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The Effect of Pressure on Fe^{57} Hyperfine Fields in Ferromagnetic *Pd*Co Alloys*

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The Fe^{57} hyperfine fields in a series of ferromagnetic *Pd*Co alloys having cobalt concentrations between 5 and 15 at.% are measured by Mössbauer spectroscopy at 297 °K and pressures up to 180 kbar. The pressure-dependent finite-temperature impurity hyperfine-field problem is parametrized in terms of the pressure dependence of the host Curie temperature, the zero-temperature hyperfine field, and the host-impurity coupling constant. In conjunction with previous data of Holzapfel *et al.* on the pressure dependence of the Curie temperatures, the present data are fitted within the molecular-field approximation with a positively pressure-dependent zero-temperature hyperfine field and a negatively pressure-dependent host-impurity coupling constant, the pressure derivatives being roughly composition independent. Implications of these results are discussed, and a comparison is made with the high-pressure Mössbauer data of Möller and Drickamer on *Pd*Fe alloys in the same composition range. Isomer-shift data are also presented briefly.

I. INTRODUCTION

The dilute alloy systems *Pd*Co and *Pd*Fe have been the subject of much interest in recent years, stemming from the pioneering work of Constant,[1] Gerstenberg,[2] and Bozorth *et al.*[3] on *Pd*Co and Crangle[4] on *Pd*Fe. The interest arises from (i) the "giant" magnetic moment assoicated with each impurity (~ 10 μ_B in very dilute cases) and (ii) the extremely long range (~ 10 Å) of the interaction which couples the impurity moments ferromagnetically. It is now understood that each Co or Fe impurity maintains an on-site magnetic moment when dissolved in the Pd host,[5,6] and because of the highly exchange-enhanced Pd susceptibility each impurity moment is able to polarize the surrounding Pd matrix out to large distances.[6–11] The impurity–polarized-host complex then comprises the giant moment, and the polarized host carries the ferromagnetic interaction which couples the impurities indirectly.[12–14] The dilute systems *Pd*Co and *Pd*Fe are very similar. *Pd*Ni differs in that a Ni impurity cannot sustain a magnetic moment in the Pd host until a critical concentration of ~2-at.% Ni is reached; then the Ni moments appear and the alloy goes ferromagnetic concurrently.[15]

The Mössbauer effect has been used extensively to study these Pd-based alloy systems in both the dilute and nondilute regimes.[16–32] Information has thereby been acquired on the strength and range of the ferromagnetic interactions, the statistical distribution and clustering of impurities, the concentration dependence and statistical smearing of the Curie temperatures, the temperature and field dependences of the bulk-alloy magnetizations, and the nature of the localized-impurity magnetic states and hyperfine interactions. The volume dependence of the magnetic properties of these systems has been investigated in magnetostriction,[33–37] compressibility,[38,39] and pressure-dependent resistance,[33] magnetoresistance,[40] and susceptibility[33,41,42] experiments. The pressure dependence of the Fe^{57} Mössbauer effect has been measured in this laboratory.[43–45]

The present work is concerned with the effects of pressure on the Fe^{57} hyperfine fields, in zero external field at 297 °K, in a series of $Pd_{1-x}Co_x$ alloys ranging from $x = 0.05$ to $x = 0.15$, at pressures up to 180 kbar. The effects are observed to be large, systematic, and nonlinear. The finite-temperature Fe^{57}-impurity hyperfine-field problem is parametrized in terms of five pressure-dependent quantities: the ferromagnetic Curie temperature T_C, the host magnetization σ_0 and local impurity moment μ_0 at $T = 0$, the host-impurity coupling parameter ζ, and the impurity hyperfine coupling constant A. The impurity hyperfine field is seen to sense mainly T_C, $H_0 = A\mu_0$, and ζ, where H_0 is the hyperfine field at $T = 0$. In conjunction with independent measurements of the pressure dependence of T_C for these alloys,[44] it is shown that the present data can be fitted semiquantitatively within the molecular-field approximation with a linearly positively pressure-dependent $|H_0|$ and a linearly negatively pressure-dependent $|\zeta|$, of the same magnitude for all alloys. Under the reasonable assumption that $d\ln H_0/dp$ and $d\ln\zeta/dp$ *must* be roughly composition independent, the above fit is seen to be unique and demonstrates, we believe, the first observation of a pressure-dependent host-impurity coupling constant in a ferromagnetic metal.

A discussion is given of some implications of the above results. It is apparent that the Fe^{57} impurity plays an active role in the effects observed here and does not probe the pressure-dependent magnetization of the host in a simple unobtrusive way. A comparison is made with pressure data for the alloy system *Pd*Fe in the same composition range as above.[45] The pronounced differences observed in the pressure-dependent properties of *Pd*Co and *Pd*Fe are attributed to direct impurity-impurity interactions, indicating that the present concentration regime cannot be considered dilute, as was previously done.[44] Isomer-shift data for the *Pd*Co alloys are also presented briefly.

II. EXPERIMENTAL DETAILS

A. Sample Preparations

The $Pd_{1-x}Co_x$ alloys (nominally 5-, 8-, 12-, and 15-at.% Co) were supplied by the Metal Science Group, Battelle Memorial Institute, Columbus, Ohio. The alloys were fabricated at Battelle by arc melting. Each charge was melted at least six times at 200 A and 30 V, held molten 20–25 sec, and quenched via water-cooled copper hearth. The weight loss in the melting process was less than 1% in each case. The arc-melted buttons were next cold rolled to about 0.1-in. thickness, homogenized in a vacuum furnace at 1125 °C for 24 h under an atmosphere of purified argon, and furnace cooled. There was no measurable weight loss. The samples were cold rolled again to 0.01 in. and were received thus at this laboratory. A $Pd_{0.91}Co_{0.09}$ alloy was fabricated here in an induction furnace from the $Pd_{0.92}Co_{0.08}$ and $Pd_{0.88}Co_{0.12}$ alloys described above. The sample was contained in a recrystallized aluminum-oxide crucible, melted in an argon atmosphere with 13–14 kW of power for 10 min, then 8–9 kW for 1 min, and allowed to cool in the furnace. No weight loss was detected after the melting process.

Since the original alloys displayed somewhat messy Mössbauer spectra with broad lines (which could result from compositional inhomogeneities and a consequent variety of Mössbauer sites), these samples were submitted to additional homogenizing treatment along with that of the newly fabricated $Pd_{0.91}Co_{0.09}$ alloy. (This homogenizing did, in fact, improve the spectra markedly.) Each sample was placed between two recrystallized aluminum-oxide chips and the resulting sandwich wrapped in molybdenum foil so that the sample itself would not contact the molybdenum. Then each bundle was sealed, with an argon atmosphere, in a separate quartz tube. These tubes were heated to 1200 °C in a resistance furnace and maintained there for 212 h, after which they were air then water cooled. The quartz tubes showed no evidence of darkening from sample vaporization. The new $Pd_{0.91}Co_{0.09}$ alloy, now in the form of a button, was cold rolled to 0.01-in. thickness, then annealed in a boat similar to those described above, under argon, at 1000 °C for 5 min and 950 °C for 8 min. It cooled over a period of about 15 min. Flame-emission analysis for Co content at this point confirmed that, to within the accuracy of the measurement ($\Delta c/c \simeq \pm 0.02$), all sample compositions were still nominal.