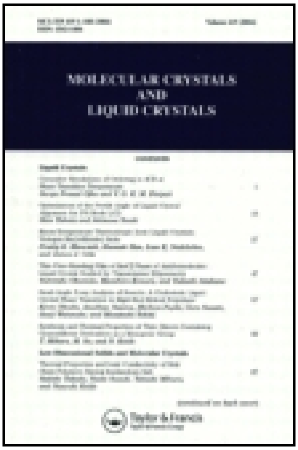

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Laser-Induced Optical Nonlinearities In Ferronematics

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LASER-INDUCED OPTICAL NONLINEARITIES IN FERRONEMATICS

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Nematic and ferronematic samples were subjected to high power laser irradiation. Optical nonlinearities such as diffraction rings and self focussing effects were examined as function of light intensity. It is shown that the self-focusing effect appearing in a ferronematic (obtained by dispersing 0.1 % yttrium iron garnet into nematic 4799 Merck) is smaller when compared to that encountered in pure nematic. A theoretical approach was elaborated to explain these optical nonlinearities in ferronematics and good agreement with the experimental findings was found.

Keywords: ferronematics; laser irradiation; nematic liquid crystals; optical non-linearities

1. INTRODUCTION

High power laser irradiation effects in nematic liquid crystals have been investigated since a long time. They consist in changes of the refractive index and therefore in laser beam propagation in these media [1–7]. Phenomena such as self-phase modulation, self-focusing, optical harmonic generation were reported. These phenomena have not been investigated in ferronematics so far and it is the aim of this paper to do that.

The pioneering paper referring to ferronematics belongs to Brochard and de Gennes [8] who examined the behaviour under magnetic field of a nematic liquid crystal (NLC) containing a small fraction of magnetic particles. These mixtures were termed ferronematics and were extensively examined experimentally by other investigators [9,10]. It has been proved

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that the critical field for magnetic Freedericksz transition was lowered in case of embedding the magnetic particles in lyotropic nematics [11] and was enhanced in case of thermotropic nematics [12]. To explain these discrepancies, Burylow and Raikher [13] developed a theoretical model relying on the assumption that the magnetic moment of the magnetic particles are oriented perpendicularly to the nematic director. Based on this assumption they estimated first the free energy of the ferronematic and then the critical field for magnetic Freedericksz transition.

In this paper a theoretical aproach is elaborated to explain nonlinear optical effects occurring in ferronematics subjected to high power laser irradiation. First the free energy density of the ferronematic was calculated considering the interaction energy between the electromagnetic wave and the liquid crystal. Then, using the model of Tabyrian *et al.* [14,15] the deviation angle of the electromagnetic wave penetrating the ferronematic, which behaves as an optical lens with a convergence f^{-1} depending on the laser power, was estimated. The theoretical results were in good agreement to the experimental findings obtained when sujecting the ferronematic (obtained by dispersing 0.1% yttrium iron garnet into nematic 4799 Merck) to high power laser irradiation.

2. THEORETICAL APPROACH

2.1. The Free Energy Density

In order to explain the optical nonlinearities induced in ferronematics by high power laser irradiation we estimated first the free energy density of the ferronematic. It consists of:

1. The elastic free energy of the pure nematic (Frank energy) given by

$$F_F = \frac{1}{2}\left[K_1(\nabla\vec{n})^2 + K_2(\vec{n}\nabla\vec{n})^2 + K_3(\vec{n}\times(\nabla\times\vec{n}))^2\right] \quad (1)$$

where K_1, K_2, K_3, are the spay, twist and bend elastic constants, respectively and $\vec{n}$ the molecular director. Assuming a planar orientation for the ferronematic and \$ $n_y = 0$ (see Fig. 1) this relationship simplifies to

$$F_F = \frac{1}{2}K_1\left(\frac{\partial n_z}{\partial z}\right)^2 \quad (2)$$

2. An interaction energy between the electromagnetic wave and the liquid crystal

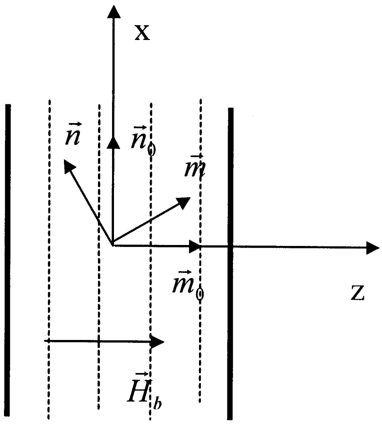

FIGURE 1 The choice of the coordinate frame and the orientation of nematic director and magnetic particle moment in the absence ($\vec{n}_0$, $\vec{m}_0$) and in the presence ($\vec{m}$, $\vec{n}$) of an external perturbation.

$$F_E = -\frac{1}{16\pi}\varepsilon_{ik}(\vec{r})E_i(\vec{r})E_k^*(\vec{r}) \tag{3}$$

where $\varepsilon_{ik} = \varepsilon_\perp \delta_{ik} + \varepsilon_a n_i n_k$ and $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the dielectric anisotropy of the nematic.

3. A dissipative energy which may be considered of the form:

$$F_d = \frac{1}{2}\gamma\, \dot{n}^2 \tag{4}$$

where γ is orientational viscosity coefficient and $\dot{n} = \dfrac{\partial n}{\partial t}$

4. A free energy due to the contribution of the magnetic particles. According to Burylov and Raikher [10], this is given by:

$$F_m = -\frac{1}{2}\chi_a\left(\vec{n}\vec{H}_b\right)^2 - M_s f\left(\vec{m}\vec{H}_b\right) + \frac{fW}{a}(\vec{n}\vec{m})^2 \tag{5}$$

In (5) χ_a is the diamagnetic anisotropy, $\vec{H}_b$ an internal magnetic field giving a saturation of the magnetization, f the volume fraction of the magnetic particles with the mean diameter a, magnetic moment $\vec{m}$ and saturation magnetization $\vec{M}_s$. W is the magnetic surface energy of the magnetic particle–nematic boundary.

According to the hypothesis of Burylov and Raikher, the magnetic moment of the magnetic particle $\vec{m}$ is perpendicular to the nematic director $\vec{n}$ when no external perturbation is present. Under the action of an electromagnetic wave of amplitude E their directions are changed as it is shown in Figure 1. Therefore, the relationship (5) simplifies to:

$$F_m = -\frac{\chi_a H_b^2 n_z^2}{2} + \frac{M_s f H_b}{2} m_x^2 + \frac{f\,W}{a}(m_x + n_z)^2 \tag{6}$$

and the total free energy density of a ferronematic is:

$$\begin{aligned} F =& \frac{1}{2} K_1 \left[\left(\frac{\partial n_z}{\partial z} \right) \right]^2 - \frac{\varepsilon_a}{16\pi} \left(E_x E_z^* + E_z E_x^* \right) n_z \\ & - \frac{\chi_a H_b^2 n_z^2}{2} + \frac{M_s f H_b}{2} m_x^2 + \frac{f\,W}{a}(m_x + n_z)^2 \end{aligned} \tag{7}$$

The Euler-Lagrange equations are:

$$\frac{\partial}{\partial z}\left(\frac{\partial F}{\partial\left(\dfrac{\partial m_x}{\partial z}\right)} \right) - \frac{\partial F}{\partial m_z} = 0, \quad \frac{\partial}{\partial z}\left(\frac{\partial F}{\partial\left(\dfrac{\partial n_z}{\partial z}\right)} \right) - \frac{\partial F}{\partial n_z} = 0 \tag{8}$$

From the first Eq. (8) we get a relationship between m_x and n_z:

$$m_x = \frac{-2W/a}{M_s H_b + 2W/a} n_z \tag{9}$$

The second Eq. (8) gives:

$$\gamma \frac{\partial n_z}{\partial t} - K_1 \frac{\partial^2 n_z}{\partial Z^2} - \chi_a H_b^2 n_z - \frac{2f\,W}{a}(n_z + m_x) = \frac{\varepsilon_a}{16\pi}\left(E_x E_z^* + E_z E_x^* \right) \tag{10}$$

Denoting:

$$C = \frac{\varepsilon_a}{16\pi}\left(E_x E_z^* + E_z E_x^* \right) \tag{11}$$

$$D = -\chi_a H_b^2 + \frac{\dfrac{2W}{a}(\,M_s f H_b\,)}{M_s H_b + \dfrac{2W}{a}} \tag{12}$$

and substituting Eq. (9) into Eq. (10) one obtains:

$$\gamma \frac{\partial n_z}{\partial t} - K_1 \frac{\partial^2 n_z}{\partial z^2} + D n_z = C \tag{13}$$

The solutions of Eq. (13) are obtained when taking into account the initial and boundary conditions. These are: $n_z(z,0)=0$ and $n_z(0,t)=n_z(d,t)=0$, where d is the liquid crystal cell thickness. The general solution is:

$$n_z(z)=\sum_{j=1}^{\infty} B_j \sin\frac{j\pi z}{d} \tag{14}$$

Introducing (14) into Eq. (13), multiplying by $\sin\frac{m\pi z}{d}$ and integrating over $(0,d)$ we get:

$$\frac{\gamma}{K_1}\frac{dB_m}{dt}+B_m\frac{m^2\pi^2}{d^2}\left(1+\frac{D^2d^2}{K_1m^2\pi^2}\right)=\frac{2C}{K_1n\gamma\pi}\left[1-(-1)^m\right] \tag{15}$$

This equation may be written as:

$$\frac{\gamma}{K_1}\frac{dB_m}{dt}+P_mB_m=R_m \tag{16}$$

where

$$\frac{m^2\pi^2}{d^2}\left(1+\frac{D}{K_1}\frac{d^2}{m^2\pi^2}\right)=P_m \tag{17}$$

$$\frac{2C}{K_1m\pi}\left[1-(-1)^m\right]=R_m \tag{18}$$

The solution of Eq. (16) is:

$$B_m=\frac{d^2\varepsilon_a}{8K_1m^3\pi^4}\frac{(E_xE_z^*+E_zE_x^*)[1-(-1)^m]}{1+\frac{D}{C_1}\frac{d^2}{m^2\pi^2}}\left[1-\exp\left(-\frac{K_1P_m}{\gamma}t\right)\right] \tag{19}$$

2.2. Optical Nonlinearities in Nematics

The optical nonlinearities in nematic liquid crystals (NLC) induced by high power laser irradiation were thoroughly investigated by Tabiryan *et al.* [14,15]. Their results may be found as a particular case of ferronematics when assuming $D=0$. Therefore $P_m=m^2\pi^2/d^2$ and B_m reduces to A_m which means:

$$A_m=\frac{\varepsilon_a d^2\left(E_xE_z^*+E_zE_x^*\right)}{8\pi^4m^3K_1}\left[1-(-1)^m\right]\left[1-\exp\left(-\boldsymbol{\Gamma}_m t\right)\right] \tag{20}$$

where

$$\Gamma = -\frac{K_1 m^2 \pi^2}{\gamma d^2} \tag{21}$$

and the general solution is:

$$n_z(z,t) = \sum_{m=1}^{\infty} A_m(t) \sin \frac{m\pi z}{d} \tag{22}$$

According to Tabiryan *et al*. [14] a simplified solution is obtained under the following conditions.

First, if the equilibrium state of the system is reached in a rather short time $\exp(-\Gamma_m t) \cong 0$; this means that the dissipative effects may be neglected and therefore n_z does not depend on time. Second, in the sum (22) only the odd number give a non-zero contribution. As estimated [14] the contribution of the mode $m = 3$ is 27 smaller when compared to that corresponding to the mode $m = 1$. Therefore, instead of the general solution (22) we may consider a simplified one:

$$n_z(z) = A_1 \sin \frac{\pi z}{d} = \frac{\varepsilon_a d^2 (E_x E_z^* + E_z E_x^*)}{4\pi^4 K_1} \sin \frac{\pi z}{d} \tag{23}$$

Assuming an oblique incidence of the electromagnetic wave to the planar NLC (see Fig. 2) one obtains:

$$E_x E_z^* + E_z E_x^* = -2|E_0|^2 \sin\beta \cos\beta \tag{24}$$

and instead of (23):

$$n_z(z) = -\frac{\varepsilon_a d^2 \left|E_0^2\right| \sin\beta \cos\beta}{2\pi^4 K_1} \sin \frac{\pi z}{d} \tag{25}$$

It has been also demonstrated by Tabyrian *et al*. that the deviation experienced by the electromagnetic wave in a nematic layer with thickness d is:

$$\theta_m = \frac{1}{\sqrt{\varepsilon_0} a_\perp} \frac{\varepsilon_a^2 |E_0|^2 d^3}{\pi^5 K_1} \sin^2\beta \cos\beta \tag{26}$$

where $a_\perp$ is the cross section of the laser beam with a bell shaped profile. Such a medium behaves like an optical lens with the convergence:

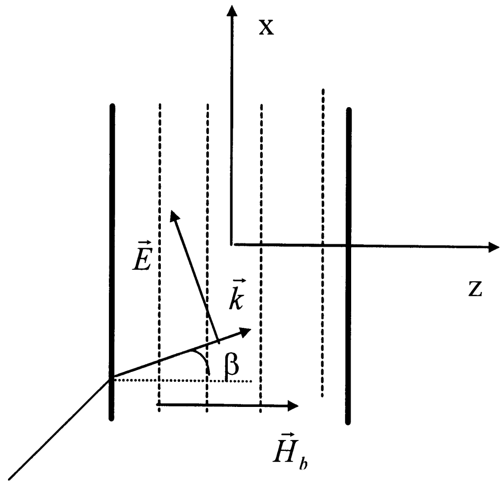

FIGURE 2 Oblique incidence of a laser beam on a nematic liquid crystal homotropically aligned.

$$f_n^{-1} = \frac{4\varepsilon_a |E_0|^2}{\sqrt{\varepsilon_a}\pi^5 K_1 a_\perp^2} \sin^2 \beta \cos \beta \tag{27}$$

Another phenomenon which is characteristic when subjecting NLC to high power radiation is due to the angular divergence which gives rise to a peculiar form of circular fringes originating from the phase difference between the periphery $\varphi\,(r \to \infty)$ and the beam center $\varphi(r = 0)$. The number of fringes is given by:

$$N = \frac{\varphi(r \to \infty) - \varphi(r = 0)}{2\pi} = \frac{\varepsilon_a^2 d^3 |E_0|^2}{\sqrt{\varepsilon_0}\lambda\pi^2 K_1} \sin^2 \beta \cos \beta \tag{28}$$

2.3. Optical Nonlinearities in Ferronematics

It is reasonable to assume that when ferronematics are involved only the first term of the Eq. (14) gives the main contribution. The coefficient B_1 is related to A_1 by means of the following relationship:

$$B_1 = \frac{A_1}{1 + \frac{D}{K_1}\frac{d^2}{\pi^2}} \tag{29}$$

Using the same procedure as before we get for the deviation angle $(\theta_m)_{fm}$ in a ferronematic the following relationship:

$$(\theta_m)_{fn} = \frac{\theta_m}{1 + \frac{D}{K_1}\frac{d^2}{\pi^2}} \tag{30}$$

and for the convergence of the focal lens:

$$f_{fn}^{-1} = \frac{f_n^{-1}}{1 + \frac{D}{K_1}\frac{d^2}{\pi^2}} \tag{31}$$

As it may be seen, when using ferronematics, the deviation angle and the convergence of the focal lens are smaller when compared to those obtained for pure nematics. The same occurs when estimating the number of diffraction rings.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Sample Preparation

The ferronematics were obtained by first mixing and then sonicating for 15 minutes a powder of yttrium iron garnet (YIG) into a nematic liquid crystal (Merck 4799). The magnetic powder had a mean diameter $a = 3 \times 10^{-8}$ m and a volume fraction of $f = 10^{-4}$. Both nematic and ferronematic were introduced by capillarity into liquid crystal cells which were previously chemically processed for planar alignment. In both cases the thicknesses of the cells were 300 µm.

3.2. Equipment

The experimental equipment is shown in Figure 3. A beam of He-Ne laser $\lambda = 633$ nm was focused by means of a lens with the focal distance $f = 25$ cm on the liquid crystal cell. This one was mounted on a support which enabled changes in its orientation with respect to the incident laser beam. It has been noticed that when the laser power was above a certain threshold, a characteristic pattern consisting in concentric rings was noticed on a projecting screen; this one was positioned at a distance $L = 1$ m from the liquid crystal cell. As it has been shown [14,15] the diffraction pattern originates from the intercepts of conical shells (corresponding to different angles of deviation θ_m) with the projecting screen. A camera provided with a red filter (to diminish the intensity of the emergent light) was used to register the diffraction pattern.

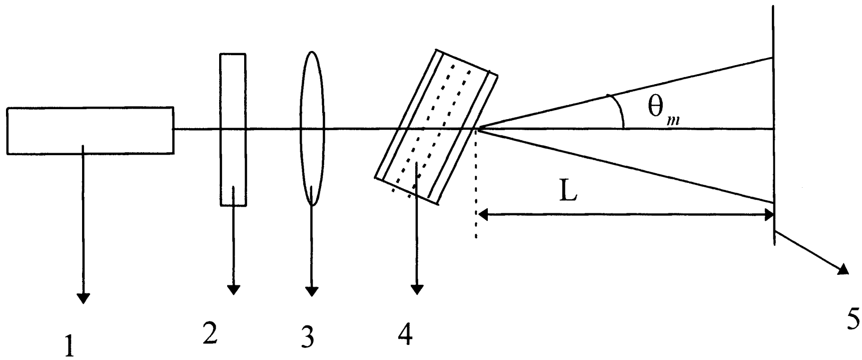

FIGURE 3 Experimental equipment : 1-He-Ne laser , 2-filter , 3-projecting lens ($f = 25\,\text{cm}$), 4-liquid crystal cell, 5-projecting screen.

3.3. Results and Discussion

First the incidence of the laser beam on the liquid crystal cell was kept constant ($\alpha = 45°$) and the laser power was gradually increased. Typical diffraction ring patterns for nematic samples subjected to a beam power of 20 mW/cm are shown in Figure 4.

When the laser power was increased, the number of diffraction rings and the lens convergence were increased. As it is shown in Figure 5 the convergences of the nematic, respectively of the ferronematic lenses increase linearly with the laser power as expected from the relationships (31). Although these dependencies are described by straight lines, the slopes are different. From (27) and (31) we can get the ratio of these slopes; it is given by:

$$r = \frac{B_1}{A_1} = \frac{1}{1 + \frac{D}{K_1}\frac{d^2}{\pi^2}} \simeq \left(1 - \frac{d^2 D}{K_1 \pi^2}\right) \tag{32}$$

and it depends on the volume fraction of the magnetic powder. Introducing the specific parameters, such as $W = 5 \times 10^{-7}\,\text{dyn/cm}$, $f = 10^{-3}$, $a = 3 \times 10^{-6}\,\text{cm}$, $M_s = 10^2$ G, $H_b = 10$ Oe and $\chi_a = 1.25 \times 10^{-7}$ we get $D = 3.3 \times 10^{-4}$. Using $d = 3 \times 10^{-2}\,\text{cm}$ and $K_1 = 2.75 \times 10^{-7}\,\text{dyn}$ it results $\frac{d^2 D}{K_1 \pi^2} = 10^{-1}$. Therefore, $r = 0.91$; this agrees with the experimental findings.

In order to explain the time evolution of the diffraction rings we estimated the time-dependent solution of the Euler-Lagrange equation. When the nematic is involved, this is given by:

$$n_z(z,t) = -\left\{1 - \exp\left(-\frac{K_1 \pi^2}{\gamma d^2} t\right)\right\} \frac{\varepsilon_a d^2 |E_0^2| \sin\beta\cos\beta}{2\pi^4 K_1} \sin\frac{\pi z}{d} \tag{33}$$

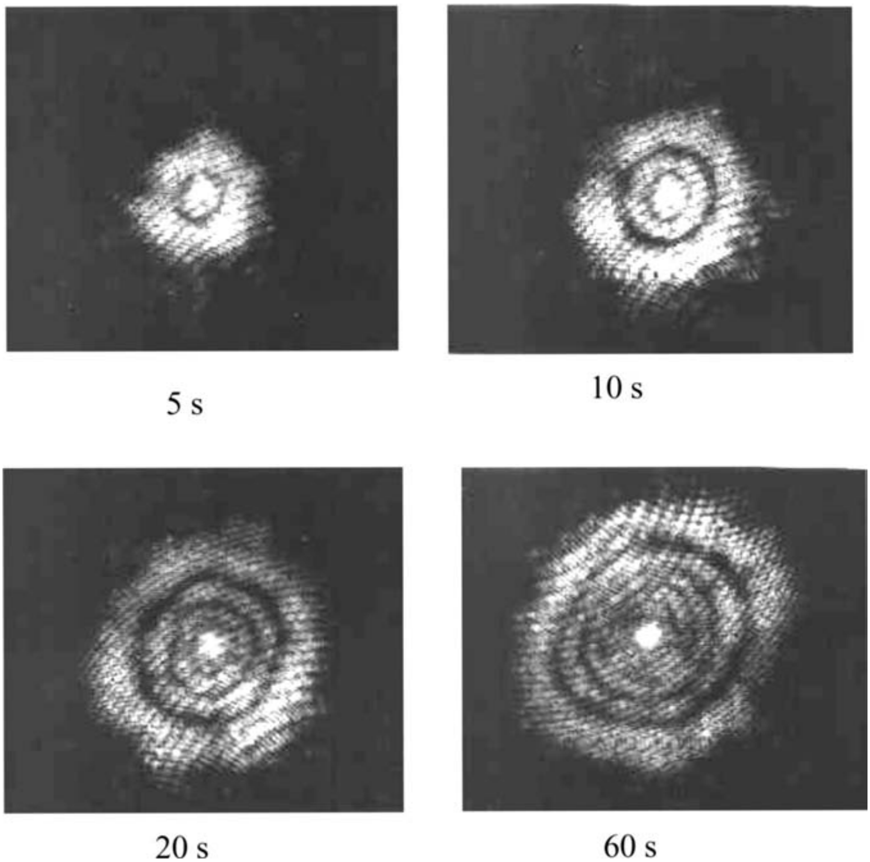

FIGURE 4 Time evolution of the diffraction patterns obtained after subjecting nematic cells to a laser beam of 20 mW.

Using the same procedure as in [14] we obtained:

$$N = \left[1 - \exp\left(-\frac{K_1\pi^2}{\gamma d^2}t\right)\right]\frac{\varepsilon_a^2 d^3|E_0|^2}{2\pi^4 K_1}\sin^2\beta\cos\beta \tag{34}$$

Therefore:

$$N = \left[1 - \exp\left(-\frac{K_1\pi^2}{\gamma d^2}t\right)\right]N_{\max} \tag{35}$$

This relationship shows that the number of rings increases with time reaching the maximum value $N_{\max}$ when $t \to \infty$. Taking into account the Eq. (28) we get:

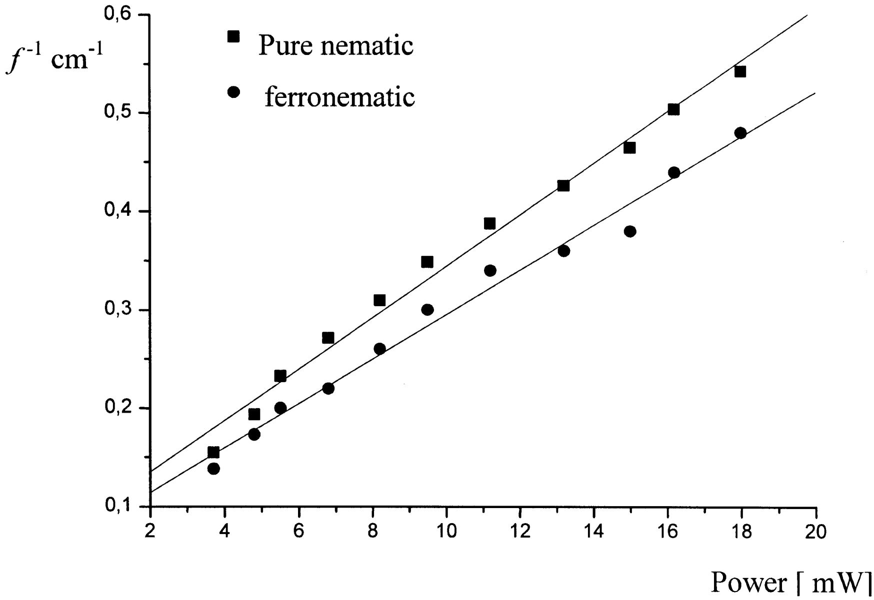

FIGURE 5 The convergence of the nonlinear lens as function of the laser beam power.

$$N_{\max} = N_n \tag{36}$$

A similar relationship is obtained for ferronematics. The maximum number of rings in this case $(N_{\max})_{fn} = N_{fn}$, is:

$$N_{fn} = \frac{N_n}{1 + \frac{D}{K_1}\frac{d^2}{\pi^2}} \tag{37}$$

The time evolution of the diffraction rings is shown in Figure 6 for both nematic and ferronematic samples. The number of the diffraction rings increases with time reaching a constant value; this result agrees with the relationships (36).

Another set of experiments were performed when keeping constant the laser power (20 mW) and changing the incidence of the laser beam on the liquid crystal cell. The results of the experiments are plotted in Figure 7. It may be noticed that the convergence of the lens f^{-1} increases linearly with the parameter $\sin^2\beta\cos\beta$, in good agreement with the relationship (27). Here β is the reflection angle experienced by the laser beam with the incidence α ($\sin\beta = n\sin\alpha$, where n is the the refractive index). As it was

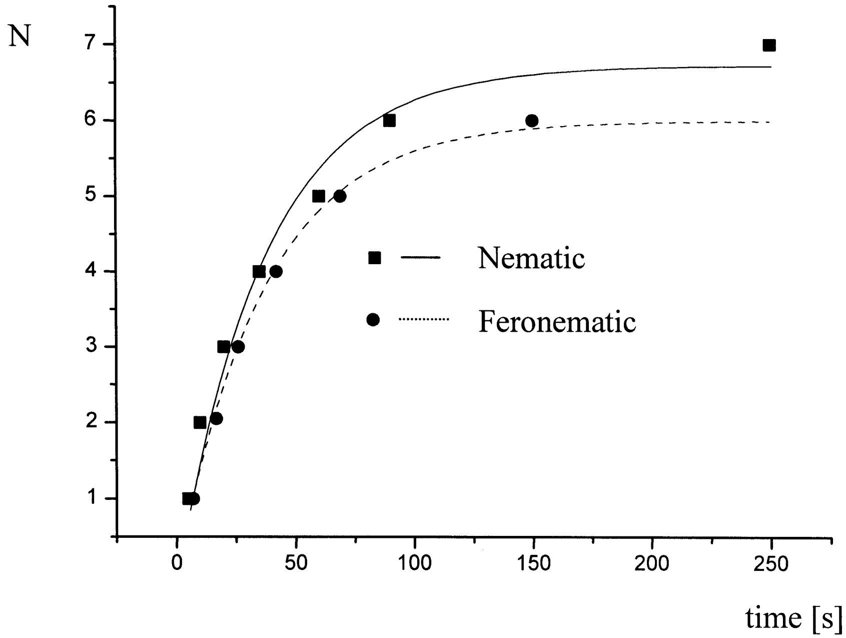

FIGURE 6 The number of diffraction rings displayed by nematic, respectively ferronematic samples (subjected to a laser power of 20 mW) as function of time.

noticed previously, the slopes for the nematic and ferronematic are different, their ratio being given by (32).

4. CONCLUSIONS

The self-focusing effects induced by a high power laser beam in ferronematics are described by similar relationships encountered for pure nematics. The only difference consists in the parameter $r = 1/(1 + Dd^2/K_1\pi^2)$ involved in the equations giving the number of the diffraction rings, the deviation angle of the laser beam and the convergence of the lens. As a result of this the effects induced by a laser beam into a ferronematic are smaller when compared to those induced in nematics. This arises because the magnetic moment of the magnetic particles are oriented perpendicularly to the nematic director and consequently the molecular orientation is hindered when the ferronematic is subjected to an electromagnetic field. This behavior is similar to that encountered when a magnetic field is acting on the ferronematic (the critical field for magnetic

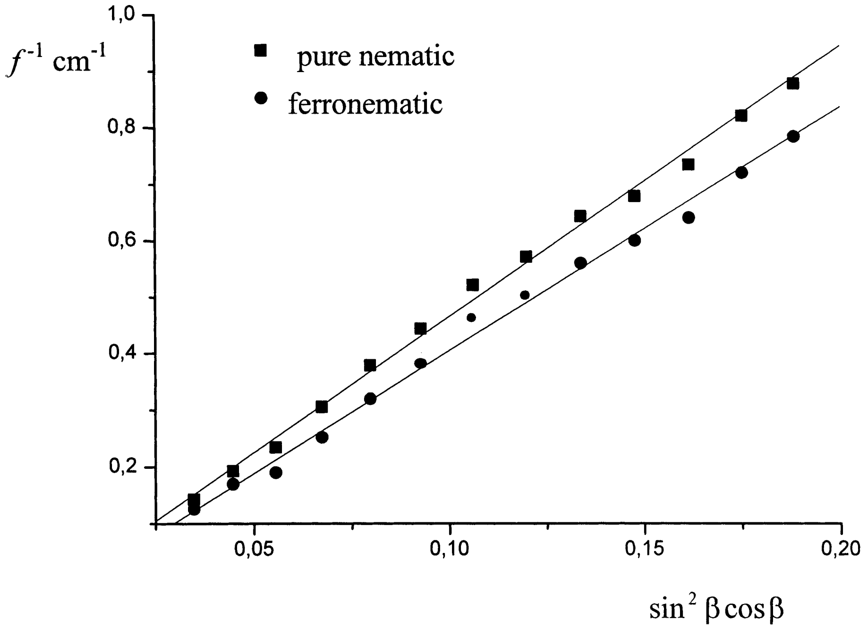

FIGURE 7 The convergence of the nonlinear lens as function of $\sin^2 \beta \cos \beta$.

Freederichsz transition is enhanced). This study provides potential application in obtaining systems with variable focal lenses which may be obtained by adding different percentages of magnetic powder into nematic liquid crystals.

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