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## Investigation of the single-pulse NMR echo origin in cobalt using additional magnetic video-pulses

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A comparative study of the formation mechanism of a single-pulse echo and its analog formed jointly by radio-frequency and two magnetic video-pulses, when their amplitude exceeds the pinning force of domain walls, is carried out. The results obtained support the assumption that the single-pulse echo signal is formed by the mechanism of local field distortions at nuclei in domain walls when they are displaced by RF pulse fronts due to the inhomogeneity of the hyperfine field and the NMR amplification factor in the domain walls.

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#### 1. Introduction

Nuclear magnetic resonance (NMR) in ferromagnets was discovered in 1959 [1]. Since then, NMR became one of the most powerful methods for studying the structure and properties of magnetically ordered substances [2–5].

Spontaneous magnetization of a ferromagnet such as cobalt polarizes s-electrons, creating an effective hyperfine field (HFF) on nuclei through the Fermi contact interaction (a brief discussion of the origin of the effective field and other features of the NMR methods in magnets is given in [2,6]). The existence of an effective field on nuclei makes it possible to observe NMR in the absence of an external constant field, which is necessary when observing NMR in non-magnetic materials.

The HFF reaches a value of the order of  $10^5-10^6$  Oe. The typical NMR linewidth in nonmagnetic materials is about 1 Oe, while in ferromagnetic materials the linewidths range from about  $10^2$  Oe in iron to greater than  $10^3$  Oe for dilute impurities in iron. The detection of NMR in magnets is greatly facilitated by the fact that the RF field acts on the nuclei through electronic magnetization. This leads to an RF field amplification by the enhancement factor  $\eta_d \sim 10^2$ within the domains and to a much stronger amplification effect in a DWs with  $\eta_w \sim 10^4$ .

For this reason, magnetic NMR spectrometers are in some respects simpler than conventional NMR spectrometers. However, they must be tuned over a wide frequency range up to 1 GHz due to the broad NMR lines in magnets. A remarkable feature of the manifestation of NMR in magnets is that in many cases the main contribution to the intensity of resonant absorption is made by nuclei located in the DWs. Since DWs are easy to control under the action of magnetic video-pulses, their use is a convenient method for studying the features of the formation of additional echo signals arising under the action of a magnetic video-pulse (MVP) [7].

For the first time, the effect of MVP on the DW dynamics in a ferrite sample was studied by Galt [8]. He showed that, under the influence of MVP, DWs are torn-off from fixing (pinning) centers and begin to move with a constant velocity v linearly depending on the MVP amplitude H:

$$v = S(H - H_0),\tag{1}$$

where S is the DW mobility,  $H_0$  is the DW pinning force.

Since an external RF field acts on the nuclei through the electronic subsystem, the explanation of the phenomena considered below should be based on the understanding of the motion of the electron magnetic moments M in the DW under the action of the MVP. The displacement of the DW, even if it is insignificant, can be accompanied by a large rotation by M. In this case, the rotation angle M inside the DW is proportional to the displacement of the DW. This process is accompanied by changes in the HFF and the factor  $\eta$  on the nuclei, proportional to the displacement of the DW, due to the anisotropy of the HFF and the inhomogeneity of the factor  $\eta$  in the DW of magnets [9].

In this work, we use a simple technique for generating an analog of a single-pulse echo (SPE) signal [10], for brevity called a magnetic echo (ME) signal, using two additional MVPs acting in combination with the RF pulse to study the process of its formation and comparison with the properties of the SPE. As is known [11], the SPE is a resonant response of an inhomogeneously broadened nuclear spin system upon the application of a single RF pulse, which is formed at a moment of time approximately equal to the length of the RF pulse after its termination, Fig. 1.



Figure 1. Oscillogram of a single-pulse echo signal of <sup>59</sup>Co nuclei in cobalt (upper trace). On the lower trace, it is shown a wave meter signal showing the position of the exciting RF pulse (the highlighted part of the trace) with NMR frequency  $\nu(\omega/2\pi) = 213$  MHz and RF pulse duration  $\tau_{\rm RF} = 20 \,\mu s$ , T = 293 K.

The mechanisms of the SPE formation can be divided into the following two classes: the first class refers to the so-called edge-type mechanisms, when the fronts of the RF pulse act similarly to two resonant RF pulses in the Hahn two-pulse echo (TPE) method, and the second class is the mechanisms of the formation of the SPE of an internal nature [12].

The important role of the fronts of RF pulses for edge mechanisms is due to the fact that it is at these instants of time that the direction of the effective magnetic field

$$\mathbf{H}_{\rm eff} = (\Delta \omega_j \, \mathbf{z} + \omega_1 \, \mathbf{y}) / \gamma_n$$

changes in the rotating coordinate system (RCS), where  $\gamma_n$  is the nuclear gyromagnetic ratio, z and y are the unit vectors in the RCS,  $\Delta \omega_j = \omega_{\text{NMR}_j} - \omega_{\text{RF}}$  is the detuning of the *j*-th isochromate,  $\omega_1 = \gamma_n \eta H_1$  is the RF field amplitude in frequency units,  $\eta$  is the RF field enhancement factor [2,3].

Such changes in the direction of  $\mathbf{H}_{\text{eff}}$  arise in the case of a non-resonant mechanism, mainly due to non-resonant effects, when the carrier frequency of an ideal RF pulse is detuned from the center frequency by an amount  $\Delta\omega_0$ , comparable to or greater than the width of the inhomogeneously broadened NMR line  $\Omega_{j\max}$ , Fig. 2.



Figure 2. (a) Precession of the *j*-isochromate of the nuclear magnetization vector  $\mathbf{m}_j$  around the direction of the effective field  $\mathbf{H}_{\text{eff}}$  in a rotating coordinate system (RCS). (b) Non-resonant excitation of a single-pulse echo at  $\Delta\omega_0 > \Omega_{j\max}$ , where  $\Delta\omega_0$  is the detuning from the center of the studied section of the NMR line with a width  $\Omega_{j\max}$ .

In addition, a similar effect also takes place for the mechanism of distortion of the fronts of the RF pulse, which inevitably arises during the generation of RF pulses due to the non-ideal properties of the components of electronic circuits [13]. In both these cases, the deviation angle  $\mathbf{H}_{\text{eff}}$  from the equilibrium direction of the nuclear magnetization vector  $\mathbf{m}$  in the RCS plays the role of the angle of rotation of the nuclear magnetization  $\mathbf{m}$  around  $\mathbf{H}_{\text{eff}}$  in the Hahn TPE echo method [10].

The internal mechanisms of the SPE formation are due to different types of nonlinearities of spin systems, for example, nuclear spin systems in weakly anisotropic magnets at low temperatures have a large dynamic frequency shift (DFS). In these systems, the resonant frequency of nuclear spins depends on the angle of deviation of the spins from their equilibrium directions, and the so-called frequency-modulated mechanism of SPE formation is effective [3]. Another internal mechanism for the formation of SPE and its secondary signals, the so-called multipulse mechanism, was proposed for systems with nonlinear dynamics of nuclear spins upon excitation by RF pulses in the presence of a large inhomogeneous broadening of the NMR line of the Larmor and Rabi types and non-equilibrium of the nuclear spin system in front of RF pulses of the sequence [12]. Such a situation is also realized in lithium ferrite [14] and at the application of low-power RF pulses in cobalt [15].

For such systems, a single-pulse excitation with a shorter repetition period  $T_{\rm R}$  compared to the longitudinal relaxation time  $T_1$  can be used to amplify the SPE signal when observing a cumulative echo [17, 18]. Within the framework of this mechanism, the SPE signal is absent during the single-pulse excitation, but it is possible to observe the signal of a two-pulse stimulated

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echo (TPSE) [19] and the SPE signal during multipulse excitation [14, 15].

It was shown in [14] that the SPE properties in lithium ferrite can be understood within the framework of a multipulse non-resonant mechanism under conditions of non-equilibrium of the spin system before a front of RF pulse in a series of exciting RF pulses [12].

The SPE in cobalt was first experimentally observed by Stearns [20], contrary to theoretical estimates indicating its absence. Particularly intense SPE signals were observed in the hexagonal close-packed (hcp) phase, which is characterized by a particularly strong HFF anisotropy. In subsequent works [13], in order to explain the observed SPE effect, a mechanism for the formation of SPE was proposed, due to distortion of the fronts of an RF pulse arising from transients in radio circuits when the RF pulse is turned on and off. In this case, the fronts of an RF pulse play the role of a pair of RF pulses in the Hahn TPE method, and the occurrence of the SPE can be explained according to the experiment.

To study the role of the inhomogeneous broadening of the NMR line due to the large anisotropy of the HFF in cobalt, a comparative study of the SPE signals in lithium ferrite and cobalt was carried out in [15] in order to elucidate the mechanism of the formation of the SPE in cobalt. To explain the obtained experimental results, an alternative internal mechanism of distortion of the fronts of the RF pulse was proposed, to account for the role of the strong anisotropy of the HFF in cobalt, taking into account the fact that when the electron magnetization M deviates from the equilibrium direction when the RF pulse is turned on and off, in the region of the fronts of the RF pulse, significant distortions of the oscillating local RF field due to the strong anisotropy of the HFF take place. It was shown in [15] that the SPE in cobalt can be formed by a distortion mechanism, which is mainly due to the anisotropy of the HFF and is effective above a certain threshold value of the RF pulse power.

Below this threshold value, the SPE is formed by a multipulse non-resonant mechanism [12], as in lithium ferrite, where a similar distortion mechanism is ineffective due to the relatively weak anisotropy of the HFF in this ferrite [14].

The role of the strong anisotropy of the HFF in cobalt was previously demonstrated in the study of the effect of modulation of the TPE decay envelope under the action of an additional weak low-frequency (LF) magnetic field in cobalt [21]. We also note that earlier in [16], in the framework of a theoretical model with a phase distortion near the fronts of the RF pulse, the inhomogeneity of the resonant field on the nuclei of a ferromagnet was taken into account due to the inhomogeneity of the NMR enhancement factor  $\eta$  in it, which made it possible to describe qualitatively the experimentally observed SPE signals <sup>59</sup>Co nuclei in Co<sub>2</sub>MnSi. The inhomogeneity of the NMR enhancement factor  $\eta$  in lithium ferrite with a high DW mobility also manifests itself in a strong modulation of the TPE decay envelope under the action of a weak LF magnetic field [22], similar to the modulation effect in cobalt, although the anisotropy of the HFF in lithium ferrite is an order of magnitude less than that of cobalt.

The purpose of this work is to show the possibility of generating a ME signal, which is an analog of the SPE signal and arises under the combined action of a RF pulse and a pair of MVP pulses, which is formed by the distortion mechanism due to the anisotropy of the HFF and the inhomogeneity of the NMR enhancement factor  $\eta$  at exposure to MVP and RF pulses, which leads to a jump-like change in the effective field  $\mathbf{H}_{\text{eff}}$  in the RCS acting on the nuclei in the DWs, as well as a comparison of its properties with those of the SPE to elucidate the mechanism of its formation.

#### 2. Experimental results and their discussion

The measurements were carried out on a phase-incoherent spin echo spectrometer [6,23] in the frequency range of 200–400 MHz at a temperature of 293 K. In the range of 200–400 MHz, a commercial Lecher-type generator with a two-wire line including two inductors with different numbers of turns was used. For pulse lengths in the range from 0.1 to 50  $\mu$ s, the maximum amplitude of the RF field produced on the sample was about 3.0 Oe, and the front steepness was no worse than 0.15  $\mu$ s. Receiver dead time is ~ 1  $\mu$ s.

The scheme of the experiment on the combined RF and MVP action is given in [6]. A pulsed magnetic field was created by a gated current stabilizer of adjustable amplitude and an additional copper coil, which made it possible to obtain magnetic field pulses of the order of 500 Oe for a sample size of  $\sim 10$  mm.

Cobalt micropowders were obtained by the melting method [23] with an average grain size of  $\sim 10 \,\mu$ m. The characteristic parameters of RF pulses are: the duration is a few microseconds, the delay between them is tens of microseconds, and the carrier frequency of 213 MHz coincides with the frequency of the nuclei in the center of the DW of the face-centered cubic phase (fcc) of cobalt.

The study of the dependence of the intensity of an SPE on the power of RF pulses in cobalt showed that at low powers of RF pulses, only a free precession signal is observed [13]. Besides it, signals of a cumulative echo [17,18] and a two-pulse stimulated echo (TPSE) [19], Fig. 3, are also observed in this case, as in the case of the multipulse mechanism of SPE formation [12,14]. The SPE signal in cobalt, formed by the distortion mechanism, occurs at a sufficiently high power of the RF pulse [13], Fig. 1.



**Figure 3.** Oscillogram of the two-pulse stimulated echo (TPSE) signal in cobalt at low power RF pulses and single exposure to a pair of RF pulses  $\tau_1 = \tau_2 = 8 \,\mu s$ ,  $\tau_{12} = 27 \,\mu s$ ,  $T = 293 \,\text{K}$ . The lower trace is a signal from wave meter, showing the location and duration of RF pulses.

To implement a single-pulse analog of the SPE signal in the SPE method, the spin system is excited by a more complex RF pulse, during which the direction of  $\mathbf{H}_{\text{eff}}$  changes sufficiently quickly at the action of a pair of MVP pulses under the condition  $\Delta \omega'_j \tau_{\text{m}} \ll 1$ , where  $\Delta \omega'_j = \sqrt{\Delta \omega_j^2 + \omega_1^2}$  and  $\tau_{\text{m}}$  is the duration of the MVP [10]. In our case, a sharp change in the direction of  $\mathbf{H}_{\text{eff}}$  within the RF pulse is achieved when the pair of MVP pulses is turned on near the trailing edge of a sufficiently long RF pulse. In this case, only one ME signal is observed, and other

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signals, which are formed by the combined action of the leading edge of the RF pulse and MVP pulses, do not interfere with it, Fig. 4.



Figure 4. Oscillogram of a single excitation by an RF pulse combined with two MVPs, generating a ME signal at the frequency of  $\nu = 213$  MHz, with an RF pulse duration of  $\tau_{\rm RF} = 60 \,\mu s$ , MVP durations of  $\tau_{\rm m} = 1 \,\mu s$ , T = 293 K. The lower trace is a wave meter signal showing the location of MVP pulses within the RF pulse.

Rapid changes in the direction of  $\mathbf{H}_{\text{eff}}$  in the RCS occur when a DW is displaced under the action of MVP pulses due to the anisotropy of the HFF and the inhomogeneity of the enhancement factor  $\eta$  in the DW.

Next, we study the properties of this analog of the SPE signal, for brevity called the magnetic echo (ME) signal, formed under the combined action of RF and a pair of MVP pulses, Fig. 5. Previously, in [24], the effect of the formation of an edge magnetic echo (EME) signal was studied under the combined action of an MVP and the trailing edge of a sufficiently long RF pulse. In this work, it was shown that the EME signal, an analog of the SPE signal that occurs in this case, is formed by the mechanism of distortion of the fronts of an effective RF pulse. It is formed jointly by an MVP pulse and the trailing edge of a sufficiently long RF pulse above a certain MVP threshold amplitude associated with the DW pinning force when the power of the RF pulse was sufficient to observe the normal SPE signal.

The impact of two MVP pulses makes it possible to control the degree of influence of the mechanism of distortion of both fronts of the effective RF pulse depending on the amplitude of the MVP. Fig.5 shows the dependence of the ME intensity on the amplitude of the MVP pulses.

When the MVP amplitudes exceed  $H_0$  for the ME observation, the ME signal is formed by the distortion mechanism and is observed under a single combined action of RF and a pair of MVPs. At the MVP amplitudes  $H < H_0$ , the ME signal is formed by a multipulse mechanism and is not observed at the application of a single combined RF and the pair of MVPs excitation, but only at repeated exposure to RF and a pair of MVPs with  $f_r$  frequency, Fig. 6a.

Next, we present the results of a comparative study of the transverse relaxation processes  $T_2$  of ordinary TPE and SPE signals with the transverse relaxation of the ME signal, Fig. 7. The figure 7 shows the amplitude dependence of the ME on the interval between two MVP pulses  $\tau$ , as well as the amplitude dependences of the SPE and TPE depending on the duration of the



Figure 5. Dependence of the intensity  $I_{\rm ME}$  of the magnetic echo (ME) on the amplitudes of the two similar magnetic video-pulses (MVPs).



Figure 6. Oscillogram of ME signal excitation by an RF pulse combined with two MVPs, at the NMR frequency  $\nu = 213$  MHz, with RF pulse duration  $\tau_{\rm RF} = 60 \,\mu \text{s}$ , MVP pulse durations  $\tau_{\rm m} = 1 \,\mu \text{s}$ , MVP amplitude H = 30 Oe, T = 293 K. (a) multiple excitations with a repetition frequency of RF and a pair of MVPs action  $f_r = 100$  Hz; (b) single excitation with RF and a pair of MVPs. The lower trace is a signal from wave meter showing the position of the MVPs within the RF pulse.

RF pulse  $\tau$  and the interval  $\tau_{12}$  between two TPE exciting RF pulses, respectively. As can be seen from Fig. 7, the SPE and ME signals are characterized by practically similar transverse relaxation times  $T_2^{\text{ME}} = T_2^{\text{SPE}} = 30 \,\mu\text{s}$ , which are somewhat shorter as compared to  $T_2$  for TPE, in accordance with [23,24].

Based on the obtained experimental results, it can be assumed that the ME signal is formed by the mechanism of distortion of the fronts of the effective RF pulse, formed by a pair of MVP pulses in combination with the RF pulse when the amplitudes of the MVP pulses exceed the threshold value of the pinning force  $H_0$ , measured by the effect of these MVP pulses on the intensity of the TPE as in [24]. It should be noted that as the amplitude of one of the MVPs decreases below  $H_0$ , the ME signal disappears. Thus, the leading role is played by the distortion  $Investigation \ of \ the \ single-pulse \ NMR \ echo \ origin \ in \ cobalt \ \dots$ 



Figure 7. The intensities I of decay envelops of ME (1), SPE (2), and TPE (3) signals depend on the duration of the effective RF pulse (curve 1) and RF pulse (curve 2)  $\tau$ , as well as the distance between RF pulses  $\tau_{12}$  (curve 3), respectively.

of the fronts of the effective RF pulse under the action of a pair of MVP pulses, which is caused by the sufficiently fast displacements of the DWs, due to the anisotropy of the HFF and the inhomogeneity of the RF field enhancement factor  $\eta$ .

The experimental dependences of the ME signal can be qualitatively understood within the framework of a simple model similar to that used in [24]. Let us assume that under the combined action of RF and a first MVP pulse, a DW reversibly shifts at a distance  $\Delta x$  proportional to the MVP amplitude:  $\Delta x \sim v \tau_d = S(H - H_0) \tau_m$ , when the MVP amplitude H exceeds the DW pinning force  $H_0$ .

Under the combined action of RF and MVP, the magnitude and direction of the effective magnetic field  $\mathbf{H}_{\text{eff}}$  at the nuclei of the  $\Delta x$  layer in the RSC changes abruptly due to the corresponding changes in the HFF and the enhancement factor  $\eta$ . According to the non-resonant model of the formation of the SPE [10], the impact of the first MVP is equivalent to the effect of the leading edge of the RF pulse during the formation of the SPE. In this case, the role of the second front of the RF pulse, rephasing of the transverse magnetization arising as a result of the action of the first MVP, is played by the second MVP. In this case, the ME amplitude will be proportional to the number of nuclei in the layer  $\Delta x$  formed when the DW is displaced by the first MVP:  $I_{\text{ME}} \sim \Delta x/L$ , where L is the width of the excited section of the DW under the influence of an RF pulse, while the jump-like change in the NMR frequency in the RCS  $\Delta w'_j = \sqrt{\Delta w'_j + w_1^2}$  must satisfy the condition  $\Delta w'_j \tau_{\text{m}} \ll 1$  at which the precession period  $T' = 2\pi/\Delta w'_j$  of nuclei in RSC must be larger than MVP duration  $\tau_{\text{m}}$ :  $T' \gg \tau_{\text{m}}$ .

As is known, the NMR spin echo phenomenon can be used to store and manipulate large amounts of information [25–28]. Examples are functional electronic devices, spin processors, and the development of quantum computers. Spin processors based on NMR spin echo can also be used for analog processing of RF pulses in order to increase the processing speed of wideband signals in real-time, which is difficult to achieve using traditional digital process engineering methods. An echo processor using the NMR spin-echo phenomenon in magnetic materials can play the role of such an analog processor. The importance of using a magnetic material is that no external magnetic field is required, and there is significant amplification of the spin echo signal from the RF amplification effect in magnetic materials. The SPE method can provide practically the same information about the spin system as the two-pulse echo method. The properties of SPE and the conditions for its observation must be known to account for the spin processor operation based on NMR single-pulse echo phenomenon. In addition, with sufficient signal strength, it is possible to develop a device for processing RF pulses based on the phenomenon of SPE. The advantage of such a system is that it is not necessary to read the pulses in an RF sequence and that the complexities associated with the need to synchronize the write and read pulses are eliminated.

#### 3. Conclusion

A comparative study of the formation mechanisms of a single-pulse echo and a magnetic echo formed upon joint excitation by an RF pulse and a pair of magnetic video-pulses has been carried out. The magnetic echo is an analog of the single-pulse echo signal generated by the mechanism of distortion of the effective RF pulse edges during the domain wall displacement when the amplitudes of magnetic video-pulses exceed the domain wall pinning force. The obtained results testify in favor of the assumption that the single-pulse echo signal in cobalt is formed due to the mechanism of distortion of the fronts of RF pulses due to the displacements of domain walls leading to distortion of the local field on the nuclei due to the anisotropy of the HFF and the inhomogeneity of the enhancement factor  $\eta$  in the domain walls for sufficiently large RF pulse powers causing the displacements of domain walls.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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