Homogeneous magnetic relaxation near second-order phase transition

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To describe correctly a phase transition in the ferromagnet we must take into account not only exchange forces, but others as well, including the dipole-dipole interaction. One method of investigating the influence of dipole forces on a second-order phase transition is to study radio-wave absorption, which occurs only because of dipole-dipole forces. In the present work we have investigated the dynamic susceptibility of yttrium-iron-garnet single crystals near the Curie point. The relaxation time of homogeneous magnetization is calculated from the values obtained for the real and imaginary parts of the susceptibility. It is shown that the temperature dependence of the relaxation time is nonmonotonic. The influence of the demagnetization factor on the relaxation time can be evaluated on the basis of experiments performed on samples of different shapes. A comparison of the experimental results with current theoretical predictions shows that they are in qualitative agreement in the hydrodynamic region. The behavior of the dynamic susceptibility in stationary external magnetic fields indicates the complex nature of the dependence of the shift of the magnetic-loss peak on the external field strength.

An analysis of spin dynamics in the paramagnetic phase must take into account not only exchange forces, but also the dipole-dipole interaction (especially at low values of the momentum transfer). It is well known (see, e.g.,[1]) that exchange forces conserve the projection of the total spin on any axis. The dipole-dipole interaction leads to nonconservation of the total spin, and can be observed in the absorption of radio waves near the phase-transition point.

Current theoretical concepts of spin dynamics in a second-order phase transition refer to the hydrodynamic region, in which the size of fluctuations is characteristically much larger than the correlation radius. Van Hove[2] has suggested that the average magnetization in the paramagnetic phase ($T > T_C$, $H = 0$, where T_C is the Curie point) should relax slowly via diffusion. To take the dipole-dipole interaction into account, Krivoglaz[3] included in the diffusion equation an additional relaxation term $\mathbf{M}/\tau$, where $\mathbf{M}$ is the magnetization and τ is the relaxation time of the homogeneous magnetization, In this case the susceptibility at $k = 0$ (k is the wave vector) takes the form

$$\chi(\omega)=\chi'(0)/(1-i\omega\tau). \qquad (1)$$

This expression is valid when $\omega\tau \ll 1$. Another condition for the validity of Eq. (1) is $4\pi\chi' \ll 1$. From Eq. (1) we have for the relaxation time

$$\tau=\chi''/\omega\chi'. \qquad (2)$$

We have used this equation to calculate the relaxation time.

Vugal'ter and Maleev[4] have shown that, as a result of the spatial anisotropy of the dipole-dipole forces, the shape of the sample has a significant effect on the relaxation of the homogeneous magnetization. For instance, for a sample in the form of a triaxial ellipsoid the susceptibility along the j-th axis is given by the expression

$$\chi_j(\omega)=\chi(0)/(1-i\omega\tau_j) \qquad (3)$$

and τ_j can be expressed in terms of the two characteristic times τ' and τ''; $1/\tau_j = 1/\tau' + N_j/\tau''$, where N_j are the principal values of the demagnetization-coefficient tensor. The dipole-dipole forces as well as other interactions (anisotropy, magnetostriction, etc.) contribute to τ'. The value of τ'' is due only to dipole forces. In the far paramagnetic region, where $\chi_\perp(0) = \chi_\parallel(0)$ ($\chi_\parallel(0)$ and $\chi_\perp(0)$ are the longitudinal and transverse parts of the susceptibility, respectively, relative to the wave vector), we find from[4] that the relaxation time does not depend on the shape of the sample. At high values of the susceptibility, when the dipole-dipole interaction becomes significantly anisotropic, the sample shape begins to have an effect on the relaxation time.

Recently Huber[5] showed that, in the long-wave case in the hydrodynamic region, the relaxation time is proportional to the distance from the transition point, i.e., $\tau \propto \xi$. On the other hand it is clear that τ should increase as the Curie point is approached. We can therefore expect the relaxation time to depend non-monotonically on ξ in a wide temperature range.

Since a relaxation mechanism is responsible for the absorption of radio waves in the vicinity of a second-order phase transition, information about homogeneous magnetic relaxation can be obtained from a study of the behavior of the susceptibility near this point. In the present work the dynamic susceptibility near T_C is investigated in single-crystal samples of yttrium-iron garnets (YIG) in the form of cylinders of length 21 mm and diameter 4 mm and spheres of diameter 10 mm. The cylinder axis was set along the [111] direction; the spheres were oriented arbitrarily with respect to the measuring coil. For the spherical samples the measuring coil was also spherical with the same number of turns per unit length of the axis, thus guaranteeing homogeneity of the radio-frequency magnetic field in the sample. The stationary external magnetic field was in the same direction as the alternating field. The amplitude of the radio-frequency field did not exceed 5 mOe.

A series resonant circuit was used to measure the real and imaginary components μ' and μ'' of the magnetic permeability of the samples; the real part of the permeability was determined by the capacitance and the imaginary part by the resistance of the tank circuit at resonance. From the measured values of μ' and μ'', together with the demagnetization factor, we calculated the real (χ') and imaginary (χ'') parts of the susceptibility χ as well as the relaxation time τ of homogene-

ous magnetization. The error in the determination of χ' and χ'' did not exceed 5–7% over the entire temperature range, starting from $\xi = 6 \times 10^{-3}$ and below, where $\xi = (T - T_c)/T_c$.

The sample temperature was measured with a platinum resistance thermometer. The temperature was held constant within 5×10^{-3} deg by a temperature stabilization system similar to that used in[6]. The Curie point was assumed to be the temperature at which the peak magnetic loss was observed. We have already shown (see[6]) that the transition point determined in this manner coincides with the Curie point determined from the scattering of polarized neutrons.

The results of our investigation of the magnetic susceptibility are shown in Figs. 1 to 5. The data for the cylindrical sample, obtained at frequencies from 1 to 3.4 MHz (Fig. 1), show a weak frequency dependence of the relaxation time at $8 \times 10^{-4} < \xi < 2 \times 10^{-3}$. A similar dependence was obtained for the sphere at frequencies from 0.6 to 1 MHz. Generally speaking, when $\omega\tau \ll 1$ the relaxation time of homogeneous magnetization should not depend on the frequency. The observed weak frequency dependence of τ can only be attributed to the failure of this condition to be completely satisfied in our case (in fact, $\omega\tau \approx 0.1$ in the considered temperature range). There are at present no theoretical arguments to explain the large differences in the relaxation times observed for different frequencies in the immediate vicinity of T_c ($\xi < 1.5 \times 10^{-4}$).

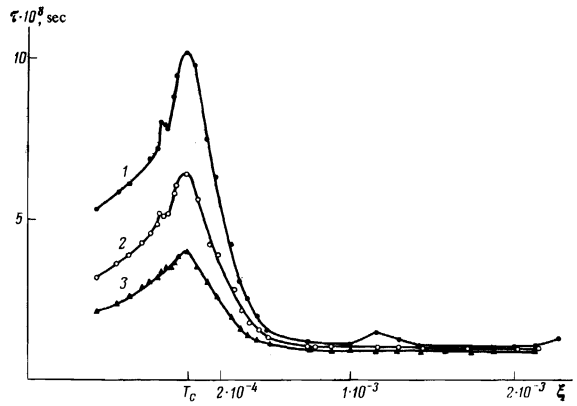

FIG. 1. Relaxation time τ as a function of the relative temperature for a cylinder at H = 0: 1) frequency 1 MHz, 2) 2 MHz, 3) 3.4 MHz.

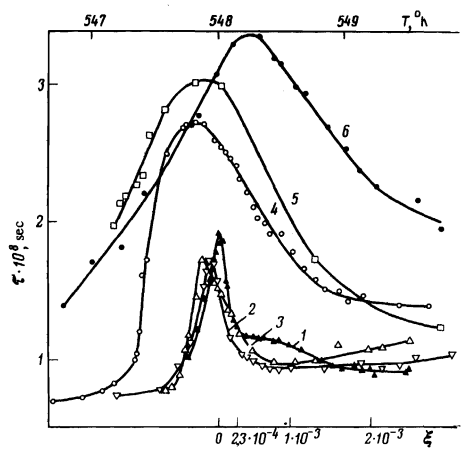

FIG. 2. Temperature dependence of the relaxation time τ for a sphere, at a frequency of 1 MHz in a stationary external magnetic field: 1) H = 0, 2) H = 3.4 Oe, 3) H = 8 Oe, 4) H = 40 Oe, 5) H = 60 Oe, 6) H = 128 Oe.

From a comparison of the relaxation times for a cylinder (Fig. 1) and a sphere (Fig. 2) at the same frequency (1 MHz) it is apparent that $\tau_{cyl} \approx \tau_{sph}$ in the far paramagnetic region ($\xi > 1 \times 10^{-3}$). We note that at $\xi > 1 \times 10^{-2}$ we obtained some experimental points indicating an increase in the value of τ with temperature, but the experimental errors in this temperature range (due to the smallness of χ at very large values of ξ) are too large for us to be certain of this behavior. Nevertheless, if this increase in the relaxation time with temperature is taken into account together with the fact that τ is independent of temperature for $6 \times 10^{-4} < 2 \times 10^{-3}$ and begins to increase for $\xi < 6 \times 10^{-4}$, we can say that the temperature dependence of the relaxation time is nonmonotonic.

In comparing the dependence of log χ' on log ξ (Fig. 3) it becomes apparent that, starting at $\xi < 8 \times 10^{-4}$, the values of χ' obtained at 1 MHz for the sphere and the cylinder diverge. This divergence cannot be eliminated by any reasonable choice of the demagnetization factor for the cylindrical sample. (Strictly speaking, for a cylinder of finite dimensions the demagnetization factor

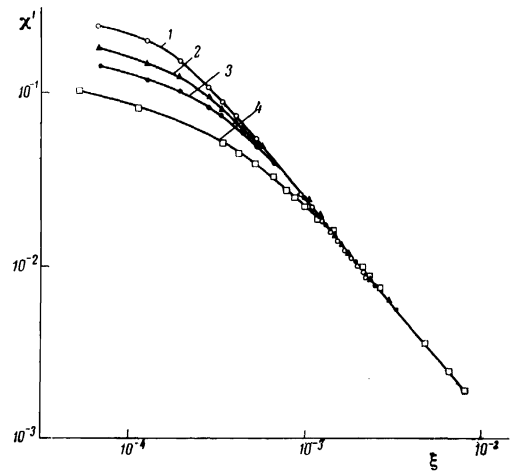

FIG. 3. Real part of the susceptibility as a function of the relative temperature in the paramagnetic region at H = 0, for a sphere (curve 4, frequency 1 MHz) and for a cylinder (curve 1, 1 MHz; curve 2, 2 MHz; curve 3, 3.4 MHz).

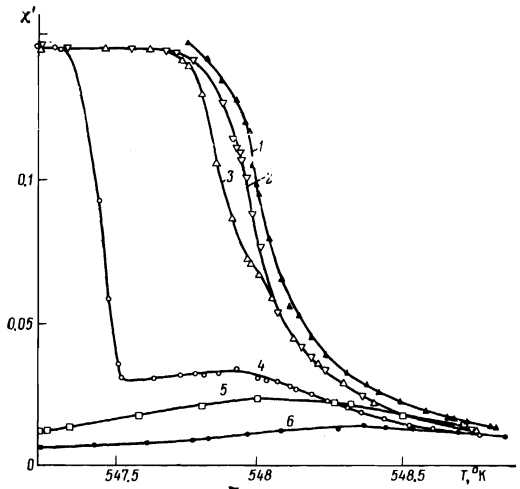

FIG. 4. Temperature dependence of the real part of the susceptibility for a sphere, at a frequency of 1 MHz in a stationary external magnetic field H: 1) H = 0, 2) H = 3.4 Oe, 3) H = 8 Oe, 4) H = 40 Oe, 5) H = 60 Oe, 6) H = 128 Oe.

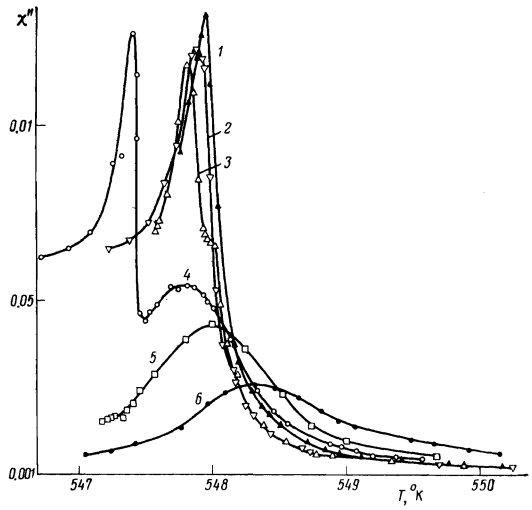

FIG. 5. Temperature dependence of the imaginary part of the susceptibility for a sphere, at a frequency of 1 MHz in a stationary external magnetic field H: 1) H = 0, 2) H = 3.4 Oe, 3) H = 8 Oe, 4) H = 40 Oe, 5) H = 60 Oe, 6) H = 128 Oe.

is a variable quantity, depending on the value of the susceptibility.) It may be suggested that the difference in the values of χ' for the sphere and the cylinder at $\xi < 8 \times 10^{-4}$ is related either to the appearance of anisotropy in the immediate vicinity of T_C in the paramagnetic phase, or to the failure of the Curie point to coincide with the temperature at which the peak magnetic loss is observed.

When $\omega\tau \ll 1$ (ω is the angular frequency) it can be assumed that the dynamic susceptibility is approximately equal to the static value $\chi'(0)$, i.e., $\chi' \approx \chi'(0)$. It is clear from Fig. 3 that in this case the temperature dependence of the susceptibility agrees with that predicted by the static theory of similarity, namely $\chi'(0) \sim \xi^{-\gamma}$, where $\gamma = 1.33$. Our measurements gave $\gamma = 1.30 \pm 0.04$. When the condition $\omega\tau \ll 1$ is no longer satisfied, the curves of χ' for different frequencies begin to diverge. This divergence vanishes in the ferromagnetic phase at $\xi > 8 \times 10^{-4}$.

The behavior of the spin system changes when a stationary external magnetic field H is applied (Figs. 2, 4, and 5). In weak fields the loss peak is shifted in the direction of lower temperatures. This shift in the transition point is evidently related to the stability of a state of inhomogeneous magnetization in an isotropic ferromagnet[7]. Above some value of the external field (around 40 Oe) the loss peak begins to shift in the direction of higher temperatures. (The existence of a "critical" field is clearly illustrated by the temperature dependence of χ' and χ'' in this field (see curve 4 in both Fig. 4 and Fig. 5).) This transition from one direction of loss-peak shift to the other may be related to the transition from a state of inhomogeneous magnetization of the sample to the state of homogeneous magnetization.

It must be noted that the susceptibility of YIG in the phase-transition region has been investigated by a number of authors[8-10]. However, the lack of data on the relaxation time and the impossibility of comparing the detailed behavior of the susceptibility because of different degrees of temperature stabilization, a number of physical differences (higher frequencies, such that $\omega\tau$ approaches 1, or polycrystalline samples), and the influence of sample shape in the immediate vicinity of T_C, makes it extremely difficult to compare the results of our work with the results by others.

An analysis of our results indicates that they are in qualitative agreement with the current theoretical concepts of the hydrodynamic region. The temperature dependence of the relaxation time is in fact observed to be nonmonotonic; in addition, the relaxation time depends on the sample shape as predicted by Vugal'ter and Maleev[4], and in particular, in the far paramagnetic region where we can assume that $\chi_\perp(0) = \chi_\parallel(0)$, $\tau_{sph} = \tau_{cyl}$. However, at $\xi < 1 \times 10^{-3}$ τ_{sph} becomes small compared to τ_{cyl} at the same relative temperature. At $\xi < 6 \times 10^{-4}$ it becomes difficult to compare samples of different shapes, because $4\pi\chi'$ is close to 1, and in general Eq. (2) cannot be used to determine the relaxation time. Unfortunately, at temperatures in the immediate vicinity of T_C ($\xi < 6 \times 10^{-4}$) there are as yet no means of calculating the relaxation time from the dynamic susceptibility data. Despite this fact, we note that the τ_{max} calculated from Eq. (2) occurs at the same temperature as the peak magnetic loss, i.e., at the temperature that we chose as the Curie point.

Thus, measurements of the radio-frequency susceptibility are an effective means of studying the dynamics of a spin system near the phase transition.

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111