \quad (8)$$

which determines in the stationary case the birefringence of the basic MBBA crystal at a given temperature.

The temperature dependences of I_1 and I_2 corresponding to the stationary values of (7) and (8) are shown in Fig. 4. The relative shift of these curves points to a change of the phase-transition temperature under the action of the helium-cadmium laser radiation. This change can be determined, starting from the equality of I_1 and I_2 , from the condition

$$\frac{\pi z}{\lambda} [\Delta n^0(\tau - \Delta\tau) - \Delta n^0(\tau)] = \Delta(\Delta n). \quad (9)$$

Here $\Delta(\Delta n)$ is the birefringence change determined from the

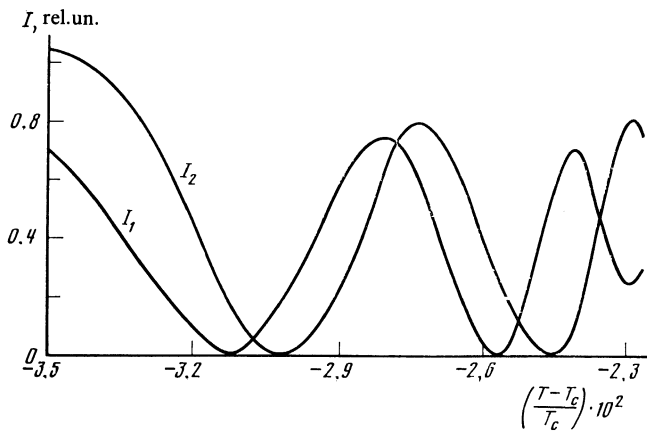


FIG. 4. Temperature dependence of the intensity of the light passing through the polarization interferometer: I_1 —illumination at $\lambda = 0.44 \mu\text{m}$, I_2 —with illumination.

shift of the experimental curves and leading to a phase advance equal to that in the absence of external illumination, while $\Delta n^0(\tau)$ is described, on the basis of an approximation of the published data,^{17,18} by the expression

$$\Delta n^0 = 0.11 + 0.43 |\tau|^{0.5} + 0.1 |\tau|. \quad (10)$$

Solution of (9), assuming conservation of the function (10) under phototransformation, yields $\Delta\tau \approx 3 \cdot 10^{-4} (\Delta T_c \approx 10^{-1} \text{ K})$. Such a shift of the phase-transition temperature leads, according to (6), to a refractive-index change $\Delta n_e \approx 5 \cdot 10^{-4} (T_c = 22^\circ \text{ C})$. This value of Δn_e corresponds to a nonlinearity parameter $\varepsilon_2 \approx 0.1 \text{ cm}^3/\text{erg}$, of the same order as that previously obtained.^{2,8}

We note that the obtained considerable changes of the birefringence under saturation conditions cannot be due to heating of the sample by light absorption. Indeed, even in the absence of heat exchange, at a power density $I_0 \approx 1 \text{ W/cm}^2$, at an absorption coefficient $\alpha \approx 25 \text{ cm}^{-1}$, and at a cell thickness $50 \mu\text{m}$ the sample heating during the time of action of the laser pulse did not exceed 0.02 K , lower by approximately an order of magnitude than the ΔT_c obtained by us. That the thermal contribution is small is indicated also by the fact that the nonlinearity parameter ε_2^T for the thermal mechanism is several orders lower than that measured in experiment.² The possible heating of the sample by the continuous emission of the testing helium-neon laser influences only the rate of sample cooling and introduces no measurement error.

Starting from the linear dependence, we can estimate the shift ΔT_c^* of the phase-transition temperature of photoconverted molecules relative to the temperature T_c of the unperturbed sample. The experimentally recorded shift $\Delta T_c \approx 0.1^\circ \text{ C}$ corresponds to a relative density $N/N_0 \approx 0.1$, therefore $\Delta T_c^* \approx 1^\circ \text{ C}$.

Measurement of the temperature dependences of the birefringence, as well as holography experiments, points thus to a shift of the temperature T_c upon phototransformation, and the value of shift leads to observable values of the nonlinearity parameter.

3. Expression (4) shows that the change of the refractive index is connected with the change of the NLC-molecule

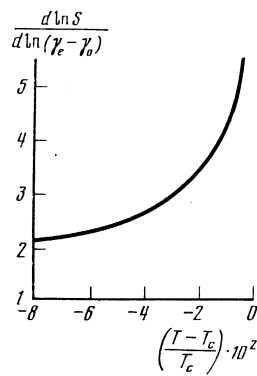


FIG. 5. Temperature dependence of the relative contribution of the change of the order parameter S and of the molecular polarizability γ to the conformational nonlinearity.

polarizability as well as with the change of the order parameter S . Using the temperature dependences of the refractive indices and the known $S(\tau)$ dependence¹⁹ we can estimate the contribution of the change of the order parameter and of the molecular polarizabilities to the nonlinearity. The temperature dependence of the anisotropy of the molecular polarizability can be calculated starting from the Vuks formula¹⁵

$$(\gamma_e - \gamma_o) = S^{-1} (\bar{\alpha}_e - \bar{\alpha}_o), \quad \bar{\alpha}_e - \bar{\alpha}_o = \frac{3}{4\pi} \frac{M}{N_A \rho} \frac{n_e^2 - n_o^2}{n^2 + 2},$$

which contains the difference of the NLC polarizabilities. Figure 5 shows $d \ln S / d \ln (\gamma_e - \gamma_o)$ vs temperature. It can be seen that the contributions of both mechanisms are comparable in size, and when the phase transition temperature is approached the contribution to the nonlinearity of the change of the order parameter increases.

4. We obtain now an expression for the components of the NLC nonlinear-susceptibility tensor due to a shift of the phase-transition temperature. In our case the third-order nonlinear polarizability $P^{(3)}$ can be written in the form

$$P_i^{(3)} \sim \alpha_{ij} E_i E_j \frac{dn_{kl}}{d\tau} \Delta\tau E_k. \quad (11)$$

Starting from this relation we can formally introduce the cubic nonlinear susceptibility tensor $\chi^{(3)}$ whose components are given by

$$\chi_{ijkl} \sim \alpha_{ij} \frac{dn_{kl}}{d\tau} \Delta\tau.$$

The symmetry of the tensor $\chi^{(3)}$ for the conformation-phototransformation mechanism admits of only four non-zero components of the tensor χ_{ijkl} .² The relations between the components χ_{ijkl} and their signs can be qualitatively understood on the basis of Fig. 2. Quantitative estimates can be obtained by using the relations for the refractive indices n_o and n_e , as well as the fact that for the wavelength $\lambda = 0.44 \mu\text{m}$ we have $\alpha_{11}/\alpha_{33} \approx 0.4$. This yields

$$\chi_{1111} = -0.075 \chi_{3333}, \quad \chi_{3311} = 0.22 \chi_{3333}, \\ \chi_{1133} = -0.5 \chi_{3333}, \quad \chi_{1111} > 0, \quad \chi_{3333} < 0.$$

Earlier experimental data^{2,8} yield the close values

$$\chi_{1111}^e = -0.1\chi_{3333}^e, \quad \chi_{3311}^e = 0.44\chi_{3333}^e,$$

$$\chi_{1133}^e = -0.44\chi_{3333}^e, \quad \chi_{1111}^e > 0, \quad \chi_{3333}^e < 0.$$

The value of χ_{3311}^e does not agree with our estimate. The discrepancy is possibly due to the insufficient accuracy of the approximation of $n_o(\tau)$ as well as to the fact that the change of the refractive index for the ordinary wave is connected not only with the shift but also with the change of the form of the function $n_o(\tau)$ during phototransformation.

CONCLUSION

It can thus be regarded as established that the main cause of the "giant" nonorientational nonlinearity of nematic liquid crystals is the conformation change of the mesogenic molecule upon phototransformation, which leads to a change of the intramolecular order parameter. The interaction of this parameter, which characterizes the intramolecular degrees of freedom of the molecule, with the macroscopic order parameter of the NLC leads to a lowering of the phase transition temperature T_c , meaning also to a change of the refractive index of the liquid crystal. The change of the refractive index is due both to a change of the order parameter S and to a change of the anisotropy of the polarizability of the mesogenic molecule. The contributions of both mechanisms to the nonlinearity are comparable far from the phase transition, and as the latter is approached the contribution of the change of the order parameter S increases.

From the proposed model it follows naturally that the change of the refractive index is positive for the ordinary wave and negative for the wave with the extraordinary polarization. Also agreeing in the main with the experimental data are the relations between the values of the individual components of the nonlinear cubic susceptibility.

The observed nonlinearity mechanism can be used hereafter to investigate the effect of impurities on the character of phase transitions in liquid crystals, inasmuch as the experiments afford a unique possibility of controlling the impurity density. To realize such investigations it is necessary to explain the form of the photoconformation transformations of mesogenic molecules. Once the intermolecular order parameter responsible for the change of the refractive index is determined, dynamic holography methods can be

used to study the interaction of this order parameter with the macroscopic parameter S . This problem is now exceedingly vital for the understanding of the nature of phase transitions in liquid crystals.

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¹⁾Unfortunately, the insufficient accuracies of the temperature stabilization and of the material purity, as well as the uncontrollable presence of oxygen and moisture in MBBA, which lead to gradual lowering of the temperature T_c (Ref. 9), have made detailed investigation of these effects impossible so far.

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