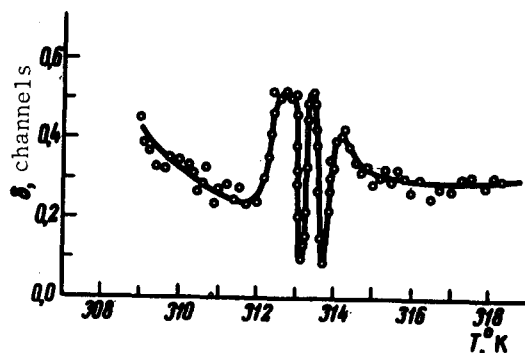Fig. 1

Fig. 2

Fig. 1. Temperature dependence of the shift δ of the resonance line in the temperature range 332 — 350°K. The shift was determined relative to a "standard" absorber (0.5 at.% Sn in Pd at room temperature). One channel corresponds to 0.031 mm/sec.

Fig. 2. The same as in Fig. 1, for the temperature range 309 — 318°K.

Since the data obtained by us on the "fine structure" of the temperature dependence of the isomer shift are so far the only available ones, and this structure has turned out to be rather complicated, we are unable at present to offer a well-founded interpretation of the observed phenomenon. It is reasonable to assume, however, that the variation of the electron density in the region of the nucleus are connected with changes in the electronic wave functions near T_C, induced by the effective exchange field. This can give rise in the electron-state density to singularities (possibly similar to the known singularities that occur in metals placed in an external magnetic field). The change in temperature brings about a change in the exchange field, and consequently a shift of the singularities as functions of the electronic-state densities relative to the Fermi level. Naturally, the abrupt changes in the densities of the electronic states on the Fermi surface can be reflected in abrupt changes of the isomer shift. If such an interpretation turns out to correspond to reality to a certain degree, then measurements of the isomer shift can serve as a new method for the spectroscopy of the electronic states in magnetic metallic systems. To explain the physical nature of the observed phenomenon we propose to carry out detailed investigations for different alloys with a better temperature resolution.

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MAGNETIC MOMENT OF Mn ATOMS IN THE $Ni_{1-x}Mn_x$ SYSTEM

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The high-frequency parts of the NMR spectra of $Ni_{1-x}Mn_x$ alloys have been resolved for the first time as a result of the different orientations of the magnetic moments of the Mn atoms. This has made it possible to estimate the local moments of Mn.

The atomic moments in the $Ni_{1-x}Mn_x$ system were investigated both experimentally and theoretically [1-4]. Neutron-diffraction investigations [2] have shown that in the disordered state we have $\mu_{Mn} = 3 \mu_B$ and $\mu_{Ni} = 0.6 \mu_B$ at concentrations x < 6 at.%, and that the moments decrease at x > 6 at.%. However, neutron-diffraction methods of determining the local magnetic moments in systems having no ideal order do not make it possible to determine the true values of the

moments, and yield only "effective" [2] or "mean" [3] values. A calculation by the coherent-potential method [4] likewise failed to yield unique values of the individual moments. Our investigation of the $Ni_{1-x}Mn_x$ by the spin-echo method has made it possible to observe new singularities of the NMR spectrum and to obtain certain estimates of the moments of the Mn atoms.

The $Ni_{1-x}Mn_x$ samples were melted from components 99.99% pure in an arc furnace in an argon atmosphere. The homogenized castings were powdered to particle dimension 50 μ and less, and subjected to ordering annealing in evacuated quartz ampules for 10 days. An x-ray diffraction analysis of the sample revealed no presence of second phases. The component concentrations in the alloys were obtained from chemical-analysis data.

The nuclear spin echo spectra of Mn^{55} were obtained by using pulses of 1 μsec duration spaced 15 μsec apart. The main results of the measurements at 4.2°K are shown in the figure.

According to [5] and our data (e.g., sample 4a in the figure), there are no signals from disordered samples at frequencies above 370 MHz, so that it can be assumed that the peak in the 340 — 350 MHz region is due to the Mn nuclei with a disordered environment of neighbors, while the high-frequency group is due to an ordered environment. It is natural to assume that the principal peak of the high-frequency group at 391.3 MHz is due to Mn nuclei in a fully ordered environment consisting of 12 Ni neighbors in the first coordination sphere, six Mn neighbors in the second, etc. Since the degree of order in all the samples is less than unity, the first sphere can contain a manganese atom with a magnetic moment either parallel to the given moment, or antiparallel, depending on its local environment: if there are two or less nearest Mn neighbors, the moment of the Mn atom is parallel to the moments of the neighbors, and if there are three and more Mn atoms in the first sphere, the moment is antiparallel [3, 6]. The ferromagnetic orientation of the nearest Mn neighbor corresponds in this case to a peak at a higher frequency, 396.4 MHz, and the antiferromagnetic orientation of the Mn neighbor in the first sphere corresponds to a peak at a lower frequency, 385.1 MHz. At low Mn concentrations, the 396.4-MeV peak is more intense than the 385.1-MHz peak, but when x increases their intensities become equalized, and at higher Mn concentrations the 385.1 MHz peak becomes more intense. This corresponds to an increase in the probability that three or more Mn atoms are included among the nearest neighbors when x is increased. For large values of x, the probability that the second Mn atom occupies the position of Ni also becomes appreciable, i.e., the probability of the appearance of a second Mn atom with an antiparallel spin orientation in the first coordination sphere. This configuration should correspond to a resonance at a frequency lower than 385.1 MHz. Indeed, in the spectra of the samples with the maximal x one observes an incompletely resolved line at ∿ 379 MHz, corresponding to this configuration.
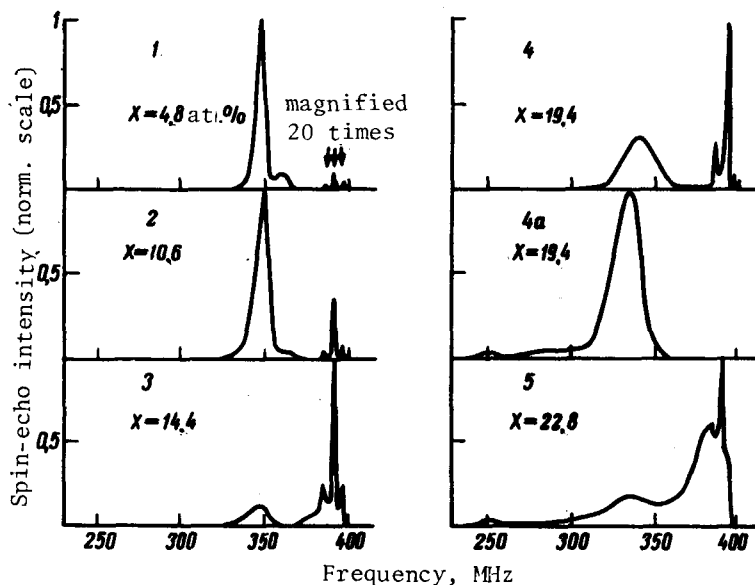
Let us present some quantitative estimates. We assume that the magnetic moments of Mn and Ni remain unchanged for configurations that are close to ordered, and are equal to 3.2 μ_B and 0.3 μ_B, respectively [1]. For a disordered environment corresponding to a mean statistical distribution of the atoms in a sufficiently large number of coordination spheres, these values, according to [2], are 3.0 μ_B and 0.6 μ_B. To calculate the hyperfine field at the Mn nuclei we shall use the phenomenological relation

$$H_{Mn} = a\mu_{Mn} + \sum_i N_i\, b_i\, \overline{\mu}_i , \qquad (1)$$

where N_i is the number of atoms in the i-th coordination sphere, $\overline{\mu}_i$ is the average magnetic moment of the atoms in the i-th sphere, and a and b_i are coefficients. The field at the Mn nuclei is negative [5]. Using (1) for the lines of the high-frequency group of the spectrum, which correspond to changes of the environment in the first coordination sphere, we obtain $b_1 = -1.7$ kG/μ_B. To determine the contribution of the next coordination spheres, it is convenient to use in addition to the high-frequency lines also the 344.5-MHz line [5], which corresponds to Mn atoms surrounded primarily by Ni atoms in several closest spheres. If only two coordination spheres are taken into account, calculation yields $b_2 = -1.93$ kG/μ_B, i.e., under these conditions the contribution of the second sphere turns out to be larger than that of the first, which is not very probable. It is natural to assume that this is due to the need of taking into account the influence of the succeeding phases. This is confirmed also by the slope of the plot of the resonant frequency against the Mn concentration for disordered samples [5] at Mn concentrations close to zero. Not being able to determine from the experimental data the individual contributions of the succeeding spheres, we took their influence into account with the aid of the "effective" coefficients. It turned out that allowance for six spheres gives good agreement between the calculations and the experimental spectra. In this approximation we have

$$H_{Mn}(\text{kG}) = -96.6\mu_{Mn} - 1.7N_1\overline{\mu}_1 - 0.59N_{2-6}\overline{\mu}_{2-6} . \qquad (2)$$

The peak at 361 MHz is an aggregate of lines with different contents of Mn atoms in the nearest coordination sphere, but not more than two in the first sphere, and the moments of Mn are oriented parallel to the given moment, since they are surrounded mainly by nickel atoms. A peak is seen only at small manganese concentrations.

The position of the peak at 250 MHz (samples 4, 4a, and 5) are quite stable and depend little on either the concentration or on the degree of order of the alloy. This indicates that this peak, like the high-frequency group, is due to a stable configuration of the atoms in the closest coordination spheres. Calculations based in [2] show that this peak corresponds to Mn atoms having an antiferromagnetic orientation relative to Ni. Calculation relative to the line shift in the high-frequency group, when the orientation of the Mn moment in the first coordination group is reversed, confirms that $\mu_{Mn} = 3.2$ μ_B in both orientation. Indeed, the 385.1-MHz peak, which is due to the change of the moment in the first sphere by -3.5 μ_B, should be shifted relative to the principal peak 391.3 MHz by 0.6 μ_B/2.9 μ_B = 21% more than the 396.5-MHz peak, which is due to a +2.9 μ_B change of the moment. But the observed relative shift is equal to 1.1 MHz/5.1 MHz = 21%. Thus, the local moment of the Mn atoms, for configurations close to ordered, remains constant both in its sublattice and in the nickel sublattice. It must be emphasized that the character of the closest environment is quite appreciably altered in this case: the average moment of the atom in the first coordination sphere changes from 0.3 μ_B in the Mn sublattice to 1.27 μ_B in the Ni sublattice (but the average moment in the six spheres changes here insignificantly and even decreases from 1.18 to 0.98 μ_B. We note that in Ni_3Mn, on going from a disordered to an ordered state, the average moment in six spheres increases by more than four times, and when the Mn concentration is increased the difference increases abruptly). Consequently, the influence of the changes in the first coordination sphereon the magnetic moment of Mn in configurations close to ordered is small. At the same time, the good agreement between the calculated resonant frequencies and the experimental ones for disordered configurations, and also the absence of intermediate resonances between the spectral lines corresponding to the ordered and disordered environments (at x < 20%) indicates that the value $\mu_{Mn} = 3.0$ μ_B for the disordered case has also been determined quite correctly. It can apparently be assumed that on going from an ordered environment to a disordered one the magnetic moment of Mn changes abruptly, at a certain configuration, from 3.2 to 3.0 μ_B. Such jumplike changes of the local moments following a change in the environment were observed earlier in the alloys Ni-Cu [7], Fe-Mo-Nb [8], and others.

The fact that allowance for six coordination spheres with the aid of "effective" coefficient gives a good interpretation of the experimental results suggests that the effective radius of the excitation of the spin density around the Mn atom is close to the radius of the sixth sphere and amounts to not less than 6 Å.

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LAW OF SPIN-ECHO DAMPING OF CONDUCTION ELECTRON IN METALS

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The damping of spin-echo signals on conduction electrons in lithium was found to obey a square-root law. The concept of spatially-inhomogeneous spin relaxation on impurities (in the interior of the metal) is introduced and may explain the observed law.

We have previously reported [1, 2] observation of conduction-electron spin echo (CESE) from spheroidal particles of metallic lithium suspended in an LiF dielectric matrix. In those investigations, the maximum time interval τ between the sounding pulses (the interval was limited by the intensity of the echo signal from the investigated samples) was $\sim 0.8 - 1$ µsec. The observed damping of the main and stimulated CESE signals was close to exponential within the limits of experimental error.

We report here preliminary results of an investigation of CESE for analogous samples, but with a larger amount of metal. Improvement of the measurement technique and an increase in the signal intensity have made it possible to increase appreciably the interval τ (to 5 µsec). It turned out, unexpectedly, that the empirical law for the CESE damping is

$$V(2\tau) = V_0 \exp(-\sqrt{2\tau/T_{eff}}). \tag{1}$$
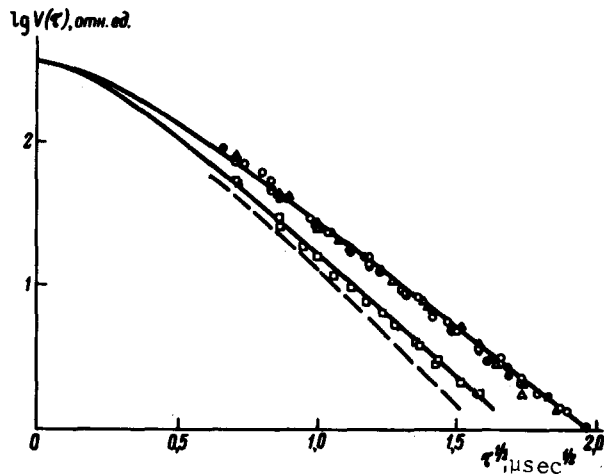
Amplitude of the signal of the main CESE, in relative units, vs. the variable interval between the sounding pulses, τ: o and Δ — experimental points at 300 and 390°K respectively without a gradient, ● and ▲ — the same but with a gradient, □ — experimental points at 77°K without a gradient, dashed line — qualitative character of the echo-signal fall-off with a gradient.