Alignment of magnetic moments of Fe and Mn atoms in a ferromagnetic Pt–Fe alloy

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Mössbauer spectroscopy with the ^{57}Fe nucleus and the low-temperature nuclear orientation of ^{54}Mn have been used to study the local magnetization of Fe and Mn in the ferromagnetic alloy $Pt_{90}Fe_{10}$. Measurements of the Mössbauer effect at 4.2 K in longitudinal external magnetic fields of 0.5 and 1 T reveal that the moments of the Fe atoms are completely ordered. The magnetic hyperfine field for ^{57}Fe is found to be 33.2 ± 0.6 T. The moments of the Mn impurity atoms, in contrast, are not collinear with the macroscopic magnetization of the sample, according to the observed temperature dependence of the effective magnetic field at ^{54}Mn in the $Pt_{90}Fe_{10}$ matrix. The magnetic field at the ^{54}Mn nucleus is 38.4 ± 0.6 T, according to a calculation based on the experimental data with allowance for the noncollinearity.

1. INTRODUCTION

Alloys of iron with platinum, like alloys of iron with palladium, are well known to be systems with giant magnetic moments, which are associated with the 3*d* impurity. The threshold concentration for the appearance of a ferromagnetism in a Pt–Fe alloy is estimated[1] to be 0.76 at.% Fe. In contrast with Fe (and Co), a Mn impurity in Pt does not form a giant moment, and it does not give rise to ferromagnetic order. In this regard, the Pt–Mn alloys differ from Pd–Mn alloys, which acquire ferromagnetic order at low Mn concentrations (up to 3 at. %). At the same concentrations of Mn in Pt, a spin glass is observed to form in the system.[2,3]

An extremely productive way to study the local magnetization of impurity atoms is to use methods based on hyperfine interactions and the detection of radioactivity: nuclear orientation, emission Mössbauer spectroscopy, and perturbed angular correlations. Methods of low-temperature nuclear orientation have been used previously[4,5] to study the magnetic behavior of a ^{54}Mn impurity in a Pt–Fe matrix with an Fe concentration up to 1 at. %. A hindered alignment of the Mn moments was observed in external magnetic fields up to 8 T. This result was attributed to the Mn–Fe interaction which causes the Mn spins to be noncollinear with the magnetization of the sample. It turns out that the extent of the deviation from collinearity increases with the Fe concentration in the alloy.

As the subject of this study we selected the alloy $Pt_{90}Fe_{10}$, with an Fe concentration well above the threshold for ferromagnetism. The measurements were carried out by methods based on the nuclear orientation of ^{54}Mn, added to the sample as an impurity, and on the Mössbauer effect involving the ^{57}Fe nucleus. The purpose of this study was to determine the nature of the magnetic ordering of the manganese and iron atoms.

2. MÖSSBAUER MEASUREMENTS

To study the degree of alignment of the Fe magnetic moments, which determine the magnetization of the $Pt_{90}Fe_{10}$ alloy, we carried out Mössbauer measurements involving the nucleus ^{57}Fe in an external magnetic field oriented parallel to the direction in which the γ rays were detected. We know that when a polycrystalline sample is polarized in a longitudinal magnetic field the intensity of the hyperfine components corresponding to transitions between Zeeman sublevels with identical spin projections ($\Delta m = 0$) is zero. The presence or absence of the second and fifth components of the spectrum of the ^{57}Fe hyperfine structure thus indicates noncollinearity and alignment, respectively, of the Fe moments.

Because of the strong absorption of 14.4-keV γ rays by platinum, the absorber for the Mössbauer studies must be quite thin. Accordingly, filings of the alloy (enriched in the isotope ^{57}Fe) were ground in a mortar, and the finely divided fraction was selected. The resultant thickness of the absorber in terms of ^{57}Fe was 0.4 mg/cm^2.

The Mössbauer-effect measurements were first carried out in a zero magnetic field over the temperature range from room temperature down to ≈ 5 K. Figure 1 shows Mössbauer spectra of ^{57}Fe measured at various temperatures. At a low temperature the spectrum consists of a well-resolved sextet, which corresponds to the hyperfine field $B_{hf} = 33.2(6)$ T. From the temperature dependence of B_{hf} we found the Curie point for the given sample: $T_C \simeq 160$ K. This value agrees well with the value of 159.5 K which has been found for an alloy with the same composition.[6]

The measurements in external magnetic fields of 0.5 and 1 T were carried out at a temperature of 4.2 K. Since the procedures used to fabricate the absorber result in a stress, which can in turn give rise to a texture, it is necessary to anneal the test sample before the measurements. It was found experimentally that annealing at 800 °C in vacuum sinters the powder. Accordingly, we used for the measurements an absorber annealed at the relatively low temperature of 450 °C. Figure 2 shows Mössbauer spectra measured in an external magnetic field of 0.5 T as a function of the duration of the annealing (a–c), along with spectra mea-

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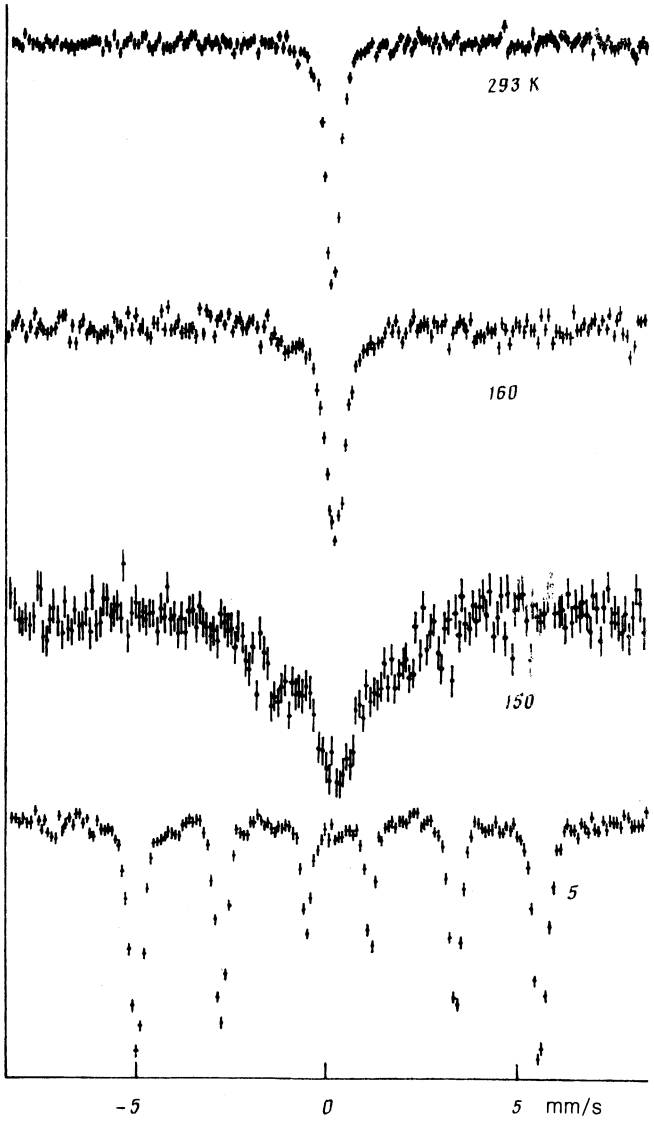

FIG. 1. Mössbauer spectra of ^{57}Fe in $Pt_{90}Fe_{10}$ in a zero external magnetic field at various temperatures. The source is ^{57}CoPd, and the detector is a resonant counter. The count rate, in arbitrary units, is plotted along the ordinate.

sured in an external field of 1 T (d). It can be seen from this figure that the annealing causes a dramatic decrease in the intensities of the inner components. The Mössbauer spectrum measured in a longitudinal magnetic field of 1 T indicates that the magnetic moments of the Fe atoms are aligned parallel to the external magnetic field.

3. NUCLEAR-ORIENTATION MEASUREMENTS

The magnetic behavior of the Mn impurity in the $Pt_{90}Fe_{10}$ ferromagnetic matrix was studied by the nuclear-orientation method at low temperatures. The test sample was prepared in the following way: A substrate of an alloy with ^{54}Mn activity (deposited from a $MnCl_2$ solution) was remelted in vacuum. The resulting regulus was rolled to a thickness $\approx 5\ \mu$m and annealed at 900 °C. The Mn concentration in the sample did not exceed 10^{-5} (the concentration of stable Mn in the radioactive ore is being taken into account here).

In the nuclear-orientation experiments, the observed quantity is the anisotropy of the angular distribution of the nuclear emission with respect to the axis of the nuclear orientation. The angular distribution of the γ rays accompanying the decay of the oriented nuclei is

$$W(\theta)=1+\sum_{k=2,4} A_k U_k Q_k B_k(T) P_k(\cos\theta), \qquad (1)$$

where A_k and U_k are coefficients which are determined by the particular nuclear decay scheme, the Q_k are corrections for the finite solid angle of the detector, and $B_k(T)$ are the orientation functions of the mother nucleus, which are determined in the case of a magnetic dipole interaction by the parameter $\mu B/(IkT)$. Here μ and I are the magnetic moment and spin of the nucleus, B is the magnetic field at the nucleus, $P_k(\cos\theta)$ are the Legendre polynomials, and θ is the angle between the orientation axis and the γ emission direction. If the nuclear parameters and the temperature are known, one can work from the measured anisotropy to determine the magnetic field acting on the nucleus. The method of low-temperature nuclear orientation also makes it possible to detect a possible deviation of the atomic magnetic moments from the direction collinear with the quantization axis, which is set by the direction of the external magnetic field. If the magnetic field acting on the nucleus is calculated with the help of expression (1) under the assumption that the nuclear-orientation axis coincides with the direction of the external magnetic field, then the effective magnetic field B_{eff} calculated in this manner turns out to differ at different measurement temperatures in the case of a deviation from collinearity. The reason is that the angular distributions of the γ emission with respect to the direction of the nuclear spin depend on the temperature. This effect makes it possible to detect noncollinearity through measurements of the temperature dependence $W(T)$ if it turns out that B_{eff} depends on the temperature (specifically, it decreases with decreasing temperature[7]).

We measured the γ anisotropy of the ^{54}Mn nuclei ($E_\gamma = 835$ keV) as a function of the external magnetic field at a constant temperature and also as a function of the temperature in external magnetic fields of 0.46 and 1.16 T. The temperature was measured by a ^{60}CoFe nuclear thermometer. The test sample and the thermometer were soldered to a heat exchanger with a developed surface and placed in the dissolution chamber of a ^{3}He–^{4}He refrigerator (the SPIN apparatus of the Joint Institute for Nuclear Research[8]). The γ emission was detected by two Ge(Li) detectors, placed at angles of 0° and 90° with respect to the direction of the orienting external field, which was in turn produced by a pair of superconducting coils.

Figure 3 shows values of B_{eff} at ^{54}Mn in the $Pt_{90}Fe_{10}$ matrix calculated from the experimental data as a function of the external magnetic field B_{ex}. We see that the magnetic field at the nucleus remains essentially constant at ~ 28 T for $B_{\text{ex}} \gtrsim 0.3$ T. This value is well below the hyperfine field for Mn in Pt, $B_{hf} = 37.5$ T (Ref. 9). The results on the effect of the temperature, found for two values of the external magnetic field, show that B_{eff} decreases with decreasing temperature (Fig. 4). As was mentioned above, this behavior is evidence that the Mn moments are not collinear with the

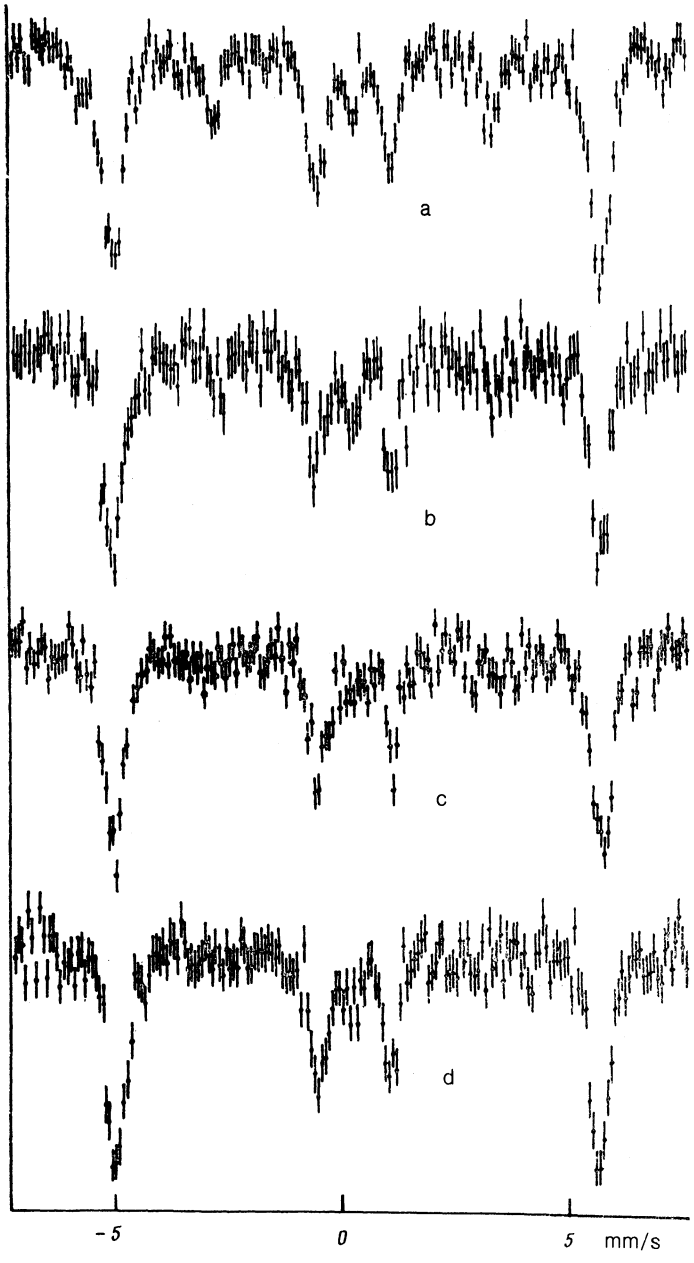

FIG. 2. Mössbauer spectra of ^{57}Fe in $Pt_{90}Fe_{10}$ in an external magnetic field at 4.2 K. The source is $^{57}CoCr$, and the detector is a scintillation detector. a—Absorber which has not been annealed; b—annealed for 18 h; c, d—annealed for 100 h. The count rate, in arbitrary units, is plotted along the ordinate.

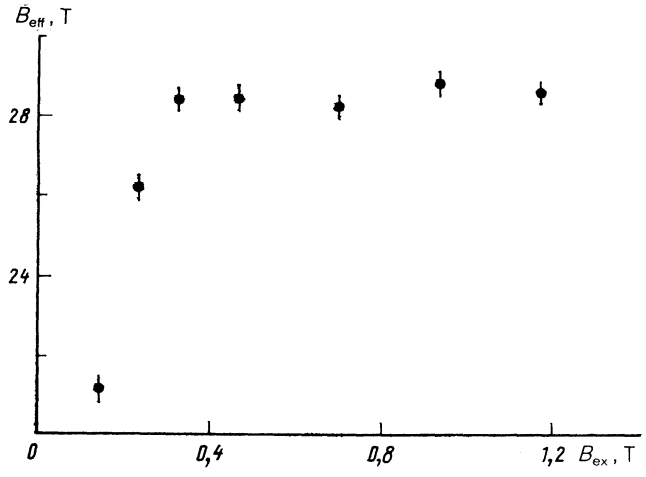

FIG. 3. B_{eff} at ^{54}Mn in $Pt_{90}Fe_{10}$ versus B_{ex} at 12 mK.

macroscopic magnetization. The latter is parallel to the alignment direction of the iron atoms, along the external magnetic field.

The experimental temperature dependence $W(T)$, along with the results measured by the two detectors, was approximated by expression (1) under the assumption of Lorentzian and Gaussian distributions of the directions of the Mn spins (a $\cos\varphi$ distribution, where φ is the angle between the Mn moments and B_{ex}). The adjustable parameters of this fit were B (the field of the nucleus), $(\cos\varphi)_m$, i.e., the value corresponding to the peak in the distribution, and the half-width Γ or the standard deviation σ of the distribution in the case of Lorentzian and Gaussian fits, respectively. The Lorentz distribution turned out to be preferable according to the χ^2 test. For both values of the external magnetic field we found $B(^{54}Mn) = 38.4(6)$ T, $(\cos\varphi)_m = 1$, and $\Gamma = 0.08$.

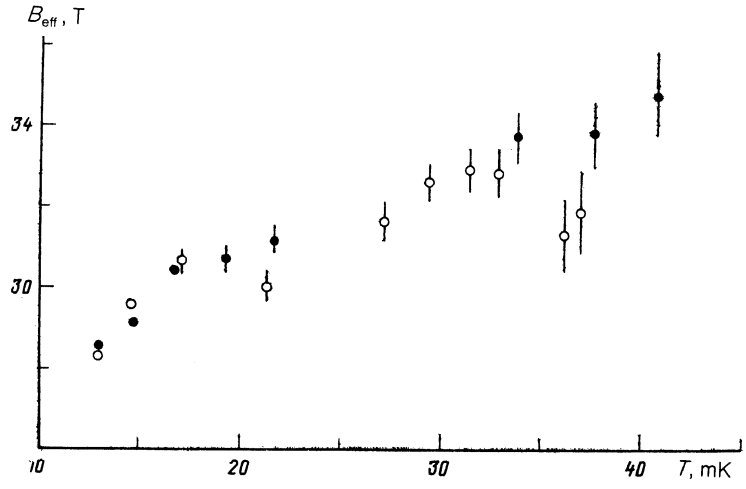

FIG. 4. Temperature dependence of B_{eff} at ^{54}Mn in $Pt_{90}Fe_{10}$. ○—$B_{ex} = 1.16$ T; ×—$B_{ex} = 0.46$ T.

4. DISCUSSION OF RESULTS

In summarizing the experimental results, we should point out the following.

1. The Mössbauer-effect measurements showed that in an external magnetic field the Fe atoms responsible for the ferromagnetism of the alloy studied here, $Pt_{90}Fe_{10}$, are aligned essentially completely with the external magnetic field. This conclusion follows from the fact that there are no central components in the spectrum of the ^{57}Fe hyperfine structure. It can thus be asserted that the axis of the macroscopic magnetization of the matrix coincides with the external field.

2. The nuclear-orientation studies of the ^{54}Mn impurity indicate that the Mn magnetic moments are not collinear with the magnetization axis of the matrix. By approximating of the temperature dependence of the γ anisotropy, with allowance for this noncollinearity (i.e., for the presence of an angle between the orientation axis and the direction of the external field), showed that the field at the ^{54}Mn nucleus in the $Pt_{90}Fe_{10}$ matrix is $B = 38.4(6)$ T. This result corresponds within the strength of the external field to the hyperfine magnetic field for Mn in Pt and Pd matrices and also in noble metals.[10]

3. The field dependence of B_{eff} at Mn indicates that B_{eff} is constant in external fields above 0.3 T.

It thus follows from these experimental results that the Mn spins in the system of Pt with 10 at. % Fe do not follow the Fe moments, which are oriented along the external magnetic field. The observed behavior of the Mn moments can be explained under the assumption that in this ferromagnetic sample, with its fairly high Curie point ($T_c = 160$ K), the distance-dependent antiferromagnetic Mn–Fe interaction plays an important role along with the ferromagnetic exchange interaction (the antiferromagnetic Mn–Mn interaction can be ignored because of the extremely low Mn concentration in the alloy). In this case, competition may arise between the two interactions. In the alloy studied, a significant fraction of the Mn atoms have Fe atoms as nearest neighbors. The moments of these nuclei in Mn are oriented antiparallel to the Fe moments. In an external magnetic field, the antiparallel bonds are broken, with the result that these Mn moments become disoriented, and there is simultaneous alignment of the Mn spins, with a small noncollinearity. Over a broad interval of the external field (beginning at the value of B_{ex} at which the Fe atoms are completely aligned) the total numbers of Mn atoms which are oriented along and opposite the field remain the same. Since the directions of the nuclear spins along and opposite the field are equivalent for the nuclear-orientation method [only even powers of $\cos\theta$ appear in (1)], the observed anisotropy and therefore the effective magnetic field at the nuclear are constant. Some of the Mn atoms evidently have an intermediate orientation (something between an orientation along the external field and one opposite it), so the value found for B_{eff} is lower than B_{hf}.

It follows from the approximation of the temperature dependence of $W(T)$ that the peak in the angular distribution of the Mn spins corresponds to the value $(\cos\varphi)_m = 1$. In other words, a majority of the atoms are aligned along (or opposite) the external field. The atoms with an intermediate orientation are not completely random, as follows from the observed temperature dependence of B_{eff}.

An alternative explanation of the observed dependence of B_{eff} on B_{ex} is again based on a competition between ferromagnetic and antiferromagnetic interactions[10] of the Mn impurity. As a result of this competition, the transverse components of the Mn moments freeze in a random way at T_f above the temperatures of the nuclear-orientation measurements. When the longitudinal components of these moments align along the field (at $B_{ex} > 0.3$ T), the total Mn moment turns out to lie at some angle with respect to the external field. Since the extent of the deviation from alignment (the canting) should not change in relatively weak external fields, the effect is seen as a "saturation" of the effective magnetic field (Fig. 3). This deviation of the system from collinearity may be responsible for the observed temperature dependence of B_{eff}.

The results of this study are consistent with the conclusion that the ^{54}Mn($Pt_{90}Fe_{10}$) system can be thought of as consisting of two spin systems: the Fe moments which are aligned along the direction of the external magnetic field and the partially noncollinear Mn moments.

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