Method of identification of the NMR origin in magnetically ordered substances: Application to the $Mn_{1+\delta}Sb$ alloy system

I. G. Kiliptari
Department of Physics, Tbilisi State University, Tbilisi 380028, Republic of Georgia
(Received 18 April 1995)

Exact computer calculations of the two-pulse echo intensity as a function of rf excitation level in magnetic materials have been carried out by taking into account the effect of variation of NMR frequencies and enhancement factors of the rf field. It is established that the profile of the spin echo amplitude curve depends upon the location of nuclear spins within domains or domain walls of a multidomain ferromagnet. The numerical results are supported by systematic experimental investigation performed in the series of $Mn_{1+\delta}Sb$ alloys by the use of spin echo techniques both in zero and applied magnetic fields up to 8 kOe. Ultimate NMR enhancement at 77 K due to the increasing contribution from the domain nuclei to the echo amplitude was found in the concentration range $0.22 \lesssim \delta \lesssim 0.26$. The obtained data may be used to study both the magnetic structure of the domain walls and structure-sensitive properties of a crystal.

I. INTRODUCTION

In pulsed spectroscopy of magnetically ordered substances a question of general interest is whether a NMR signal from a multidomain ferromagnet arises from domains or domain walls. This problem is interesting since echo formation of either type is characterized by a number of specific features which influence the shape and width of resonance spectra. For example, in ferromagnetic materials with axially symmetric hyperfine (hf) interaction the domain-wall NMR spectrum shows a double-peaked structure which is attributed to nuclei located at the center and at the edge of domain walls; at the same time the use of domain resonance is generally preferable for resolving the NMR line structure due to the anisotropic magnetic and electric hf interactions.[1–3]

At present, depending on a problem under consideration, domain or domain-wall resonances are used to study quadrupolar effects in the nonequidistant system of energy levels,[3,4] magnetic perturbation in the vicinity of a solute atom,[5] anisotropic contributions to the hf field and their relationship to the magnetocrystalline anisotropy,[6–8] the structure and dynamical characteristics of the domain walls,[9,10] and so on. It is evident that the efficiency of such investigations is determined by the possibility of the overall separation of the corresponding signals.

On the other hand, in some cases it becomes essential to maximize the echo intensity by means of superposition of the domain and domain-wall signals. The main point of this problem is that in a multidomain ferromagnet the zero field NMR signals appear to arise from nuclei in domain walls which occupy only a small portion ($\sim 10^{-3}$) of the whole volume of the crystal; therefore, a major fraction of the nuclear spins does not contribute to the NMR signal.[11] Hence the problem reduces to one of establishing optimum operating conditions which would increase the relative contribution of domain nuclei to the resultant echo intensity. For this purpose, Pogorelii and Kotov[12] have suggested using temperature dependence of the uniaxial magnetic anisotropy. However, the temperature region which yields the largest spin-echo amplitude in hexagonal cobalt amounts to 540–550 K severely limiting the possibility of the practical application of the observed effect.

The origin of the spin-echo signal is generally governed by several factors which may be classified into two groups. The first group considers structure-sensitive characteristics of a magnet such as the value of the magnetocrystalline anisotropy, mobility, and thickness of domain walls, etc., while the second group allows for the excitation conditions involving the presence of the external magnetic field, the amplitude of the driving radio frequency (rf) pulses, and so on.[13–15] Most of the existing methods used to indicate the origin of the NMR signals deal with the second group. These methods utilize the effect of external magnetic field on the spin-echo intensity[7,8,14] and the analysis of the NMR spectra shape at various rf levels.[4,15] Recently[16] we have reported that similar information may be provided by measuring the echo amplitude as a function of the delay time between the two pulses due to the difference in relaxation mechanisms for the domain and domain-wall nuclei.

In the present paper, we report on an approach to the identification of the NMR origin in magnets by studying the variation of two-pulse echo intensity with applied rf field. The possibilities of the proposed method are demonstrated on an $Mn_{1+\delta}Sb$ alloy system which allows one to vary the contribution of both the domain and domain-wall nuclei to the overall echo intensity.

II. DOMAIN AND DOMAIN-WALL RESONANCES IN MULTIDOMAIN FERROMAGNETS

A. Enhancement factor distribution functions

Most features of NMR in magnetically ordered substances are closely related to the existence of tremendous

internal fields at the nuclei, H_n, due to the strong hf interaction of the electronic-nuclear spin system. As a result, because of the coherent rotation of the electronic magnetization in the exciting rf field, h, the resonance transitions of nuclear spins are induced not directly, but via the transverse component of the local field, ΔH_n, enhanced by a factor η, so that $\eta=\Delta H_n/h$.[11]

For a monodomain ferromagnet magnetically aligned along an axis of easy magnetization the enhancement factor of the rf field is given by[14]

$$\eta_D=\frac{H_n}{H_0+H_A}\ , \tag{1}$$

where H_0 and H_A are the external magnetic and static anisotropy fields, respectively.

Actually, Eq. (1) applies to an averaged value of the domain enhancement $\bar{\eta}_D$. In the real ferromagnet, due to the various kinds of structural inhomogeneities (like internal strains, imperfections, dislocations, etc.) the magnitude, orientation, and dynamical characteristics of the electronic moments are randomized over different sites of the crystal lattice giving rise to a spread in the values of most of the characteristic parameters of NMR.[11] This can be attributed first to the longitudinal component of the hf field which leads to the variation in the NMR frequencies, ω_n, about the center frequency, ω_0. This kind of broadening is usually introduced by means of the inhomogeneous line-shape function which is assumed to be the Gaussian, of the form

$$g(\Delta)=\left[\frac{\ln 2}{\pi}\right]^{1/2}\sigma^{-1}\exp[-\ln 2(\Delta/\sigma)^2]\ , \tag{2}$$

where $\Delta=\omega_n-\omega_0$ and σ is the NMR line half-width.

Another important effect due to inhomogeneity of the magnetic susceptibility is a random variation of transversal components of the hf field seen by the nuclei which can be represented as a spread in values of enhancement factors. This effect is commonly described in terms of the enhancement factor distribution function which may be expressed as

$$F_D(\eta)=\left[\frac{\ln 2}{\pi}\right]^{1/2}\Delta\eta_D^{-1}\exp\left\{-\ln 2\frac{(\eta-\bar{\eta}_D)^2}{\Delta\eta_D^2}\right\}\ , \tag{3}$$

$\Delta\eta_D$ being the width of the $F_D(\eta)$ function, which is believed to be a measure of structural inhomogeneities in the crystal. As an example, supposing that $\Delta\eta_D\ll\bar{\eta}_D$, we obtain an almost unique distribution of enhancement factors typical for the domain nuclei in high purity magnetic materials.

An entirely different source of inhomogeneity is associated with the domain walls where the gradients of magnetization are of the greater magnitude. In this case, the primary mechanism of enhancement is due to the wall motion in the rf field which leads to the positional dependence of η values along the normal to the plane of the wall (x axis). Particularly, as was shown by Stearns,[17] for 180° Bloch walls, which typically occur in the easy-axis crystals, the corresponding dependence is of the form

$$\eta=\eta_W\,\mathrm{sech}(x/d)\ , \tag{4}$$

where d is the wall thickness and η_W is the maximum enhancement at the center of the wall.

Because of the large angles of rotation of the electronic moments due to the wall displacement in the rf field, average enhancement factors in the domain walls are expected to be much greater than in the domains. However, as will be discussed in Sec. IV, in appropriate experimental conditions the contribution of the domain resonance may sharply increase and become comparable with that of the domain walls.

In addition, as far as treating resonance phenomena in magnets one must take into account that the domain walls are usually trapped by the above mentioned imperfections and impurities giving rise to inhomogeneous displacement and damping of the wall motion.[18] This results in distribution in the wall mobilities which may be considered as a spread in values of the maximum enhancement factor at the center of the wall η_W.[19] (According to Refs. 9 and 18, neither a difference in masses nor a variation in the natural frequencies of the domain walls would contribute so appreciably to the inhomogeneity of η_W values.)

Since in this special case the main source inhomogeneity is of the same origin as for the domain nuclei, the wall distribution function over maximum enhancement factors may be given as

$$F(\eta_W)=\left[\frac{\ln 2}{\pi}\right]^{1/2}\Delta\eta_W^{-1}\exp\left\{-\ln 2\frac{(\eta_W-\bar{\eta}_W)^2}{\Delta\eta_W^2}\right\}\ , \tag{5}$$

where $\bar{\eta}_W$ is the value of enhancement at the center of the wall averaged in the region of the sample and $\Delta\eta_W$ is the width of the corresponding distribution.

Following Ref. 19, let us introduce the domain-wall joint distribution function which allows for both foregoing sources of inhomogeneity and, apart from unimportant coefficients, can be written as

$$F_W(\eta)=\eta^{-1}\int_0^{\infty}\exp\left\{-\ln 2\frac{[(\varepsilon^2+\eta^2)^{1/2}-\bar{\eta}_W]^2}{\Delta\eta_W^2}\right\}d\varepsilon\ , \tag{6}$$

with $\varepsilon=(\eta_W^2-\eta^2)^{1/2}$. Throughout the present study, the distribution of enhancements given by Eqs. (3) and (6) is assumed to be independent of the rf power level and the relaxation effects are neglected.

Thus, for the required response of macroscopic magnetization to the action of resonant pulses we have[16,19]

$$I(h,t)=\int_0^{\infty}\eta F_{D,W}(\eta)d\eta\int_{-\infty}^{+\infty}\mu^{+}(\eta,\Delta,h,t)g(\Delta)d\Delta\ , \tag{7}$$

where $\mu^{+}=\mu_x+i\mu_y$ represents the contribution of a single spin packet to the total magnetization which for the two-pulse echo produced by pair of rectangular pulses of equal length, τ_p, and amplitude, h, is given by[20]

$$\mu^+/\mu_0=\frac{\chi^3}{\Phi^3}\sin^2\frac{\Phi\tau_p}{2}\left[\frac{2\Delta}{\Phi}\sin^2\frac{\Phi\tau_p}{2}-i\sin\Phi\tau_p\right]\exp[i\Delta(\tau-t)]\ . \quad (8)$$

Here $\Phi=(\chi^2+\Delta^2)^{1/2}$, μ_0 is the equilibrium value of magnetization, $\chi=\gamma h\eta$ the pulse amplitude expressed in frequency units, γ the gyromagnetic ratio of the resonating nuclei, τ the spacing between the pulses, and t the time following the trailing edge of the second pulse.

B. Numerical results

The numerical calculations of Eq. (7) were performed according to the procedure described previously.[19] Since the position of a single-peaked echo near time $t=\tau$ is slightly dependent upon the rf strength, we initially computed the echo wave forms for about 30 different values of h, other parameters being fixed, and then plotted the $I(h)$ dependence for the maximum intensity of the signal.

The results of the calculations for the domain resonance intensity as a function of rf pulse amplitude are shown in Fig. 1. As one can see from Fig. 1(a), in the case of almost unique distribution of the rf fields ($\Delta\eta_D/\overline{\eta}_D \lesssim 0.05$) the echo amplitude displays damped oscillations. However, even a small spread in η values smears out completely the oscillations in the $I(h)$ curve. Thus, as the rf power is gradually increased the echo intensity rises rapidly, goes through a maximum at a certain value of $h=h_m^D$, and then decreases monotonically,

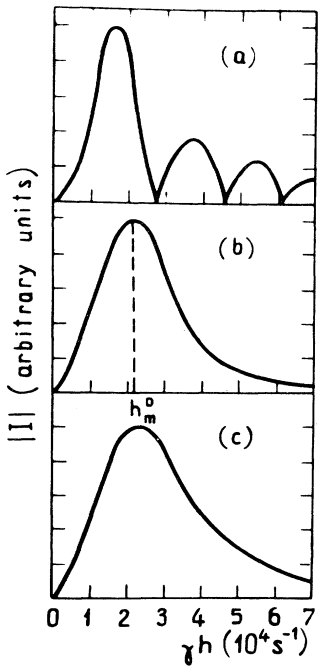

FIG. 1. Computer plots of the domain spin-echo intensity as a function of γh obtained by numerical integration of Eq. (7) for $F_D(\eta)$ of Eq. (3) for the following values of $\Delta\eta_D/\overline{\eta}_D$: (a) 0.03; (b) 0.25, and (c) 1.00. Parameters used in the calculations are $\overline{\eta}_D=500$, $\tau_p=0.1$ μs, and $\sigma=6\times10^6$ s^{-1}.

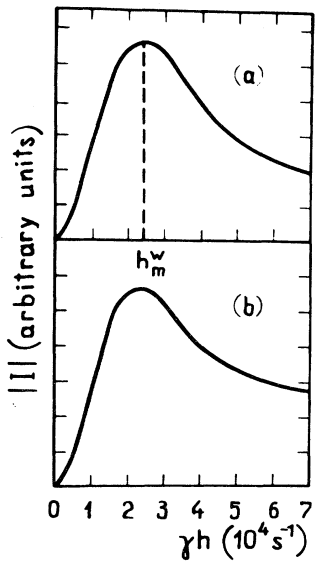

FIG. 2. Computer plots of the domain-wall spin-echo intensity as a function of γh obtained by numerical integration of Eq. (7) for $F_W(\eta)$ of Eq. (6) for the following values of $\Delta\eta_W/\overline{\eta}_W$: (a) 0.625 and (b) 1.00. Parameters used in the calculations are $\overline{\eta}_W=500$, $\tau_p=0.1$ μs, and $\sigma=6\times10^6$ s^{-1}.

as illustrated in Figs. 1(b) and 1(c).

The domain-wall nuclei, exhibit similar behavior as shown in Fig. 2; however, for all reasonable values of parameters used in our calculations the domain-wall $I(h)$ curves beyond the maximum intensity, for $h>h_m^W$, fall much more slowly compared to the corresponding domain curves. In other words, the $I(h)$ curves for the domain nuclei are more symmetrically shaped about the center at $h=h_m$ than those of the domain-wall nuclei. For this reason, the general shape of $I(h)$ may be used to indicate the origin of the NMR signal.

As to the optimum turning angles required to maximize the echo amplitude, we note that they show no significant difference for the domain and domain-wall resonances and for relatively short pulses ($\tau_p^{-1}\gtrsim\sigma$) are equal to

$$\alpha_m^{D,W}=\gamma h_m^{D,W}\overline{\eta}_{D,W}\tau_p=1.2\pm0.1 \text{ rad} . \quad (9)$$

Equation (9) shows that in order to maximize the intensity of both of the signals at a given value of h_m the difference in enhancement factors for the domain and domain-wall nuclei should be as small as possible.

III. EXPERIMENTAL DETAILS AND OBTAINED RESULTS

A. Sample preparation

Experimental investigation has been carried out on the easy-axis $Mn_{1+\delta}Sb$ ferromagnetic compounds, which crystallize in the hexagonal NiAs-type structure. On the whole about ten samples with excess Mn content δ ranging from 0.049 to 0.325 have been prepared. After grind-

TABLE I. Values for various parameters of $Mn_{1+\delta}Sb$ at 77 K. The hf field, H_n, at manganese nucleus is virtually independent of the cation concentration and is equal to 242.0 kOe.

Composition δ	$\lvert K_1\rvert$ (10^6 erg/cm^2)	M_s (Oe)	H_A (kOe)	$\bar{\eta}_W \times 10^3$ at $H_0=0$	$\bar{\eta}_D$ at $H_0=0$	$\bar{\eta}_D$ at $H_0=3.5$ kOe
0.049	4.00[a]	720[a]	11.1[b]	4.0[c]	~22[b]	~17[b]
0.128	1.75[a]	630[b]	5.6[b]	3.2[c]	~43[b]	~27[b]
0.245	<0.04[a]	492[b]	<0.16[b] 0.12[c]	2.7[c]	2000[c]	67[c]

[a]Measured values in Ref. 21.
[b]Estimated values from the results reported in Refs. 21 and 22.
[c]Values of parameters obtained in this work to an accuracy of about 10%.

ing, the appropriate amounts of Mn (purity 99.9%) and Sb (purity 99.999%) were sealed in evacuated quartz tubes, heated for about 4 h at 1000 °C and then slowly cooled to room temperature (15 °C/h) being kept for 1 h at 900 and 800 °C. After that the samples were reground to fine particles from 1 to 80 μm in diameter, annealed at 550 °C for 725 h, and then rapidly quenched in water. Both chemical and x-ray-diffraction analyses were performed before measurement, which defined the exact composition of the alloys and identified the existence of a single phase in the range of concentrations studied.

The choice of the system under investigation is motivated by a strong dependence of its magnetoanisotropic properties on the interstitial Mn content. That is, according to magnetic measurements,[21,22] upon deviating from stoichiometry to the high Mn side the saturation magnetization M_s and the absolute value of the magnetocrystalline anisotropy constant K_1 sharply decrease, so that the latter at 77 K in the vicinity of $\delta \approx 0.24$ goes through zero. Numerical values of the corresponding parameters for several of the samples studied, together with the anisotropy field calculated by the formula $H_A = 2|K_1|/M_s$ (Ref. 11) are listed in Table I. Because of the significant difference in magnetic anisotropy the domain enhancement factors for alloys with different Mn content are expected to vary by several orders of magnitude thereby changing the contribution of the domain nuclei to the overall echo intensity.

B. Experimental apparatus and excitation conditions

All the experiments presented were performed at 77 K using a phase-incoherent NMR spectrometer which was essentially the same as described by other investigators (see, for example, Ref. 13). The value of the rf level at the sample was estimated in the usual way by measuring the induced voltage in a single turn coil fitting closely around the cylindrical powdered samples.[14,18] The bandwidth of the detector (~4 MHz) was wide enough to respond adequately to the excited range of frequencies even at the high power level. A maximum field at the sample produced by the rf pulses was about 2.9 Oe.

The $I(h)$ dependence was measured for signals of single-peaked form generated by two relatively short pulses ($\tau_p \lesssim 1$ μs). Since the echo intensity and shape are extremely sensitive to the relative turning angles of the two pulses,[23,34] at each power level the equality of rf amplitudes and widths were carefully monitored. The measurements in a static magnetic field were performed with the aid of a subsidiary circuit connected to the rf oscillator through a coaxial cable.

It is well known that in order to sweep out the domain walls the external magnetic field should be larger than the demagnetizing field which for spherical particles is of the form $H_D = 4/3\pi M_s$.[14] Using the numerical values listed in Table I we find that for a $Mn_{1.049}Sb$ alloy having maximum saturation magnetization the removal of the walls occurs at a field of about 3.0 kOe; therefore, above this value the NMR signals in all samples are expected to originate from the domain nuclei.

C. Spin-echo amplitude curves in $Mn_{1+\delta}Sb$

Figure 3 represents some typical experimental plots of the echo intensity as a function of the rf excitation level. The $I(h)$ dependence in zero applied field for most of the samples studied is similar to that shown in Fig. 3(a). Specific features, however, exhibit alloys synthesized in the narrow concentration range $0.22 \lesssim \delta \lesssim 0.26$; namely,

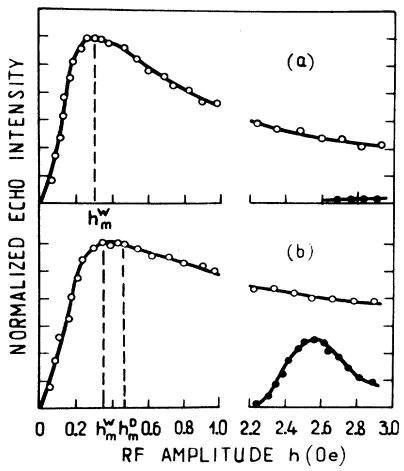

FIG. 3. The normalized intensity of the ^{55}Mn NMR in $Mn_{1+\delta}Sb$ versus applied rf field for the samples with δ (a) 0.128 and (b) 0.245. Resonance frequency 255.5 MHz; temperature $T=77$ K. White circles: $\tau_p=0.2$ μs, $H_0=0$. Solid circles: $\tau_p=1.0$ μs, $H_0=3.5$ kOe. Solid lines in Figs. 3 and 4 are merely to guide the eye.

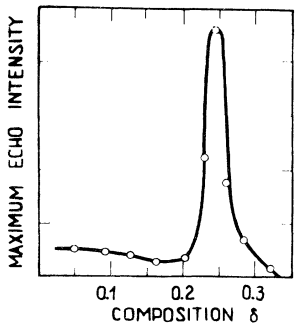

FIG. 4. Variation of the ^{55}Mn spin-echo intensity in $Mn_{1+\delta}Sb$ as a function of excess Mn concentration δ in zero applied field. For each sample, both pulse durations and amplitudes have been carefully adjusted in order to maximize the echo intensity.

for these alloys the zero field $I(h)$ curves show a broad maximum followed by a more gradual decrease in echo intensity for fields $h > h_m$, as seen in Fig. 3(b).

A further prominent feature of the system under consideration is a strong compositional dependence of the largest spin-echo amplitude produced under optimum excitation conditions of the unhomogeneously broadened NMR line. Figure 4 shows that with increasing Mn content the echo intensity somewhat decreases, while the maximum of the echo amplitude curve shifts to higher h values; next, as δ varies from 0.22 to 0.26, the signal intensity rises sharply, reaches a maximum, and finally again decreases in magnitude. Consequently, the echo amplitude for a $Mn_{1.245}Sb$ sample is about one order of magnitude larger than for the nearly stoichiometric alloy with $\delta = 0.049$.

Attempts to study the $I(h)$ dependence in the saturation region, i.e., when $H_0 > 3.0$ kOe, for most of the samples of the $Mn_{1+\delta}Sb$ system ended in failure. In the best case, at the largest available rf field strength, only hardly visible echoes buried in the noise level could be detected, as seen from Fig. 3(a). Again, unusual characteristics are exhibited by alloys with Mn composition ranging from 0.22 to 0.26. For these samples, the NMR signals are clearly observable over a wide range of driving rf fields, whereas the $I(h)$ curve at $H_0 = 3.5$ kOe has a much more symmetrical form than that in zero applied field [see Fig. 3(b)]. As H_0 is further increased, the spin-echo intensity curve for the alloy with $\delta = 0.245$ smoothly shifts towards larger magnitudes of h and at $H_0 \gtrsim 4.0$ kOe the maximum of echo intensity moves away from the range of rf amplitudes generated by our spectrometer.

IV. DISCUSSION AND CONCLUSIONS

The results of the present paper suggest that the profile of the spin-echo amplitude curve provides information of the origin of the NMR signals in multidomain ferromagnets. Indeed, oscillatory behavior of the echo intensity as a function of the rf level decisively indicates the domain origin of a signal. However, such a behavior is believed to characterize resonance phenomena in the case of almost unique distribution of the rf fields like the domain resonance in good-quality single crystals.[25] For most polycrystalline bulk crystals and thin ferromagnetic films having inhomogeneously distributed enhancement factors the echo intensities show a smooth dependence upon h with more or less pronounced maximum at optimum turning angles.[18,19] It should be emphasized that the symmetry properties of the $I(h)$ curves for the domain and domain-wall resonances significantly differ from each other providing a basis for identification of the echo origin.

Indeed, the analysis of the experimental data in the series of $Mn_{1+\delta}Sb$ alloys shows that for Mn concentration varying over the range $0.049 < \delta < 0.22$ and $\delta < 0.26$ the zero field NMR signals predominantly originate in the domain walls. However, the possibility of observing the domain resonance in this set of alloys is somewhat restricted. This arises from the fact that in highly anisotropic materials with great saturation magnetization, the removal of the domain walls takes place under the influence of sufficiently strong magnetic fields, which sharply decrease the echo intensity and shift its maximum to relatively high values of h. Particularly, the domain enhancements $\bar{\eta}_D$ in these alloys estimated from Eq. (1) are found to be so small that the maximum of echo amplitude is expected to occur for $h > 5.5$ Oe, which is not available with our equipment. Another effect which also reduces the echo intensity is the spread in frequencies of the domain nuclei due to the variation of the demagnetizing fields from different-shaped particles.[18] Thus we conclude that such materials are of particular interest in a view of restoring the form of the domain-wall distribution function $F_W(\eta)$ according to the method proposed recently,[19] since in this case over a wide range of applied rf fields the effect of domain resonance on the observable $I(h)$ dependence can generally be neglected.

An essentially different situation holds for weak ferromagnets with small values of magnetic anisotropy such as the present samples in the range $0.22 \lesssim \delta \lesssim 0.26$. Here, sufficiently strong nuclear signals are detectable even in the saturated state, while the shape of the $I(h)$ curves indicates the domain origin of signals. This point is further justified by an almost exponential decrease of the echo intensity as a function of time interval between the pulses in contrast to a nonexponential decay taking place for the domain-wall resonance.[16] The values of enhancement factors within the domains and at the center of domain walls obtained from Eq. (9) for several of the samples under investigation are listed in Table I.

In zero applied field for the samples with δ ranging from 0.22 to 0.26 the shape of $I(h)$ is more complicated, and cannot be attributed to either of the sites of magnetically ordered crystal. To clarify the subject let us return for a moment to the experimental data obtained for a $Mn_{1.245}Sb$ alloy under the magnetic field of 3.5 kOe. First, using the experimental value of h_m^D from Eqs. (1) and (9) we find that the anisotropy field, H_A, in this sample is 120 ± 15 Oe, which is within the possible range of values calculated from magnetic measurements.[21] Then using the same equations we estimate the domain reso-

nance enhancement factor, $\bar{\eta}_D$, and optimum value of rf amplitude, h_m^D, in zero applied field which differ from the corresponding values measured for the domain-wall nuclei, $\bar{\eta}_W$ and h_m^W, by no more than a factor 1.5 [see Fig. 3(b) and the data of Table I]. Thus, in this alloy both of the signals are contributing to the resultant echo intensity giving rise to a broad maximum in the $I(h)$ curve. It should be noted here that the NMR frequencies for the domain and domain-wall resonances within the accuracy of ± 100 KHz coincide with each other which is characteristic of a crystal with small anisotropy in the hf fields.

Further clear evidence of the mixture of signals coming from the domains and domain walls is the marked increase in echo intensity for a $Mn_{1.245}Sb$ alloy in comparison to other alloys of the system studied. As is well known,[12,14] the spin-echo intensity is proportional to the enhancement factors of the resonating nuclei as well as to the number of spins contributing to the resonance signal. As δ is raised from 0.049 to 0.24 the maximum enhancement factors for the domain-wall nuclei show a tendency to decrease due to greater damping of the domain-wall motion,[18] while the domain enhancements monotonically increase because of the compositional dependence of the magnetic anisotropy.[21] Consequently, with increasing Mn content the difference in enhancement factors and, thus, in the values of h_m for the domain and domain-wall nuclei gradually decreases. However, the sharp rise in echo amplitude at 77 K is observed only for alloys synthesized in the narrow concentration range $0.22 \lesssim \delta \lesssim 0.26$ when most of the nuclei in the sample are contributing to the NMR signal. We believe that by more careful adjustment of the manganese content in the vicinity of $\delta = 0.24$ one should obtain still more prominent enhancement of the echo intensity.

It is of special interest that an ultimate enhancement of the NMR signal can be, in principle, obtained in any temperature region. However, the concentration of excess Mn atoms required to maximize the echo intensity should be chosen by taking into account the temperature dependence of the magnetic anisotropy. For example, at 293 K the largest spin-echo amplitude was found to occur for the sample with $\delta \approx 0.1$ which, in agreement with magnetic measurements,[21] corresponds to the minimum of the magnetocrystalline energy at room temperature.

Finally, it is worth noting that the domain resonance studies may provide a possibility to avoid some principle difficulties which arise in analysis of $I(h)$ for the domain-wall nuclei. Indeed, as was outlined in Sec. II A, the explicit form of a joint distribution function $F_W(\eta)$ is determined, in general, by two different functions (4) and (5) depending upon the magnetic structure of the domain walls and structure-sensitive properties of a sample, respectively. Therefore, even having reliable information of the form of $F_W(\eta)$ function from experimentally observed $I(h)$ dependence,[19] it seems impossible to separate the contributions of these factors if other supporting data are not available. Our analysis shows that such data may be, in principle, obtained from the domain resonance $I(h)$ dependence, since the form of the latter is mainly governed by the spatial distribution of structural imperfections over the volume of the sample.

ACKNOWLEDGMENTS

The author gratefully acknowledges M. I. Kurkin and L. L. Buishvili for their constant interest in this work and many helpful discussions.

[1] A. C. Gossard, V. Jaccarino, and J. P. Remeika, J. Appl. Phys. **33**, 1187 (1962).
[2] M. A. Butler, Int. J. Magn. **4**, 131 (1973).
[3] C. H. Cobb, V. Jaccarino, and M. A. Butler, Phys. Rev. B **7**, 307 (1973).
[4] A. A. Shemyakov and V. A. Klochan, Zh. Eksp. Teor. Fiz. **101**, 1014 (1992) [Sov. Phys. JETP **74**, 544 (1992)].
[5] T. M. Shavishvili and I. G. Kiliptari, Phys. Status Solidi B **92**, 39 (1979).
[6] M. Kawakami, T. Hihara, Y. Koy, and T. Wakiyama, J. Phys. Soc. Jpn. **33**, 1591 (1972).
[7] C. W. Searle, H. P. Kunkel, S. Kupca, and I. Maartense, Phys. Rev. B **15**, 3305 (1977).
[8] H. P. Kunkel and C. W. Searle, Phys. Rev. B **23**, 65 (1981).
[9] A. Hirai, J. A. Eaton, and S. W. Searle, Phys. Rev. B **3**, 68 (1971).
[10] A. V. Zalessky and I. S. Zheludev, At. Energy Rev. **1**, 133 (1976).
[11] M. I. Kurkin and E. A. Turov, *NMR in Magnetically Ordered Substances: Theory and Applications* (Nauka Press, Moscow, 1990) (in Russian).
[12] A. N. Pogorelii and V. V. Kotov, Zh. Eksp. Teor. Fiz. Lett. **13**, 561 (1971) [JETP Lett. **13**, 400 (1970)].
[13] R. L. Streever, Phys. Rev. **134A**, 1612 (1964).
[14] R. W. N. Kinnear, S. J. Campbell, D. H. Chaplin, and G. V. H. Wilson, Phys. Status Solidi A **58**, 507 (1980).
[15] V. S. Pokatilov, S. V. Kapelnitskii, and V. N. Karazeyev, Fiz. Tverd. Tela (Leningrad) **32**, 1982 (1990) [Sov. Phys. Solid State **32**, 1155 (1990)].
[16] I. G. Kiliptari, Fiz. Tverd Tela (St. Petersburg) **35**, 1232 (1993) [Sov. Phys. Solid State **35**, 627 (1993)].
[17] M. B. Stearns, Phys. Rev. **162**, 496 (1967).
[18] M. B. Stearns and J. F. Ullrich, Phys. Rev. B **4**, 3825 (1971).
[19] I. G. Kiliptari and M. I. Kurkin, Fiz. Met. Mettaloved. **8**, 81 (1992) [Sov. Phys. Met. Metallogr. **74**, 136 (1992)].
[20] A. L. Bloom, Phys. Rev. **98**, 1105 (1955).
[21] T. Okita and Y. Makino, J. Phys. Soc. Jpn. **25**, 120 (1968).
[22] Y. Yamaguchi and H. Watanabe, J. Phys. Soc. Jpn. **48**, 435 (1980).
[23] W. B. Mims, Phys. Rev. **141**, 499 (1966).
[24] I. G. Kiliptari, Fiz. Tverd. Tela (St. Petersburg) **35**, 496 (1993) [Sov. Phys. Solid State **35**, 257 (1993)].
[25] V. D. Doroshev, V. A. Klochan, N. M. Kovtun, and V. N. Seleznev, Phys. Status Solidi A **9**, 679 (1972).