Competing interactions and spin-glass-like features in the UCu_2Ge_2 system

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The experimental results of low field, frequency-dependent complex ac susceptibility including both the linear and nonlinear responses are reported for the UCu_2Ge_2 system, where the ferromagnetic long-range order is observed below $T_c \approx 105$ K and antiferromagnetic transition occurs at $T_N \approx 45$ K. The data show various spin-glass-like features in the intermediate FM-AFM temperature regime.

I. INTRODUCTION

The neutron diffraction investigations[1] and the magnetic properties of the uranium intermetallic system UCu_2Ge_2 (Ref. 2) with the $ThCr_2Si_2$-type crystal structure show some unusual behavior. The interest in this material stems from the fact that it shows the onset of ferromagnetism (FM) at a fairly high temperature ~105 K followed by a transition to antiferromagnetism (AFM) at low temperatures ~45 K. It is apparent from recent measurements of dc magnetization and low-field susceptibility that the transition from FM to AFM state is a gradual one over a large temperature range.[3] In spite of the evidence for an AFM state below 45 K from neutron measurements there remain several questions regarding the exact nature of this FM-AFM transition and especially of the magnetic phase in the transition regime.[3]

In this communication we present the results of detailed measurements of linear and nonlinear susceptibility in fields in the range of 4–16 Oe at different frequencies (19 Hz–1.37 kHz) for the temperature range 56–120 K, i.e., the FM-AFM regime. Our aim was to look for possible spin-glass-like features, as the study of the nonlinear susceptibility is an important experimental tool in identifying the spin glass (SG) and reentrant spin glass (RSG).[4–6] We have made a systematic study of the nonlinear susceptibility of a system showing metamagnetic behavior. (UCu_2Ge_2 shows such behavior below 70 K.[3]) In the following sections we present our data for the field and frequency dependence of the ac susceptibility and seek to interpret the results in the light of various theoretical models.

II. EXPERIMENTAL

Ac susceptibility has been measured for the same sample for which the dc magnetization and resistivity data were reported along with details of preparations.[3] In the present experiment the sample dimensions are 4.55 mm×1.18 mm×1.11 mm with weight 50.17 mg. The experiment was carried out for different ac fields in the range of 4–16 Oe using a L'ATNE mutual inductance bridge for different frequencies in the range of 137 Hz–1.37 kHz. The ac susceptometer with calibration details is described in Ref. 7. The mutual inductance bridge has been used to balance out the linear term χ_1, i.e., both the in-phase χ_1' and out-of-phase χ_1'' components at the fundamental frequency ω (19, 137, 1370 Hz). The nonlinear terms in the ac susceptibility experimental configuration can be defined as[8]

$$\chi_2^t h_0 = \chi_2 h_0 + \chi_4 h_0^3 + \cdots ,$$

$$\tfrac{3}{4}\chi_3^t h_0^2 = \tfrac{3}{4}\chi_3 h_0^2 + \tfrac{15}{16}\chi_5 h_0^4 + \cdots ,$$

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) in Sec. IV]. The nonlinear susceptibilities χ_2^t and χ_3^t are observed as higher harmonic components of frequencies 2ω and 3ω. These quantities including both in-phase and out-of-phase components have been measured by using the residual output of the bridge as the signal input of a PAR 124A lock-in amplifier (LIA). The reference of the LIA is driven at 2ω and 3ω with the help of a Z80A microprocessor-based phase-locked loop which also monitors the phase of the primary current and adjusts the phase (in phase and quadrature) of the reference signal accordingly. However unlike as in 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, for the higher harmonic term χ_3, we present only the magnitude $|\chi_3^t|$. The values of $|\chi_2^t|$ were within the our experimental error for the temperature range of interest, i.e., 55–90 K and are not shown. For small h_0 the higher-order terms like χ_4, χ_5, etc., which are usually small, can be neglected.

III. RESULTS

The ac susceptibility results for UCu_2Ge_2 sample measured at 4 Oe are shown in Fig. 1. The linear term, consisting of dispersion χ_1' and absorption χ_1'' at the fundamental frequency ω (137 Hz), and the nonlinear term $|\tfrac{3}{4}\chi_3^t h_0^2|$ corresponding to 3ω are shown in the same figure. The high temperature peak ~105 K corresponds to the PM-FM transition (T_c) and the sharp drop in $\chi_1'(T)$ for $T \le T_c$ is consistent with earlier measurements.[3] We believe that in the present work the most interesting feature is the low temperature anomaly in the temperature range 55–75 K, which is clearly distinct in

$\chi_1''(T)$ and in the nonlinear $|\chi_3'|$ data. In order to understand these features we restrict ourselves to this temperature region, rather than region near T_c which has been studied.[1,2] It may be noted that this temperature range under investigation corresponds to the FM-AFM regime for the sample where $T_c \approx 105$ K, $T_N \approx 45$ K. In Fig. 2 we present the data for $\chi_1'(T)$, $\chi_1''(T)$, $|\frac{3}{4}\chi_3' h_0^2|$ for $\omega \sim 137$ Hz in different fields. It is surprising to see that a well-defined peak in the latter two quantities appears at 16 Oe but is slightly less distinct at the fields 4 Oe, 8 Oe. The peak in $\chi_1''(T)$ and $|\chi_3'|$ occurs at $T \sim 65$ K for 16 Oe, $T \sim 69.5$ K for 8 Oe, and $T \sim 72$ K for 4 Oe. Moreover the peak in the nonlinear term is somewhat similar to the distinct peak that appears in the spin-glass problem particularly in the "reentrant" type of spin glass.[6,9] We discuss these aspects in detail in the subsequent section. In order to understand the origin of this peak in $\chi_3'(T)$ and $\chi_1''(T)$, we have extended our measurements to different frequencies in the range of 19–1370 Hz at 16 Oe. Here the highest field was chosen since the peak at 16 Oe is more distinct, so that the effect of frequency on this peak temperature can be more easily studied. We present the data for the frequency dependence in Fig. 3. However it is clear from Fig. 3 that the peak temperature in $\chi_1''(T)$ shifts to higher values as ω increases, i.e., the peak at 19 Hz is ~ 62.9 K, at 137 Hz is ~ 64.9 K, and at 1370 Hz is ~ 66.5 K. In $|\chi_3'|$ the peak temperature at 19 Hz is ~ 63.3 K, at 137 Hz is ~ 65 K, and at 1370 Hz is ~ 68 K. More importantly the temperature dependence of $|\chi_3'|$ below this peak temperature appears to be similar to that in ideal spin-glass systems.[4,5] Thus the field and frequency dependence of the peak temperature clearly suggests that the relaxation behavior which occurs in the FM-AFM regime may be akin to that in spin-glass systems. We wish to stress that the detailed frequency and field dependent ac susceptibility data, including the nonlinear part, is lacking for metamagnetic materials.

IV. DISCUSSION AND CONCLUSION

Moriya and Usami[10] theoretically discussed the various magnetic phase transitions possible in strongly interacting itinerant-electron systems and the possibility of the coexistence of ferro- and antiferromagnetism. They con-

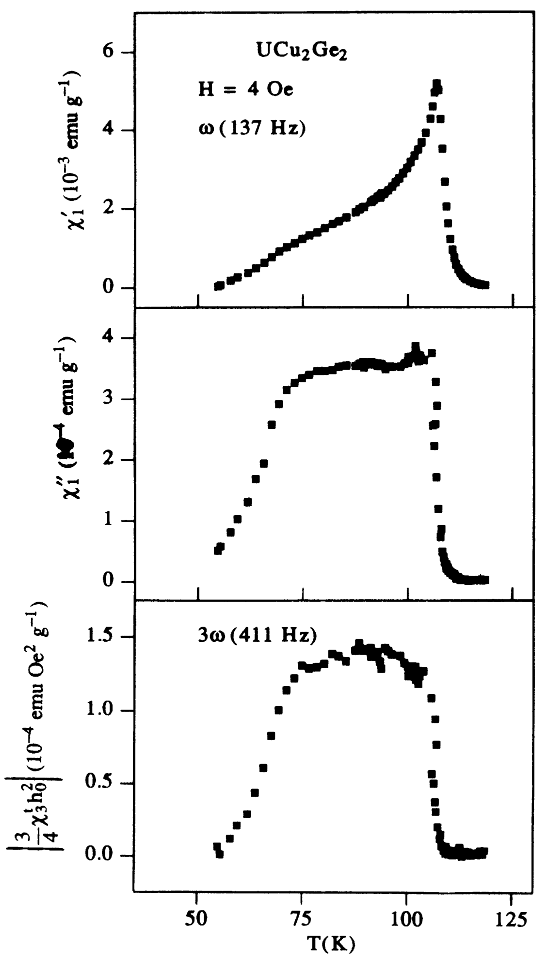

FIG. 1. The linear $\chi_1'(T)$, $\chi_1''(T)$, and nonlinear $|\frac{3}{4}\chi_3' h_0^2|$ ac susceptibility of UCu_2Ge_2 as a function of temperature measured at a fundamental frequency ω (137 Hz) at 4 Oe.

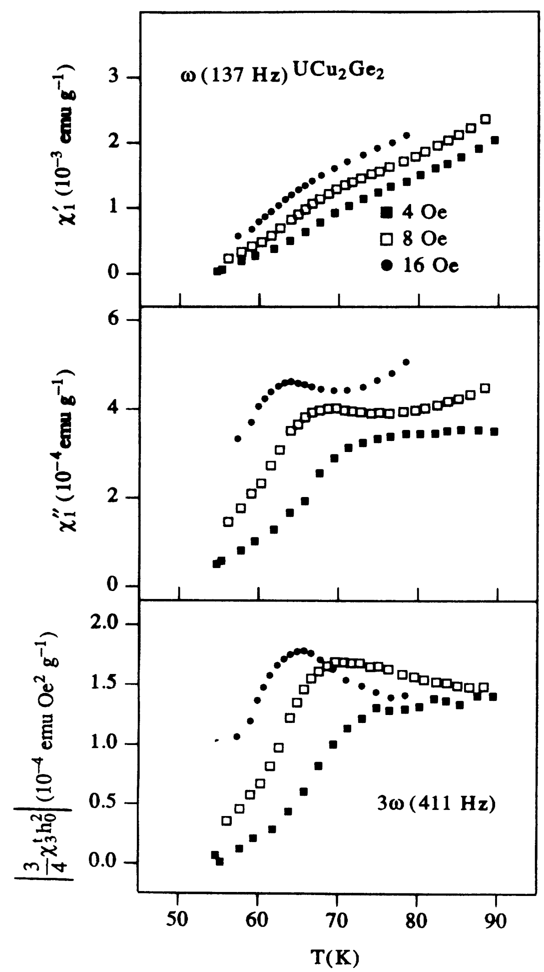

FIG. 2. The linear and nonlinear ac susceptibility measured at different fields (■) 4 Oe, (□) 8 Oe, (●) 16 Oe for a fundamental frequency ω (137 Hz).

sidered an itinerant electron system and expressed its free energy as a function of the uniform and staggered components of the magnetization, retaining terms up to fourth order. The coefficients of the terms of various orders can, in principle, be calculated for different systems if their electronic band structure is known. The possible equilibrium states, which Moriya and Usami obtained by minimizing the free energy, are rich in variety. The possibility they discuss of PM-AFM-FM transitions has already obtained experimental support for $(Hf_{1-x}Ta_x)Fe_2$ (Ref. 11) and $(Zr_{1-x}Nb_x)Fe_2$ (Ref. 12) hexagonal Laves phase compounds. In UCu_2Ge_2 which exhibits PM-FM-AFM transitions, the intermediate regime of coexisting FM and AFM seems to be quite interesting. In UCu_2Ge_2, the uranium $5f$ wave functions are quite extended and hybridize substantially with the conduction band, providing both itinerant character and strong electronic correlation, reminiscent of the strongly correlated electrons in the isostructural compound URu_2Si_2, a heavy fermion system. Thus the observation of metamagnetic behavior has a natural explanation within

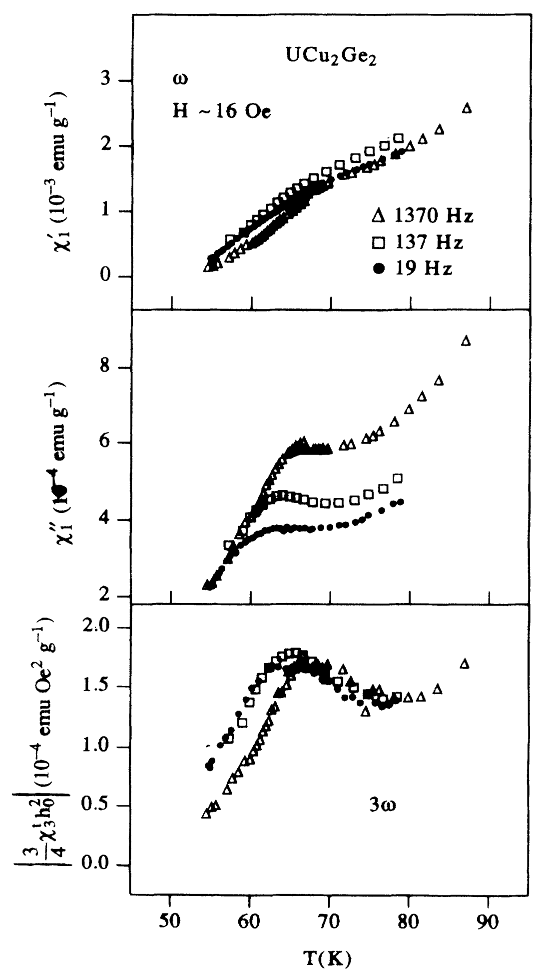

FIG. 3. The linear and nonlinear ac susceptibility measured at different frequencies (●) 19 Hz, (□) 137 Hz, (△) 1370 Hz for 16 Oe.

the Moriya-Usami's model,[10] as has also the influence of anisotropy, which has been subsequently treated by Isoda[13] as an extension of Moriya-Usami's model. Alternatively it is interesting to note that the behavior of the field and frequency-dependent parts of the linear and nonlinear susceptibilities of UCu_2Ge_2 in the so called FM-AFM transition regime has a striking similarity to that observed in what have been considered "reentrant" spin-glass systems.[9] Focusing on the nonlinear term we can express the magnetization in the presence of a magnetic field for a FM as[8]

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

where m_0 is the spontaneous magnetization, χ_1 is the linear susceptibility, χ_2 and χ_3 are the nonlinear susceptibilities. It is important to note that in Eq. (1) χ_2 can only be observed if there is 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 an odd power series in h as[14,15]

$$m = \chi_1 h + \chi_3 h^3 + \cdots . \qquad (2)$$

It has been shown both theoretically and experimentally that at least in some systems the nonlinear term χ_3 diverges at T_f characterizing the SG transition.[14,15] Recently we obtained the nonlinear terms χ_2 and χ_3 as the second and third harmonic components (2ω and 3ω) of the signal directly from the ac susceptibility experiment, rather than by estimating these quantities from $M(H)$ data for "reentrant" spin-glass systems (RSG) (for example, FeMnSi, NiFeAu) near T_f.[9] The idea behind such an experiment is that the presence of χ_2 (i.e., a 2ω signal) supports the existence of a FM moment. Also the distinct nondivergent peak in the nonlinear response, as in the case of NiMn (Ref. 16) and PdFeMn,[6] is an intrinsic feature of the RSG transition and can be considered to be one of the criteria for choosing T_f experimentally. Theoretical calculations suggest that the observation of this peak may be the longitudinal response due to the cooperative spin freezing in the sample.[17] However the applicability of these mean-field theories to real systems is not clear. Thus the unique measurements of χ_3 and χ_2 can reveal whether the system shows FM-SG behavior (RSG), and if so whether the SG state coexists with FM state, i.e., whether long-range order is present.

In UCu_2Ge_2, within our experimental accuracy (better than 10^{-5} emu/gm) we did not see any signature of the χ_2 term in the range 55–90 K which suggests that there is no effective internal field in this temperature range. On the other hand, the peak in χ_3^t for UCu_2Ge_2 is clearly distinct at ~16 Oe and is also frequency dependent (Figs. 2 and 3). It may be noted that in the χ_3^t term (Fig. 3), for $T < T_f$ the behavior is similar to that observed in many SG systems.[4,5] However in the absence of any detailed study of the field and frequency dependence of the χ_3 term in RSG systems, it is difficult to make any further comparison. The observation of higher harmonics in susceptibility was to be expected as magnetization measurements in UCu_2Ge_2 show nonlinear behavior.[2] However it is premature to say whether the observed similarity with the spin-glass systems is just a coincidence or there is a subtle link between these two (apparently) different families of magnetic system. A microscopic probe-like neutron measurement is required here to distinguish between spin-glass-like features and the presence of a canted regime in this UCu_2Ge_2 system. It is not clear what would provide the randomness and frustration in this system which is normally considered essential for spin-glass behavior. Even though the sharp drop in χ_1' (Fig. 1 and see, also, Fig. 1 in Ref. 3) suggests the presence of strong temperature-dependent anisotropy in the system, we see no reason to assume this anisotropy to be random in nature. However it should be noted here that the coexistence of FM and AFM phases is also possible within the framework of the anisotropic Ising model with competing interaction (ANNNI model) which has been discussed in detail.[2]

To conclude, we report the frequency dependence of the low-field complex ac susceptibility, including the nonlinear behavior, for the UCu_2Ge_2 system in the FM-AFM temperature regime. The highlight of the present work is that such a study has been considered for the first time (to our knowledge) in metamagnetic materials. The results reveal some spin-glass-like features. The role of competing interactions, the possibility of coexistence of FM and AFM order in terms of the Moriya and Usami model, the role of anisotropy and its relevance to ANNNI model are considered. However it should be stressed that microscopic neutron measurements are required in order to arrive at any definite conclusion. Other studies of detailed thermomagnetic history effects may be helpful in elucidating the complex magnetic behavior. We believe that it will be interesting to perform similar experiments on samples where a well-defined canting phase in the region of coexistence has been characterized by bulk and neutron measurements.[18,19] Such measurements for the related Al-doped $CeFe_2$ pseudobinaries are in progress..

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