The effect of pressure on the Curie point of dilute Pd–Co alloys

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The effect of hydrostatic pressure (of up to 12 kbar) on the Curie temperature (Θ_c) of solid Pd-Co(x) solutions ($1.55 \leq x \leq 9.6$ at.%) has been investigated. It has been found that, for these alloys, the quantity $d\,\Theta_c/dP$ is positive and increases from 2×10^{-4} to 5×10^{-4} deg/bar as the cobalt concentration is increased from 1.55 to 9.6 at.%. The changes in the Debye temperatures of the Pd-Co alloys upon their transition into the ferromagnetic state have been estimated from the $d\Theta_c/dP$ values. The estimates agree with the Debye-temperature changes observed in γ-resonance experiments on impurity tin atoms in solid Pd–Co solutions.

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A small addition of cobalt to palladium leads, as is well known, to ferromagnetism in this system at low temperatures.[1] Recently, it was demonstrated with the aid of the Mössbauer effect on impurity tin introduced in small quantities into solid Pd–Co(x) solutions ($0.3 \leqslant x \leqslant 5$ at.%) that, upon the transition of the solid solutions into the ferromagnetic state, their Debye temperature increases by more than 10%.[2] It might have been supposed that such an increase in Θ_D is largely determined by the change in the phonon spectrum due to the spontaneous volume magnetostriction. If this assumption is correct, then the change that occurs in the Θ_D of the alloy during the magnetic transition can be related to the change in the Curie temperature Θ_c of the alloy under pressure. In this connection, the effect of hydrostatic pressure on the Curie temperatures of solid Pd–Co solutions was investigated in the region of low Co concentrations.

For the measurements we prepared solid Pd–Co solutions with the following cobalt contents (in at.%): 1.55; 2; 4; 4.7; 9.6; and we also used Pd–Co(4.0)–Sn(0.12) and Pd–Co(5.0)–Sn(0.12) samples$^{1)}$ that had been prepared earlier for γ-resonance experiments.[2]

As the starting materials we used palladium with a resistance ratio $R_{300}/R_{4.2} \approx 3000$ and cobalt of 99.999% purity. The calculated amounts of the starting materials were alloyed in a high-frequency furnace. Then portions of the obtained samples were rolled into foils. Foil strips of dimensions $60\times8\times0.085$ mm were rolled into cylinders (such a sample shape allowed us to carry out simultaneous measurements of the temperature dependence of the resistance and the magnetic moment of the given alloy). The other portions of the samples were left in the form of small solid balls. All the samples were annealed in a vacuum for 8–10 hours at 950 °C. The values of the Curie points of the samples were determined simultaneously from both the temperature dependence of the magnetic moment and the kink in the electrical conductivity curve. The investigation of the effect of pressure on the Curie points of dilute Pd–Co alloys was carried out in a beryllium bronze bomb. The bomb served at the same time as a thermal unit of large heat capacity. The sample (to be investigated) either in the form of a cylinder rolled from a strip of foil, or in the form of a small solid ball was placed in a fixed location in the active volume of the bomb. A Cu–Cu (0.1 at.% of Fe) thermocouple was fixed directly to the sample. The working medium for pressure transmission in the bomb was a mixture of dehydrated transformer oil and kerosene. The pressure was determined from the resistance of a manganin-wire manometer.$^{2)}$ After the production of the pressure, the bomb with the sample was placed in a cryostat and cooled. The measurement of the temperature dependence of the magnetic moment of the sample was carried out in a constant external magnetic field of intensity 50 Oe.

Figure 1 shows typical dependences of the magnetization and resistance of a sample on temperature for two cases: before and after the application of pressure. It can be seen that in each case the Curie temperatures found from the kink in the electrical conductivity curve and from the data on the magnetization coincide with each other. In the range of pressures used (up to 12 kbar), the Curie temperatures of the alloys increased in direct proportion to the applied pressure (a typical dependence is shown in Fig. 2).

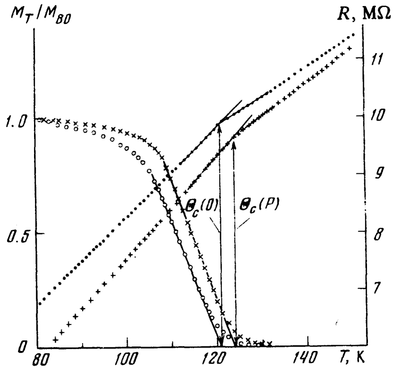

FIG. 1. Temperature dependence of the resistance, (●, +), and of the magnetic moment, (○, ×), for a Pd–Co alloy (2.4 at.%) (simultaneous measurements): ○), ●) without hydrostatic pressure; ×), +) under a pressure ~8.0 kbar. The magnitude of the moment of the sample at any temperature has been expressed relative to the moment at 80 K.

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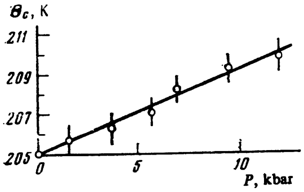

FIG. 2. The variation of the Curie temperature of a Pd–Co alloy (4.7 at.%) under hydrostatic pressure.

The data obtained for the dependence of the quantity $d\Theta_c/dP$ on the cobalt content in the investigated alloys are presented in Fig. 3. It can be seen from the figure that the quantity $d\Theta_c/dP$ is positive for all the investigated alloys and depends essentially on their composition. According to the data obtained by Holzapfel *et al.*[4] for Pd–Co alloys in the region of relatively high cobalt concentrations (from 5 to 15 at.%), the quantity $d\Theta_c/dP$ is also positive. However, it was conjectured in the indicated investigation that, for alloys with cobalt concentrations less than 3 at.%, we should expect a change in the sign of $d\Theta c/dP$. As can be seen from Fig. 3, such a change does not occur, at least down to a cobalt concentration of 1.5 at.%. Furthermore, the $d\Theta_c/dP$ values obtained by us for the 5- and 9.6-at.% Pd–Co alloys exceed almost by a factor of two the values obtained in[4] for analogous alloys.

One of the causes of such a discrepancy is, apparently, the nonlinear dependence of the change in the Θ_c of an alloy when measured in a wide range of pressures (in[4] the minimum applied pressure was more than 20 kbar). Another cause could be the distinctive features of the experimental procedures. For the measurement of the Θ_c of the Pd–Co alloys, Holzapfel *et al.* used the Mössbauer effect (the temperature-scanning method). In this case into the Pd–Co matrix was introduced a small quantity of Fe^{57}. But it is known[5–7] that the Curie temperature in the Pd–Fe system decreases with pressure. Thus, the $d\Theta_c/dP$ values for Pd–Co alloys with an Fe^{57} impurity could prove to be too low.

Also noteworthy is the fact that the behavior of dilute Pd–Fe alloys under pressure differs significantly from the behavior of Pd–Co alloys. For Pd–Fe alloys the quantity $d\Theta_c/dP$ is negative and is an order of magnitude smaller in absolute value than for Pd–Co alloys. According to the data obtained by Fawcett *et al.*,[6] the resistance of Pd–Fe alloys under pressure increases. At the same time, for dilute Pd–Co alloys, as follows from Fig. 1, hydrostatic pressure leads to a decrease in the resistance.

The data of the present measurements (Fig. 3) show that the presence of a small tin impurity in the Pd–Co matrix does not, apparently, affect the $d\Theta_c/dP$ value for the dilute alloy. In the region of cobalt concentrations of 3 to 5 at.%, the quantity $d\Theta_c/dP = 4\times10^{-4}$ deg/bar. Using these data and Belova and Nikolaev's results,[8] we can estimate the change that can occur in the Debye temperature of the alloys as a result of the magnetic transition.

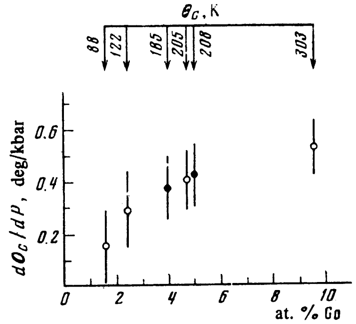

FIG. 3. Dependence of the quantity $d\Theta_c/dP$ on the cobalt content in palladium. The points ● are data for Pd–Co alloys with a tin impurity (0.12 at.%).

According to the indicated investigation, for a face-centered cubic lattice, the change in the Debye temperature due to a magnetic transition can be represented in the form

$$\frac{\Delta\Theta_D}{\theta_D} \approx \left[\left(\frac{1}{16Ms_t^2} + \frac{4\gamma}{\mathscr{K}a^3}\right)\frac{a}{J}\frac{\partial J}{\partial a} + \frac{1}{48Ms_t^2}\frac{a^2}{J}\frac{\partial^2 J}{\partial a^2}\right]U_{\text{exc}},$$

where $\Delta\Theta_D = \Theta_p - \Theta_f$ is the change occurring in the Debye temperature of the crystal as a result of the transition from the paramagnetic state into the ferromagnetic state, γ is the Grüneisen constant (for pure palladium, $\gamma = 2.45$[9]), $\mathscr{K}$ is the bulk modulus, s_t is the transverse sound velocity, a is the lattice constant, J is the exchange integral, and $U_{\text{exc}} \sim k\Theta_c$ is the mean exchange energy. Assuming direct proportionality between the Curie point Θ_c and the exchange integral, we can write[8]

$$\frac{a}{J}\frac{\partial J}{\partial a} = -3\mathscr{K}\frac{1}{\Theta_c}\frac{\partial\Theta_c}{\partial P}.$$

Thus, the change in Θ_D connected with the derivative $\partial J/\partial a$ can be represented in the form

$$\frac{\Delta\Theta_D}{\Theta_D} \approx -\left(\frac{1}{16Ms_t^2} + \frac{4\gamma}{\mathscr{K}a^3}\right)3\mathscr{K}k_B\frac{\partial\Theta_c}{\partial P}.$$

The estimate made in this way shows that we can expect a 4–6% increase in the Debye temperature upon the transition of the alloy into the ferromagnetic state. Taking into consideration the approximate character of the formula used in the estimation, we can consider the obtained result to be in sufficiently good agreement with the data obtained in the Mössbauer investigations, thus confirming the important role played by magnetostriction.

In conclusion, the authors consider it their pleasant duty to express their gratitude to A. P. Kir'yanov, V. M. Belova, V. I. Nikolaev, I. Ya. Korenblit, and E. F. Shender for useful discussions.

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[1] In brackets everywhere we indicate the impurity amounts in atomic percent.

[2] According to Itskevich,[3] in this case the magnitude of the pressure in the bomb can be found from the relation $P = \Delta R/KR$, where R is the resistance of the manometer before the production of the pressure, ΔR is the change in the resistance of the manometer under the action of the pressure, and K is a coefficient equal, on the average, to 2.5×10^{-6} bar^{-1} at all temperatures above 80 K.

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Effect of dispersionless phonons on the kinetics of electrons in one-dimensional conductors

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A theory is constructed for the influence of dispersionless phonons on the diffusion and localization in one-dimensional systems. The conductivity of perfect one-dimensional crystals, is described by the Drude formula, as a result of exact cancellation of an extensive class of non-ladder diagrams. In imperfect crystals (impurities, structural disorder), the dispersionless phonons do not disturb the localization, and the localization length and form factor, as well as the dielectric constant, are preserved. The low-frequency asymptotic behavior of the conductivity, however, becomes different, taking the form $\omega^2\ln^3\omega$ instead of $\omega^2\ln^2\omega$. In addition, if the free path times for scattering by impurities and phonons (τ_i and τ_{ph}) satisfy the inequality $\tau_i \gg \tau_{ph}$, then the characteristic time that limits the region of the low-frequency asymptotic behavior is no longer the free-path value τ_i but the diffusion value τ_i^2/τ_{ph}. The results are valid in the region of relatively low temperatures $T \ll \omega_0$(ω_0 is the phonon frequency) and in the region of weak scattering $\tau_i T \gg 1$ and $\tau_{ph} T \gg 1$. The considered model is apparently of greatest importance from the point of view of the interaction of electrons with fully-symmetrical intramolecular phonons, for which the frequencies are quite high so that the upper bound on T is not very stringent. We conclude with a qualitative examination of the conductivity under certain other conditions.

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1. INTRODUCTION

The main singularity of the electron spectrum of one-dimensional disordered systems is that all the states are localized in them.[1] Therefore such systems have no static electric conductivity or diffusion.[2,3] These statements are valid for real systems only at zero temperature, when the electrons interact only with the static potential of the impurities.

At finite temperatures real phonons produce for the electrons a time-dependent potential that gives rise to transitions between the electron states with different energies. By the same token, the interaction with the phonons differs radically from the interaction with the impurities, which leads to localization of the electrons on account of the exact interference of waves scattered by different impurities. One should expect the change of the energy in scattering by phonons to disturb the interference and to lead to qualitatively new effects.

In our preceding paper[4] we considered the influence of weak electron-phonon interaction on the kinetics of electrons localized in an impurity field. The conditions were indicated under which the interaction with the phonons causes transitions between localized states of the electrons, disturb the localization partially, and by the same token give rise to a finite conductivity. In this case the electron motion consists of hops over distances on the order of the localization length in the impurity field, which follow each other in intervals on the order of the time of scattering by the phonons.

Gogolin *et al.*[5] have indicated the mechanisms through which the interaction with the phonons can intensify the localization of the electrons. These mechanisms in conjunction with the previously obtained results[4] have made it possible to propose an explanation for the experimental data on the temperature dependence of the electric conductivity and the dielectric constant of TCNQ salts with structural disorder.

Thus, the influence of the phonons on the electron kinetics in one-dimensional conductors is manifold—they can disrupt as well as enhance the localization. To determine the relative importance of the different types of electron-phonon interaction, it is therefore necessary