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Hysteresis and relaxation behavior in diluted $A_2Fe_{1-x}In_xCl_5 \cdot H_2O$ $(A=K,Rb)$

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Low-anisotropy antiferromagnetic compounds of the general formula $A_2Fe_{1-x}In_xCl_5 \cdot H_2O$ $(A=K,Rb)$ exhibit a net spontaneous magnetization below their transition temperature (T_N) when they are cooled in very small applied magnetic fields. This magnetization is observed when the easy axis of the system is aligned parallel to the field and is found to saturate at fields of a few Oe. Hysteresis loops M vs H have been obtained at several temperatures for single crystals of samples with different concentrations x. For low values of x the loops are not symmetric and show a series of well-defined jumps. This behavior suggests that a structure of domains is present, the jumps being associated with avalanche-type movements of the antiferromagnetic domain walls. The coercive fields are of the order of a few hundreds of Oe and for similar x are lower in the Rb-based samples than in the K ones. When the applied field is removed below T_N some of the samples of the dilute rubidium derivative show relaxation effects. The drop in M after the field is removed is two orders of magnitude larger than the drop expected from the susceptibility term $(\chi_{\parallel}H)$. The origin of the net magnetization in this and in other dilute antiferromagnets has not yet been established but net domain wall magnetization of antiferromagnetic domains is suggested as the most consistent mechanism. [S0163-1829(97)01029-1]

I. INTRODUCTION

The presence of a net spontaneous magnetization in site diluted low-anisotropy antiferromagnets, such as $A_2Fe_{1-x}In_xCl_5 \cdot H_2O$ $(A=K,Rb)$,[1–3] $Mn_{1-x}Zn_xF_2$,[4] and $(CH_3NH_3)Mn_{1-x}Cd_xCl_3 \cdot 2H_2O$,[5] has been a subject of research interest in recent years. The appearance of such net magnetization (M) is observed when the sample is cooled in the presence of a low magnetic field $(H<10$ Oe) applied along the easy axis of antiferromagnetic alignment. The measured magnetization M in an applied field parallel to the easy axis is given by $M=M_r+\chi_{\parallel}H$ ($\chi_{\parallel}$ being the antiferromagnet parallel susceptibility). In the diluted $A_2Fe_{1-x}In_xCl_5 \cdot H_2O$ $(A=K,Rb)$ systems the magnitude of M_r is large when compared to the contribution of the $\chi_{\parallel}H$ term to M at fields of the order of a few Oe. This occurs even for small values of x.

The observed M_r points always along the easy axis, increases with decreasing temperature T, and saturates in fields of only a few Oe with saturation value $M_{r,s}$. One of the suggestions for the origin of M_r lays in the formation of antiferromagnetic domains nucleated at T_N whose walls are pinned by the impurities. Then domain walls would contribute with a net magnetization due to the presence of uncompensated magnetic moments at their surface. The magnitude of the measured $(M_{r,s})$ varies greatly from one system to another. Compared to the saturation value M_s of the sublattice magnetization $M_{r,s}$ is of the order of 0.001% in $Mn_{1-x}Zn_xF_2$ with $x=0.25$,[4] 0.3% in $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$ with $x=0.14$,[1,2] and reaches more than 1% in some samples of $Rb_2Fe_{1-x}In_xCl_5 \cdot H_2O$ with $x=0.15$.[3] It has been found that the dependence of M_r with the reduced temperature $\tau=T/T_N$ is the same in $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$ and $Mn_{1-x}Zn_xF_2$ independently of the value of x. This implies a universal type of behavior for M_r that extends to temperatures well below T_N.[2–4,6]

Recently, a very small M_r of the order of 0.01% of M_s has also been observed in pure antiferromagnet $Rb_2FeCl_5 \cdot H_2O$.[7] It is remarkable that the temperature dependence of $M_r[M_r(T)]$ for this undiluted system also obeys the same mentioned universal behavior.[3,6] The temperature dependence of the sublattice magnetization of this antiferromagnet, determined from neutrons experiments,[8] follows a very different behavior than that found for $M_r(T)$. If the remanent magnetization is due to a volumetric effect then M_r should scale with M_s. This lack of scaling suggests that M_r is not due to a volume effect and it is in favor of the proposed mechanism that relates M_r with uncompensated moments at the surface of antiferromagnetic domains.

However, not all the samples of the diluted compound $Rb_2Fe_{1-x}In_xCl_5 \cdot H_2O$ show the same temperature dependence of M_r. Even between samples of nearly equal x, very different temperature dependences of M_r have been observed and differences in $M_{r,s}$ of more than an order of magnitude have been found. This problem will be addressed in this paper.

We also study here the magnetic behavior of M_r under the cycling of the applied field (hysteresis) since it provides information on the nature of the domain structure that seems to be established when the system is cooled through T_N in the presence of an axial field. The relaxation process that follows the removal (or the application) of the magnetic applied field can also furnish extra information on the striking behavior of the magnetization processes. A preliminary account on some topics of this work has been published elsewhere.[9]

II. EXPERIMENTAL DETAILS AND RESULTS

A. Experimental details

The compounds $A_2MCl_5 \cdot H_2O$, $A=$Rb of K and $M=$Fe or In, are isomorphous and crystallize in the orthorhombic space group $Pnma$.[10,11] Substitution of Fe by In is possible at any concentration. Single crystals of $A_2Fe_{1-x}In_xCl_5 \cdot H_2O$, were grown from appropriate solutions of Rb(K)Cl, $FeCl_3 \cdot 6H_2O$ and $InCl_3$. No difference was observed between large and small crystals grown from the same starting solution and taken out of the solution at the same time. The value of x in the crystals was determined by atomic absorption, optical absorption in the wavelength range between 900 and 550 nm, and/or from the susceptibility at temperatures well above T_N, e.g., above $5T_N$. We estimate an accuracy of 10% for the values of x. Structurally, $A_2FeCl_5 \cdot H_2O$ is formed by discrete $[FeCl_5(H_2O)]^{2-}$ octahedra that are hydrogen bonded together.[12] Magnetically, the two materials are easy-axis antiferromagnets with two collinear interpenetrating sublattices.[7] For the undiluted, $x=0$, compounds the ordering temperature is $T_N = 10{,}1$ K and the ratio between the anisotropy field and the exchange field is $\alpha = H_A/H_E = 3.4 \times 10^{-3}$ (Ref. 10) for the $Rb_2FeCl_5 \cdot H_2O$. The values of these parameters are, respectively, 14.06 K and 8.5×10^{-3} for $K_2FeCl_5 \cdot H_2O$.[13]

The magnetic measurements were carried out in a Quantum Design superconducting quantum interference device (SQUID) magnetometer and in an EGG vibrating sample magnetometer (VSM). Since the magnetic coercive field in the hysteresis curves is small, a good resolution of M at low fields was obtained using a small copper coil to generate the fields below 100 Oe. Higher fields were obtained by means of a superconducting magnet. All the measurements were made in single crystals with the easy antiferromagnetic axis oriented parallel to the direction of the applied field. The compensation of the local axial field (Earth's field and neighbors' equipment) was achieved by setting a very small current through the small copper coil until no net magnetization was obtained from the sample after repeated cycles through its transition temperature. Occasional checks using a fluxgate probe indicated that the field was compensated within ± 0.002 Oe at the sample site. Usually the local axial field was lower than 0.5 Oe.

In the experiments where the magnetic field was varied to obtain the hysteresis curves the sweeping rate was slow enough to ensure that the measured magnetization was not affected by the time response of the VSM that was set at 1 sec. In some relaxation experiments the sample was cooled through T_N in an applied axial field [field cooling (FC) process] provided either by the axial copper coil or by the superconducting magnet, depending on its strength. After a given temperature below T_N was well stabilized, the field was set to zero and M was measured as a function of time. In another set of experiments the time response of the magnetization to an applied field was measured after cooling the sample at $H=0$ [zero field cooling (ZFC)] to a given temperature below T_N.

B. Hysteresis behavior in $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$

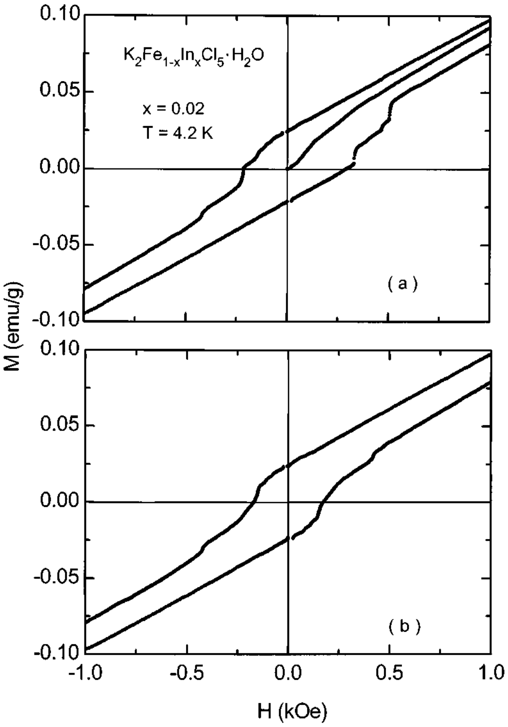

FIG. 1. Hysteresis curves for a sample of $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$ ($x=0.02$), at $T=4.2$ K. The extreme values of the applied field in the cycle are (a) ± 10 kOe and (b) ± 30 kOe.

Figure 1(a) shows a typical hysteresis loop measured at $T=4.2$ K, obtained for a sample of $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$ ($x=0.02$). When the sample is cooled in a zero axial field (the ZFC procedures include the axial field compensation), a zero magnetization is obtained. As H increases there is an increase in M that resembles that of systems with spontaneous magnetization starting from a domain state with $M=0$ at $H=0$. For fields above 5 kOe (not shown) the magnetization curve follows the straight line $M = M_{r,s}(T) + \chi_{\parallel} H$. When the field is decreased from a high positive value (we reversed it at 10 kOe), M decreases linearly with slope $\chi_{\parallel}$ down to $M_{r,s}(T)$ at $H=0$. After changing the sign of the field, the linear behavior persists for several tens of Oe and then a series of jumps occurs when increasing the negative field. For large negative values of H the linear behavior is again obtained, now with a constant negative $M_{r,s}(T)$. The subsequent decrease of H from large negative values leads again to a linear behavior of M. This linear behavior persists as the sign of the field is reversed again and is followed by a new series of jumps that seem not correlated to those observed in the negative field. The result is an asymmetric hysteresis curve. If the same cycle is repeated subsequently several times the jumps will occur always at the same values of H and with the same magnitude of M. By warming up the sample above T_N and cooling it down to the same T, the hysteresis cycle will be repeated exactly. This pattern is not altered by the magnitude of the field H in which the sample is FC through T_N, provided its direction is the same and that its values is of the order of a few Oe. We also observed that if the sign of the field in the FC process is reversed, then the new isothermal hysteresis curve relates to the previous one by an inversion operation through the origin ($M=H=0$).

The most striking effect in these curves in the existence of the series of well-defined magnetization jumps and the resultant asymmetric hysteresis curve. The actual fields at which the jumps occur depend on each particular sample. Even samples of the same batch generate jumps at different values of H and of different magnitudes. The steps are better resolved in samples with small x.

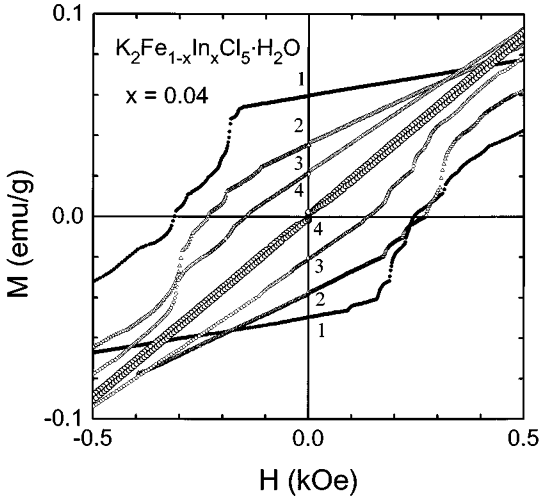

FIG. 2. Hysteresis loops in $K_2Fe_{1-x}In_xCl_5{\cdot}H_2O$, $x = 0.04$ at temperatures (1) 2.0 K, (2) 8.0 K, (3) 10.5 K, and (4) 16.0 K.

The isothermal hysteresis curves can be turned symmetric if at any temperature below T_N the turning field of the hysteresis cycle is extended to values that exceed the critical field of the antiferromagnetic phase. This is well illustrated in Fig. 1(b) where the field was extended to ±30 kOe in the hysteresis cycle for the same sample shown in Fig. 1(a) and at the same T. In undiluted $K_2FeCl_5{\cdot}H_2O$ the antiferromagnetic to spin-flop transition at 4.2 K occurs at 28.2 kOe.

The role of the temperature in the isothermal magnetization loops is well illustrated in Fig. 2 for a sample with $x = 0.04$ ($T_N = 13.5$ K). Each one of these loops was obtained after FC through T_N in a field of $H = 10$ Oe. The range of the cycle is in the interval between -10 to 10 kOe. Only the region between -0.5 and $+0.5$ kOe is shown. Note the asymmetry of the curves, particularly the extension of the linear region occurring after the field is reversed. The position of the jumps and the number of resolved steps change with temperature. Note that the slopes of the linear region decrease with decreasing temperature because the parallel susceptibility of the underlying antiferromagnetic structure decreases with T.

C. Hysteresis in $Rb_2Fe_{1-x}In_xCl_5{\cdot}H_2O$

Hysteresis curves were also obtained for the Rb compound. Figure 3 shows the loops obtained for a sample with $x = 0.15$ (sample RB232) at two different temperatures. The sample was FC in a field of 10 Oe. Since the concentration x is not too small the structure of jumps is not so clearly seen, only a single jump is noticeable just after the field changes sign. The transition to a spin-flop-like phase for this sample is at 13.6 kOe at 2 K. Then the entire cycle between -10 and 10 kOe occurs with the system inside the antiferromagnetic phase. Note that at 2 K the cycle is not closed at 1 kOe. This indicates that the reversibility region is attained at higher fields. Note also that the linear region just after the field change sign is considerably reduced in this case.

Figure 4 shows the hysteresis curve measured for another sample (RB1) also with $x = 0.15$ but from another batch. In this case the isothermal magnetization loop shows several differences with respect to the previously discussed sample (RB232): (i) the magnitude of the remanent magnetization is more than one order of magnitude larger than the previous one, (ii) the point where the magnetization starts to decrease is attained for small positive (negative) values of the applied field, that is, at $H = 0$ the magnetization is well below the value that will be obtained by extrapolating the liner region of the loops to $H = 0$, (iii) the width of the loop for comparable temperatures is smaller in this case, (iv) the reversibility region is attained already at 200 Oe for this sample. As we will show in the next section the decrease in M in the hysteresis loop on approaching $H = 0$ and the huge difference in the magnitude of M between these two samples with similar concentrations x is directly related to the relaxation behavior observed in the RB1 sample.

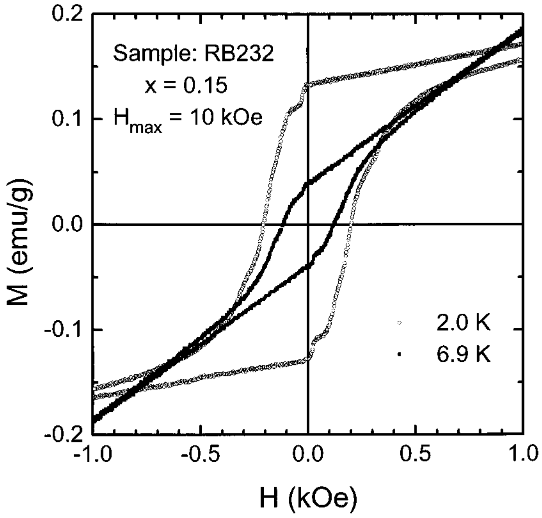

FIG. 3. Hysteresis curves for the sample RB232 with $x = 0.15$ at $T = 6.9$ and 2 K. Only the portion of the loop between ±1 kOe is shown.

D. Relaxation effect in $Rb_2Fe_{1-x}In_xCl_5{\cdot}H_2O$

Our relaxation measurements on the diluted systems $A_2Fe_{1-x}In_xCl_5{\cdot}H_2O$ (A = K,Rb) reveal that when the field

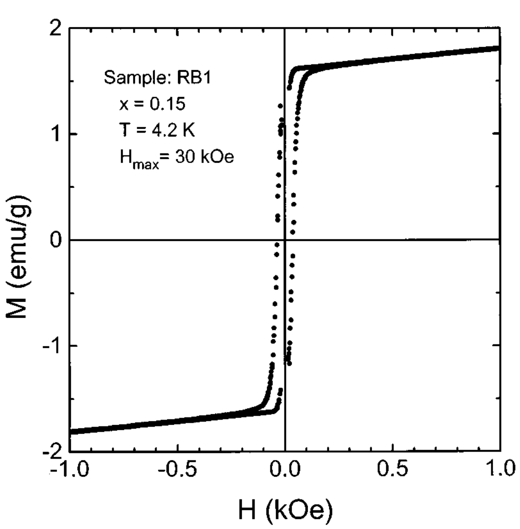

FIG. 4. Hysteresis curve for sample RB1 with $x = 0.15$ obtained at $T = 4.2$ K. Only the portion of the loop between ±1 kOe is shown.

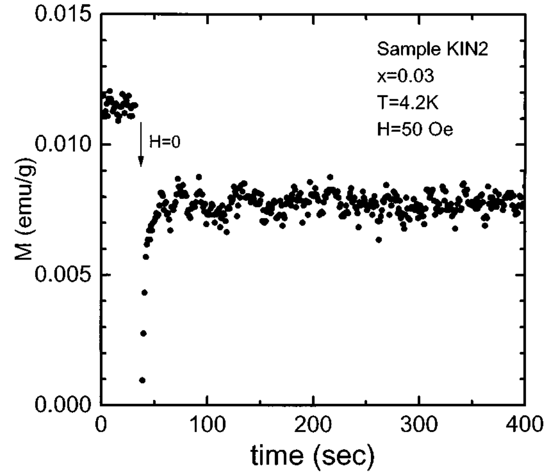

FIG. 5. Time dependence of the magnetization for a sample of $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$ ($x = 0.03$). The arrow indicates the time in which the field was removed. The sample was cooled in a field of 50 Oe.

in which the sample was cooled is removed below T_N, only some of the samples of the Rb system exhibit relaxation effects. Reducing the field to $H = 0$, should always lead to a decrease of M by an amount $\chi_\parallel H$, independent of the occurrence of a subsequent relaxation process.

Figure 5 is representative of the results obtained in the samples of $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$. The initial jump in $M(t)$ is due to the $\chi_\parallel H$ term. The data are for a sample with $x = 0.03$ but higher concentrations exhibit the same behavior. The absence of relaxation is characteristic of all the diluted samples of $K_2Fe_{1-x}In_xCl_5 \cdot H_2O$.

The results for the Rb system are more complex because only some of the sample show relaxation. We report here on results obtained for two of the many samples studied. These are the two Rb samples with $x = 0.15$ whose hysteresis curves have been presented in the previous section. Figure 6 shows at 4.2 K the time dependence of M of sample RB232 for a period of 120 min after the field was removed. The sample was cooled in a field of 20 Oe. The jump due to $\chi_\parallel H$ after the field was set to zero is barely visible. No relaxation is found in this case. Figure 7 shows the result for $M(t)$ obtained for the sample RB1. In this case the sample was cooled at a field of 75 Oe. After the field is removed at 4.2 K a huge decrease ΔM in $M(t)$ occurs followed by a subsequent relaxation process that even includes jumps in $M(t)$. The initial decrease in ΔM in M is at least two orders of magnitude bigger than the $\chi_\parallel H$ term at 75 Oe. Note also that $M(t = 0)$ is about one order of magnitude bigger than in the RB232 sample (see Fig. 6).

The temperature dependence of the relaxation in sample RB1 is illustrated in Fig. 8. An applied field of 10 Oe was used in the FC procedure. The increase in the values of $M(T)$ with the decreasing T reflects the temperature dependence of the remanent magnetization. Note that the initial drop ΔM in M, decreases with decreasing temperature.

The field cooling FC dependence of the relaxation process for the RB1 sample is shown in Fig. 9 at 4.2 K for fields range from 2 to 35 Oe. Note that the relaxed $M(t)$ tends to a saturation value for FC fields over 10 Oe. The initial magnetization also tends to slow down with increasing FC field. The $\chi_\parallel H$ contribution to M is typically of 1% of the total magnetization at $H = 20$ Oe (see Fig. 6 for comparison).

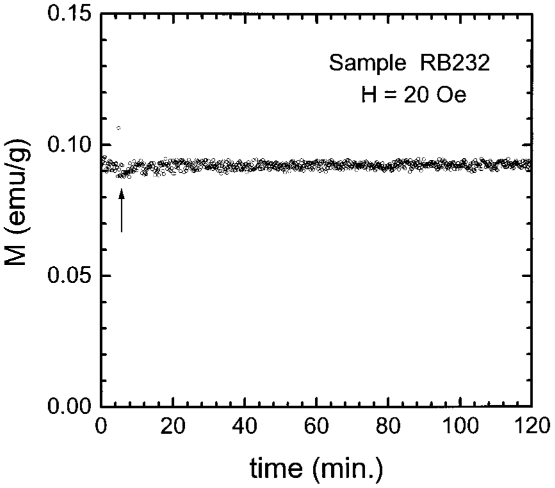

FIG. 6. Time dependence of the magnetization in sample RB232 at $T = 4.2$ K. The sample was cooled in a field of 20 Oe. The arrow indicates the instant when H was set to zero.

In another experiment with this same sample a small field was applied after the sample was cooled in a zero axial field to a temperature below T_N. We observed that this field induces a magnetization in the sample. The resulting increase in M is more than two orders of magnitude greater than the contribution due to the $\chi_\parallel H$ term. The subsequent removal of the field results in a reduction of M, however, a remanent magnetization slowly decaying in time persists. The resulting M vs time curve is shown in Fig. 10. The times when a field of 10 Oe is applied and removed are indicated. The magnetization reduction $\Delta M = 0.26$ emu/g, after the removal of H

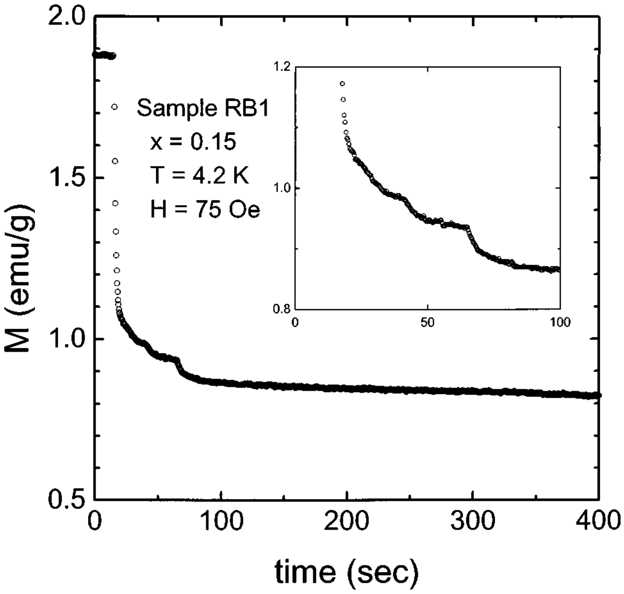

FIG. 7. Time dependence of the magnetization in sample RB1 at $T = 4.2$ K. The sample was FC in a field of 75 Oe. After the initial decrease in $M(t)$ occurs, some jumps are observed along the time (see inset).

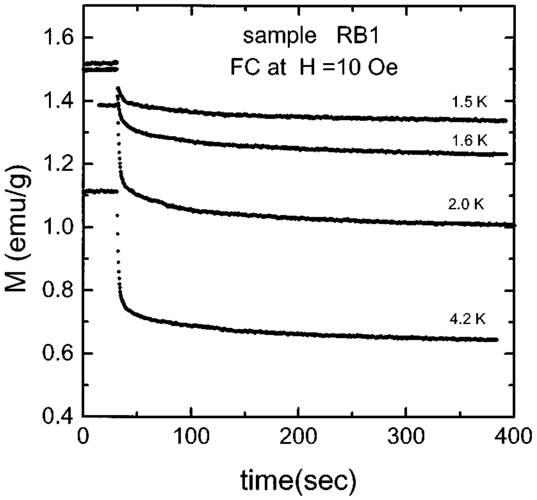

FIG. 8. Temperature dependence of the relaxation in the RB1 sample. The sample was always cooled in a field of 10 Oe. The initial drop of $M(t)$ in the curves occurs when H is set to zero.

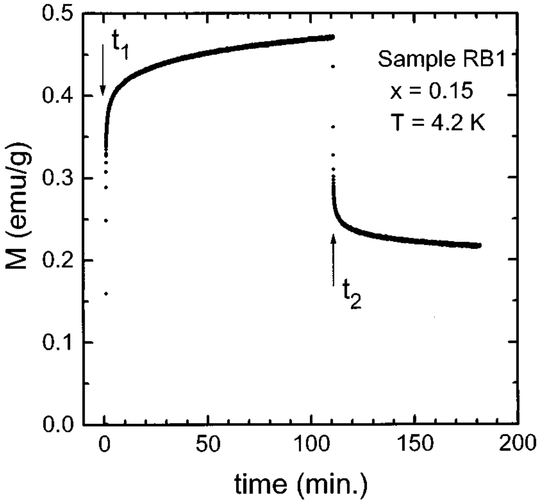

FIG. 10. Magnetization of sample RB1 at 4.2 K, after cooled in a zero applied field. At t_1 a field of 10 Oe is applied and removed at t_2.

is more than 100 times bigger than the corresponding contribution of the term $\chi_{\parallel}H$ (0.002 emu/g).

III. DISCUSSION

A. Hysteresis

When the sample is cooled in the absence of an axial field $(H=0)$ a state with no net magnetization $(M=0)$ is obtained. The subsequent application of an axial field leads to the increase of M. In Fig. 1(a) the results for a sample of $K_2Fe_{1-x}In_xCl_5\cdot H_2O$ is illustrated. The behavior is similar to the one that occurs in systems where a structure of domains with an overall zero magnetization exists at $H=0$. The application of a field reorients the domains and a net nonzero magnetization is obtained. In the antiferromagnet (AF) systems studied here the nonzero magnetization observed should also be attributed to the formations of domains in the crystal. Such domains can have uncompensated magnetic moments due to the dilution, their wall being stabilized by nonmagnetic impurities as well as for defects that may occur in the crystal. By increasing H the magnetization continues to increase until M_r reaches the saturation value. As already pointed out M_r in these samples is typically of the order of 0.1% of the magnetization of the antiferromagnetic sublattice. The subsequent linear increase of M at higher fields is due to the contribution of the antiferromagnetic $\chi_{\parallel}H$ term to M. This behavior should persist until the limit of thermodynamic stability of the AF phase is attained at the spin-flop field (H_{SF}). In the K samples with low x, the linear behavior corresponding to the $\chi_{\parallel}H$ term persists for few tens of Oe after the sign of H is reversed. That is, the magnetic moments in the domains are pinned and a minimum field opposed to their direction is needed to reverse them. This dipinning field depends on T (see Fig. 2) and varies from sample to sample. The effect of the demagnetizing field H_d should be taken into account in crystals where M_r is large. For a sample with M_r of the order of 0.1% of M_s as for the sample shown in Figs. 1 and 2, H_d is of the order of one Oe at helium temperatures. In some cases the beginning of the reversal process may occur just when H is reversed as shown in Fig. 3 for a sample of $Rb_2Fe_{1-x}In_xCl_5\cdot H_2O$ with $x=0.15$ (RB232). Here the estimated H_d is about 6 Oe. Curves similar to those shown in Fig. 3 were observed in samples of the K derivative with equivalent concentration.[9] As a general trend we noticed that the increase in x leads to a tendency to the reduction of the applied reverse field where the depinning starts. In samples where the magnitude of the magnetization at $H=0$ is much larger (as occurs for sample RB1 in which $M_{r,s}$ is about 4% of M_s) the demagnetizing field can reach several tens of Oe and this field can be sufficient to unchain the reversal process before the applied field $H=0$ is attained. For the sample RB1 whose hysteresis loop is shown in Fig. 4, a value of $H_d=60$ Oe has been estimated assuming an ellipsoidal shape of the sample.

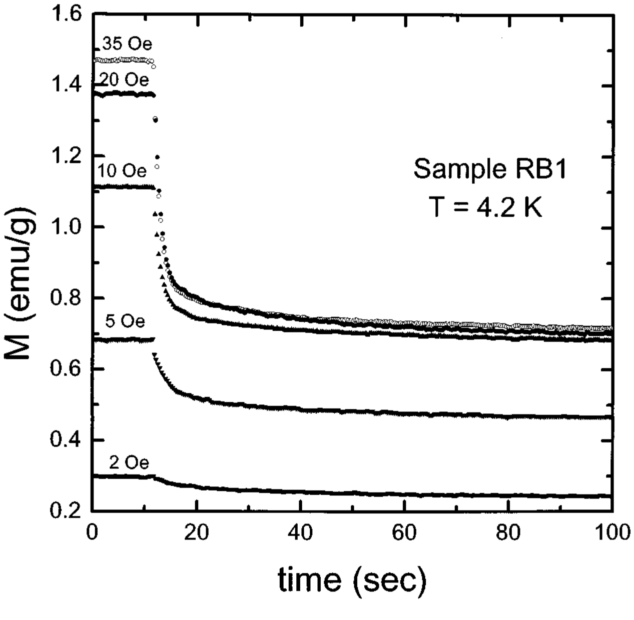

FIG. 9. Cooling field (FC) dependence of the relaxation in the RB1 sample at 4.2 K.

It seems that due to a lower anisotropy the Rb diluted system is softer than the K one at a comparable concentration. This can be inferred from the width of their hysteresis curves (roughly twice the coercive field) at comparable reduced temperatures. Although we systematically found that the width in the Rb diluted system is always smaller than in the K ones, Rb samples with nearly equal concentration can yield different values for the width. For instance, this can be seen by a quick look at Figs. 3 and 4 for the two Rb samples discussed here. Even taking into account the demagnetization fields in the RB1 sample, the width of the RB232 sample of comparable concentration is still higher than for RB1.

Since the magnetization of the single crystal saturates at a few hundreds of Oe it is interesting to compare the condition under which a monodomain will reverse under a depolarizing field. This field depends on the anisotropy K of the system and also on the magnitude of the magnetization as $H_c = 2K/M$.[14] The anisotropy of the $A_2\mathrm{FeCl}_5 \cdot \mathrm{H}_2\mathrm{O}$ ($A = \mathrm{K,Rb}$) systems is very small (the ratio α of the anisotropy to exchange fields is 8.5×10^{-3} for $A = \mathrm{K}$ and 3.4×10^{-3} for $A = \mathrm{Rb}$). This implies a smaller coerciveness for the Rb system considering that in general the size of M_r is higher in the Rb samples. This can also qualitatively explain why the RB1 sample that has an $M_{r,s}$ ten times larger than the RB232 sample has a much lower width in the hysteresis curve.

Another remarkable feature are the jumps observed in the hysteresis loops shown in Figs. 1 and 2. These jumps are seen more clearly in samples with small concentrations x. Our observations indicate that in a hysteresis loop measured at a given temperature T below T_N, the position and the magnitude of the jumps do not depend on the strength of the field at which the sample was FC to that temperature. Also the position and magnitude of the jumps do not change after successive cycling through T_N. The structure of domains seems to be established in the crystal when the sample passes through T_N. Even after a warming up of the sample above T_N and cooling it again to the same T, the position and magnitude of the jumps remains the same as long as the direction of the applied field is preserved. This behavior suggests that the domains are not formed in a random way but that their walls always go through the same set of nonmagnetic ions or defects. In the samples with lower values of x where several jumps are clearly resolved, the loops at a given T, are asymmetric with respect to the field. The observed inversion of the curve through the origin when the direction of the applied cooling field is reversed confirms the suggestion proposed for the domain formation. The jumps seem to originate in abrupt changes in the domain structure; e.g., abrupt domain-wall motion with the avalanche caused by groups of magnetic moments at the wall sites that align themselves in the local field. The position of the jumps changes with the temperature indicating a thermally assisted process. As the temperature is lowered the jumps also become more resolved because of the reduction of the thermal fluctuations. This is clear in Fig. 3.

We observed that the hysteresis loops become symmetric when the sample is driven through H_{SF} (both for the positive and negative H) [Fig. 1(b)]. It appears that when the sample enters into another ordered phase the ''memory'' concerning the pattern of domains, formed at T_N in the AF phase, is lost. The curve is symmetric because while lowering the field starting from the spin-flop phase (either for positive or negative H) the conditions under which the antiferromagnetic state is reached are symmetric with respect to the field.

The main features observed in the hysteresis curves for both diluted systems can be summarized as follows. (1) Steps are observed in the hysteresis curves only when the field is increasing either to positive or negative values of H. (2) When the field is decreasing the magnetization is linear in H except in some Rb-based sample where the nonlinearity close to $H = 0$ can be attributed to the size of H_d. (3) Steps are more resolved in samples with lower concentrations. (4) The steps occur in a given sample always at the same values of the field but the position of successive steps does not appear to be correlated. (5) Hysteresis curves are asymmetric in the sample with several resolved jumps when the loop is entirely cycled in the antiferromagnetic phase. If the sample is driven out of the AF phase by raising the upper value of the cycling field then the resulting loop is symmetric.

The observed jumps occur always at certain values of the field at a given temperature and this excludes the usual Barkhausen-like jumps in the hysteresis curves that are due to stochastic reversals of small groups of magnetic moments in the field, and that are not field reproducible. We have samples such as the one shown in Fig. 1, where almost one third of $M_{r,s}$ can be reversed in a single jump. This seems to indicate that domain wall motion occurs at the same time throughout the sample as an avalanche. A simple model, that may show jumps at fixed field values, is one in which magnetic states in multiple potential wells generated by the disorder are shifted by the action of the external field. This makes possible the occurrence of thermally assisted transition between these states favoring those where the moments are in the field direction. In addition the occurrence of tunneling processes between magnetic states cannot be disregarded. In avalanche-type processes it is suggested[15] that the primary event occurs through a microscopic tunneling that causes a local heating HdM; this can thermally nucleate wall motions that spread throughout the sample in a short time scale.

B. Relaxation

The models developed to account for the dynamic behavior of domains in diluted antiferromagnets in a field have been a focus of discussion for some time. The random-field (RF) induced domains are expected to grow with time with a logarithmic dependence,[16–18] and the excess magnetization predicted to occur in this case should also decay. The reported behavior is strongly dependent on the anisotropy of the system investigated. The width of the domain wall and its pinning either by the RF, when H is present, or by the nonmagnetic ions, is an important factor in determining its relaxation behavior. In low-anisotropy systems where the width of the wall is large the suppression of the RF should lead to an increase in the domains size and to a decay in M with time. In high-anisotropy systems where the domains walls are thin the nonmagnetic sites are the agents that stabilize the domain walls. In this case almost no relaxation is observed after the field is removed. These RF effects and the excess magnetization involved occur at fields well above those addressed in this work.[19–21] The relaxation effect observed here in some samples of the Rb system are certainly related to the disorder in the system and to its domain structure but not to RF effects.

In our discussion we must consider first that all the diluted samples of the K compound investigated do not show any sign of relaxation. These samples, although with different concentration x, have in common a reduced magnetization curve that thus implies a scaling behavior. It was also found that the Rb-based samples with dilution that do not show relaxation in M_r as well as the undiluted $Rb_2FeCl_5H_2O$ system also follow the same universal curve for the reduced magnetization.[7] In contrast, it was found that the samples of the Rb system that show relaxation effects have a reduced magnetization curve that departs considerably from the universal behavior followed by the K-based diluted samples and by $Rb_2FeCl_5H_2O$. As already pointed out in the undiluted Rb compound, M_r has a temperature dependence different than that of the sublattice magnetization. This seems to suggest that the uncompensated magnetic moments are at the surface of antiferromagnetic domains and do not follow the temperature dependence of the moments at the bulk of the domains. Uncompensated moments at the surface of domain walls would be ''loosely'' correlated to their neighbors in the bulk. At the transition temperature the domains form and their excess magnetization is oriented by the small local fields in the crystal.

Samples of the Rb system that exhibit strong relaxation effects always show a larger remanent magnetization than samples with similar concentrations that do not show signs of relaxation. This is exemplified by sample RB1 ($x=0.15$) whose relaxation behavior is shown in Figs. 7–10. Here the values attained by $M_r(T)$ are an order of magnitude larger than in sample RB232 also with $x=0.15$. In these strongly relaxing samples there is always a significant decrease ΔM in M just after H is removed. In fact, in sample RB1, ΔM is two orders of magnitude larger than the expected $\chi_{\parallel}H$ reduction in M. After this initial decrease in M, jumps in $M(t)$ have been observed in several samples. These are clearly visible in Fig. 7 for the RB1 sample. In some cases, jumps were observed more than an hour after the field was suppressed. Jumps were also occasionally observed when a small field was applied after a zero field cooling process of the sample. These jumps can in principle also be attributed to avalanche processes that occur along the time. Typical time constants for the subsequent decaying of M after the initial decrease ΔM are of the order of 10^2 sec. Due to the occurrence of jumps in time, it is not possible to assign a single time constant to the decaying process. The relaxation curves shown in Fig. 7 were taken in a bath of He at 4.2 K. This temperature is very stable and it is hard to conciliate these events (jumps) with thermally assisted processes. It could be possible, however, that local temperature fluctuations could induce transitions between magnetic states as was suggested at the end of the discussion in Sec. III A of the hysteresis behavior.

The relaxation curves obtained at different temperatures below T_N (Fig. 8) reveal that ΔM is temperature dependent. In Fig. 11 the value of the magnetization for sample RB1 before and 10 min after the cooling field of 10 Oe was removed is shown as a function of temperature. There is a region in temperature where the relaxation is more pronounced. The difference in M between the two curves in Fig. 11 is mainly due to the initial drop ΔM. When the sample was cooled in zero field down to a temperature below T_N no net moment is present. Figure 10 shows the results of the application of a small field of 10 Oe at 4.2 K after cooling the sample in zero field. This induces a magnetization that exceeds by two orders of magnitude the expected magnetization due to the $\chi_{\parallel}H$ antiferromagnetic term (0.002 emu/g). This magnetization is still one third of that obtained when FC the sample to that temperature in a field of 10 Oe. It might be argued that the response of the sample to the applied field could have its origin in a paramagnetic contribution of ions in the crystal. However, the subsequent suppression of the field does not lead to the former zero magnetization initial state. Instead, after an initial decrease a slowly decaying moment remains in the sample. Paramagnetic responses will tend to increase with T^{-1}. Such a tendency is not observed in Fig. 11 where the relaxed and nonrelaxed magnetizations are plotted as a function of temperature. The relaxed magnetization in the curves of Fig. 9 obtained at 4.2 K for various cooling fields show a tendency to saturation above 10 Oe. As in the previous discussion of Fig. 8 the initial decrease ΔM in $M(t)$ after the suppression of the field cannot be attributed to a paramagnetic contribution since $\Delta M/H$ is not constant.

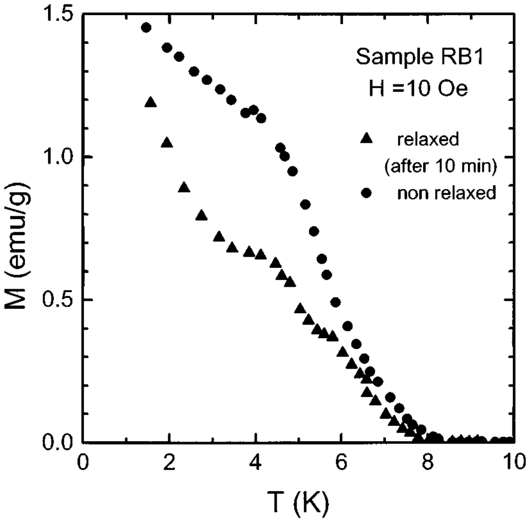

FIG. 11. Magnetization M_r as a function of temperature for sample RB1 obtained with the FC field applied (nonrelaxed) and without (relaxed). The sample was FC in a field of 10 Oe. The relaxed points were obtained 10 min after the field was removed. Between each pair of point the sample was warmed up above T_N and then FC to the next temperature.

From the above discussion it seems that in the Rb-based samples that exhibit these relaxation effects many uncompensated moments (typically of the order of 1% of the magnetic moments in the antiferromagnetic sublattice for the RB1 sample) are rather free to orient themselves in a low field. These moments, however, still maintain some correlation with the antiferromagnetic ''substrate.'' In contrast, in the samples of the K-based compounds, the magnetic moments are more fixed and the application of a small field of about 10 Oe does not result in an increase of M such as the one observed in the strongly relaxing Rb-based samples. To conciliate these two contrasting behaviors, we suppose that there are two main kinds of uncompensated magnetic moments. In one kind, the moments are moderately pinned by the nonmagnetic sites or defects and a field of the order of a hundred Oe is needed in order to revert its moments. These moments do not relax and typical hysteresis curves generated by them are shown in Figs. 1 and 2 for the K-based diluted system and in Fig. 3 for the RB232 sample. The resulting remanent magnetization of these moments follows the universal reduced magnetization curve discussed in Refs. 3 and 6. Although all samples are supposed to have moments of this kind, this is the only group present in the samples that follows a universal behavior. The second group is constituted by very loosely pinned magnetic moments that are easily reoriented by a very small field typically of the order of an Oe. These kind of moments would also contribute to M, however, its response to a small field would be more effective. Sample RB1 would be a representative example of a system possessing both kinds of pinned magnetic moments. These very loosely pinned moments would also be responsible for the departure of the total reduced magnetization of these samples from the universal curve. It should be remarked that this is a rather simplified picture and in practice a distribution of pinned moments can be present.

The unusual behavior observed in the relaxing Rb samples when compared with the K-based system may seem contradictory considering their close analogy. We found that diluted Rb-based systems appear to be very sensitive to the conditions under which the crystals were grown. Although crystals from the same batch give consistent results, crystals from different solutions with nearly equal concentrations can develop remanent moments that are an order of magnitude apart. This might likely be associated with the dynamics of the crystallization process of each particular batch. Such an unusual behavior may be due to the structural properties of these systems. As we mentioned before the $A_2\mathrm{FeCl}_5\mathrm{H}_2\mathrm{O}$ ($A = \mathrm{K,Rb}$) are isostructures, however, due to the large ionic radius of the Cs ion, $\mathrm{Cs}_2\mathrm{FeCl}_5 \cdot \mathrm{H}_2\mathrm{O}$ has a different crystal structure. It may be that the Rb system is close to the stability edge of its structure and consequently more subject to the formation of defects in the process of crystallization. This can create an additional contribution to the disorder of the lattice. The fact that the undiluted ($x = 0$) Rb system shows also a remanent moment[7] supports this argument since in that case the small remanent magnetization observed can in principle be attributed to uncompensated moments pinned by defects and dislocations in the crystal. The smaller anisotropy of the Rb system associated with its closeness to a ''structural edge'' may lead to the appearance of almost free magnetic moments that easily respond to a small applied field below T_N.

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