Magnetic memory effect in FePd nanoparticles prepared by sonoelectrochemistry

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Abstract: Magnetic memory effect was newly observed in hard magnetic FePd nanoparticles prepared by sonoelectrochemistry. The applied magnetic field was changed in defined protocols during the magnetic relaxation process and the magnetic memory effect occurs when the field changes are larger than the critical field change of around 25 Oe. This magnetic memory effect can be explained by the magnetic reversal processes in strong interaction systems. The width of the energy barrier distribution of these hard magnetic particles is related to the critical field change deduced from this study.

1. Introduction

Hard magnetic properties of FePd nanoparticles have attracted interests due to their potential applications in ultrahigh density magnetic recording media [1-11]. FePd alloy nanoparticles are chemical stable and have the type $L1_0$ ordered structure with large uniaxial magnetocrystalline anisotropy of $K_u \sim 1.8 \times 10^7$ erg cm^{-3}. In previous studies, FePd were prepared by various methods, however the ordered $L1_0$ phase transition varies with preparing methods [4-11]. We have reported the hard magnetic properties of FePd nanoparticles [12] synthesized by sonoelectrochemical method, which was developed to make nanoparticles [13] and successfully used in preparation of FePt nanoparticles [14]. Upon the annealing at 450-600°C FePd nanoparticles have $L1_0$ order phase and their hard magnetic properties were investigated in dependence on the nominal compositions [12].

The slow dynamics such as non-exponential relaxation, ageing and memory effects are one the most interesting topics in magnetism. These phenomena provide many interesting information and are commonly observed in various systems, such as polymers [15], granular materials [16] and especially

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in spin-glass [17-20]. In spin-glass systems, the magnetic reversal process can be explained by the hierarchical model for weakly interacting nanoparticles or can be understood by a modification of the distribution energy barriers for superparamagnetic ones [21]. These phenomena however were rarely observed in strongly interacting systems, such as hard magnetic ones. Only an anomalous magnetic viscosity in exchange-spring magnet was reported [22]. In this study, we newly present the magnetic memory effect in FePd hard magnetic nanoparticles.

2. Experimental

FePd nanoparticles were synthesized by sonochemical reaction using a Sonic VCX 750 ultrasound emitter within 90 minutes and described elsewhere [12]. Palladium (II) acetate [$Pd(C_2H_3O_2)_2$] and iron (II) acetate [$Fe(C_2H_3O_2)_2$] were mixed with distilled water in a 150 ml flask and was ultrasonicated with power of 375 W, frequency of 20 kHz. The FePd nanoparticles with the nominal composition of Fe:Pd equal 1.5:1 were collected from the ultrasonicated solution by using a centrifuge with alcohol at 9000 rpm for 30 minutes. The collected powders were left for drying at $70^{o}C$-$75^{o}C$ then were annealed at temperatures of 550°C under continuous flow of (N_2 + Ar) gas at heating rate of $5^{o}C$/min for 1 h. The morphology and structure of samples were investigated and reported elsewhere [12]. Hysteresis loop and magnetic reversal processes of samples were studied at room temperature by using a Vibrating Sample Magnetometer (VSM, DMS 880).

3. Results and discussion

Fig. 1 shows magnetic hysteresis loop at room temperature of the FePd nanoparticles annealed at 550°C. Sample has hard magnetic properties with the saturation magnetization M_S of 103 emu/g, the coercivity H_C of 2100 Oe, the magnetic squareness M_r/M_S of 0.65 (M_r is the remanent magnetization).
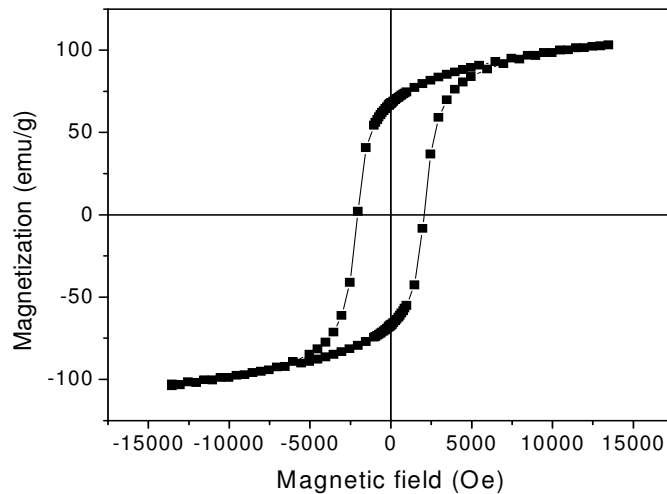

Figure 1. Hysteresis loop at room temperature of FePd nanoparticles annealed at $550^{o}C$.

The magnetic viscosity or the time dependence relaxation of the FePd nanoparticles were measured under a negative magnetic field after applied an external magnetic field of 13.5 kOe. The magnetic relaxation follows an exponential function of time and the magnetic viscosity $S = dM/d(\ln t)$ can be calculated. Fig. 2 illustrates the measured field dependence at room temperature of the magnetic viscosity S of the studied sample, which deduced from the time dependence magnetic relaxation measurement shown in the inset. The magnetic viscosity shows the maximum at around - 2000 Oe, which is close to the value of the coercivity H_C of 2100 Oe.
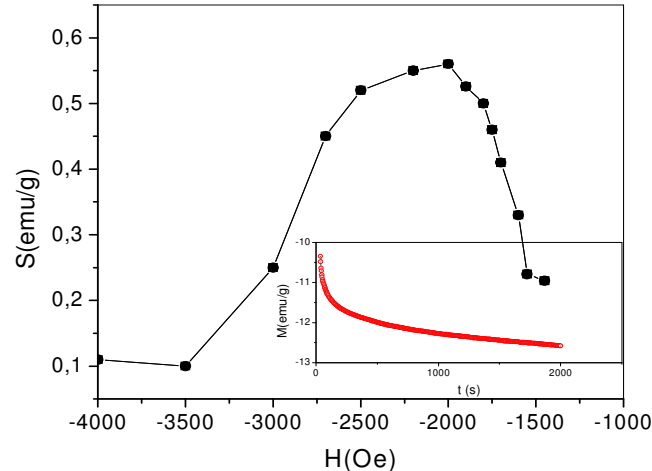

Figure 2. Applied field dependence of the magnetic viscosity S of studied FePd nanoparticles deduced from the time relaxation of the magnetization shown in the inset.

During the relaxation at H_1 = - 2000 Oe, the magnetic field was switched to a "smaller" value of $H_2 = – 1550$ Oe (H_2 has smaller absolute value) and was kept it for $\Delta t = 300$ s before switched back the magnetic field to initial value of $H_1 = – 2000$ Oe as shown in Fig. 3. After the H_2 was applied, the magnetization suddenly changed to the smaller value and kept almost constant. The relaxation continued when the magnetic field switched back to the initial value of H_1. The magnetization value is almost same as the value before switching the magnetic field. This behavior repeated at $t = 900$ s when the magnetic field was switched again. The amounts of changes of the magnetization ΔM are the same at the switching as can be seen in the Fig. 3.

We repeated the relaxation measurements with different values of $\Delta H = H_2 - H_1$ and the longer keeping times Δt of H_2 switching and the results were similar (all data not shown). Fig. 4 shows one of these results with $\Delta H = 100$ Oe and $\Delta t = 600$ s. The ΔM in this case is smaller than the ΔM in Fig. 3, where the ΔH is larger of 450 Oe. The curves t_1', t_2' and t_3' are obtained by shifting the curves t_1, t_2 and t_3 by a time period Δt, indicating the continuously relaxation of the magnetization if the field was not switched. The inset shows that all of these curves have same magnetic viscosity. In other words, we presented the magnetic memory effect in the FePd nanoparticles. The magnetic memory effect even occurs when the magnetic field changes from H_1 to H_2 and then to H_3 which is smaller than H_2, after that turns back to H_1, as indicated in Fig. 5 where the ΔH is large of 200 Oe.
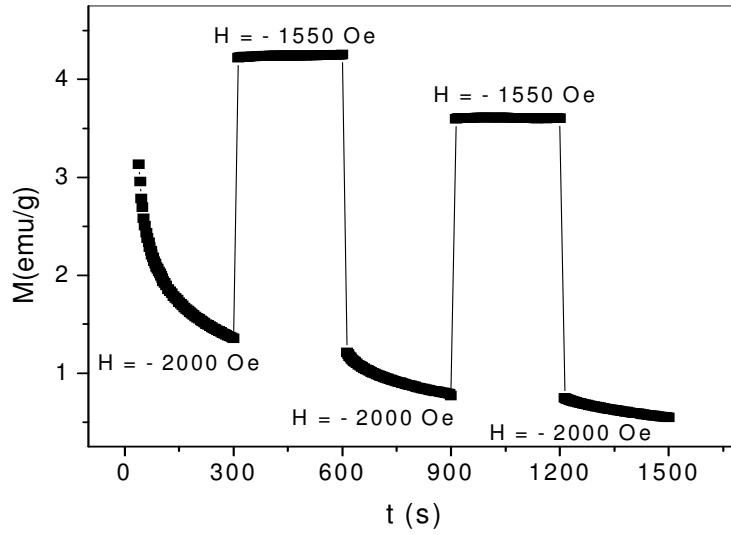
Figure 3. The magnetic relaxation of studied FePd nanoparticles under the field $H_1 = - 2000$ Oe and $H_2 = - 1550$ Oe as the function of time.
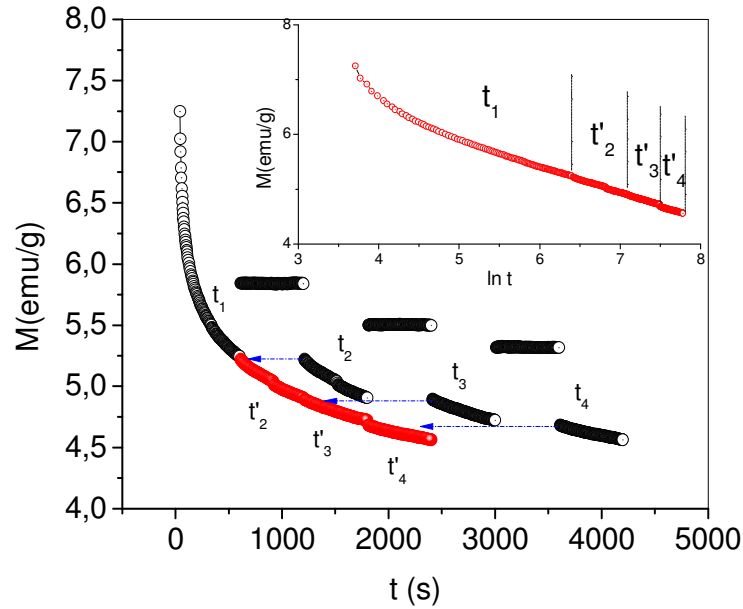

Figure 4. The magnetic relaxation of studied FePd nanoparticles under the field change of $\Delta H = 100$ Oe as the function of time. The inset shows the logarithmic dependence as time varies of the magnetization.
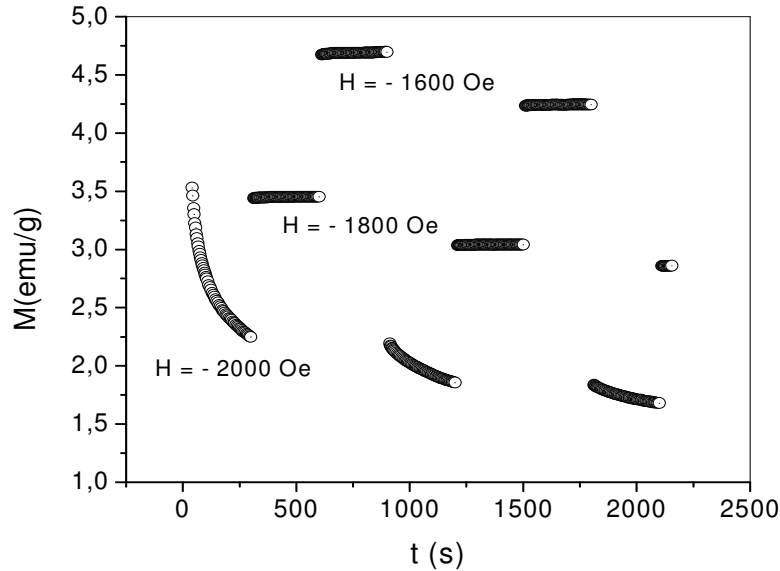
Figure 5. The magnetic relaxation of studied FePd nanoparticles under the field $H_1 = -2000$ Oe, $H_2 = -1800$ Oe and $H_3 = -1600$ Oe as the function of time.

It is clear that the magnetization will continuously relax with no such memory effect if there is no change of the magnetic field. Therefore there is a critical small change of the magnetic field ΔH, where the magnetic memory effect starts to occur. This ΔH should be smaller than 100 Oe which already shown in Fig. 4. Fig. 6 shows the relaxation of the magnetization with $\Delta H = 25$ Oe, i.e. $H_2 = -1975$ Oe, where the magnetic memory effect is not presented. The magnetization decreases almost linearly during the time Δt and there is a gap between the curves t_1, t_2' and t_3' as shown in the inset (t_2' and t_3' are observed by shifting the curves t_2 and t_3 by Δt, similarly to the process in Fig. 4), which shows that the magnetic viscosities deduced from these curves are different. It is not like in Fig. 3 and 4, after the magnetic field switched back to the initial value H_1, the value of the magnetization M is not same as the value of M just before the switching from H_1 to H_2. This measurement was repeated (data not shown) and the similar results were observed at $H_2 = -1990$ Oe ($\Delta H = 10$ Oe) but the magnetic memory effect still occurs at $H_2 = -1950$ Oe ($\Delta H = 50$ Oe). Hence it can be said that the critical ΔH_{crit} is around 25 Oe.
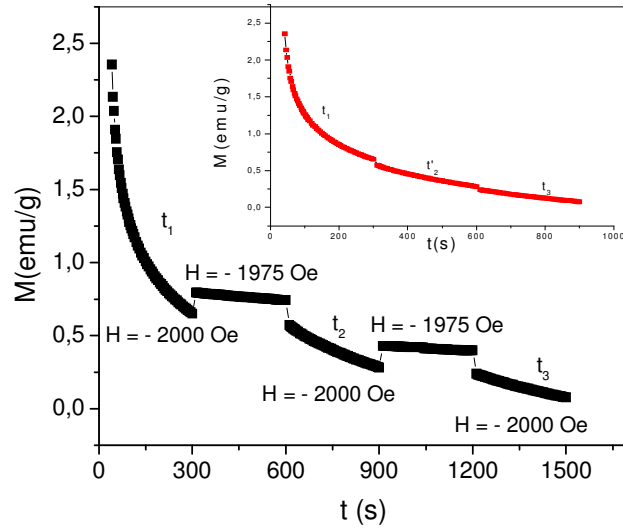
Figure 6. The magnetic relaxation of studied FePd nanoparticles under the field H_1 = - 2000 Oe, H_2 = - 1975 Oe as the function of time.

During the magnetic relaxation at H_1, instead of changing the magnetic field to "smaller" field, we changed it to "higher" field of H_2 and the results were shown in Fig. 7 with H_1 = - 1600 Oe and H_2 = - 2100 Oe. The huge change of the magnetization was observed at the switch of the magnetic field. After the field switching from H_1 to H_2, the magnetization continues to relax with different viscosity. When the field returned to the initial value, the magnetization has smaller change compared to the previous one. The magnetic memory effect does not occur in this case. However, when the magnetic field repeats the protocol similarly as in the Fig. 3 and 4, the magnetic memory was again observed.
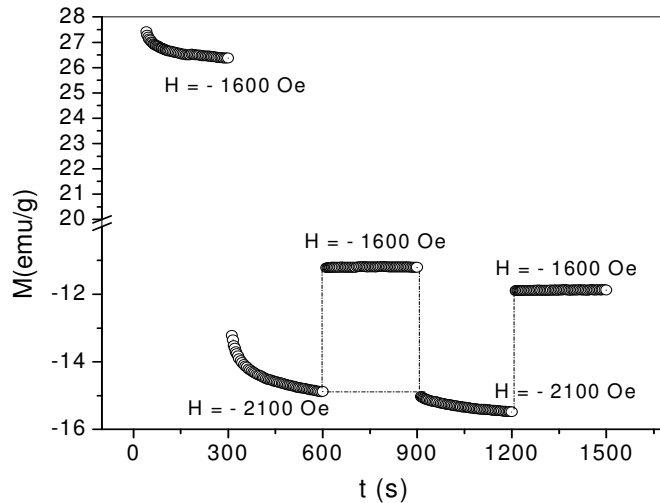

Figure 7. The magnetic relaxation of studied FePd nanoparticles under the field H_1 = - 1600 Oe, H_2 = - 2100 Oe as the function of time.

These results can be understood by the energy barrier distribution, which may relate to the particles size distribution [23]. The energy barrier for the magnetic relaxation is changed by changing the applied magnetic field at the same temperature. When the ΔH is large, the energy barrier increases significantly and caused a halt in the relaxation of the magnetization. In other words, the magnetic moments in the sample cannot reverse because of high-energy barriers. By switching the field to initial value, the reversal process will continue, as the field was not changed indicated by curves t'$_2$ and t'$_3$ in Fig. 4. The small discontinuations can be assigned to the lag due to the changing field in VSM.

When the ΔH is small as shown in Fig. 6, the changes of energy barriers are also small and have the broad distribution because of the large size distribution of the sample. In this case, there are some magnetic moments with higher energy than others can overcome the energy barriers and the magnetic reversal process still occurs after switching the field. The relaxation rate is quite small in this case then the decreasing of the magnetization seems to be linear in this time period. These mean that if the sample contains only one size of the particles or exactly the same energy barriers, any increase of the magnetic field will halt the reversal process. On the other hand, if the distribution is broad, ΔH_{crit} should be high in order to observe magnetic memory effect. It can be said that the ΔH_{crit} is closely related to the width of the energy barrier distribution.

4. Conclusion

Magnetic memory effect of FePd nanoparticles prepared by sonoelectrochemistry were systematically studied and showed the critical field change of 25 Oe, where the magnetic reversal process starts to occur. These results can be explained by the conventional magnetic reversal model in hard magnetic systems and provide some information related to the distribution of the energy barriers.

Acknowledgment

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