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Relaxation process of magnetic moments in chromium-doped Invar alloys

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Invar alloys $Ni_{31}(Fe_{69-x}Cr_x)$ $x = 3$, 4, 6 and 8 (at.%) in the temperature range 120–400 K have been studied by magnetic and Messbauer methods. It is find out the absence of a magnetic hyperfine structure of ^{57}Fe nuclei in the magnetically ordered temperature range for alloys with $x = 6$ and 8. The magnetic structure of the alloys can be represented as a system of superparamagnetic clusters, the magnetic moments of which relax between differently populated energy levels. The phenomenon is explained by the increased frequency of relaxation of magnetic moments between these levels.

Keywords: invar alloy, magnetization, Curie temperature, magnetic moment, relaxation process, Messbauer effect.

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Invars are commonly defined as Fe–Ni alloys with an fcc lattice containing 25–40 at.% of Ni. Their physical properties have several abnormal features. Among these are the behavior of the thermal coefficient of linear expansion, a strong compositional dependence of the saturation magnetization, and high values of magnetic susceptibility of the paraprocess and the magnetostriction [1]. The magnetic structure of invar alloys is heterogeneous with ferro- and antiferromagnetic components possibly coexisting [2,3]. Depending on the alloy composition, each of these components may be present in the form of clusters in the matrix of the other component [3].

In the present study, we examine the temperature dependences of the Mössbauer effect and the magnetization of invar alloys with an admixture of chromium $Ni_{31}(Fe_{69-x}Cr_x)$ ($x = 3$, 4, 6, and 8 at.%). The aim of this research was to compare the temperatures of magnetic ordering derived from the results of magnetic (Curie temperature T_C) and Mössbauer (T_M) measurements. Note that T_C of an alloy doped with chromium decreases (depending on the chromium concentration), although the slope of the magnetization curve in the region of T_C remains fairly steep. This is the reason why invar alloys doped with chromium are used in thermal energy converters as thermomagnetic materials with predefined values of T_C [4,5].

Magnetization measurements were performed in the temperature interval of 120–400 K using a VM2-A vibration magnetometer. Mössbauer spectra of ^{57}Fe nuclei were measured with a YaGRS-4M spectrometer with a $^{57}Co(Cr)$ γ-radiation source. Samples for Mössbauer measurements had the form of disks 15 mm in diameter and 30 μm in thickness that were cut out of alloy ingots and polished with diamond paste. Temperature measurements were performed by introducing the studied sample into a temperature chamber (combined with a cryostat) with a smooth adjustment of temperature within the 120–400 K interval. The system of automatic temperature control maintained the set temperature with an error of $\pm 0.5°$. Curie temperature T_C of the alloy was determined by analyzing the temperature dependence of magnetization, and T_M was assumed to correspond to the onset of broadening at half-maximum of absorption lines of paramagnetic Mössbauer spectra.

Figure 1 shows the temperature dependences of saturation magnetization $M_s(T)$ of alloys, while Fig. 2 shows the Mössbauer spectra of alloys with $x = 3$ and 8 at.%. The spectra take the shape of relaxation ones, which typically feature significant broadening of outer absorption lines of the magnetic hyperfine structure (MHFS) of ^{57}Fe nuclei [6]. Figure 3 presents the temperature dependences of the onset of broadening of absorption lines of paramagnetic Mössbauer spectra for $Ni_{31}(Fe_{69-x}Cr_x)$ alloys. As the tem-

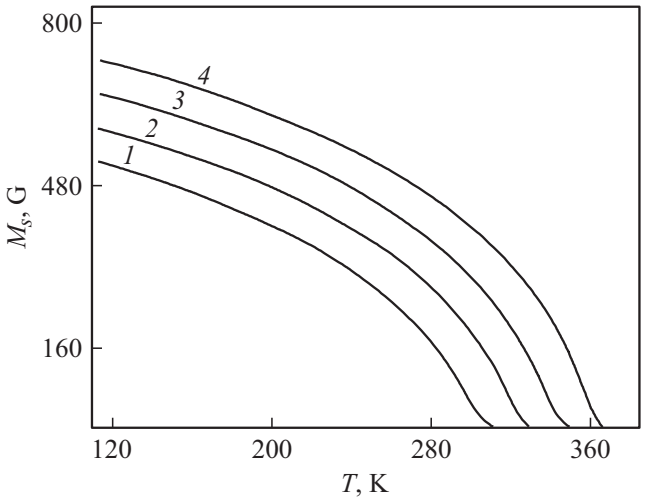

Figure 1. Temperature dependences of the saturation magnetization of $Ni_{31}(Fe_{69-x}Cr_x)$ alloys. $x = 8$ (*1*), 6 (*2*), 4 (*3*), and 3 at.% (*4*).

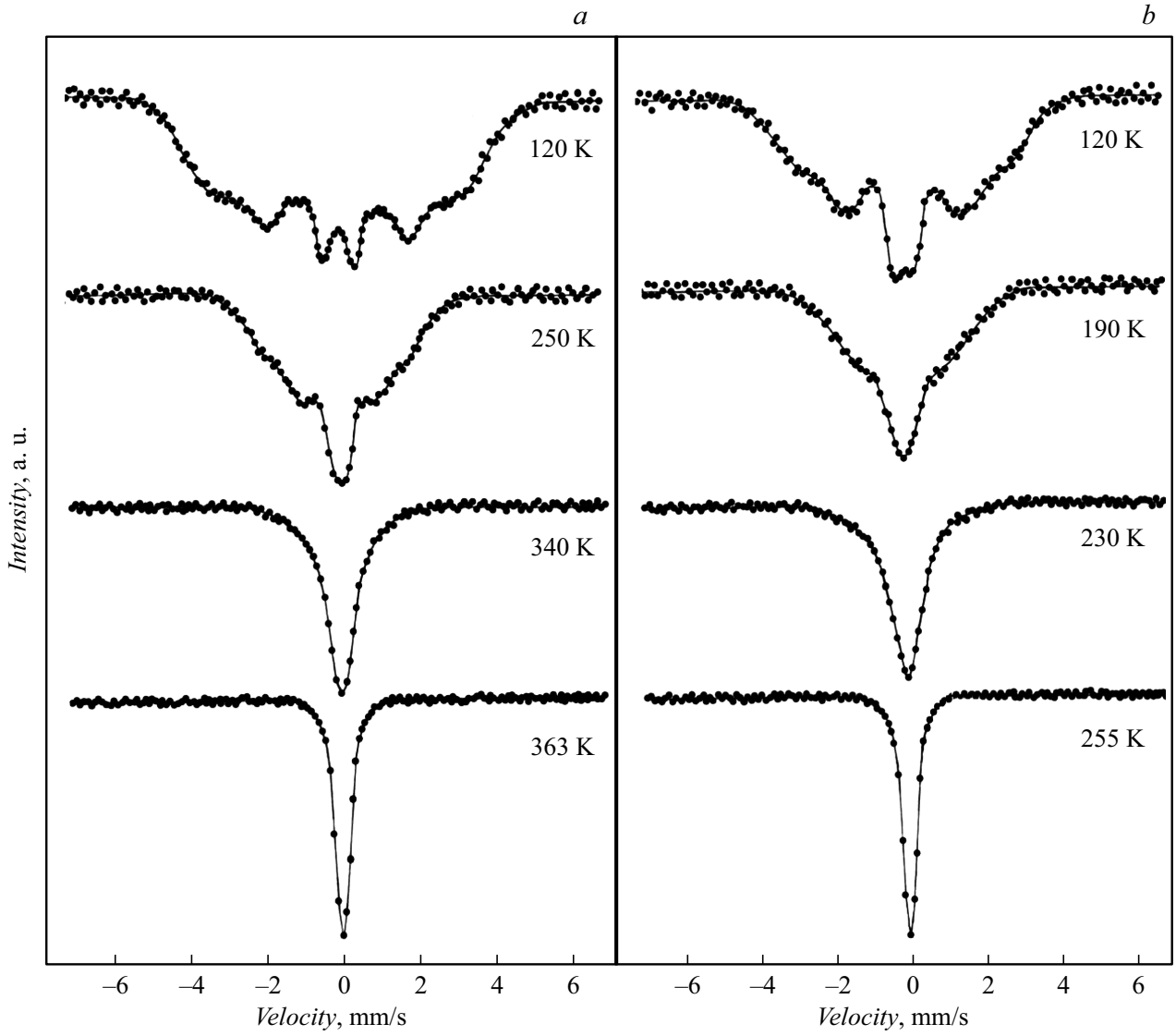

Figure 2. Mössbauer spectra of $Ni_{31}(Fe_{69-x}Cr_x)$ alloys at different temperatures. $x = 3$ (a) and 8 at.% (b).

The values of magnetic ordering temperatures (in kelvins) of $Ni_{31}(Fe_{69-x}Cr_x)$ alloys determined in magnetic (T_C) and Mössbauer (T_M) measurements

Temperature	x, at.%			
	3	4	6	8
$T_C(\pm 3\,\mathrm{K})$	363	344	323	302
$T_M(\pm 3\,\mathrm{K})$	363	344	290	255

perature decreases, the absorption line broadening gradually becomes more pronounced due to the manifestation of unresolved MHFS of ^{57}Fe nuclei in the spectrum. The Univem MS utility was used to analyze the measured spectra. The obtained values of T_C and T_M are listed in the table.

It can be seen that the T_C and T_M values for alloys with $x = 6$ and 8 at.% differ: $T_M < T_C$. Note that Mössbauer spectroscopy is used successfully in research into structural and magnetic properties of magnetic materials containing nanometer-sized particles or clusters [7–9]. It has already been demonstrated experimentally that the antiferromagnetic component of invar alloys of the studied compositions may assume the form of clusters in a ferromagnetic matrix [2,3]. Curie temperature T_C of the ferromagnetic component in such alloys is significantly higher than Néel temperature T_N of the antiferromagnetic component [3]. At the same time, owing to the exchange interaction between these two components, the antiferromagnetic component does not enter a paramagnetic state until T_C of the ferromagnetic component is reached [3]. Thus, the bulk of the ferromagnetic alloy matrix becomes closer in its nature to a system of superparamagnetic clusters [3].

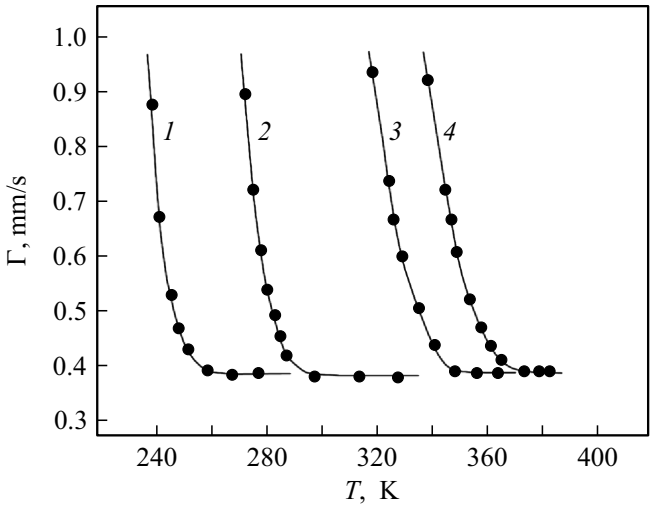

Figure 3. Temperature dependences of the onset of broadening of absorption lines of paramagnetic Mössbauer spectra for $Ni_{31}(Fe_{69-x}Cr_x)$ alloys. $x = 8$ (*1*), 6 (*2*), 4 (*3*), and 3 at.% (*4*).

This is evidenced by the relaxation form of Mössbauer spectra (Fig. 2). Taking these features of the magnetic structure in the region of T_C into account, one may present the bulk of the ferromagnetic alloy matrix as a system of superparamagnetic clusters with their magnetic moments relaxing between differently populated energy levels [10]. More specifically, the relaxation between cluster states with opposite directions of the magnetic moment proceeds as a transition between different energy level values. One direction then dominates over the other within the time interval of relaxation of the magnetic moment of a cluster between opposite directions.

The following relation is a prerequisite for MHFS observation in Mössbauer experiments [6]:

$$\omega_L \tau_s \geqslant 1, \qquad (1)$$

where ω_L is the Larmor precession frequency of nuclear spins in effective magnetic field H_e at the nucleus and τ_s is the relaxation time of electron spins producing this field (i.e., the characteristic time of fluctuation of the effective field at the nucleus).

It is evident that condition (1) is not fulfilled for alloys with $x = 6$ and 8 at.% at $T \geqslant T_M$ (i.e., $\omega_L \tau_s < 1$ in the vicinity of T_C). In view of the difference in population of energy levels, the direction of magnetizing field H will dominate over the opposite direction in magnetization of a system of superparamagnetic clusters within the time interval of relaxation of magnetic moments. Naturally, a magnetically ordered state will be observed in such a system as a collective phenomenon regardless of the relaxation time of magnetic moments. However, if relation (1) is not fulfilled in the relaxation process of magnetic moments of clusters, Mössbauer spectra will not register the MHFS of ^{57}Fe nuclei in such a system; i.e., the nuclear spin has no time in this case to precess about the direction of the effective magnetic field produced by the electron spin in the region of the nucleus. At $T = T_C$, the time interval of relaxation of magnetic moments of clusters between opposite directions evens out, and the system enters a paramagnetic state.

Thus the magnetic structure of chromium-doped invar alloys in the vicinity of Curie temperature T_C may be presented as a system of superparamagnetic clusters with their magnetic moments relaxing between differently populated energy levels. The lack of the MHFS of ^{57}Fe nuclei in the magnetically ordered temperature region is attributable to the elevated frequency of relaxation of magnetic moments of clusters between these levels.

Conflict of interest

The authors declare that they have no conflict of interest.

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