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1993 J. Phys.: Condens. Matter 5 L437

(http://iopscience.iop.org/0953-8984/5/36/002)

J. Phys.: Condens. Matter 5 (1993) L437–L442. Printed in the UK

LETTER TO THE EDITOR

Low-temperature magnetization and spin-wave excitation in nanocrystalline ferromagnets

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Received 6 July 1993

Abstract. Magnetization measurements in the temperature range 1.5–300 K have been made on two series of samples of nanocrystalline ferromagnets: $Fe_{60}Co_{30}Zr_{10}$ and $Fe_{73.5}CuNb_3Si_{13.5}B_9$. It was found that at low temperatures magnetization decreases in accordance with the Heisenberg-model prediction for both series of samples. The coefficients B and C were obtained by fitting $M_s(T)$ to two terms, $T^{3/2}$ and $T^{5/2}$, from the Bloch law. It is shown that for both series of samples the B and C coefficients are larger in the amorphous state than in the nanocrystalline state; larger values of spin-wave stiffness constants D were obtained in nanocrystalline FeCoZr samples with grain size $d < 40$ nm than in the amorphous state; with increasing grain size a much larger value for D was observed for nanocrystalline FeCoZr with a grain size of 140 nm. The results present a reasonable agreement with predictions for nanocrystalline materials with two or three different magnetic phases.

The interest in magnetic nanocrystalline materials has been growing recently. In particular, many studies [1–5] have been made on the typical $Fe_{73.5}CuNb_3Si_{13.5}B_9$ nanocrystalline ferromagnets which show excellent soft magnetic properties. In our previous investigations [6] a series of $Fe_{60}Co_{30}Zr_{10}$ nanocrystalline ferromagnets with grain sizes of 15–300 nm were produced. The grain-size dependence of magnetic properties and magnetization processes have been analysed in the light of the random-anisotropy model and structural potential theory, respectively. The TEM micrographs show that the series of nanocrystalline FeCoZr [6] and FeCuNbSiB [2] are composed of very fine crystalline grains embedded into an amorphous matrix. In such heterogeneous alloys the behaviours of low-temperature magnetization and spin-wave excitation were expected to be different from those in amorphous and crystalline systems. In this paper we present data on the coefficients B and C, as well as spin-wave stiffness constant D for both series of samples in the amorphous and nanocrystalline states.

The experimental procedure was as follows. Amorphous $Fe_{60}Co_{30}Zr_{10}$ ribbons were produced by rapid quenching using the single-roller technique in an Ar atmosphere. The samples were then crystallized by annealing in vacuum for 10 min above their crystallization temperature. A series of samples with increasing grain size of 15–300 nm was obtained by varying the annealing temperature from 800 K up to 1300 K. The amorphous $Fe_{73.5}CuNb_3Si_{13.5}B_9$ samples were produced in the Institute of Physics of the Slovak

0953-8984/93/360437+06$07.50

Academy of Sciences at Bratislava. They were subsequently crystallized at 825 K for times ranging from 30 to 300 min in order to vary the dimensions of the nanocrystals.

The observations of microstructure in samples of FeCoZr alloys were performed with the TEM Siemens Elmiskop 102 in the Max-Planck-Institut für Metallforschung in Stuttgart. The low-temperature magnetization was measured by an extracting sample magnetometer with a field of up to 1.6 T in the temperature range 1.5–300 K.

Figure 1 shows TEM micrographs and diffraction patterns of the crystallized FeCoZr alloys for annealing temperatures of 900 K and 1200 K. For amorphous FeCoZr the micrographs exhibit only homogeneous contrast; the electron diffraction pattern shows a typical diffuse ring characteristic of the amorphous state. After annealing at a temperature above the crystallization temperature ultrafine grain structure appears homogeneously in the amorphous matrix, as shown in figure 1(*a*). The weak diffuse diffraction rings in the diffraction patterns of figure 1(*b*) indicate the presence of residual amorphous phase. The results of x-ray diffraction and the temperature dependence of saturation magnetization for amorphous FeCoZr after heating up to 1100 K [7] reveal that the grains are composed of two ferromagnetic phases, α-Fe(Co) and the Laves phase of $(Fe,Co)_2Zr$.

The average grain size was measured by a Videoplan. It was found that below an annealing temperature $T_a = 1000$ K the grain size increases slowly with increasing annealing temperature, and its distribution is homogeneous. At annealing temperatures above 1000 K the grain size increases more significantly and the distribution of the grains becomes more inhomogeneous (see figure 1(*c*)).

TEM investigation [1] indicated that the $Fe_{73.5}CuNb_3Si_{13.5}B_9$ alloy annealed at 823 K for 1 h is composed of ultrafine grains of BCC Fe solid solution, with diameters of about 10 nm.

Neutron-diffraction and magnetization measurements [8] have demonstrated that (1) the amorphous ferromagnets do indeed exhibit well defined spin-wave excitations at long wavelengths, which satisfy a normal ferromagnetic dispersion relation

$$hw(q) = \Delta_g + Dq^2 + Eq^4 + \dots \tag{1}$$

as expected for crystalline ferromagnets, where D is the spin-wave stiffness constant and (2) at low temperature, the magnetization decrease in amorphous ferromagnets is adequately described by the Heisenberg-model prediction, as in crystalline ferromagnets,

$$\Delta M(T)/M(0) = [M(0) - M(T)]/M(0) = BT^{3/2} + CT^{5/2} + \dots \tag{2}$$

where B and C are coefficients. B, C and D are related through the expressions

$$B = \zeta(\tfrac{3}{2})\left[g\mu_B/M(0)\right]\left(k_B/4\pi D\right)^{3/2} \tag{3}$$

$$C = \zeta(\tfrac{5}{2})\left[g\mu_B/M(0)\right]\left(k_B/4\pi D\right)^{5/2}\tfrac{3}{4}\pi\langle r^2\rangle \tag{4}$$

where $\zeta(\frac{3}{2}) = 2.612$, and $\zeta(\frac{5}{2}) = 1.341$ are the Riemann functions and $\langle r^2\rangle$ is the average mean-square (MS) range of the exchange interaction.

For the nanocrystalline alloys investigated, which are ferromagnetic multiphase materials composed of a ferromagnetic amorphous interface and different ferromagnetic nanocrystalline grains, the temperature dependence of the saturation magnetization could be decomposed neglecting any interaction among the phases, as Herzer did [2]

$$M(T) = v_1M_1(T) + v_2M_2(T) + v_3M_3(T) \tag{5}$$

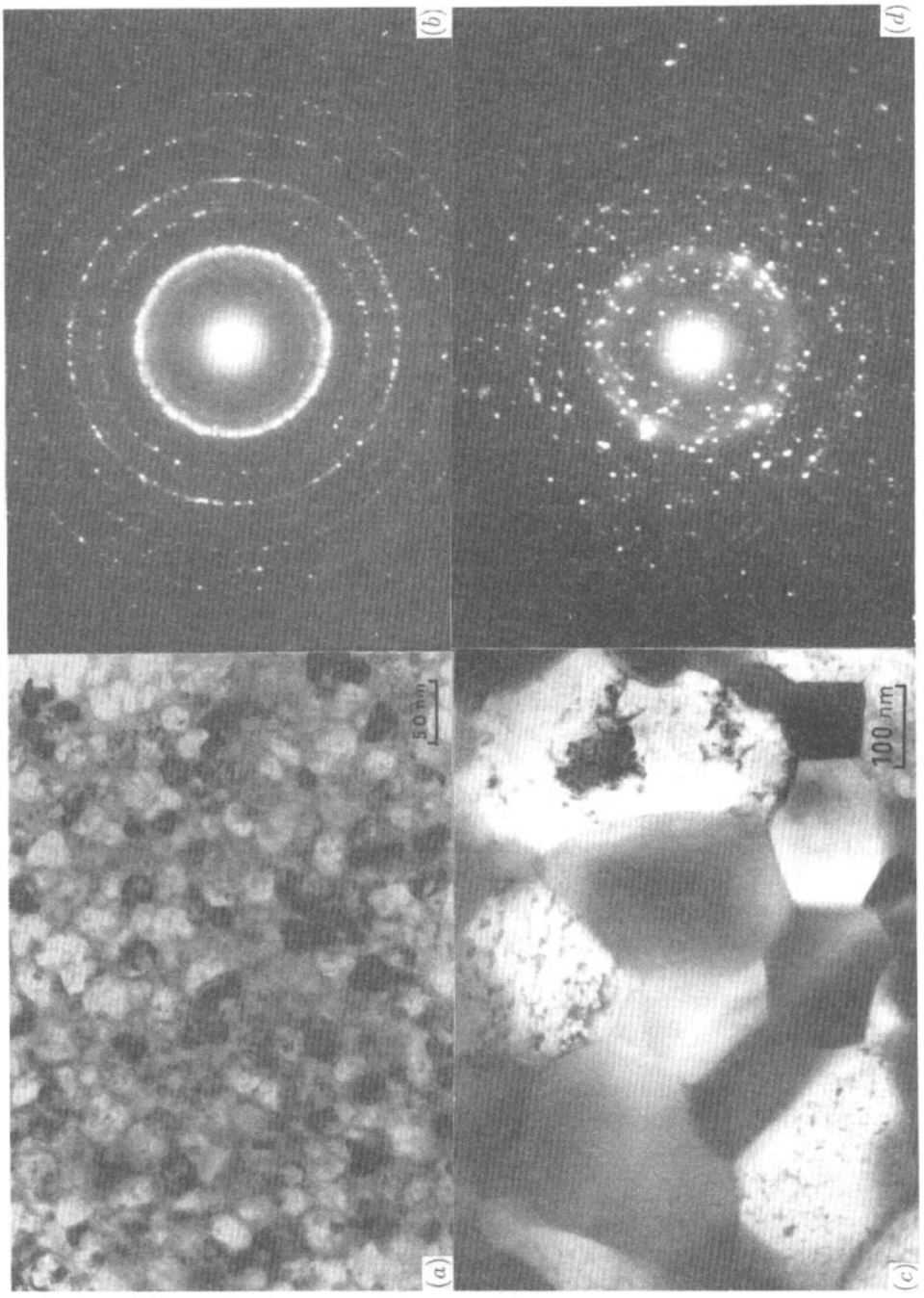

Figure 1. TEM micrographs and diffraction patterns of $Fe_{60}Co_{30}Zr_{10}$ alloys: (*a*, *b*) annealed at 900 K; (*c*, *d*) annealed at 1200 K.

Table 1. Spin-wave parameters for amorphous and nanocrystalline alloys: A, amorphous; F, nanocrystalline alloys with grain size d prepared by annealing at temperature T_a.

Samples	T_a (K)	d (nm)	B' (10^{-5} K$^{-3/2}$)	C' (10^{-8} K$^{-5/2}$)	D (meV Å^2)
A-FeCoZr	—	—	0.81	1.61	195
F_1-FeCoZr	850	15	0.69	0.31	215
F_2-FeCoZr	900	20	0.72	0.56	209
F_3-FeCoZr	950	25	0.68	0.24	216
F_4-FeCoZr	1200	140	0.39	0.56	313
A-FeCuNbSiB	—	—	1.69	3.09	126
F_1-FeCuNbSiB	823, 0.5 h	10 [1]	1.28	1.62	159
F_2-FeCuNbSiB	823, 2 h	—	1.29	1.05	158
F_3-FeCuNbSiB	823, 5 h	—	1.26	1.45	161

with v_i being the volume fractions and $M_i(T)$ (i = 1, 2, 3) the saturation magnetizations of the different magnetic phases, whose individual temperature dependence is described by (2). In this way, the magnetization $M(T)$ of (5) could be represented by

$$\Delta M(T)/M_0 = B'T^{3/2} + C'T^{5/2} + \ldots \tag{6}$$

with $B' = (v_1M_{01}B_1 + v_2M_{02}B_2 + V_3M_{03}B_3)/M_0$, $C' = (v_1M_{01}C_1 + v_2M_{02}C_2 + V_3M_{03}C_3)/M_0$ and $M_0 = v_1M_{01} + v_2M_{02} + v_3M_{03}$. Obviously, B' and C' for the multiphase system depend on the volume fractions v_i and coefficients B_i and C_i of the different magnetic phases. Using (6) to fit the experimental data of the temperature dependence of the magnetization (in the range 1.5 K $< T <$ 300 K), coefficients B' and C' were obtained for FeCoZr and FeCuNbSiB alloys in the amorphous and nanocrystalline states with different crystal sizes. For comparison table 1 gives the spin-wave parameters for the two series of samples. Figures 2 and 3 show experimental data and fitted curves for FeCoZr and FeCuNbSiB alloys, respectively. It is evident from table 1 that the values of B' and C' are larger in the amorphous than in the nanocrystalline state. Consequently, a smaller value of D was observed in the amorphous state for both series of samples. In addition, for the samples of FeCoZr alloys with grain size $d <$ 25 nm, B' and D exhibit no distinguishable difference, but for the sample with a larger grain size of d = 140 nm, D rises significantly. Previous investigation [8] proved that the spin-wave stiffness constant D is smaller in amorphous than in crystalline ferromagnets, due to the distribution of exchange interactions resulting from the structural fluctuations in amorphous materials. In nanocrystalline ferromagnets B_i for the ultrafine grain phases is smaller than for the amorphous matrix, leading to a smaller B' than in the amorphous state. With increasing grain sizes the volume fraction of amorphous phases decreases, resulting in a decrease of B' and an increase of D. Probably, for nanocrystalline FeCuNbSiB, the grain size did not grow very much, as observed by Hofmann *et al* [9]. Consequently, B' and D exhibit nearly the same values for different annealing times.

To summarize, the microstructure of FeCoZr and FeCuNbSiB nanocrystalline ferromagnets and its influence on the low-temperature magnetization and spin-wave excitations were studied. The results could be summarized as follows.

(1) By annealing amorphous FeCoZr in the temperature range 850–1200 K, ultrafine grains with increasing size (15–300 nm) were obtained. The microstructure of the nanocrystalline ferromagnet is composed of three magnetic phases: α-Fe(Co), $(Fe,Co)_2Zr$ and the amorphous matrix.

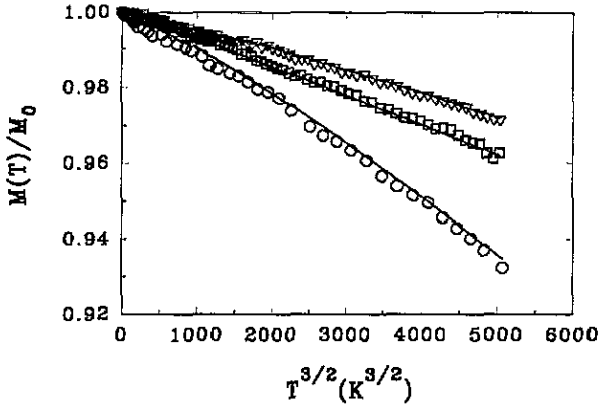

Figure 2. Reduced spontaneous magnetization plotted against $T^{3/2}$ in $Fe_{60}Co_{30}Zr_{10}$ alloys: ○, amorphous; □, annealed at 950 K; ▽, annealed at 1200 K; —, fitted curves.

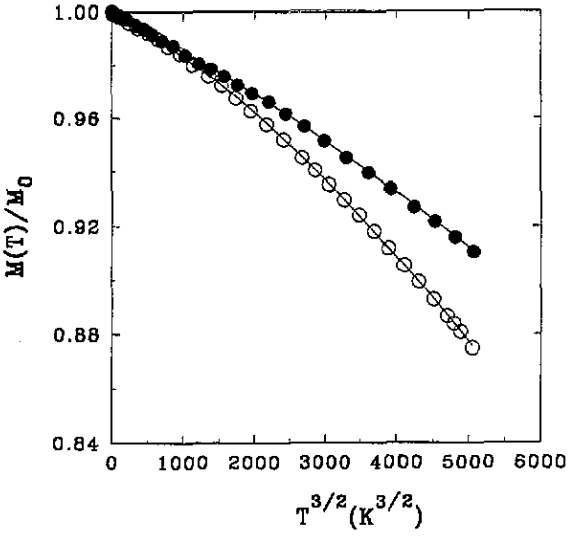

Figure 3. Reduced spontaneous magnetization plotted against $T^{3/2}$ in $Fe_{73.5}CuNb_3Si_{13.5}B_9$: ○, amorphous; ●, annealed at 823 K for 0.5 h; —, fitted curves.

(2) The low-temperature magnetization in both series of nanocrystalline samples decreases with increasing temperature and satisfies the Heisenberg-model equation. The larger value for the exchange stiffness D in the nanocrystalline than in the amorphous state and its dependence on grain size are in good agreement with predictions for ferromagnetic multiphase materials composed of amorphous and ultrafine-grain phases.

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