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Relaxation processes of magnetic moments in ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$

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Experiments on the Mössbauer effect on the ^{57}Fe nuclei in ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$ revealed the absence of the magnetic hyperfine structure of ^{57}Fe nuclei in the magnetically ordered temperature range. The phenomenon is explained by the increased frequency of relaxation of the iron ions' magnetic moments in ferrite. Investigation performed in external magnetic fields has shown that in ferrite there exist iron spin groups with different relaxation times.

Keywords: ferrite, magnetic moment, magnetization, Curie temperature, relaxation process, Mössbauer effect.

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The paper reports the results of studying the Mössbauer effect on ^{57}Fe nuclei in spinel-structure ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$ in the temperature range of 80–350 K. The study has revealed that the magnetic hyperfine structure (MHFS) of iron nuclei in the magnetically ordered temperature range decreases to zero: the iron nuclei MHFS decreases to zero at $T = 160$ K, while the ferrites̕ Curie temperature is $T_c = 320$ K.

Magnetization was measured with vibration magnetometer VM2-A (Fig. 1). The figure shows that the Curie temperature of ferrite is $T_c = 320$ K. Mössbauer spectra of the ^{57}Fe nuclei were recorded using spectrometer YaGRS-4M equipped with the ^{57}Co (Cr) source of γ radiation.

Fig. 2 presents the ferrite Mössbauer spectra measured at different temperatures. The spectra are similar in shape to relaxation spectra which are characterized by broadening of the MHFS lines, especially of the external ones [1]. At the temperature of 117 K, a quadrupole doublet (QD) whose intensity increases with increasing temperature manifests itself in the spectrum jointly with MHFS. At $T = 160$ K,

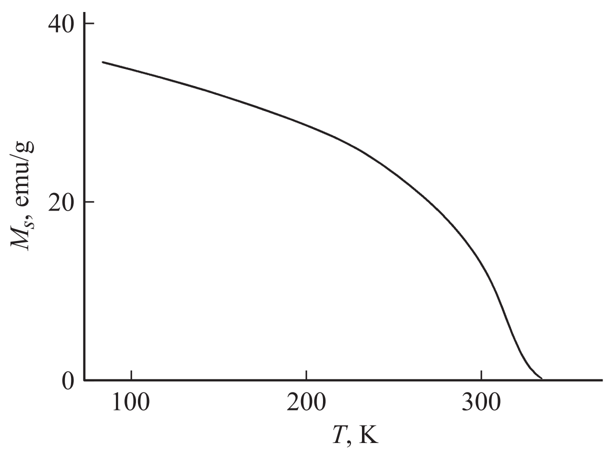

Figure 1. Saturation magnetization M_s of ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$ versus temperature.

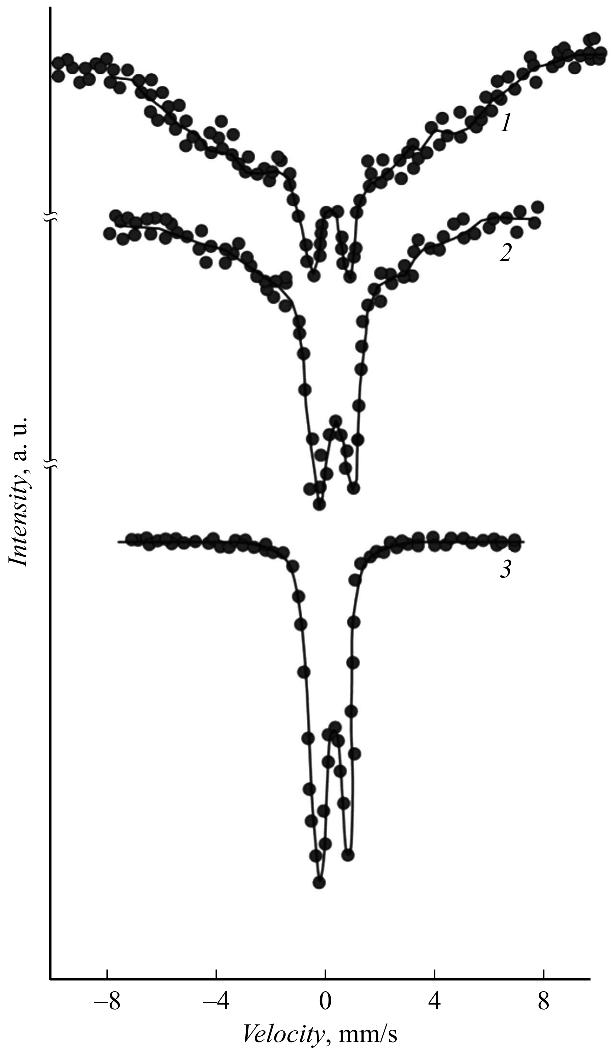

Figure 2. Mössbauer spectra of ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$ obtained at different temperatures. T, K: *1* — 80, *2* — 117, *3* — 160.

MHFS of iron nuclei disappears, and only asymmetric QD with splitting $\Delta = 0.81$ mm/s remains in the spectrum. In the rest temperature range, the spectrum shape does not change.

Taking into account the cation distribution in Ni–Gd ferrites $(Gd_x^{2+}Fe_{1-x}^{3+})_A[Ni_{1-x}^{2+}Fe_{1+x}^{3+}]_BO_4$ [2], the QD asymmetry may be explained by superposition of a single iron ion line in the A sublattice onto symmetric QD produced by iron ions in the B sublattice (Fig. 3); the ratio of resonance line areas $S_A/S_B = 0.18$ matches the ferrite cation distribution (curve *1* in Fig. 3). The obtained spectra were analyzed using the UnivemMS code.

Typically, the iron nuclei MHFS in ferrites decreases to zero at T_c [1]; therefore, investigation of the mechanism of the MHFS abnormal absence in the magnetically ordered temperature range is of undoubted interest.

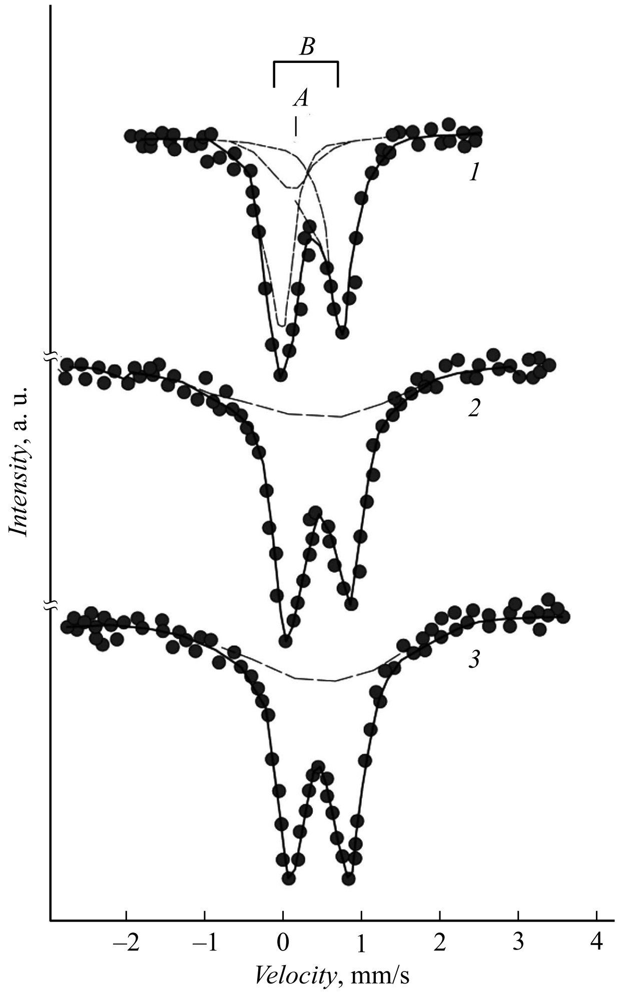

Figure 3. Mössbauer spectra of ferrite $Ni_{0.3}Gd_{0.7}Fe_2O_4$ at $T = 295$ K with accounting for the $(Gd_{0.7}^{2+}Fe_{0.7}^{3+})_A[Ni_{0.3}^{2+}Fe_{1.3}^{3+}]_BO_4$ cation distribution in the absence and in the presence of the external magnetic field. H, kOe: *1* — 0, *2* — 6, *3* — 15. *1* — 0, *2* — 6, *3* — 15. The field is oriented parallel to the sample plane and perpendicular to the γ-ray propagation. A and B are the sublattices.

Note that we have revealed a similar phenomenon in invar alloys $Ni_{31}(Fe_{69-x}Cr_x)$, where $x = 6$ and 8 [3]. In these alloys, the MHFS decreases to zero in the magnetically ordered temperature range is explainable by the presence in the alloys of superparamagnetic clusters whose magnetic moments relax at increased frequency between differently populated energy levels [4]. Note that Mössbauer spectroscopy is successfully used in studying the structural and magnetic properties of magnetic materials [1.5–7]. Let us show that in this case the absence of MHFS in the magnetically ordered temperature range is associated with another relaxation process of the iron spins. Let us proceed from the fact that iron ion spins in the cluster relax collectively, i. e. the disappearance or appearance of MHFS for all iron nuclei of the given cluster should occur simultaneously in the case of superparamagnetic relaxation. Fig. 3 presents the ferrite Mössbauer spectra measured in external magnetic fields at $T = 295$ K. One can see that, in the field $H = 6$ kOe, the QD asymmetry decreases, and at the same time some of the iron nuclei begin to exhibit unresolved MHFS (curve *2* in Fig. 3). In the field $H = 15$ kOe (curve *3*), the QD asymmetry completely disappears, which indicates the transition of all the A sublattice iron ions to the ferromagnetic state. In the case of superparamagnetic relaxation, all iron nuclei in the sublattices would exhibit MHFS simultaneously.

The revealed phenomenon may be explained within the relaxation model [8] where the Mössbauer spectrum shape is governed by the ratio between the ion spin relaxation frequency and hyperfine interaction Larmor frequency. As per [8], MHFS of iron nuclei disappears at the ion spin relaxation frequency $\lambda \approx 1000\,s^{-1}$. This is because at such an ion spin relaxation frequency the nuclear spin has no enough time to precess near the direction of the hyperfine magnetic field created in the vicinity of the nucleus by the ion spin.

Based on the results of studies conducted in external magnetic fields, we may note the following.

1. The ion spin relaxation time increases in the external magnetic field.

2. In the A and B sublattices of ferrite, there exist groups of iron spins with different relaxation times. This is evident from the fact that in both sublattices the number of iron nuclei exhibiting MHFS increases with increasing external magnetic field, i. e. not all the sublattice iron nuclei exhibit MHFS simultaneously at the same external field. The fact that at a certain temperature paramagnetic QD arises in the spectrum jointly with MHFS with subsequent increase in its intensity with increasing temperature also indicate the presence in ferrite of a group of iron spins with different relaxation times. The existence of such spin groups may be explained provided the fact is taken into account that magnetic dilution leads to the emergence in the ferrite A and B sublattices of magnetically nonequivalent iron ion positions differing from each other in their immediate environment.

Apparently, iron nuclei belonging to ions that have a greater number of active exchange couplings with neighboring magnetic ions retain MHFS up to higher temperatures and are the first that exhibit MHFS in the external magnetic field.

Conflict of interests

The authors declare that they have no conflict of interests.

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