

Synopsis

This work has been devoted to the study of phase transformations involving chemical ordering and magnetic properties evolution in bulk Fe-Pt alloys composed of nanometer-sized grains. Nanocrystalline $\text{Fe}_{100-x}\text{Pt}_x$ ($x = 40-60$) alloys have been prepared by mechanical ball milling of elemental Fe and Pt powders at liquid nitrogen temperature. The as-milled Fe-Pt alloys consist of $\sim 100 \mu\text{m}$ sized particles constituted by randomly oriented grains having an average size in the range of 10-40 nm. Depending on the milling time, three major microstructure types have been obtained: samples with a multilayer-type structure of Fe and Pt with a thickness of 20-300 nm and a very thin (several nanometers) A1 layer at their interfaces (2 h milled), an intermediate structure, consisting of finer lamellae of Fe and Pt (below approximately 100 nm) with the A1 layer thickness reaching several tens of nanometers (4 h milled) and alloys containing a homogeneous A1 phase (7 h milled).

Subsequent heat treatment at elevated temperatures is required for the formation of the L_{10} FePt phase. The ordering develops via so-called combined solid state reactions. It is accompanied by grain growth and thermally assisted removal of defects introduced by milling and proceeds rapidly at moderate temperatures by nucleation and growth of the ordered phases with a high degree of the long-range order. The mechanism of ordering into the L_{10} structure depends on the alloy composition and the precursor microstructure. Heating of the $\text{Fe}_{100-x}\text{Pt}_x$ ($x = 40; 45; 50; 55$) alloys with the initial A1 structure induces the $\text{A1} \rightarrow \text{L}_{10}$ transformation, whereas in the $\text{Fe}_{50}\text{Pt}_{50}$ powders with the multilayer-type microstructure, the L_{10} formation proceeds via a metastable state according to the sequence: $\text{Fe/Pt precursor} \rightarrow \text{L}_{10} + \text{L}_{12} \rightarrow \text{L}_{10}$. The results based on the *in-situ* neutron powder diffraction and differential scanning calorimetry measurements show that for the equiatomic composition the $\text{A1} \rightarrow \text{L}_{10}$ transformation rate is fastest and a deviation from the equiatomic concentration significantly decelerates the ordering.

The combination of the chemically highly ordered L_{10} FePt and L_{12} Fe_3Pt and/or FePt_3 phases, formed after an isothermal heat treatment of the $\text{Fe}_{100-x}\text{Pt}_x$ ($x = 40 - 60$) powders with multilayer-type microstructure, yields a wide range of magnetic phenomena. In these alloys, the nanometer scale multilayer structure is preserved after annealing. In Pt-rich alloys, the decoupling of the L_{10} FePt lamellae by paramagnetic L_{12} FePt_3 results in the increased coercivity. In heat treated Fe-rich $\text{Fe}_{100-x}\text{Pt}_x$ powders, composed of the high magnetisation L_{12} Fe_3Pt and hard magnetic L_{10} FePt phases, adjusting the microstructure of the as-milled alloys modifies the extent of exchange coupling between the phases. In the 2 h milled and subsequently annealed Fe-rich Fe-Pt alloys, despite small grains, only a weakly pronounced exchange coupling is observed between the L_{12} Fe_3Pt and L_{10} FePt phases. Due to rather thick Fe_3Pt layers in terms of the soft magnetic phase domain-wall parameter, only those Fe_3Pt grains that are located at the interface between the Fe_3Pt and L_{10} lamellae are exchange-coupled to the hard magnetic L_{10} phase. In these powders, the lower (than predicted for non-interacting randomly oriented particles) remanence may be explained by the presence of dipolar interactions leading to magnetisation reversal already in positive applied fields. Due to the presence of the soft magnetic Fe_3Pt and/or A1 phases, weakly

coupled to the hard magnetic $L1_0$, the magnetisation reversal in these powders is noncooperative.

The phases in the annealed $Fe_{100-x}Pt_x$ powders milled for 4 h are distributed in the form of very fine lamellae. This nanometer-scale microstructure gives rise to large effects of exchange interactions between crystallites of the phases. As a result, the demagnetisation curves of the heat treated Fe-rich $Fe_{100-x}Pt_x$ samples show a single-phase magnetic behaviour, although several magnetic phases are present in the powders, and in both equiatomic and Fe-rich $Fe_{100-x}Pt_x$ alloys a significant remanence enhancement is observed. As a result of remanence enhancement, much improved energy density $(BH)_{max}$ values (up to 121 kJ/m^3) are obtained in the heat treated Fe-rich $Fe_{100-x}Pt_x$ powders milled for 4 h.

In the heat treated $Fe_{100-x}Pt_x$ powders milled for 4 h, due to the strong intergrain exchange coupling magnetisation reversal is more uniform compared to that in the 2 h milled and annealed powders. The demagnetisation curves of the former alloys are highly reversible. Nevertheless, the open recoil loops of the Fe-rich powders indicate deviations from the *ideal* exchange-spring-magnet behaviour. The recoil loop area decreases with decreasing soft magnetic phase fraction and no recoil hysteresis is observed in the essentially single $L1_0$ -phase $Fe_{50}Pt_{50}$ sample. In a two-particle interaction model elaborated in the present work, the existence of hysteresis in recoil loops has been shown to arise from insufficient coupling between the low- and the high-anisotropy particles. The recoil loop area turns out to be a function of the interaction strength, anisotropy of the particles, i.e. switching field distribution, and the fraction of the low-anisotropy component. The model reveals the main features of magnetisation reversal processes observed experimentally in exchange-coupled systems. In particular, it has been shown that if two particles interact, a *reversible* magnetisation rotation is possible also when the easy axes of the particles are parallel to each other and to the applied field, provided additional conditions are fulfilled.

Neutron diffraction has been used for the investigation of the magnetic structure of ordered and partially ordered nanocrystalline Fe-Pt alloys. It has been shown that the Fe moment in $L1_0$ -type Fe-Pt alloys is sensitive to composition and almost insensitive to the degree of order in the investigated compositional range. In particular, the Fe moments decrease with increasing Fe content, which is primarily a consequence of the lattice volume reduction.