Superparamagnet in an inverted state

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We consider the relaxation of a superparamagnet in an external magnetic field from a strong-nonequilibrium inverted state into the equilibrium state at various temperatures. The equations corresponding to the Fokker-Planck equation are uncoupled both by analytic approximation and by direct computer integration of rather long chains of equations. The existence is established of a critical temperature $T_c \approx 0.3$ MHV/k, at which the character of the relaxation of the transverse components of the magnetic moment of the superparamagnet changes. At $T < T_c$ the ferromagnetic type of relaxation predominates: during the first stage of the relaxation process the transverse-relaxation parameter is negative and the transverse components increase. At $T > T_c$ the paramagnetic relaxation predominates: phase randomization due to thermal fluctuations makes the transverse-relaxation parameter negative and the transverse components attenuate from the very start of the relaxation process.

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1. INTRODUCTION. PARAMAGNETIC AND FERROMAGNETIC TYPES OF RELAXATION

We consider an arbitrary spin system at equilibrium, situated in an external magnetic field directed along the z axis: $H_z = H$. At an instant of time $t = 0$ we reverse "instantaneously" (i.e., within a time much shorter than all the characteristic times of the system) the direction of the magnetic field: $H_z = -H$. At this instant the spin system turns out to be in an inverted strong-nonequilibrium state and relaxes subsequently to a new equilibrium state corresponding to the magnetic field $H_z = -H$. This situation is of interest because for a certain time, so long as the spin-level population remains inverted, the system has maser properties at the frequency of the corresponding magnetic resonance (EPR, FMR, etc.).

It is clear that, depending on the concrete properties of the spin system, the process of relaxation from a strong-nonequilibrium inverted state to an equilibrium state can vary greatly in character. Let us examine, for example, the evolution of such a process in a classical paramagnetic or a classical ferromagnetic system. For a paramagnet situated in an external magnetic field H_z, the general solution of the Bloch equation takes the form

$$M^{\pm} = M_0^{\pm} \exp(\pm i\gamma H_z t - \Gamma_\perp t), \quad M_z = M_z^{\infty} + (M_z^0 - M_z^{\infty}) \exp(-\Gamma_\parallel t), \quad (1.1)$$

where $\Gamma_\perp$ and $\Gamma_\parallel$ are respectively the transverse and longitudinal relaxation parameters $\tau_\perp = \Gamma_\perp^{-1}$ and $\tau_\parallel = \Gamma_\parallel^{-1}$ are the corresponding relaxation times; $M^{\pm} = M_x \pm iM_y$; $M_0^{\pm}$ and M_z^0 are the initial values of the magnetization components, and M_z^{∞} is the value of the component M_z at $t \to \infty$.

Expressions (1.1) describe the behavior of the paramagnet both in the normal state ($H_z = H$) and in the inverted state ($H = -H_z$). In the normal state, exact equilibrium of the system corresponds to value $M_0^{\pm} = 0$ and $M_z^0 = M_z^{\infty} = M$. We, however, shall assume that at $t = 0$ the equilibrium is not exact and there are certain deviations from it, albeit small: the $M_0^{\pm}$ are not equal to zero, and $M_z^0 \neq M$. The investigation should be carried out in the presence of such deviations, since it is practically impossible to produce an ideal inverted state; for example, inexact antiparallelism of the reversed and initial magnetic fields is equivalent to specifying certain initial values of the transverse magnetization components, etc.

It is seen from (1.1) that the character of the relaxation of the transverse components is the same in the normal state (if the reversal of the magnetic field was produced at the instant $t = 0$) as in the inverted state: both precess[1)] and decrease to zero with a relaxation time $\tau_\perp$. The longitudinal magnetization component in the normal state decreases or increases (depending on the sign of the deviation $\Delta M_z^0 = M_z^0 - M$ from the equilibrium state) and tend to M:

$$M_z = M + \Delta M_z^0 \exp(-\Gamma_\parallel t). \quad (1.2)$$

In the inverted state we have $M_z^{\infty} = -M$ and

$$M_z = -M + (M + M_z^0) \exp(-\Gamma_\parallel t), \quad (1.3)$$

i.e., M_z changes from $M_z^0 \approx M$ at $t = 0$ to $-M$ at $t \to \infty$.

For a ferromagnet at low temperatures, described by the Landau-Lifshitz equation, the process of homogeneous relaxation proceeds with conservation of the modulus of the magnetic moment M, and the general solution takes the simplest form in a spherical coordinate system:

$$\operatorname{tg}(\theta/2) = \operatorname{tg}(\theta_0/2) e^{-\xi x}, \quad \varphi = x + \varphi_0, \quad x = \gamma H_z t; \quad (1.4)$$

here ξ is a dimensionless relaxation parameter, and θ_0 and φ_0 are the initial azimuthal and polar angles. It is seen that in the normal state ($H_z = H$) the value of θ relaxes from θ_0 to zero, while in the inverted state ($H_z = -H$) it changes from θ_0 to $\theta = \pi$. In circular projections we have for the normalized magnetization vector $\mathbf{m} = \mathbf{M}/M$

$$m^{\pm} = \frac{m_0^{\pm} e^{\pm i x} \operatorname{sch} \xi x}{1 + m_z^0 \operatorname{th} \xi x}, \quad m_z = \frac{m_z^0 + \operatorname{th} \xi x}{1 + m_z^0 \operatorname{th} \xi x}. \quad (1.5)$$

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For comparison with the paramagnet, let us examine (1.5) during the initial stage of the relaxation process, when the deviations from the "old" equilibrium position (i.e., from $M_z = M$) are small enough:

$$M^{\pm} \approx M_0^{\pm} \exp(\mp i\gamma H_z t - \xi\gamma H_z t), \quad M_z \approx M + \Delta M_z^0 \exp(-2\xi\gamma H_z t) \qquad (1.6)$$

($\Delta M_z^0 \approx -M_0^+ M_0^- / 2M$). In the normal state, in order for these expressions to be correct it suffices to stipulate smallness of the initial deviations from the equilibrium position, since at $H_z = H$ the deviations of the system from the equilibrium position will only decrease. In the inverted state ($H_z = -H$) the deviation of the system from the old equilibrium state will grow in the course of time. In this case the requirement that the initial deviations be small is not sufficient and another restriction is imposed on the time interval Δt during which the deviations of M_i from the old equilibrium state can still be regarded as small. Taking all these restrictions into account, we can compare expressions (1.6) with expressions (1.1)–(1.3) for the paramagnet.

It is seen that for the ferromagnet the transverse relaxation time $\tau_\perp$, the longitudinal relaxation time $\tau_\parallel$, and the relaxation parameters $\Gamma_\perp$ and $\Gamma_\parallel$ corresponding to them are given by

$$\Gamma_\perp = 1/\tau_\perp = \xi\gamma H_z, \quad \Gamma_\parallel = 1/\tau_\parallel = 2\xi\gamma H_z. \qquad (1.7)$$

In the normal state these quantities are positive and the transverse components of the ferromagnet decrease to zero, as do the transverse components of the paramagnet. The longitudinal component of the ferromagnet, tending to an equilibrium value M, can only increase, for in contrast to the paramagnet the initial deviation ΔM_z^0 can only be negative; this is due to the conservation of the modulus of the magnetic moment. The last circumstance determines also the ratio of the transverse and longitudinal relaxation times: $\tau_\perp / \tau_\parallel = 2$ whereas for a paramagnet we have $\tau_\perp \ll \tau_\parallel$.

Much stronger qualitative difference between a ferromagnet and a paramagnet occur in the case of relaxation from a strong-nonequilibrium inverted state. In the inverted state $\Gamma_\perp$ and $\Gamma_\parallel$ and the times $\tau_\perp$ and $\tau_\parallel$ corresponding to them reverse sign. We recall that a negative damping parameter is a positive growth parameter and a negative relaxation time is a positive self-excitation time. Indeed, the transverse component of a ferromagnet still increase as they precess, in contrast to the transverse component of a ferromagnet. The longitudinal component of both paramagnets and ferromagnets in the inverted state decrease, but the character of this process is different: whereas for a ferromagnet (1.6) the rate of decrease of M_z is proportional to the initial deviation from the position of the old equilibrium and vanishes if there is no such deviation at the instant $t = 0$, for the paramagnet (1.3) the rate of decrease of M_z is practically independent of the deviation from the old equilibrium position. The ferromagnet type of relaxation in the inverted state, corresponding in the first stage to negative relaxation times, is shown in Fig. 1a, while the paramagnetic type of relaxation is shown in Fig. 1c (the situation corresponding to Fig. 1b will be discussed in the next section.

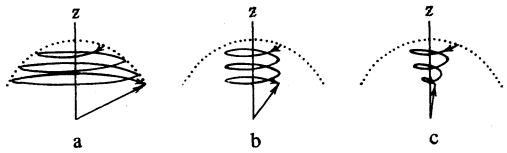

FIG. 1. Initial period of the relaxation of a spin system from an inverted state: a—ferromagnetic type of relaxation, b—relaxation of superparamagnet at the critical temperature $T = T_c$, c—paramagnetic type of relaxation.

It should be noted that we are considering here uniform motion of the magnetic moment of a ferromagnet, and do not consider the possibility of decay of the homogeneous precession into spin waves. For bulky ferromagnets the probability of such a decay in the inverted state is very large,[1] and this is one of the obstacles to the development of a ferromagnetic maser. For sufficiently small ferromagnetic particles, however, which will be investigated in the following sections, the assumption of uniform motion is acceptable.

The purpose of the present work is to investigate the behavior, in the inverted state, of a superparamagnet i.e., of a system of small ferromagnetic particles that do not interact with one another, at a nonzero absolute temperature.

In the next section we consider the initial stage of the relaxation of such a system in an approximation equivalent to the approximation (1.6) for a ferromagnet. The main results of this section were published in abbreviated form earlier.[3] In Sec. 3 the relaxation of a superparamagnet from a strong-nonequilibrium inverted state to a state of new equilibrium is investigated in the entire time interval.

2. INITIAL STAGE OF MAGNETIZATION RELAXATION

The Fokker-Planck equation for a superparamagnet whose energy density U does not contain spatially inhomogeneous terms, was written out by Brown[3]:

$$\frac{M}{\gamma}\frac{\partial W}{\partial t} = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left\{\sin\theta\left[\left(\xi\frac{\partial U}{\partial\theta} + \frac{1}{\sin\theta}\frac{\partial U}{\partial\varphi}\right)W + \xi\frac{kT}{V}\frac{\partial W}{\partial\theta}\right]\right\} + \frac{1}{\sin\theta}\frac{\partial}{\partial\varphi}\left[\left(\xi\frac{1}{\sin\theta}\frac{\partial U}{\partial\varphi} - \frac{\partial U}{\partial\theta}\right)W + \xi\frac{kT}{V}\frac{1}{\sin\theta}\frac{\partial W}{\partial\varphi}\right]; \qquad (2.1)$$

Here $W = W(\theta, \varphi, t)$ is the probability density of finding the magnetic moment in the solid angle $d\Omega = \sin\theta\, d\theta\, d\varphi$, V is the volume of the ferromagnetic particle, M is the magnetization per unit volume, $\gamma = \gamma_0/(1+\xi^2)$, γ_0 is the gyromagnetic ratio, ξ is the dimensionless relaxation parameter of the Landau-Lifshitz equation, T is the temperature, and k is the Boltzmann's constant.

It is easy to show that the infinite chain of equations for the moments, which corresponds to Eq. (2.1), is of the form

$$\frac{1}{\gamma}\frac{\partial\langle m_x^l m_y^n m_z^s\rangle}{\partial t} = s\langle H_x^e m_x^l m_y^{n+1} m_z^{s-1}\rangle - n\langle H_x^e m_x^l m_y^{n-1} m_z^{s+1}\rangle + l\langle H_y^e m_x^{l-1} m_y^n m_z^{s+1}\rangle - s\langle H_y^e m_x^{l+1} m_y^n m_z^{s-1}\rangle + n\langle H_z^e m_x^{l+1} m_y^{n-1} m_z^s\rangle - l\langle H_z^e m_x^{l-1} m_y^{n+1} m_z^s\rangle + \xi[l\langle H_x^e m_x^{l-1} m_y^n m_z^s\rangle - (l+n+s)\langle H_x^e m_x^{l+1} m_y^n m_z^s\rangle + n\langle H_y^e m_x^l m_y^{n-1} m_z^s\rangle - (l+n+s)\langle H_y^e m_x^l m_y^{n+1} m_z^s\rangle + s\langle H_z^e m_x^l m_y^n m_z^{s-1}\rangle$$

$$-(l+n+s)\langle H_z^e m_x^l m_y^n m_z^{s+1}\rangle]+\xi\frac{kT}{MV}[l(l-1)\langle m_x^{l-2}m_y^n m_z^s\rangle +n(n-1)\langle m_x^l m_y^{n-2}m_z^s\rangle+s(s-1)\langle m_x^l m_y^n m_z^{s-2}\rangle -(l+n+s)(l+n+s+1)\langle m_x^l m_y^n m_z^s\rangle], \quad (2.2)$$

where $\mathbf{H}^e=-\partial U/\partial\mathbf{M}$ is the effective magnetic field; $\mathbf{m}=\mathbf{M}/M$; l, n, and s run through all the integer values from zero to infinity.

We consider the first three equations of this chain for the case of an isotropic spherical superparamagnet, when $\mathbf{H}^e=\mathbf{H}$:

$$\begin{aligned}\gamma^{-1}\langle\dot m_x\rangle&=-\langle m_y\rangle H_z+\langle m_z\rangle H_y-\xi[(\langle m_x^2\rangle-1)H_x\\&\quad+\langle m_x m_y\rangle H_y+\langle m_x m_z\rangle H_z+\langle m_x\rangle H_T],\\ \gamma^{-1}\langle\dot m_y\rangle&=\langle m_x\rangle H_z-\langle m_z\rangle H_x-\xi[(\langle m_y^2\rangle-1)H_y\\&\quad+\langle m_x m_y\rangle H_x+\langle m_y m_z\rangle H_z+\langle m_y\rangle H_T],\\ \gamma^{-1}\langle\dot m_z\rangle&=-\langle m_x\rangle H_y+\langle m_y\rangle H_x-\xi[(\langle m_z^2\rangle-1)H_z\\&\quad+\langle m_x m_z\rangle H_x+\langle m_y m_z\rangle H_y+\langle m_z\rangle H_T],\end{aligned} \quad (2.3)$$

where $H_T=2kT/MV$ is the effective thermal field.

In this section we calculate the mean values contained in (2.3) by approximating roughly the nonequilibrium distribution function $W(t)$ by a function that deviates little from equilibrium. When momentum-inverted systems are considered there are always two equilibrium states, "old" and "new." Assume that prior to the instant $t=0$ the old equilibrium state W_0 was produced in a constant field $H_z=H$. The field is reversed instantaneously at the instant $t=0$: $H_z=-H$. The resultant strong-nonequilibrium inverted state is described exactly at the first instant by the old equilibrium function W_0, since the distribution function is not altered by an instantaneous change of the system parameters. Relaxation begins to a new equilibrium state W_∞, which is reached at $t\to\infty$. All the approximations based on small changes of the distribution function are valid here in two cases:

1) During the initial time interval Δt, so long as the deviations of the nonequilibrium distribution function $W(t)$ from the old equilibrium function W_0 remain small (in the sense of smallness of the deviations of the statistical moments).

2) In the asymptotic limit at $t\gg\tau$ (τ is the relaxation time), when the deviation of $W(t)$ from the new equilibrium function W_∞ become small.

The validity of these approximation is based on the smallness of the difference $W_0-W(t)$ or $W_\infty-W(t)$; it is immaterial here whether W_0 is the equilibrium value or not, so long as it is known exactly.

We consider here only the first of these two cases, but write out all the expressions in such a way that they apply both to the inverted state (when the magnetic field is reversed at $t=0$ from $H_z=H$ to $H_z=-H$) and to the normal state (when nothing happens at $t=0$ and the value $H_z=H$ existing at $t<0$ is preserved also at $t>0$). The latter variant, accurate to the direction of $\langle\mathbf{M}\rangle$, simulate also the asymptotic relaxation behavior corresponding to case 2.

For a superparamagnet, the old equilibrium state prevailing prior to the instant $t=0$ is described in a constant field $H_z=H$ by the Gibbs distribution function

$$W_0=\frac{\sigma\exp(\sigma m_z)}{4\pi\,\mathrm{sh}\,\sigma} \quad (2.4)$$

where $\sigma=HMV/kT$. Thus, in the equilibrium state the average values of the transverse components of the magnetization are equal to zero, and $\langle m_z\rangle_0$ is determined by the Langevin function

$$\langle m_x\rangle_0=\langle m_y\rangle_0=0,\qquad \langle m_z\rangle_0=\mathrm{cth}\,\sigma-\sigma^{-1}\equiv L(\sigma). \quad (2.5)$$

(Here and below the subscript 0 denotes averaging over the distribution function W_0.)

If we consider only small deviations of the system from the equilibrium state, then the nonequilibrium distribution function (see, e.g., [4]) can be represented in the form

$$W(t)=W_0[1+a_x m_x+a_y m_y+a_z(m_z-\langle m_z\rangle_0)], \quad (2.6)$$

where a_i are certain functions of the time. It is more convenient for us to rewrite the distribution function (2.6) in different form. To this end we calculate with its aid the expectation values of the deviation from the equilibrium state:

$$\begin{aligned}&\langle m_x\rangle=a_x\langle m_x^2\rangle_0,\quad \langle m_y\rangle=a_y\langle m_y^2\rangle_0,\\&\langle m_z\rangle-\langle m_z\rangle_0=a_z(\langle m_z^2\rangle_0-\langle m_z\rangle_0^2).\end{aligned} \quad (2.7)$$

Determining from this the a_i and substituting them in (2.6) we get

$$W(t)=W_0\left[1+\frac{\langle m_x\rangle m_x}{\langle m_x^2\rangle_0}+\frac{\langle m_y\rangle m_y}{\langle m_y^2\rangle_0}+\frac{(\langle m_z\rangle-\langle m_z\rangle_0)(m_z-\langle m_z\rangle_0)}{\langle m_z^2\rangle_0-\langle m_z\rangle_0^2}\right]. \quad (2.8)$$

This representation is formally more convenient for it enables us to obtain from (2.3) uncoupled equations not for the functions a_i, but for the expectation values $\langle m_i\rangle$ of the magnetization projections. In fact, substituting (2.8) in (3.2) and calculating the resultant equilibrium averages over the distribution function (2.4)

$$\begin{aligned}&\langle m_x^2\rangle_0=\langle m_y^2\rangle_0=L/\sigma,\quad \langle m_z^2\rangle_0=1-2L/\sigma,\\&\langle m_x^2 m_z\rangle_0=\langle m_y^2 m_z\rangle_0=(1-3L/\sigma)/\sigma,\\&\langle m_x m_y\rangle_0=\langle m_x m_z\rangle_0=\langle m_y m_z\rangle_0=\langle m_x m_y m_z\rangle_0=\langle m_x m_z^2\rangle_0=\langle m_y m_z^2\rangle_0=0,\end{aligned} \quad (2.9)$$

we obtain in final form equations that describe the weak deviation of the average magnetic-moment projections of a superparamagnet from the equilibrium state:

$$\begin{aligned}&\langle\dot m_x\rangle+\gamma H_z\langle m_y\rangle+\langle m_x\rangle/\tau_\perp=\gamma L(\eta H_x+H_y),\\&\langle\dot m_y\rangle-\gamma H_z\langle m_x\rangle+\langle m_y\rangle/\tau_\perp=\gamma L(\eta H_y-H_x),\\&\langle\dot m_z\rangle+(\langle m_z\rangle-L)/\tau_\parallel=-\xi\gamma L H_T(1-H_z/H).\end{aligned} \quad (2.10)$$

The transverse relaxation time $\tau_\perp$, the longitudinal relaxation time $\tau_\parallel$, and the corresponding relaxation parameters $\Gamma_\perp$ and $\Gamma_\parallel$ are determined by the expressions

$$\Gamma_\perp=1/\tau_\perp=\xi\gamma(aH_z+H_T),\quad \Gamma_\parallel=1/\tau_\parallel=\xi\gamma(bH_z+H_T), \quad (2.11)$$

where

$$a=\frac{1}{L}-\frac{3}{\sigma},\quad b=\frac{2}{\sigma}\frac{L^2\sigma+3L-\sigma}{\sigma-2L-L^2\sigma},\quad \eta=\xi\left(\frac{1}{L}-\frac{1}{\sigma}\right). \quad (2.12)$$

If the deviation from the equilibrium state is not connected with the change of the z projection of the magnetic field, then $H_z=H$ and the term in the right-hand side of

the third equation of the system (2.10) vanishes; in this form, this system describes, for example, the magnetic resonance of an isotropic superparamagnet in the normal (non-inverted) state.[5,6]

We consider the system (2.10) in the absence of a transverse magnetic field: $H_x = H_y = 0$; then the general solution for any constant H_z is

$$\langle m^{\pm}\rangle = \langle m_0^{\pm}\rangle \exp(\pm i\gamma H_z t - \Gamma_\perp t),$$
$$\langle m_z\rangle = L + \Delta m_z^0 \exp(-\Gamma_\parallel t) - \xi\gamma L H_T \left(1 - \frac{H_z}{H}\right) \frac{1-\exp(-\Gamma_\parallel t)}{\Gamma_\parallel} \qquad (2.13)$$

where $\Delta m_z^0 = \langle m_z^0\rangle - L$.

The main feature of these expressions is that the relaxation parameters (2.11) contained in them consist of two parts. The first, "ferromagnetic" part is proportional to the magnetic field and reverses sign in the inverted state, in analogy with the relaxation parameters of the ferromagnet (1.7). The second, "paramagnetic" part is proportional to the temperature and does not change when H changes, just as the relaxation parameters (1.1) of a paramagnet.

Let us examine in greater detail the relaxation parameters $\Gamma_\perp$ of the transverse components of the magnetization. In the normal state we have

$$\Gamma_\perp = \Gamma_2 = \xi\omega_0(1/L - 1/\sigma), \qquad (2.14)$$

where $\omega_0 = \gamma H$ is the precision frequency. The temperature dependence of Γ_2 is shown in Fig. 2 (curve q_2) and has been discussed in detail earlier.[5,6]

In the inverted state, the parameter of the transverse relaxation is determined by the expression

$$\Gamma_\perp = \Gamma_2' = -\xi\omega_0(1/L - 5/\sigma). \qquad (2.15)$$

Its temperature dependence is also shown in Fig. 2 (curve q_2'). At low temperatures Γ_2' is negative and the initial deviation of the transverse components does not attenuate, and increases instead—in analogy with (1.6). At high temperatures $\Gamma_2' > 0$ and the transverse components attenuate—in analogy with (1.1). The functions Γ_2 and Γ_2' tend to a common asymptotic limit $2\xi\omega_0/\sigma$ as $T \to \infty$.

Particular interest attaches to the critical temperature T_c at which Γ_2' reverses sign, and the relaxation time τ_2' becomes infinite:

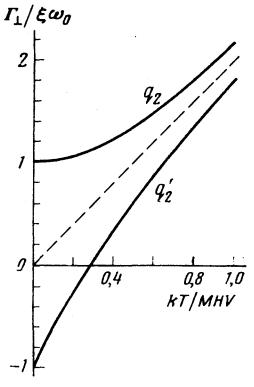

FIG. 2. Temperature dependence of the normalized relaxation parameter of the transverse components of the magnetization in the normal ($q_2 = \Gamma_2/\xi\omega_0$) and inverted ($q_2' = \Gamma_2'/\xi\omega_0$) states.

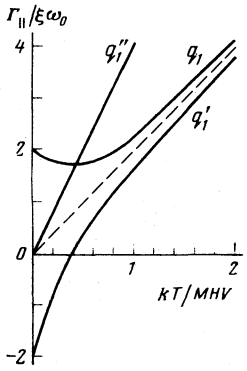

FIG. 3. Temperature dependence of the normalized relaxation parameters of the longitudinal magnetization component in the normal ($q_1 = \Gamma_1/\xi\omega_0$) and inverted ($q_1' = \Gamma_1'/\xi\omega_0$, $q_1'' = \Gamma_1''/\xi\omega_0$) states.

$$T_c = 0.28MHV/k. \qquad (2.16)$$

We consider now the behavior of the longitudinal magnetization component m_z. In the normal state ($H_z = H$) the third term in the right-hand side of the corresponding expression (2.13) vanishes, and it takes a form analogous to (1.2) and (1.6):

$$\langle m_z\rangle = L + \Delta m_z^0 e^{-\Gamma_1 t}. \qquad (2.17)$$

It should be noted (this pertains to both the normal and the inverted state) that the Δm_z^0 formed at the initial state cannot be arbitrary. In fact, besides the natural requirement that it be small, Δm_z^0 is subject to one more condition

$$\Delta m_z^0 \leqslant (1 - L^2 - \langle m_0^+\rangle\langle m_0^-\rangle)/2L, \qquad (2.18)$$

which is a consequence of the inequality $\sum_i \langle m_i\rangle^2 \leqslant 1$, valid for any instant of time and at any temperature. It is seen that at high temperatures, when L is much lower than unity, both positive and negative Δm_z^0 can be formed; as $T \to 0$, however, when $L \to 1$, m_z^0 can only be negative.

The longitudinal relaxation parameter Γ_1 is defined by

$$\Gamma_1 = \frac{1}{\tau_1} = \frac{2\xi\omega_0 L}{\sigma(\sigma - 2L - \sigma L^2)}. \qquad (2.19)$$

Its temperature dependence is shown in Fig. 3 (curve q_1). At $T = 0$ the quantity $\Gamma_1 = 2\Gamma_2$ first decreases with increasing temperature, and then increases and tends to the same asymptotic value as Γ_2.

In the inverted state, an important role is assumed in the relaxation of the m_z component by the term connected with the thermal force. Since our analysis is valid in this case only for sufficiently short times, we expand the exponentials in series; then $\Gamma_\parallel$ cancels out in the term connected with the thermal force and we obtain the expression

$$\langle m_z\rangle = L(1 - \Gamma_1'' t) + \Delta m_z^0(1 - \Gamma_1' t) \qquad (2.20)$$

with two longitudinal relaxation times, τ_1' and τ_1'':

$$\Gamma_1' = \frac{1}{\tau_1'} = \frac{2\xi\omega_0}{\sigma}\,\frac{2\sigma - 5L - 2\sigma L^2}{\sigma - 2L - \sigma L^2},$$
$$\Gamma_1'' = \frac{1}{\tau_1''} = \frac{4\xi\omega_0}{\sigma}. \qquad (2.21)$$

We see that (2.20) is valid for times satisfying the relations

$$\Gamma_1''t\ll 1, \quad \Delta m_z^0\Gamma_1't/L\ll 1. \tag{2.22}$$

The first term in (2.20) is similar to Eq. (1.3) for an inverted paramagnet (provided that $\Gamma_{\parallel}t\ll 1$); the second term, as well as the time-dependent term in the component m_z of an inverted ferromagnet (1.6), is proportional to Δm_z^0. The temperature dependence of the parameters (2.21) is shown in Fig. 3. The parameter Γ_1', which is the "inverted" parameter Γ_1, reverses sign at a certain critical temperature $T_c'\approx 0.38\,MHV/k$; with increasing temperature it tends to the same asymptotic limit as Γ_1. The parameter Γ_1'' is equal to zero at $T=0$ and increases linearly with increasing T; it is this parameter which plays the principal role in the relaxation of the component m_z at $T\neq 0$.

Thus, the analysis performed in this section allows us to draw the following conclusion concerning the character of the initial relaxation period of an inverted system of noninteracting quasiclassical spins: At $T=0$ the system is described by the Landau-Lifshitz equation and a ferromagnetic type of relaxation is realized (Fig. 1a): the transverse components increase, the longitudinal ones decreases, and the modulus of the magnetic moment is conserved. At $T\neq 0$, owing to thermal fluctuations, dephasing of the transverse components takes place and the modulus of $\langle \mathbf{m}\rangle$ is no longer conserved, but one of the attributes of the ferromagnetic type of relaxation—the increase of the transverse components in the initial period—is preserved up to the critical temperature T_c. At this temperature the initial deviation of the transverse components during the initial stage of the relaxation process remains unchanged (Fig. 1b): the rate of rotation of the magnetic moment to the new equilibrium position (ferromagnetic type of relaxation) is balanced out by the rate of dephasing of the transverse components (paramagnetic type of relaxation) At $T>T_c$ the paramagnetic type of relaxation prevails (Fig. 1c), and the system is described by the Bloch equation: the initial deviation of the transverse components is damped.

The critical temperature T_c at which a change in the type of relaxation takes place for particles of 120 Å diameter with a magnetization $M=10^3$ G in an external field $H=200$ Oe lies in the range of room temperatures; the critical temperature can be easily regulated by varying H.

3. APPROXIMATION OF THE MAGNETIZATION RELAXATION PROCESS IN THE ENTIRE TIME INTERVAL

We consider the system (2.2) at $H_x=H_y=0$. The values $l=n=0$ correspond to an infinite chain of equations, which contains only different moments of the projection m_z:

$$\frac{1}{\gamma}\frac{d\langle m_z^s\rangle}{dt}=\xi sH_z(\langle m_z^{s-1}\rangle-\langle m_z^{s+1}\rangle)$$
$$+\xi\frac{kT}{MV}[s(s-1)\langle m_z^{s-2}\rangle-s(s+1)\langle m_z^s\rangle]. \tag{3.1}$$

The values $l=1$, $n=0$ and $l=0$, $n=1$ correspond to two chains for $\langle m_x m_z^s\rangle$ and $\langle m_y m_z^s\rangle$, which take in terms of circular variables the form

$$\frac{1}{\gamma}\frac{d\langle m^{\pm}m_z^s\rangle}{dt}=\pm iH_z\langle m^{\pm}m_z^s\rangle+\xi H_z[s\langle m^{\pm}m_z^{s-1}\rangle-(s+1)\langle m^{\pm}m_z^{s+1}\rangle]$$
$$+\xi\frac{kT}{MV}[s(s-1)\langle m^{\pm}m_z^{s-2}\rangle-(s+1)(s+2)\langle m^{\pm}m_z^s\rangle]. \tag{3.2}$$

This system is simplified by making of change of variables such as to exclude the precession motion

$$m^{\pm}=m(t)\exp(\pm i\gamma H_z t\pm i\varphi_0), \tag{3.3}$$

where $m(t)$ is the amplitude of the transverse magnetization components. As a result, (3.2) takes the form

$$\frac{1}{\gamma}\frac{d\langle mm_z^s\rangle}{dt}=\xi H_z[s\langle mm_z^{s-1}\rangle-(s+1)\langle mm_z^{s+1}\rangle]$$
$$+\xi\frac{kT}{MV}[s(s-1)\langle mm_z^{s-2}\rangle-(s+1)(s+2)\langle mm_z^s\rangle]. \tag{3.4}$$

We consider first the chain of equations (3.1). The first equation of this chain is

$$d\langle m_z\rangle/dt=-\xi\gamma[H_z(\langle m_z^2\rangle-1)+H_T\langle m_z\rangle]. \tag{3.5}$$

If we make the simple uncoupling $\langle m_z^2\rangle\approx\langle m_z\rangle^2$, then this equation is easily integrated and we have

$$\langle m_z\rangle=\alpha_0\frac{\langle m_z^0\rangle+\sigma_z^{-1}+\alpha_0\,\mathrm{th}(\alpha_0\xi x)}{\alpha_0+(\langle m_z^0\rangle+\sigma_z^{-1})\,\mathrm{th}(\alpha_0\xi x)}-\sigma_z^{-1}, \tag{3.6}$$

where

$$\alpha_0=(1+\sigma^{-2})^{1/2}, \quad \sigma_z=MVH_z/kT, \quad x=\gamma H_z t.$$

It follows therefore that in the normal state as $t\to\infty$ the quantity $\langle m_z\rangle$ tends to the expression

$$L_0=(1+\sigma^{-2})^{1/2}+\sigma^{-1}. \tag{3.7}$$

A plot of this function is shown in Fig. 4. Actually $\langle m_z\rangle$ should tend as $t\to\infty$ to the Langevin function L, which is also shown in Fig. 4. From a comparison of these curves it is seen that although the approximating curve L qualitatively accounts for the course of the curve L, the quantitative deviations are nevertheless appreciable. We therefore construct a solution for $\langle m_z\rangle$ with account taken of the next equation of the chain (3.1), which is of the form

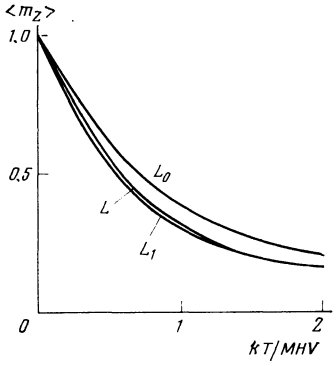

FIG. 4. Temperature dependence of the Langevin function and of its approximating functions L_0 and L_1.

$$\frac{1}{\gamma}\frac{d\langle m_z^2\rangle}{dt}=2\xi H_z(\langle m_z\rangle-\langle m_z^3\rangle)+\xi H_T(1-3\langle m_z^2\rangle). \qquad (3.8)$$

Effecting in this equation the uncoupling $\langle m_z^3\rangle \approx \langle m_z\rangle\langle m_z^2\rangle$, and differentiating (3.5) and combining it with (3.8), we obtain for m_z a nonlinear second-order differential equation

$$\frac{d^2\langle m_z\rangle}{dt^2}+2\xi\gamma[H_z\langle m_z\rangle+2H_T]\frac{d\langle m_z\rangle}{dt}$$
$$+2\xi^2\gamma^2H_T[H_z(\langle m_z\rangle^2-1)+{}^3/_2H_T\langle m_z\rangle]=0. \qquad (3.9)$$

We have obtained the exact solution of this equation

$$\langle m_z\rangle=\alpha\frac{\langle m_z^0\rangle+3/2\sigma_z+\alpha\,\mathrm{th}(\alpha\xi x)}{\alpha+(\langle m_z^0\rangle+3/2\sigma_z)\,\mathrm{th}(\alpha\xi x)}-\frac{3}{2\sigma_z}, \qquad (3.10)$$

where $\alpha=[1+(3/2\sigma)^2]^{1/2}$.

In the normal equilibrium state, the value of $\langle m_z\rangle$ corresponding to (3.10) is

$$L_1=[1+(3/2\sigma)^2]^{1/2}-3/2\sigma. \qquad (3.11)$$

A plot of this expression is also shown in Fig. 4. We see that this curve hardly differs from the Langevin function. On the other hand, at $T=0$ Eq. (3.10) goes over into (1.5). Thus, in the two known limiting cases (at $T=0$ for arbitrary time and as $t\to\infty$ for arbitrary temperature) expression (3.10) is a good approximation.

We now examine the chain of equations (3.4). The first equation of this chain is

$$d\langle m\rangle/dt=-\xi\gamma(H_z\langle mm_z\rangle+H_T\langle m\rangle). \qquad (3.12)$$

Putting in this equation $\langle mm_z\rangle \approx \langle m\rangle\langle m_z\rangle$, substituting $\langle m_z\rangle$ from (3.10), integrating, and substituting the result in (3.3), we obtain ultimately

$$\langle m^{\pm}\rangle=\frac{\langle m_0^{\pm}\rangle\alpha\,\mathrm{sch}(\alpha\xi x)}{\alpha+(\langle m_z^0\rangle+3/2\sigma)\,\mathrm{th}(\alpha\xi x)}\exp\left(\pm ix-\xi\frac{\gamma kT}{MV}t\right). \qquad (3.13)$$

At $T=0$ this expression goes over into (1.5).

For the case of small deviations from the old equilibrium position we obtain from (3.10) and (3.13) expressions close to the results of the preceding section. Thus, for the $m^{\pm}$ components we obtain the relaxation parameter in the form

$$\Gamma_\perp=\xi\gamma(L_1H_z+H_T), \qquad (3.14)$$

which approximates well the corresponding expression in (2.11). For m_z in the inverted state we obtain expression (2.20) with two relaxation parameters:

$$\Gamma_1'=\frac{2\xi\omega_0}{\sigma}(1-\sigma L_1),\quad \Gamma_1''=\frac{4\xi\omega_0}{\sigma}, \qquad (3.15)$$

which approximate expressions (2.21) quite well.

To verify the correctness of the analytic expressions (3.10) and (3.13) in the entire range of times and temperatures, the chains of differential equations (3.1) and (3.4) were integrated numerically for the inverted state up to $s=30$. A comparison of the results of the numerical calculation (Figs. 5 and 6, dashed curves) with the analytic expressions (3.10) and (3.13) (the solid curves in the same figures) shows that the latter are a good approximation of the real situations. Figures 5 and 6 show the behavior of the components of the magnetization in time for relaxation from the inverted state to a new equilibrium state at various temperatures. The value of the normalized temperature σ^{-1} is marked on each curve.

It is seen from Fig. 5 that with increasing temperature the relaxation time of the $\langle m_z\rangle$ magnetization component decreases. In addition, a change takes place in the character of the relaxation; at low temperatures ($\sigma^{-1}=0.01$) the value of $\langle m_z\rangle$ decreases slowly at the first instant, and this corresponds to the ferromagnetic type of relaxation (to rotation of the almost-conserved magnetic moment); at high temperatures ($\sigma^{-1}=0.3$), conversely, at the first instant of time the rate of relaxation of $\langle m_z\rangle$ is maximal, just as in a paramagnet.

An even greater change in the temperature dependence of the relaxation is observed for the transverse components (Fig. 6). At low temperatures the transverse components first increase sharply, and then decrease,

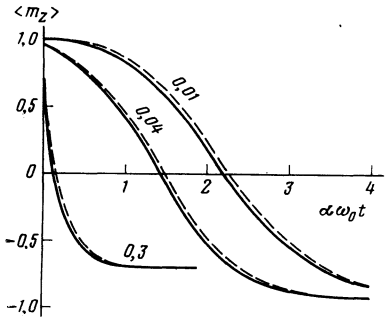

FIG. 5. Dependence of $\langle m_z\rangle$ on the time at different temperatures; the values of the normalized temperature kT/MHV are indicated on each curve. The solid curves correspond to the analytic expression (3.10), and the dashed ones were calculated with a computer.

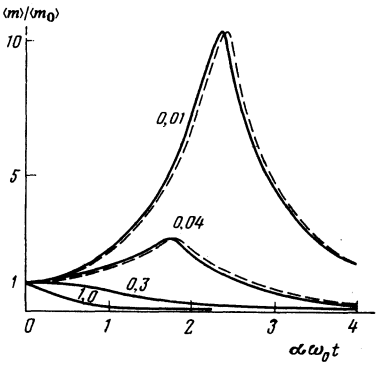

FIG. 6. Dependence of the relative amplitude of the transverse components $\langle m^{\pm}\rangle$ on the time at different temperatures; the values of the normalized temperature kT/MHV are indicated on each curve. The solid curves correspond to the analytic expression (3.13), and the dashed ones were calculated with a computer.

because the main process is ferromagnetic rotation of the magnetic moment. With increasing temperature, the maximum decreases and shifts to the left. At a temperature corresponding to the critical value ($\sigma_c^{-1} \approx 0.3$), the type of relaxation changes: the growth of the transverse components as a result of the rotation of the magnetic moment is offset in a certain time interval by their decrease as a result of the phase randomization; the character of the relaxation corresponds in this case to Fig. 1b. At temperatures higher than T_c the relaxation of the transverse components has a paramagnetic character—the decisive mechanism is not the phase randomization.

[1)]Naturally, the precession direction is reversed when the sign of H_z is reversed.

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Optical and electro-optical properties of confocal cholesteric textures

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Textures with a period greatly exceeding the equilibrium pitch P_0 of the helix were observed in a cholesteric liquid crystal layer with homotropic boundary conditions at thicknesses on the order of P_0. It is shown that the known methods of measuring the pitch on a confocal texture can be used only at thicknesses that exceed the equilibrium pitch by an order of magnitude. The appearance of peaks on the voltage-contrast and transition characteristics of the transition from the cholesteric to the nematic liquid crystal is attributed to changes in the electric field of the diffraction-reflection intensities.

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A cholesteric liquid crystal (CLC) placed in a sandwich cell between glasses prepared for planar orientation, forms the well known Granjean texture. Such "planar" boundary conditions do not disturb the helical structure of the CLC (they are compatible with it), and their influence reduces to a change in the pitch of the helix, and only in thin cells (of thickness on the order of the pitch) does the helix become completely unbound.[1] At the same time, confocal textures formed by a CLC in a cell with glasses prepared for homotropic orientation of the molecules, are quite complicated (since these boundary conditions are not compatible with the helical character of the CLC) and have not been investigated to any extent. The need for a more detailed investigation of these textures at arbitrary layer thicknesses arises, in particular, when attempts are made to explain the observed difference[2,3] between the equilibrium helix pitch, determined microscopically and from the angle of diffraction of the laser beam by the confocal structure,[4,5] on the one hand, and the pitch measured by the wedge method, on the other.[6]

The untwisting of the cholesteric helix in an electric field (the CLC → NLC transition, where NLC stands for nematic liquid crystal) in sandwich cells also goes through a stage of formation of confocal textures,[7,8] and in this case this is satisfactory explanation of the complicated behavior of the voltage-contrast curves of the CLC → NLC transition[9] or for the reasons for the appearance of peaks on the oscillograms of the relaxation photoresponse of the untwisted helix.[10,11]

The task of the present paper is to investigate in detail the optical properties of various types of confocal textures of CLC and their variations in an electric field, for the purpose of obtaining a correct approach to a procedure for measuring the pitch of a helix on a confocal structure and the investigation of the voltage-contrast curves of CLC → NLC effect.

EXPERIMENTAL PROCEDURE

The textures produced in a CLC layer under homotropic boundary conditions were investigated in wedge-shaped sandwich cells, which are convenient for observation of the variation of the optical properties of the layer with increasing thickness. The wedge taper was set by means of Teflon liners of various thicknesses,

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