# Static susceptibility of thin films of quasi-uniaxial magnets at phase transitions accompanied by domain-structure formation

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A theoretical and experimental study is made of the behavior of the static susceptibility of thin films of quasi-uniaxial magnets near lines of spontaneous (near the Curie point) and orientational phase transitions. Allowance is made for the contributions to the susceptibility from the homogeneous component of the magnetization, the domain structure, and fluctuations. It is shown that the domain structure plays an important role in shaping the response of magnets of finite size to an external magnetic field. It is established that the behavior of the susceptibility in the homogeneous state is not described by simple power laws with definite critical exponents.

The study of anomalies of the magnetic susceptibility at spontaneous (near the Curie or Néel point) and orientational phase transitions yields valuable information on the critical behavior of magnets. Different aspects of this problem have attracted continuing interest among theorists and experimenters; a detailed bibliography can be found in the many reviews and monographs (see, e.g., Refs. 1–9). In magnets of finite size, phase transitions involving a lowering of the symmetry are often accompanied by the formation of inhomogeneous magnetic states, i.e., domain structures,[10,12] which must be taken into account both in the construction of a realistic theory and in the interpretation of experimental results. The various types of domain structures arising at spontaneous and orientational phase transitions in thin films of quasi-uniaxial magnets and the transformations between different domain structures were analyzed in Refs. 12–15. It can be stated *a priori* that the existence of domain structure will affect the critical behavior of the various thermodynamic quantities, but such questions have even to this day remained essentially outside the purview of investigators.

In this present paper we make a theoretical and experimental study of the static susceptibility of thin films of quasi-uniaxial magnets at spontaneous (near the Curie point) and orientational phase transitions. It is shown that the domain structure plays an important role in shaping the response of these objects to changes in a homogeneous external magnetic field. The questions discussed in this paper have a direct bearing on the correct calculation and experimental determination of the critical exponents of the thermodynamic quantities.

## 1. EXPERIMENT

The experiments were done on quasi-uniaxial films of mixed iron garnets of various compositions which were grown by liquid-phase epitaxy on gadolinium-gallium garnet substrates having various crystallographic orientations (the essential parameters of the investigated films are given later on in the text). The term "quasi-uniaxial" means that the uniaxial component (described by the uniaxial anisotropy constant $K_u$) is dominant in the magnetic anisotropy energy, but that there are also weak cubic and rhombic components (described by the constants $K_c \ll K_u$ and $K_p \ll K_u$, respectively). The axis of easy magnetization in real films generally deviates from the normal to the surface $\mathbf{n}\|\mathbf{e}_z$ by a small angle $\varphi_u \ll 1$. The saturation magnetization $M_O$ of the films is much less than the uniaxial anisotropy field $H_u = 2K_u M_0^{-1}$, so that in the absence of an external magnetic field $\mathbf{H}$ the vector $\mathbf{M}$ in the central part of the domains is approximately perpendicular to the surface. An explicit expression for the anisotropy energy is given in Ref. 15; we will adhere to the notation adopted in that paper. In the theoretical part of the present paper we will often use the normalized anisotropy constants $\beta_i = K_i(2\pi M_O^2)^{-1}$, $i = u, p, c$.

For isothermal ($T = 290$ K) orientational phase transitions induced by a static magnetic field $\mathbf{H}_\perp = \mathbf{H}_y = H_y \mathbf{e}_y$, we investigated the dependence of the two components $\chi_{zz}$ and $\chi_{yz}$ of th static magnetic susceptibility tensor on the field $H_\perp$ (for $\mathbf{H}_\perp = \mathbf{H}_z = H_z \mathbf{e}_z = \text{const}$) or on the field $H_\parallel$ (for $H_\perp = \text{const}$), and for the spontaneous phase transitions near the Curie point we studied $\chi_{zz}(T)$ for $H = \text{const}$.

### 1.1. Experimental techniques

The measurements were made by a magnetooptical method for both the orientational and spontaneous phase transitions. In the first case the film under study was mounted on a turntable between the pole pieces of an electromagnet producing a uniform static magnetic field $\mathbf{H}_\perp \| \mathbf{e}_y$ of up to 5 kOe parallel to the surface of the film. The turntable permitted rotation of the film about the normal $\mathbf{n}$ to its surface by an arbitrary angle $\varphi_H$. A field $\mathbf{H}_\| \| \mathbf{n}$ of up to 500 Oe was produced by a pair of coreless coils placed on the two sides of the films. Plane-polarized light with $\mathbf{k}\|\mathbf{n}$ from a helium-neon laser with a working wavelength of 0.6328 $\mu$m was focused onto the surface of the film by a long-focus lens. The diameter of the illuminated region of the film ($\approx 1$ mm$^2$) was much larger than the period of the domain structure. The light transmitted through the film was collected by a wide-aperture lens and directed onto a Wollaston prism, which split the light beam into two beams of mutually orthogonal

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polarization, which proceeded to fall on the sensitive areas of two identical photodiodes. The signal from the photodiodes was sent to a *dc* differential amplifier. The principal axes of the Wollaston prism were oriented at a 45° angle to the plane of polarization of the laser light incident on the film; this arrangement provided an effective suppression of the laser noise, and the signal at the output of the differential amplifier was to good accuracy proportional to the *z* component of the magnetization averaged over the area of the illuminated region of the film. With this layout we could take the curves of $M_z(H_\parallel)$ for $H_\perp = \text{const}$ and $M_z(H_\perp)$ for $H_\parallel = \text{const}$.

For studying the field dependence of the susceptibilities $\chi_{zz} = \partial M_z/\partial H_\parallel$ and $\chi_{yz} = \partial M_z/\partial H_\perp$, provisions were made for the use of modulating coils capable of producing alternating magnetic fields $\tilde{h}_\parallel$ and $\tilde{h}_\perp$ with strengths of the order of a few oerstads at a frequency $\sim 1$ kHz. The field $\tilde{h}_\parallel$ was produced by a single-layer flat coil, with from 5 to 10 turns and an inside diameter of $\approx 1$ mm, placed directly on the surface of the film. The field $\tilde{h}_\perp$ was produced by a pair of miniature coils wound directly on the film; the coil for producing $\tilde{h}_\parallel$ was placed in the gap between these coils. For measuring the susceptibility, the signal from the photodiodes was sent to a balanced amplifier, then to a narrow-band amplifier tuned to the modulation frequency, and finally to a lock-in detection circuit. The field dependence of $\chi_{zz}$ and $\chi_{yz}$ could also be determined by direct differentiation of the $M_z(H_\parallel)$ and $M_z(H_\perp)$ curves. Although this procedure is more difficult and gives a low absolute accuracy, in a number of cases it yields more-useful information than does the modulation technique (see below).

An analogous procedure was used for the spontaneous phase transitions in the neighborhood of the Curie point $T_C$. In this case the film under study was placed on a heated stand having a cylindrical flange on top of which a cover glass was placed. Static magnetic fields $H_\parallel$ and $H_\perp$ of up to 300 Oe were produced by Helmholtz coils; the arrangement for producing the modulating field $\tilde{h}_\parallel$ was analogous to that used for the orientational phase transitions. The temperature was measured by a copper-constantan thermocouple with one junction in a dewar of liquid nitrogen and the other cemented to the film right next to the probed region.

The temperature and field curves of $M_z$ and $\hat{\chi}$ were recorded on an *XY* recorder.

### 1.2. Orientational phase transitions

Figure 1a shows the $M_z(H_\parallel)$ curves at $T = 293$ K for various values of $H$ in film No. 1 [composition $(YGdYbBi)_3(FeAl)_5O_{12}$; substrate orientation (100); thickness $L = 8.0$ $\mu$m; saturation magnetization $M_O = 10.93$ G; angle of deviation of the easy axis from the normal $\varphi_u = 10°$; uniaxial, cubic, and rhombic anisotropy constants $K_u = 2905$, $K_c = 1080$, and $K_p = 128$ erg/cm$^3$]. Because of the complexity of the anisotropy of the film, a field $H_\perp$ induces an appreciable component $M_z$ even in the absence of a field $H_\parallel$. The slight hysteresis on separate regions of the curve is due to the fact that the nucleation and annihilation of domains occurs as a first order phase transition. In a small neighborhood of the apex ($H_{\perp C} = 710$ Oe, $H_{\parallel C} = 50$ Oe) of the curve on which the homogeneous state loses stability, i.e., the "critical parabola" $H^{(U\to St)}(H_\parallel)$ (see Ref. 15), this phase transition becomes second order, and the hysteresis vanishes. Hysteresis effects are also absent, of course, when $H_\perp > H_{\perp C}$. Inside the critical parabola the change in $M_z$ occurs mainly through the motion of domain walls, outside this parabola it occurs mainly by a change in the orientation of the vector **M**. The position of the inflection point on the $M_z(H_\parallel)$ curve is determined by the value of $H_\perp$; for $H_\perp = 0$ the inflection is observed at $H_\parallel = 0$, while for $H_\perp = H_{\perp C}$ it occurs at $H_\parallel = H_{\parallel C}$ (the values of $H_{\perp C}$ and $H_{\parallel C}$, in turn, depend on the azimuthal orientation of the film with respect to the field $H_\perp$, i.e., on the angle $\varphi_H$).[15]

If we ignore fluctuation effects, then on the stability-loss line of the homogeneous state (the SLHS line) for ideal films there would be either a divergence of the susceptibility and a jump in $M_z$ (a first order phase transition) or else a jump in the susceptibility and a kink on the $M_z(H_\parallel)$ or $M_z(H_\perp)$ curves (a second order phase transition). In real films (even films of very good quality), which unavoidably contain microscopic defects and have slightly inhomogeneous properties, these features get smoothed out, since the nucleation or annihilation of domains occurs over the course of some finite change in the magnetic field. This circumstance has a particularly strong effect on a first order phase transition. For this reason the procedure for determining the position of the SLHS line becomes somewhat tentative, but the uncertainty can be avoided by making a reasonable ap-

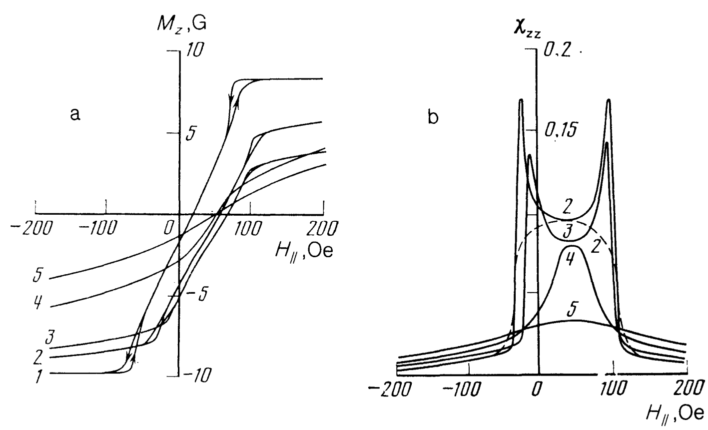


FIG. 1. Curves of $M_z$ (a) and $\chi_{zz}$ (b) versus the field $H_\parallel$ for film No. 1 at $T = 293$ K. On curves 1–5: $H_\perp = 50$, 400, 500, 700, and 900 Oe.

proximation to the curves. We checked that the position of the critical parabola had been determined correctly by making visual observations of the nucleation of the domain structure (see Ref. 15).

The $\chi_{zz}(H_{\parallel})$ curves obtained by differentiating the $M_z(H_{\parallel})$ curves for film No. 1 in the case of a decreasing field $H_{\parallel}$ are represented by the solid curves in Fig. 1b. The overshoots on curves 2 and 3 correspond to the nucleation of a domain structure by way of a first order transition. When $\chi_{zz}$ is measured by the modulation technique, the overshoots are less pronounced. At a sufficiently large amplitude of the modulating field $\tilde{h}_{\parallel}$ the overshoots vanish altogether and the $\chi_{zz}(H_{\parallel})$ curves exhibit only the jumps characteristic of a second order phase transition (the dashed curve, for $H_{\perp} = 500$ Oe). The reason for this is that for a strong but slow modulation (with an amplitude of the modulating field comparable to the width of the hysteresis loop and a period considerably longer than the characteristic relaxation times) the film is found in a single-domain state over a certain part of the modulation period and in a state with a domain structure over the remainder of the period. As a result, the jumpy parts of the $M_z(H_{\parallel})$ curves become smoother, causing the overshoots to vanish from the magnetic-field curve of the susceptibility.

Figure 2 shows curves of $\chi_{zz}(H_{\perp})$ at $T = 293$ K for various values of $H_{\parallel}$ (the curves were taken on decreasing $H_{\perp}$) for film No. 2 [composition $(YEu)_3(FeGa)_5O_{12}$, substrate orientation (111), $L = 6.1$ $\mu$m, $\varphi_u = 1.53°$, $K_u = 4346.2$ erg/cm$^3$, $K_p = 292.5$ erg/cm$^3$, $K_c = 689.1$ erg/cm$^3$]. The apex of the critical parabola for the chosen azimuthal position of film No. 2 has the coordinates $H_{\parallel C} = -10.92$ Oe, $H_{\perp C} = 968$ Oe. For a second order phase transition ($|H_{\parallel} - H_{\parallel C}| \lesssim 10$ Oe) the susceptibility increases smoothly as the domain structure is being nucleated (curve 1); on the lines of the first order phase transitions the change in $\chi_{zz}$ is discontinuous[1)] (curves 2–4). Beginning

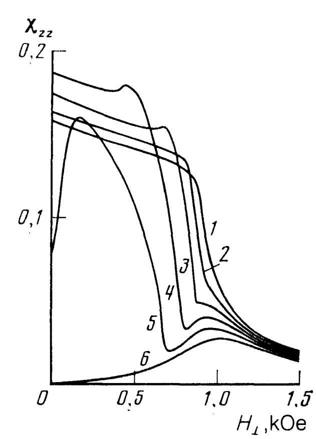


FIG. 2. Curves of $\chi_{zz}(H_{\perp})$ for film No. 2 at $T = 293$ K. On curves 1–6: $H_{\parallel} = -11$, 11, 33, 44, 55, and 77 Oe.

with values $|H_{\parallel} - H_{\parallel C}| \gtrsim 20$ Oe a "high-field" peak appears on the susceptibility $\chi_{zz}$ in the uniformly magnetized state, and with increasing $|H_{\parallel} - H_{\parallel C}|$ it shifts toward higher values of $H_{\perp}$ and decreases rapidly in amplitude (curves 2–6). At large values of $|H_{\parallel} - H_{\parallel C}|$, when the straight line $H_{\parallel} = \text{const}$ on the $H_{\parallel}H_{\perp}$ plane no longer intersects the SLHS line, the $\chi_{zz}(H_{\perp})$ curves exhibits only the high-field peak (curve 6). Figure 2 shows only the curves for positive values of $H_{\parallel} - H_{\parallel C}$; the situation is similar for negative values of $H_{\parallel} - H_{\parallel C}$.

The curves of $M_z(H_{\perp})$ for $H_{\parallel} = \text{const}$ for film No. 1 are shown in Fig. 3a. The curves exhibit a kink where the domain structure is nucleated (curves 2–4 and 6–10). The kink vanishes at $H_{\parallel} = H_{\parallel C} = 50$ Oe (curve 5) and also for large values of $|H_{\parallel} - H_{\parallel C}|$, when the straight lines $H_{\parallel} = \text{const}$ do not intersect the SLHS line (curves 1 and 11). In the latter case the $\chi_{yz}(H_{\perp})$ curves shown in Fig. 3b

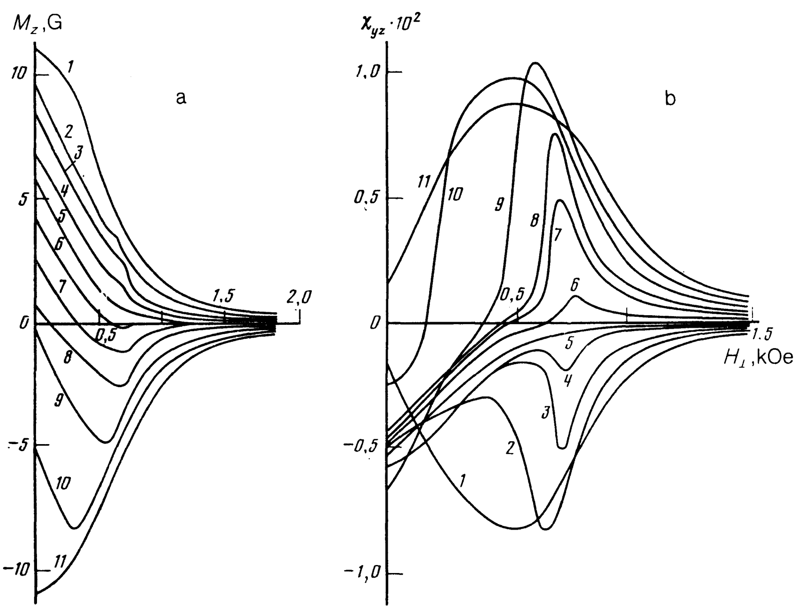


FIG. 3. Curves of $M_z$ (a) and $\chi_{yz}$ (b) versus the field $H_{\perp}$ for film No. 1 at $T = 293$ K. On curves 1–11: $H_{\parallel} = 138$, 92, 69, 58, 50, 46, 34.5, 28, 0, $-46$, and $-69$ Oe.

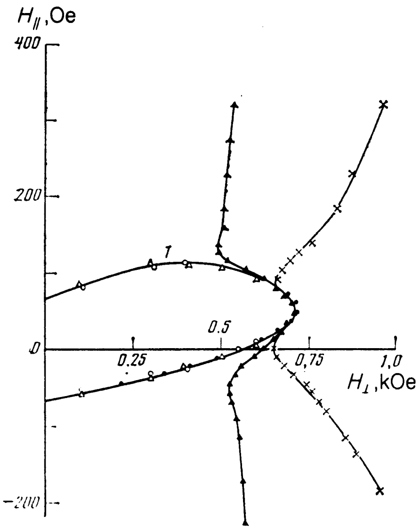


FIG. 4. Line on which the homogeneous state loses stability (the SLHS line; curve 1) and the position of the peaks of $\chi_{zz}(H)$ (×) and $\chi_{yz}(H_{\perp})$ (▲) on on the $H_{\parallel}H_{\perp}$ plane for film No. 1 at $T = 293$ K. The position of the SLHS line was determined from the curves of $M_z(H_{\perp})$ (●), $M_z(H_{\parallel})$ (Δ), and $\chi_{zz}(H_{\parallel})$ (○).

have a broad peak which decreases in amplitude and shifts to higher fields $H_{\perp}$ as $|H_{\parallel} - H_{\parallel C}|$ increases (curves 1 and 11). In the region of small $|H_{\parallel} - H_{\parallel C}|$ the peak on the $\chi_{yx}(H_{\perp})$ curves shifts to lower values of $H_{\perp}$ with increasing $|H_{\parallel} - H_{\parallel C}|$; this peak is due to the nucleation of a domain structure (curves 2–4, 6–10). Curve 5, for $H_{\parallel} = H_{\parallel C}$, does not exhibit any features.

A graphic representation of the position of the lines of maximum $\chi_{zz}$ and $\chi_{yz}$ on the $H_{\parallel}H_{\perp}$ plane for film No. 1 is given in Fig. 4, which also shows the SLHS line as determined from the curves $M_z(H_{\perp})$, $M_z(H_{\parallel})$, and $\chi_{yz}(H_{\perp})$. Because of the smearing of the first order phase transition in real films, the curves for the $\chi_{yz}$ peaks in the existence region of the domain structure in Fig. 4 do not coincide with the

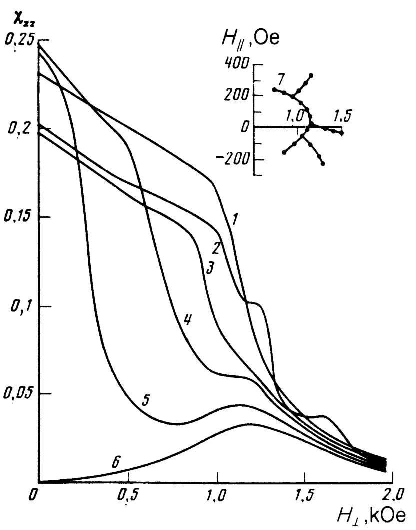


FIG. 5. Curves of $\chi_{zz}(H_{\perp})$ for film No. 3 at $T = 293$ K. The inset shows the SLHS line (7) and the position of the peaks at $\chi_{zz}(H_{\perp})$ on the $H_{\parallel}H_{\perp}$ plane. On curves 1–6: $H_{\parallel} = 58, 0, -46, -92, -138$, and $-207$ Oe.

SLHS line.[2)] In the region of small $|H_{\parallel} - H_{\parallel C}|$ the difference in the positions of the lines becomes negligibly small, since the nucleation of the domain structure occurs by way of a second order phase transition.

For several films with the (110) orientation, e.g., for film No. 3, which had a strong anisotropy in the basal plane ($K_p/K_u \approx 0.3$), an additional (third) high-field peak on the $\chi_{zz}(H_{\perp})$ curves was observed in a certain angular region $\varphi_H \approx 10°$ (see Fig. 5). The curves giving the position of all the observed $\chi_{zz}(T)$ peaks on the $H_{\parallel}H_{\perp}$ plane are given in the inset in Fig. 5; also shown is the SLHS line. The origin of the additional high-field peak of $\chi_{zz}$ remains unclear at present, because the theory (which, to be sure, was developed under the assumption $K_p \ll K_u$) predicts the presence of only two extrema (see below). However, in the presence of strong anisotropy in the basal plane a change in $H_{\perp}$ can lead to an appreciable azimuthal rotation of the vector **M**, and this can affect the behavior of the susceptibility. It is also conceivable that the film might be layered.[16]

In films with a slight anisotropy in the basal plane [usually with the (111) and (100) orientations), the lines of the $\chi_{zz}$ and $\chi_{yz}$ peaks on the $H_{\parallel}H_{\perp}$ plane (the "whiskers") are symmetrically arranged with respect to the critical parabola; for films with the (110) orientation there is a pronounced asymmetry, especially for those values of $\varphi_H$ at which $H_{\parallel C}$ reaches a maximum.

### 1.3 Spontaneous phase transitions

The phenomena observed at spontaneous phase transitions near the Curie point have many traits in common with those which occur at orientational transitions. The curves of $\chi_{zz}(T)$ for $H_{\parallel} = \text{const}$ at temperatures $T \lesssim T_C$ exhibit a discontinuity due to the transition of the film to an inhomogeneous state; this anomaly shifts to lower temperatures with increasing $H_{\parallel}$. For $H_{\perp} = 0$, when the SLHS line $T^{(U\to St)}(H_{\parallel})$ in the $H_{\parallel}T$ plane is symmetric with respect to the $T$ axis, the $\chi_{zz}(T)$ curves in the paraphase generally exhibit a faint "high-temperature" ($HT$) peak in a narrow temperature region about 10 K above $T_C$ in addition to the two symmetric "low-temperature" (LT) peaks predicted by the theory. All these peaks shift to higher temperatures with increasing $|H_{\parallel}|$.

The $\chi_{zz}(T)$ curves at various values of $H_{\parallel}$ for film No. 4 [composition $(YGdYbBi)_3(FeAl)_5O_{12}$, orientation (111)] are given in Fig. 6; the inset shows the curves describing the position of the LT and HT peaks on the $H_{\parallel}T$ plane and also the position of the SLHS line.

The cause of the HT peak of $\chi_{zz}$ in the paraphase is unknown. It has been conjectured[17] that the HT peak is due to inhomogeneity of the film over its thickness. Offered as evidence for this view[17] was the fact that the HT peak disappeared after the film was annealed in air at a temperature of 1200 °C. In our films a high-temperature annealing also eliminated the HT peak. However, a magnetooptical study of the change in the structure of the films during the annealing showed (see Ref. 18) that annealing at temperatures of 1200 °C and higher does not increase the homogeneity of the samples. On the contrary, because of the expansion of the

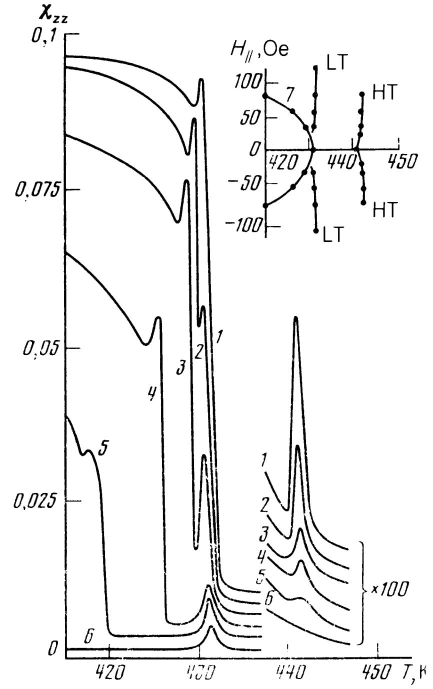


FIG. 6. Curves of $\chi_{zz}(T)$ for film No. 4 in the neighborhood of the Curie point for $H_{\perp}=0$. The inset shows the SLHS line (7) and the position of the peaks of $\chi_{zz}(T)$ on the $H_{\parallel}T$ plane. On curves 1–6: $H_{\parallel}=0$, 25, 33, 58.3, 83.3, and 125 Oe. Each curve has been shifted vertically from the next for legibility.

film at the free surface and the intense interdiffusion of ions between the film and substrate, the changes in the parameters of the film over its thickness were much greater than they had been before the annealing.[18] It seems likely that the HT peak vanishes after annealing precisely because the film becomes highly inhomogeneous. To check this hypothesis we studied a large number of films with different degrees of inhomogeneity of the parameters over thickness. It turned out that the HT peak was present only in rather homogeneous samples, in which (for example) the compensation temperature did not vary more than 10 K over the thickness. Moreover, if in those films which did have an HT peak the probe light beam was shifted from the center to the periphery of the film, where the inhomogeneity of the properties over the thickness is largest, the value of $\chi_{zz}$ at the point of the HT peak decreased rapidly and the width of the HT peak increased, until the peak had disappeared completely.

When the thickness of the films was gradually reduced by mechanical polishing, the HT peak remained, and the ratio of the values of $\chi_{zz}$ at the points of the HT and LT peaks (for the same value of $H_{\parallel}$) stayed practically constant. This shows that the HT peak is not due to a thin surface inversion layer at the film—substrate interface, as such a layer generally has a higher Curie temperature.[19]

It has been established that the HT peak exists only in films having a strong uniaxial anisotropy ($\beta_u \gg 1$) and a small anisotropy in the basal plane ($\beta_u \gg \beta_p, \beta_c$). For these films the domain structure typically becomes highly amorphous at a spontaneous second order phase transition. Strong anisotropy in the basal plane is evidently unfavorable for the existence of the HT peak. This surmise is confirmed by the influence of $H_{\perp}$ on the HT peak: with increasing $H_{\perp}$ the HT gets smeared out and gradually vanishes.

One possible cause of the HT peak might be the existence of bound states of magnons (solitons) in the paraphase.

## 2. CALCULATION OF THE SUSCEPTIBILITY OF QUASI-UNIAXIAL FERROMAGNETIC FILMS AT ORIENTATIONAL PHASE TRANSITIONS

For studying the field dependence of the static magnetic susceptibility tensor

$$\chi_{ik}=-\frac{1}{V}\frac{\partial^2 F}{\partial H_i\,\partial H_k} \qquad (1)$$

in a quasi-uniaxial iron garnet film of thickness $L$ in the neighborhood of an orientational phase transition, let us (following Ref. 15) write the free energy of the magnet as a sum of three terms:

$$F=F^{(0)}+F^{(d)}+F^{(fl)}, \qquad (2)$$

where

$$F^{(0)}(\mathbf{m}_0)=2\pi M_0^2 V[f_A(\mathbf{m}_0)-2(\mathbf{m}_0\mathbf{h})-m_{0z}^2] \qquad (3)$$

is the free energy due to the presence of the homogeneous magnetization component $\mathbf{m}_0$ induced in the film by an external magnetic field $\mathbf{H}=4\pi M_0(h_\perp \mathbf{e}_y + h_\parallel \mathbf{e}_z)$; $F^{(d)}$ and $F^{(fl)}$ are the domain part and fluctuation part of the free energy; $f_A$ is the normalized magnetic anisotropy energy (see Ref. 15); $\mathbf{m}=\mathbf{M}/M_0$ ($M_0$ is the saturation magnetization), and $V$ is the volume of the sample. In a homogeneous phase $F^{(d)}=0$; in an inhomogeneous phase $F$ can validly be represented as a sum of three terms only in the domain of application of the theory developed in Ref. 15. By defining the tensor $\hat{\chi}$ in the form (1), we are seeking the spatially averaged response of the entire film to a uniform magnetic field $\mathbf{H}$.[3)] It follows from (1) that the tensor $\hat{\chi}$ also can be written in the form

$$\hat{\chi}=\hat{\chi}^{(0)}+\hat{\chi}^{(d)}+\hat{\chi}^{(fl)}. \qquad (4)$$

### 2.1. Contribution of the homogeneous component of the magnetization

Let us determine the contribution to the susceptibility due to the homogeneous magnetization component $\mathbf{m}_0$. For ideal uniaxial films, the equation of state in neglect of domain structure and magnetization fluctuations is of the form

$$h_\perp m_{0z}-h_\parallel m_{0y}-\beta_u^* m_{0z}m_{0y}=0, \qquad (5)$$

where $\beta_u^*=\beta_u-1$. Using the relation $m_{oi}^2=1$ and introducing the notation $\mathbf{S}=\mathbf{h}/\beta_u^*$, we find

$$m_{0z}^4+2S_z m_{0z}^3+(S^2-1)m_{0z}^2-2S_z m_{0z}-S_z^2=0, \qquad (6)$$

i.e., the function $m_{0z}(\mathbf{h})$ is described by a fourth-order equation, which complicates the analysis. Nevertheless, certain information about the behavior of the susceptibility can be obtained without solving Eq. (6).

On general grounds the function $4\pi\chi_{zz}^{(0)} = \partial m_{0z}/\partial h_{\parallel}$ must have a maximum as a function of $h_\perp$ for $h_\parallel$ = const. Let us find the locus of the extrema on the $h_\parallel h_\perp$ plane. Differentiating (5) with respect to $h_\parallel$, we find that

$$4\pi\chi_{zz}^{(0)} = (h_\perp m_{0y}^{-3} - \beta_u^*)^{-1}.$$

Since $\partial\chi_{zz}^{(0)}/\partial h_\perp = 0$ at the maximum, we have

$$m_{0y} = 12\pi h_\perp \chi_{yy}^{(0)}, \tag{7}$$

where $4\pi\chi_{yy}^{(0)} = \partial m_{0y}/\partial h_\perp$. Determining $\chi_{yy}^{(0)}$ with the aid of (5) and substituting the resulting expression into (7), we get the extremum condition

$$m_{0z} m_{0y}^{-1} = \alpha_\parallel^{1/2}, \tag{8}$$

where $\alpha_\parallel = (h_\parallel/2h_\perp)^{2/3}$. Combining (8) with the equation of state (5), we find the equation of the line describing the position of the extrema on the $h_\parallel h_\perp$ plane:

$$\alpha_\parallel^3 - {}^3/_4\alpha_\parallel + {}^1/_4(1 - S_y^{-2}) = 0,$$

which has the solution

$$|h_\parallel| = 2h_\perp\{\cos[\pi/3 + {}^1/_3\arccos(1 - \beta_u^{*2}h_\perp^{-2})]\}^{3/2}. \tag{9}$$

For $h_\perp \to \beta_u^* + 0$ the field $h_\parallel$ goes to zero, and expression (9) reduces to

$$h_\perp \approx \beta_u^*[1 + {}^3/_2(h_\parallel/2\beta_u^*)^{2/3}], \tag{10}$$

i.e., the extremum of $\chi_{zz}^{(0)}$ is described approximately by a critical exponent of 2/3. In the neighborhood of the point $h_\parallel = 0$, $h_\perp = \beta_u^*$ we can obtain an explicit expression for $\chi_{zz}^{(0)}$ at the extremum, viz.,

$$4\pi\chi_{zz}^{(0)} \approx {}^1/_3(4\beta_u^*)^{-1/3}h_\parallel^{-2/3} \approx {}^1/_2(h_\perp - \beta_u^*)^{-1}.$$

For $h_\perp \gg \beta_u^*$ the curves for the extrema (9) approach the asymptotes

$$|h_\parallel| = h_\perp/\sqrt{2} - \sqrt{3}\beta_u^*/2,$$

i.e., the critical exponent approaches unity.

The initial (small $h_\parallel$) parts of curves (9) are inaccessible to observation because of the "screening" by the SLHS line, whose equation for an ideally uniaxial film is[15]

$$h_\perp \approx \beta_u(1 - {}^3/_2 h_\parallel^2).$$

Curves (9) in the case under discussion are symmetric "whiskers" which intersect the SLHS line and come together on the abscissa at the point $h_\perp = \beta_u^* = \beta_u - 1$.

The equation of the lines describing the position of the extrema of $\chi_{yz}^{(0)}$ on the $h_\parallel h_\perp$ plane is

$$\alpha_\perp^3 - 3\alpha_\perp^2 - 4(1 - S_y^{-2}) = 0,$$

where $\alpha_\perp = (2h_\parallel/h_\perp)^{2/3}$; hence

$$|h_\parallel| = {}^1/_2 h_\perp\{1 - 2\cos[\pi/3 + {}^1/_3\arccos(2\beta_u^{*2}h_\perp^{-2} - 1)]\}^{3/2}. \tag{11}$$

In the neighborhood of the point $h_\perp = \beta_u^* + 0$, $h_\parallel = 0$ expression (11) reduces to

$$h_\perp \approx \beta_u^*[1 + {}^3/_8(2h_\parallel/\beta_u^*)^{4/3}], \tag{12}$$

i.e., the critical exponent is equal to 4/3. Under these approximations we have on the lines of the extrema

$$-4\pi\chi_{yz}^{(0)} \approx {}^1/_3(2/\beta_u^*)^{2/3}h_\parallel^{-1/3} \approx {}^1/_3(\beta_u^*)^{-3/4}[{}^1/_6(h_\perp - \beta_u^*)]^{-1/4}.$$

The asymptotes of curves (11) for $h_\perp \to \infty$ are the straight lines

$$|h_\parallel| = \sqrt{2}h_\perp - \sqrt{{}^3/_2}\beta_u^*.$$

The susceptibility component $\chi_{yy}^{(0)}$ does not have a maximum as a function of $h_\perp$. If $h_\parallel = 0$, then

$$4\pi\chi_{yy}^{(0)} = (\beta_u^*)^{-1}\theta(\beta_u^* - h_\perp),$$

where $\theta(x)$ is the Heaviside step function; for $h_\parallel \neq 0$ the jump in the susceptibility $\chi_{yy}^{(0)}$ at $h_\perp = \beta_u^*$ is smoothed out, and $\chi_{yy}^{(0)}$ becomes a smooth, decreasing function of $h_\perp$.

The following relations are often useful for calculating the components of the susceptibility tensor:

$$\begin{aligned} 4\pi\chi_{zz}^{(0)} &= m_{0y}^3 h_{yy}^{-1} = m_{0y}^3 m_{0z} h_{yz}^{-1} = m_{0z} m_{0y}^2 h_{zz}^{-1}, \\ -4\pi\chi_{yz}^{(0)} &= m_{0y}^2 m_{0z} h_{yy}^{-1} = m_{0y}^2 m_{0z}^2 h_{yz}^{-1} = m_{0y} m_{0z}^2 h_{zz}^{-1}, \\ 4\pi\chi_{yy}^{(0)} &= m_{0z}^2 m_{0y} h_{yy}^{-1} = m_{0y} m_{0z}^3 h_{yz}^{-1} = m_{0z}^3 h_{zz}^{-1}, \end{aligned} \tag{13}$$

where

$$h_{yy} = h_\perp - \beta_u^* m_{0y}^3, \quad h_{yz} = h_\perp m_{0z}^3 + h_\parallel m_{0y}^3; \quad h_{zz} = h_\parallel + \beta_u^* m_{0z}^3,$$

and, hence,

$$\chi_{zz}^{(0)} = (m_{0y}/m_{0z})^2 \chi_{yy}^{(0)} = -(m_{0y}/m_{0z})\chi_{yz}^{(0)}.$$

In the neighborhood of the point $h_\perp = \beta_u^* + 0$, $h_\parallel = 0$ the condition $m_{0y} \gg m_{0z}$ is satisfied; this implies that $\chi_{zz}^{(0)} \gg |\chi_{yz}^{(0)}| \gg \chi_{yy}^{(0)}$. We note that the component $\chi_{yz}^{(0)}$ is negative, since $m_{0z}$ decreases with increasing $h_\perp$.

For real films[4)] (having cubic and rhombic anisotropy components and a deviation of the easy axis from the normal) the analysis of the behavior of the susceptibility can be carried out only for $m_{0z} \ll 1$. Straightforward but laborious calculations show that all the formulas given above remain valid for real films in which the uniaxial anisotropy constant obeys $\beta_u \gg \max\{\beta_p, \beta_c, \beta_u^2 \varphi_u\}$, provided the following substitutions are made: $\beta_u^* \to \beta u_{\text{eff}}^*$, $h_\parallel \to h_{\parallel\text{eff}}$, where

$$\beta_{u\,eff}^* = \beta_u^* + {}^1/_2\beta_p + {}^1/_2\beta_c - \beta_{2c}, \qquad h_{\parallel\,eff} = h_\parallel + \beta_{uc} + \beta_{pc}^* + \beta_{1c} \tag{14}$$

(see Ref. 15 for notation). The equation of the SLHS line (the critical parabola) for real films is of the form

$$h_\perp = h_{\perp c}[1 - {}^3/_2(h_\parallel - h_{\parallel c})^2], \tag{15}$$

and the coordinates of the critical parabola peak are determined by the expressions

$$\begin{aligned} h_{\perp c} &= \beta_u + {}^1/_2\beta_p + {}^1/_2\beta_{pc} - \beta_{2c} - 2\pi D^{1/2} L^{-1}\mu^{-1/2}, \\ h_{\parallel c} &= -\beta_{uc}(1 + \beta_u^{-1}) - \beta_{pc}^* - \beta_{1c}(1 + 3\beta_u^{-1}), \end{aligned} \tag{16}$$

where $D$ is the inhomogeneous-exchange interaction constant and $\mu \approx 1 + \beta_u^{-1}$. The curves representing the locus of the maxima of $\chi_{zz}^{(0)}$ and $\chi_{yz}^{(0)}$ on the $h_\parallel h_\perp$ plane are determined by Eqs. (9) and (11) after substitution (14) is made for $\beta_u^*$ and $h_\parallel$ and are asymmetric with respect to the critical parabola (15). The coordinates of the point $(h_\perp^*, h_\parallel^*)$ at which the "whiskers" come together are given by $h_\perp^* = \beta_{u\text{eff}} - 1$ and $h_{\parallel\text{eff}} = 0$, i.e.,

$$h_\perp^* = \beta_u - 1 + {}^1/_2\beta_p + {}^1/_2\beta_{pc} - \beta_{2c} = h_{\perp C} - 1 + 2\pi D^{1/2} L^{-1} \mu^{-1/2},$$

$$h_\parallel^* = -\beta_{uc} - \beta_{pc}^* - \beta_{1c} = h_{\parallel C} + \beta_{uc}\beta_u^{-1} + 3\beta_{1c}\beta_u^{-1}. \qquad (17)$$

If the condition $m_{0z} \ll 1$ does not hold, the solution cannot be found in analytical form. Nevertheless, it is obvious that in the region of large $h_\perp$ the critical exponents for the maxima of $\chi_{zz}^{(0)}$ and $\chi_{yz}^{(0)}$ will differ from 2/3 and 4/3, respectively.

### 2.2. Domain-structure contribution

Let us begin our study of the domain contribution $\chi^{(d)}$ to the susceptibility by considering films which have anisotropy in the basal plane and a moderately high uniaxial anisotropy ($\beta_u \gtrsim 1$); an orientational phase transition in such a film is accompanied by the formation of a stripe domain structure, while the formation of two-dimensional domain lattices is energetically unfavorable.[15] In this case there are two critical points on the SLHS line: $K_1$ and $K_2$, with coordinates $H_{\parallel K}$ and $H_{\perp K} = H_\perp^{(U\to\text{St})}(H_{\parallel K})$ which are symmetric with respect to the apex of the critical parabola. On the interval $K_1K_2$ the nucleation of the domain structure occurs as a second order phase transition, while outside this interval it is a first order transition.[13,15,20] Following Refs. 13, 15, and 20, we write the domain part of the free energy in the form

$$F^{(d)} = \frac{-2\Delta^2 M_0 V [\varepsilon^{(U\to\text{St})}]^2}{9pH_{\perp C}} \left(1 + \frac{R\varepsilon^{(U\to\text{St})}}{12p^2}\right) \qquad (18)$$

in regions which are far from the critical points, i.e., for $\varepsilon^{(U\to\text{St})} \ll p$, and as

$$F^{(d)} = -1.346\Delta^2 M_0 V H_{\perp C} [\varepsilon^{(U\to\text{St})}]^{3/2} \qquad (19)$$

in the neighborhood of the critical points, i.e., for $1 \gg \varepsilon^{(U\to\text{St})} \gg p$. Here $\Delta = 8\pi^2\mu^{-1}L^{-1}D^{1/2}M_0$ is the shift of the field of the orientational phase transition due to the finite size of the film; $p = 1 - \eta_c^2\eta_k^{-2}$, $\varepsilon^{(U\to\text{St})} = (H_\perp^{(U\to\text{St})} - H_\perp)\Delta^{-1}$, $R = \frac{161}{108} - \frac{39}{2}\eta_c^2\eta_k^{-2} + \frac{123}{16}\eta_c^4 \times \eta_k^{-4}$; for the remaining notation see Ref. 15. Substituting (18) and (19) into (1), we find that far from the critical points

$$\chi_{yy}^{(d)} = {}^4/_9 M_0 (pH_{\perp C})^{-1}(1 + {}^1/_4 R\varepsilon^{(U\to\text{St})}p^{-2}),$$

$$\chi_{yz}^{(d)} = a\chi_{yy}^{(d)} - {}^2/_9\eta_c\Delta\,\varepsilon^{(U\to\text{St})}\,(\pi\eta_k^2 H_{\perp C})^{-1}, \qquad (20)$$

$$\chi_{zz}^{(d)} = a^2\chi_{yy}^{(d)} - {}^1/_3\,\varepsilon^{(U\to\text{St})}\,\Delta(1 - {}^3/_4 p)(\pi^2 p^2 M_0)^{-1},$$

where $a = 3\eta_c h_{\perp C}(4\pi M_0)^{-1}$, and in the neighborhood of the critical points

$$\chi_{yy}^{(d)} = 0.1956 M_0 H_{\perp C}^{-1} [\varepsilon^{(U\to\text{St})}]^{-1/2}, \qquad (21)$$

$$\chi_{yz}^{(d)} = a\chi_{yy}^{(d)}, \quad \chi_{zz}^{(d)} = a^2\chi_{yy}^{(d)}.$$

On the line of second order phase transitions the components $\chi_{yz}^{(d)}$ and $\chi_{zz}^{(d)}$ have discontinuities which are proportional to $|\eta_c| = |H_\parallel - H_{\parallel C}|$ and $\eta_C^2 = (H_\parallel - H_{\parallel C})^2$, respectively, and which increase on approaching the critical points as $|H_{\parallel K} - H_\parallel|^{-1}$. In the neighborhood of the critical points we have $\hat\chi^{(d)} \propto (\varepsilon^{(U\to\text{St})})^{1/2}$. Since the function $R = R(H_\parallel)$ changes sign at

$$H_\parallel = H_{\parallel M}^{(1,2)} = H_{\parallel C} \pm 0.281\,|H_{\parallel K} - H_{\parallel C}|,$$

the component $\chi_{yy}^{(d)}$ increase with distance from the line of phase transitions in the field interval $H_{\parallel M}^{(2)} < H < H_{\parallel M}^{(1)}$, while outside this interval it decreases. The components $\chi_{yz}^{(d)}$ and $\chi_{zz}^{(d)}$ always decrease with distance from the line of phase transitions. Near the critical points the region of $H_\perp$ values in which $\hat\chi^{(d)}$ increases sharply becomes very narrow. We note that in order to calculate correctly the behavior of $\hat\chi^{(d)}$ with increasing distance from the line $H_\perp^{(U\to\text{St})}(H_\parallel)$, we must take the nonlinear field dependence of the magnetization rigorously into account, which is difficult to do using perturbation theory.

In the case of a large uniaxial anisotropy ($\beta_u \to \infty$) and a negligible anisotropy in the basal plane ($\beta_p \to 0$, $\beta_c \to 0$, $\varphi_u \to 0$), the phase diagram has a critical point $C$ of the first order phase transition ($H_\perp = H_{\perp C}$, $H_\parallel = 0$) from the homogeneous state to a hexagonal domain lattice; a first order phase transition from the hexagonal lattice to a stripe domain structure is also possible (see Refs. 14, 15, and 21). Following Refs. 15 and 21, we write the free energy of the hexagonal lattice in the form

$$F^{(d)} = -\frac{2^7 H_{\perp C} H_\parallel^4 V}{3^6 \cdot 5^3 \pi^8 M_0^3}(\varepsilon_1^{1/2} + 1)^3 (3\varepsilon_1^{1/2} - 1), \qquad (22)$$

where

$$H_\perp^{(L\to U)}(H_\parallel) = H_{\perp C}[1 + 8H_\parallel^2(45\pi^4 M_0^2)^{-1}]$$

is the equation of the line on which the hexagonal lattice loses stability against a transition to the homogeneous state, and

$$\varepsilon_1 = 45\pi^4 M_0^2 (8H_{\perp C}H_\parallel^2)^{-1}(H_\perp^{(L\to U)} - H_\perp).$$

Using (1) and (22), we obtain the components of the tensor $\hat\chi^{(d)}$ as

$$\chi_{yy}^{(d)} = 4M_0(15H_{\perp C})^{-1}(1 + \varepsilon_1^{-1/2}),$$

$$\chi_{yz}^{(d)} = -a_1\chi_{yy}^{(d)} - 64H_\parallel(3^3 \cdot 5^2\pi^2 M_0)^{-1}(1 + \varepsilon_1^{1/2}), \qquad (23)$$

$$\chi_{zz}^{(d)} = a_1^2\chi_{yy}^{(d)} + 2^{11}H_{\perp C}H_\parallel^2(3^5 \cdot 5^3\pi^8 M_0^3)^{-1}(1 + {}^3/_2\varepsilon_1^{1/2} + {}^1/_2\varepsilon_1),$$

where

$$a_1 = 16H_\parallel H_{\perp C}(45\pi^4 M_0^2)^{-1}.$$

It follows from (23) that $\hat\chi^{(d)}$ for the hexagonal lattice has a reciprocal square-root singularity in the neighborhood of the critical point $C$. The field dependence of $\hat\chi^{(d)}$ for the stripe domain structure is again described by Eqs. (20); $\hat\chi^{(d)}$ is discontinuous on the line of first order phase transitions from the stripe domain structure to the hexagonal lattice.

In the intermediate case of finite uniaxial anisotropy and nonzero anisotropy in the basal plane, the critical point $C$ is split into two triple points $R_1$ and $R_2$, at which there is a coexistence of the homogeneous state, the stripe domain structure, and one of the hexagonal lattices.[15] The tensor $\hat{\chi}^{(d)}$ does not exhibit singularities at these points.

### 2.3. Allowance for fluctuations

Let us determine the susceptibility corrections $\hat{\chi}^{(fl)}$ due to static fluctuations of the magnetization. In the homogeneous phase the Hamiltonian of a ferromagnet is written in the harmonic approximation as

$$H=\frac{V}{2}\sum_{n=1}^{\infty}\sum_{\mathbf{k}_\perp}\Omega^2_{\mathbf{k}_\perp,n}|X_{\mathbf{k}_\perp,n}|^2, \qquad (24)$$

where $X_{\mathbf{k}_\perp,n}$ are the normal coordinates expressed in terms of a linear combination of the amplitudes of the static fluctuations of the magnetization, and

$$\Omega^2_{\mathbf{k}_\perp,n}=2\pi\omega_n^2\omega_M^{-2}=2\pi[D(k_\perp^2+\tilde{q}_n^2-2k_c^2)+h_\perp-h_\perp^{(U\to\mathrm{St})}+\tilde{q}_n^2(k_x^2+\mu k_y^2+\tilde{q}_n^2)^{-1}]. \qquad (25)$$

Here $\omega_n$ are the spin-wave resonance frequencies [see Eq. (9) in Ref. 20]; $\omega_M^2\approx 16\pi^2\mu\beta_u g^2M_0^2$, where $g$ is the gyromagnetic ratio; $k_c=(\pi/\mu L^2D^2)^{1/4}$ is the critical value of $k_\perp$; $n$ is the number of the mode; and $\tilde{q}_n$ is the wave vector describing the distribution of $\mathbf{m}(\mathbf{r})$ over thickness of the film. Equation (25) is obtained from the Landau-Lifshitz equation in the approximation of a thick slab[5)] ($L\gg D^{1/2}$) and a large uniaxial anisotropy ($\beta_u\gtrsim 1\gg Dk_c^2=\varkappa_c^2$; see Refs. 12 and 15). The resonance frequency $\omega_n$ as a function of $\mathbf{k}_\perp$ has a minimum at $k_\perp=k_{cn}\approx\tilde{q}_n^{1/2}$. In the neighborhood of the points $k_{cn}$ we find[12] from the boundary conditions that $\tilde{q}_n=\pi nL^{-1}$. Far from the points $k_{cn}$ this relation does not hold; however, since the main contribution to the thermodynamic quantities comes from fluctuations in the region of phase space in which $k_\perp\sim k_{cn}$, we can ignore this circumstance. The fluctuation part of the free energy is given by the expression[5]

$$F^{(fl)}=-T\ln Z=-\frac{T}{2}\sum_{n=1}^{\infty}\sum_{\mathbf{k}_\perp}\ln(2\pi T/VM_0^2\Omega^2_{\mathbf{k}_\perp,n}), \qquad (26)$$

from which we get

$$\chi_{yy}^{(fl)}=T(4VM_0^2)^{-1}\sum_{n=1}^{\infty}\sum_{\mathbf{k}_\perp}\Omega^{-4}_{\mathbf{k}_\perp,n},\quad \chi_{yz}^{(fl)}=a\chi_{yy}^{(fl)},$$
$$\chi_{zz}^{(fl)}=a^2\chi_{yy}^{(fl)}-3TH_{\perp c}\sum_{n=1}^{\infty}\sum_{\mathbf{k}_\perp}(32\pi^2M_0^2\Omega_{\mathbf{k}_\perp,n})^{-1}. \qquad (27)$$

In the neighborhood of the SLHS line ($\varepsilon^{(U\to\mathrm{St})}\ll 1$) the tensor $\hat{\chi}^{(fl)}$ is of the form

$$\chi_{yy}^{(fl)}={}^1/_2T(I_2+{}^1/_2I_1\varkappa_c^{-2})(16\pi^2M_0^2)^{-1},\quad \chi_{yz}^{(fl)}=a\chi_{yy}^{(fl)},$$
$$\chi_{zz}^{(fl)}=a^2\chi_{yy}^{(fl)}-3TH_{\perp c}I_1(64\pi^3M_0^3)^{-1}, \qquad (28)$$

where

$$I_1=V^{-1}\sum_{\mathbf{k}_\perp}i_1\approx\frac{1+b}{4\pi DL(2\varepsilon^{(U\to\mathrm{St})}+1)^{1/2}}K\left[\left(1+\frac{2\varepsilon^{(U\to\mathrm{St})}}{\Gamma}\right)^{-1/2}\right],$$

$$I_2=V^{-1}\sum_{\mathbf{k}_\perp}i_2\approx-\frac{(1+b)M_0}{2DL\varepsilon^{(U\to\mathrm{St})}(2\varepsilon^{(U\to\mathrm{St})}+\Gamma)^{1/2}\Delta}E\left[\left(1+\frac{2\varepsilon^{(U\to\mathrm{St})}}{\Gamma}\right)^{-1/2}\right],$$

$$i_j=[D(k_\perp^2-2k_c^2)+h_\perp-h_\perp^{(U\to\mathrm{St})}+\varkappa_c^4D^{-1}(k_\perp^2-\Gamma k_y^2)^{-1}]^{-j},\quad j=1,2;$$

$\Gamma=1-\mu^{-1}\approx(1+\beta_u)^{-1}$, $b$ is a coefficient of order unity, and $K(x)$ and $E(x)$ are elliptic integrals of the first and second kind, respectively. In deriving Eq. (28) we have used the relations

$$\sum_{n=1}^{\infty}\Omega^{-4}_{\mathbf{k}_\perp,n}\approx\frac{1}{4\pi^2}\left(i_2+\frac{i_1}{2\varkappa_c^2}\right),\qquad \sum_{n=1}^{\infty}\Omega^{-2}_{\mathbf{k}_\perp,n}\approx\frac{i_1}{2\pi}.$$

For films in which $\beta_u$ is not too large, we can write expression (28) in the region $\varepsilon^{(U\to\mathrm{St})}\ll\Gamma$ as

$$\chi_{yy}^{(fl)}=\frac{(1+b)T}{512\pi^4\Gamma^{1/2}D^{3/2}M_0^2\varepsilon^{(U\to\mathrm{St})}}\left(1+\varepsilon^{(U\to\mathrm{St})}\frac{2\Gamma+1}{\Gamma}\ln\frac{8\Gamma}{\varepsilon^{(U\to\mathrm{St})}}\right),$$
$$\chi_{yz}^{(fl)}=a\chi_{yy}^{(fl)}, \qquad (29)$$
$$\chi_{zz}^{(fl)}=a^2\chi_{yy}^{(fl)}-\frac{3TH_{\perp c}(1+b)}{1024\pi^4\Gamma^{1/2}DLM_0^3}\ln\frac{8\Gamma}{\varepsilon^{(U\to\mathrm{St})}}.$$

In this case the components $\chi^{(fl)}$ are proportional to $(\varepsilon^{(U\to\mathrm{St})})^{-1}$, i.e., there are singularities governed by an exponent $\tilde{\gamma}=1$. Critical behavior of this kind is characteristic for two-dimensional systems in the harmonic approximation.[6)]

For films in which $\beta_u\to\infty$ ($\Gamma\to 0$), the field dependence of the tensor $\hat{\chi}^{(fl)}$ is described by Eqs. (29) in a small neighborhood of the SLHS line ($\varepsilon^{(U\to\mathrm{St})}\gg 1$), while for $1\gg\varepsilon^{(U\to\mathrm{St})}\gg\Gamma$ it is determined by the expressions

$$\chi_{yy}^{(fl)}=(1+b)T[1024\cdot 2^{1/2}\pi^3D^{3/2}M_0^2(\varepsilon^{(U\to\mathrm{St})})^{3/2}]^{-1}(1+2\varepsilon^{(U\to\mathrm{St})}),$$
$$\chi_{yz}^{(fl)}=a\chi_{yy}^{(fl)}, \qquad (30)$$
$$\chi_{zz}^{(fl)}=a^2\chi_{yy}^{(fl)}-3TH_{\perp c}(1+b)(1024\cdot 2^{1/2}\pi^4DLM_0^3)^{-1}.$$

In the limiting case when $\Gamma\to 0$ the fluctuational corrections to $\hat{\chi}$ have a still stronger singularity, governed by an exponent $\tilde{\gamma}=3/2$. This is due to the growth of the phase volume of the fluctuations on decreasing anisotropy of the film in the basal plane.[7)] These results are in agreement with Ref. 22, where it was shown that the correlations of the fluctuation in isotropic systems increase strongly at the phase transition to the inhomogeneous state. Far from the SLHS line ($\varepsilon^{(U\to\mathrm{St})}\gg 1$) the fluctuational corrections to $\hat{\chi}$ are of the form

$$\chi_{yy}^{(fl)}=T(512\pi^3D^{3/2}M_0^2)^{-1}\ln[16\pi M_0\Delta^{-1}(\varepsilon^{(U\to\mathrm{St})})^{-1}], \qquad (31)$$

$$\chi_{yz}^{(fl)} = a\chi_{yy}^{(fl)}, \quad \chi_{zz}^{(fl)} = a^2\chi_{yy}^{(fl)}.$$

In this case the tensor $\hat{\chi}^{(fl)}$ has the logarithmic singularities characteristic of three-dimensional uniaxial systems, in which the fluctuations are suppressed by long-range dipolar forces.[23] In particular, an analogous singularity occurs in the fluctuational correction to the specific heat in the neighborhood of $T_C$ in an infinite uniaxial ferroelectric.[24,25] For $\varepsilon^{(U\to St)} \lesssim 1$ there is a transition from the critical behavior characteristic of infinite three-dimensional uniaxial structures having a dipole interaction ($\varepsilon^{(U\to St)} \gg 1$) to the behavior characteristic of two-dimensional systems with no dipole interaction ($\varepsilon^{(U\to St)} \ll 1$). Since the parameter $\varepsilon^{(U\to St)}$ is proportional to the thickness of the film, the region of two-dimensional behavior shrinks as $L$ increases. We note that for $\varepsilon^{(U\to St)} \ll 1$ the singular behavior of $\hat{\chi}^{(fl)}$ is governed by the contribution of the critical mode with $n = 1$, while for $\varepsilon^{(U\to St)} \gg 1$ it is determined by the contributions from all the modes.

In the domain phase the regular domain structure creates a periodic potential in the $xy$ plane, and consequently the fluctuation spectrum acquires a band nature.[21,26] It can be shown that in the domain of application of our theory ($|\varepsilon^{(U\to St)}| \ll 1$), the singular behavior of $\hat{\chi}^{(fl)}$ is governed by fluctuations associated with critical modes whose correlation lengths go to infinity as one approaches the stability-loss lines of the phases $[h_\perp = h_{\perp i} = f_i(h_\parallel)]$ and with acoustic modes which destroy the translational order in the domain structure.

Let us first consider the singular contribution to $\hat{\chi}^{(fl)}$ from fluctuations associated with the critical modes. The fluctuation part of the free energy in this case is of the form

$$F^{(fl)} = -\frac{T}{2} \sum_i \sum_{\mathbf{Q}} \ln (2\pi T / V M_0^2 \Omega_{i,\mathbf{Q}}^2),$$

where $i$ is the number (index) of the critical mode, and the form of the functions $\Omega_{i,\mathbf{Q}}$ is given in Ref. 15; the wave vector $\mathbf{Q}$ is reckoned from the points $\{\mathbf{k}_\perp\}$ in phase space at which $\Omega_{i,\mathbf{Q}}$ has a minimum.

For example, for $\beta_u \gtrsim 1$, we have in the neighborhood of the line of second order phase transitions from the stripe domain structure to the homogeneous state

$$\chi_{yy}^{(fl)} = T(32\pi^4\Gamma^{1/2}D^{3/2}M_0^2|\varepsilon^{(U\to St)}|)^{-1}, \quad \chi_{yz}^{(fl)} = a\chi_{yy}^{(fl)}, \quad \chi_{zz}^{(fl)} = a^2\chi_{yy}^{(fl)}, \tag{32}$$

i.e., here again the singular behavior of $\hat{\chi}^{(fl)}$ in the neighborhood of the stability-loss lines of the phases is described by an exponent $\tilde{\gamma} = 1$, which is characteristic of two-dimensional systems in the harmonic approximation. This circumstance allows us to treat a magnetic film (where the theory is applicable) as a model of a two-dimensional system (see Refs. 15 and 27). If $\beta_u \to \infty$, then

$$\chi_{yy}^{(fl)} \propto |H_{\perp i} - H_\perp|^{-1/2}, \quad \chi_{yz}^{(fl)} \propto H_\parallel |H_{\perp i} - H_\perp|^{-3/2}, \quad \chi_{zz}^{(fl)} \propto H_\parallel^2 |H_{\perp i} - H_\perp|^{-5/2}. \tag{33}$$

Let us turn to an analysis of acoustical fluctuations. Since a magnetic film in the neighborhood of the phase transition can be treated as a two-dimensional system, acoustical fluctuations destroy the long-range translational order in the regular domain structure with the formation of Berezinskiĭ phases, which are characterized by different degrees of ordering.[28–31] Such transitions were considered in Refs. 15 and 27. Using the relations of scaling theory, we can estimate the singular contribution to $\chi^{(fl)}$ in the neighborhood of the transition from the Berezinskiĭ phase to the ferrimagnetic liquid-crystal phase[5,31]:

$$\chi_{ik}^{(fl)} \propto \tilde{\varepsilon}(0)^{-2} R_+^{-2},$$

where $\tilde{\varepsilon}(0) = h_\perp - h_{\perp R}^{(s)}$ is the shift of the field $h$ relative to the phase transition field $h_{\perp R}^{(s)}$, $R_+ \propto \exp(\varepsilon^{-\nu}(0))$ is a parameter which determines the average distance between free magnetic dislocations in the domain structure: $\nu = 0.5$ and 0.369 for the stripe domain structure and hexagonal domain lattice, respectively. In the neighborhood of the phase transition point, $h_\perp = h_{\perp R}^{(s)} + 0$, the components of $\hat{\chi}^{(fl)}$ fall off exponentially, i.e., this singularity is extremely weak.

Magnetic defects of the regular domain structure such as free dislocations and disclinations, which exist in the ferrimagnetic liquid crystal and ferrimagnetic liquid, respectively, can cause the aforementioned anomalies of the tensor $\hat{\chi}^{(fl)}$ to become smeared out.

#### 2.4. Spontaneous phase transitions

Let us describe the procedure for generalizing the theory developed in Secs. 2.1–2.3 to the case of spontaneous phase transitions near the Curie point $T_C$ in the presence of a weak magnetic field $h_\parallel \ll 1$ (the field $h_\perp$ is assumed to be zero). The discussion will be limited to quasi-uniaxial films having no cubic anisotropy and no deviation of the easy axis from the normal ($\beta_c = 0, \beta_p \neq 0, \varphi_u = 0$). In this case the magnetization component $m_{0z} \ll 1$ induced by the field $h_\parallel$ satisfies the equation[13–15,21]

$$\delta m_{0z}^3 + (1-\xi) m_{0z} - h_\parallel = 0, \tag{34}$$

where $\xi = \xi(T)$ and $\delta \approx$ const are the coefficients of $m^2$ and $m^4$ in the expansion of the free energy. It follows from (34) that the components of $\chi_{zz}^{(0)}$ have a maximum as functions of $T$ (the maximum of the paraprocess[32]) at

$$1-\xi(T) = {}^3/_4 \delta^{1/3} (2h_\parallel)^{2/3}, \tag{35}$$

and on the line of extrema we have

$$4\pi\chi_{zz}^{(0)} = {}^2/_3 \delta^{-1/3} (2h_\parallel)^{-2/3} = {}^1/_2 [1-\xi(T)]^{-1}. \tag{36}$$

To obtain the explicit temperature dependence we set $\xi(T) = \xi_0'(T_0 - T)$, where $T_0$ is the Curie temperature in an unbounded medium and $\xi_0' = (\partial\xi/\partial T)_{T=T_0}$. As a result, we find that the position of the lines of extrema on the $H_\parallel T$ plane is given by the equation

$$T(H_\parallel) = T_0 - (\xi_0')^{-1} [1 - {}^3/_4 \delta^{1/3} (H_\parallel / 2\pi M_0)^{2/3}]. \tag{37}$$

The equation of the SLHS line (the critical parabola) is[13,15,21]

$$T^{(U\to St)}(H_\parallel) = T_0 - (\xi_0')^{-1} [\xi_c + 3\delta H_\parallel^2 (16\pi^2 M_0^2)^{-1}], \tag{38}$$

where

$$\xi_c=\xi_0'(T_0-T_c)=2\pi L^{-1}D^{1/2}\mu^{-1/2}\ll 1,$$

and $T_C$ is the Curie temperature of the film (the temperature at which the domain structure is nucleated for $H_\parallel = 0$). The point of intersection of the curves given by (37) satisfies the relation

$$T^*=T_0-(\xi_0')^{-1}=T_C-(\xi_0')^{-1}(1-\xi_c)<T_c,$$

i.e., again in the case of spontaneous phase transitions the initial parts of the curves (37) are "screened" by the SLHS line $T^{(U\to\mathrm{St})}(H_\parallel)$.

The domain and fluctuation contributions to the susceptibility $\chi_{zz}$ can be calculated using the theory developed in Secs. 2.2 and 2.3 by setting $H_{\parallel C} = 0$ in the corresponding formulas and making the following substitutions: $H_{\perp C}\to 8\pi\delta M_0$, $\varepsilon^{(U\to\mathrm{St})}\to(T^{(U\to\mathrm{St})}-T)\Delta_T^{-1}$, $\Delta\to 4\pi\xi_0'\Delta_T M_0$, $\Gamma\to\beta_p\beta_u^{-2}\ll 1$, where $\Delta_T = T_0 - T_C$.

### 2.5. Discussion of the results

The main results of our studies can be summarized as follows:

1. The experimental behavior of the peaks of the components $\chi_{zz}$ and $\chi_{yz}$ of the susceptibility tensor in the homogeneous state is in good agreement with the conclusions of the theory. For example, the solid lines in Fig. 7 show (for film No. 2) the calculated position on the $H_\parallel H_\perp$ plane of the SLHS line $H_\perp^{(U\to\mathrm{St})}(H_\parallel)$ [see Eq. (15)] and of the curves for the extrema of $\chi_{zz}^{(0)}$ and $\chi_{yz}^{(0)}$ [see Eqs. (9), (11), and (14); the coordinates of the calculated point of intersection of the curves are $H_\perp^* = 831$ Oe, $H_\parallel^* = -6.92$ Oe]. The dashed lines show the curves for the extrema of $\chi_{zz}^{(0)}$ and $\chi_{yz}^{(0)}$ as calculated by the approximate formulas (10) and (12). We see that the experimental points conform well to the solid lines and deviate significantly from the dashed line, i.e., the observable parts of the curves [the parts outside the line $H_\perp^{(U\to\mathrm{St})}(H_\parallel)$] are not described by simple power laws with critical exponents 2/3 and 4/3.

2. The experimental dependence of $\chi_{zz}$ and $\chi_{yz}$ on the field $H_\perp$ (or on the temperature) at the orientational (or spontaneous) phase transition is also described satisfactorily by the theory. On the line $H_\perp^{(U\to\mathrm{St})}(H_\parallel)$ [or $T^{(U\to\mathrm{St})}(H_\parallel)$] we observe the susceptibility discontinuity due (see Secs. 2.2 and 2.3) to the domain-structure ($\hat{\chi}^{(d)}$) and fluctuation ($\hat{\chi}^{(fl)}$) contributions. The calculated curves of $\chi_{zz}(H_\perp)$ and $\chi_{yz}(H_\perp)$ for films No. 2 at different values of $H_\parallel$ are shown by the solid lines in Fig. 8 (inside the critical parabola, where the domain structure exists, the curves are shown by dashed lines, since our theory does not apply in this region). The fluctuation part $\hat{\chi}^{(fl)}$ of the susceptibility (see Sec. 2.3) always diverges on the line $H_\perp^{(U\to\mathrm{St})}(H_\parallel)$ (only the component $\chi_{yz}$ for $H_\parallel = H_{\parallel C}$ is not affected by fluctuations) and then falls off rapidly, becoming comparable to $\hat{\chi}^{(0)}$ at $|H_\perp - H_\perp^{(U\to\mathrm{St})}|\sim 10^{-2}$–$10^{-3}$ Oe (for typical film parameters). The domain part $\hat{\chi}^{(d)}$ (see Sec. 2.2) diverges for $\eta_c>\eta_k$, where the nucleation of the domain structure occurs by way of a first order phase transition, and then decays rapidly, becoming comparable to $\hat{\chi}^{(0)}$ at

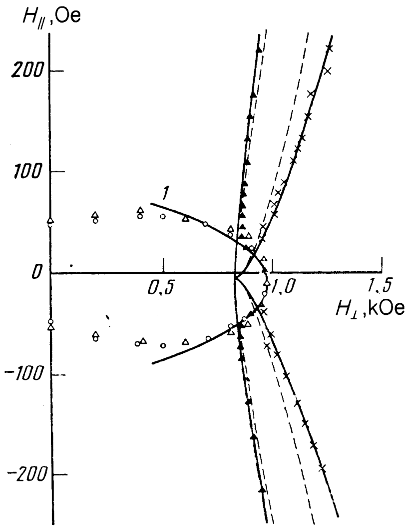


FIG. 7. Calculated positions of the SLHS line (1) and of the peaks of $\chi_{zz}^{(0)}(H_\perp)$ and $\chi_{yz}^{(0)}(H_\perp)$ on the $H_\parallel H_\perp$ plane for film No. 2. The solid lines are the exact solution, the dashed lines are the approximate solution; the legend for the experimental points is the same as in Fig. 4.

$|H_\perp - H_\perp^{(U\to\mathrm{St})}|\sim 10^{-2}$–$10^{-1}$ Oe. For this reason, on the scale chosen in Fig. 8, the corresponding parts of the curves are represented by vertical lines. Although the theory developed in Secs. 2.2 and 2.3 cannot be used to describe the behavior of $\chi_{zz}$ and $\chi_{yz}$ away from the line of phase transitions in the interior of the existence region of the domain structure, we can nevertheless state that in thick films at small values of $H_\parallel$ and for $H_\perp\to 0$ the component $\chi_{yz}$ goes to zero and $\chi_{zz}$ goes to $C_W(4\pi)^{-1}$, where $C_W\geqslant 1$ is a constant which depends on the ratio $W=L/l_w$ ($C_W = 1$ for $W\to\infty$, and $C_W\to\infty$ for $W\to 0$), where $l_w$ is the characteristic length of

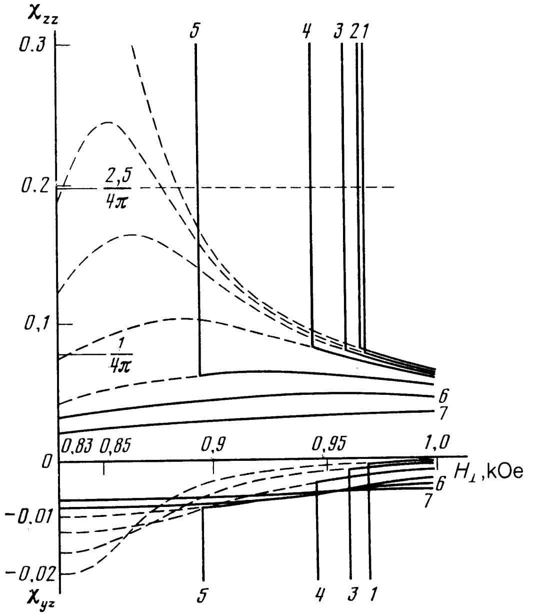


FIG. 8. Theoretical dependence of $\chi_{zz}$ and $\chi_{yz}$ on $H_\perp$ for film No. 2 ($H_{\parallel C} = -10.55$ Oe, $H_\parallel^* = -6.92$ Oe). On curves 1–7: $H_\parallel = -10.55$, $-6.92$, $-0.32$, 6.27, 19.47, 32.66, and 59 Oe.

the material.[33] For film No. 2 this ratio is $L/l_w = 7.2$, and so, according to Ref. 33, $C_W = 2.5$ (see Fig. 8). Behavior similar to that described has been observed for $\hat{\chi}$ in films with a small anisotropy in the basal plane (see Fig. 2); for a large anisotropy in the basal plane the behavior of $\hat{\chi}$ becomes more complex (Figs. 1b, 3b, and 5).

3. In real films the narrow spike that, according to the theoretical predictions, should appear on $\chi_{zz}$ at the line of phase transitions as a result of fluctuations and the onset of domain structure is smoothed out because of microscopic defects and inhomogeneities in the samples. This spike would be much more apparent at spontaneous transitions than at orientational transitions (cf. Figs. 2 and 6). However, the field dependence of $\chi_{yz}$ at orientational phase transitions exhibits a very pronounced "domain-fluctuation" peak even at second order phase transitions; the kinks on the $M_z(H_\perp)$ curves are also clearly visible (Fig. 3). This is because, first, $\chi_{yz}^{(0)}$ and $\chi_{yz}^{(d,fl)}$ have opposite signs and, second, $\chi_{yz}^{(0)} \ll \chi_{zz}^{(0)} \approx (4\pi)^{-1}$, while $\chi_{yz}^{(d,fl)}$ is considerably larger than $\chi_{zz}^{(d,fl)}$ at small values of $|H_\parallel - H_{\parallel C}|$ (see Secs. 2.1–2.3).

The results obtained in this study are convincing evidence that the domain structure and the actual anisotropy of the objects of study must be taken into account in designing and interpreting experiments on phase transitions in samples of finite size. For example, even an approximate determination of the critical exponents from the experimental curves of the susceptibility peaks becomes impossible in principle if the position of the point $(H_\parallel^* H_\perp^*)$ for an orientational phase transition or the value of $T^*$ for a spontaneous phase transition is unknown. The fluctuational contribution to the susceptibility in samples of finite size always adds to the domain-structure contribution, and this circumstance is usually not taken into account in the analysis of experimental results.

In conclusion, we note that although the theory developed in this paper gives a rather good description of the experiments, it must be kept in mind that it is essentially only a first correction to the Landau thoery, which is known to be inapplicable in a narrow region near lines of phase transitions, where one must take some other approach.[5]

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[1)]There is no jump on curve 5 because the line $H_\parallel = 55$ Oe intersects the stability-loss curve at a small angle.

[2)]One more reason for the discrepancy is that the $\chi_{yz}(H_\perp)$ curves were measured by the modulation technique. This also affects the initial parts of the curves for the high-field peaks of $\chi_{zz}$.

[3)]This orientational phase transition is isomorphic to the spontaneous phase transition near the Curie point $T_C$ (see below), and so the field dependence of the tensor is analogous to the temperature dependence of the specific heat near $T_C$.

[4)]Generally speaking, in such films the component $m_{0x}$ is nonzero, i.e., the susceptibility tensor has nonzero components $\chi_{xx}^{(0)}$, $\chi_{xy}^{(0)}$, and $\chi_{xz}^{(0)}$, but they are very small since $m_{0x} \ll m_{0z} \ll m_{0y} \approx 1$.

[5)]This approximation permits neglect of the contribution of the surface modes to the thermodynamic quantities.

[6)]The singular behavior of the tensor $\hat{\chi}^{(fl)}(H)$ is analogous to the behavior of the specific heat $C(T)$ near the Curie point, i.e., the exponent $\tilde{\gamma}$ is the same as the exponent $\alpha$, which for two-dimensional systems with no dipole interaction is equal to unity in the harmonic approximation.[5]

[7)]In the absence of anisotropy in the basal plane ($\beta_u \to \infty$) the function $\omega_n(\mathbf{k}_\perp)$ is minimum on a circle of radius $|\mathbf{k}| = k_c$.


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