Behavior of nonlinear dynamic susceptibility above the Curie point of cubic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$

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(Submitted 11 May 1984)
Zh. Eksp. Teor. Fiz. **87**, 2129–2141 (December 1984)

An experimental investigation was made of the behavior of the nonlinear dynamic susceptibility in the region of a phase transition in cubic weakly anisotropic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$. It was found that above the Curie point T_C there were two temperature ranges, scaling and anomalous, differing in respect of the nature of the critical phenomena. In the scaling range, corresponding to $4\pi\chi_0 \lesssim 25$, the observed behavior of the linear and nonlinear susceptibilities was in satisfactory agreement with the predictions of a theory based on similarity (scaling) considerations. The anomalous range adjoined directly T_C. In this range there were phenomena which could not be explained by the current ideas on the nature of second-order phase transitions and they included hysteresis on the magnetic field scale, a "residual" second harmonic signal which did not disappear for a long time after the external static field was switched off, and anomalous behavior of the susceptibility at low frequencies. The phenomena observed in the anomalous region were similar to those encountered in spin glasses.

Nonlinear properties of the dynamic susceptibility of ferromagnets near the Curie point have not yet been investigated in detail either experimentally or theoretically.

We shall report the results of the first experimental study of the behavior, in the paramagnetic phase, of higher harmonics of the magnetization induced by an external alternating magnetic field. Let us consider this problem first in greater detail. Consider a sample kept at a temperature $T > T_C$ and subjected to an alternating magnetic field $h = h_0 \sin\omega t$. We assume that the demagnetization factor of this sample is zero. Therefore, when the field amplitude is sufficiently low, the magnetization can clearly be represented as a series

$$m(h) = \chi_0(\omega)h + \chi_2(\omega)h_0^3 + \chi_4(\omega)h_0^5 + \dots . \quad (1)$$

Here, $\chi_0(\omega)$ is the usual dynamic susceptibility and $\chi_{2n}(\omega)$, where $n = 1,2,3\dots$, are nonlinear dynamic susceptibilities which can be expressed in terms of spin correlation functions of order $2n+2$. The corresponding expressions for the static case $\omega = 0$ can be found in Ref. 1. We shall consider below sufficiently low frequencies and use the static limit $\omega \to 0$ both for χ_0 and χ_{2n}. The dependences of χ_0 and χ_{2n} on the relative temperature $\tau = (T - T_C)/T_C$ are given by[1,2]

$$\chi_0 = \frac{b_0(g\mu)^2}{kT_C v_0}\tau^{-\gamma}, \quad \chi_{2n} = \frac{1}{\tau^\gamma} b_{2n}\frac{(g\mu)^2}{kT_C v_0}\left[\frac{(g\mu)^2}{kT_C}\frac{1}{\tau^{(5-\eta)\nu/2}}\right]^{2n}, \quad (2)$$

where the standard symbols for the g factor and the Bohr magneton μ are used; v_0 is the volume of a unit cell per one magnetic atom; b_0 and B_{2n} are constants; η is the Fisher exponent; ν and γ are the critical exponents for the correlation length and susceptibility, respectively.

The Fisher exponent is small and we shall ignore it. In the case of an isotropic ferromagnet, we have $\nu \approx 2/3$. In this approximation, the susceptibilities are described by

$$\chi_0 \propto \tau^{-4/3}, \quad \chi_2 \propto \tau^{-14/3}, \quad \chi_4 \propto \tau^{-24/3}, \dots . \quad (3)$$

It should be stressed that the expansion (1) is valid only on condition that the magnetic field amplitude is sufficiently small[1]:

$$g\mu h_0 \ll kT_C\tau^{5/3}. \quad (4)$$

Clearly, this fairly rigid condition must be satisfied when comparisons are made of experimental and theoretical results. In such a comparison it is essential to allow also for another important circumstance. As pointed out above, the expressions for χ_{2n} are currently available only for the static limit. Therefore, in experimental studies of the nonlinear susceptibility in terms of higher harmonics of the magnetization it is necessary to ensure also that the frequency of an alternating magnetic field is low compared with the characteristic energy of critical fluctuations. According to the current ideas (see, for example, Ref. 3), one can distinguish two temperature ranges depending on the relative importance of the dipole forces: $4\pi\chi_0 \ll 1$ (exchange range) and $4\pi\chi_0 \gg 1$ (dipole range). In the exchange range the characteristic energy of critical fluctuations is $\Omega_e \approx kT_C\tau^{5/3}$ and the reciprocal of the relaxation time of a homogeneous magnetization is governed by the dipole forces and is given by

$$\Gamma_H = \frac{(g\mu)^2}{v_0}\frac{(g\mu)^2}{kT_C v_0}\tau^{-1}. \quad (5)$$

In this case the susceptibility χ_{2n} is independent of ω on condition that

$$\omega \ll \Gamma_H \ll \Omega_e. \quad (6a)$$

In the dipole range the characteristic energy of critical fluctuations and the reciprocal relaxation time Γ_0 of the homogeneous magnetization are quantities of the same order and the amplitudes χ_{2n} are independent of ω on condition that

$$\omega \ll \Gamma_0. \quad (6b)$$

It is usual to consider theoretically two variants of the behavior of Γ_0 in the dipole range: normal or "soft," described by

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$$\Gamma_0=\text{const}\frac{(g\mu)^2}{v_0}\left[\frac{kT_Cv_0}{(g\mu)^2}\right]^{1/4}\tau^{4/3}, \tag{7a}$$

and "hard" described by

$$\Gamma_0=\text{const}\frac{(g\mu)^2}{v_0}\left[\frac{kT_Cv_0}{(g\mu)^2}\right]^{1/4}\tau^{2/3}. \tag{7b}$$

It follows that the most stringent restrictions on ω have to be imposed in studies of the dipole range, because then in the limit $T\to T_C$ we have $\Gamma_0\to 0$ and, consequently, the condition (6b) is obeyed if $\omega\to 0$ on approach to the Curie point. In addition to the condition (4), this is a fundamental circumstance which must be allowed for in experiments of this kind.

It is clear from Eq. (3) that on increase in the harmonic number the dependence of χ_{2n} on τ becomes increasingly stronger and away from T_C the higher harmonics of the induced magnetization begin to fall rapidly in amplitude. For this reason in our experiments we were able to determine the behavior of the nonlinear susceptibility only in the dipole range.

EXPERIMENTS

Harmonics of the magnetization induced by an external alternating field will clearly give rise to (in a search coil wound on a sample) higher harmonics of the emf $E_{2n+1}(t)$. The corresponding expressions for these harmonics can easily be found from Eq. (1):

$$\begin{aligned}E_3(t)&=-A_3\cos 3\omega t\\&=-p\omega\eta_0[{}^3/_4\chi_2+{}^{15}/_{16}\chi_4h_0^2+\ldots]h_0^3\cos 3\omega t,\\E_5(t)&=A_5\cos 5\omega t=p\omega\eta_0[{}^5/_{16}\chi_4+\ldots]h_0^5\cos 5\omega t,\ldots,\end{aligned} \tag{8}$$

where the coefficient p is governed by the number of turns and by the geometry of the search coil and η_0 is the space factor of the coil; A_3, A_5, ... are the amplitudes of higher harmonics of the emf.

It follows that measurements of the amplitudes of higher harmonics of the emf induced in a search coil provide a direct method for investigating the nonlinear properties of the susceptibility. This was the method used in our experimental study of critical phenomena in cubic weakly anisotropic ferromagnets $CdCr_2S_4$ ($T_C\approx 84$ K) and $CdCr_2Se_4$ ($T_C\approx 128$ K). We used single-crystal samples shaped to form rings in order to aviod demagnetization effects: the outer and inner diameters and the thicknesses were 2.7, 1.5, and 0.3 mm in the case of $CDCr_2S_4$ and 5.4, 2.0, and 2.1 mm in the case of $CdCr_2Se_4$. The plane of the ring in both samples was parallel to a (111) plane. A single-layer toroidal coil with 36 turns was wound uniformly on each sample. The same coil was used to measure the higher harmonic signal and to apply a magnetic field to a sample.

In the case of a toroidal coil the space factor could be determined only approximately from the dimensions of the sample and the volume inside the coil. Therefore, the absolute value of the susceptibility was found subject to a systematic error associated with the inaccuracy of the determination of the absolute value of the self-inductance of the coil and of the space factor (estimates gave $\eta_0\approx 0.5$ for cadmium chromium sulfide and $\eta_0\approx 0.7$ for cadmium chromium selenide).

The frequency and amplitude of the alternating field were selected on the basis of the conditions (4), (6a), and (6b). Measurements were carried out at frequencies 12.8 and 25.65 kHz satisfying the condition $\omega\ll\Gamma_0$. In fact, according to Ref. 4, in the case of $CdCr_2S_4$ in the dipole range the value of Γ_0 varied from 1×10^8 Hz for $\tau\approx 1\times 10^{-2}$ to 1×10^6 Hz for $\tau\approx 1\times 10^{-4}$. An estimate of the critical field from the conditon (4), i.e., from $g\mu h_{00\,\text{cr}}=kT_C\tau_{5/3}$, gave $h_{0\,\text{cr}}\approx 0.1$ Oe for $\tau=1\times 10^{-4}$ and $h_{0\,\text{cr}}\approx 200$ Oe for $\tau=1\times 10^{-2}$. These estimates indicated that the range of permissible values of the magnetic field was fairly wide ranging from a few millioersted (when measurements were made near T_C) to tens of oersted at temperatures sufficiently far from the Curie point. However, when the measuring coil was used also to create the field in the sample, there was a further constraint on the value of the field associated with the power dissipated in the coil and this was additional to the condition (4). Heating of the coil by the current could naturally alter considerably the temperature field near the sample and, therefore, result in a difference between the readings of the measuring thermometer and the temperature of the sample. For these reasons the maximum amplitude of the alternating field did not exceed 3 Oe. For this value of h_0 the higher harmonics were measured in the range $\tau<1\times 10^{-3}$. Estimates indicated that the power which was then dissipated in the coil was approximately 1×10^{-4}W, which should not affect significantly the results of our measurements. In fact, the thermal power in the thermostat needed to raise the temperature of the sample from 77.5 K (which was the temperature of the outer jacket of the thermostat) to temperatures corresponding to the critical region was approximately 0.3 W in the case of $CdCr_2S_4$ and 3 W in the case of $CdCr_2Se_4$, so that the maximum change in the temperature of the sample caused by the power 1×10^{-4}W dissipated in the coil did not exceed 5×10^{-3} K, which was clearly unimportant in the range $\tau<1\times 10^{-3}$. On closer approach to T_C the measurements were carried out in much weaker fields ($h_0<0.1$ *Oe*) so that in this case again we could ignore the change in the temperature field near the sample.

The magnetic field created in the coil was found from the coil geometry, number of turns, and value of the current. The relative stability of the voltage generated in a fundamental-frequency oscillator was at least 1×10^{-4}. The magnetizing current was determined to within 0.1%.

Sufficiently far from T_C the signal due to the higher harmonics was 10^3–10^5 times less than the signal at the fundamental frequency. Therefore, it was necessary to adopt a number of measures in order to eliminate stray signals that were unrelated to the quantities being measured. With this in mind the relative content of the higher harmonics in the initial fundamental-frequency current was reduced to 3×10^{-6} by filters, and overloading of a selective amplifier was avoided by placing band-rejection filters with an attenuation factor of at least 10^3 at the fundamental frequency. A block diagram of the circuit used to measure the signals due to the higher harmonics was given in Fig. 1. A resistance R_0 was selected to be sufficiently high to satisfy $R_0\gg\omega L_x$ (L_x is the

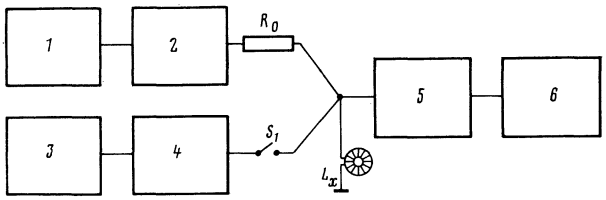

FIG. 1. Block diagram of the apparatus used in measurements of the higher harmonic signals: 1) oscillator supplying the voltage at the fundamental frequency f_0: 2) low-pass filter; 3) variable source of a static current; 4) low-pass filter with a high internal impedence; 5) band-rejection filter tuned to the fundamental frequency f_0; 6) selective microvoltmeter; R_0 is a limiting resistor; L_x is a measuring coil with the investigated sample; S_1 is a switch controlling the current responsible for the magnetization.

inductance of the measuring coil and the sample). Therefore, the measured harmonic voltage was practically equal to the emf of this harmonic. However, it should be pointed out that we determined only the amplitude of the higher harmonic and did not analyze its phase. In processing the data we used the relationship $A_m^2 = A_t^2 + A_n^2$, where A_m is the measured quantity, A_t is the true value of the harmonic signal, and A_n is the intrinsic noise of the selective amplifier (in our experiments the value of A_n was at least 0.1–0.2 μV). The experimental error did not exceed 2% when the signal was less than 2 μV; when the signal was stronger, the error decreased to 0.5%. The error in the determination of A_t clearly depended on the relationship between A_m and A_n.

We used a thermostat described in detail in Ref. 4. We also considered inRef. 4 the selection of the Curie point. In the present investigation we found T_C from the position of the maximum of the third harmonic signal with an error of $(10–15)\times 10^{-3}$K. The temperature fo the sample was stable to within $(2–3)\times 10^{-3}$ K.

The earth's magnetic was attenuated by Permalloy screens. The residual field was directed at some angle to the plane of the ring sample and its magnitude did not exceed 10–15 mOe.

RESULTS

The results of our investigation of the behavior of the higher magnetization harmonics indicated that at temperatures $T > T_C$ we could distinguish two ranges, scaling and anomalous, in which the critical phenomena were of different nature. This conclusion was in agreement with the results obtained by us earlier[4] in a study of the dynamic susceptibility above T_C.

Scaling range

The scaling range for the static susceptibility was between $4\pi\chi_0 \ll 1$ and $4\pi\chi_0 \approx 25$, i.e., it covered both the exchange and dipole ranges discussed earlier. The value $4\pi\chi_0 \approx 25$, considered in the limit of a systematic error in the static susceptibility, corresponded to a relative temperature $\tau_b \approx 1\times 10^{-2}$ for $CdCr_2S_4$ and $\tau_b \approx 3\times 10^{-3}$ for $CdCr_2Se_4$. This boundary of the scaling range was deduced from the change in the nature of the behavior of the investigated quantities as a function of ω and τ at the above values of τ_b.

The scaling range is so called because of the satisfactory agreement between the experimental results and those of a thoery based on static and dynamic similarity considerations. We shall consider below the behavior of the static susceptibility, critical damping coefficient Γ_0, and amplitude χ_2 associated with a four-particle spin correlation function in the scaling range.

1. Static susceptibility. Measurements of the linear susceptibility were carried out by a phase method[4] at frequencies satisfying the condition $\omega \ll \Gamma_0$. The dependence of χ_0 on τ obtained in the scaling range (Fig. 2) was of the power-law type with the critical exponent γ amounting to 1.38 ± 0.02 for $CdCr_2S_4$ and to 1.34 ± 0.01 for $CdCr_2Se_4$. In the scaling range these values of the critical exponent γ of the susceptibility, as well as the values of the static susceptibility itself, were very close to the results reported in Ref. 5, where the behavior of the static susceptibility in the critical region of these substances was deduced from measurements of he magnetization carried out using a vibration magnetometer.

2. The behavior of Γ_0 in the scaling range was determined for $CdCr_2S_4$ in our earlier study.[4] According to the results reported there, the value of Γ_0 in the exchange range increased on approach to the Curie point, in agreement with the theoretical predictions, and at $4\pi\chi_0 \approx 1$ the behavior of the homogeneous relaxation changed, whereas in the dipole range right up to $\tau \approx 1\times 10^{-2}$ the dependence $\Gamma_0(\tau)$ was close to the result of the "hard" variant of the dipole dynamics ($\Gamma_0^{\exp} \propto \tau^{0.8}$).

3. Behavior of higher dynamics of the magnetization was determined only in the dipole range (as already pointed out earlier). At relative temperature corresponding to the

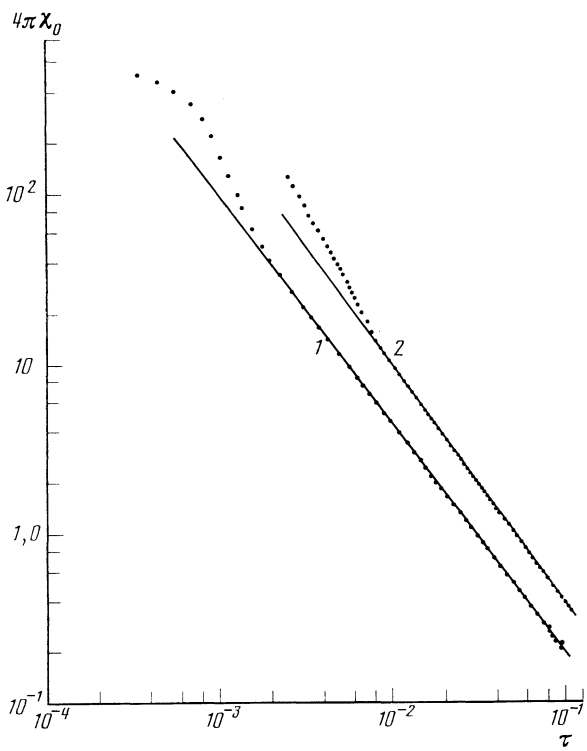

FIG. 2. Dependences of χ_0 on τ: 1) $CdCr_2Se_4$; 2) $CdCr_2S_4$.

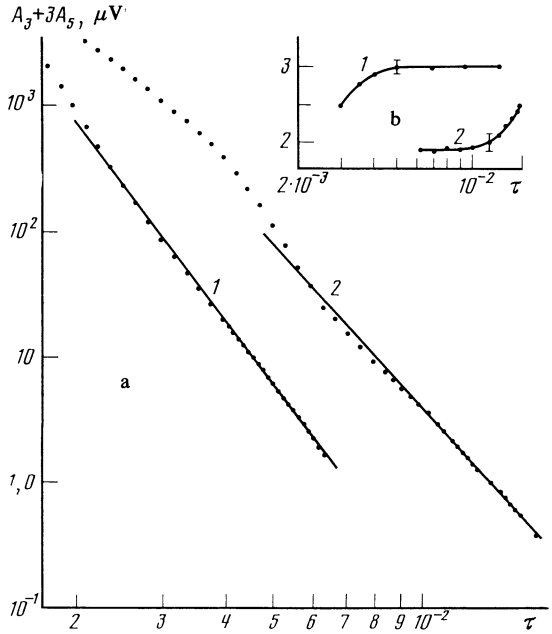

FIG. 3. a) Dependences of $A_3 + 3A_5$ on τ, which—according to Eq. (9b)—are identical with the dependence $\chi_2(\tau)$. b) Temperature dependences of the power exponent in the dependence of $A_3 + 3A_5$ on h_0: 1) $CdCr_2Se_4$; 2) $CdCr_2S_4$; $f_0 = 25.65$ kHz, $h_0 = 2.3$ Oe.

scaling range the higher harmonic signals were fairly small in the absolute sense and they were only slightly greater than the intrinsic noise level in the selective amplifier, amounting to 10^{-4}–10^{-5} of the fundamental-frequency signal. Therefore, we could determine reliably only the temperature dependences of the third and fifth harmonics. However, in the scaling range the amplitudes of these harmonics were relatively close to one another (for example, measurements carried out in a field $h_0 = 2.3$ Oe indicated that $A_5 \approx 0.3A_3$ for $CdCr_2S_4$ and $A_5 \approx 0.1A_3$ for $CdCr_2Se_4$ when $\tau = 1\times10^{-2}$) and we could expect the harmonics with higher numbers to have relatively larger amplitudes. Clearly, one would have to allow then for the second and higher terms in Eq. (8). However, because of the absence of data on the amplitudes of the higher harmonics we had to confine ourselves to the second term inthe expression for the amplitude of the third harmonic:

$$A_3 = -p\omega\eta_0[{}^3/_4\chi_2 + {}^{15}/_{16}\chi_4 h_0^2]h_0^3, \quad A_5 = {}^5/_{16}p\omega\eta_0\chi_4 h_0^5, \quad (9a)$$

and hence

$$A_3 + 3A_5 = -{}^3/_4 p\omega\eta_0\chi_2 h_0^3. \quad (9b)$$

The temperature dependences of the third and fifth harmonics in Eq. (9b) were used to determine the dependence of $(A_3 + 3A_5) \propto \chi_2$ on τ (Fig. 3a). In the scaling range this dependence was a power law for both substances and the exponents were 4.7 ± 0.2 for $CdCr_2S_4$ and 5.2 ± 0.2 for $CdCr_2Se_4$, quite close to the expected value $10/3 + \gamma \approx 4.7$ [see Eq. (2)]. The following comment should be made. It is clear from Eq. (9b) that a quantity $A_3 + 3A_5$ should be proportional to h_0^3, which is the criterion used in a comparison of the experimental and theoretical data. In the case of $CdCr_2Se_4$ this proportionality was obeyed throughout the scaling range (curve 1 in Fig. 3b) when the amplitude of the alternating field was altered threefold. However, in the case of $CdCr_2S_4$ the dependence of $A_3 + 3A_5$ on h_0 was close to quadratic (curve 2 in Fig. 3b) although in this case there was an almost complete agreement between the experimental and predicted exponents in the dependence $\chi_2(\tau)$. This type of dependence of $A_3 + 3A_5$ on h_0 and τ may possibly be due to a stronger than assumed influence of the higher terms ignored in the expressions for the amplitudes of the third and fifth harmonics. A more accurate determination of the temperature dependence of χ_2 would clearly require measurements of higher-order harmonics of the emf. On the other hand, the interval of relative temperatures in which the dependence $\chi_2(\tau)$ was determined was far too narrow to draw the final conclusions on the numerical value of the exponent in this dependence. However, we cannot exclude the possibility that the observed behavior of the higher harmonics is due to an earlier manifestation of the interactions that give rise to the anomalous range.

Anomalous range

Although the anomalous behavior of the dynamic susceptibility near T_C has been observed for a number of substances,[6–8] a detailed study of this effect was first carried out by us[4] for $CdCr_2S_4$. According to the results reported there, in the direct vicinity of the Curie point (at least in the range $\tau \lesssim 1\times10^{-3}$) a fairly strong dispersion of the real part of the susceptibility was observed in the low-frequency range and is was found that $\chi' \propto \log(1/\omega)$. The magnetic losses were fairly high and partically independent of the frequency. It was also noted in Ref. 4 that the dependence $\Gamma_0(\tau)$ observed near T_C differed from the theoretically predicted behavior of the characteristic energy of critical fluctuations in the dipole range.

An investigation of behavior of higher harmonics of the magnetization indicated that the signals due to these harmonics also behaved anomalously near the transition point. It is clear from Fig. 4 that the temperature dependence of the third harmonic signal obtained for both substances near T_C was definitely nonmonotonic.

We recall that the boundary between the scaling and anomalous ranges was deduced from a change in the nature of the behavior of the linear and nonlinear parts of the susceptibility. It is clear from Figs. 2 and 3a that at $\tau_b \approx 1\times10^{-2}$ in the case of $CdCr_2S_4$ and at $\tau_b \approx 3\times10^{-3}$ in the case $CdCr_2Se_4$ the dependences $\chi_0(\tau)$ and $\chi_2(\tau)$ began to lose their power-law nature; within the limits of the error in the determination of τ_b itself and of the absolute value of χ_0, the boundary of the transition to the anomalous behavior of the susceptibility corresponded to practically the same value of the static susceptibility for both substances: $4\pi\chi_0 \approx 25$. This boundary of the anomalous range is supported also by the observed sharp deviation of the dependence $\Gamma_0(\tau)$ from the power law at $\tau \approx 1\times10^{-2}$ (see Ref. 4).

Hysteretic and "residual" phenomena

In subsequent investigations of the phenomena in the anomalous range we used the method of even harmonics

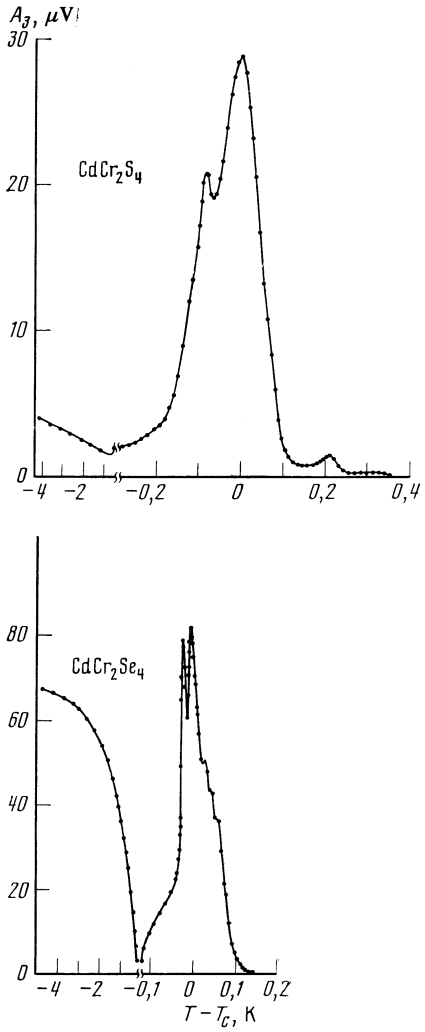

FIG. 4. Dependences of the third harmonic signal on $T-T_C$; $f_0 = 25.65$ kHz; $h_0 = 20$ mOe ($CdCr_2S_4$); $h_0 = 10$ mOe ($CdCr_2Se_4$).

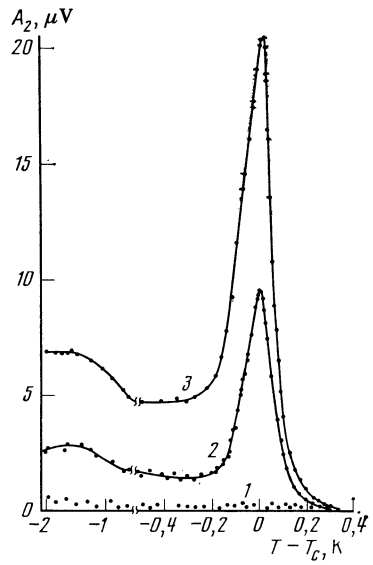

FIG. 5. Dependences of the second harmonic signal on $T-T_C$ obtained in different fields H_0 ($CdCr_2S_4$, $f_0 = 25.65$ kHz, $h_0 = 20$ mOe): 1) $H_0 = 0$; 2) $H_0 = 5$ mOe; 3) $H_0 = 20$ mOe.

which is essentially a very sensitive magnetometer. In fact, the even harmonics of the magnetization (and we shall speak later only of the second harmonic) appear only in the case when a static magnetic moment appears in a sample (irrespective of whether it is due to the spontaneous magnetization or is induced by an external static magnetic field). If a static field H_0 is applied parallel to an alternating field, then (in the first approximation) it follows from the expansion (1) that the emf due to the second harmonic is

$$E_2(t) = A_2 \sin 2\omega t = 3p\omega\eta_0 H_0 h_0^2 \chi_2 \sin 2\omega t. \quad (10)$$

It is clear from this expression that if $H_0 = 0$ then $A_2 = 0$, whereas if $H_0 \neq 0$, then both χ_2 and A_2 tend to infinity when T approaches T_C (naturally, subject to the criterion that the fields H_0 and h_0 are low). In fact, in the absence of a static magnetic field the second harmonic signal observed in our experiments was practically undetectable at temperatures $T > T_C$ or it only slightly exceeded the intrinsic noise level of the amplifier (curve 1 in Fig. 5), which could be due to insufficient screening of the terrestrial magnetic field. The low level of the second harmonic observed at $T < T_C$ was clearly due to the appearance in the ferromagnetic phase of regions with the opposite direction of the magnetization. In fields $H_0 \neq 0$ the second harmonic signal assumed its maximum value at the Curie point (curves 2 and 3 in Fig. 5). One should point out that, in contrast to the temperature dependence of the third harmonic signal, the dependence of A_2 on $T - T_C$ had no singularities at $T > T_C$. Experiments on the second harmonic were also carried out at frequencies 12.8 and 25.65 kHz in static and alternating magnetic field amplitudes satisfying the condition of their smallness given by Eq. (4).

The most interesting results were obtained in the following two series of experiments. In the first series carried out at a fixed temperature we determined the dependence of the second harmonic signal on the static magnetic field. This was done as follows. When a thermal equilibrium was established in a sample, a field of specific polarity was applied gradually (at a rate not exceeding 10^{-3} Oe/sec) and the dependence $A_2(+H_0)$ was recorded at a fixed amplitude of an alternating field while the static field was varied from $H_0 = 0$ to $H_{0\max} \approx 0.1$ Oe, followed by a return to zero static field. Next, the polarity of the field was reversed and the dependence $A_2(-H_0)$ was recorded in the same way. It should be pointed out that on approach to T_C, in accordance with Eq. (10), the amplitude of the second harmonic was found to be proportional to the static field and independent of its polarity, and it vanished for $H_0 = 0$. This behavior of the second harmonic was characteristic both of the scaling and anomalous ranges when τ was comparable with τ_b. However, beginning from $\tau \approx 4\times10^{-3}$ ($T - T_C \approx 0.30$–0.35 K) for $CdCr_2Se_4$ and $\tau \approx 1.4\times10^{-3}$ ($T - T_C \approx 0.15$–0.20 K) for $CdCr_2Se_4$, we observed hysteretic phenomena (Fig. 6c), i.e., variation of the static field caused the second-harmonic signal to disappear at some value H_C different from zero; for both substances the temperatures of appearance of hysteresis corresponded to $4\pi\chi_0 \approx 40$–50. This value of the static susceptibility was obtained as a result of direct extrapolation

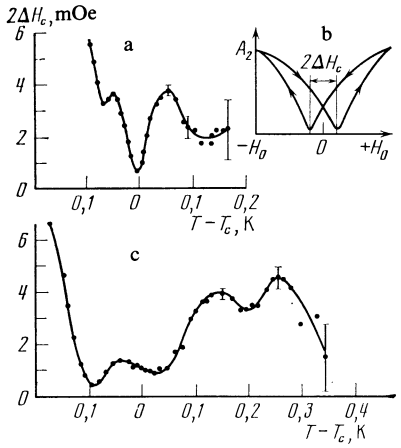

FIG 6. Dependences of the "coercive force" $2\Delta H_c$ on $T - T_C$ obtained from the experimental values of the second harmonic signal determined in a static magnetic field H_0 ($f_0 = 25.65$ kHz $h_0 = 20$ mOe) applied to $CdCr_2Se_4$ (a), and $CdCr_2S_4$ (b), and the dependence $A_2(H_0)$ illustrating hysteretic phenomena in the anomalous range (c).

to the anomalous range of the dependence $\chi_0(\tau)$ found experimentally for the scaling range. By analogy with hysteretic phenomena in ferromagnets, the value of $2\Delta H_C$ could be called the "coercive force." It was found that the value of $2\Delta H_C$ itself depended on the amplitude of the alternating field: it decreased, when h_0 was increased. The dependences of $2\Delta H_C$ on $T - T_C$ obtained for both samples were nonmonotonic with a minimum near T_C (Figs. 6a and 6b). The coercive force began to rise as we moved away from the Curie point in the ferromagnetic phase range and already at $T - T_C = -2$ K it was several times greater than the value of $2\Delta H_C$ obtained at $T > T_C$. It should be pointed out that hysteretic phenomena were also exhibited by the third harmonic signal. On the other hand, there was no hysteresis of the linear part of the susceptibility (within the limits of the experimental error).

The second series of experiments was carried out as follows. When the static and alternating magnetic fields were switched off, a sample was heated to temperatures corresponding to the scaling range. Then, only the static field $H_0 \approx 0.1$ Oe was applied and in this field the sample was cooled to a given temperature. When a thermal equilibrium was reached in a sample, the static field was switched off (its value fell from 0.1 Oe to zero in a time not exceeding 10^{-3} sec). After 1–2 min, the dependence of the second harmonic signal on the amplitude of the alternating field was determined by recording first $A_2(h_0)$ while the field amplitude was increased from $h_0 = 0$ to $h_{0\,max} \approx 0.3$ Oe (forward run); then h_0 was reduced from $h_{0\,max}$ to zero and the reverse run of the dependence $A_2(h_0)$ was recorded (in both cases the rate of variation of h_0 did not exceed 10^{-3} Oe/sec). Next, the sample was heated again and the procedure for the determination of the dependence $A_2(h_0)$ during the forward and reverse runs was repeated at a new value of the equilibrium temperature. In control experiments carried out in the absence of a static magnetic field a similar procedure was used to determine the "background" dependences of the second harmonic signal on the alternating field.

Among the results of this series of experiments we should point out the following.

Firstly, a residual signal was observed. The dependence $A_2(h_0)$ (curve 1 in Figs. 7a and 7b) obtained during the forward run after the static field was switched off indicated that the second harmonic signal was considerably higher than the signal during the reverse run. One should point out that the

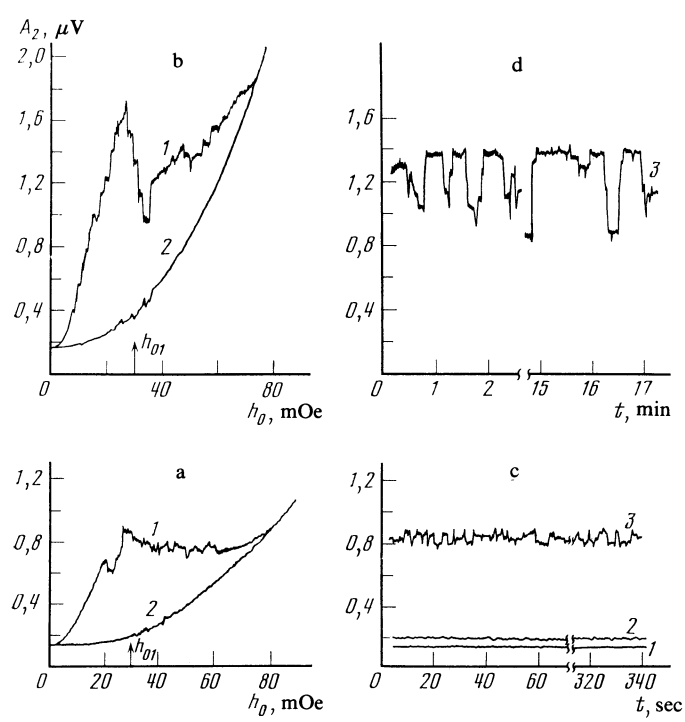

FIG. 7. a), b) Dependences of the second harmonic signal on the amplitude of the alternating magnetic field (curve 1 represents the forward run and curve 2 the reverse run). c), d) Time dependences of the second harmonic signal: 1) intrinsic noise level in the selective amplifer; 2) second harmonic signal during the reverse run; 3) "residual" signal (forward run) obtained at different relative temperatures: $\tau = 1.2 \times 10^{-3}$ (a,c) and $\tau = 4.8 \times 10^{-4}$ (b,d); $f_0 = 25.65$ kHz $h_{01} = 30$ mOe.

dependence $A_2(h_0)$ recorded during the reverse run (curve 2 in Figs. 7a and 7b) coincided with the background dependence of A_2 on h_0 obtained in control experiments in which a sample was cooled in the absence of a static magnetic field. (The background level of the second harmonic signal was independent of the "direction" of variation of h_0.) However, the difference between the residual and background signals was important only when the alternating field amplitude satisfied the condition of smallness given by Eq. (4). In the range $h_0 > h_{0\,cr}$ the dependences $A_2(h_0)$ obtained in these two experiments were practically identical. It was also established that the residual signal level was independent, within a wide range, of the static field in which a sample was cooled.

It should be stressed that the residual signal effect was significant when the separation from T_C was the same as that in observations of hysteretic phenomena, i.e., when τ was $4\pi\chi_0 \approx 40$–50 for both samples. In contrast to the temperature dependence of the amplitude of the second harmonic (Fig. 5), obtained in a static applied field H_0, the temperature dependence of the residual signal for a fixed value of h_0, like the dependence of $2\Delta H_C$ on $T - T_C$, was nonmonotonic with a minimum near the Curie point and a steep rise of the residual signal away from T_C in the ferromagnetic phase range.

Secondly, when the amplitude of the alternating signal was reduced from the maximum value to zero, the residual signal returned to its background value (curve 2 in Figs. 7a and 7b), as observed during demagnetization of ferromagnetic samples.

Thirdly, for certain amplitudes of the alternating field the residual signal became unstable in time and exhibited abrupt changes (Figs. 7c and 7d). The amplitudes of the discontinuities and the intervals between them were governed by the proximity to T_C. At temperatures close to the temperature of appearance of anomalous phenomena the fluctuations of the residual signal level were fast and small (Fig. 7c). Near T_C the interval between the discontinuities could reach several tens of seconds and fluctuations of the residual signal level increased (Fig. 7d). This effect resembled the so-called Barkhausen jumps observed during magnetization switching in ferromagnetic cores. One should point out that when the static field was removed, there was no significant time decay of the residual signal, excluding the fluctuations of its level mentioned above. However, all these results were preliminary and the final conclusions on the time dependence of the residual signal could only be made after special investigations.

Although the nature of these anomalous phenomena was not yet clear, the observations obtained in two series of determinations of the dependences $A_2(H_0)$ and $A_2(h_0)$ allowed us to draw the conclusion that long-lived formations appeared above T_C and these were some kind of precursors of the future domain structure. No thermal hysteresis of the coercive force or of the residual signal level of the second harmonic were observed in these experiments. Similar "memory" phenomena above T_C were encountered also in a study of the critical behavior of the dynamic susceptibility in static magnetic fields applied to spherical single-crystal samples of yttrium iron garnet.[9] Clearly, these phenomena were also due to the appearance of some formations at $T > T_C$. It is possible that formations or states of this kind occur in many ferromagnetic substances exhibiting a second-order phase transition. This hypothesis is supported by the anomalous behavior of the linear dynamic susceptibility observed near T_C for some ferromagnets (see, for example, Refs. 6–9).

CONCLUSIONS

The results of our investigation indicate a complex pattern of critical phenomena in the paramagnetic phase of weakly anisotropic cubic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$. The data obtained on the behavior of the linear and nonlinear susceptibilities above T_C suggest that in the range $T > T_C$ we can distinguish two ranges of values of the static susceptibility, scaling and anomalous, differing in respect of the behavior of the susceptibility. In the scaling range, corresponding to $4\pi\chi_0 \approx 25$, the observed behavior of the linear susceptibility, of the amplitude χ_2 associated with a four-particle spin correlation function, and of the critical dumping factor Γ_0 are in satisfactory agreement with the conclusions reached in a theory based on dynamic and static similarity considerations. In the range of temperatures adjoining directly the Curie point (anomalous range) the dynamic susceptibility behaves anomalously as a function of ω and τ and metastable formations of nature not yet understood may be postulated. The phenomena observed in the anomalous range are generally very similar to the situation encountered in spin glasses. In fact, the logarithmic frequency dependence of the real part of the susceptibility had hitherto been reported only for spin glasses[10] and the phenomenon of a residual second harmonic signal, observed in experiments involving cooling in a static magnetic field which is then switched off, are to some extent analogous to the thermoremanent magnetization characteristic of the spin glass state. The dipole forces play a special role in the phenomena occurring in the anomalous range. We recall that the boundary of the anomalous range of both investigated substances corresponds to practically the same value of the static susceptibility. The appearance of hysteretic and residual phenomena also occurs at a specific value of $4\pi\chi_0$. We cannot exclude the possibility that various defects, which are unavoidably present in real crystals, play some role in these anomalous phenomena. On the other hand, as pointed out above, the anomalous behavior of the susceptibility above T_C is typical of various ferromagnets and ferrimagnets. These observations suggest that a new so far unknown state of magnetic materials appears in such cases above the Curie point. Actually, the problem of the nature of the anomalous range will require further theoretical and experimental investigations.

The authors are deeply grateful to G. M. Drabkin and S. V. Maleev for their constant interest in this investigation and valuable discussions of the results. The authors are grateful to K. G. Nikiforov and T. G. Aminov for suppling the samples used in these investigations.

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Translated by A. Tybulewicz