FERROMAGNETISM IN DILUTE SOLUTIONS OF GADOLINIUM IN PALLADIUM

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Ferromagnetism sometimes occurs when small amounts of transition elements are dissolved in metals of high magnetic susceptibility. The outstanding examples are the dilute solutions of Fe or Co in Pd,[1,2] where giant magnetic moments occur due to the polarization of the matrix in the vicinity of the impurities. In the most dilute of these alloys the range of the ferromagnetic coupling seems to extend over distances several times greater than the interatomic distance.

In view of the fairly recent discovery[3,4] that rare earths form solid solutions to a limited extent in palladium, it is obviously of interest to investigate magnetic coupling effects in these alloys. Peter et al.[3] have reported that in an alloy $Pd_{97}Gd_3$ the dependence of magnetic susceptibility on temperature is indicative of a paramagnetic Curie temperature of +12°K, suggesting the possible existence of a ferromagnetic state at lower temperatures.

A set of Pd-Gd alloys was made up by arc melting appropriate quantities of specially refined pure palladium and Johnson Matthey gadolinium together under very clean conditions in an argon atmosphere. Their nominal compositions ranged up to 9.7 atomic percent of Gd. As yet they have not been analyzed chemically. However, there was very little loss of total weight during alloy preparation. That true solid solutions occur in every case was verified by x-ray diffraction analysis. Over the whole range up to 9.7 atomic percent, the lattice parameter increased smoothly and continuously with increasing gadolinium content. The synthetic compositions are therefore assumed to be correct to a first approximation. Iron, nickel, or cobalt were not present as impurities in the starting materials.

The magnetizations (σ) of the small ellipsoidal specimens were measured as a function of magnetic field strength (H) at various constant temperatures (T), using a Sucksmith ring balance in the form which allows independent control of field and field gradient.[5] The maximum applied field was about 20 kilo-oersteds and the lowest temperature about 1.5°K. Measurements were relative to the magnetization of pure nickel, to which the absolute data of Weiss and Forrer[6] were assumed to apply. The Curie points were determined by plotting graphs of H/σ against σ^2 for various constant temperatures, and finding by linear interpolation the Curie temperature θ at which the linear graph of H/σ against σ^2 passes through the origin. To determine the magnetic moments the linear high-field parts of the (σ, H) isothermals corresponding to the lowest two temperatures used for each alloy were extrapolated to $H = 0$. These extrapolated values were plotted against T^2 and this graph was extrapolated to $T^2 = 0$ to give the respective saturation magnetizations (at $T = 0$). In converting the saturation magnetizations to numbers of Bohr magnetons, the Bohr magneton was assumed to be equivalent to 5586 emu per gram molecular unit.

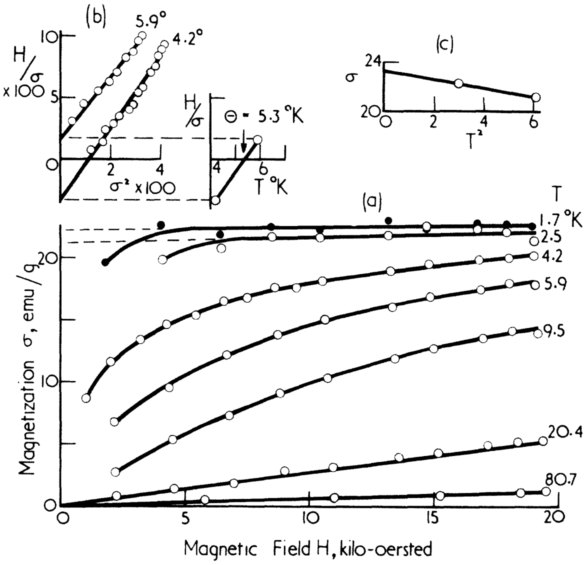

FIG. 1. Experimental data for 7.2 atomic percent PdGd alloy. (a) The magnetization-field isothermals; (b) the determination of the Curie temperature; and (c) the extrapolation by which the saturation magnetic moment is determined.

As an example, the data obtained for the 7.2 atomic percent alloy are illustrated in Fig. 1. For the other alloys the general pattern was similar, except that in the most dilute alloys where the Curie points were lowest, fewer useful isothermals could be measured. The Curie temperatures are probably accurate to within ±0.2°K and the magnetic moments to within ±0.5 emu/g.

Figure 2 shows how the Curie points (a) and the magnetic moment per added Gd atom (b) vary with the Gd content of the alloys.

The main features apparent from the data shown in Fig. 2 seem to be the following: (1) Apparently normal ferromagnetism persists in PdGd alloys down to less than 1 atomic percent of Gd, indicative of some kind of long-range interaction between the Gd impurity atoms. (2) The Curie temperatures for the same amount of solute are an order of magnitude smaller in the PdGd alloys than they are in PdFe or PdCo alloys. (3) In all the alloys the observed moment per Gd atom is significantly smaller than the expected number of 7 Bohr magnetons usually associated with Gd atoms. The amount by which the measured value falls short of the free Gd value is shown in Fig. 2(c), where its dependence on the Gd content is plotted.

It is unlikely that this apparent deficiency below the free-gadolinium magnetic moment is due to crystalline electric field effects partially quenching an orbital part of the magnetic moment, because Gd is expected to be in an $^8S_{7/2}$ state with no orbital moment. It is probably due to the spin polarization of the conduction electrons associated with the palladium matrix and located around the Gd impurity atoms being aligned oppositely to the Gd spins. Peter et al.[4] have shown that the Gd g value measured by electron paramagnetic resonance is significantly lower in $Pd_{97}Gd_3$ (at about 20°K) than in Gd, for this reason.

The explanations of the relatively large value of the deficiency and also of the long range of the ferromagnetic coupling implicit in the observation of finite Curie temperatures in alloys containing as little as 1% of Gd are probably to be found in the recent paper by Giovannini, Peter, and Schrieffer.[7] They report that exchange interactions between valence electrons strongly modify the form of the induced polarization surrounding a magnetized impurity dissolved in Pd. Instead of the damped oscillatory form given by the Rudermann-Kittel-Yosida theory for the variation of the polarization with radius from the impurity center, valence-electron exchange interactions cause a displacement so that no reversed polarization occurs at all until the radius is very large. Thus separated Gd atoms in the random alloy are coupled together ferromagnetically when their associated "clouds" of induced polarization overlap, and this occurs consistently over a wide range of separations. And little or no part of the valence-electron polarization is balanced out by opposing effects at different radii.

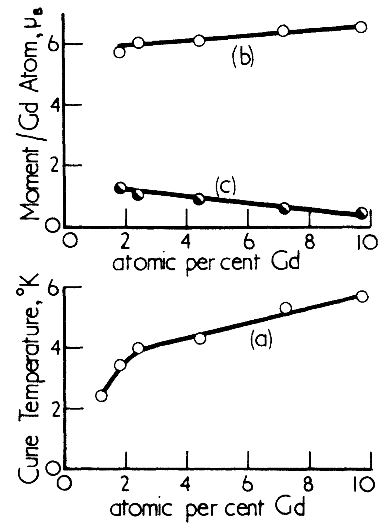

FIG. 2. The dependence on the Gd content of (a) the Curie temperature; (b) the magnetic moment per impurity Gd atom (in Bohr magnetons); and (c) the amount by which (b) is less than the $7\mu_B$ usually associated with Gd.

The Curie points are lower in the PdGd alloys than in PdFe and PdCo possibly because the net moment per solute atom is lower and the interaction therefore weaker. In the PdFe and PdCo alloys the solute and the conduction-electron magnetizations add together.

Ferromagnetism also occurs in other solutions of rare earths in Pd, although with lower Curie points. Further work is in progress on these alloys.

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