Second-harmonic nonlinear response of a cubic ferromagnet in the critical paramagnetic neighborhood of T_c

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We present the results of an experimental and theoretical investigation of the second-harmonic nonlinear longitudinal response of the magnetization M_2 of the cubic ferromagnet $CdCr_2Se_4$ in the critical paramagnetic neighborhood of T_c when the latter is placed in a constant magnetic field. The experimental data were obtained for the phase components of M_2 in the exchange region under both weak- and strong-field conditions. The Re M_2 part of the signal due to the nonlinearity of the magnetization curve allows us to determine χ_2. Analysis of Im M_2 showed that the contribution to this signal is due not only to the frequency dispersion of the susceptibility χ_2, which has a static limit, but also to the dynamic portion of the response, which has no static limit. The latter arises due to the influence of the AC field on the relaxation processes. In the limiting strong- or weak-field cases the experimental data for Im M_2 are well described by using well-known limiting formulas for $\Gamma_\parallel$ (Refs. 1, 8). An interpolation expression is proposed for $\Gamma_\parallel$ which allows us to describe the behavior of Im M_2 from the region of weak fields to the limits of the region of strong fields. The value of the spin-diffusion coefficient D_0 is determined.

It is well known that ferromagnets possess a pronounced nonlinearity in their magnetic properties in the fluctuation-dominated paramagnetic neighborhood of T_c. Ref. 1 contains a theoretical analysis of a number of dynamic effects caused by this phenomenon, as well as a discussion of the very few papers in which it is investigated experimentally. However, these phenomena are still very ill-studied.

The nonlinear contribution to the response of a ferromagnetic to an external uniform harmonic magnetic field of amplitude h_0 and frequency ω leads to the generation of higher harmonics of the excitation frequency. Since the magnetization of a ferromagnet above T_c is an odd function of the field, only odd harmonics can appear when the field has no constant component H. Although the even harmonics are an even function of h_0, they are odd in H; therefore, in order to observe them we need a constant field.

This paper is devoted to a theoretical and an experimental investigation of the second-harmonic magnetization M_2 of a cubic ferromagnet, caused by nonlinearity in its longitudinal response ($\mathbf{H}\|\mathbf{h}_0$), under conditions where $M_2 \sim h_0^2$. In this case, the expression for $M_2(t)$ induced by a field $h(t) = h_0 \sin \omega t$ can be written in the following form:

$$M_2(t) = -{}^1/_2 h_0^2 (\chi_2(\omega) e^{-2i\omega t} + \chi_2^*(\omega) e^{2i\omega t}). \qquad (1)$$

Here $\chi_2(\omega)$ is the second-order dynamic susceptibility, which describes the nonlinear longitudinal response and is expressed in terms of the fourth-order spin Green's function of the magnet.[1,2] Using this relation and symmetry considerations, we can determine the general properties of $\chi_2(\omega)$.[1,2] Let us list several of these. By virtue of the reality of the response, $\chi_2(\omega) = \chi_2^*(-\omega)$. The function $\chi_2(\omega)$ is odd in H, and since the entire susceptibility is described by a retarded response, it is analytic in the upper-half ω plane. In contrast to the linear susceptibility, the quantity Im $\chi_2(\omega)$ for $\omega > 0$ is not required to have a definite sign, because the condition Im $\chi_1(\omega) > 0$ follows from the requirement of positiveness of the energy absorbed by the body located in the external field, while the contribution to the absorption from Im $\chi_2(\omega)$ is negligibly small.[1] In the fluctuation-dominated neighborhood of T_c, the asymptotic behavior of χ_2 in ω and H can be determined in a number of simple cases from considerations of dynamic similarity.[1] The detailed functional form of $\chi_2(\omega)$ in the critical region is not analyzed.

We know of only a few experimental papers regarding the investigation of the nonlinear dynamic response of ferromagnets to an external field above T_c.[3,4] In these appears the dependence of the amplitudes of the higher harmonics on $\tau = (T - T_c)/T_c$ is investigated for $H = 0$ (without separating the phase components) for the cubic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$ in the dipole region of temperatures $4\pi\chi > 1$. It was observed that in the immediate vicinity of T_c the magnets possessed anomalous dynamic properties, which up to now have not received any theoretical explanation. In the region $20 \geqslant 4\pi\chi > 1$, where only the odd harmonics were observed, it was possible to carry out a comparison with similarity theory in one limiting case: at sufficiently low frequencies, the dependence on τ of the third harmonic amplitude of the magnetization M_3 under conditions where $M_3 \propto h_0^3$ was found to agree with its static limit: $M_3/h_0^3 \propto \partial^3 M/\partial H^3 (H=0)$. Data on the amplitudes of those higher-harmonic components that reduce to zero as $\omega \to 0$ (i.e., those that have no static limit) in the normal dipole region were not obtained, due to the small contribution they make to the signal in the region of frequencies employed in experiment. The problem of isolating this contribution is further complicated by the fact that these authors did not separate the phase components of the signals they recorded. Let us note that, generally speaking, even harmonics have not been studied in the dipole region, let alone in the exchange region ($4\pi\chi < 1$) where the magnitude of the signal is greatly reduced in proportion to the decrease in the value of the susceptibility.

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The experimental method used in this paper allowed us to operate with weak signals. We have demonstrated this previously in our studies of solid magnetically-dilute paramagnets and their alloys under conditions where single-particle nonlinear effects predominate.[2,5,6] In this case we showed that it is possible to investigate paramagnets in which no EPR signal is observed due to large line widths or appreciable splitting in zero-field.[7] The second harmonic was also reliably observed in high-grade crystals of La_2CuO_4, a material which is currently under intense study and which, as is well-known, exhibits no EPR signal. This being the case, there is clearly no advantage in studying M_2 for this material, in the absence of any detailed interpretation of the results of investigations of M_2 in magnets that undergo phase transitions. We recall that in La_2CuO_4 there is a phase transition to the antiferromagnetic state.

One of the reasons for the appearance of a nonlinear response in the single-particle approximation is the field dependence of the spin-relaxation time.[6] As we will see, nonlinear effects of this kind also arise in concentrated magnets that undergo a phase transition, in which the influence of the AC magnetic field on the relaxation process reduces to a dependence of the relaxation rate Γ on the instantaneous value of the field $H(t) = H + h(t)$. The part of $\chi_2(\omega)$ that corresponds to this effect does not have a static limit. The primary contribution, which is $\propto \omega/\Gamma$, enters into the phase component $M_2^{\sin} \propto \mathrm{Im}\ \chi_2(\omega)$. A second and more trivial reason for the appearance of the second harmonic in the longitudinal response is the nonlinearity of the magnetization curve $M(H)$. This part of $\chi_2(\omega)$ has a static limit, and contributes primarily to the phase component $M_2^{\cos} \propto \mathrm{Re}\ \chi_2(\omega)$.

In this paper we investigate the properties of $\chi_2(\omega)$, both theoretically and experimentally, for the cubic ferromagnet $CdCr_2Se_4$ ($T_c \approx 129$ K) in the exchange regions of temperature and field $[4\pi\chi(T,H) < 1]$ and at comparatively low frequencies for which the Lorenz formula correctly describes the linear response.

The purpose of this paper is to obtain experimental data in the exchange region under both weak- and strong-field conditions, and to analyze these features quantitatively based on existing theoretical ideas. In addition, in this paper we address the question of how to isolate from the total signal that portion which is due to the effect of the AC field on the relaxation processes, which in our view is the most interesting part.

The dependence of $\Gamma_{\parallel}$ on H for a ferromagnet was analyzed in Ref. 8. It is important to emphasize that not all of this dependence leads to a nonlinear response. The theoretical investigation of this group of questions is the subject of Sec. 1 of the paper, where we determine the function $\chi_2(\omega)$ and propose an interpolation expression that describes the behavior of $\Gamma_{\parallel}(H)$ from the region of weak fields up to the limits of the strong-field region. This expression is required for our analysis of the experimental data. In Sec. 2 we briefly describe our experimental method. In Sec. 3 we present the results of our experimental investigation of the τ-dependence of the two phase components of $\chi_2(\omega)$ at a fixed frequency $f = 15.7$ MHz for two values of the field H, along with a discussion of the experimental results.

Comparing theory and experiment allows us to determine the rate of longitudinal relaxation of the magnetization $\Gamma_{\parallel}(\tau,H)$ in the exchange region. As far as we know, this kind of data on the H-dependence of $\Gamma_{\parallel}$ has never been obtained from investigations of the longitudinal linear response.

We discuss how the τ-dependence of the attenuation $\Gamma_{\parallel}$ for fixed H obtained in this paper differs from the τ-dependence of $\Gamma_{\perp}$ obtained previously using the EPR method.[9]

1. $\chi_2(\omega)$ FOR A CUBIC FERROMAGNET IN THE EXCHANGE REGION

It is well-known that for not-too-high frequencies the linear longitudinal susceptibility of a cubic ferromagnet in the exchange region is described by the Lorenz formula in the uniform limit, with $\Gamma_{\parallel}$ determine by dipole forces that are included via perturbation theory.[1,8] The behavior of $\chi_{\parallel}^{(1)}(\omega)$ can be obtained from the Bloch equations. Under the same conditions, the Bloch equations can also be used to describe the behavior of $\chi_{\parallel}^{(2)}(\omega)$, if these equations are modified in an obvious way to include the nonlinearity of the magnetization curve and the requirement that the fields relax to the instantaneous value of the magnetic field (this approach is used in the analysis of single-particle nonlinear effects in the high-temperature region, where the nonlinearity of the magnetization curve can be neglected[3,7]):

$$(\partial/\partial t)(\Delta M(t)) = -\Gamma_{\parallel}(\omega_0(t))[\Delta M(t) - \chi_{\parallel}^{(1)} h(t) - \chi_{\parallel}^{(2)} h^2(t)]. \quad (2)$$

Here $\Delta M(t) = M(t) - M_0$ is the deviation of the magnetization from its static equilibrium value caused by the AC field $h(t)$, while $\chi_{\parallel}^{(1)} = \partial M/\partial H$, $\chi_{\parallel}^{(2)} = (1/2)\partial^2 M/\partial H^2$. In essence, we have expanded the equilibrium magnetization on the right side of the equation in a power series in $h(t)$, which allows us to represent the relaxation to an instantaneous value of magnetic field explicitly while taking into account the nonlinearity of the magnetization curve $M(H)$. The latter is treated in an obvious way by the term involving $\chi_{\parallel}^{(2)}$.

Let us turn to an analysis of $\Gamma_{\parallel}$ for a ferromagnet. For $\Gamma_{\parallel}$ we will use the uniform-limit expressions obtained in Refs. 1 and 8, in which we take into account the influence of the AC field. Let us recall that in Refs. 1 and 8 $\Gamma_{\parallel}$ was calculated within the relaxation time formalism, which involves the use of pair correlators of the spin density for the total spin of the system obtained by averaging with respect to an equilibrium distribution function. The attenuation $\Gamma_{\parallel}$ is determined by the decay of the uniform mode into exchange fluctuation modes, a process that is mediated by dipole forces. A magnetic field can change $\Gamma_{\parallel}$ in two ways. First of all, the field affects the equilibrium distribution function, leading to a "static" H-dependence of both the mode amplitudes and the excitation spectrum. Secondly, the equations of motion change their form in the presence of a field. The transverse components of the spins precess in the field, leading to a shift in the dispersion frequency of the transverse pair correlators by ω_0 and to the appearance in the longitudinal correlator of components that depend on $(\omega \pm \omega_0)$ due to the interaction of the longitudinal and transverse modes. We emphasize that this shift is a consequence of the fact that the exchange Hamiltonian and the interaction with H commute. As a result, the function $\Gamma_{\parallel}(\omega,H)$ given in Ref. 8 has both static and dynamic (dispersive) H-components, the latter consisting of a shift by ω_0 and $2\omega_0$ in those terms that have the form of functions of $\omega \pm \omega_0$ and $\omega \pm 2\omega_0$. In the low-frequency region, where the dispersion of $\Gamma_{\parallel}$ is not important (the corresponding criteria will be given below), the response can be analyzed on the basis of the Bloch equations with $\Gamma_{\parallel}(\omega,H) \approx \Gamma_{\parallel}(0,H)$. In this case the effect of the AC field on the relaxation processes will be manifested as a modulation of the precession frequency. In the low-frequency region, this effect reduces simply to replacing the static field H by its instantaneous value $H(t) = H + h(t)$ in the expression for the precession frequency $\omega_0 = g\mu H$, in complete analogy with the single-particle case.

From Eq. (2) it is easy to determine $\chi_{\parallel}^{(2)}(\omega)$:

$$\chi_{\parallel}^{(2)}(\omega) = \frac{\Gamma_{\parallel}}{-2i\omega+\Gamma_{\parallel}}\chi_{\parallel}^{(2)} - i\omega\frac{g\mu(\partial/\partial\omega_0)\Gamma_{\parallel}}{(-2i\omega+\Gamma_{\parallel})(-i\omega+\Gamma_{\parallel})}\chi_{\parallel}^{(1)}, \tag{3}$$

where $\Gamma_{\parallel} = \Gamma_{\parallel}(H,\omega_0,\tau)$ and $\chi_{\parallel}^{(1,2)} = \chi_{\parallel}^{(1,2)}(H,\tau)$.

The first term in (3) is due to the nonlinearity of $M(H)$. Its structure is analogous to $\chi_{\parallel}^{(1)}(\omega)$, since it measures the deviation of $\Delta M(t)$ from $\chi_{\parallel}^{(2)}h^2(t)$. The second term is a consequence of interference between the dynamic part of the linear response, given by the difference $\Delta M(t) - \chi_{\parallel}^{(1)}h(t)$, and the linear term in the expansion of $\Gamma_{\parallel}[\omega_0(t)]$ with respect to $h(t)$. The quantities $\Gamma_{\parallel}$ and $\chi_{\parallel}^{(1)}$ are even, while $(\partial/\partial\omega_0)\Gamma_{\parallel}$ and $\chi_{\parallel}^{(2)}$ are odd functions of H. It is easy to see that $\chi_{\parallel}^{(2)}$ possesses the correct symmetry and analytic properties.

We now consider the properties of $\Gamma_{\parallel}(H,\omega_0,\tau)$ and determine the limits on admissible values of ω in (3). Let us recall the frequency scales in the problem. The characteristic energy for critical fluctuations is $\Omega(\tau) \propto T_c\tau^{5/3}$ [here we have taken into account the fact that the Fisher index η in three-dimensional systems is negligibly small, while the critical index for the correlation length is $\nu \approx 2/3$ (see Ref. 1)], and the dipole attenuation in zero field $\Gamma_0(\tau) \propto \omega_d{}^2(T_c\tau)^{-1}$, where $\omega_d = 4\pi(g\mu)^2/v_0$ is a characteristic dipole energy. Here v_0 is the volume of an elementary magnetic cell. In the exchange region $\Omega(\tau) \gg \Gamma_0(\tau)$. The field is weak if $\omega_0 \ll \Omega(\tau)$, and strong in the opposite case.

The dependence of $\Gamma_{\parallel}$ on ω and H in a weak field is well known.[8] Let us recall the results of Refs. 1 and 8, where it was shown that in the hydrodynamic region ($g \ll \varkappa$, where q is the momentum of the critical fluctuations and æ is the inverse correlation length) the appearance of irregular dispersion corrections to Γ_0 with respect to H and ω (where Γ_0 is the Huber attenuation in zero field) is caused by the nontrivial frequency and field dependences of the kinetic coefficients ($L_{\parallel} = \chi_{\parallel}\Gamma_{\parallel}$), which in turn are due to the existence of singular points associated with the decay of the critical modes. The main contribution to the correction to Γ_0 is caused by double decays.[1,8] The most interesting interval is $\Omega(\tau) \gg \omega_0 \gg \Gamma_0$, where important dispersion corrections to Γ_0 appear that are caused by diffusive modes. We note that the characteristic scale over which these corrections vary as a function of ω and H is not $\Omega(\tau)$, but rather the Huber attenuation $\Gamma_0 = \gamma_0\omega_d^2(T_c\tau)$. The corresponding expression for $\omega = 0$ has the form

$$\Gamma_{\parallel}(\omega_0,\tau) = \Gamma_0 + \Delta\Gamma_{\parallel}{}^h = \gamma_0\omega_d{}^2/(T_c\tau)\,[1 - C_{\parallel}(\omega_0/T_c)^{1/2}\tau^{-5/6}]$$
$$= \gamma_0(\omega_d{}^2/T_c)\tau^{-1} - [1+2^{5/2}]\,b\,(120\pi)^{-1}(\omega_d{}^2/T_c)C^{-3/2}h^{1/2}\tau^{-11/6}. \tag{4}$$

Here γ_0 and C are positive coefficients of order unity, $C_{\parallel} \propto D_0^{-3/2}$, $D = D_0\tau^{1/3} = Cd^2T_c\tau^{1/3}$ is the coefficient of spin diffusion, d is the magnetic lattice constant, and β is the amplitude of the critical susceptibility.[9] The dependence of D on τ in (4) enters into the overall exponent that τ is raised to in the second term. The diffusion correction to (4) is the primary term that determines the H dependence of $\Gamma_{\parallel}$ in a weak field. The regular corrections are of order $[\omega_0/\Omega(\tau)]^2$ and $[H/\Omega(\tau)]^2$, and we can neglect them. If $\omega \neq 0$ holds, then in place of ω_0 in (4) there appear terms with $\omega - \omega_0$ and $\omega + \omega_0$. From this there arises a frequency limitation $\omega \ll \omega_0$ in (3), which is more restrictive than the usual condition for applicability of the pole approximation $\omega \ll \Omega(\tau)$. This is quite natural, because in (3) the factor $\partial\Gamma_{\parallel}/\partial\omega_0$ is due to the dispersion of $\Gamma_{\parallel}$.

The expression for the attenuation in the region of small fields $g\mu H = \omega_0 \gg \Omega(\tau)$, where the primary energy scale is determined by the field and the characteristic size of the critical fluctuations becomes the quantity

$$R_c^{(h)} = \varkappa_h{}^{-1} = d\left(\frac{T_c}{g\mu H}\right)^{2/5},$$

is also well-known:[1]

$$\Gamma_0{}^h = \gamma_{\parallel}{}^h\frac{\omega_d{}^2}{T_c}\left(\frac{T_c}{g\mu H}\right)^{3/5}. \tag{5}$$

Here $\gamma_{\parallel}^h$ is a constant coefficient of order unity.

In order to analyze the experimental data we had to have an expression for $\Gamma_{\parallel}$ that was correct both in weak and strong fields. Because its calculation does not appear to be possible, we present here an interpolation formula which reduces to expression (4) for $\Gamma_{\parallel}$ in a weak field, is in agreement with dynamic similarity, takes into account the anisotropy of fluctuations in the field, and finally—this is very important—contains a "dynamic" dependence on ω_0 and a "static" dependence on H:

$$\Gamma_{\parallel}(H,\omega_0,\tau) = \gamma_0\frac{\omega_d{}^2}{T_c}\tilde{\chi}_{\perp}^{3/4}\frac{\chi_{\perp}}{\chi_{\parallel}}\left\{\left[C_1\left(\frac{\omega_0}{T_c}\right)^2\tilde{\chi}_{\perp}^{5/2}+1\right]^{-1}\right.$$
$$\times\left[1+C_2\left(1-\frac{\chi_{\parallel}}{\chi_{\perp}}\right)\right]$$
$$\left.-C_{\parallel}\left(\frac{\omega_0}{T_c}\right)^{1/2}\tilde{\chi}_{\perp}^{5/8}\left[1+C_3\left(1-\frac{\chi_{\parallel}}{\chi_{\perp}}\right)\right]\right\}. \tag{6}$$

Here $\chi_{\perp} = M/H$, $\chi_{\parallel} \equiv \chi_{\parallel}^{(1)} = \partial M/\partial H$ are the transverse and longitudinal susceptibilities, $C_{\parallel}$ and C_i are fitting parameters ($i = 1,2,3$), $C_1 > 0$, and $\tilde{\chi}_{\perp}$ is a dimensionless susceptibility, i.e., $\chi_{\perp}$ normalized in such a way that $\tilde{\chi}_{\perp} = \tau^{-4/3}$ under weak-field conditions in the exchange region. A specific expression for $\tilde{\chi}_{\perp}$ will be given below. By introducing $\chi_{\parallel}$ and $\chi_{\perp}$ into the expression for $\Gamma_{\parallel}$, we can transform the dependence on τ into a scaling dependence on H and τ, which makes it possible for us to describe the behavior of $\Gamma_{\parallel}$ as we pass from the weak-field to the strong-field regime. The dependence on ω_0 is exhibited in explicit form, while the "static" dependence on H is contained in $\tilde{\chi}_{\perp}$ and $\chi_{\parallel,\perp}$. The terms with C_2 and C_3 describe the anisotropy.

Let us describe the considerations that led us to calculate Γ in the form (6). Consider the term with ω_0^2 in a weak field, where $\tilde{\chi}_\perp^{-5/2} \approx (\tau^{5/3})^2 \sim \Omega^2(\tau)$, $\chi_\parallel/\chi_\perp \approx 1$. It is not difficult to show that this expression corresponds to the contribution to $\Gamma_\parallel$ from the decay of the uniform mode into two transverse modes or a transverse and a longitudinal mode, a contribution coming from the region of momenta $q \sim \varkappa$. The appearance of ω_0^2 is caused by the spectrum of transverse modes which has the form $\omega \pm (\omega_0 + b^* g\mu H q^2/\varkappa^2) + i\Gamma_q$, where $b^* \sim 1$ and $\Gamma_q \sim \Omega(\tau)$ when $q \sim \varkappa$ (see Refs. 8, 10). We omit an interference term proportional to $\omega_0 H$ which appears along with ω_0^2. In an arbitrary field τ is replaced by $\tilde{\chi}_\perp^{-3/4}$ in the term with ω_0^2, the diffusion term, and the overall factor of τ^{-1} in (4). This implies that for arbitrary fields we must replace $\varkappa$, which is the weak-field effective scaling of the momentum for decay into transverse modes, by the natural static scale $\varkappa_\perp \propto \chi_\perp^{-1/2}$ for the transverse modes. The overall factor of $\chi_\perp/\chi_\parallel$ in (6) includes the fact that the perturbation-theory plot corresponding to $\Gamma_\parallel$ is proportional to $\chi_\parallel^{-1}$. Finally, the distinction between decays into two transverse modes and a longitudinal mode is taken into account in a simple way, i.e., by the ratio of the amplitudes of these modes in the terms with C_2 and C_3. We will see that Eq. (6) contains enough parameters to reconstruct the function $\Gamma_\parallel(H,\omega_0,\tau)$ from the experimental data.

We also need expressions for $\chi_{\parallel,\perp}$. These susceptibilities can be determined from the experimental data, since for $\omega \ll \Gamma_\parallel$ we have, according to (3), that

$$\mathrm{Re}\ \chi_\parallel^{(2)}(0) = \chi_\parallel^{(2)} = 1/2(\partial/\partial H)\chi_\parallel.$$

We will use the known two-parameter representation for the dimensionless $\chi_{\parallel,\perp}$ (Ref. 11) which is written in a form that is convenient for us as follows:

$$\tilde{\chi}_\parallel(\tau,h) = [\varphi(x)/\tau]^{4/3} = A^{4/5}[1-\varphi(x)]^{2/5}\{1-a[1-\varphi(x)]\}^{4/5}h^{-4/5}, \quad (7)$$

$$x = h/\tau^{5/3} = A[1-\varphi(x)]^{1/2}\{1-a[1-\varphi(x)]\}\varphi^{-5/3}, \quad (8)$$

$$\tilde{\chi}_\perp(\tau,h) = \frac{\tilde{\chi}_\parallel(\tau,H)}{1-a[1-\varphi(x)]}, \quad \frac{\chi_\parallel}{\chi_\perp} = \frac{\tilde{\chi}_\parallel}{\tilde{\chi}_\perp}. \quad (9)$$

Here A, a are positive coefficients. In this approximation the theoretical value of $a = 2\gamma/3 = 8/9$, $h = g\mu H/T_c$. The function $\varphi(x)$ decreases monotonically from unity to zero as x varies from zero to infinity. The parameters A and a have a clear physical meaning. The function $\tilde{\chi}_\parallel(\tau,h)$ has a maximum with respect to τ for fixed h, whose position is determined by the coefficient a. In this case the value of the amplitude of $\tilde{\chi}_\parallel$ at its maximum is proportional to $A^{4/5}$. The susceptibility $\chi_\perp$ for h = const decreases monotonically with increasing τ. The maximum anisotropy $\chi_\parallel/\chi_\perp = 1 - a$ is reached for $\tau = 0$. The reason for introducing a two-parameter representation for $\chi_{\parallel,\perp}$ is obvious—it enables us to include not only the τ-dependence of χ but also its h-dependence, so that the susceptibility is fully specified in an arbitrary field. Including the correct scale factor is not difficult when we use experimental data[9] based on the static susceptibility.

2. EXPERIMENTAL METHOD

In our experiments we investigated the longitudinal nonlinear response to a harmonic magnetic field in the presence of a constant field $[H(t) = H + h_0 \sin \omega t]$ at the second harmonic 2ω of the excitation frequency. The phase components of the second harmonic of the magnetization M_2 were recorded for the sample under study as a function of the magnitude of the constant magnetic field for different values of temperature. The frequency of the AC field was $f = \omega/2\pi = 15.7$ MHz. Although the experimental setup and method of calibrating the phase of the reference voltage at 2ω in order to separate out the phase components of M_2 were essentially described in Ref. 7, there are certain differences, which we will touch on briefly, associated with the need to make measurements in a constant magnetic field H. They are reflected in Fig. 1. In this setup, the high-frequency (hf) coil of *1* (Fig. 1), which creates the AC magnetic field within which the sample under study is placed, is situated in a separate electromagnetic shield *2* and thermal isolation shield *3*, and is taken out from the remaining high-frequency elements of the operating detector. The coil is connected to these elements by means of a coaxial cable *4*. This is done so as to decrease the influence of the constant field H on the remaining high-frequency elements of the detector, which is manifest in these elements as a parasitic signal that depends on H.[7] The hf coil is placed in the gap of an electromagnet together with sweeping coils *5* for the magnetic field H and output elements of two thermal stabilization systems—one system for stabilizing the temperature of the hf coil and one for stabilizing the sample temperature, in which we used nitrogen vapor as a cooling agent. Separation of the fluxes of the two systems was ensured by a quartz diaphragm *6*.

The measurements were carried out for two values of the constant magnetic field, $H = 1.5$ kOe and $H = 3$ kOe, both on single-crystal $CdCr_2Se_4$ and on samples consisting of groups of small single crystals placed on the surface of the quartz sample holder *7* in a single layer. The size of these small single crystals was less than $35 \times 15 \times 15\ \mu m^3$. This type of sample *8* ensured a considerably larger surface for

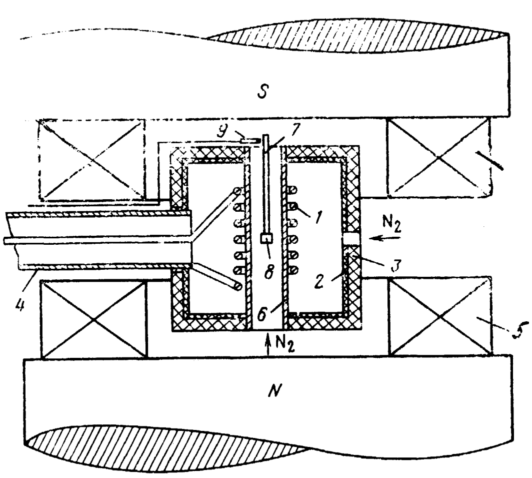

FIG. 1. Schematic of a portion of the operating detector placed in an electromagnet: 1—high-frequency coil, 2 and 3—electromagnetic and thermal isolation shields, respectively, 4—coax, 5—scanning coil for magnetic field, 6—quartz partition, 7—quartz sample holder, 8—sample under study, 9—thermocouple.

thermal contact with the heat sink of the thermal stabilization system. Comparison of the experimental results for a single crystal and the polycrystalline samples did not show any appreciable difference in the shape and magnitude of the signal. We note that the signal shape, both for a large single crystal and for the small single crystals, can roughly be represented as an ellipsoid with axes that differ from one another by a factor of two, the large axis being oriented parallel to the field. The experimental data near T_c allow us to estimate the influence of absorption of the HF power due to dielectric and magnetic losses at the sample temperature, and to choose the amplitude of the HF field h_0 such that these effects are smaller than the measurement error at that temperature, which for reasons we will describe below was rather large, about 0.2 K.

Yet another limitation on the amplitude of the hf field h_0 at zero constant field follows from the condition $g\mu h_0 \ll \Omega(\tau)$. In our experiment the closest approach to T_c was the value $\tau = 3.88 \cdot 10^{-3}$, at which $h_0^{\text{crit}} = 96$ Oe. In our case, where the constant fields $H = 1.5$ kOe and 3 kOe are comparatively large and the dipole interactions considerably suppressed, the limitation on h_0 was less severe. From these considerations we chose an amplitude $h_0 = 6$ Oe.

The measurements were carried out in the temperature range 129.5 to 146 K. The temperature was measured with a thermocouple *9* (Fig. 1) made of copper-constantan, with the temperature of one of its junctions fixed at 0 °C. Calibration of the thermocouple was accomplished by tying its readings to specific well-determined temperature points 0 °C, 100 °C, $-$ 195.8 °C, using well known fifth-degree polynomial approximations to describe the temperature curve of the thermocouple.[12] The zeroth-order approximation for the coefficients of the polynomial was obtained from tables of data in Ref. 13 for a sample copper-constantan thermocouple. Corrections to the coefficients for the real thermocouple were determined by matching at the temperature points described above. The error in determining the temperature came to 0.1 K. Since the thermocouple was located a certain distance from the sample under study in order to keep it from being heated by the hf field, a temperature gradient was present, and consequently an additional error in determining the sample temperature. The total error in measuring its temperature came to $\leqslant$0.2 K.

Matching of the thermocouple readings to the temperature of the sample was accomplished using the Curie temperature as a reference. In order to determine the Curie point we investigated a known feature of the behavior of the second harmonic of the magnetization M_2 in a weak field H—the fact that the temperature dependence of its amplitude is a maximum at T_c (Ref. 3). The portion of the experimental results used to determine T_c, i.e., the phase components of M_2 near T_c as a function of H, were obtained by symmetric scanning of H through zero field and are shown in Fig. 2. The scanning frequency of H was 1 Hz, and the amplitude of the AC field in this case was $\approx$0.3 Oe. The character of the dependence of M_2 on H below T_c became very complicated and is not the subject of analysis in this paper. We note only that, depending on the value of H, excess noise appeared over and above the noise input from the detector at a certain $\Delta T = T - T_c$ below T_c, which disappeared as H increased without changing the temperature of the sample. We also observed hysteresis in several regions of H (but not on all curves), along with the appearance of additional extrema of the signal whose position changed as we varied the temperature. This behavior is apparently due to domain-formation processes.

3. RESULTS AND DISCUSSION

The experimental results for nonzero values of the constant magnetic field H (the pedestal) equal to 1.5 kOe or 3 kOe for a constant scanning range ΔH (about 70 Oe) are straight lines with an accuracy (5–10)%, whose slope changes as a function of temperature. We calculated the ratios $\Delta M_2^{\cos}/\Delta H$ and $\Delta M_2^{\sin}/\Delta H$ (the slope angles) because the τ-dependence of the derivatives is considerably stronger, with characteristic special points (a zero or a maximum). The ratios $\Delta M_2^{\cos}/\Delta H$ and $\Delta M_2^{\sin}/\Delta H$ are respectively the real and imaginary parts of the second harmonic response. Figures 3 and 4 show these experimental points in arbitrary units as a function of $\Delta T = T - T_c$ for $H = 1.5$ kOe. The error present in the experimental curves was caused primarily by errors in measuring sample temperature. In reality the variation of the sample temperature causes a change in the amplitude of the signal, which is proportional to the derivative with respect to τ of the experimental curve. Therefore, the errors are unequal at different

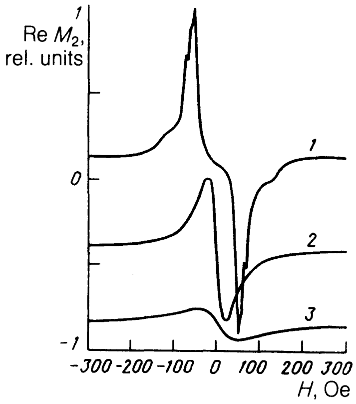

FIG. 2. Real part of the second harmonic signal as a function of H for symmetric scanning of H around zero field near T_c: 1—$T = 127.6$ K, 2—$T = 129$ K, 3—$T = 129.9$ K.

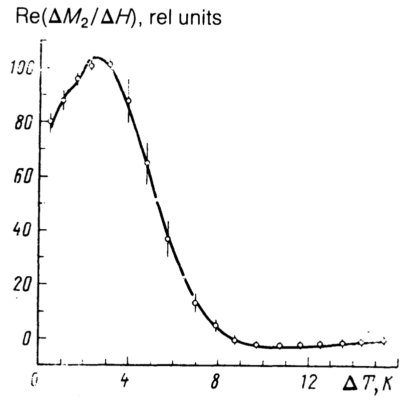

FIG. 3. Dependence of $\text{Re}(\Delta M_2/\Delta H) = \Delta M_2^{\cos}/\Delta H$ on ΔT for $H = 1.5$ kOe. The parameters of the theoretical curve are $a = 0.7 \pm 0.02$, $A = 0.33 \pm 0.1$.

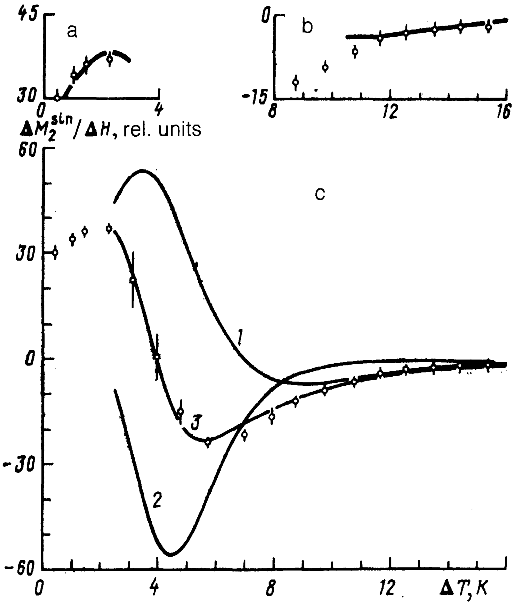

FIG. 4. Dependence of $\mathrm{Im}(\Delta M_2/\Delta H) = \Delta M_2^{sin}/\Delta H$ on ΔT for $H = 1.5$ kOe. a—strong-field region $2.5\ \mathrm{K} \geqslant \Delta T \geqslant 0.5$ K; in the approximation we used the limiting formula (5) for $\Gamma_\parallel$; $\gamma_\parallel^h = 3.9 \pm 0.8$. b—weak-field region $\Delta T \geqslant 12$ K: the limiting formula (4) was used to approximate $\Gamma_\parallel$; $C_\parallel = 1.45 \pm 0.7$. c—approximation of the curve in the region $\Delta T \geqslant 3$ K. The interpolation formula (6) was used for $\Gamma_\parallel$; $C_1 = 0$; $C_2 = 1.6 \pm 0.1$; $C_3 = -2.14 \pm 0.15$; $C_\parallel = 1.49 \pm 0.1$ 1—F_1 (theoretical curve)—dynamic part, which does not have a static limit, caused by the ω_0-dependence of $\Gamma_\parallel$. 2—F_2 (theoretical curve)-contribution from frequency dispersion of χ_2. 3—$F_1 + F_2 = \Delta M_2^{sin}/\Delta H$. The normalizing factor is the same for a, b, c.

points on the experimental curves. The minimum error was determined by the input noise of the detector.

Because the theoretical results presented above, which were used for the analysis of the experimental data, were obtained to lowest order in h_0 (i.e., $\sim h_0^2$), it should be noted that we carried out a preliminary check of the dependence of the signal amplitude on h_0 both in a field $H = 1.5$ kOe and in $H = 3$ kOe. It showed that the quadratic dependence on h_0 is obeyed up to an amplitude $h_0 \approx 20$ Oe in the temperature range $T \geqslant T_c$. In the experiments, as we have already noted above, we used an amplitude $h_0 \approx 6$ Oe.

We note also that over the entire temperature range used in the experiment $0 \leqslant \Delta T \leqslant 20$ K and in the presence of the constant fields $H = 1.5$ or 3 kOe, the following equation holds:

$$4\pi\chi_{\perp,\parallel} < 1, \tag{10}$$

as is clear from the experimental data regarding the magnetic susceptibility[9] and theoretical estimates which are easy to derive using the results of Ref. 1. The reason that condition (10) is fulfilled is obviously suppression of the phase transition by the magnetic field. In the immediate neighborhood of T_c, where the anisotropy of the susceptibility is considerable, condition (10) for $H = 1.5$ kOe naturally holds less well for $\chi_\perp$: $4\pi\chi_\perp(\tau = 0) \approx 0.6$, and better for $\chi_\parallel$: $4\pi\chi_\parallel(\tau = 0) \approx 0.18$ (Ref. 9), i.e., we can assume that our experiments, in which the minimum value of the temperature difference is $\Delta T \approx 1$ K, took place in the exchange region.

We must also determine the region over which the other important condition is fulfilled, i.e., the weak-field region:

$$g\mu H < \Omega \approx kT_c\tau^{5/3}, \tag{11}$$

where k is Boltzmann's constant. It is not difficult to verify that $g\mu H \approx \Omega$ holds for $CdCr_2Se_4$ at $\Delta T \approx 2.65$ K when $H = 1.5$ kOe, i.e., condition (11) is fulfilled for $\Delta T > 2.65$ K. Let us also recall once more that[1]

$$\tau_d = (\omega_d Z/T_c)^{3/4}, \tag{12}$$

where $Z = z_0 S(S+1)/3$ ($z_0 \approx 1$); for $CdCr_2Se_4$ we have $\tau_d \approx 1.56 \cdot 10^{-2}$, i.e., $\Delta T_d \approx 2$ K. In the presence of a constant field $H \geqslant 1.5$ kOe, as mentioned above, and a sample whose shape is close to ellipsoidal, such a value of ΔT_d allows us to assume that the corrections to the magnetization are small (the estimates indicate less than 10%) in the temperature range $\Delta T > 2$ K.

The frequency limitation was mentioned previously. The frequency becomes comparable to Ω for $\Delta T \leqslant 0.09$ K. Therefore, in the range of temperatures that we studied in our experiments, i.e., $\Delta T \geqslant 1$ K, we may assume that the frequency is small according to this criterion. Furthermore, data from EPR investigations[9] show that for $CdCr_2Se_4$ the width of the resonance transition in the exchange region satisfies $\Gamma \geqslant 30$ Oe in field units, or $\Gamma \geqslant 2\pi \cdot 10^8$ in frequency units. An estimate shows that in our case $\omega/\Gamma \leqslant 0.16$, so that in analyzing the experimental data we may neglect terms of higher order in ω/Γ.

In order to analyze the experimental data we make use of expression (3) for $\chi_\parallel^{(2)}(\omega)$, taking (6)–(9) into account. From this, by virtue of the smallness of ω/Γ we obtain

$$\frac{\Delta M_2^{\cos}(H,\tau)}{\Delta H} = \mathrm{Re}\left(\frac{\Delta M_2(H,\tau)}{\Delta H}\right) \approx h_0^2 \frac{d}{dH}\chi_\parallel^{(2)} = h_0^2 \frac{d^2}{dH^2}\chi_\parallel^{(1)}, \tag{13}$$

$$\frac{\Delta M_2^{\sin}(H,\tau)}{\Delta H} \approx h_0^2 \frac{\omega}{\Gamma_\parallel}\left\{\left[\frac{d^2\chi_\parallel^{(1)}}{dH^2} - \frac{d\chi_\parallel^{(1)}}{dH}\frac{d\Gamma_\parallel}{dH}\frac{1}{\Gamma_\parallel}\right] - \frac{g\mu}{\Gamma_\parallel}\left[\Gamma_\parallel'\frac{d\chi_\parallel^{(1)}}{dH} + \chi_\parallel^{(1)}\left(\frac{d\Gamma_\parallel'}{dH} - \frac{2\Gamma_\parallel'}{\Gamma_\parallel}\frac{d\Gamma_\parallel}{dH}\right)\right]\right\} = \mathrm{Im}\left(\frac{\Delta M_2\ (H,\tau)}{\Delta H}\right) = F_1 + F_2, \tag{14}$$

where $\Gamma_\parallel' = \partial\Gamma_\parallel/\partial\omega_0$; F_1 describes the dispersive contribution to $\chi_\parallel^{(2)}$, taking into account the static field dependence of $\Gamma_\parallel$ (this is necessary since we are interested in $\Delta M_2^{sin}/\Delta H$ rather than M_2^{sin} which is obtained in experiment), and F_2 describes the contribution due to the function $\Gamma[\omega_0(t)]$.

We compared the experimental data with the theory, taking into account our estimates of the boundaries of the weak-field region and the value of τ_d, in the temperature range $\Delta T \geqslant 2$ K. Because the interpolation formula (6) proposed above gives the required field dependence of $\Gamma_\parallel$ in the strong-field region[1] and a correct expression for $\Gamma_\parallel$ in the weak-field region, we expect it to describe the behavior of $\Gamma_\parallel$ adequately in the intermediate region as well. Expression (13), which contains three fitting parameters (a, A, and the normalization) taking into account (7)–(9), was used as an approximation for $\Delta M_2^{\cos}/\Delta H$, i.e., the curve shown in Fig. 3. Here and in what follows, we determine the fitting parameters and the normalization factor are by standard methods, i.e., minimization of the sum of squares of the deviations of the theoretical curve from the experimental points. In this case we obtain the following values of these parameters: $a = 0.7 \pm 0.02$; $A = 0.33 \pm 0.01$. The theoretical curves are shown in Figs. 3 and 4 (the solid curves).

Using Eq. (14) and taking (7)–(9) into account, we constructed an approximation for $\Delta M_2^{\sin}/\Delta H$; the results are shown in Fig. 4. In this case we made use of the values of a and A we had already found without further changes. In Fig. 4b we show the results of our approximation using the exact expression (4) for $\Gamma_\parallel$, which is valid for the tail of the experimental curve in the weak-field region, and in that region of temperatures where we can neglect the anisotropy of the critical fluctuations, i.e., $\Delta T \geqslant 12$ K. As fitting parameters we used $C_\parallel$ and the normalization. We obtained the value $C_\parallel = 1.45 \pm 0.7$. The error in determining $C_\parallel$ is large, since in this region of ΔT the experimental curve is rather featureless. Continuation of this theoretical curve into the region $\Delta T < 12$ K, where, as is clear from Fig. 5, considerable anisotropy is already present, shows a significant disagreement with the experimental points, which attests to the importance of including the temperature dependence of the anisotropy of critical fluctuations even under weak field conditions in the exchange region.

In Fig. 4a we present the results of approximating the experimental curves $\mathrm{Im}(\Delta M_2/\Delta H)$ using Eq. (5) for $\Gamma_\parallel$, which is valid in the strong-field region $g\mu H > \Omega(t)$. The approximation was carried out only for the first four points, for which the strong-field condition was fulfilled approximately. Retaining the normalization constant obtained in the previous case for the tail of the experimental curve, we found that $\gamma_\parallel^h = 3.9 \pm 0.8$. It is true that at these temperatures the proximity to the dipole region, which is not included in the magnetization calculations, can introduce a considerable amount of error.

In Fig. 4b we show the results of approximation using

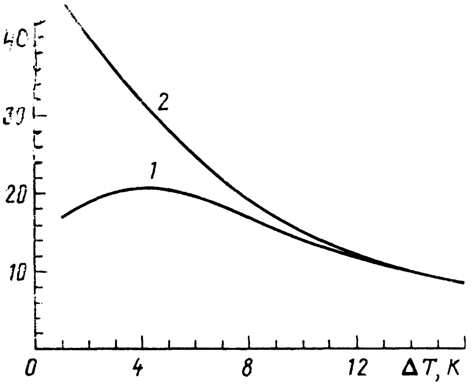

FIG. 5. Dependence of $\chi_\parallel$ (curve 1) and $\chi_\perp$ (curve 2) on ΔT for $H = 1.5$ kOe.

the interpolation expression (6) for $\Gamma_\parallel$. In this expression there are four fitting parameters plus the normalization, i.e., five parameters in all. Despite the large number of parameters to be determined the problem is correctly posed, since the presence of a maximum, minimum, and zero on the experimental curve already gives the necessary number of equations. We note that in the limit $C_1 = C_2 = C_3 = 0$ for the experimental points corresponding to Fig. 4b (in the tail), use of Eq. (6) as an approximation for $\Gamma_\parallel$ gives the same results for the constant $C_\parallel$ and the normalization factor as does the use of the exact expression (4) for the weak field.

In the region of temperatures $\Delta T \geqslant 3$ K, use of the approximation leads to the following results. The coefficient C_1, i.e., the field correction to the Huber attenuation, is found to equal zero. The values of the remaining parameters are as follows: $C_2 = -1.6 \pm 0.1$; $C_3 = -2.14 \pm 0.15$; and $C_\parallel = 1.49 \pm 0.1$. It is clear from this that the value of the constant $C_\parallel$ to good accuracy coincides with the value obtained in the limiting case for the tail of the experimental curve. The normalizing factor also coincides with the factor obtained in this limiting case to within 20%. Curves *1* and *2*, which are shown in Fig. 4c, reflect the dependence on ΔT of the functions F_1 introduced into (14) (the dispersion contribution from $\chi_\parallel^{(2)}$) and F_2 [the contribution caused by the dependence of $\Gamma_\parallel$ on $\omega_0(t)$]. The parameters C_2 and C_3 have values ~ 2, i.e., they are well-defined. The fact that they are contained in the factors that determine the effect of the anisotropy attests to their considerable influence on the relaxation, even in the weak-field region, starting at temperatures where the anisotropy of χ becomes appreciable. This agrees with the conclusion arrived at from the previous analysis. The coefficient $C_\parallel$ for the diffusion correction to $\Gamma_\parallel$, which depends on $\omega_0(t)$, can be correlated with results obtained by EPR method[9] in the exchange region under conditions $\chi_\perp \approx \chi_\parallel$. From Ref. 1, by taking into account the difference in the constant coefficients ($\Gamma_\parallel^h \sim 1 + 2^{5/2}$, $\Gamma_\perp^h \sim 5/2 + 2^{1/2}$), isolating an overall factor $h^{1/2}(\omega_d^2/T_c)\tau^{11/6}$, taking into account that in our case, according to (6), γ_0 is an overall factor for $\Gamma_\parallel$, and using the value of the constants from Ref. 9, we find for the diffusion correction

$$\Delta\Gamma_\perp{}^h \approx -3{,}46\cdot 10^{-2} C^{-3/2}(\omega_d{}^2/T_c)h^{1/2}\tau^{-11/6}.$$

In our case

$$\Delta\Gamma_\parallel{}^h \approx -C_\parallel(\omega_d{}^2/T_c)h^{1/2}\tau^{-11/6} = -5{,}88\cdot 10^{-2} C^{-3/2}(\omega_d{}^2/T_c)h^{1/2}\tau^{-11/6},$$

where $C_\parallel = 1.49 \pm 0.1$, as we proposed above. From this we obtain $C \approx 0.115$ and $D_0 = (24.5 \pm 6)$ MeV·Å^2, which, taking into account the errors, is rather close to the value $D_0 \approx 15 \pm 3$ of Ref. 9.

In Fig. 5 we show the dependence of the quantities $\chi_\parallel$ and $\chi_\perp$, normalized according to the results of measuring the static susceptibility,[9] on ΔT. The character of the variation of $\chi_\parallel$ and $\chi_\perp$ in the presence of a constant field H is found to be in agreement with the experimental data obtained by measuring the static magnetic susceptibility.[9] In Fig. 6 we show the dependence of $\Gamma_\parallel = \Gamma_0 + \Delta\Gamma_\parallel^h = \Gamma_\parallel^{(1)} - \Gamma_\parallel^{(2)}$ on ΔT (curve *3*), plotted according to Eq. (6) while using the val-

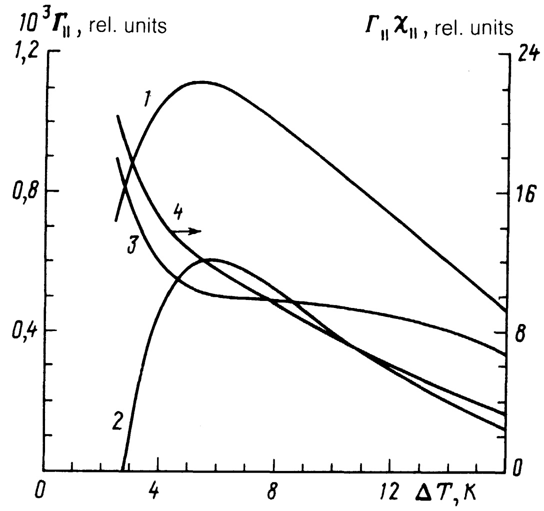

FIG. 6. Dependence of $\Gamma_\parallel$, Γ_1, Γ_2, and $L_\parallel = \Gamma_\parallel \chi_\parallel$ on ΔT for $H = 1.5$ kOe: 1—Γ_1 (in the weak-field region this is the Huber attenuation); 2—Γ_2 (in the weak-field region this is the ω_0-dependent diffusion correction); 3—$\Gamma_\parallel = \Gamma_1 - \Gamma_2$; 4—$L_\parallel = \Gamma_\parallel \chi_\parallel$.

ues of the parameters C_i ($i = 1$–3) and $C_\parallel$ presented above. Here we also show the ΔT-dependences of the individual parts of $\Gamma_\parallel$, i.e., $\Gamma_\parallel^{(1)}$ in curve *1* (in a weak field this is the Huber attenuation) and $\Gamma_\parallel^{(2)}$ in curve *2* (in a weak field this the h-dependent correction from the diffusion mode), as well as the dependence of the quantity $L_\parallel = \Gamma_\parallel \chi_\parallel$ on ΔT in curve *4* (here $L_\parallel$ is the kinetic coefficient of Ref. 1). The transformation of the function $\Gamma_\parallel^{(2)}$ which takes place when we pass from the weak-field regime to the strong-field regime is interesting. For the weak-field regime we have expression (4). As τ decreases the "diffusion" term decreases, and then changes sign, in the intermediate region. This transformation is completely natural, since in a strong field only one scale remains for the characteristic energy—the energy $g\mu H \sim \Omega(H)$—and the frequency region $[g\mu H \ll \Omega(\tau)]$ that is the origin of the "diffusion" term in $\Gamma_\parallel$ disappears. It is clear from Fig. 4c that the expression (14) gives a rather good description of the experimental data. This implies that the parametrization we have proposed here with a minimum number of parameters adequately reflect the features of the behavior of $\Gamma_\parallel$.

The attenuation $\Gamma_\parallel(\tau)$ shown in Fig. 6 increases monotonically as τ decreases. At the same time the function $\Gamma_\perp(\tau)$, which describes the EPR line width, has a maximum at fixed H whose position is determined by the field.[9] In order to understand the origin of this sharp difference in the behavior of $\Gamma_\parallel$ and $\Gamma_\perp$, let us compare the τ-dependence of the kinetic coefficients $L_{\parallel,\perp} = \Gamma_{\parallel,\perp}\chi_{\parallel,\perp}$. The coefficient $L_\parallel(\tau)$ (Fig. 6, curve *4*) increases monotonically as $\tau \to 0$. Using the data of Ref. 9, we can verify that the same character is seen in the function $L_\perp(\tau)$ for $H =$ const. Thus, the difference in the behavior of $\Gamma_\parallel$ and $\Gamma_\perp$ in a field originates primarily in the anisotropy of χ.

The analysis of the experimental data given above was carried out for $H = 1.5$ kOe, where the results are more reliable in the sense of measurement accuracy. In order to control the accuracy of the results obtained, we carried out measurements at $H = 3$ kOe and the same value of h_0 as we used for $H = 1.5$ kOe. To this end the experimental points for $H = 1.5$ kOe were recalculated in the region $H = 3$ kOe, using the following expressions:

$$\frac{\Delta T_3}{\Delta T_{1.5}} = \left(\frac{H_2}{H_1}\right)^{3/5} \approx 1.52, = \frac{A_3^{\cos}}{A_{1.5}^{\cos}} = \left(\frac{H_1}{H_2}\right)^{14/5} \approx 0.144,$$

14 $$\frac{A_3^{\sin}}{A_{1.5}^{\sin}} = \left(\frac{H_1}{H_2}\right)^{11/5} \approx 0.218$$

obtained from scaling relations between h and τ. Here $H_1 = 1.5$ kOe, $H_2 = 3$ kOe, $A^n = \Delta M_2^n/\Delta H$, $n = \cos$, $\sin$; $\Delta T_3 A_3^n$ are the values in the region $H = 3$ kOe obtained from the experimental values $\Delta T_{1.5}$, $A_{1.5}^n$ are for $H = 1.5$ kOe. The results of the recalculation, together with the experimental points for $H = 3$ kOe, are shown in Figs. 7a, 7b. There is good agreement between theory and experiment for $\mathrm{Re}(\Delta M_2/\Delta H)$ with a disagreement which goes somewhat beyond the range of experimental error only in the region of maximum signal in the vicinity of the boundary of the strong-field region. Satisfactory agreement is also obtained for the dispersive part of the response $\mathrm{Im}(\Delta M_2/\Delta H)$. The rather small difference is most likely connected with the difference in the frequency dispersion of $\chi_2(\omega)$ for $H = 1.5$ kOe and $H = 3$ kOe. In the analysis above we used the static limit for Re $\chi_2(\omega) \approx$ Re $\chi_2(0)$, and assumed that Im $\chi_2(\omega) \sim \omega/\Gamma$. Because $\Gamma_\parallel$ ($H = 1.5$ kOe; τ) $> \Gamma_\parallel$ ($H = 3$ kOe; τ), this condition is fulfilled badly for $H = 3$ kOe, which also is the origin of a certain disagreement in the time dependence when the "scaling" method is used for recalculation. These factors obviously explain the small discrepancy in $\mathrm{Re}(\Delta M_2/\Delta H)$ shown in Fig. 7a, which was mentioned above. The fact is that $\mathrm{Re}(\Delta M_2/\Delta H)$ contains a small admixture from relaxation effects that is proportional to $\omega/\Gamma_\parallel$ $\mathrm{Im}(\Delta M_2/\Delta H)_{\mathrm{rel}} = \omega/\Gamma_\parallel F_2$.

We also note that we have not included the contribution of the spin lattice relaxation Γ_{sp} to $\Gamma_\parallel$ in Eq. (6), which is caused by spin-phonon coupling.[9] The fact is that this term does not contribute to the relaxation part of the response $\Delta M_2^{\sin}/\Delta H$, which is proportional to $\partial\Gamma_\parallel/\partial\omega_0$, since, like χ, it depends only on the static field and is only weakly affected by its second part (proportional to the term in $\Gamma_\parallel$) while its value is known from Ref. 9.

CONCLUSION

The results of these investigations of the phase components of the second-harmonic longitudinal nonlinear response of the cubic paramagnet $CdCr_2Se_4$ in the presence of a constant field in the critical paramagnetic vicinity of T_c (in the exchange region, under weak-field conditions up to the edge of the strong-field condition) have shown that Re M_2 is caused by that part of χ_2 which has a static limit. The use of the well-known two-parameter representation for $\chi_\parallel^{(1)}$ (Ref. 11) makes it possible to obtain $\chi_\parallel^{(1)}$ in the exchange region of longitudinal fields by applying standard approximation procedures to the experimental curve. This allows us to carry out a detailed analysis of the imaginary part of M_2 and to isolate that portion which is caused by the frequency dispersion of χ_2 from the dynamic part, which does not have a static limit. This latter contribution is due to the effect of the AC field on the relaxation process. The mechanism that creates this response is revealed in the dependence of $\Gamma_\parallel$ on ω_0. We emphasize that the dependence on the static field,

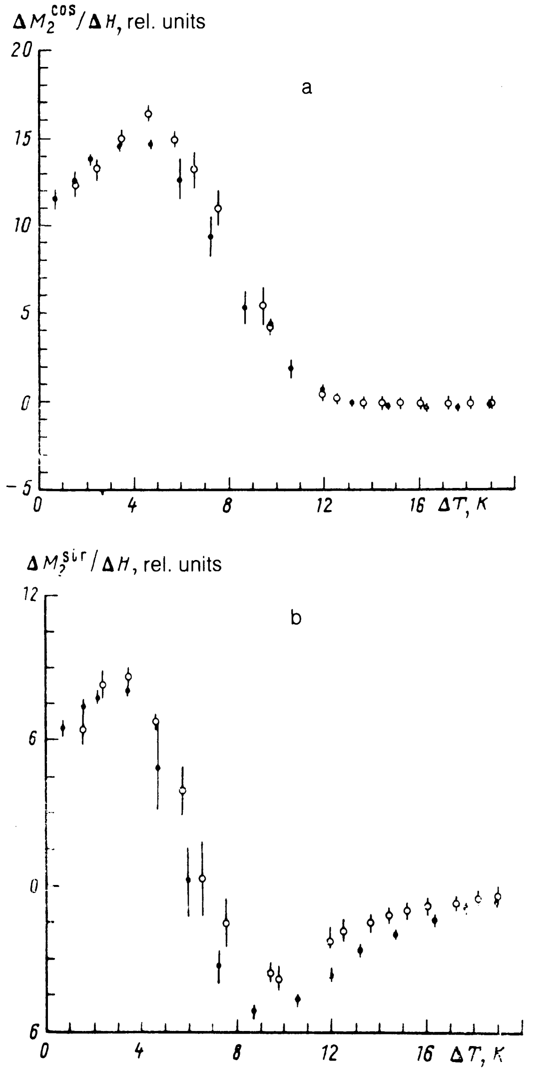

FIG. 7. Experimental results for $\Delta M_2/\Delta H$ for $H = 1.5$ kOe and $H = 3$ kOe as a function of ΔT. The points with $H = 1.5$ kOe (and their error bars) are recalculated using the scaling relations, where the scale is set at the experimental points for $H = 3$ kOe: (○)—$H = 3$ kOe; (●)—$H = 1.5$ kOe.

e.g., in χ, which is the limiting form of the τ-dependence in the region of small τ, does not lead to the appearance of the response mentioned above. In the limiting cases of weak and strong fields the experimental data for $\Gamma_\parallel$ are correctly approximated when expressions are used for $\mathrm{Im}(\Delta M_2/\Delta H)$ that are well-known from the theory corresponding to these limiting conditions,[1,8] which allows us to determine the constant coefficients in the expressions for $\Gamma_\parallel$ mentioned above.

We have presented an interpolation expression for $\Gamma_\parallel$ that accurately describes the behavior of $\Gamma_\parallel$ in the weak-field limit and takes into account anisotropy as the edges of the strong-field region are approached, and also contains a portion which is dependent on $\omega_0(t)$. Approximations of the experimental points for the imaginary part of $\Delta M_2/\Delta H$ that take into account the results of approximations for $\mathrm{Re}(\Delta M_2/\Delta H)$ allow us to obtain well-defined coefficients for $\Gamma_\parallel$ and to define the coefficient of spin diffusion D_0. We note that the coefficient C_1 in the ω_0-dependent correction to $\Gamma_\parallel$ turns out to equal zero in the approximating procedure, which confirms the sensitivity of the dynamic part of $\mathrm{Im}(\Delta M_2/\Delta H)$ to its ω_0-dependence. Using our interpolation formula, we obtain a phenomenological τ-dependence for $\Gamma_\parallel$ and its parts $\Gamma_\parallel^{(1)}$ and $\Gamma_\parallel^{(2)}$ from the region of weak fields up to the edge of the strong-field region. While the behavior of $\Gamma_\parallel$ as a function of τ differs from the behavior of $\Gamma_\perp$, the behaviors of the kinetic coefficients $L_\parallel \approx \Gamma_\parallel \chi_\parallel$ and $L_\perp \approx \Gamma_\perp \chi_\perp$ coincide qualitatively in that both coefficients decrease monotonically with increasing τ. This implies that the difference in behavior of Γ is essentially caused by the anisotropy of χ.

Especially noteworthy is the agreement in the limiting case of weak field between the descriptions of the experimental data for $\mathrm{Im}(\Delta M_2/\Delta H)$ using both the well-known limiting expression (4) from theory and the interpolation formula (6) for $\Gamma_\parallel$ in the limiting case where anisotropy of critical fluctuations is not included, along with the values of the constants obtained in this case for the diffusion term in $\Gamma_\parallel$ and the normalization constant. Within the limits of experimental error, these same values of the constants are obtained when the largest part of the experimental curve, including the intermediate range of field values, is approximated by the interpolation formula for $\Gamma_\parallel$.

We also note that this paper is the first example of a quantitative description of the nonlinear dynamic response caused by the influence of an AC field on relaxation processes. A quantitative description of this response has not been attempted before even for the simplest systems, where the single-particle approximation is applicable. Our work here show the effectiveness of the use of experimental approaches of this kind in investigating concentrated magnets with exchange interaction, at least in the critical paramagnetic vicinity of the phase transition point.

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