Homogeneous magnetic relaxation in yttrium–iron–garnet in the vicinity of the phase transition

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Results are presented of an experimental investigation of the dynamics of homogeneous magnetization during the second-order phase transition in yttrium–iron–garnet (YIG) single crystals of different shapes. It is shown that the homogeneous relaxation significantly depends on both the quantity $4\pi\chi_{st}$ and the relation between the alternating-field frequency at which the investigation is carried out and the characteristic energies. It is shown that, starting from the temperatures corresponding to $4\pi\chi_{st}\approx 1$, the characteristic energy of the dipole interaction becomes frequency dependent, which indicates the inapplicability in this case of the Lorentz relation between the dynamic susceptibility and the homogeneous-relaxation time. This is a fundamental fact in the investigation of homogeneous relaxation by radio-frequency techniques. The temperature dependences of the homogeneous-relaxation time and the static susceptibility are determined in the exchange region. It is found that the phase transition in YIG is accompanied by anomalous phenomena that manifest themselves in the evolution and absorption of heat by the sample and in the appearance of additional distinctive features in the temperature dependence of the homogeneous-relaxation time.

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Experimentally, the critical dynamics of a ferromagnetic substance undergoing a second-order phase transition is investigated, as a rule, by two mutually complementary methods: with the aid of inelastic neutron scattering and by measuring the susceptibility in an alternating magnetic field in the case when $\lambda\gg L$ (λ is the wavelength of the field and L is the characteristic sample dimension). Critical neutron scattering yields primarily information about the dynamics of the inhomogeneous magnetization, $\mathbf{M}(\mathbf{q},\omega)$. Experiments performed by means of this method (see, for example, Refs. 1 and 2) have shown the applicability of the dynamic scaling law hypothesis at temperatures at which the interactions violating the law of conservation of total spin are unimportant, the most important of these interactions in cubic ferromagnets being the dipole interaction. The relaxation of the homogeneous magnetization, $\mathbf{M}(0,\omega)$, which is the order parameter, is due to the dipole forces, and its behavior in the critical region is usually investigated by radio-frequency methods, in which the temperature dependences of the real, χ', and imaginary, χ'', parts of the susceptibility are determined.

In the paramagnetic phase the dynamics of the homogeneous magnetization has been determined for two limiting values of the static susceptibility χ_{st}: when $4\pi\chi_{st}\ll 1$ (the exchange region)[3] and $4\pi\chi_{st}\gg 1$ (the dipole region).[4] In the exchange region, the characteristic dynamic-exchange-scaling energy, $\Omega_t\sim T_C\tau^{5\nu/2}$, is much greater than the quantity $\Gamma_0=1/t_0$, where τ is the relative temperature ($\tau=T/T_C-1$), ν is the critical exponent of the correlation length, and t_0 is the homogeneous magnetic relaxation time. In this case the dynamic susceptibility $\chi(\omega)$ can be expressed in terms of Γ_0 and the relation between ω and Ω_t:

$$\chi(\omega)=\chi_{st}\frac{\Gamma_0\gamma_t(\omega/\Omega_t)}{-i\omega+\Gamma_0\gamma_t(\omega/\Omega_t)}\tag{1}$$

($\gamma_t\approx 1$ when $\omega\ll\Omega_t$). According to Huber,[3] in the exchange region

$$\Gamma_0=\omega_0(\omega_0/T_C)^{1/4}(4\pi\chi_{st})^{5/4},\tag{2}$$

where $\omega_0=4\pi(g\mu_B)/v_0$ is the characteristic energy of the dipole interaction and v_0 is the volume of the unit cell.

At large values of the susceptibility an important role in the dynamics of the spin system should be played by demagnetization effects, and for the case when $4\pi\chi_{st}\gg 1$ Maleev has shown[4] that the characteristic dynamic-scaling energy Ω_d decreases, as T_C is approached, like

$$\Omega_d\sim\omega_0(\omega_0/T_C)^{1/4}(4\pi\chi_{st})^{-1/2};\tag{3}$$

in this case the quantity Γ_0 in (1) is of the order of Ω_d and the relation between Γ_0 and the susceptibility has the form

$$\chi(\omega)=\chi_{st}\frac{\Gamma_0\gamma_d(\omega/\Omega_d)}{-i\omega+\Gamma_0\gamma_d(\omega/\Omega_d)}\tag{4}$$

($\gamma_d\approx 1$ when $\omega\ll\Omega_d$).

Thus, when $\omega\ll\Omega_t$ for the exchange region and when $\omega\ll\Omega_d$ for the dipole region, (2) and (4) formally coincide, and, consequently, using the simple Lorentz form of these expressions, we obtain for the computation of the homogeneous-relaxation time from the susceptibility data the simple formula:

$$t_0=\frac{1}{\omega}\frac{\chi''}{\chi'}.\tag{5}$$

Generally speaking, at sufficiently high frequencies of the alternating magnetic field—such that ω is comparable to, or greater than, the characteristic energies Ω_t and Ω_d—the quantity Γ should differ from its static value Γ_0. In this case a simple relation of the type (5) between Γ_0 and $\chi(\omega)$ will cease to be valid.

It can be seen from the short review of the theoretical concepts that the critical relaxation of the homogeneous magnetization is determined essentially by the quantity

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$4\pi\chi_{st}$: as the transition point is approached, the value of the relaxation time should decrease in the exchange region and increase in the dipole region. On the other hand, for the transition region ($4\pi\chi_{st}\approx 1$) there are at present, as far as we know, no theoretical notions. Obvious difficulties also arise in the theoretical analysis of the behavior of the dynamic susceptibility at ω comparable to Ω_I and Γ_0. It follows from the analysis, carried out in Ref. 5, of the asymptotic behavior of $\Gamma(\omega)$ and $\chi(\omega)$ for $\omega\gg\Omega_I, \Omega_d$ that $\mathrm{Re}\chi(\omega)$ can assume in this case negative values and, consequently, at values of the alternating-field frequency higher than the characteristic energies, the magnitude of the susceptibility should decrease with increasing ω.

The dependence of Γ on ω for $\omega\gg\Omega_d$ does not allow us to speak of $1/\Gamma_0$ as a relaxation time. This is a fundamental circumstance in the experimental investigation of the homogeneous relaxation by radio-frequency techniques. For $\omega\to 0$ (or, which is the same thing, for $\omega\ll\Omega_I, \Omega_d$) the homogeneous-relaxation time and the dynamic susceptibility are related by the simple dependences (3) and (4). However, as T_C is approached, the magnitude of the characteristic dynamic-scaling dipole energy decreases, and can be equal to the value of ω (the condition $\omega\ll\Gamma_0$ is especially easily violated in the dipole region because of the smallness of the quantity Γ_0). In this case the dipole energy begins to depend on the frequency, and Γ can no longer be considered a constant. As T_C is approached further, i.e., as $T\to T_C$, it becomes impossible to determine the value of the homogeneous-relaxation time from the susceptibility data. This fundamental circumstance should always be taken into consideration in the analysis of experimental data on the critical behavior of the dynamic susceptibility.

In the present paper we give the results of an experimental investigation of the relaxation of the homogeneous magnetization in YIG single crystals in the vicinity of the second-order phase transition (some partial data were given in Ref. 6). The experimental samples had the forms of a cylinder with $N=1.26$ ($l=21$ mm, diameter 4 mm), a sphere (diameter 10 mm), and a torus ($12\times4\times4$ mm). To measure the permeability of the samples, we used a series resonant circuit (the procedure is described in detail in Ref. 7). The measurements were carried out in a frequency band: for the cylindrical sample, in the (1–3.8)-MHz range; for the torus, in the (0.02–1)-MHz range; and for the sphere, in the (1–0.5)-MHz range. In the experiments with the torus the increase of the relative accuracy of the measured quantities to 0.05% allowed the determination of susceptibility values starting from values $\chi\sim10^{-4}$. The direction of the radio-frequency magnetic field in the experiments on the sphere and cylinder coincided with the easy-magnetization axis [111]. The amplitude of the variable field did not exceed 10 mOe; the terrestrial magnetic field was decreased with the aid of shields to 60 mOe. The sample temperature was measured with a thermocouple glued to the sample surface. The stability of the temperature on a sample was not worse than $(2-3)\times10^{-3}$ deg. The permeability measurements were carried out under conditions of steady thermal equilibrium on the sample, a situation which was monitored by the constancy in time of the measuring-thermocouple's readings and of the measured physical quantities.

From the measured permeability values we computed the susceptibility of the material with allowance for the demagnetization factor, N, and the space factor. Here we should take into consideration the difficulty of the exact determination of the space factor for the sample inside the pickup loop. It is known that the fringing fields of an induction coil with an experimental sample placed in it lead to a situation in which not the true value of the permeability, μ, but the so-called effective permeability, μ_e, of the core is measured in the experiment. The space factor η takes into account the effect of the fringing fields on the value of the measurable permeability, η depending in the general case not only on the relation between the geometrical volumes of the induction coil and the sample, but also on the magnitude of the permeability of the sample. The last circumstance is very important for the investigation of the critical behavior of the susceptibility, when the range of its variation is fairly wide. In the general case the calculation of the connection between μ and μ_e is quite complex. The existing relations[8] are suitable only for $\mu\approx1$ and $\mu\gg1$. Only in the case of a homogeneous field in the sample can we sufficiently correctly compute the susceptibility of the material from the μ_e values. Thus, for a spherical sample in a spherically closed coil, it is easy to show that

$$\chi'-i\chi''=\frac{\{(\mu_e'-1)([2\eta-(\mu_e'-1)]-\mu_e''^2\}-i\cdot2N_{sp}\mu_e''}{N_{sp}\{[2\eta-(\mu_e'-1)]^2+\mu_e''^2\}} \tag{6}$$

where $N_{sp}=4\pi/3$ and η is the ratio of the volumes of the sample and the coil. For low sample-permeability values, it can be assumed that the space factor is equal to the ratio of the volumes of the sample and the pickup coil. We used such a definition of η in the computation of the susceptibility of the cylindrical sample. For a toroidal coil with close winding the fringing fields are, as is well known, fairly weak, which allows us to neglect their influence. In spite of the different degrees of approximation in the determination of the space factor in the $4\pi\chi_{st}<1$ case, good agreement between the susceptibility values for all the samples can be observed. It should, however, be borne in mind that, for large μ_e values, the computed magnitude of the susceptibility of the material is quite "sensitive" to the values of N and η.

From the critical behavior of the dynamic susceptibility (Figs. 1 and 2)[1)] we must first of all separate out the frequency dependences of χ' and χ'' of the toroidal sample. A change in the nature of the dependence $\chi''(\omega)$ occurs at a temperature corresponding to $4\pi\chi_{st}\approx1$. Starting from this temperature, the quantity χ'' decreases as the frequency of the alternating field increases, whereas for $4\pi\chi_{st}\ll1$ the dependence $\chi''(\omega)\sim\omega$. For $4\pi\chi_{st}\approx1$, the real part of the susceptibility also becomes frequency dependent. In this case there exists in the covered frequency range a simple empirical relation between χ' and ω: $\log\chi'=K\log\omega+\mathrm{const}$ (Fig. 3), such a dependence being observable only in the paramagnetic phase; for $T\to T_C$ the dependence $\chi(\omega)$ does not

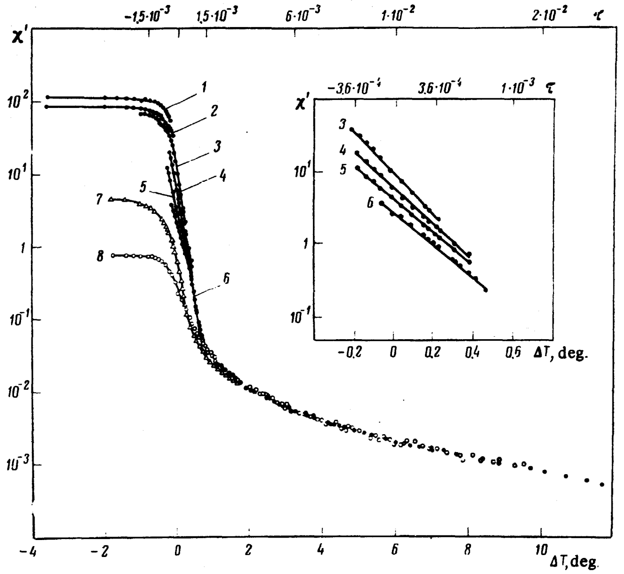

FIG. 1. Temperature dependence of the real part of the susceptibility of samples of different shapes. The curves 1–6, obtained respectively at the frequencies 0.03; 0.1; 0.2; 0.3; 0.5; and 1 MHz, pertain to the torus; the curve 7 (1 MHz) pertains to the cylinder; and the curve 8 (1 MHz), to the sphere.

have a simple form. The exponent K is itself temperature dependent, and assumes its maximum value at the transition point (Fig. 4a); here the dependence of $\log K$ on $\log\tau$ (Fig. 4b) represents a set of straight lines, i.e., $K\sim\tau^{y}$. A frequency-dependent real part of the susceptibility is observed not only for a torus, but also for a cylinder. However, in this case the quantity K is smaller than for a torus, and the dependence $\chi'(\omega)$ disappears in the ferromagnetic phase when $T-T_C>-2$ deg (for a torus the dependence $\chi'(\omega)$ exists right up to room temperatures). For a spherical sample, χ' is, within the limits of the experimental error, virtually frequency independent. The connection between the magnitude of the χ' dispersion and the sample shape is, apparently, not accidental. Indeed, it can be seen from Fig. 1 that there is relatively good agreement between the susceptibility values for all the samples in the region $4\pi\chi'<1$, but at lower temperatures the $\chi'(T)$ functions for samples of different shapes differ. Although the computed susceptibility value is, as has already been noted above, quite sensitive to the values of η and N for large magnitudes of μ_e, it turned out to be impossible to make, by a judicious choice of the space factor, the $\chi'(T)$ functions of all the samples coincide in the entire temperature range. The dependence of the quantity χ' on the shape of the body begins to manifest itself when $4\pi\chi\approx1$, the value of χ' decreasing with increasing N at a fixed temperature. The dependence on sample shape is also characteristic of the imaginary part of the susceptibility.

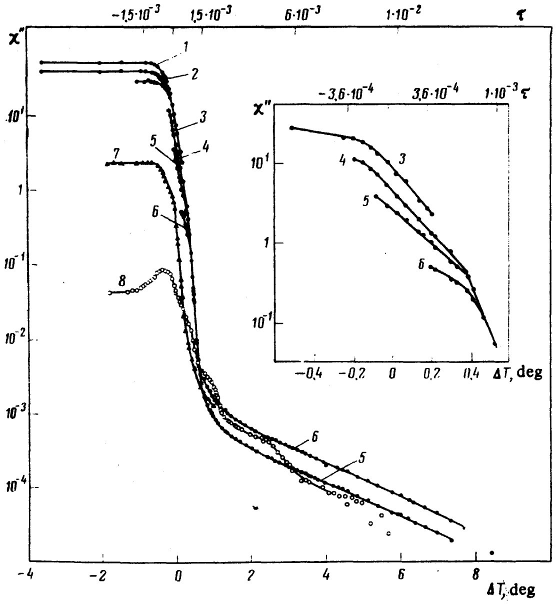

FIG. 2. Temperature dependence of the imaginary part of the susceptibility of samples of different shapes. The curves 1–6, obtained respectively at frequencies of 0.03; 0.06; 0.1; 0.3; 0.5; and 1 MHz, pertain to the torus; the curve 7 (1 MHz) pertains to the cylinder; and the curve 8 (1 MHz), to the sphere.

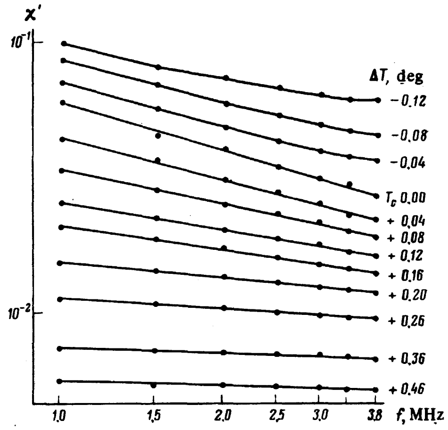

FIG. 3. Dependence of the real part of the susceptibility on frequency for the cylinder at different distances from T_C.

In the ferromagnetic phase we can distinguish two temperature regions where $\chi'(T)$ behaves differently: for $|\tau|<1\times10^{-3}$, $\chi'\sim\tau^{0.5}$, while in the region of lower temperatures the dependence of χ' on T virtually disappears. Such behavior of the susceptibility is characteristic of all the samples.

Knowing the dependences $\chi'(T)$ and $\chi''(T)$, we can, using the expression (5), determine the dependences

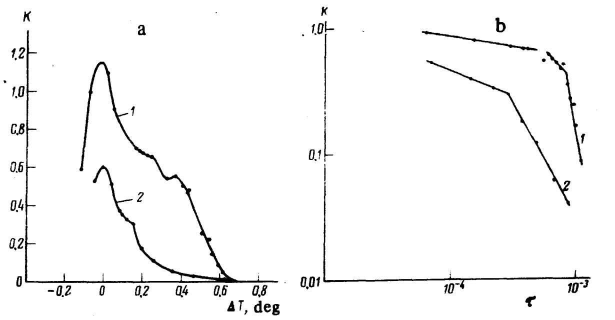

FIG. 4. a) Dependence of the exponent, K, of the variation of the real part of the susceptibility with frequency on temperature: 1) torus, 2) cylinder. b) Dependence of K on the relative temperature: 1) torus, 2) cylinder.

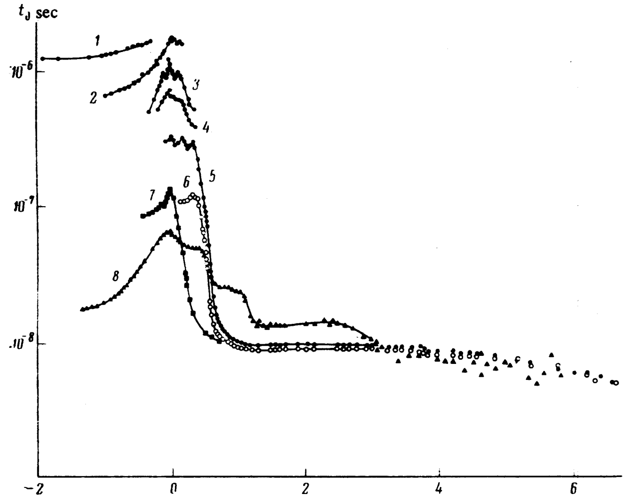

FIG. 5. Temperature dependence of the uniform-relaxation time computed from the expression (5) for samples of different shapes. The curves 1–6, obtained respectively at frequencies of 0.06; 0.1; 0.2; 0.3; 0.5; and 1 MHz, pertain to the torus; the curve 7 (1 MHz) pertains to the cylinder; and the curve 8 (1 MHz), to the sphere.

$t_0(T)$ and $\chi_{st}(\tau)$. Although, as has already been noted, the expression (5) is valid only if $\omega \ll \Omega_l$, Ω_d, it seems reasonable for the discussion of the behavior of the characteristic dipole energy, Γ, to compute t_0 and χ_{st} for $\omega > \Gamma_0$, bearing in mind here the limited applicability of the Lorentz form of the expression for the dynamic susceptibility. From the presented dependences $t_0(T)$ (Fig. 5) and $\chi_{st}(\tau)$ (Fig. 6), we should notice the following. First, in the exchange region the homogeneous-relaxation time does not increase with distance from T_C, as predicted in Ref. 3. Secondly, for $\tau > 1\times10^{-2}$ we have $\chi_{st} \sim \tau^{-1.75}$, while in the region $1\times10^{-2} > \tau > 1\times10^{-3}$ the susceptibility $\chi_{st} \sim \tau^{-1.33}$. Thirdly, for $4\pi\chi' \geqslant 1$ ($\tau \leqslant 1\times10^{-3}$), the computed values of t_0 and χ_{st} of all the samples begin to depend on frequency and body shape. And, finally, the appearance, besides the maximum in t_0, of additional distinctive features in the dependence $t_0(T)$ in the paramagnetic phase, features which are due, in our opinion, to the Curie point. These distinctive features are characteristic of both the torus and the spherical sample, for which they turned out to be the most strongly pronounced. These anomalies in $t_0(T)$ were also accompanied by the liberation and absorption of heat in the sample. This conclusion was drawn on the basis of the following arguments.

For the thermostat used in our experiments[7] there should obtain a linear dependence between the equilibrium values of the thermostat temperature, T_t, and the sample temperature, T. Only in this case can we assume the temperature field in the thermostat to be invariable. The thermal perturbations can give rise to a redistribution of power in the thermostat heaters, which will lead to a change in the temperature field. And, consequently, in this case the measuring thermocouple can, in the presence of a temperature gradient in the sample unit, indicate a change in the sample temperature different from the actual change. Therefore, in our experiments it was always necessary to monitor the linearity of the dependence $T(T_t)$. In the investigations with spherical samples we observed deviations from linearity in this dependence in the vicinity of T_C. For the purpose of ascertaining the causes of this phenomenon, the thermal coupling between the thermostat and the sample was weakened (measures were taken such that the heat exchange between them was effected primarily through heat radiation). If the rate of some thermal perturbations in the sample unit is higher than the rate of heat transfer between the thermostat and the sample, then the thermal conditions can be called quasi-adiabatic. Under such quasi-adiabatic (owing to the slackening of the heat transfer) conditions, we observed that, in the case when there is deviation from linearity of $T(T_t)$, the changes in the sample temperature always began earlier than the changes in the thermostat temperature, and what is more control experiments without a sample indicated the absence of such effects. Thus, the source of thermal perturbations could only be the sample. This allowed us to put forward the hypothesis that heat was liberated and absorbed by the sample, it having been established that the decrease of the sample temperature then occurs along the downward, and the increase along the upward, slopes of the additional distinctive features in $t_0(T)$.

The magnitude of the change in the sample temperature was affected by the finite value of the thermal coupling between the thermostat and the sample. This circumstance did not allow us to carry out a quantitative analysis of the observed effect, and its presence can best of all be judged from the change in the character of the dependence $T(T_t)$; but a very rough estimate yielded a maximum change in the sample temperature of the order of 0.1 deg. It was also observed that deviations in the dependence $T(T_t)$ increased as T_C was approached. Similar phenomena with the same character of the de-

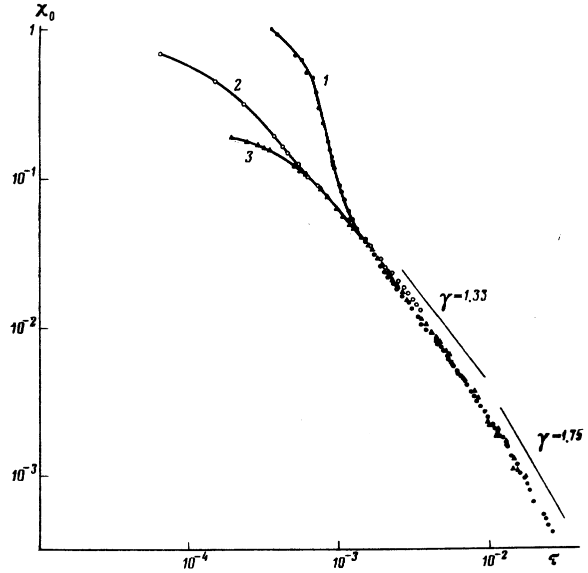

FIG. 6. Dependence of the static susceptibility, computed from the expression (5), on the relative temperature: the curve 1 (frequency 0.5 MHz) is for the torus; 2 (1 MHz), for the cylinder; and 3 (1 MHz), for the sphere.

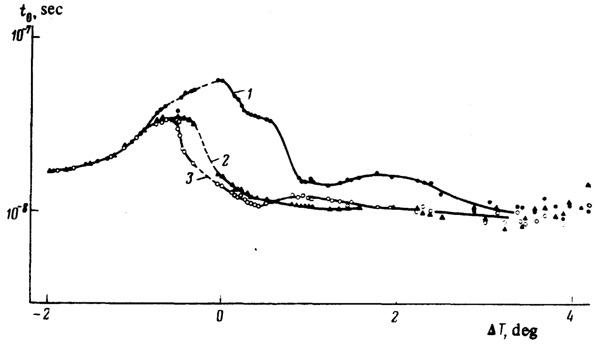

FIG. 7. The dependence $t_0(T)$ obtained for the sphere in the case when a constant magnetic field was applied by the "slit" method (frequency 0.5 MHz). The curve 1) $H=0$, 2) $H=4.3$ Oe, and 3) $H=8.5$ Oe.

pendence on distance from the transition point were also observed in the ferromagnetic phase. We should emphasize that the thermal effects developed also in the absence of an alternating magnetic field.

An analysis of the dependence $T(T_t)$ in experiments on samples of other shapes indicated similar deviations from linearity in $T(T_t)$ near T_C, but in these experiments the magnitude of the effect was influenced by a stronger thermal coupling between the thermostat and the sample.

In order to elucidate the character of the observed anomalies, we performed experiments with a spherical sample in the presence of a constant external magnetic field. The application of the field was effected in two ways. In one case the applied field was switched on during only a cycle of the passage through the critical temperature region. In the other procedure, conditionally called the "slit" method, the field was switched on only after the establishment of thermal equilibrium in the sample. In the settled equilibrium, the permeability measurements were carried out first in the absence of a field, then a 4.3-Oe field was switched on and the measurements were repeated, after which the strength of the external field was increased to 8.5 Oe. At the end of the measurements the constant field was switched off, and the temperature of the thermostat was changed. This measurement procedure was repeated upon the establishment in the sample of the next equilibrium temperature.

The dependence $t_0(T)$ obtained in the case when the external field was applied by the "slit" method is shown in Fig. 7. It can be seen that the constant field led not only to the smoothing out of the curve, but also to a shift toward the lower-temperature region of the distinctive features of this dependence. The field also enhanced the heat release in the sample, which was indicated by the larger—than in the absence of a field—deviations from linearity of $T(T_t)$ and the larger changes in the sample temperature. Since the enhancement of the thermal effects in the sample changed the temperature field in the thermostat to a considerable degree, the value of the new equilibrium sample temperature could (especially in the vicinity of T_C) differ strongly from the value of the previous equilibrium temperature, or be quite close to it. This circumstance complicated the temperature equidistance of the investigations. It is possible that it is precisely for this reason that there is, in Fig. 7, no t_0 peak connected with the transition point.

The changes in the sample temperature turned out to be very large in the experiments with an $H=21.5$-Oe external field that was constantly switched on. In this experiment, because of the considerable pumping, which arose owing to the heat release in the sample, of the temperature stabilization system of the thermostat, we could not carry out measurements in the vicinity of T_C.

In the absence of a constant magnetic field the dependence of χ' on T (Fig. 1) does not have distinctive features, at least within the limits of the measurement errors. However, even a brief application of a constant magnetic field caused the appearance of features in this dependence (Fig. 8). Such behavior of the susceptibility (especially at $H=0$ in the case when the field is applied by the "slit" method) is surprising. It turns out that the spin system in the paramagnetic phase possesses a "memory." Moreover, the "storage" state is quite stable if we take into consideration the fact that the establishment of thermal equilibrium during the transition from one equilibrium temperature to another under conditions of heat release by the sample can last for several hours and the fact that the constant magnetic field was switched off in this case.

As we know, the few published investigations of the critical behavior of the dynamic susceptibility were carried out largely on samples of cylindrical shape with N differing little from each other.[10-12] Therefore, a comparison of the behavior of the homogeneous relaxation in samples of one and the same material but having different shapes is of definite interest. Here the results obtained on a toroidal sample are "purer" for compari-

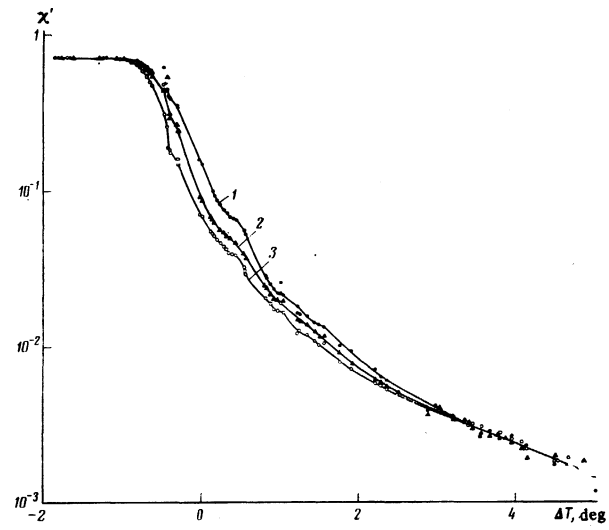

FIG. 8. The dependence $\chi'(T)$ obtained for the sphere in the case when a constant magnetic field was applied by the "slit" method (frequency 1 MHz). The curve 1) $H=0$, 2) $H=4.3$ Oe, and 3) $H=8.5$ Oe.

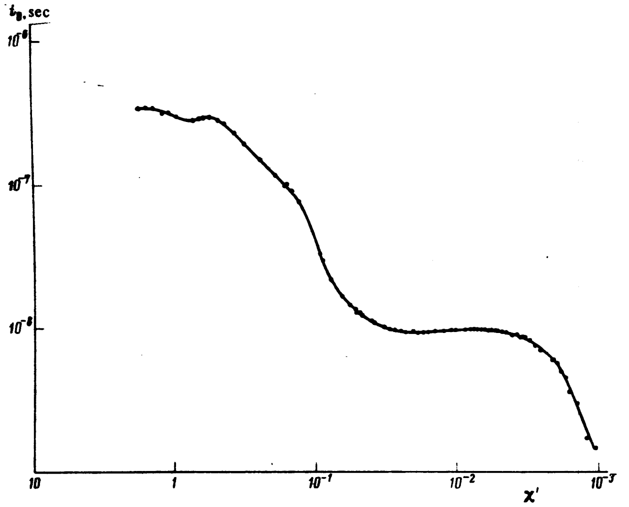

FIG. 9. The dependence $t_0(\chi_{st})$ for the torus (frequency 0.5 MHz).

son with the existing theoretical ideas because of the absence of a demagnetization factor.

The results of the performed investigations showed that the dynamics of the homogeneous magnetization in the paramagnetic phase for the transition depends significantly on the quantity $4\pi\chi_{st}$, which, of course, is not surprising, since the homogeneous relaxation in cubic ferromagnetic substances is wholly determined by the dipole forces.

From the analysis of the obtained results we can draw the following main conclusions.

First, the frequency dependence of the t_0 and χ_{st} values computed from (5) indicates that, starting from the temperatures corresponding to $4\pi\chi_{st}\approx 1$, the characteristic energy Γ depends on the frequency, which can be due only to the violation of the condition $\omega \ll \Gamma_0$. As has already been indicated above, this is of fundamental importance in the investigation of the relaxation of the homogeneous magnetization by radio-frequency methods, since, if special measures are not taken, then because $\Gamma_0 \to 0$ as the Curie point is approached the condition $\omega t_0 \ll 1$ will always be violated. But the transition in the region of very low frequencies entails well-known experimental difficulties. Apparently, this fundamental circumstance was not taken into account in the analysis of the temperature dependence of the homogeneous-relaxation time computed from (5) in Refs. 10 and 11. The dependence $t_0(T)$ given in these papers does not have a power form, but the increment in t_0 with temperature decreases as $T \to T_C$. In our opinion, such behavior of $t_0(T)$ can be due only to the above-named cause, so that in this case the application of the Lorentz expression for the computation of the homogeneous-relaxation time from data on the susceptibility becomes incorrect. For this reason it is virtually impossible to observe the behavior, predicted in Ref. 4, of the relaxation time in the dipole region. This can be seen from the dependence $t_0(\chi_{st})$ (Fig. 9). (Kötzler was, apparently, the first to use such a representation of the dependence of the relaxation time on the magnitude of the static susceptibility, a representation which is made convenient by the explicit absence of distances for T_C.) The decrease of the quantity χ' with increasing value of the frequency of the variable field is, possibly, also connected with the fact that $\Gamma_0 \to 0$ as $T \to T_C$. According to Maleev,[5] in the asymptotic limit $\omega \gg \Gamma_0$ the real part of the susceptibility should assume negative values. An estimation from (3) yields $\Gamma_0 = 2.5\times 10^7$ sec^{-1} for $4\pi\chi_{st}\approx 10$, but this value of the characteristic energy is not large enough for the asymptotic condition to be fulfilled in our frequency band.

Secondly, in the exchange region ($4\pi\chi_{st} \ll 1$) the dependence $t_0 \sim \tau$, predicted in Ref. 3, is not observed, although the estimation of the quantity t_0 from (2) yields reasonable agreement with the homogeneous relaxation time value (of the order of 10^{-8}) computed from the experimental data for $4\pi\chi_{st}\approx 10^{-2}$, so that the condition $\omega \approx \Omega_l$ is certainly satisfied. Such a discrepancy between theory and experiment can be explained by the fact that the static susceptibility behaves in the exchange region substantially differently above and below the temperature corresponding to $\tau \approx 1\times 10^{-2}$. For $\tau > 1\times 10^{-2}$ the exponent $\gamma \approx 1.75$, while in the region $3\times 10^{-3} < \tau < 1\times 10^{-2}$, γ is close to 1.33—the value given by the scaling law hypothesis for three-dimensional ferromagnets. Thus, it is possible that there occurs at $\tau \approx 1\times 10^{-2}$ some kind of phenomenon reminiscent of the crossing-over phenomenon, but for YIG the causes of its appearance are as yet completely unclear. On the other hand, it is known that for the majority of paramagnets[13] the relaxation time of the spin system increases with decreasing temperature, so that it is quite possible that the critical relaxation at large distances from T_C occurs in the "background" of the normal relaxation of a paramagnet. It should be noted that the dependence $t_0 \sim \tau$ is absent not only in a two-sublattice ferromagnet, which is what YIG is; in Ref. 10 its absence in the cubic ferromagnet EuS is also noted, though the values of $4\pi\chi_{st}$ given there are not much smaller than unity.

With regard to the anomalous phenomena accompanying the second-order phase transition in YIG in the paramagnetic region and the effect of sample shape on the behavior of the dynamic susceptibility, we can at present draw conclusions of only conjectural nature. In the first place, it should be noted that the anomalous phenomena apparently occur in all the investigated samples. Indeed, the deviation from linearity of the dependence $T(T_t)$ near T_C was observed in the experiments on all the samples; additional distinctive features in $t_0(T)$ were observed for the torus and especially for the sphere. Although for the cylinder they did not appear in their explicit form, the change in the character of the dependence $K(\tau)$ (Fig. 4b) indicated indirectly the presence of the anomalies in this case.

It is very tempting to attribute the anomalous effects to the ferromagnetic phase, but such an explanation meets with a number of difficulties, of which the most fundamental consists in the absence in this case of the critical-slowing-down phenomenon, which, on the basis of the most general physical arguments, should occur. And, consequently, it is necessary then to relate the maximum of the magnetic losses ($\mathrm{tg}\,\delta_{max}$) observed in all the investigations with a different phenomenon. However, in Ref. 9 it was shown that the Curie point determined from $tg\delta_{max}$ coincides with the T_C determined from polarized-neutron transmission. It should be noted that in all the presented experiments the maximum of $tg\delta$ and the maximum of t_0 computed from (5) were observed at one and the same temperature. Furthermore, experiments performed by us with the aid of a method similar to the method described in Ref. 14 on the determination of the behavior of the spontaneous magnetization in a spherical sample indicated the coincidence with respect to temperature of the Curie point found from the data of such an investigation and the one found from the magnetic-loss peak. An analysis of the experimental data on the critical behavior of the susceptibility in external constant magnetic fields also indicated that $\partial\chi/\partial H$ has a maximum at a temperature that we take for T_C. Let us note that the maximum of $\partial\chi_{st}/\partial H$ in Ref. 14 also corresponded to the Curie point. All this allows us to hope that the maximum of the tangent of the magnetic-loss angle is due to the critical slowing-down phenomenon.

The most problematic is the question of impurities and the various kinds of defects of the sample as sources of the observed anomalous phenomena. In view of the fact that the impurities, which for solids are virtually unavoidable, were not controlled in our experiments, this question remains open, and requires further investigations in this direction. However, a neutron-diffraction analysis of the spherical sample, for which the anomalies were most strongly pronounced, indicated that the degree of its mosaic order was 10′, which allows us to regard this sample as perfect from the crystallographic standpoint. And what is more, experiments performed on a sample of exactly the same dimensions, but consisting of several large lumps showed that for it the anomalies in $t_0(T)$ were smoothed out in magnitude to a greater degree and that the sample-temperature changes connected with the heat release in the sample were less abrupt. We should note here that the anomalous phenomena were observed at the same temperatures (or, correspondingly, at the same values of the susceptibility) as in the perfect sample. Such a difference in the behavior of the anomalies of the two spherical samples, as well as a comparison of the character of the anomalies in samples of other shapes, may indicate a decisive influence of the boundaries (or shape) on the pattern of the phenomena accompanying the phase transition in YIG.

The distinctive features in $t_0(T)$ are, possibly, connected with the appearance of the effect of heat evolution and absorption in the sample, as well as with the distinctive features in the dependence of K on τ. It seems reasonable to assume that the second-order phase transition in YIG is accompanied by the formation and destruction of some kind of structures having, apparently, a fluctuation character. Whether such formations have a metastable character is, for the present, difficult to say; for the solution of this problem requires special investigations. The nature of such type of structures can be related with spin waves, whose appearance in the paramagnetic phase has been discussed for a long time.[15] If the assumption of the existence of fluctuation formations is true, then the difference in the susceptibility values for the various samples for $4\pi\chi_{st}>1$ becomes understandable, since in this case the susceptibility will be determined also by the susceptibility of the structures.

From the temperature behavior of the real part of the susceptibility in the ferromagnetic phase we can determine the temperature dependence of the anisotropy constant near T_C. In a ferromagnet the susceptibility in weak fields is determined by the reversible rotation of the magnetization, χ'_a, and the reversible motion of the domain walls, χ'_{dc}, with $\chi'_a \sim I^2/k_a$, $\chi'_{dc} \sim I/k_a^{1/2}$, where I is the spontaneous magnetization and k_a is the anisotropy constant. Since in this approximation the temperature dependences $\chi'_a(T)$ and $\chi'_{dc}(T)$ have the same character, we can find from the experimentally obtained exponents of the dependences $I(T)$ and $\chi'(T)$ the exponent for the dependence $k_a(T)$. For the critical exponent, β, of the spontaneous magnetization, two values were obtained in Ref. 14: $\beta = 0.75$ in the region $10^{-4} < |\tau| < 10^{-3}$ and $\beta = 0.38$ for $|\tau| > 1\times10^{-3}$. It follows from our data that for these same temperature regions the exponents of the temperature dependence of the real part of the susceptibility are respectively equal to ~ 0.5 and ~ 0.0. Thus, in the region $|\tau| < 1\times10^{-3}$, $k_a \sim \tau^1$, while $k_a \sim \tau^{0.75}$ for higher temperatures. The dependence $k_a \sim \tau$ agrees with the prediction of Ref. 16 and with the data for some ferrites.[17] The change in the character of the dependence $\chi'(T)$ at $|\tau| \approx 1\times10^{-3}$ apparently indicates a transition from a state of homogeneous magnetization to a formed domain structure, but to split the susceptibility obtained from the experimental data into parts connected with the reversible rotation of the magnetization and the reversible motion of the domain walls is not possible. The difference in the susceptibility values for samples of different shapes in the ferromagnetic region is clearly due to the difference in the domain structures.

From the above discussion emerges quite a complicated picture of the dynamics of the homogeneous magnetization in YIG. The point is not only that the critical relaxation significantly depends on the quantity $4\pi\chi_{st}$, but also that the behavior of the dynamic susceptibility is also determined by the relation between the variable-magnetic-field frequency at which the investigation is carried out and the characteristic energies. The behavior of the homogeneous relaxation in the critical region is significantly complicated by the phenomena accompanying the phase transition, phenomena whose nature is for the present not clear to us. All this allows us at present to carry out only a semiquantitative comparison with the existing theoretical ideas, which, besides, pertain to the asymptotic cases.

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[1] Because of the considerable (by three orders of magnitude) change in the permeability of the torus, it was not possible to cover the entire temperature range at one frequency. There-

fore, the dependences $\chi'(\Delta T)$, $\chi''(\Delta T)$, and $t_0(\Delta t)$ are given in this case for a few frequencies.

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Dielectric-metal phase transition in V_3O_5

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Results are presented of a comprehensive investigation of a phase transition in V_3O_5, which is identified on their basis as dielectric–metal phase transition (DMPT). The entire investigated aggregate of properties (kinetic, magnetic, optical, structural) is interpreted is best fashion within the framework of a model with strong electron–electron correlations of the Mott-Hubbard type in both the low- and the high-temperature phases. Above the phase-transition temperature, V_3O_5 is a poor metal with a strong degree of disorder, a small mean free path, and localized magnetic moments. The DMPT that takes place at 430 K is apparently connected with differentiation of the charge and with the spatial localization of the vanadium ions with different valences.

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1. INTRODUCTION

Dielectric-metal phase transitions (DMPT) have been observed in many transition-metal compounds.[1] In particular, they are typical of vanadium oxides. By virtue of the variable valence (2–5), vanadium forms an entire series of oxides with both integer valence (VO, V_2O_3 VO_2) and with intermediate valence, such as the Magneli phase V_nO_{2n-1} ($n = 3-9$). DMPT take place in almost all the oxides. The only exceptions have up to now been taken to be V_7O_{13}, which remains metallic down to the lowest temperatures, and V_2O_5, which exhibits semiconductor properties.

In this paper we summarize the results of a comprehensive investigation of the phase transition previously observed[2] in V_3O_5 and interpret this transition as a DMPT. We study the kinetic characteristics (resistivity, thermoelectric power, Hall effect), the magnetic properties, and the optical characteristics (reflection coefficient in the visible and infrared), carry out structural investigations, and determine from differential thermal analysis (DTA) data the thermodynamic characteristics of the transition. We investigate also the influence of doping on the V_3O_5 properties.

An analysis of the entire aggregate of obtained data allows us to conclude that at 430 K a dielectric-metal phase transition takes place in V_3O_5. It is apparently similar in character with that of the transitions in other Magneli phases and is due mainly to spatial ordering of the V^{+3} and V^{+4} ions (the analog of Wigner crystallization). Thus, V_3O_5 can be regarded on the basis of its properties as belonging to the common series of vanadium oxides that undergo DMPT.

We report here successively the results of an experimental investigation of various properties of V_3O_5 (Secs. 3, 4) and then carry out a theoretical analysis that allows us to draw conclusions concerning the behavior of V_3O_5 at $T < T_c$ and $T > T_c$ and concerning the mechanism of the transiton (Sec. 5). We formulate also a number of questions that remain unanswered to this day.

2. TECHNOLOGY OF PRODUCTION OF V_3O_5 CRYSTALS

The V_3O_5 single crystals were grown by the method of chemical transport reaction in a closed volume, using $TeCl_4$ as the carrier.[3] To obtain single crystals of V_3O_5 of the lower limit of the homogeneity region the