Relaxation in a transition to a spin-glass state in a dilute PdFe alloy

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A Mössbauer-effect technique has been used to observe the transition to a spin-glass state in the alloy PdFe (0.06 at. %) at $T_f \approx 0.1$ K. The use of the isotopes ^{57}Fe and ^{57}Co as probes has made it possible to determine the range of relaxation times below T_f: from 10^{-9} to $\gtrsim 10^{-2}$ s.

Study of the relaxation of spins in spin glasses is of decisive importance for reaching an understanding of the properties of such systems and for resolving the question of the existence of a phase transition. Mössbauer studies of dilute alloys of PdFe, which are well known as systems with a huge magnetic moment at an impurity, have revealed an unusually slow relaxation of the electron spins as the temperature is lowered.[1,2] It was shown in Ref. 2 that the relaxation in the alloy PdFe (0.01 at. %) is largely determined by spin-spin interactions of the RKKY type, which are responsible for the formation of a spin glass.

In the present study we used the method of emission Mössbauer spectroscopy to study the alloy PdFe, whose impurity iron concentration was 0.06 ± 0.005 at. %, according to x-ray spectral microanalysis. A PdFe(^{57}Co) radioactive source based on this alloy was prepared by the method of Ref. 2. The measurements were carried out in a ^{3}He–^{4}He dissolution refrigerator over the temperature interval from 25 mK to 4.2 K. The Mössbauer γ rays were detected by a resonant detector.

It can be seen from the spectra measured in a zero magnetic field (Fig. 1) that an isolated line undergoes a broadening with decreasing temperature, and (below 0.2 K) a hyperfine structure arises. This hyperfine structure then converts into a completely resolved six-component spectrum. The temperature dependence of the quantity $1/\Delta\Gamma$ [$\Delta\Gamma = \Gamma - \Gamma_0$, where Γ is the width of the line at half-maximum at the measurement temperature, and $\Gamma_0 = 0.42(2)$ mm/s is the instrumental linewidth] is shown in Fig. 2 for the temperature interval in which the spectrum is a broadened singlet. This dependence is linear, without any structural features, suggesting that no phase transition occurs. The observed broadening of the line is a consequence of the slowing of the electronic relaxation of the spins of the Fe.

For a mathematic description of the spectra we use the model of Ref. 3, according to which the shape of the Mössbauer spectrum is calculated for conditions corresponding to a stochastic isotropic relaxation of the hyperfine field. In our case, that approach implies the same relaxation of the Fe spin, since in a dilute alloy the hyperfine field at the Fe is proportional to its spin. We used this model to analyze not only the relaxation-broadened singlets but also the spectra with a hyperfine structure which

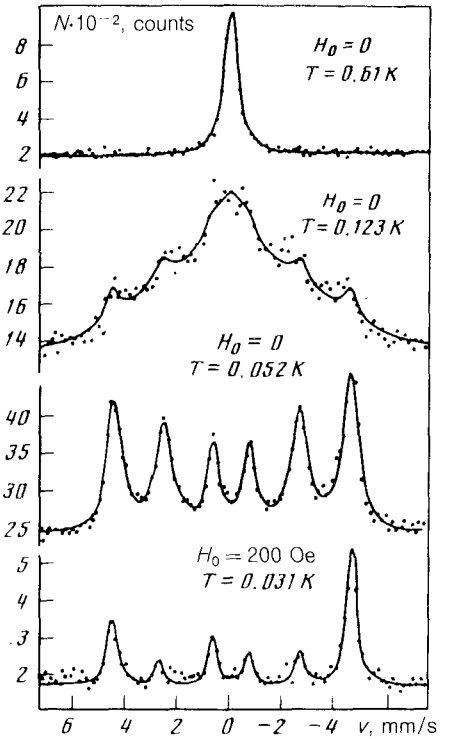

FIG. 1. Mössbauer spectra of the alloy PdFe (0.06 at. %). The solid lines are the results of a computer fit.

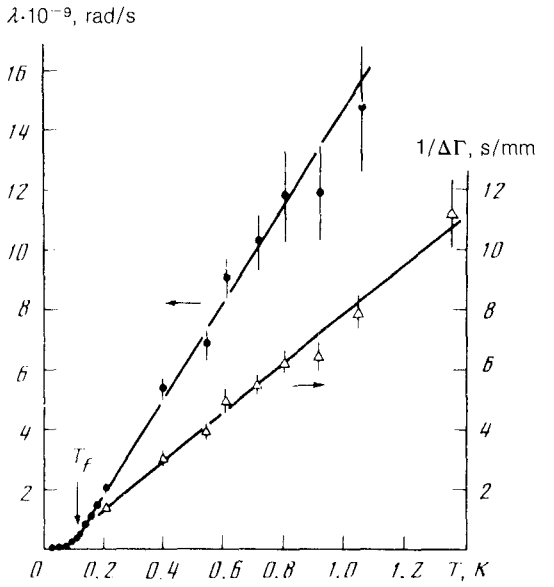

FIG. 2. Temperature dependence of the reciprocal line broadening ($1/\Delta\Gamma$) and of the relaxation frequency (λ).

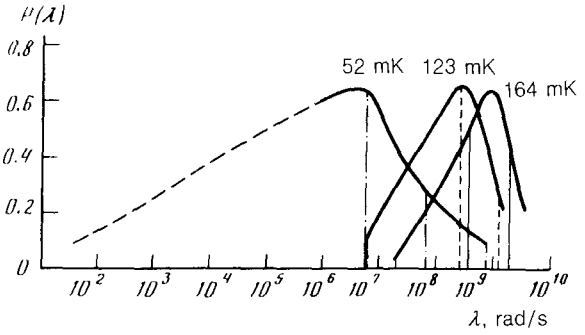

FIG. 3. Approximate shape of the distribution of relaxation frequencies above and below T_f. The vertical line segments correspond to partial frequencies found from the processing of the spectra.

arise at a lower temperature. A satisfactory description of the spectra was achieved under the assumption that there is a distribution of relaxation frequencies, which we found approximately in the form of histograms corresponding to a discrete set of frequencies (Fig. 3). The values of the relaxation frequencies found by the numerical processing are shown in Fig. 2 as functions of the temperature. For spectra with a hyperfine structure [the points on the $\lambda(T)$ curve below 0.2 K] we took averages over the frequency distributions, $\bar{\lambda}$. Over a broad temperature range, the resulting temperature dependence $\lambda(T)$ is linear [as is $1/\Delta\Gamma(T)$], but near $T = 0.1$ K the nature of this temperature dependence changes abruptly, so that the points at temperatures below 60 mK lie essentially on the temperature axis at the scale of this figure. We regard the observed change in the temperature dependence of the relaxation frequency at $T_f \approx 0.1$ K as an indication of the existence of a magnetic transition at this temperature.

The appearance of magnetic order below T_f is confirmed by the asymmetry of the Mössbauer spectra, which can be reliably seen at temperatures below 70 mK. At $T = 52$ mK (Fig. 1), for example, the ratio of the intensities of the outermost components of the spectrum is 1.22 ± 0.03. This asymmetry is known to result from a polarization of the ^{57}Co parent nucleus by the effective hyperfine field. In order to create a nuclear polarization, the hyperfine field would have to relax over a time longer than or comparable to the nuclear spin-lattice relaxation time, which is[4] about 10^{-2} s for ^{57}Co in Pd. Since the hyperfine field is proportional to the spin of the impurity Co, the latter requirement means that there must be a nonzero Edwards-Anderson order parameter $\langle \mathbf{S}(0)\mathbf{S}(\tau)\rangle$, where $\mathbf{S}$ is the spin of the magnetic atom, and the time τ is no shorter than 10^{-2} s. In the system of impurity spins below the transition temperature, there are accordingly both relaxation times $\tau \gtrsim 10^{-2}$ s and times 10^{-9}–10^{-6} s, which determine the shape (the linewidths and the pedestal) of the ^{57}Fe Mössbauer spectrum. At $T > T_f$ and near T_f, the spectrum of relaxation times has a width of 10^{-10}–10^{-7} s; as a result of the transition (at $T < T_f$), this spectrum spans the huge range 10^{-9}–$\gtrsim 10^{-2}$ s (Fig. 3; the low-frequency part of the distribution at $T = 52$ mK is simply a rough indication of the behavior). It is believed[5] that a relaxation behavior of this sort is a distinctive feature of spin glasses.

That the spin configuration at $T < T_f$ is noncollinear, as it would have to be for

spin glasses, follows from measurements in a longitudinal magnetic field. The presence of the "forbidden" second and fifth components (see the lower spectrum in Fig. 1) is evidence that the Fe spins become disoriented with respect to the applied field; this is a consequence of the noncollinear exchange fields at the magnetic atoms.

An interpretation of the observed relaxation picture can be summarized as follows: The spectrum of relaxation times at $T > T_f$ is due to the existence of magnetic clusters of various sizes (within which the interaction between spins satisfies the condition $V > T$), and the change in the relaxation regime below T_f is evidence of the appearance of a degenerate state. Here this degenerate state is characterized by macroscopically long times, corresponding to transitions through "infinite" barriers.[5]

We note in conclusion that the possibility of using both ^{57}Fe and the ^{57}Co parent nucleus as probes means that the data obtained here can be related not only to the lifetime of the excited Mössbauer state of ^{57}Fe (10^{-7} s) but also to the nuclear spin-lattice relaxation time of ^{57}Co ($\sim 10^{-2}$ s in Pd). The net result is to make the Mössbauer effect apparently the only method with such a wide "window" of relaxation times accessible to observation. This circumstance is particularly important specifically with respect to spin glasses, where the relaxation spectrum can stretch from 10^{-13} s up to minutes or hours.

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