

Multiple nuclear spin echoes in the $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy

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The formation of multiple nuclear spin echo has been investigated in ordered $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy on ^{55}Mn nuclei. Fractional echo signals were first observed for metallic ferromagnetics. The appearance thereof has been explained in terms of high ordering degree and regular distribution of electric field gradients in such kind of intermetallic compounds.

Исследовано формирование многократного ядерного спинового эха в упорядоченном сплаве $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ на ядрах Mn^{55}. Впервые наблюдались дробные эхо-сигналы в металлических ферромагнетиках. Их появление связано с высокой степенью упорядочивания и однородностью распределения градиентов электрического поля в таких интерметаллических соединениях.

The presence of nuclear spin echo on ^{55}Mn in nearly stoichiometric Ni_2MnGa alloys [1–3] at room temperature and above is nontrivial by itself. It is possible only due to a high degree of orbital localization [1]. Here, we report the study of multiple spin echoes arising as a result of multi-quantum transitions between ^{55}Mn Zeeman levels split due to quadrupole interaction.

The $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy was studied using nuclear spin echo technique on ^{55}Mn and ^{70}Ga nuclei at 2.2 K. The powdered alloy for NMR experiments was prepared from single crystal by milling. Then the powder (about 200 μm grain size) was quenched from 1200 K down to room temperature. Finally, it was annealed at 1070 K and homogenized at 720 K for 90 hours. Measurements of martensite transformation and Curie temperature performed by low-field ac magnetic susceptibility technique, the saturation magnetization as well as X-ray diffraction experiments have shown that the powder parameters are identical to those for the bulk alloy.

It has been shown before [1–3] that the NMR spectrum (on ^{55}Mn nuclei) for ordered alloys of a nearly stoichiometric composition consists of a single line ($\omega/2\pi \sim$ 310 MHz) slightly broadened ($\Delta\omega/2\pi \sim$ 7–10 MHz) due to quadrupole interaction caused by deviation of the lattice symmetry from cubic one below T_m. A deviation in composition as well as disorder may result in the appearance of additional low-frequency broad satellite lines [2, 3].

The NMR spectrum of the ordered $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy is presented in Fig. 1 ($n = 1$). It also contains a low-frequency satellite line. The characteristic feature of nuclear spin echo in this compound is the possibility to observe additional echo signals arising at the time moments $t \neq \tau$ (here τ is the time interval between exciting rf pulses and $t = n\tau$, t is the time after the second pulse) and especially the fractional echo signals ($t = 1/2\tau$ and $3/2\tau$) (see Fig. 2). The spectra of these signals are also shown in Fig. 1 ($n \neq 1$). The parameters of the rf pulses correspond to the optimal excitation of echo signals at $n = 1/2$ and $3/2$.

The multi-quantum transitions $\Delta m > 1$ are possible for nuclei with spin $I > 1/2$ between the Zeeman levels $\hat{I}_z|m\rangle = m|m\rangle$, m being the magnetic quantum number. It has been

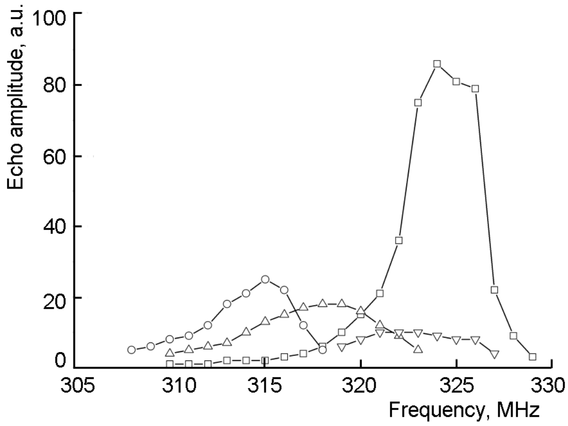

Fig. 1. ^{55}Mn NMR spectra in $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy at 4.2 K for "usual" nuclear spin echo $t = \tau$ (□) and additional multiple spin echoes $t = 1/2\tau$ (o), $t = 2\tau$ (Δ), and $t = 3\tau$ (∇). The intensities of the additional signals are substantially increased.

shown [4–6] that such transitions result in formation of multiple echo signals $t = n\tau$, where n is integer and $n \le 2I + 1$. To excite optimally the multi-quantum transition, it is necessary to use an increased power of the first exciting rf pulse as compared to the second one [7]. Quadrupole interaction in some cases can lead to the formation of fractional echo signals (n is non-integer) [8, 9]. These "fractional" echo signals can be caused both by single- ($\Delta m = 1$) and multi-quantum ($\Delta m > 1$) transitions. For instance, for $I = 5/2$ the echo signal arising in the time moment $t = 3/2\tau$ is caused by multi-quantum transitions, while the $1/2\tau$ signal, by single-quantum ones [8].

The $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy is the first metal magnetic compound where fractional echo signals have been observed. Before, this was possible only for diamagnetic crystals [8, 10] and magnetic semiconductor $CdCr_2Se_4$ [9]. It should be noted that the fractional echo signals were observed in the ordered $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy after its homogenizing annealing at 720 K for 90 h.

As mentioned above, the fractional signals have quadrupole origin, so as a result, their spectra include only quadrupole components. Besides, the following condition should be met for the possibility to observe the signals in experiment:

$$\omega_0 > \omega_q \ge \Delta\omega_q > \Delta\omega_0, \quad \Delta\omega_0 \to 0, \qquad (1)$$

where ω_0, ω_q, $\Delta\omega_0$, $\Delta\omega_q$ are the Zeeman and quadrupole interactions and their inhomogeneities, respectively. The spectrum of the $1/2\tau$ echo signal is entirely situated in the satellite region of the main signal ($n = 1$) spectrum. Hence, only the nuclei which contribute to the satellite line satisfy the condition (1). The alloy under study contains an excess of Ni ions and a deficit of Mn ones (in comparison with stoichiometry). As a result, some of the Mn positions should be occupied by nickel. First of all, this will cause a decreasing in the Zeeman fields on neighboring Mn nuclei, which results in the decreasing of NMR frequency. This can be a reasonable explanation of the satellite line appearance in the spectrum observed experimentally. Second, this should result in a wide distribution of local fields on gallium nuclei, which was also confirmed experimentally: a weak echo signal was observed on ^{70}Ga nuclei in a wide frequency range (45–100 MHz) [3]. Finally, the occupation of manganese positions by nickel should result in appearance of large electric field gradients on nearest manganese nuclei, which results in formation of fractional echo signals. It is to note that for disordered alloys, large electric field gradients should arise on ^{55}Mn nuclei. However, as is mentioned above, no fractional echo signals were observed for the disordered compound. This fact can be easily explained taking into consideration the inhomogeneity of hyperfine and quadrupole interaction in such material. For the disordered alloy the broad distribution of hyperfine and quadrupole interaction has to be expected due to the chaotic character of lattice and stacking defects. This results in the violation of condition (1) and impossibility to observe fractional echo signals.

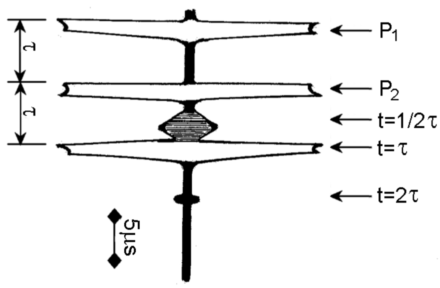

Fig. 2. Oscillogram of multiple echo signals in $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ alloy at 4.2 K. The intensities of exciting pulses and "usual" spin echo are substantially decreased.

As is mentioned above, the replacement of Mn ions for Ni ones should cause electric field gradients on neighboring Mn sites. The direction of the gradients with respect to the martensitic axes (axes of magnetic anisotropy) depends on position of particular Mn neighbors. Such angular distribution of the gradients gives rise to the inhomogeneity of $\Delta\omega_q$ (1). In an ideal crystal, the substituting nickel ions should be homogeneously distributed through the crystal lattice, i.e. the crystal should have a modulation translation symmetry. The value of quadrupole interaction ω_q (1) is mainly defined by martensitic distortion.

Fractional echo signals appear only after the homogenizing annealing, i.e. when irregular distortions due to the defects practically disappear and the lattice perfection is close to maximum. This means that the fractional echo signals arise as a result of angle scattering of quadrupole interaction. For NMR spectrum, this corresponds to satellite line (Fig. 1).

The NMR spectra on Ga nuclei situated near Ni ions on Mn sites were observed both for homogeneous and inhomogeneous alloys. Unfortunately, due to the low signal/noise ratio, it is impossible to reveal the difference between them.

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Багатократне ядерне спінове відлуння у сплаві $Ni_{54.1}Mn_{20.7}Ga_{25.2}$

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Досліджено формування багатократного ядерного спінового відлуння в упорядкованому сплаві $Ni_{54.1}Mn_{20.7}Ga_{25.2}$ на ядрах Mn^{55}. Вперше спостерігалися дробові сигнали відлуння у металічних феромагнетиках. Їх поява пов'язана з високим ступенем впорядкування та однорідністю розподілу градієнтів електричного поля в таких інтерметалічних сполуках.