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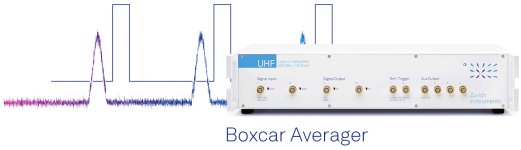
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Ac susceptibility studies of ferrimagnetic $FeCr_2S_4$ single crystals

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Ac linear and nonlinear susceptibilities, χ_0 and χ_2, of ferrimagnetic $FeCr_2S_4$ single crystals were measured in the temperature range from 4.2 to 300 K for different driving ac and applied dc magnetic fields in the frequency range of $10^{-1}-10^3$ Hz. For high driving ac fields the real part of χ_0 exhibits a cusp at around $T_m \approx 60$ K correlated with the onset of dc magnetization irreversibilities. The imaginary part of χ_0 shows a strong increase below 100 K and nonmonotonic temperature dependence with a maximum shifted toward low temperatures with an increase in the driving field. Both real and imaginary parts of the linear susceptibility, χ_0' and χ_0'', show a pronounced frequency dependence between 90 and 20 K with a maximal difference at around 60 K. Below the Curie temperature the real part of the nonlinear susceptibility, χ_2, exhibits a broad negative peak which is strongly shifted towards low temperatures with an increase in the driving field. No clearly divergent behavior of χ_2' around T_m is observed. The field and temperature dependencies of χ_0 and χ_2 are explained by domain wall pinning. Spin-glass-like magnetic anomalies at temperatures below 60 K are attributed to changes in the domain structure and the appearance of additional pinning centers suggested to result from a structural lattice transformation.
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INTRODUCTION

The ternary spinel magnetic semiconductor compound $FeCr_2S_4$ has recently attracted much interest due to its colossal magnetoresistance, similar to that of manganese perovskites,[1,2] and its half-metallic behavior.[3,4] It orders ferrimagnetically below 170 K and shows strong magnetocrystalline anisotropy with a saturation field of about 100 kOe at 4.2 K.[5–7] In the paramagnetic state the susceptibility follows Curie–Weiss law with an asymptotic temperature $\theta \approx -(260\pm 40)$ K,[5] indicating the dominance of strong antiferromagnetic Fe–Cr interactions.[8] Recently, spin glass-like (SGL) behavior was observed in the magnetic properties of $FeCr_2S_4$ single crystals at temperatures below $T_m \approx 60$ K, at which the low field magnetization exhibits a cusp.[9,10] Below T_m a difference between zero field (ZFC) and field cooled (FC) magnetization, coercivity and remanence appears. In addition, at temperatures, of $T<20$ K, a strong time-dependent relaxation, shifted FC hysteresis loops, *S*-shaped magnetization curves and a sharp increase in the coercivity were found. The irreversible magnetic effects in $FeCr_2S_4$ were attributed to disorder originating from cation defects. Even for small quantities, these defects were suggested to produce substantial fluctuations of exchange interactions similar to the well known spin-glass compound $CdCr_{2-2x}In_{2x}S_4$.[11–13] The role of high magnetocrystalline anisotropy and rearrangement of the domain structure with temperature have also been discussed.[10] However, the dc data did not allow one to distinguish between domain and spin glass effects. Ac susceptibility measurements are known to provide complementary information about the origin of such magnetization anomalies and the ground state of magnetic materials. In this article we report results of linear and nonlinear ac susceptibility measurements on the same $FeCr_2S_4$ single crystals that were studied in Refs. 9 and 10.

EXPERIMENT

Magnetic susceptibility was measured in the temperature range of $4.2\ \text{K}<T<250$ K utilizing an Oxford ac susceptometer and a superconducting quantum interference device (SQUID) magnetometer (Quantum Design). In-phase χ' and out-of-phase χ'' signals were recorded simultaneously by a two-channel lock-in amplifier operated at fundamental frequency ν and the third harmonic, 3ν. For low values of the driving field (typically 1 Oe_{rms}) the output signal at frequencies ν and 3ν is proportional to linear χ_0 and nonlinear χ_2 susceptibilities, respectively.[14] Measurements were performed on plate-like samples with ac and dc magnetic fields applied perpendicular to the (111) and (110) surface planes. The samples were cut from octahedron-like single crystals of about 3 mm edge length. Details of the single crystals' growth and of sample composition control are described elsewhere.[10]

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the real χ_0' and imaginary χ_0'' parts of the linear ac susceptibility for one of the single crystalline samples studied at a frequency of 1000 Hz for an ac driving field of 1 Oe_{rms}. Both the real

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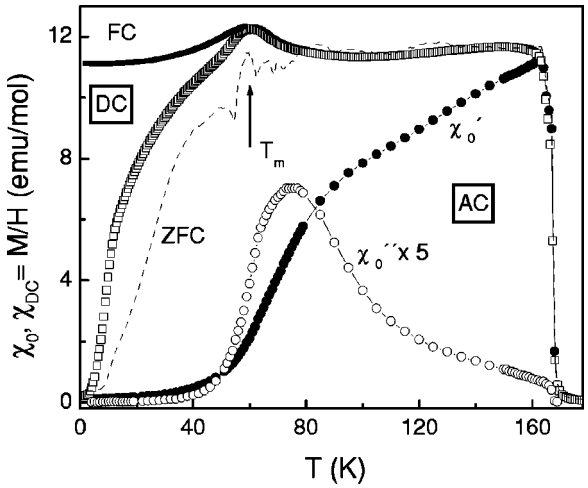

FIG. 1. Temperature dependences of the real χ_0' and imaginary χ_0'' parts of the linear ac susceptibility and of the dc ZFC and FC susceptibilities M/H of a $FeCr_2S_4$ single crystal (sample 35x, plane {111}). Open and closed squares represent dc data for M/H for magnetic field of 100 Oe, for 2.5 Oe (dotted line).

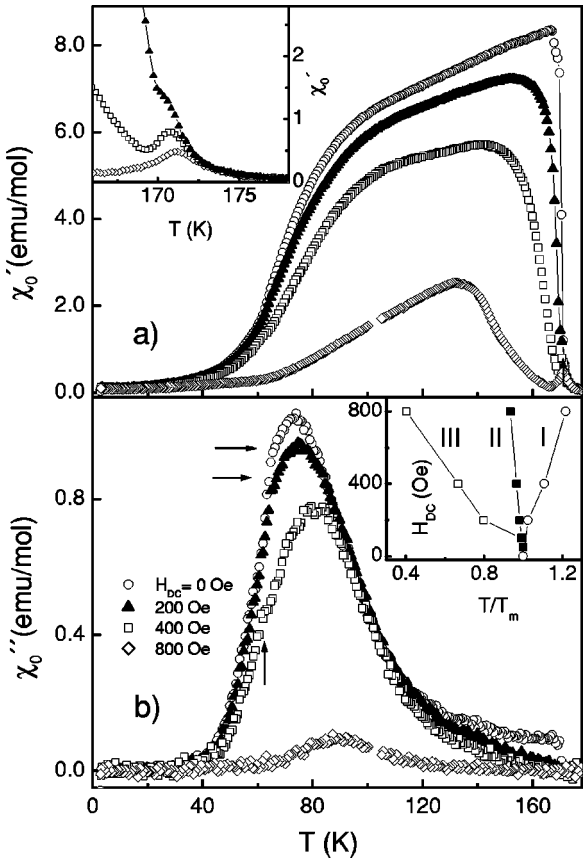

FIG. 2. Temperature dependences of the real χ_0' (a) and imaginary χ_0'' (b) parts of the linear ac susceptibility of a $FeCr_2S_4$ single crystal (sample 35h, plane {110}) at different applied dc magnetic fields H_{dc}. Driving field H_{ac} = 1 Oe, frequency ν = 1000 Hz. The arrows in (b) indicate the temperatures at which the dc and ac susceptibilities exhibit an anomaly at around T_m. Inset in: (a) Peaks in the region of the Curie temperature. Inset in: (b) Temperature shifts of the maximum of the imaginary part of susceptibility χ_0'' (curve I), of the maximum of dc ZFC susceptibility (curve II) and of the splitting point of dc FC and ZFC susceptibility (curve III) as a function of H_{dc}.

and imaginary parts of the susceptibility exhibit nonmonotonic behavior with the temperature. Between 170 and 160 K the real part of the susceptibility is typical for a material at the phase transition from a paramagnetic to a long-range (ferrimagnetic) ordered state. It shows the Hopkinson maximum[15] below the Curie temperature, $T_C \approx 167$ K, determined by the Kouvel–Fisher method.[16] With a temperature decrease down to 130 K, χ_0' decreases linearly and then below 100 K more rapidly. The imaginary part, χ_0'', which characterizes magnetic losses, increases smoothly between 160 and 130 K. Below 100 K, χ_0' increases strongly, reaching a maximum at the temperature at which the real part χ_0' shows an inflection point. For this ac driving field magnitude no cusp around $T_m \approx 60$ K was recorded for χ_0', in contrast to the dc susceptibility also presented in Fig. 1. Even at the lowest frequency available ($\sim 10^{-2}$ Hz) a significant difference between the ac susceptibility χ_0'' and the ZFC dc susceptibility was observed below T_c. We will show that this behavior can simply be attributed to the difference in values of ac driving and dc applied magnetic fields.

Figure 2 demonstrates the variation of the real and imaginary parts of the linear susceptibility for different applied dc magnetic fields. The usual Hopkinson maximum in the real part of the susceptibility for zero dc field is modified due to magnetocrystalline anisotropy. This maximum becomes broadened and is finally suppressed by application of the DC magnetic field [see Fig. 2(a)]. At the same time, close to T_c a sharp peak in χ_0' appears that is shifted toward higher temperature with an increase in the dc magnetic field [see the inset of Fig. 2(a)]. Peaks of this type were observed earlier in amorphous Fe–Ni and Fe–Zr re-entrant spin glasses[17–19] and were attributed to critical fluctuations.[17,20] In the absence of a dc field a sharp kink in the imaginary part is observed at the Curie temperature. This kink is suppressed by applying a dc magnetic field. The loss peak of χ_0'' detected for $60\,\text{K} < T < 90\,\text{K}$ is shifted towards higher temperatures with an increase in the dc field [see the inset in Fig. 2(b), curve I] in contrast to the behavior of a canonical spin glass.[21] The temperature at which the cusp in the dc susceptibility is observed is nearly independent of the applied field (see curve II in the inset of Fig. 2). The splitting point of the FC and ZFC curves in the dc data, however, shifts toward low temperatures with an increase in the applied magnetic field (see curve III in the inset of Fig. 2) the same as for canonical spin glasses.[21] Overall, the magnetic ac response deviates from pure SG behavior. It may be attributed rather to relaxation of the domains in a ferromagnetic (FM) or mixed state in which SG and FM coexist.

Since spin glass and a ferromagnet exhibit different frequency dependence of χ_0 for freezing of the spins and domains, respectively, the susceptibility was measured as a function of frequency in the range of 10^{-1}–10^3 Hz. In Fig. 3 the temperature dependences of the linear susceptibilities χ_0' and χ_0'' are presented for various frequencies for an ac driving field of 1 Oe without a dc magnetic field. Between T_c and 100 K both χ_0' and χ_0'' depend only very weakly on frequency as expected for a ferro-or ferrimagnet in a long-range ordered state.[21] However, below 90 K, when the real

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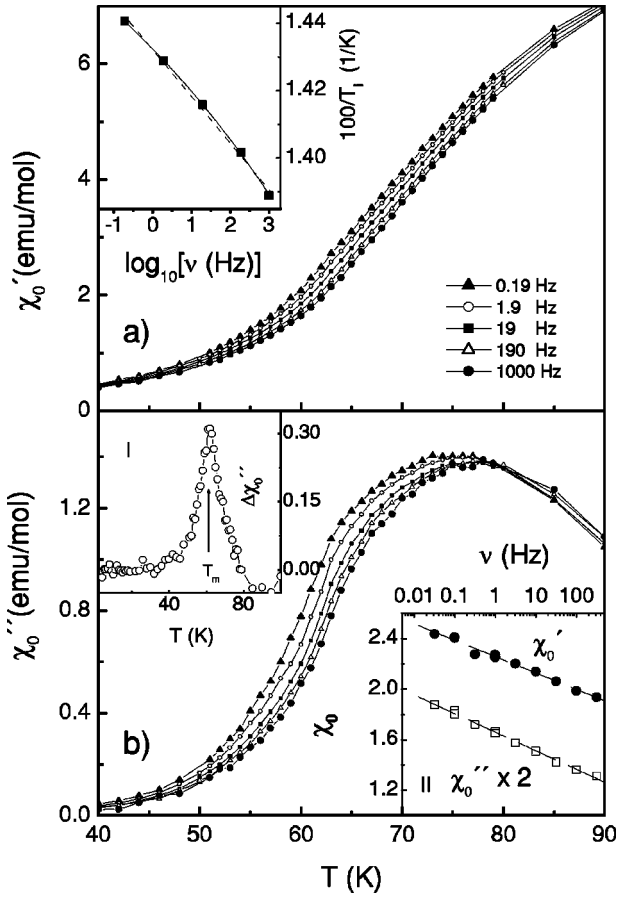

FIG. 3. Real χ_0' (a) and imaginary χ_0'' (b) parts of the linear ac susceptibility for sample 35x at different frequencies as a function of temperature. Inset in: (a) Frequency shift of the inflexion point of the real part of the susceptibility χ_0' vs the inverse temperature fitted by Arrhenius (dotted line) and Vogel–Fulcher (solid line) laws. Inset in: (b) Temperature dependences of the difference of the imaginary part of the susceptibility χ_0'' for the highest and lowest measured frequencies (I). Frequency dependence of the real χ_0' and imaginary χ_0'' parts of the susceptibility at a temperature of 61.5 K on a semilogarithmic scale (II).

part χ_0' starts to decrease sharply with the temperature, a pronounced frequency dependence is found. The imaginary part of the susceptibility χ_0'' shows similar frequency dependence between 90 and 20 K. The inflection point of $\chi_0''(T)$ is shifted toward higher temperatures with an increase in frequency. Below 20 K, χ_0 is frequency independent. No anomaly in the ac susceptibility χ_0' was detected in the temperature range of $4.2\,\mathrm{K}<T<20\,\mathrm{K}$, in contrast to the dc data, which show a strong decrease of ZFC magnetization accompanied by a similar increase of remanence and coercivity in this range.[10] Obviously, below 20 K, the pinning barriers for the domains are already too high to allow any contribution to the low field ac response.

To estimate the shift of χ_0 as a function of the frequency of the driving field in the absence of a clear anomaly of χ_0' at T_m and a relatively broad maximum of χ_0'', we used the temperature T_i at which the real part χ_0' shows an inflection point. The dependence of T_i^{-1} on the frequency is shown in the inset of Fig. 3(a). From the maximal shift ΔT_i we get $\Delta T_i/[T_i\Delta(\log\omega)]\sim 0.015$, which is closer to the value of the frequency shift of the cusp in χ_0' in spin glasses than for superparamagnets (see Table 3.1 in Ref. 21). Fitting of the experimental data to a thermally activated process, described by the Arrhenius law $\omega=\omega_0\exp(-E_a/k_BT)$ [see the dashed line in the inset of Fig. 3(a)], yields unrealistic values of $\omega_0\approx 10^{104}\,\mathrm{Hz}$ and activation energy $E_a\approx 16\,500\,\mathrm{K}$. This seems to indicate the cooperative character of the freezing process in $FeCr_2S_4$ being similar to that of spin glasses. Therefore we applied the Vogel–Fulcher law $\omega=\omega_0\times\exp[-E_a/k_B(T_i-T_0)]$ [see the solid line in the inset of Fig. 3(a)], and achieved a satisfactory fit to our data with values of $\omega_0\approx 6\times 10^{15}\,\mathrm{Hz}$, $E_a\approx 290\,\mathrm{K}$ and $T_0\approx 61.3\,\mathrm{K}$. The value of T_0 is close to $T_m=61.5\,\mathrm{K}$, the temperature at which a cusp is observed in the dc measurements of this sample. The significance of this coincidence is not clear, but it may reflect changes in the relaxation processes occurring at this temperature, connected with the critical slowing down at least of a part of the relaxation time distribution. Keeping this in mind, it is important to note that the frequency opening of the $\chi_0''(T)$ curve, which characterizes the difference in magnetic loss for various frequencies, as, e.g., given by $\Delta\chi_0''(T)=\chi_0''(0.19\,\mathrm{Hz})-\chi_0''(1000\,\mathrm{Hz})$, also exhibits a maximum at T_m [see the upper left inset of Fig. 3(b)]. Furthermore, χ_0' and χ_0'' decrease proportional to $\log\nu$ in the range of $\nu=10^{-2}-10^3\,\mathrm{Hz}$ at a temperature of 61.5 K, shown in the lower inset of Fig. 3(b). For χ_0' this type of dispersion is similar to the one found in spin glasses below the freezing temperature.[22] However, the negative slope in the $\chi_0''(\omega)$ dependence around the freezing temperature contradicts the assumption of spin glass.

Previous magnetization measurements of our samples[9,10] have shown that long-range order persists at all temperatures below T_c. Therefore, the contrast in the ac and dc susceptibility behavior may result from the domain wall pinning effect. This is demonstrated in Fig. 4(a), where the temperature dependences of the linear susceptibility χ_0' are shown for several values of ac driving field. H_{ac}. Only for high driving fields, which overcome the pinning forces, does the ac susceptibility value approach the dc value in Fig. 1. This is consistent with the temperature behavior of the imaginary component χ_0''. For higher driving fields blocking of the domain walls occurs at lower temperatures. With an increase in the driving field the maximum of χ_0'' is shifted toward lower temperatures [Fig. 4(b)]. For fields above 5 Oe, magnetic losses associated with χ_0'' reach saturation. At the same time the anomaly at around 60 K is present for all values of the driving field [see, for instance, the 0.2 Oe curve in Fig. 4(b) and the inset in Fig. 4(b), where derivatives of χ_0'' for higher driving fields are shown]. The development of the cusp in χ_0' with an increase in the driving field, on the one hand, and the field independent features in the $\chi_0'(T)$ and $\chi_0''(T)$ curves at around 60 K, on the other hand, may indicate two different contributions to the relaxation process. One of them may be attributed to domain wall pinning due to the temperature variation of the magnetocrystalline anisotropy, while the other, connected with the appearance of the coercivity accompanied by changes of the domain structure, is probably due to structural transformation which is suggested to occur at T_m ($\sim 60\,\mathrm{K}$). Our recent pressure investigation showed a direct relation between the magnetic anomaly at T_m and

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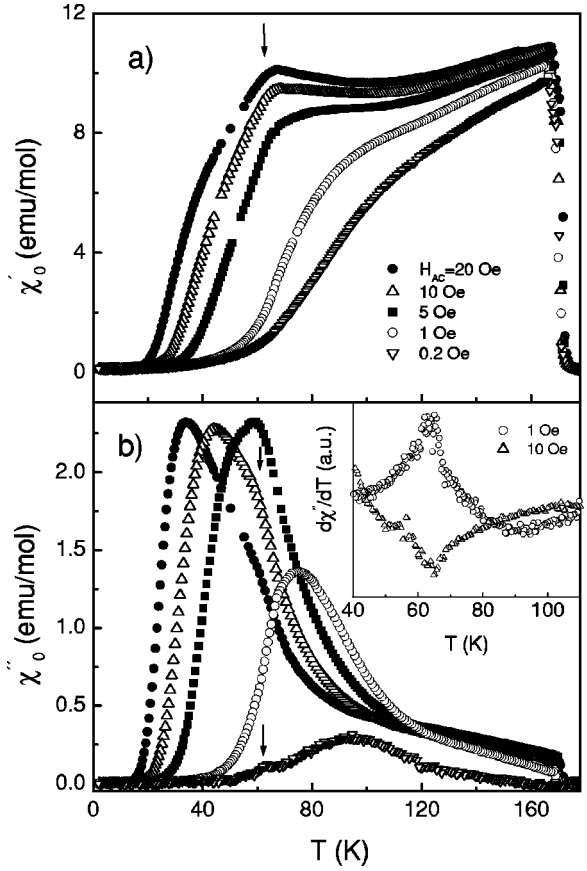

FIG. 4. Temperature dependences of the real part χ'_0 (a) and imaginary part χ''_0 (b) of the linear ac susceptibility at different driving ac fields at a frequency $\nu = 333$ Hz (sample 35x). Inset: Temperature dependences of derivatives of χ''_0 for driving fields of 1 and 10 Oe.

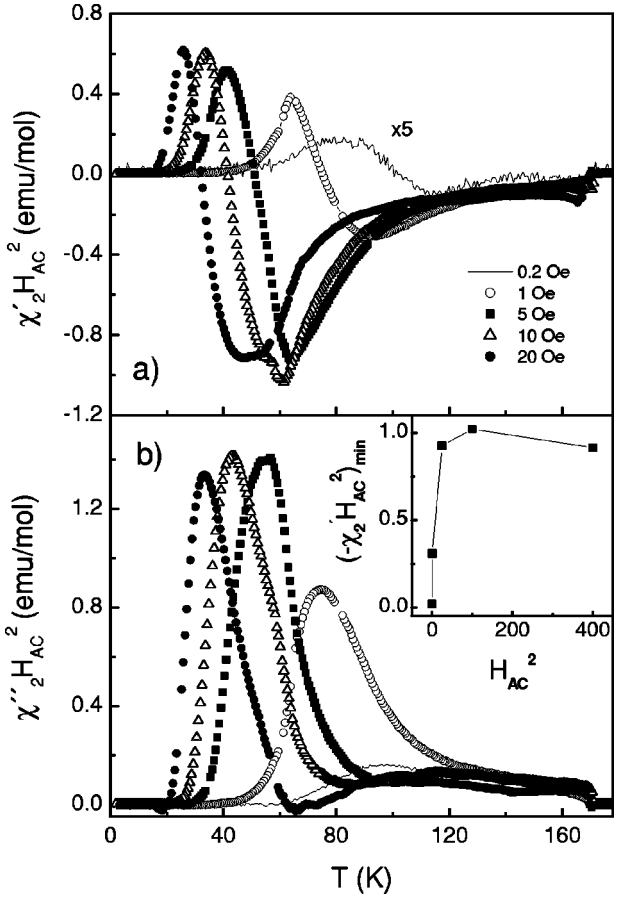

FIG. 5. Temperature dependences of the real part $\chi'_2 H_{ac}^2$ (a) and imaginary part $\chi''_2 H_{ac}^2$ of the nonlinear ac susceptibility of a $FeCr_2S_4$ single crystal (sample 35x) at different ac driving fields at frequency $\nu = 333$ Hz. Inset: Dependence of $\chi'_2 H_{ac}^2$ of the minimum on the magnitude of H_{ac}^2.

structural lattice distortions and it strongly supports such a transformation.[23]

In order to study spin glass-like anomalies in dc and ac susceptibilities in more detail, we performed measurements of the nonlinear susceptibility which is expected to diverge at the freezing temperature for a canonical spin glass.[24–26] The temperature dependence of the real part of the nonlinear susceptibility, χ'_2, is shown in Fig. 5(a) for different values of the ac driving field. For the lowest driving field of 0.2 Oe, the $\chi'_2(T)$ curve exhibits a broad negative minimum between 130 and 100 K which becomes more pronounced and is shifted toward low temperatures for higher driving fields. For lower temperatures a distinct maximum is observed that becomes sharper for higher driving fields. The behavior of this low temperature maximum corresponds to the loss features reflected by the imaginary part of the nonlinear susceptibility, χ''_2, and thus follows relaxation dynamics. Moreover, χ''_2 shows a temperature dependence similar to that of the imaginary part of the linear susceptibility χ''_0 [see Fig. 5(b)]. The nonlinear susceptibility χ''_2 is also strongly influenced by the driving field. Around 60 K a structure in the $\chi''_2(T)$ curves is observed. Although some small features at T_m can be also seen in $\chi'_2(T)$, the absence of a clear divergent-like anomaly in χ'_2 is in contrast with the canonical spin glass picture.[27] The expected behavior of χ'_2 in the case of a spin glass-like phase may, however, be masked by domain effects. In addition, for large values of H_{ac}, the dependence of the minimum value of $\chi'_2 H_{ac}^2 = f(H_{ac}^2)$ shows saturation (see the inset of Fig. 5), indicating that contributions of higher order terms to the nonlinear susceptibility cannot be neglected.[14] These factors prevent a comparison with existing theoretical models and a clear distinction between a re-entrant spin glass and a pure domain scenario is not possible.

The data obtained for different values of the ac driving field show that both linear and nonlinear susceptibilities are strongly affected by domain wall movement. Similar results were observed in re-entrant spin glass NiFeAu alloys,[28] UCu_2Ge_2 (Ref. 29) and UCoGa.[30] For the first two materials the ac susceptibility exhibits a pronounced field dependence. For the latter compound, which also shows complicated magnetic behavior in fields far below the Curie temperature, the ac response was found to be strongly affected by domain wall pinning effects due to the very high magnetocrystalline anisotropy. Furthermore, just below the transition temperature, the ac susceptibility of UCoGa exhibits a noticeable frequency dependence.[30] Some strongly anisotropic compounds, like, e.g., $TbFe_{11}Ti$ (Ref. 31) and Nd_2Co_{17},[32] also exhibit a cusp in the magnetization at temperatures far below T_C, similar to that found in $FeCr_2S_4$ crystals at T_m. The appearance of such an anomaly is a consequence of a spin–reorientation (SR) transition due to competition of the anisot-

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ropy of RE and TM ions in these compounds. Since magnetocrystalline anisotropy in $FeCr_2S_4$ is also of considerable magnitude ($K_1 \sim 3-6\times10^6$ erg/cm^3 at 4.2 K)[6,7] it is important to clarify its influence on the magnetization anomalies observed.

In hard magnetic compounds pinning of the domain walls in low fields causes a cusp in the susceptibility χ_{ZFC} at the transition temperature, whereas broad maxima in $\chi_{ZFC}(T)$ curves are observed for soft magnets.[33–37] The $FeCr_2S_4$ crystals show rather soft ferromagnetic behavior for applied fields between 2.5 and 100 Oe, i.e., the dc susceptibility exhibits a demagnetization limited plateau[38] at temperatures of $130\,\mathrm{K} < T < T_C$. In this temperatures range, the almost linear dependence of the magnetization on the applied field as well as the absence of hysteresis indicate that domain wall movement is not affected by the anisotropy field. For temperatures below 60 K the shape of the ZFC dc susceptibility curve depends significantly upon the applied field (see the curves for 100 and 2.5 Oe in Fig. 1). However, magnetization reversal in low fields[10] as well as spin glass-like magnetization irreversibilities do not depend on the relative orientation of the magnetic field to the crystallographic axes. Therefore, the role of the anisotropy field in blocking domain wall movement does not seem to be dominant. The nonmonotonic behavior of χ_0' also indicates that the change in pinning of the domain walls below 90 K is not due to a continuous increase of the anisotropy field with a decrease in temperature. Although an increase in the anisotropy field should reduce the domain wall width, the sudden onset of coercivity for temperatures below 60 K (Ref. 10) indicates the appearance of new pinning centers, which strongly influence domain wall movement.

In considering the possibility of a spin–flip transition it is necessary to mention that the strong magnetocrystalline anisotropy of $FeCr_2S_4$ originates from the tetrahedrally coordinated Fe^{2+} ions,[6,39] whereas the contribution of the octahedrally coordinated Cr^{3+} ions to the magnetic anisotropy is negligible.[40,41] Therefore the SR transition due to a competition mechanism should be of no importance. High field magnetization measurements of our crystals do not show a change of the easy axis of magnetization in the (110) plane. In this case the ⟨001⟩ axis is the easy axis for all temperatures (the results will be presented elsewhere). Therefore, another type of transformation, for example, one similar to that observed in highly anisotropic hexagonal R_2Co_{17} (R=Tb, Dy, Ho) compounds, might be considered. These compounds do not show a spin–reorientation transition, but exhibit a susceptibility anomaly due to the appearance of anisotropy in the basal plane.[42,43] It is important to note here that the pressure experiments[23] mentioned above also revealed a relation of the magnetic anomaly at around 60 K in $FeCr_2S_4$ with the appearance of a low symmetry anisotropy component in the (010) plane due to lattice distortions. Furthermore, a sign reversal of the spontaneous transverse magnetoresistance at T_m has been found for the magnetic field applied in a hard direction, which indicates substantial changes in the domain structure at this temperature.[23] We suppose that due to strong spin–lattice coupling lattice distortions are responsible for the low temperature changes in magnetic behavior. For example, the structural transformation suggested at T_m may change the defect structure, introducing new phase boundaries, e.g., twinning in the case of tetragonal distortions, that act as additional pinning centers for domain wall movement as well as result in rearrangement of the domain structure.

Finally, we want to mention that the suggestion of structurally induced magnetic changes in $FeCr_2S_4$ is in general agreement with the earlier investigations of the Mössbauer effect, which indicated the presence of low symmetry crystalline fields at low temperatures in this compound.[6] Unusual peculiarities of the Mössbauer spectra, like quadrupole splitting below the Curie temperature or sharp changes of the electric field gradient on the Fe site at 10 K, were explained by lattice distortions induced, for example, by dynamic or static Jahn–Teller effects.[44–47] Although earlier neutron diffraction and low temperature x-ray studies of polycrystalline $FeCr_2S_4$ did not find deviation from cubic symmetry,[48,49] it seems that the sensitivity of these measurements was not adequate to detect such small distortions. Nevertheless, the latter investigations of powdered single crystals did indeed reveal unusual broadening of the Bragg peaks at low temperatures, possibly due to inhomogeneous lattice distortions.[50] In spite of other explanations of the anomalies of the Mössbauer effect in $FeCr_2S_4$, like, for example, orbital ordering,[46,51] the structural changes, in our opinion, are most likely the driving force for the observed low temperature magnetic anomalies.

CONCLUSION

The magnetic behavior of ferrimagnetic $FeCr_2S_4$ single crystals was studied by linear and nonlinear ac susceptibility. Strong dependence of χ_0 and χ_2 on the value of the ac driving field indicates an important contribution by the domain wall pinning effect. A cusp in the real part of χ_0 at around $T_m \approx 60\,\mathrm{K}$ observed for high driving ac fields correlates with the onset of magnetic irreversibilities in the dc susceptibility. The frequency dependence of the linear susceptibility χ_0 as well as a broad negative peak of the real part of the nonlinear susceptibility χ_2' below the Curie temperature cannot be explained within a purely spin glass picture. The presence of distinct susceptibility anomalies at around T_m indicates that spin glass-like magnetic behavior below 60 K is connected with changes in the domain structure and the appearance of additional pinning centers, which we suggest result from a structural lattice transformation.

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