

Linear and nonlinear ac susceptibility of the canted-spin system: $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$

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The linear and nonlinear ac susceptibilities of the canted-spin system $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$ have been measured using the mutual-inductance-bridge method as a function of temperature (50–100 K), frequency (19–1370 Hz) with different static biasing fields (12–36 Oe) as well as ac fields in the range of 4 to 24 Oe. An attempt has been made to compare the “canted-spin” system with the so-called “reentrant spin-glass system” from the ac-susceptibility point of view, since the “canting” plays a significant role in the reentrant spin-glass system, below the Curie temperature.

I. INTRODUCTION

A number of crystalline as well as amorphous magnetic systems (e.g., Ni-Mn, Au-Fe, Cr-Fe, Fe-Al, amorphous Fe-Zr, FeNiSiB, CrFeSi, etc.) are found to exhibit “reentrant” behavior depending on the concentration. This regime shares the characteristics of both spin-glass (SG) and ferromagnetic (FM) ordering.^{1,2} Theoretical investigations of the SG transitions which are generally used to explain the experimental results are based particularly on the mean field model suggested by de Almeida and Thouless³ (AT) and by Gabay and Toulouse (GT).⁴ Accordingly, when $\mathbf{J}_0 \geq \mathbf{J}$, the longitudinal FM state is followed by canted FM state, characterized by a freezing of the transverse components of spin accompanied by weak irreversibility at some critical temperature T_{GT} . At a lower temperature T_{AT} ($T_{AT} < T_{GT}$), this transition is followed by a crossover from weak to strong irreversibility. Thus it is clear that below T_C (for $T < T_{GT}$) the spin becomes locally “canted.” A very similar picture was put forward by Mookerjee.⁵ The role of spin canting and ferromagnetism in the reentrant spin-glass (RSG) alloy AuFe has been investigated through Mössbauer and other magnetic measurements.⁶ From an experimental point of view, as far as the ac susceptibility (ACS) is concerned, RSG systems are characterized by a sharp rise in the in-phase component $\chi'_1(T)$ at a T_C signifying a (PM) paramagnetic-FM transition, followed by a drop in the same at a less well defined temperature T_f , indicating spin freezing.

Recently, we performed an ac susceptibility experiment in the intermetallic compound UCu_2Ge_2 ,⁷ where a FM to antiferromagnetic (AFM) transition occurs via a canted phase. This system is of interest because in contrast with alloys and disordered systems as mentioned above, here the magnetic transitions are taking place within an ordered compound. Our results in the broad regime of FM to AFM transitions suggest features which are observed commonly in the SG-RSG systems. As a consequence, we believe that it will be interesting to perform experiments

on systems where a well defined canting phase exists in the region of coexistence of ferromagnetic and antiferromagnetic moments as characterized both by bulk and neutron measurements. We identify CeFe_2 substituted with Al, $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$ as one such system. The interest in the material stems from the fact that the transition from the FM ($T_C \sim 200$ K) state to the AFM ($T_N < 40$ K) state is not a sharp one, rather taking place gradually through a regime of increasing canting of the moments, separating FM and AFM phases.^{8,9} It should be emphasized here that this canting is not random and in contrast to pure SG systems its long range order has been established by neutron measurements.⁸

Therefore, this system is not a spin-glass system. Thus, macroscopic properties of many systems (e.g., canted, RSG systems) undergoing multiple magnetic phase transitions may show some common features (e.g., a drop in ACS experiments) although the microscopic natures and underlying physics could be quite different. In general at a canting transition $|\mathbf{m}|$ is conserved but $\langle m_z \rangle$ decreases, whereas in RSG systems a transverse spin-glass-like order sets in, keeping the FM order parameter unaffected, so that $\langle m_z \rangle$ is conserved but $(|\mathbf{m}|)$ increases.¹⁰

This motivated us to investigate experimentally in detail linear and nonlinear ac susceptibility for the “canted” system $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$. We particularly choose nonlinear ac susceptibility as such type of measurements have received considerable attention in recent times in SG-RSG systems. This is because a possible phase transition in SG systems could only be proved or disproved if one concentrated on the nonlinear susceptibility as the divergent behavior suggests a phase transition.^{11–14} Also it may be mentioned that in the thermodynamic sense whether the transition to RSG state is a true phase transition or not is still not clear. To our opinion such an approach will be useful to compare RSG, intermetallic compounds like UCu_2Ge_2 with canted spin systems as the canting plays an important role in RSG systems below the Curie temperature. Indeed this detailed study

has been undertaken exclusively focusing on the canted spin system.

II. EXPERIMENTAL DETAILS

The samples used in the present experiment are the same as in Ref. 9 and the typical dimensions are $l = 4.61$ mm, $b = 1.24$ mm, $d = 1.06$ mm, and $wt = 35.13$ mg. The ac susceptibilities χ'_1 and χ''_1 (linear components) were measured using a L'atne mutual inductance bridge. The sample movement and the data acquisition were controlled by a Z-80 microprocessor and the details of the apparatus are given in Ref. 15. The third harmonic was measured by feeding the "noise" (after balancing the linear components) output from the adder stage of the bridge to the input of a lock-in amplifier (LIA). The reference of the LIA was driven externally by a square wave signal generated (with zero crossings matched with the primary current) by another microprocessor. This digitally simulated phase-locked loop was capable of generating higher harmonics both phase locked and 90° phase shifted with respect to the primary current. In this way both in-phase and quadrature components of the higher harmonics could be measured.

Focusing on the nonlinear term, we can express in general the magnetization in the presence of a magnetic field for a ferromagnet as

$$m = m_0 + \chi_1 h + \chi_2 h^2 + \chi_3 h^3 + \dots, \quad (1)$$

where m_0 is the spontaneous magnetization, χ_1 is the linear susceptibility, and χ_2 and χ_3 are the nonlinear susceptibilities. It is important to note that χ_2 can be observed only if there is a spontaneous magnetization because for a FM m has no inversion symmetry with reference to the applied field. Thus for a direct PM-SG transition m is expressed as the odd power series of h as

$$m = \chi_1 h + \chi_3 h^3 + \dots. \quad (2)$$

The nonlinear term in the ac-susceptibility experimental configuration can be defined as¹⁶

$$\begin{aligned} \chi_2^t h_0 &= \chi_2 h_0 + \chi_4 h_0^3 + \dots, \\ \frac{3}{4} \chi_3^t h_0^2 &= \frac{3}{4} \chi_3 h_0^2 + \frac{15}{16} \chi_5 h_0^4 + \dots, \end{aligned}$$

where χ_2 and χ_3 are the coefficients of h^2 and h^3 in the expansion of the magnetization m [see Eq. (1)]. The nonlinear susceptibilities χ_2^t and χ_3^t are observed in ac-susceptibility experiments as $\chi_2^t h_0$, $\frac{3}{4} \chi_3^t h_0^2$ proportional to higher harmonic signals of frequencies 2ω , 3ω , respectively. However, unlike the case of χ_1 , i.e., $\chi_1 = \chi'_1 + i\chi''_1$, where we present both the in-phase and out-of-phase components separately, we present only the magnitude of $|\chi_3^t|$ for the higher harmonic term χ_3 . For small h_0 the higher-order terms like χ_4 , χ_5 , etc., which are usually small, can be neglected. Thus the measurements of χ_3 (3ω signal) and χ_2 (2ω signal) can reveal whether the system shows SG-like behavior and if so whether the SG state coexists with FM state, i.e., whether long range order is present.¹¹ The presence of a distinct divergentlike

peak in the nonlinear susceptibility χ_3 (i.e., 3ω signal) is an intrinsic feature of the RSG transition and can be considered to be one of the criteria for choosing T_f experimentally. The static dc field H_{dc} , parallel to H_{ac} was produced using a solenoid coil. Both the ac and dc fields were applied along the length of the sample.

III. RESULTS AND DISCUSSION

The cubic CeM_2 , Laves-phase intermetallic compounds ($M = Fe, Co, Ni$) have anomalously small lattice constants compared with those of the corresponding compounds of all other rare earths. $CeCO_2$ and $CeNi_2$ are paramagnets while $CeFe_2$ has an ordered moment. Also the ferromagnetic cubic Laves-phase compound $CeFe_2$ is a special case among the AFe_2 compounds ($A = Y, Zr, Sc, Lu$). Its Curie temperature T_C and the iron magnetic moment μ_{Fe} ($T_C = 230$ K, $\mu_{Fe} = 1.24\mu_B$) are somewhat smaller than those of other similar compounds. Eriksson *et al.*¹⁷ suggested that the $4f$ electrons in CeM_2 are itinerant in nature and hybridize with $3d$ electrons giving rise to various anomalies. The $3d$ - $4f$ hybridization plays an important role in making alloying substitution on either site of $CeFe_2$ important. Indeed metallic impurities such as Al on the Fe sites destabilize ferromagnetism, even in some cases causing a total loss of magnetism.^{8,9}

A neutron study by Kenedy and Coles⁸ suggests for the parent compound $CeFe_2$ the existence of ordered moments developed on both Fe and Ce sites at T_C which are coupled antiparallel so that $\mu_{Ce}/\mu_{Fe} = -0.3$. But in $Ce(Fe_{1-x}Al_x)_2$ ($x = 0.02-0.08$) no Ce moment contributions are seen in the FM phase. At a lower temperature AFM components appear accompanied by lattice distortion although its development is inhibited by the presence of the FM component. In their study the broad region of overlap between the two magnetic phases is the result of an extended region of canted spins. For $Ce(Fe_{1-x}Al_x)_2$, $x = 0.035$, close to that in the present work, the overlapping region of temperature is $\sim 50-120$ K. It is significant to note that the lattice distortion appearing with the antiferromagnetic components is prominent in the overlapping region. This spin-canted phase of these compounds represents a continuous spin reorientation involving changing magnitudes of ferro- and antiferromagnetic components. It is also clear from the measurements of a low-temperature canted-spin phase in $CeFe_2$ that the tendency towards antiferromagnetic order is present in the parent compound itself and the metallic substitution simply enhances this tendency. The present ACS investigation covers this region of overlap of two magnetic phases in Al-substituted $CeFe_2$.

For the present investigation we have chosen to study the behavior of the real and imaginary (absorption) parts of the linear components χ'_1 , χ''_1 and nonlinear components χ_2^t , χ_3^t of the ac susceptibility as a function of temperature, frequency, and amplitude of the ac field as well as different dc biasing fields. These types of experiment are generally used to characterize the SG-RSG system. This will enable us to compare the ideal SG-RSG systems.

Figure 1 shows the temperature dependence of the lin-

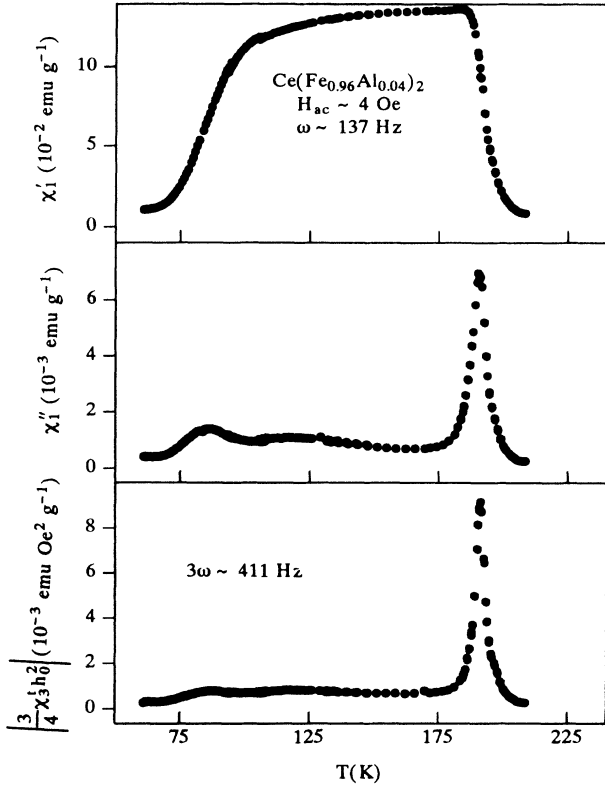


FIG. 1. The linear susceptibilities $\chi_1'(T)$, $\chi_1''(T)$ and nonlinear ac susceptibility $|\frac{3}{4}\chi_3^t h_0^2|$ of $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$ as a function of temperature measured at a fundamental frequency ω (137 Hz) at 4 Oe.

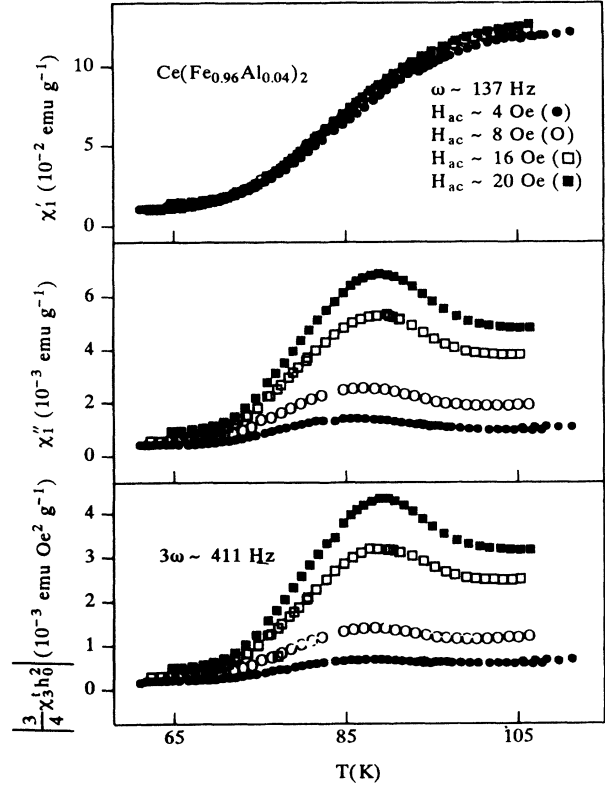


FIG. 2. The linear and nonlinear ac susceptibilities measured at different ac fields for a fundamental frequency ω (137 Hz).

ear χ_1' , χ_1'' susceptibilities and nonlinear susceptibility $|\frac{3}{4}\chi_3^t h_0^2|$. The sharp rise in χ_1' as well as a peak in χ_1'' and χ_3^t at $T = 190$ K corresponds to the PM-FM transition.⁹ Around 100 K $\chi_1'(T)$ starts dropping and reaches zero at around 60 K. This drop in χ_1' is accompanied by a peak in $\chi_1''(T)$ as well as χ_3^t at $T \sim 85.3$ K. In order to understand the origin of this peak, we focus our attention in this low-temperature regime rather than the well defined PM-FM transition region near $T_C \sim 190$ K.^{8,9,18}

Figure 2 shows the linear and nonlinear ACS components as a function of temperature at different ac fields in the range of 4–20 Oe, measured at $\omega \sim 137$ Hz. It appears from the $\chi_3^t(T)$ (3ω signal) and $\chi_1''(T)$ data that the peak shifts to higher temperature with increasing ac field. The peak position occurs at around $T \sim 88.7, 87.6, 86.3, 85.3$ K for $H_{ac} \sim 20, 16, 8, 4$ Oe, respectively.

Figure 3 shows the frequency dependence of χ_1' , χ_1'' , χ_3^t measured in the frequency range 19 Hz–1.37 kHz at $H_{ac} \sim 16$ Oe. This peak temperature ~ 88 K does not show any appreciable shift with frequency within our temperature measurement accuracy ≤ 0.05 K. In an ideal SG system one would expect the frequency dependence.¹² In materials like UCu_2Ge_2 also we found the peak temperature in χ_3^t shifts as much as 5 K in the same frequency range 19 Hz–1.37 kHz.

The second harmonic components $|\chi_2^t h_0|$ as defined earlier have been measured as a function of temperature for different ac fields in the range 8–24 Oe at $2\omega \sim 274$

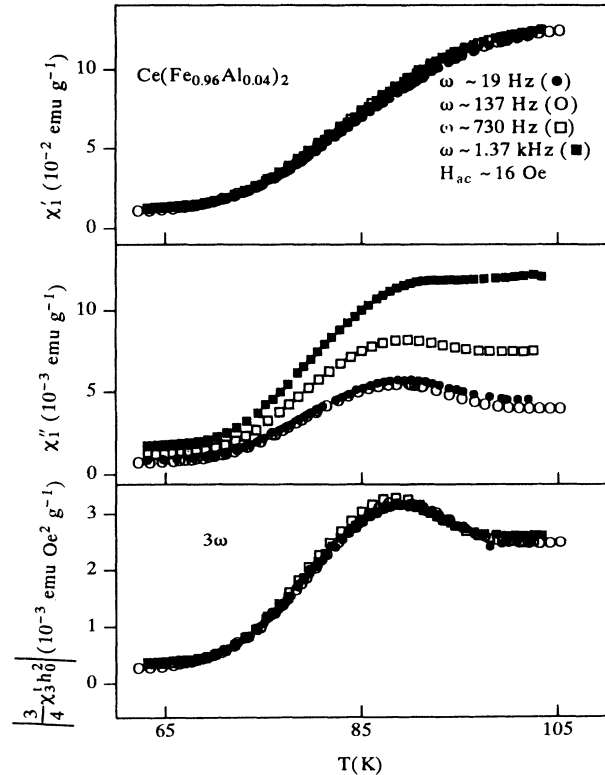


FIG. 3. The linear and nonlinear ac susceptibilities measured at different frequencies for H_{ac} (16 Oe).

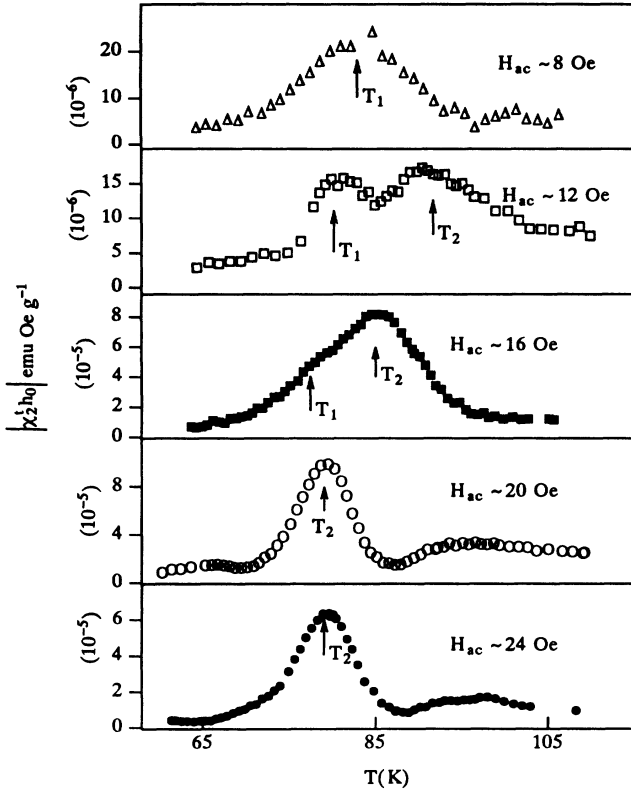


FIG. 4. The temperature dependence of the second harmonic signal $|\chi_2^t h_0|$ ($2\omega = 274$ Hz) measured at different ac fields for a fundamental frequency ω (137 Hz).

Hz, and the results are shown in Fig. 4. It may be noted that the temperature regime of interest (60–100 K) lies in the FM-AFM overlapping region with the presence of long range order. The present experimental observation of the presence of χ_2^t term supports the presence of long range order. The χ_2^t results show two well defined peaks T_1 , T_2 at temperatures $T_1 = 82.3, 80, 78.5$ K and $T_2 = 101.2, 88.7, 86.5$ K for $H_{ac} = 8, 12, 16$ Oe, whereas for 24 and 30 Oe the lower-temperature peak T_1 is not as significant but the peak T_2 occurs at 80.8 and 80 K, respectively. We have observed such double peaks in the RSG-like alloy $(\text{NiFe})_{25}\text{Au}_{75}$.¹¹ Since no theoretical guidance is available concerning the second harmonic component, the results are not analyzed further, but it certainly requires further attention.

In Fig. 5 we show the results of the measurements of $\chi_1'(T)$, $\chi_1''(T)$, $|\frac{3}{4}\chi_3^t h_0^2|(T)$ at an ac field ~ 4 Oe, $\omega \sim 137$ Hz superimposed with dc fields ranging from 12 to 36 Oe. In $|\chi_3^t|$ the peak temperature, 85.3 K for $H_{ac} \sim 4$ Oe does not show any significant change with the change of dc field. Here the dc field is applied under a zero-field-cooled (ZFC) condition. For a number of systems like AuFe ,¹⁹ $(\text{Fe}_x\text{Ni}_{1-x})_{(1-y)}\text{Mn}_y$,^{20,21} NiMn ,²² the presence of an anomalous double-peaked structure in the temperature dependence of the ACS in presence of static biasing field in the vicinity of the RSG transition is considered to be the reminiscent of the two reentrant phase boundaries

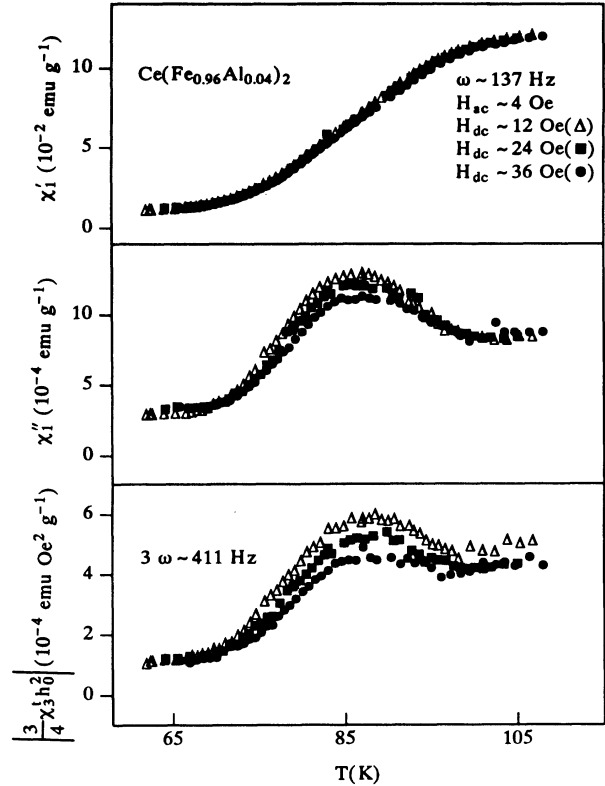


FIG. 5. The temperature dependence of the linear and nonlinear ac susceptibilities measured at H_{ac} (4 Oe) superimposed with different static biasing fields at a fundamental frequency ω (137 Hz).

(AT,GT) predicted by the isotropic vector spin model. Thus, the real and imaginary components χ_1' , χ_1'' clearly show strong irreversibility at the T_{AT} transition as can be seen from ACS experiments, performed under ZFC and FC conditions in an external static field. Similar measurements are shown in Fig. 6 where FC and ZFC data do not show any feature as in RSG systems. The starting point of our discussion is based on the nonlinear susceptibility results of SG and RSG systems. Suzuki²³ has proposed for the SG system that the third derivative of magnetization with respect to magnetic field, $\chi_3 = \frac{\partial^3 m}{\partial h^3}$, diverges at T_f and its sign is negative as $h \rightarrow 0$. Focusing on the nonlinear susceptibility in RSG systems, it has been found from the analysis of susceptibility isotherms in the vicinity of the RSG transition that a weak, nondivergent, anomalous peak in the nonlinear susceptibility occurs at T_f . This is based on the experimental evidence of the RSG systems in NiMn, PdMn, amorphous FeZr, etc. The model simulations indicate that it is the longitudinal response to transverse SG freezing. This is consistent with Mössbauer data on FeZr.²² Moreover, Katori and Suzuki²⁴ have predicted theoretically that the longitudinal susceptibility in vector spin systems should exhibit a complementary nonlinear anomaly in response to transverse spin freezing. It may be recalled that experimentally the nonlinear susceptibility in the RSG regime

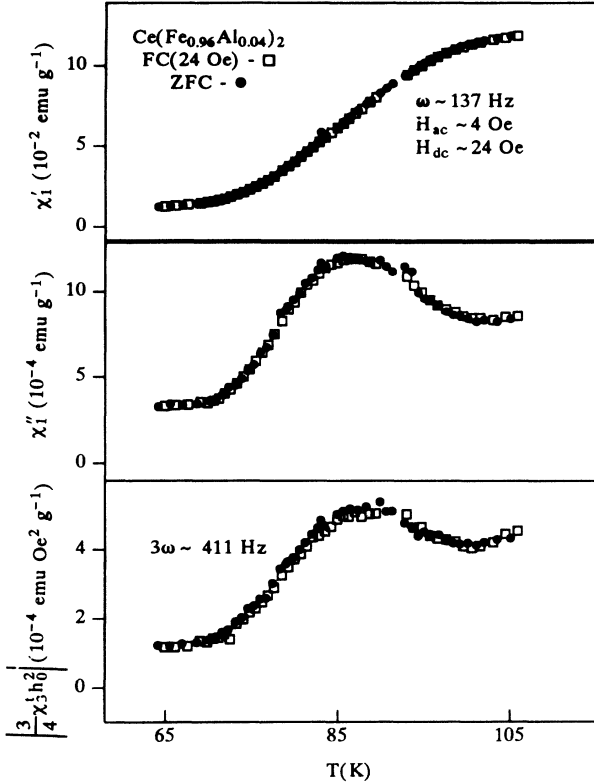


FIG. 6. The linear and nonlinear ac susceptibilities measured under zero-field-cooled (ZFC) and field-cooled (FC) conditions ($H_{dc} = 24$ Oe) at H_{ac} (4 Oe) at a fundamental frequency ω (137 Hz).

has not much been investigated even though considerable attempts have been made to understand the direct PM-SG transition. The present consensus on RSG system is as follows. Based on the mean field Heisenberg SG model⁴ with short range order and anisotropy, Campbell and Senoussi¹ argued that for a Heisenberg system with predominantly ferromagnetic interaction but with a proportion of antiferromagnetic bonds FM order first sets in at T_C with domain structure and at a lower temperature T_k local transverse spin components begin to freeze in with each of the individual spins, more or less canted with respect to the local domain magnetization direction. In zero field there is a FM domain structure for all temperatures below T_C . So in the low-temperature state both FM and SG order coexists. The drop in $\chi'_1(T)$ in ACS experiments at a lower temperature in RSG systems can be explained by the hindrance of domain wall motion in the presence of the Dzyaloshinsky-Moriya (DM) anisotropy which appears with the canting of spins. The physical picture is supported by Mössbauer, electron spin resonance (ESR), and magnetization experiments.

Focusing on our $\chi'_1(T)$, $\chi''_1(T)$ data it is known that below T_C a large number of FM domains is formed within which the spins are ferromagnetically coupled and aligned in a particular direction. The FM state responds to low fields by domain wall movements. At a lower temperature when the spins get canted an interaction of the

DM type containing the cross product term $(\mathbf{S}_i) \times (\mathbf{S}_j)$ must appear. Because of the presence of this term, the response of the domain wall to the field is inhibited and finally the wall motion is hindered. Hence we observe the low-temperature drop in $\chi'_1(T)$, as well as a peak in $\chi''_1(T)$. As we increase the amplitude of the ac field the low-temperature peak in $\chi''_1(T)$ as well as in $|\frac{3}{4}\chi_3^t h_0^2|$ data shifts to higher temperature as seen in Fig. 2. This may be because the more we increase the magnitude of the ac field the more the domain wall motion becomes irreversible. The amplitude of $\chi''_1(T)$ (Fig. 6) shows systematic suppression with increasing dc field as observed in RSG systems.²⁵ We earlier pointed out that the observation of the peak in the nonlinear susceptibility in the RSG system is due to the longitudinal response to transverse spin freezing. However, in the absence of any such glassy behavior in the present study the origin of the peak in the nonlinear χ_3^t data is due to some typical nonlinearity of the magnetization curve of this canted spin system¹⁸ and the ACS technique measures actually the slope $\frac{\partial m}{\partial h}$ of the $m-h$ curve. Indeed the magnetization data on canted systems like $\text{Fe}(\text{PdPt})_3$ show nonlinearity at the canting transition.²⁶ In $\text{FePd}_{1.6}\text{Pt}_{1.4}$ the magnetization instead of rising monotonically below $T_C \sim 270$ K reaches a sharp maximum near 145 K and then gradually decreases. The magnetic transition at ~ 145 K corresponds to a rapid but continuous change from a simple FM to a ferrimagnetic state with canted Fe moments. The ACS results, linear as well as nonlinear, should exhibit a significant change in $\chi'_1(T)$, $\chi''_1(T)$, and $\chi_3^t(T)$ data at this temperature.

The results of the ACS experiments done in the presence of superimposed dc fields under ZFC and FC conditions as well as the frequency dependence of linear and nonlinear susceptibilities may be useful to compare the “canting” and the SG-RSG systems from the macroscopic point of view. Such types of experiment for RSG systems reveal irreversible behavior at temperatures below T_{AT} whereas there is no reason to expect such behavior for the canting systems. For our system ZFC and FC results do not show any difference. Kunkel *et al.* has also recently shown that for CeFe_2 the nonlinear term deduced from the susceptibility data does not show any double-peaked structure as observed in RSG systems.²⁷ It is not clear also why the nonlinear data for UCu_2Ge_2 show a frequency dependence like SG-RSG systems, whereas the present data do not show any features in the same frequency range of investigation. However, caution has to be exercised in that GT and AT transitions are derived from the Ising or the Heisenberg model, while strictly speaking these models are not suitable for an itinerant system in which the spin fluctuates not only in the transverse direction but also in the longitudinal direction.

IV. CONCLUSION

Measurements of low field linear and nonlinear ac susceptibility as a function of ac field ($H_{ac} = 4\text{--}20$ Oe), frequency (19 Hz–1.37 kHz) biasing with a static dc field ($H_{dc} = 12\text{--}36$ Oe) under FC and ZFC conditions have been reported on the “canting” system $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$.

This canting is not random, its long range order being clearly shown in neutron measurements. The drop in the in-phase component $\chi_1'(T)$ along with a peak in the out-of-phase component $\chi_1''(T)$, nonlinear term $\chi_3^t(T)$ appears to be the same as in a RSG system. However, we believe the origin of the peak in the nonlinear term $\chi_3^t(T)$ is due to some typical nonlinearity of the magnetization below the canting transition. Also the careful study of such effects as frequency dependence, FC, and ZFC data with a superimposed static field does not show any of the significant features which are commonly observed in SG-RSG samples, such as irreversibility. Such a study has been undertaken exclusively for canting systems with a view to comparing the canting system with an ideal

“RSG” system in their ac-susceptibility behavior, as this technique is commonly used to characterize the magnetic phase transition. However, in the absence of any theoretical work on the ac response of canting systems, it is difficult to draw any detailed conclusions concerning its origin.

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