Nuclear spin-latice relaxation of ^{60}Co in $(Pd_{1-x}Pt_x)Co_1$ at infralow temperatures

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The method of nuclear orientation at low temperatures is used to investigate nuclear spin-lattice relaxation of ^{60}C in palladium- and platinum-based allows with giant magnetic moments. The observed increase of the relaxation rate on going from $Pd_{99}Co_1$ to $Pt_{99}Co_1$ differs radically from the results for Pd and Pt nuclei in pure metals, for which the relaxations are practically equal.

INTRODUCTION

Dilute solutions of Co in Pd and Pt have a number of intersting properties, foremost amnog which is the onset of giant magnetic moments due to the Co impurity atom, and a transition into an ordered (ferromagnetic) state even at low Co concentrations. The existing differences between the values of the giant moments, and also of critical Co concentrations for the onset of ferromagnetism in **Pd**Co and **Pt**Co seem to be more readily quantitative than qualitative. However, investigations of magnetic hyperfine interactions in **Pd**Co and **Pt**Co have revealed a more substantial difference between these systems, as manifested in opposite signs of the magnetic hyperfine field at the Co nucleus—positive in Pd and negative in Pt.[1-3] This experimental fact has so far not been satisfactorily explained.

The magnetic hyperfine field for $3d$ atoms is determined primarily by polarization of the core, which makes a negative contribution to the field. The positive contribution that predominates for Pd can be attributed both to polarization of the outer $4s$ electrons (conduction electrons) and to the orbital d-moment.[4] The same effects contribute also to the spin-lattice relaxation (SLR).[4]

SLR in metals can be characterized by one parameter—the Korringa constant C_K, which is connected at thigh temperatures T with the SLR time T_1 by the simple relation $C_K = T_1T$. The main contributions to the SLR are made by contact interaction with the conduction electrons and the core electrons (C_c), the orbital momentum of the d electrons (C_{orb}), and the indirect interaction with the conduction electrons via spin waves (C_{sw}). The resultant value of the relaxation constant (its reciprocal characterizes the relaxation rate) is defined by the relation

$$C_K^{-1}=C_c^{-1}+C_{orb}^{-1}+C_{sw}^{-1}+\dots \qquad (1)$$

We have undertaken a study of the SLR of ^{60}Co in the ferromagnetic alloys $(Pd_{1-x}Pt_x)_{99}Co_1$ with $x = 0$, 0.2 and 1 by the method of nuclear orientation at low temperatures, since an investigation of SLR as a dynamic process can yield additional information on hyperfine interactions.

SLR IN SYSTEMS OF ORIENTED RADIOACTIVE NUCLEI

The quantity observed in nuclear-orientation experiments is the angular-distribution function $W(\theta,t)$ of the radiation from the nucleus. For the case of axial symmetry, the γ-radiation angular-distribution function is

$$W(\theta,t)=\sum_{k=0,2,\dots} A_kB_k(\rho_m(t))P_k(\cos\theta), \qquad (2)$$

where θ is the angle between the nuclear-orientation axis and the detection direction, A_k are parameters determined by the nuclear decay scheme, and P_k (cos θ) are Legendre polynomials. The orientation functions

$$B_k=[(2I+1)(2k+1)]^{1/2}\sum_m(-1)^{I+m}\begin{pmatrix} I & I & k \\ m & m & 0\end{pmatrix}\rho_m \qquad (3)$$

depend on $2I + 1$ relative populations ρ_m (I is the nuclear spin). In the equilibrium state, the parameters ρ_m are determined by the Boltzmann distribution $\rho_m(0) \propto \exp(-E_m/kT_L)$, where E_m is the energy of the mth sublevel of the nucleus and T_L is the crystal-lattice temperature ($\Sigma\rho_m = 1$).

If the lattice temperature is changed, the populations of the nuclear sublevels also change and the nuclear spin system approaches the equilibrium state corresponding to the new temperature. Assuming a weak interaction between the nuclear spins, the populations $\rho_m(t)$ are described by the equations

$$\frac{d\rho_m}{dt}=\sum_{m'}(W_{m'm}\rho_{m'}-W_{mm'}\rho_m). \qquad (4)$$

The probabilities of the transitions between the sublevels $W_{m'm}$ differ from zero only if $m' - m = \pm 1$

$$W_{m+1,m}=\frac{\Delta E}{2kC_k}[I(I+1)-m(m+1)]/[1-\exp(-X_L)],$$
$$W_{m,m+1}=W_{m+1,m}\exp(-X_L). \qquad (5)$$

Here $X_L = \Delta E/kT_L$, $\Delta E = E_{m+1} - E_m > 0$ (Ref. 4).

For the case of pure magnetic dipole interaction, the nuclear sublevels are equidistant with $\Delta E = \mu B_{hf}/I$, where μ is the nuclear magnetic moment and B_{hf} the magnetic hyperfine field. Equation (4) can be written in the form

$$d\rho/dt=R\rho, \qquad (6)$$

where R is a three-diagonal matrix with elements

$$R_{mm'}=\begin{cases} W_{m'm}, & m'\neq m, \\ -\sum_{l\neq m}W_{ml}, & m'=m. \end{cases}$$

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The solution of (6) is

$$\rho(t)=e^{Rt}\rho(0), \tag{7}$$

or in different form

$$\rho_m(t)=\rho_m(0)+\sum_{i=1}^{2I} a_{m_i}\exp(\lambda_i t), \tag{8}$$

where λ_1 are the eigenvalues of the matrix R and $\rho_m(0)$ is the population of the unperturbed (initial) state. We see thus that the populations $\rho_m(t)$, and accordingly $B_k(\rho_m)$ and $W(t)$, are in general multiexponential functions of the time. This means that the SLR cannot be described by a single exponential (with one time constant $\lambda = 1/T_1$). This multiexponential process can be described, however, by a single relaxation constant C_K.

EXPERIMENT

Samples of the investigated alloys were activated by two different methods: neutron irradiation in a reactor and thermodiffusion followed by remelting. The finished samples were foils 1.2–8 μm thick and $\approx 1.5\times 2$ mm in area, with ^{60}Co activity 0.07–0.18 MBq. The samples were annealed in a vacuum at 800 °C directly prior to the experiment.

The SLR was measured in the present study by a modified variant of the thermocycling method proposed by Klein.[5] The method is based on rapidly changing the crystal-lattice temperature from one value to another by successive switching the high-frequency heating on and off. The spin-system relaxation to the new lattice temperature is determined from time dependence of the angular anisotropy of the τ radiation of the oriented ^{60}Co nuclei. The measurements were performed with the SPIN setup.[6]

Figure 1 shows a diagram of the heat exchanger + sample unit. The heat exchanger 1, made of sintered silver powder, is connected by silver wire 2 to a small silver base 3 to which investigated sample was secured with tin-lead solder. The heat exchange with the sample, in a teflon holder, were placed in the chamber of ^{3}He–^{4}He dilution refrigerator. An rf field (10 MHz) was produced by coil 4 at the location of the base with the sample. An external magnetic field B_{ex} produced by a pair of superconducting coils was applied parallel to the sample plane. The system parameters were chosen such that the heat exchanger was at the dilution-chamber temperature during the entire cycle, and a temperature change ocurred only on the base. This is accomplished by making the heat-exchanger area quite large (more than 10 m^2), so that the Kapitza resistance at the boundary between the heat exchanger and the liquid can be neglected compare with the thermal resistance of the silver wire (R_w).

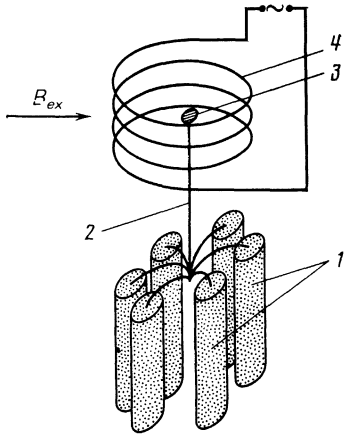

FIG. 1. Diagram of heat exchanger + sample unit for the measurement of nuclear SLR (the callouts are explained in the text).

The decisive condition of the experiment is the relation $\tau \ll T_1$, where τ is the temporal thermal constant of the base + heat exchanger system and T_1 is the characteristic relaxation time of the nuclear spins. This condition imposes an upper limit on the thermal resistance R_w, since $\tau = R_w c$, where c is the specific heat of the base. At low temperatures (when the lattice specific heat is low) c consists of the specific heat c_e of the conduction electron and the nuclear specific heat c_N. The decisive contribution to c of our sampes is made in the working temperature range by the nuclear specific heat of 1 at.% ^{59}Co. To decrease the specific heat it is therefore necessary to use small samples. In our case $R_w \approx 10^4$ K/W, $c_e \approx 2\cdot 10^{-9}$ J/K, and $c_N = (1\text{–}5)\times 10^{-7}$ J/K (at $T = 15$ mK]. This leads to the estiate $\tau \approx 1 - 5$ ms. To verify experimentally this estimate we used a ^{54}MnCu sample (the so-called fast thermometer) with a characteristic relaxation time $\approx 10\,\mu$s at 30 *mK* (Ref. 7). This sample was soldered to the base of the heat exchanger together with an inactive $Pt_{99}Co$ serving as a thermal ballast. Measurement of the time dependence of the angular anisotropy of the τ rays from the oriented ^{54}Mn nuclei (in an external magnetic field 1.2 *T*) yielded $\tau \approx 1$ *ms*, in agreement with the calculated estimate.

RESULTS AND DISCUSSION

We first measured the magnetic hyperfine fields of the investigated alloys B_{hf} at ^{60}Co. The samples together with a ^{54}MnNi nuclear thermometer were first soldered to the heat exchanger with expanded surface and placed inside the dilution-refrigerator chamber. The angular anisotropy of the ^{60}Co γ rays was measured at a fixed temperature (in the range 1–40 mK) at an angle $\theta = 0°$. Expression (2) for the stationary case and $\theta = 0°$ is of the form

$$W(0)=1+F_2U_2Q_2B_2(T)+F_4U_4Q_4B_4(T), \tag{9}$$

where F_k are parameters determined by the characteristics of the γ transition, U_k are the disorientation parameters due to preceding unobserved transitions, and Q_k is the correction for the finite solid angle of the detector. Since all the nuclear parameters of ^{60}Co are well know, measurements of the temperature dependence of $W(0)$ yields B_{hf}. The value of $W(0)$ was defined as the ratio of the counting rate at low temperature to that at ≈ 600 mK, when the radiation is isotropic. The γ photons were recorded with a Ge (Li) detector directed along the external magnetic field that determines the nuclear-orientation axis. Measurements in external fields 0.2–1.2 *T* have revealed the existence of magnetic saturation at $B_{hf} > 0.5$ *T*. The obtained hyperfine magnetic fields listed below agree with published values.[8]

x	0	0,2	1
$B_{\text{hf}}(T)$	21,6±0,5	10,5±0,4	−19,3±0,5
C_{K} (с·К)	(4,9±0,3)·10^{-3}	(4,6±0,8)·10^{-4}	(9,6±0,8)·10^{-5}

To improve the statistical accuracy, the γ photons were

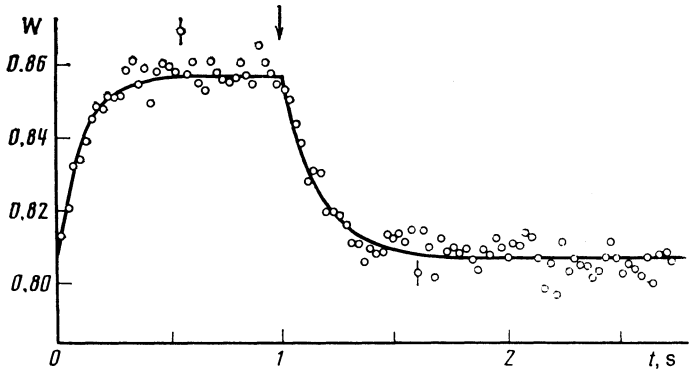

FIG. 2. Time dependence of the angular anisotropy of the γ radiation of ^{60}Co in $Pd_{99}Co_1$ ($B_{ex} = 1.2$ T, $T_i = 16.2$ mK, $T_f = 19.3$ mK). The experimental points are the results of summing $6 \cdot 10^4$ cycles.

recorded in the SLR measurements with an NaI(Tl) scintillation detector crystal of 150 mm diameter and 100 mm thickness. The rf-heating pulse duration and the off-duty cycle were chosen such that the nuclear spin system managed to reach equilibrium with the latice during the time of the rf pulse duration and in the interval between the pulses. The pulse power must be sufficient to obtain on the sample-carrying base a temperature jump that ensures an acceptable signal/noise ratio. On the other hand, the sample must not be excessively overheated by the rf field, for this lowers the accuracy with which the final temperature of the nuclear system is determined. The average power (which can be regulated by varying the off-duty cycle), should not be too high, to prevent an excessive temperature rise in the dilution-refrigerator chamber (i.e., to prevent a rise of the initial temperature).

Each thermal cycle begins at the instant when the rf heating is turned on. Synchronized with this instant is the start of the multichannel analyzer that sums the information of the successive thermal cycles.[9] Figures 2 and 3 illustrate the obtained time dependences of the anisotropy of the γ radiation of ^{60}Co in $Pd_{99}Co_1$ and $Pt_{99}Co_1$ in a 1.2 T external magnetic field. The rf heating was turned on at $t = 0$, and the instant of its shutoff is marked by an arrow in Fig. 2 or 3.

The experimental results were reduced by approximating with a multiexponential dependence with one parameter—the Korringa constant C_K (solid lines in the figures). The initial (T_i) and final (T_f) temperatures needed for the calculation were determined from the equilibrium sections of the time spectrum. Typical experimental values are $T_i = 15$–18 mK and $T_f = 19$–25 mK. The obtained values of C_K were listed above.

Attention is called to the large difference between the values of C_K of ^{60}Co in $Pd_{99}Co_1$ and $Pt_{99}Co_1$. This difference cannot be due solely to the difference in the density of the electronic states (ESD) near the Fermi level of the Pd and Pt matrices). According to present notions[10] the SLR rate in metals is proportional to the square of the ESD. In fact, in Pd and Pt matrices whose ESD are approximately equal [2.281 eV^{-1} (Ref. 11) and 19.85 eV^{-1} (Ref. 12), respectively], the SLR rates of the nonmagnetic impurities ^{45}Sc (Ref. 13), ^{51}V (Ref. 14), ^{93}Nb (Ref. 14) and ^{103}Rh (Ref. 15) hardly differ (the SLR rates for the given atom are compared in the two matrices). Nor are the relative SLR rates $1/\gamma_N{}^2 C_K$ is the nuclear gyromagnetic ratio) of the matrix nuclei (^{105}Pd and ^{195}Pt), different in Pd and Pt, being $\approx 0.9 \cdot 20^{-14}$ s·T^{2}K^{-1} (Ref. 16) and $1.0 \cdot 10^{-14}$ s·T^{2}K^{-1} (Ref. 17), respectively. We see that this is not so for the magnetic impurity Co.

The difference between the values of C_K of ^{60}Co in **Pd**Co and **Pt**Co cannot be attributed to the presence of the orbital d-momentum of Co in Pd. Thus, the assumption that an unquenched (fully or partially) orbital momentum of Co is the source of the positive hyperfine field in Pd seems implausible. The causes of the acceleration of the Co relaxation in $(Pd_{1-x}Pt_x)_{99}Co_1$ with increase of x, and also of the reversal of the sign of B_{hf} on going from Pd to Pt, must apparently be sought in the singularities of the local interaction of Co with the matrix atoms that surround it. Calculations of the hyperfine field and of the SLR time, based on consideration of local interactions of the impurity atoms and the matrix atoms, were recently performed for nonmagnetic impurities and only for iron and nickel matrices (see, e.g., Ref. 18).

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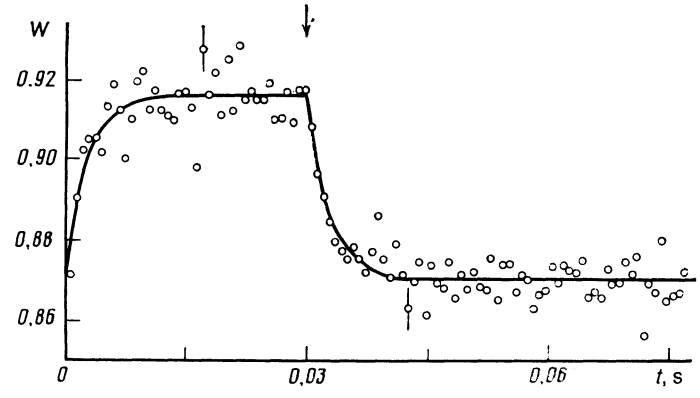

FIG. 3. Time dependence of the angular anisotropy of the γ radiation of ^{60}Co in $Pt_{99}Co_1$ ($B_{ex} = 1.2$ T $T_i = 17.4$ mK $T_j = 22.8$ mK). The experimental points are the result of summation of about $8 \cdot 10^4$ cycles.

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[4)]The positive hyperfine field observed at Co in Pd can be explained by assuming that the coupling of the Co atom with its surrounding Pd atoms is antiferromagnetic. The magnetic moment of Co is then antiparallel to the sample magnetization and the hyperfine field is negative to the Co moment.

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