# "Magnetic anomaly" of the probability of the Mössbauer effect in dilute Pd-Co alloys

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The change of the $\gamma$ resonance absorption probability in a magnetic transformation is investigated by the Mössbauer effect for impurity tin introduced in small amounts in Pd-Co ($x$) solid solutions ($0.3 \leq x \leq 5.0$ at.%). It is shown that the increase of the area of the experimental absorption spectra as the alloys go over into the ferromagnetic state is determined to a large degree by the change produced in the phonon spectrum by spontaneous magnetostriction. An estimate of the Debye temperature of the alloys in the ferromagnetic state yields the value $(310 \pm 10)$°K.

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Dilute solid solutions of Pd with 3d metals have attracted attention because of the peculiarities of their magnetic properties. These alloys are characterized by the appearance of ferromagnetic order at a very low magnetic-impurity concentration (0.005 at.% Fe,[1] 0.03 at.% Co[2]), and also the onset of "giant localized moments" that are several times larger than the moments possessed by the d shell in pure 3d metals. Experiments on neutron scattering by dilute Pd-Co alloys and Pd-Fe alloys[3] have made it possible to attribute the existence of giant moments to the presence of clusters, i.e., regions of polarized Pd atoms surrounding each atom of the magnetic impurity. Investigation of solid Pd-Co solutions with Sn impurity[3,4] have revealed a number of interesting features of the magnetic hyperfine interaction for the Co and Sn atoms. At low Co concentration in Pd, the effective magnetic field at the Sn-atom nuclei increases monotonically with increasing Co content,[4] whereas the field at the nuclei of the Co atoms decreases monotonically.[2,5]

It was of interest to obtain data on the lattice dynamics of such alloys. To this end we used the Mössbauer effect on impurity tin atoms introduced in small amounts in Pd-Co solid solutions to investigate the change of the probability f′ of $\gamma$-resonant absorption when the alloys go over into the ferromagnetic state. The measurements were made at fixed temperatures, 80 and 4.2°K.

The Curie temperatures $T_C$ of the alloys varied in a wide range with changing concentration of the magnetic impurity Co. According to the theory[6] and the experimental data,[7] the value of f′ and its temperature dependence for Sn impurity atoms in monatomic metal matrices are determined by the Debye Waller factor if the Debye temperature of the matrix is replaced by the effective Debye temperature:

$$\Theta_{\text{eff}} = \Theta_D (M/M_{\text{imp}})^{1/2},$$

where $\Theta_D$ is the Debye temperature of the matrix, M the mass of the matrix atom, and $M_{\text{imp}}$ is the mass of the impurity atom.

Measurements with Sn impurity atoms in binary solid solutions (e.g., Pd-Ag) have shown[8] that in this case, too, the temperature dependence of the resonant-absorption probability is determined by the parameter $\Theta_{\text{eff}}$. We shall show below that an analogous one-parameter description of f′ holds also for a small Sn impurity in a Pd-Co matrix.

We prepared for the measurements Pd-Co solid solutions with Co contents 0.3, 0.5, 1.0, 1.6, 2.0, 2.5, 3.0, 4.0, and 5.0 at.%. The procedure used to prepare alloys and to monitor their composition was described earlier.[4,9] For investigations with the aid of the Mössbauer effect, we introduced in the obtained solid solutions a small amount of Sn (0.12 at.%) enriched with $Sn^{119}$ (88.7%). It was ascertained earlier[4] that the small amount of tin does not affect the magnetic properties of these alloys. The samples were rolled to form a foil and annealed in a vacuum for 5 hours at t = 900°C. At room temperature, when all the alloys were paramagnetic, measurements of the Mössbauer effect in annealed samples have shown that the areas of the absorption curves agree with accuracy not worse than 3%.

Our measurements of the absorption spectra were made with both a single-channel and a multichannel spectrometer, with the compound $SnO_2$ or $CaSnO_3$ as the source. The spectra of the alloys in the ferromagnetic state were broadened line with unresolved hyperfine structures. The shift of the spectrum when the alloys went into the ferromagnetic state did not exceed 0.03 mm/sec. The experimental data were reduced with a BÉSM-4 computer. The areas of the experimental absorption spectra were determined by numerical integration of the resonance curves.

The results obtained for alloys with different contents of the magnetic impurity Co are shown in Fig. 1. We see that as the alloys go over into the ferromagnetic state the areas of the experimental absorption spectra become appreciably larger. The figure shows also that at T = 80°K, when some of the alloys Pd—Co(x)—Sn(0.12)[1)] were paramagnetic, the values of $S_{\text{exp}}$ for these alloys agreed within the limits of errors with the analogous value for the paramagnetic alloy Pd-Sn(0.12), the value of which f′ is known.[7]

The observed increase of $S_{\text{exp}}$ when the alloys become ferromagnetic can be due to the magnetic field at the nuclei of the Sn atoms, resulting from the hyperfine interaction. To take the possible influence of this field into account, we have calculated the areas of the absorption spectra as functions of the magnetic field H and of the effective absorber thickness $C_a$.

As is known,[10] the area of the absorption curve S is given in the general case by

$$S = \alpha f_a \int_{-\infty}^{+\infty} \{1 - \exp[-C_a \sigma(x)]\} dx, \qquad (1)$$

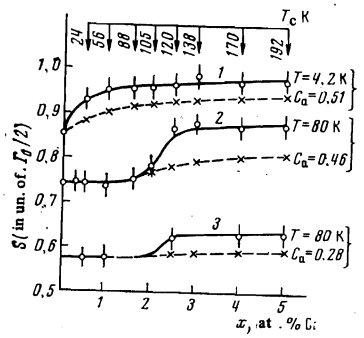


FIG. 1

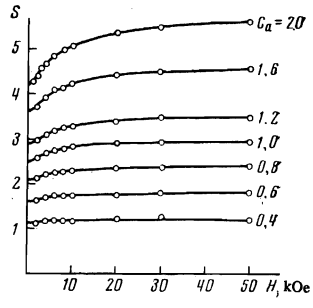


FIG. 2

FIG. 1. Plots of the areas S of the absorption spectra for the alloys Pd–Co(x)–Sn(0.12) vs. the content x of the magnetic cobalt impurity: ○–experimental values, X–calculated values with account taken of the magnetic hyperfine interaction for the tin impurity atoms. The indicated values of $C_a$ pertain to alloys in the paramagnetic state. Curves 1 and 2 were obtained with an $SnO_2$ source (experimental parameter $\alpha f_S = 0.58$), the thickness of the absorbers was 65 ± 2 $\mu$. Curve 3 was obtained with $CaSnO_3$ source (parameter $\alpha f_S = 0.69$), absorber thickness 40 ± 2 $\mu$.

FIG. 2. Calculated values of the area of the absorption spectra (in units of $\Gamma_0/2$) as functions of the magnetic field and of the effective absorber thickness.

where $f_S$ is the probability of recoilless $\gamma$-quantum emission, $\alpha$ is the relative contribution of the resonant quanta to the total counting rate registered by the detector, $\sigma(x)$ is the effective resonant-absorption cross section; $C_a = \sigma_0 f' n_a A$ is a dimensionless parameter, which is the effective thickness of the absorber (here f′ is the resonant-absorption probability, $n_a$ is the number of atoms of the investigated isotope per $cm^3$ of absorber, and A is the geometric thickness of the absorber).

In the presence of magnetic splitting we have for the resonant $Sn^{119}$ (or $Fe^{57}$) nuclei

$$\sigma(x)=\sum_{i=1}^{3}\left[\frac{\beta_i}{(x+\delta_i)^2+1}+\frac{\beta_i}{(x-\delta_i)^2+1}\right],$$

where $\beta_i$ and $\delta_i$ are respectively the relative intensity and position of each of the six components of the hyperfine magnetic splitting. For an unpolarized absorber, the intensities of the components $\beta_i$ are in a ratio 3 : 2 : 1 : 1 : 2 : 3, and their positions $\delta_i$ in the case of resonant $Sn^{119}$ nuclei is given in terms of the magnetic field (kOe) H by the relations ±0.437H, ±0.323H, and ±0.208H kOe.

An exact analytic dependence of S on $C_a$ is obtained only in the case of a single absorption line of Lorentz shape with a natural half-width[10]

$$S=\alpha f_s \pi C_a \exp(-C_a/2)\,[I_0(C_a/2)+I_1(C_a/2)], \qquad (2)$$

where $I_0(x)$ and $I_1(x)$ are Bessel functions of imaginary argument of zero and first order.

In the most general case, the connection between S, H, and $C_a$ can be obtained by numerically integrating (1) with a computer. The results of the computer calculations are shown in Fig. 2 in the form of plots of S against H for various values of the parameter $C_a$ (the experimental parameter $\alpha f_S$ was assumed equal to unity in the calculations). As seen from the figure, for thin absorbers ($C_a < 1$) there should be observed only a small change of the area. In the case of relatively large effective thicknesses of the sample ($C_a > 2$), the presence of the magnetic field can greatly influence the experimental area $S_{exp}$. This is precisely why all the measurements were made on sufficiently thin samples with $C_a \lesssim 0.5$.

We have previously obtained the dependence of the effective magnetic field at the nuclei of the Sn atoms on the Co concentration in the alloys Pd–Co(x)–Sn(0.2). If we use these data on plot on Fig. 1 the calculated values that take into account the influence of the hyperfine interaction (marked by crosses on the figure), then we see that the calculated curves lie much lower than the experimental ones. This indicates that the increase of $S_{exp}$ when the alloys go over into the ferromagnetic state cannot be due to the hyperfine interaction only It appears that this increase must be ascribed to the change produced in the phonon spectrum by the magnetic transformation.[11] Using formula (2) and the results of the calculations, we calculated from the data on $S_{exp}$ the values of the parameter $C_a$ and then of f′ for Pd-Co alloys in the paramagnetic and ferromagnetic states.

Knowing f′, we can estimate the Debye temperature of the alloys in the ferromagnetic state if we assume for the Debye temperature of the paramagnetic alloy the value $\Theta_D = (271 \pm 2)°K$.[12] In this case the Debye temperature of the ferromagnetic alloy turns out to be $(310 \pm 10)°K$, i.e., its relative change when the alloy goes over into the ferromagnetic state is ~14%. It appears that the increase of the Debye temperature of Pd-Co solid solutions as they go over into the ferromagnetic state is due to the spontaneous magnetostriction.

If this assumption is correct, then the change of $\Theta_D$ of the alloy during the magnetic transition should be associated with a change of its Curie point under pressure. Using a pressure up to 10 kbar, we investigated the influence of hydrostatic compression on the Curie point of Pd-Co alloys. It was found that in the Co concentration range from 3 to 5 at.% we have $dT_c/dP \approx (+0.4 \times 10^{-3})$ deg/bar. The observed change of $\Theta_D$ and the value of $dT_c/dP$ are in good agreement with estimates based on the data of Belova and Nikolaev,[13] who considered the question of the influence of the magnetic transformation on the $\gamma$ resonance with allowance for the spontaneous volume magnetostriction.

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[1)]The quantities in the parentheses represent the impurity contents in at. %.

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