Anomalous fluctuations of the second harmonic of the magnetization of $CdCr_2S_4$ near the Curie point

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Results are presented of investigations of anomalously large fluctuations of the magnetization second-harmonic residual signal observed near T_C in $CdCr_2S_4$ after cooling the sample in a constant magnetic field which is subsequently turned off. Random residual-signal oscillations were observed only in a definite interval of the alternating magnetic field amplitudes, with characteristic narrow bands in which the anomalous fluctuations practically vanish. A simple empirical law governing the onset of these bands is obtained. It is suggested that the anomalous fluctuations of the residual second-harmonic signal of the magnetization is of dynamic origin.

INTRODUCTION

It was shown earlier[1,2] that for the weakly anisotropic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$ it is possible to distinguish above T_C two temperature regions (named scaling and anomalous in Ref. 1) whose boundary corresponds to a static susceptibility $4\pi\chi_0 \sim 25$ for both substances. A feature of the scaling region ($4\pi\chi_0 < 25$) is a satisfactory agreement between the experimentally determined behavior of the linear and nonlinear susceptibilities and the predictions of the static and dynamic scaling theory. At $4\pi\chi_0 > 25$ (anomalous region), however, the observed behavior of the dynamic susceptibility as a function of the frequency and of the degree of deviation from T_C was unusual from the standpoint of the prevailing premises. It was found that in the range 10^2–10^5 Hz the imaginary part of the linear susceptibility depends little on the frequency, the temperature dependence of the amplitude of the third-harmonic magnetization is nonmonotonic, etc. As noted in Ref. 2, the anomalous region is similar in many of its properties to the spin-glass state. The nature of the anomalous phenomena, however is still unclear. It is possible that uncontrollable impurities and various types of defects, which are inevitably present in real crystals, can play a decisive role in the onset of the anomalous region. Nonetheless, notwithstanding the absence at present of a clear understanding of the nature of the anomalous region, it is promising to study the interesting, at times unique, physical properties observed in this temperature region.

One of the interesting phenomena observed in the anomalous region is the formation, above T_C, of a "residual" second-harmonic magnetization signal, although it follows from symmetry considerations that there should be no even harmonics in the spectrum of the magnetization induced in a ferromagnet by an alternating magnetic field $h = h_0 \sin \omega t$. Experiments,[1] however, have shown that at $\tau < 1\cdot 10^{-2} (\tau = (T - T_c)/T_c$ the second harmonic of the EMF induced in a measuring coil by applying to the sample a combined constant and alternating magnetic field remains different from zero after the constant field is turned off, and the residual signal exceeds substantially the second-harmonic background signal due to the incomplete shielding against the earth's magnetic field ($H_{res} \sim 10$–20 mOe). Since the even harmonics differ from zero only when the sample is in a magnetized state, it was natural to identify the residual second-harmonic signal with the remanent magnetization produced in the sample following the action of the constant magnetic field.

The most remarkable feature of the behavior of the remanent magnetization in an alternating magnetic field is the rather strong fluctuation of the second-harmonic residual signal level, which exceeds considerably the amplitude of the noise band of the selective amplifier. It is natural to assume that the anomalously large fluctuations of the residual signal are of dynamic origin. Owing to higher-order spin correlations, a ferromagnet near T_C is a strongly nonlinear medium.[3] Obviously, in this case, a ferromagnet placed in an alternating magnetic field can be regarded as a dynamic nonlinear system acted upon by a harmonic driving force. On the other hand, it is known that under the action of a driving force nonlinear system can exhibit a stochastic behavior. It is not excluded therefore that the observed fluctuations of the residual magnetization were due to dynamic chaos.

To pursue the phenomenon, which in our opinion is of interest, experiments were set up, aimed primarily at investigating the temporal behavior of the residual second-harmonic signal (fo the remanent magnetization) in a wide range of amplitudes of the alternating magnetic field. We report here the results of these experiments.

EXPERIMENT

Investigations of the temporal behavior of the second-harmonic residual-signal level were carried out on undoped ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$,[1)] which have a cubic magnetic symmetry (the respective anisotropy constants are $K_1 = 6\cdot 10^3$ and $K_2 = 1.5\cdot 10^3$ erg/cm^3). The single-crystal samples were rings with inside and outside diameters and heights 2.7, 1.5, and 0.3 mm ($CdCr_2S_4$) or 5.4, 1.0, 1.2 mm ($CdCr_2Se_4$), respectively, cut so that the ring planes coincided with the (111) plane. The initial single crystals (with resistivity not more than 10^6 $\Omega\cdot$cm) were of perfect form. Attesting to the magnetic homogeneity of the sample can be the fact that the temperature dependence of the second-harmonic amplitude, obtained on application of a sufficiently weak constant mangetic field, had only one maximum at

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$T = T_C$. Note that the Curie point in are experiments was detemined from the temperature position of the maximum of the third-harmonic amplitude in the absence of a constant magnetic field (see Ref. 2 for details). As a control, experiments were performed with two likewise undoped $CdCr_2Se_4$ samples taken from one batch. The control experiments have shown that the values of T_C differed by not more than 0.02 K, and the temperature values of the anomalous region, determined in this case from the positions of the singularities in the temperature dependence of the third-harmonic amplitude, were the same for both samples.

The block diagram of the measurement setup is shown in Fig. 1. The current (at the fundamental frequency f) that determines the amplitude of the alternating magnetic field in the sample was set by an attenuator and a limiting resistor. The voltage picked off the measuring coil was fed to a selective amplifier tuned to the frequency $2f$, whose internal noise did not exceed 0.3 μV. The investigated signal from the output of the amplifier passed through a converter (with an integration constant not larger than 0.5 s) to an automatic x-y plotter (with a recording time constant not larger than 0.05 s), used to plot the residual signal as a function of either the alternating magnetic field amplitude or the time. Note that we measured in the experiments the maximum amplitude of the second harmonic without analysis of its phase. The constant magnetic field, turned on during the cooling of the sample, was produced by the same induction coil, fed in this case by a dc source. The thermostat is described in detail in Ref. 2.

Two important circumstances had to be taken into account in these experiments. First, the amplitude of the residual signal A_{20} is small both in absolute value (A_{20} does not exceed several μV), and compared with the fundamental-frequency voltage A_0 picked off the measuring coil. In our experiments the minimum ratio A_{20}/A_0 was approximately 10^{-4}. Consequently, the relative content of the second harmonic in the initial voltage of the fundamental-frequency generator should not exceed 10^{-5}. This requirement was met by connecting to the output of the generator an LC filter F_2. Second, the residual signal is small also compared with the third-harmonic amplitude. And since the third-harmonic emf increases with increase of the ac field amplitude more rapidly than that of the second ($A_{3w} \propto h_0^3$ as against $A_{2w} \propto h_0^2$), this circumstances obviously becomes important

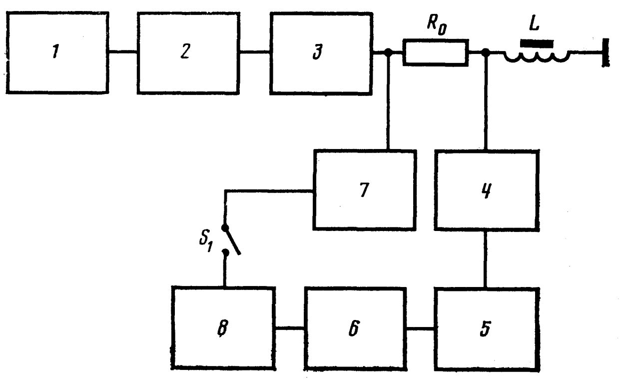

FIG. 1. Block diagram of the measurements: 1—fundamental-frequency generator; 2—attenuator; 3—LC filter F_2; 4—RC filter $F_{1,3}$; 5—selective amplifier; 6, 7—ac to dc voltage converters; 8—automatic plotter; R_0—ballast resistor; L—measuring coil with sample.

in measurements made in a wide range of field amplitudes. To prevent overloading the amplifier with large (compared with the residual) signals of frequency f and $3f$, we used an RC rejection band filter that suppressed the signals of the fundamental and of the third harmonic by not less than a factor of 10^3.

The procedure used to investigate the behavior of the remanent magnetization in an alternating field was similar in many respects to the one used in Ref. 1. The sample was also overheated to relative temperatures $\tau \geqslant 1 \cdot 10^{-2}$. A constant magnetic field $H_0 = 0.1$ Oe was then turned on, and the sample was cooled in the absence of an alternating field to the specified temperature. This value of H_0 was determined by the fact that starting with $H_0 > 0.03$ Oe the residual signal was independent of the constant field in which the sample was cooled. When thermal equilibrium was reached, the constant field was turned off and the dependence of the residual signal on the amplitude of the alternating field was determined, with h_0 varied first from 0 to $h_0^{\max} = 0.35$ Oe (direct variation), and then with the field amplitude decreased to zero (reverse variation). The rate of change of h_0 did not exceed as a rule 10^{-3} Oe/s. Control $A_{20}(h_0)$ were similarly obtained for the direct and reverse variations when cooling without a constant magnetic field. After the end of these measurements the sample was again overheated to high temperatures, cooling was repeated in a constant field to the same temperature at which the dependence of A_{20} on h_0 was determined, and after establishment of thermal equilibrium we started the investigation of the time dependence of the residual-signal level at various alternating-field amplitudes. The field amplitude was gradually increased from 0 to 0.35 Oe in steps of 1–5%, and at each value of h_0 the automatic plotter recorded, over an observation time from 5 to 60 min, the time dependence of the residual-signal level. These investigations were carried out at different temperatures both above and below T_C.

RESULTS

The main results of the investigation of the behavior of the remanent magnetication in alternating magnetic field were obtained in experiments with $CdCr_2S_4$ ($T_C \approx 84$ K) at a fundamental frequency 26.65 khz. Remanent magnetization and the anomalous fluctuations of the second-harmonic level were observed also in $CdCr_2Se_4$, but no detailed investigations of the behavior of the remanent magnetization of this substance were carried out.

We note first that the residual second-harmonic signal appeared in the anomalous region at $\tau \approx 4 \cdot 10^{-3}$ ($\Delta T = 0.3 - 0.4$ K), i.e., at the same temperature as the start of the hysteresis in the constant magnetic field and the start of the nonmonotonic temperature behavior of the third-harmonic signal.[1]

We begin the analysis of the resultant behavior of the remanent magnetization with the dependence of the residual signal itself on the alternating-field amplitude. Examples of this dependence of different deviations from T_C are shown in Fig. 2. Curves 1 and 2 in this figure pertain to the dependences obtained respectively when h_0 increases from 0 to $h_0^{\max}$ (direct variation) and when h_0 is decreased from $h_0^{\max}$ again to zero (reverse variation). Analysis of the plots obtained has shown that, first, at small field amplitudes we

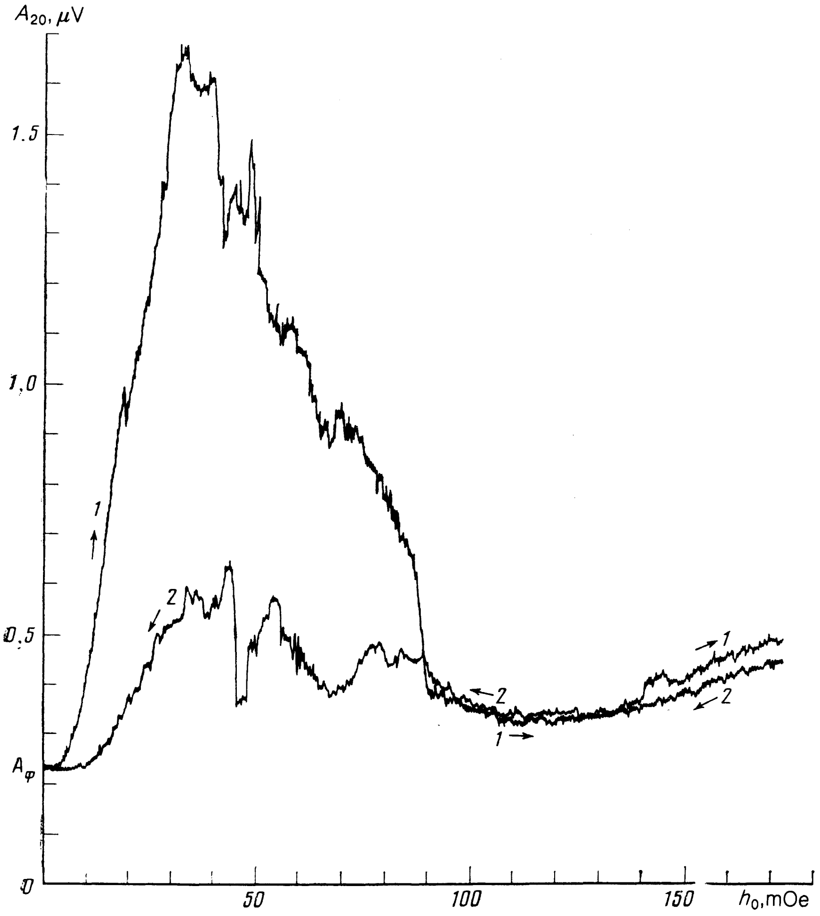

FIG. 2. Residual second-harmonic signal (in μV) vs the alternating magnetic field amplitude (in mOe): 1—forward dependence; 2—reverse dependence; A_f—level of intrinsic noise of amplifier; $\tau = 1\cdot 10^{-4}$.

have $A_{2\omega} \propto h_0^2$. This, generally speaking, was the expected result. Indeed, if the field amplitude is so small that the following condition is met[4]

$$g\mu h_0 \ll kT_C\tau^{5/3}, \qquad (1)$$

where g is the Landé factor and μ the Bohr magneton, the expression for the field-induced magnetization can be represented by a series in odd powers of h_0. If, however, a constant magnetic field collinear with the alternating one is applied to the sample, the magnetization spectrum acquires also even harmonics. In the case of a weak field, the expression for the second-harmonic emf is, in first-order approximation,

$$\varepsilon_2(t) = A_{2\omega}\sin 2\omega t = 2p\omega\chi_2 h_0^2 H_0 \sin 2\omega t. \qquad (2)$$

Here p is determined by the geometry and by the number of turns of the measuring coil, and χ_2 is the nonlinear susceptibility ($\chi_2 \propto \tau^{-14/3}$ in the case of an isotropic ferromagnet[4,5]). Thus, in fields that meet the weakness condition, the second-harmonic amplitude should be proportional to the square of the field amplitude. In the case of the residual signal, however, this law was satisfied only at very small field amplitudes. Starting with a certain value of h_0, the dependence of $A_{2\omega}$ on h_0 began to deviate noticeably from quadratic. Thus, at $\tau = 1.5\cdot 10^{-3}$ the deviation from relation (2) for the direct variation was observed starting with $h_0 \approx 15$ mOe, and at $\tau = 1\cdot 10^{-4}$ starting with $h_0 \approx 10$ mOe, whereas according to (1) the critical values of the field for the same τ are 10 and 0.1 Oe, respectively. Of course, the validity of condition (1) in the anomalous region is questionable. Experiment has shown, however, that the temperatures of the singularities in the $A_{3\omega}(T)$ dependence, including the maximum of $A_{3\omega}$ at T_C, remain unchanged if the field amplitude satisfies the smallness condition. If, however, the condition (1) is not met, the form of the temperature dependence of the third-harmonic signal changes, viz., the singularities at $T > T_C$ vanish, and the $A_{3\omega}$ maximum corresponding to the Curie point at small h_0 shifts towards lower temperatures. Note that the field interval in which the second-harmonic residual signal is proportional to the square of the field amplitude becomes narrower as T_C is approached. This temperature dependence, however, it weak and is apparently not monotonic.

In addition, the $A_{2\omega}(h_0)$ dependence is characterized also by the presence of a maximum whose value increases as $T \to T_C$ and whose position on the h_0 scale is practically independent of T at $T - T_C < 0.2$ K.

It should be noted, finally, that the forward and reverse $A_{2\omega}(h_0)$ variations are different. This difference is most noticeable at small field amplitudes. It has turned out that the inverse dependence of $A^{2\omega}$ on h_0 practically coincides with the control dependence obtained by cooling in the absence of a constant magnetic field. The nonzero residual second-harmonic signal in the control experiment is apparently due to insufficient shielding against the earth's magnetic field. Thus, at first glance the $A_{2\omega}(h_0)$ dependence has hysteresis properties. However, if one starts to increase the amplitude of the alternating field again from zero without turning on the constant one, the resultant "secondary" direct variation will not differ really from the reverse $A_{20}(h_0)$ variation. The reproducibility of the residual signal at one and the same field amplitude, except in the region of weak h_0, is quite poor, and the difference between the values of A_{20} for a fixed value of h_0 can reach 20%.

The most interesting turned out to be the time dependence of the residual second-harmonic signal. A distinction can be made between two temperature regions with different character of the fluctuations of the residual-signal level. In the interval $0.2 < T - T_C < 0.4$ K these fluctuations are negligible and barely exceed the noise corridor of the selective amplifer. At $T - T_C < 0.2$ K, however, the time dependence of the residual signal becomes qualitatively different: the residual-signal level begins to oscillate randomly at rather high amplitudes, and the character of these fluctuations is now determined by the value of the alternating field. Let us describe this phenomenon in greater detail, using as examples the time dependence of the residual signal at two temperature points: $\tau = 1.5 \cdot 10^{-3} (T - T_C = 0.12$ K) and $\tau = 1 \cdot 10^{-4} (T - T_C = 0.008$ K), see Figs. 3a and 3b. These figures show fragments of the time dependence of the level A_{20} at various h_0. On the left of each fragment is indicated the residual signal in μV, and on the right the alternating field amplitude, corresponding to this value of A_{20}, in mOe. At $T - T_C > 0.2$ K in the entire field-amplitude interval, and at $T - T_C < 0.2$ K only in weak fields, the fluctuations of the residual-signal level are small (see, e.g., $h_0 = 22.1$ mOe in Fig. 3a). With increase of field amplitude, however, the residual-signal fluctuations increase rapidly, and at certain values of h_0 random oscillations set in, and the residual signal itself and the amplitude of the oscillations of this level are independent, in a definite field-amplitude interval, of the value of h_0 (see $h_0 = (42.9–50.1)$ mOe in Fig. 3a and $h_0 = (17.7–19.4)$ mOe in Fig. 3b). Note that this field-amplitude interval is quite narrow ($\Delta h_0 / h_0 \approx 30\%$). Although the reproducibility of the values of h_0 at which the anomalously large fluctuations of the residual signal set in is quite good, we were nevertheless unable to determine the temperature dependence of the onset of this phenomenon. It can only be noted that at $\tau > 1 \cdot 10^{-2}$ the random oscillations set in at larger values of h_0 than in the immediate vicinity of T_C. With further increase of the field amplitude the character of the residual-signal flucturations changes somewhat (see $h_0 = 56.9$ mOe in Fig. 3a), and the residual signal itself becomes already dependent on the field amplitude. With further increase of the field, however, the residual-signal fluctution amplitude begins to decrease gradually to the noise value, and further increase of h_0 no longer leads to the onset of anomalous fluctuations. The foregoing can be illustrated for the dependence of the residual-signal fluctuation amplitude on the alternating-field amplitude (Fig. 4). A characteristic feature of this dependence is the presence of "stability windows," which are rather narrow field-amplitude intervals in which the fluctuations decrease abruptly in magnitude (see $h_0 = 51.3$ mOe in Fig. 3a and $h_0 = 20.3$ and 35.6 mOe in Fig. 3b). Note that the appearance of stability windows in the h_0 scale exhibits a simple empirical regularity

$$h_{n+1} = k h_n, \quad k = 1.28 \pm 0.05, \tag{3}$$

where n is the number of the window ($n = 1$ pertains to the first window corresponding to the smallest field amplitude

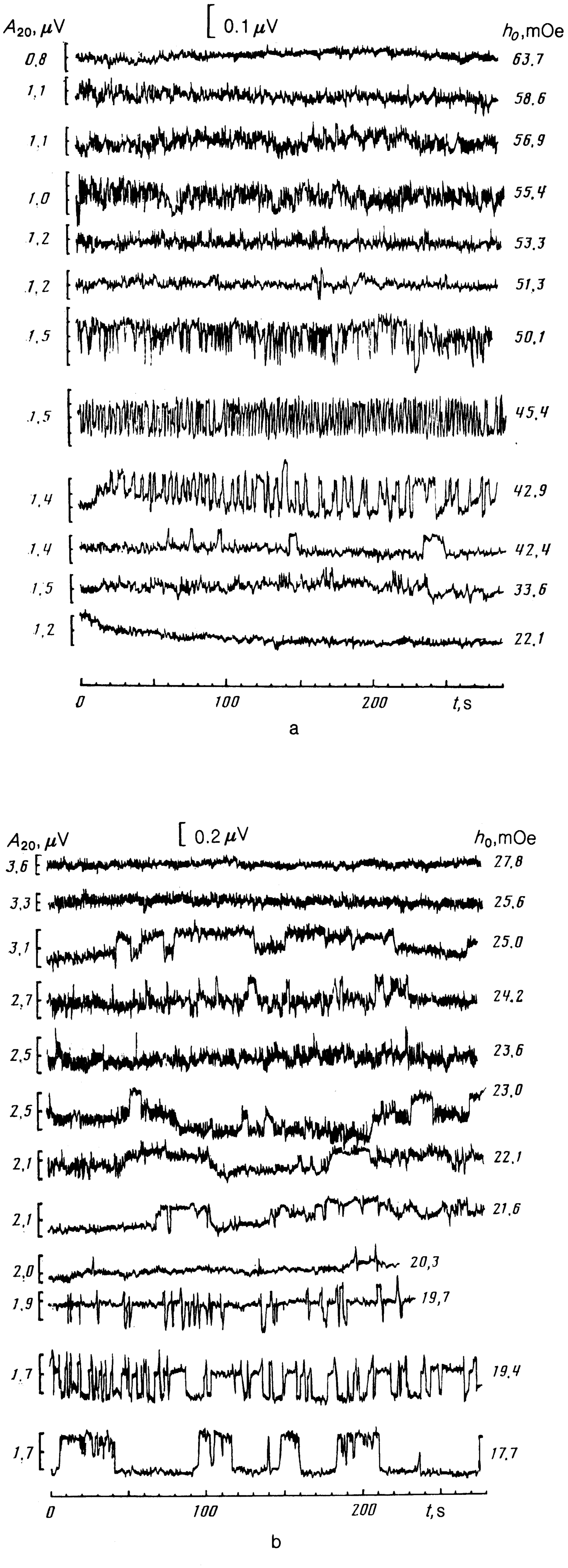

FIG. 3. Fragments of time dependence of the residual second-harmonic signal: *a* — $\tau = 1.5 \cdot 10^{-3}$; *b* — $\tau = 1 \cdot 10^{-4}$.

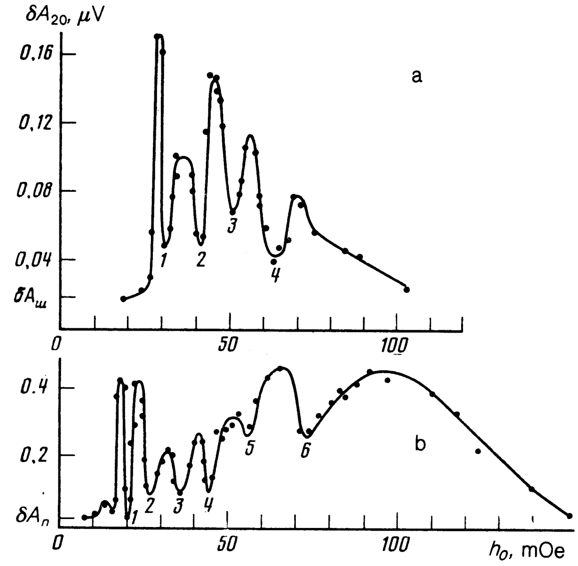

FIG. 4. Amplitude of residual-signal level pulsations vs alternating-field amplitude: a—$\tau = 1.5\cdot10^{-3}$, b—$\tau = 1\cdot10^{-4}$. The numbers label the "stability windows"; δA_n—selective-amplifier noise-interval amplitude.

at which the anomalous fluctuations decrease abruptly in magnitude).

The described evolution of the residual-signal fluctuations with change of the alternating-field amplitude pertain to the forward variation. The temporal behavior of the residual signal in the reverse variation was not investigated.

Thus, a feature of a large interval of alternating-magnetic-field amplitudes is the existence of the following: a) regions of h_0 in which the residual-level fluctuations of the second-harmonic are commensurable with the amplitude of the noise band of the selective amplifier, and b) field-amplitude regions with chaotic behavior of the residual signal, alternating with narrow h_0 zones in which the A_{20} fluctuations are as a rule insignificant. This type of behavior of the residual magnetization in an alternating field was observed in the entire anomalous region of temperatures above T_C. In the ferromagnetic phase, the anomalous fluctuation vanished at a rather large deviation from T_C (at $T - T_C \approx -1$ K), although the residual signal itself, at a fixed h_0, started to increase rapidly with deviation of T_C into the ferromagnetic phase. It can be stated that the described pattern of the behavior of the residual magnetization is almost symmetrical about the Curie point. The time dependence of the second-harmonic residual signal, however, was not investigated in detail for $T < T_C$.

CONCLUSION

Near the Curie point, a ferromagnet is a strongly nonlinear medium, and it is reasonable to search in the thermodynamic properties of a nonlinear system acted upon by a harmonic driving force for an explanation of the observed behavior of the remanent magnetization in an alternating magnetic field. To this end it would be necessary, obviously, to choose a suitable dynamic model and compare the results of the analysis with the experimental data. However, the lack of clarity of the nature of the anomalous temperature region in which the random oscillations of the second-harmonic residual signal were produced, prevents us from choosing a suitable model from carrying out at least a qualitative analysis. It is apparently possible therefore to discuss at present the observed phenomenon only by comparison with the behavior of dynamic systems with driving forces. The most suitable for this purpose are systems described by the Duffing equation[6]

$$\ddot{x}+\gamma\dot{x}+x^3=F_0 \sin \omega t, \tag{4}$$

where F_0 is the amplitude of the driving force and γ is an intrinsic parameter of the system.

An example of such a system is a series *RLC* circuit with nonlinear inductance,[7] which can apparently be used as an analog of our case. Analysis of dynamic systems described by Eq. (4), particularly the referred-to *RLC* circuit, carried out mainly by numerical methods, has shown that chaotic oscillations can set in at certain values of the intrinsic parameters and of the external-force parameters. As a rule, the stochastic regime is observed in a narrow range of the external-force parameter, and when they are varied the dynamic chaos can be interrupted in a number of cases by so-called stability windows, which correspond to harmonic oscillations. One might mention also such features of the behavior of dynamic systems as the transformation of the noise spectrum, doubling of the period with approach to chaos, and others (for more details on the results of the study of the behavior of dynamic systems and on the physical picture of the phenomena that evolve in them see, e.g., the reviews in Refs. 6 and 8). Our experiments, however, were not aimed at a search for these features.

Thus, the behavior of the remanent magnetization in an alternating magnetic field is similar in many respects to the behavior of a dynamic system acted upon by a driving force. In the present paper, however, we have confined ourselves, to an investigation of the actual fact of the onset of anomalous flucutations of the residual second-harmonic signal when only one parameter of the external force is changed, viz., the amplitude of the alternating field, and accordingly to the determination of the h_0 interval in which they were observed. More detailed investigations of the dynamic characteristics of this phenomenon are therefore necessary to draw final conclusions concerning the random nature of the remanent-magnetization fluctuations.

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[1)]The $CdCr_2Se_4$ samples were grown at the Institute of Organic and Inorganic Chemistry of the USSR Academy of Sciences, and the $CdCr_2S_4$ samples at the Institute of Applied Physics of the Moldavian SSR.

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