NMR investigation of domain wall dynamics and hyperfine field anisotropy in magnets by the magnetic video-pulse excitation method

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Abstract. Two-pulse nuclear spin echoes were studied experimentally depending on the time of application and pulse amplitudes of the DC magnetic field-magnetic video-pulses (MVP) as well as on the value of the external magnetic field. The measurements were performed with nanopowders and polycrystals of metallic cobalt, in lithium ferrite and half metal Co₂MnSi. Two types of dependences of these signals on time of application of MVP with respect to moments of application of exciting radio-frequency pulses were established, which were determined by the degree of anisotropy of local hyperfine fields. The mechanisms of influence of the pinning and mobility of domain walls on the revealed specific features of the signals under study are also discussed. It is shown that temporal spectra of the MVP effect on two-pulse echoes in multidomain magnets are determined by the parameters of domain walls and can be used for qualitative and quantitative characterization of the domain wall dynamics of magnets.

For characterization of microscopic magnetic properties of magnets the method of MVP excitation was used [1] by which the influence of MVP field on the nuclear spin echo in magnetic materials was studied. The investigation includes the determination of nuclear spin echo dependences in a sample on the amplitude $H_d$ and duration $\tau_m$ of MVP applied in the interval between RF pulses or between the second RF pulse and echo signal (asymmetric action), as well as, during RF pulse action, Figure1a (symmetric action).

The decrease of echo signal intensity at switching-on of the MVP in intervals between RF pulses or between RF pulse and echo signal is related with the breakdown of phase coherence in a system of processing nuclear moments because of change in the local nuclear frequencies caused by hyperfine field (HFF) anisotropy under the action of MVP displacing domain walls.

The switching-on MVP symmetric in respect of the second RF pulse results in the fact that the first and the second RF pulses successfully excite nuclei changing their positions in domain walls defining on their resonance frequencies and the RF field gain factor ($\eta$) [1]. At sufficiently short RF pulses the resonance frequency change could be neglected but the change of $\eta$ results in the decrease of echo intensity.
Figure 1. a) Timing diagrams of the relative intensity $I/I_{\text{max}}$ dependence of the two-pulse echo (●) on
the temporal location of a magnetic video-pulse of $H_d=5$ Oe, for $^{57}$Fe NMR in lithium ferrite at:
$\tau_1 = \tau_2 = 0.8 \mu s$, $\Delta \tau = 21 \mu s$, $\tau_d = 3 \mu s$, $f_{\text{NMR}}=74.0$ MHz, $H_d=0$. $\tau_1$, $\tau_2$, $\Delta \tau$, $\tau_d$ are RF pulse durations,
time interval between them and magnetic pulse duration, correspondingly;

b) Timing diagrams of the intensity dependence of the two-pulse echo on the temporal location of a
magnetic video-pulse of $H_d$, in cobalt thin magnetic films at: $\tau_1 = \tau_2 = 1.5 \mu s$, $\Delta \tau = 9 \mu s$, $\tau_d = 3 \mu s$,
$H_d=10$ Oe, $f_{\text{NMR}}=216.5$ MHz, $I_0$ – echo amplitude at $H_d=0$;

c) NMR spectrum of Co film (●) and frequency spectra diagrams for magnetic video-pulse influence
for symmetric (▲) and asymmetric (▼) application at: $\tau_1 = 1.3 \mu s$, $\tau_2 = 1.5 \mu s$, $\Delta \tau = 9 \mu s$, $\tau_d = 3 \mu s$,
$H_d=10$ Oe, [1].

As it follows from Figures 1–2, the essential difference takes place for the MVP action on the two-
pulse echo in different magnets.

Figure 2. Timing diagrams (TD) of the intensity dependence of the two-pulse echo on the temporal
location of the magnetic pulse with duration $\tau_d$ in Co$_2$MnSi (a) for $^{59}$Co NMR at: $\tau_1 = 1.1 \mu s$, $\tau_2 = 1.4$
$\mu s$, $\Delta \tau = 10 \mu s$, $\tau_d = 2 \mu s$, $f=145.5$ MHz, $H_d=550$ Oe and (b) for $^{55}$Mn NMR at: $\tau_1 = \tau_2 = 3 \mu s$, $\Delta \tau = 7$
$\mu s$, $\tau_d = 2 \mu s$, $f=354$ MHz, $H_d=300$ Oe.

c) Two-pulse echo intensity dependences on magnetic video-pulse amplitude $H_d$ in Co$_2$MnSi (Figure 2)
1) for $^{59}$Co spin echo for symmetric (▲) and asymmetric (▼) influence at: $\tau_1 = \tau_2 = 2 \mu s$, $\Delta \tau = 10 \mu s$, $\tau_d = 3$
$\mu s$, $f_{\text{NMR}} =145$ MHz., and 2) for $^{55}$Mn for symmetric (▲) and asymmetric (▼) influence at: $\tau_1 = 0.8 \mu s$, $\tau_2 = 0.9 \mu s$, $\Delta \tau = 8 \mu s$, $\tau_d = 1.6 \mu s$, $f_{\text{NMR}} =353$ MHz. $I_0$ – echo amplitude at $H_d=0$.[1]

The dependence of its intensity on the time of application of a MVP for cobalt thin magnetic film
(TMF), and $^{59}$Co echo signal in Co$_2$MnSi (Figure 1b and 5a), correspondingly, differs considerably
from closer-to-each-other dependences for lithium ferrite (Figure 1a) and $^{55}$Mn echo signal
dependence in Co$_2$MnSi (Figure 2b). The spectral diagrams of MVP influence of TMF under the
influence of MVP are given in Figure 1c, which correspond to the symmetrical and asymmetrical actions of MVP. As can be seen from Figs. 1 and 2, the MVPs noticeably affect the echo spectra.

Note the difference in the TD of $^{59}$Co and $^{55}$Mn in Co$_2$MnSi. This indicates the difference in the anisotropy of HFF for these nuclei.

So, the type of TDs is defined mainly by the hyperfine field anisotropy of corresponding nuclei which is small for $^{57}$Fe and $^{59}$Mn as compared for $^{59}$Co positions.

The reason for this could be understood from Figure 2c, where reduced echo intensity dependences on the MVP amplitude is shown for $^{55}$Mn and $^{59}$Co positions in Co$_2$MnSi. It is seen that for these positions amplitude dependences for asymmetric action of MVPs differ much stronger than ones for symmetric influence.

While a TD type is defined mainly by the anisotropic part of hyperfine interaction the degree of suppression of echo signals by MVP strongly depends on the DW mobility.

Note also that TDs in case of half metallic Co$_2$MnSi give visual pictures describing different hyperfine anisotropies at the $^{59}$Co and $^{55}$Mn sites.

In the cases we studied, the type of the TD was determined by the anisotropy of HFF and did not change for the given nucleus with increasing mobility of DW in the material (for instance, for polycrystalline cobalt and TMF of cobalt).

We can qualitatively understand the origin of two types of TD from the form of amplitude dependences of the MVP effect in Co$_2$MnSi, Figure 2c. The forms of dependences of echo signals on the symmetric and asymmetric effects for two different locations in one sample of Co$_2$MnSi show how the movement of the same DWs affects the echo signals from $^{59}$Co and $^{55}$Mn nuclei, which essentially differ in the anisotropy of HFF. It is clear that the diagrams of the MVP effect are close in the symmetric excitation case and differ essentially in the asymmetric effect case. These peculiarities can be understood if we assume that the diagrams of the symmetric effect are mainly determined by the mobility of DWs, i.e. by the coercive force of the sample, whereas the diagrams of the asymmetric MVP effect are mainly determined by the anisotropy of HFF. Hence we can now understand the dependence of the echo signal under the symmetric MVP effect with application of the external magnetic field $H_0=1000$ Oe in lithium ferrite, Figure 3. The obtained result testifies that the remained DWs are fixed in stronger pinning centers with application of the external field and hence that the echo signals are less suppressed under the symmetric MVP effect.

![Figure 3](image)

Figure 3. Amplitude dependence of the TPE intensity at MVP $H_d$, $H_0=0$ Oe (●) and $H_0=1000$ Oe (□)

Different behaviors of these dependences at different values of $H_0$ are clearly expressed in Figure 3. To achieve the same intensity of TPE at $H_0=0$ and $H_0=1000$ Oe, different values of $H_d$ are necessary.

As was mentioned above, recording the dependence of echo signal intensity on the amplitude $H_d$ ($I(H_d)$), it is possible to determine quantitatively the value of the MVP amplitude causing the shift in the DW equal to its effective thickness. This fact allows us to assess easily and quantitatively the DW pinning and the coercive force of the sample for the nuclei contributing the intensity of the NMR spin echo signal.
Figure 4 shows the experimental data on the dependences of the echo signal intensity on the MVP amplitude $H_d$ under the symmetric MVP effect for cobalt nanopowder with mean grain diameter 100 nm, synthesized by electron beam technology [2], cobalt nanopowder with grain diameter 22 nm, produced by Sun Innovations, Inc., USA, and polycrystalline cobalt powder, grain size ~50 μm.

As is well known, a decrease in the grain size of cobalt powder from the order of 1 μm to ~150 nm is accompanied by a significant increase in the coercive force of the sample associated with the decreasing size and the increasing part of surface effects. This fact also reflects on the dependence of the echo intensity on the value of the MVP. In all samples the echo signal intensity hardly changed up to the magnetic field values of the order of an average demagnetizing field because of the presence of DW. However, in the case of nanopowders, the mobility of DW decreases significantly, which should reflect on the character of the $I(H_0)$ dependence of the echo intensity on the value of the external magnetic field $H_0$. Besides, because of the increase in the coercive force, a much more powerful external field will be needed to suppress the echo signal in nanopowders. The measurement results on the dependence of the echo signal intensity on the value of the external magnetic field support the above reasoning, Figure 5.

The DW pinning can also increase when the grain sizes of cobalt nanopowders have decreased after their treatment with the help of a nanomill. In Fig. 6 the dependences of the echo signal intensities on the value of the external magnetic field are shown for polycrystalline cobalt powder with mean grain size ~50 μm and carbon nanopowder doped with cobalt nanoclusters (mean size ~50 nm) [3], and for the latter nanopowder after its grinding with a nanomill for half an hour, similar to the dependences shown in Figure 5.
Figure 6. Dependences of the echo intensity on the value of the external magnetic field for Co powder with grains ~50 μm in size (♥) and carbon nanopowder doped with cobalt nanoclusters with grains ~50 nm in size (○), and for the latter nanopowder after grinding with a nanomill for half an hour (●).

A similar effect should be expected in the result of annealing of the sample. For instance, Figure 7 shows the results of similar investigation for TMF cobalt samples fabricated in the form of films ~2 μm thick by the electroless deposition on quartz plates and annealed at 500°C for 3-4 hours [4]. They were also considerably magnetically softer than the cobalt powder with grains ~50 μm in size.

Figure 7. Dependence of the echo signal intensity for the cobalt film on the value of the external magnetic field before (●) and after (□) annealing.

Thus, based on the investigation of echo intensity dependence on the value of the external magnetic field, magnets can be classified by the degree of their magnetic rigidity or by the mobility of DW. For instance, in Fig.8 it is seen that, of half metal Co₂MnSi, cobalt and lithium ferrite samples, the magnetically softest sample is lithium ferrite, while half metal Co₂MnSi is the magnetically hardest one.

Figure 8. Dependence of the NMR echo intensity for 55Co nuclei in Co₂MnSi (■), in cobalt (○), and for 57Fe in lithium ferrite (●) on the value of the external magnetic field.
It should be noted that, using the technique of chemical vapor deposition (CVD) [3], we produce carbon powders doped with magnetic nanoclusters with average diameter ~50 nm that have the coercive force of different degree depending on the temperature in the reactor chamber. The fabricated magnetic nanopowders could be both relatively magnetically soft and magnetically harder.

In Figure 9 it is shown the amplitude diagrams of the symmetric MVP effect for the nanopowders prepared at T=1200°C and 700°C.

![Graph showing the amplitude diagrams of the symmetric MVP effect.]

At the same time, the variation in the degree of magnetic softness of the sample, i.e. the decrease in the DW pinning does not change the character of TDs of the MVP effect, because they are determined by the value of anisotropy of HFF of the nuclei.

**Conclusion**

In the result of experimental investigation of different types of TDs of the MVP effect, the existence of TDs of two types for the magnets under study was revealed. The type of TD is mainly determined by the anisotropy of HFF. We also studied the mechanism of the impact of the size of magnetic powder grains, and the pinning and mobility of domain walls on the revealed specific features of two-pulse echo signals observed under the MVP effect.

**Acknowledgments**

This work is supported by the Science and Technology Center in Ukraine (STCU) and the Shota Rustaveli National Science Foundation (SRNSF) Targeted Initiative Program 6081 grant.

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