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Dipole−dipole interaction in superparamagnetic nanocrystalline Fe$_{63.5}$Cr$_{10}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$

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Cr-substituted Finemet-type nanocrystalline alloy (Fe$_{63.5}$Cr$_{10}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$) has been studied by differential scanning calorimetry, x-ray diffraction, Mössbauer spectroscopy, and magnetic measurements. The Curie temperature of the remaining amorphous phase decreases as the crystalline volume fraction increases, reaching values below room temperature. This feature makes the alloy adequate for studying the magnetic decoupling of the (Fe,Si) nanocrystals at moderated temperatures and, in particular, the superparamagnetic relaxation in broad temperature and crystalline fraction ranges. It was shown that the anomalous dependence of the coercive field on the annealing temperature can be satisfactorily explained assuming a dipolar-type interaction between the crystallites. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1383017]

I. INTRODUCTION

The Finemet alloy is composed of Fe, Si, and B, with small additions of Cu and Nb. It is obtained as a glassy material and after proper heat treatments a granular Fe-rich phase crystallizes with a mean grain size of the order of 10 nm, which remains embedded in the residual amorphous matrix. This alloy has attracted much scientific attention since its discovery by Yoshizawa *et al.*[1] not only due to its outstanding soft magnetic properties, which makes it adequate for many industrial applications, but also for its two-phase nature which allows us to perform quite a number of fundamental studies in magnetism regarding the coupling of nanocrystals.

The soft magnetic character of this family of nanocrystalline alloys has been explained on the basis of the random anisotropy model[2] extended to two-phase systems.[3] According to this model, when the crystalline volume fraction is high enough to allow the interaction between the particles, the effective anisotropy of the system is averaged out since the magnetization of the individual particles cannot follow their own magnetic easy axes. Studies of the evolution of the effective anisotropy distribution of these alloys with the crystalline volume fraction[4,5] provide results that can be explained by the previously mentioned model.

The fact that these alloys are composed of a nanocrystalline ferromagnetic phase embedded in a ferromagnetic or a paramagnetic amorphous matrix allows the study of the coupling of the nanoparticles via the matrix. This coupling which is responsible for the softness of the material can be controlled in different ways: either by changing the average distance between the nanograins with the help of proper heat treatments or by modifying the Curie temperature of the matrix introducing elements such as Cr or Mo in the precursor alloy.[6−11]

When the sample is measured at high enough temperatures—approaching the ferro-paramagnetic transition of the residual matrix—the coercivity increases signifying that the matrix is unable to transmit interaction between the particles.[12] At higher temperatures the particles are completely isolated and show superparamagnetism because of their small size.[13] The first experimental evidence of this behavior in a Finemet-type alloy was given for a sample with 8 at. % Cr content.[6] More recently, this behavior has been shown not only for Cr- and Mo-containing alloys,[14] but also for a composition close to the usual Finemet-type alloys.[15]

In this article differential scanning calorimetry (DSC), x-ray diffraction, bulk magnetic measurements, and Mössbauer spectroscopy results on Fe$_{63.5}$Cr$_{10}$S$_{13.5}$B$_9$Cu$_1$Nb$_3$ nanocrystalline alloys are presented. The amorphous alloy has a Curie temperature much lower than that of the usual Finemet due to the relatively high Cr content, and after devitrification the T_c of the remaining amorphous matrix is reduced with the increase of the crystalline volume fraction. This effect makes possible the study of superparamagnetic behavior at temperatures well below the crystallization temperature of the remaining amorphous phase.

II. EXPERIMENT

Fe$_{63.5}$Cr$_{10}$Sr$_{13.5}$B$_9$Cu$_1$Nb$_3$ amorphous alloy has been obtained as a ribbon (∼10 mm wide and ∼25 μm thick) by a melt spinning technique. Its amorphous character was checked by x-ray diffraction and electron microscopy (TEM)

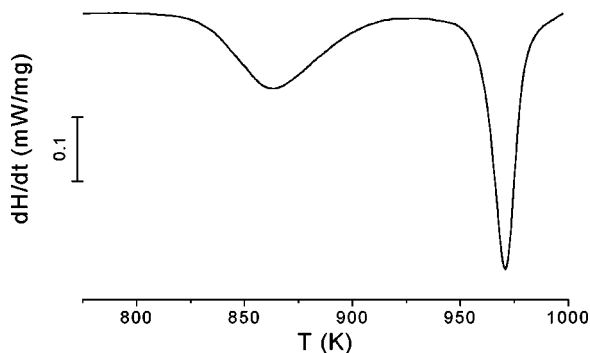
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FIG. 1. DSC plot of the as-cast alloy at 20 K/min heating rate.

TABLE I. Annealing-temperature (T_a) dependence of crystalline volume fraction (p_x) and average grain size (D_x) determined from x-ray diffraction; mean magnetic moment of the nanocrystals ($\langle\mu\rangle$) and their mean size (D) calculated from the Langevin fitting of the magnetization curves and ratio of the Fe atoms found in the nanocrystalline phase (p_{Fe}) by Mössbauer spectroscopy.

T_a (K)	p_x	D_x (nm)	$\langle\mu\rangle$ ($10^3 \mu_B$)	D (nm)	p_{Fe}
775	undetectable	undetectable	18 ± 7	5.2 ± 0.7	undetectable
800	0.066	9 ± 2	41 ± 6	6.8 ± 0.5	0.172
825	0.197	12 ± 2	112 ± 6	9.6 ± 0.5	0.350
850	0.385	13 ± 2	113 ± 6	9.6 ± 0.5	0.516

measurements. Samples were isochronally annealed for 1 h in a halogen-lamp furnace under argon atmosphere. DSC scans were performed up to 1000 K in a Perkin–Elmer DSC-7 in Ar purge gas. Mössbauer spectra were recorded at room temperature with a 50 mCi ^{57}Co (Rh) source.

Room temperature quasistatic hysteresis loops were measured with a homemade loop tracer described elsewhere.[16] A vibrating sample magnetometer (VSM) equipped with an electric furnace was used for the determination of the magnetization curves. The temperature dependence of the magnetic susceptibility was recorded by lock-in technique (with a frequency of 6 kHz and ac field of 20 mOe) in the temperature range between 77 and 300 K.

III. RESULTS AND DISCUSSION

A. Structural characterization

The devitrification of the studied alloy takes place in two main stages, as evidenced by the two well-resolved exothermal peaks shown in the DSC thermogram of Fig. 1. The first one with an onset temperature of 825 K corresponds to the nanocrystallization of the (Fe$_3$Si-type) grains. The second one—related to the crystallization of the residual amorphous phase—has an onset temperature of about 950 K. The partial substitution of Fe by Cr results in the stabilization of the amorphous matrix against nanocrystallization. The first DSC peak temperature is about 50 K higher for the Cr-containing alloy, in agreement with the trend shown by previously published data.[6–8,17]

X-ray study did not detect changes in the lattice parameter of the nanophase as the Cr content of the alloy is increased.[7,18,19] This observation suggests that Cr is excluded from the nanocrystals and it is in agreement with the absence of significant changes in the Curie temperature of the Fe$_3$Si-type phase with the Cr content of the precursor alloy. The Mössbauer spectra of different preannealed samples will be discussed below and they also indicate that the nanocrystals do not contain an appreciable amount of Cr.

As the onset temperature of crystallization is increased with respect to the Finemet alloy, the addition of Cr increases the temperature range in which the sample can be studied without increasing the crystalline volume fraction of preannealed samples. Therefore, this composition is ideal for the study of the magnetic behavior of the nanocrystals.

The nanocrystalline volume fraction is shown in Table I for isochronal 1 h annealing of the sample in the first crystallization stage. These values have been obtained by fitting the profile of the main x-ray diffraction peak by the superposition of two pseudo-Voight functions:[20] one representing the amorphous matrix and the other corresponding to the nanocrystalline phase. As an example, Fig. 2 shows the fitted x-ray peak profile of the sample annealed for 1 h at 825 K. The crystalline volume fraction can be obtained from the ratio of the integral intensities of the crystalline contribution and the total intensity.

The Mössbauer spectra measured at room temperature for some typical annealings are shown in Fig. 3. They consist of two main contributions: a narrow central doublet corresponding to the residual amorphous phase and several magnetic sextets originating from the precipitated crystalline phase. In the case of the sample annealed at 800 K the doublet of the residual amorphous phase shows some magnetic broadening which is an indication that its Curie temperature is near but somewhat above room temperature. For the samples annealed at higher temperatures the residual amorphous phase is definitely paramagnetic at room temperature as shown by the well-resolved quadrupole splitted spectra (the typical value of their quadrupole splitting is 0.484(4) mm/s). The isomer shift of this quadrupole doublet is +0.026(4) mm/s with respect to pure Fe.

The rest of the spectra corresponds to the precipitated crystalline phase and shows remarkable similarity. Their
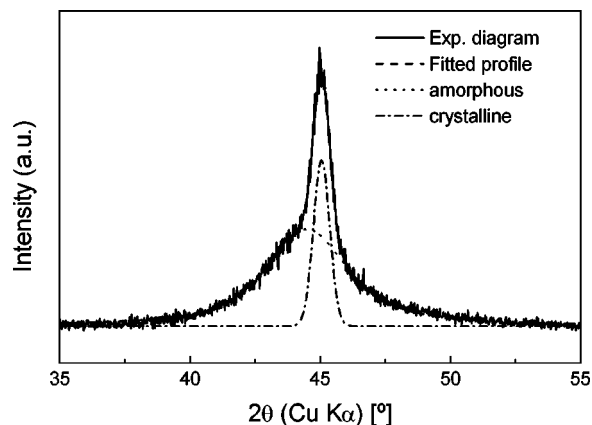

FIG. 2. Fitting of the diffraction profile corresponding to the sample annealed for 1 h at 825 K. The different contributions from the amorphous halo and the peak corresponding to the nanophase are shown.
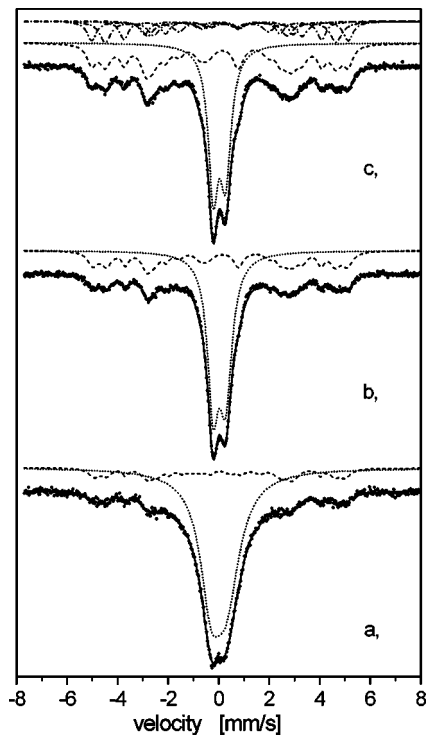
FIG. 3. Room-temperature Mössbauer spectra of nanocrystalline $Fe_{63.5}Cr_{10}Nb_3Sr_{13.5}B_9Cu_1$ annealed at 800 (a), 825 (b), and 850 K (c), respectively. The full lines are the fitted curves, the components corresponding to the residual amorphous phase are dotted lines and the components of the precipitated Fe_3Si-type nanocrystallites are marked with dashed lines, respectively. For the latter case the four subcomponents are also shown by dot-dashed lines.
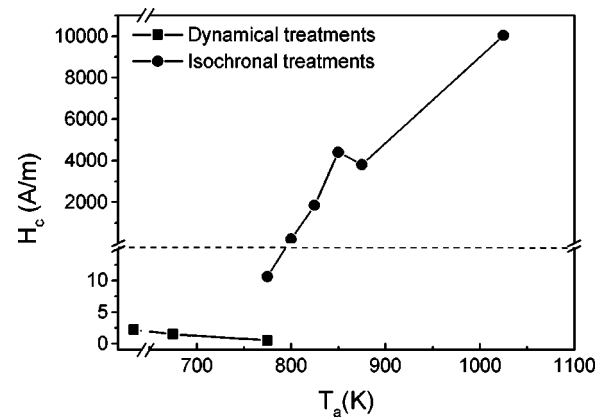

FIG. 4. Influence of annealing temperature on the room-temperature coercivity. (Dynamical treatment: continuous heating at 20 K/min. Isochronal treatment: annealing for 1 h).

relative contribution to the total spectra is increasing with increasing annealing temperature. The area under the respective curves corresponds to the percentage of Fe atoms in that phase and is given in Table I (p_{Fe}). Satisfactory description of the subspectra of the crystalline phase is obtained by four somewhat broadened six-line patterns as shown in Fig. 3(c). The hyperfine fields of the sextets for the sample annealed at 850 K are as follows: 31.5, 28.4, 24.3, and 19.0 T, respectively. These values correspond rather well to those measured by nuclear magnetic resonance spin-echo in Fe_3Si with excess Fe[21] however, their intensities are somewhat different. The values increase slightly with increasing annealing temperature, the increase is about 0.4 T between 800 and 850 K annealing temperatures. This hints at slightly increasing Curie temperatures of the crystalline Fe_3Si-type phase either due to the more ordered state or to the decreasing Si content of this alloy phase. Both possibilities are in line with the changing intensities of the six-line components corresponding to Fe atoms sitting in different environments. (The highest value of the hyperfine fields is that of Fe atoms with 8 Fe nearest and 6 Si next-nearest neighbors (B sites of the Fe_3Si structure), the lowest is that of Fe atoms on (A,C) sites with 4 Fe and 4 Si nearest and 6 Fe next-nearest neighbors, the rest is corresponding to the different environments created by the Fe excess in this structure[21] and is not the subject of the present discussion). No contribution was observed which can be assigned to dissolved Cr in this structure. Because of the complicated structure of the spectra (caused by the excess

Fe) only larger amount of dissolved Cr would result in a well-resolved component.

As seen from the Mössbauer measurements the great majority of Cr is enriched in the residual amorphous matrix. This is well in line with the general observation that early transition metals (Cr) favor to be coordinated by metalloid atoms (B, Si) with which they have a large chemical affinity (large negative heat of mixing). The DO_3 structure of the precipitated Fe_3Si-type phase evidenced clearly by Mössbauer spectroscopy is difficult to be detected by x-ray diffraction in the early stage of nanocrystallization.

B. Room temperature hysteresis loops

The room-temperature magnetic softness of this alloy is lost as nanocrystallization proceeds. According to the random anisotropy model,[3] which is used to explain the magnetic properties of Finemet-type alloys, the reduction of the average distance between the crystallites favors the coupling of the grains and the magnetic softening of the sample should return at high nanocrystalline fractions.

The dependence of the coercivity on annealing temperature is presented in Fig. 4. It is obvious that heat treatments that only produce stress relaxation of the samples reduce the coercivity. As soon as the first nanocrystals appear (annealing for 1 h at 775 K), an increase in coercivity is observed as in conventional Finemet-type alloys. Further annealing of the alloy at higher temperatures (in the temperature range of the first crystallization stage) does not reduce the value of the coercive field, in contrast to the Finemet alloys, although grain size remains around 10 nm and the crystalline volume fraction is increased (see Table I). Annealing the sample in the temperature range of the second crystallization stage (above 875 K) produces further hardening of the alloy, as more boride-type (magnetically hard intermetallic compound) phases appear.

C. Thermomagnetic properties

The decrease of the Curie temperature of the residual amorphous matrix below room temperature was previously inferred from low-field thermomagnetic experiments per-

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formed in a Perkin–Elmer thermobalance,[8,9] and confirmed by low temperature thermomagnetic measurements.[22] The Curie points obtained are in line with the Mössbauer results, indicating that the Curie temperature for the sample annealed at 800 K is near room temperature while those for the other two samples lie below it.

The features which might confirm that a sample shows superparamagnetic relaxation are (1) the absence of hysteresis and (2) the collapse of the magnetization curves as a function of H/T (applied field over temperature) for different temperatures.[13] In the case of Finemet-type alloys, the blocking temperature of the Fe_3Si-type nanoparticles is well below room temperature. However, the presence of the ferromagnetic residual amorphous phase prevents the observation of superparamagnetic behavior below its Curie temperature. The upper temperature limit of superparamagnetic relaxation is imposed by the T_c of the Fe_3Si-type phase and by the thermal stability of the specific sample, in order to avoid structural changes during the measurements.

The low Curie temperature of the amorphous matrix and its decrease with increasing crystalline volume fraction makes it possible to observe superparamagnetic relaxation in this alloy at relatively low temperatures. Also, the temperature range at which the different samples can be studied without changing the microstructure is much wider than for the alloys without Cr,[15] as the temperature at which structural changes take place increases with the volume fraction of the nanocrystallites.

In the case of conventional nanocrystalline Finemet-type alloys, the small cluster size at the beginning of the nanocrystallization results in a maximum of the room-temperature coercivity as a function of the crystalline volume fraction.[15] It is associated with a reduced coupling between the particles. In the case of the high Cr-content alloy studied in this article, room-temperature coercivity does not decrease with the increase of the crystalline fraction during the first crystallization stage. As the values of grain size and crystalline volume fraction remain below those observed for conventional Finemet alloys[18] reducing the coupling between the grains, superparamagnetic behavior is expected to be observed in these samples at higher temperatures.

Results in the literature[12] indicate that coercivity increases with temperature near the Curie temperature of the amorphous matrix. As the crystalline volume fraction increases, the maximum value of coercivity is also shifted to temperatures higher than T_c (am.). The temperature dependence of the coercivity of the samples annealed at different temperatures has been measured in the VSM and the results are similar for all of them. As an example, Fig. 5 shows the data corresponding to the sample annealed at 825 K. The value of coercivity at temperatures near 77 K has also been estimated using the loop tracer to be of the order of a few A/m. The joint results of the two measurements suggest that coercivity should have a maximum below room temperature for all the samples, confirming that the Curie temperature of the residual amorphous phase is near or below room temperature. This finding is in agreement with Mössbauer spectroscopy and ac-susceptibility measurements. The temperature at which coercivity presents a maximum seems to
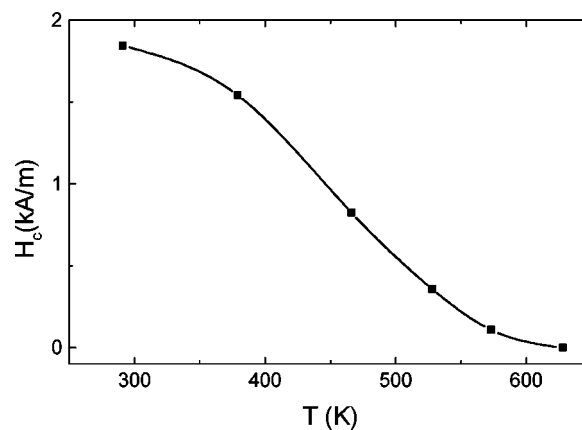

FIG. 5. Temperature dependence of coercivity for the sample annealed at 825 K for 1 h.

decrease with annealing temperature for this composition.

The temperature range where superparamagnetic relaxation can be detected shifts to slightly higher temperatures as well as the magnetization contribution originating from the superparamagnetic particles is enhanced with the increase of the crystalline volume fraction. In Fig. 6 the collapse of the reduced magnetization curves as a function of H/T is presented for the sample annealed for 1h at 825 K (H and T range from 0 to 6 kOe and from 630 to 710 K, respectively). It has to be noted that the absence of hysteresis in these curves is an experimental fact related to the high measurement temperature. The fitting of these curves to the Langevin function gives the magnetic moment of the particles and the estimate of their mean size. The results are given in Table I, together with the mean grain sizes calculated from x-ray diffraction. The values of the magnetic mean grain size are in good agreement with TEM observations[18] and x-ray results (Table I). Its dependence on the annealing temperature is similar to that observed in Finemet-type alloys,[19] with an increase of the grain size at the first stages of nanocrystallization that reaches a stable value at higher annealing temperatures. Although there is a slight systematic difference between the particle sizes obtained from the different tech-
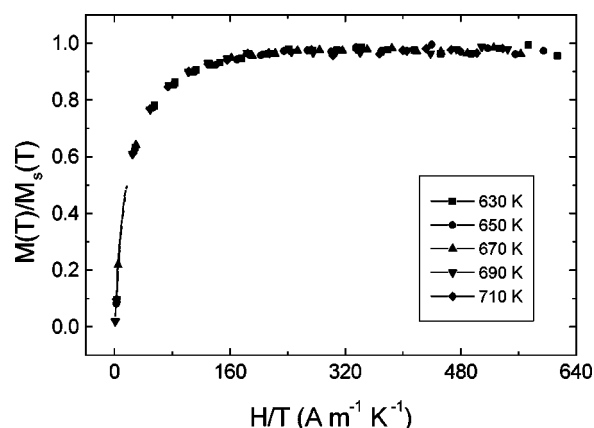

FIG. 6. Reduced magnetization curves of the sample annealed at 825 K for 1 h, showing a typical superparamagnetic behavior, without any trace of hysteresis at these relatively high temperatures.
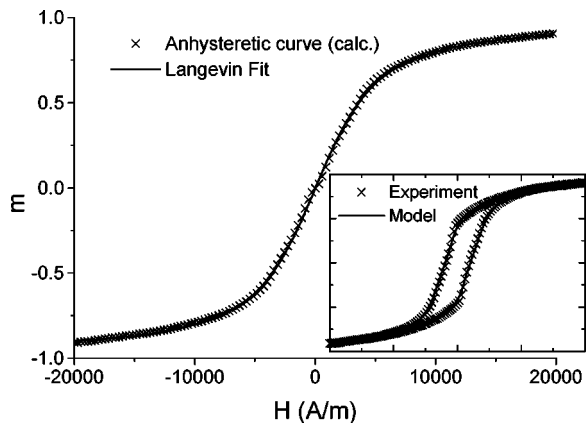
FIG. 7. Anhysteretic magnetization curve (crosses), obtained from the averaging of the branches of the hysteresis loop of the sample annealed at 825 K, together with its Langevin fit (continuous line). The magnetization normalized to its saturation value is denoted by *m*. Inset: Original hysteresis loop measured at room temperature (crosses) and its modellization (continuous line). The axis ranges of the inset are the same as those of the main figure.
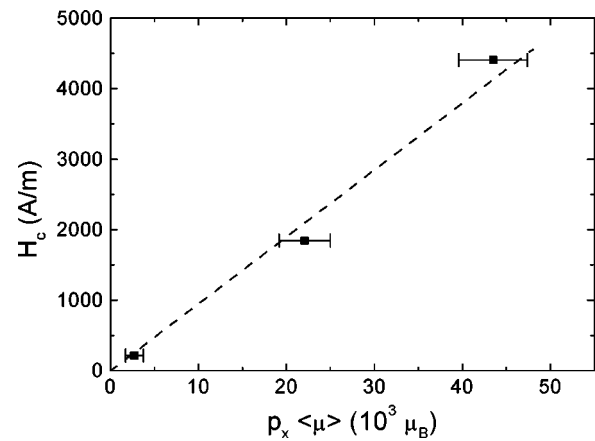

FIG. 8. Measured coercive field vs the product of the crystalline volume fraction and the mean magnetic moment of the grains for the samples annealed at 800, 825, and 850 K for 1 h. The dashed line is the best-fit line crossing the origo.

niques, however, it does not exceed the magnitude of the error bar.

A model has been developed recently for the description of hysteresis based on dipolar interactions in granular magnetic systems.[23] When the ferromagnetic particles embedded in a paramagnetic matrix interact via dipolar mechanism, the reduced half-sum of the two branches of the hysteresis loop is nearly coincident with the Langevin function of noninteracting moments. For samples with T_c (am.) below room temperature, this makes possible to check whether the interaction between the Fe_3Si-type particles at room temperature is of the dipolar type. In this case the reduced half-sum of the branches of their hysteresis loops is expected to be described by a Langevin function.

The reduced half-sum of the branches of the hysteresis loop (anhysteretic curve) together with its Langevin fit is shown in Fig. 7 for the sample annealed at 825 K for 1 h. The inset to this figure shows the original hysteresis loop measured at room temperature and the shape of the hysteresis loop predicted by the model of Allia *et al.*,[23] with the same axis ranges as the main figure. The agreement between the calculated anhysteretic curve and its Langevin fit is rather good, as well as the correlation between model and experimental hysteresis loops. The fitted mean magnetic moment of the particles at room temperature is of $175\,000 \pm 3000$ μ_B, obtaining a reasonable agreement with the values obtained from the Langevin fitting of the superparamagnetic magnetization curves (Table I). This value is certainly higher than that obtained for the experimental anhysteretic curves measured at around 675 K for the same annealing temperature (see Fig. 6). However, two effects should be taken into account. The first one is the temperature dependence of the magnetization of the particles and the second is that at temperatures near T_c (am.), a part of the matrix surrounding the particles can be magnetically coupled to them, giving an apparent larger size.

For samples annealed at lower temperatures the hysteresis loops are too abrupt to be fitted to Langevin functions

using reasonable particle sizes, which clearly indicates that the interactions are too strong at temperatures near T_c (am.). For annealing temperatures higher than 825 K the calculated room-temperature anhysteretic curves do not follow the Langevin form. Although in these cases the Curie temperature of the residual amorphous matrix decreases, the average distance between the particles also decreases, leading to stronger interactions between them.

The successful application of the model of Allia *et al.*[23] for the sample annealed at 825 K for 1 h suggests that the interactions between the nanocrystalline particles are of a dipolar nature. In order to find a correlation between the coercive field and the dipolar field, the coercive field measured at room temperature is plotted in Fig. 8 as a function of the product $\langle\mu\rangle p_x$. Here $\langle\mu\rangle$ is the mean magnetic moment of the grains obtained from Langevin fits of the magnetization curves in the high-temperature limit and p_x is the volume fraction of the nanocrystalline phase deduced from x-ray data (Table I). This product should be proportional to the dipolar field and contains only directly measured quantities. The rather good proportionality found between the coercive field and $\langle\mu\rangle p_x$ indicates that H_c is mainly determined by dipolar interactions acting between the grains. Using the volume-fraction data deduced from the ratio of the Fe atoms found in the Fe_3Si-type phase by Mössbauer spectroscopy (p_{Fe} in Table I) instead of p_x, a similar trend is obtained.

The dipolar field can be roughly estimated from the measured data. Using the form of $H_d \sim \langle\mu\rangle/R^3$ for the dipolar field H_d, where R is the average distance between the nanocrystalline particles, values of $H_d \sim 80.2$ kA/m (1.01 kOe), 231 kA/m (2.90 kOe), and 455 kA/m (5.72 kOe) are obtained for the samples annealed at 800, 825, and 850 K, respectively. Here R is calculated from the average size (D) of the particles (assumed to be cubic for simplicity) and p_x according to $R = D/p_x^{1/3}$. The values of H_d obtained this way are only estimates of the order of magnitude for the measured coercive fields since H_c is known to be determined by sev-

eral structure-sensitive factors besides the dipolar field. Therefore, the difference between the measured coercive fields and calculated dipolar fields might be associated with the microstructure of the alloys. For achieving a better agreement, the nontrivial summing of the dipolar fields should also be taken into account.

IV. CONCLUSIONS

A Finemet-type alloy has been studied in which 10 at. % Fe is substituted by Cr. This Cr addition enhances the thermal stability of the amorphous phase against nanocrystallization and results in a slightly smaller mean grain size of the Fe_3Si-type nanophase. Cr is excluded from the nanocrystals during the first crystallization stage, being mainly present in the amorphous matrix.

This enrichment in Cr of the residual amorphous matrix progressively decreases its Curie temperature, reaching values below room temperature. In contrast to the Cr-free alloys where the Curie temperature of the residual amorphous phase is above room temperature (RT), the present composition does not show soft magnetic behavior (at RT) at any transformed fraction values.

The magnetization of different nanocrystalline samples shows superparamagnetic relaxation in broad crystalline volume fraction and temperature ranges, assisted by the reduced value of T_c (am.) together with the enhanced stability of the samples against nanocrystallization. The mean grain sizes estimated from the Langevin fittings of the magnetization curves are in good agreement with x-ray data.

It was shown that the anomalous dependence of the coercive field on the annealing temperature can be satisfactorily explained by dipolar-type interaction between the crystallites.

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