

## Field-induced reentrant superconductivity in the Heusler alloy system $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$

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The ternary compound  $\text{Pd}_2\text{YSn}$  crystallizing in the cubic Heusler ( $L2_1$ ) structure is superconducting with a superconducting transition temperature ( $T_c$ ) of 4.55 K. The superconducting and magnetic phase diagram of the pseudoternary system  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  ( $x=0-1$ ) has been investigated. The superconducting  $T_c$  of  $\text{Pd}_2\text{YSn}$  decreases with the replacement of Y by Dy and superconductivity is completely suppressed at a critical concentration of Dy corresponding to  $x \sim 0.35$ . The samples with  $x \geq 0.5$  are antiferromagnetically ordered with Néel temperatures ranging from 5 K for  $x=0.5$ —15 K for  $x=1.0$  ( $\text{Pd}_2\text{DySn}$ ). The superconducting samples with  $x > 0$  show reentrant behavior under external applied fields. The temperature dependence of the upper critical field in  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  ( $x=0.0, 0.1, \text{ and } 0.15$ ) has been measured and is analyzed in terms of the Werthamer-Helfand-Hohenberg theory including the pair-breaking effects due to paramagnetic rare-earth ions.

### I. INTRODUCTION

The observation of superconductivity in ternary compounds with rare-earth atoms occupying regular lattice sites has led to many new and interesting phenomena.<sup>1</sup> These include the unusually high upper critical fields in  $R\text{Mo}_6\text{S}_8$  ( $R$  denotes rare earth) compounds,<sup>2</sup> reentrant behavior,<sup>3</sup> coexistent of long-range magnetic order with superconductivity,<sup>1,4</sup> etc. Recently, field-induced superconductivity has been observed<sup>5</sup> in the system  $\text{Eu}_{0.25}\text{Sn}_{0.25}\text{Mo}_6\text{S}_{7.28}\text{Se}_{0.72}$  due to the presence of a negative exchange interaction between the conduction electron spins and the rare-earth spins. The studies on pseudoternary systems between magnetic and superconducting members of the series have been used profitably to get information on both the magnetism and the mechanism of superconductivity. Results of such studies on various systems have been summarized in some recent reviews.<sup>1,2</sup>

The formation of the rare earth containing Heusler-type compounds of the composition  $\text{Pd}_2\text{RSn}$  ( $R = \text{Tb-Lu, Y, and Sc}$ ) has been recently reported in the literature.<sup>6</sup> These compounds crystallize in the cubic  $L2_1$  structure with lattice parameter  $a$  of the order of 6.6–6.8 Å. The rare-earth atoms are located in a cubic environment of Pd atoms and the smallest  $R$ - $R$  separation is  $\sim 4.6$  Å. The  $\text{Pd}_2\text{RSn}$  compounds with  $R = \text{Tm, Yb, Lu, Y, and Sc}$  are superconducting with the Y-containing compound having the highest superconducting transition temperature ( $T_c$ ) of 4.55 K in this series.<sup>6</sup> The Tb and Dy containing compounds studied by us order antiferromagnetically at 9 and 15K, respectively.<sup>7</sup> The Ho and Er containing compounds have also been reported<sup>6</sup> to order magnetically at 2 and 0.7 K, respectively, though our ac susceptibility measurements on these compounds, down to 1.2 K failed to exhibit any transitions. This may be due to slight differences in stoichiometry. It has been shown<sup>7</sup> that the crystalline electric field (CEF) effects are important in Tm and Yb compounds and give rise to a nonmagnetic ground state for the  $\text{Tm}^{3+}$  ion in  $\text{Pd}_2\text{TmSn}$ . The ground state of

$\text{Yb}^{3+}$  ions in  $\text{Pd}_2\text{YbSn}$  has a considerably reduced moment compared to the free-ion value. This enables superconductivity not to be suppressed in Tm and Yb compounds. In fact,  $\text{Pd}_2\text{YbSn}$  shows coexistence of ordered magnetism (presumably of the antiferromagnetic type) with superconductivity below 0.23 K.<sup>6,8</sup>

In this paper we report the results of superconducting and magnetic measurements on the pseudoternary system  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$ . This work was undertaken to study the magnetic and superconducting phase diagram of this system and, in particular, to investigate the effect of magnetic rare-earth ions on the upper critical field of  $\text{Pd}_2\text{YSn}$ . The variation of the superconducting transition temperature as a function of  $x$  as well as the temperature dependence of the upper critical field for various superconducting compositions is presented. For superconducting Dy-containing samples, reentrant behavior is observed under external applied fields. The suppression of  $T_c$  by magnetic Dy impurities is discussed in terms of the Abrikosov-Gor'kov theory.<sup>9</sup> The results of the critical-field measurements are analyzed on the basis of the Werthamer-Helfand-Hohenberg (WHH) theory<sup>10</sup> modified by Fischer<sup>11</sup> to include the effects due to paramagnetic ions. These measurements enable us to estimate the values of  $N(0)J_{sf}^2$  where  $N(0)$  is the density of states per atom per spin direction and  $J_{sf}$  is the exchange constant for exchange interaction between the conduction-electron spins and the rare-earth spins.

### II. EXPERIMENTAL PROCEDURE

The pseudoternary compounds  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  with  $x$  ranging from 0 to 1 were prepared by arc melting stoichiometric amounts of the constituent elements followed by annealing at 800°C for four days. Powder x-ray diffraction patterns obtained between 300 and 5 K revealed that these are single-phase compounds crystallizing in the cubic Heusler ( $L2_1$ ) structure. The superconducting and magnetic transition temperatures were measured

in the range 1.3–30 K by using an ac susceptibility bridge operating at a frequency of 900 Hz. The dc susceptibility measurements were made using a superconducting quantum interference device (SQUID) magnetometer in the temperature range 5–300 K. The upper critical-field measurements were carried out in an apparatus based on the principle of flux penetration depth<sup>12</sup> operating at a frequency of 45 kHz. These measurements were made in a pumped-<sup>3</sup>He cryostat where temperatures down to 0.44 K could be attained and with a continuous ramping magnetic field of up to 50 kOe. The data were taken with the temperature held constant and the field being swept up from zero and then down at a rate of 0.1 kOe/min. The shapes of the transition in zero field as well as in applied fields were comparable within error. Because of the lower temperature capability of the skin-depth measuring apparatus, the data obtained from these measurements have been used for all the analyses reported here. The upper critical field was taken as the field required to drive 90% of the sample in the normal state.

### III. RESULTS AND DISCUSSIONS

The compounds  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  form single-phase solid solution in the entire range from  $x=0$  to 1. The ionic sizes of Y and Dy are nearly the same and hence there are no appreciable changes in the lattice parameter. This is advantageous because the effects of changes in the lattice parameter on superconductivity (i.e., the nonmagnetic effects) are minimal and can be neglected. Further, the crystal structure is cubic and remains so down to 5 K for all the samples except for  $x=1.0$  and hence no anisotropic effects are expected for this series unless the crystalline electric field split ground state of the  $\text{Dy}^{3+}$  ion is a  $\Gamma_8$  quartet. The compound  $\text{Pd}_2\text{DySn}$ , however, shows a structural transformation to a low-symmetry phase below 50 K.<sup>13</sup>

#### A. Magnetic and superconducting phase diagram

As remarked earlier, the compound  $\text{Pd}_2\text{YSn}$  is superconducting with a  $T_c$  of 4.55 K while, from bulk-magnetization studies,  $\text{Pd}_2\text{DySn}$  has been found to order antiferromagnetically with a Néel temperature ( $T_N$ ) of 15 K (14 K from  $\chi_{ac}$ ). Figure 1 shows the complete superconducting and magnetic phase diagram of the system  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$ . The superconducting  $T_c$  of  $\text{Pd}_2\text{YSn}$  drops rapidly on substitution of Y by Dy. On the Dy-rich side, the magnetic ordering temperature decreases in a linear fashion as Dy is replaced by Y. When extrapolated from the superconducting and the magnetic ends, the two lines meet in a very narrow concentration range where superconductivity and magnetic order may coexist. This coexistence range depends very much on the approximations involved in extrapolating from the superconducting end (*vide infra*). The dashed line at 1.2 K parallel to the temperature axis represents the limit down to which we have carried out ac susceptibility measurements on all these samples.

The presence of antiferromagnetic ordering in  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  ( $x \geq 0.5$ ) is also inferred from the susceptibility measurements on these compounds. Figure 2 shows

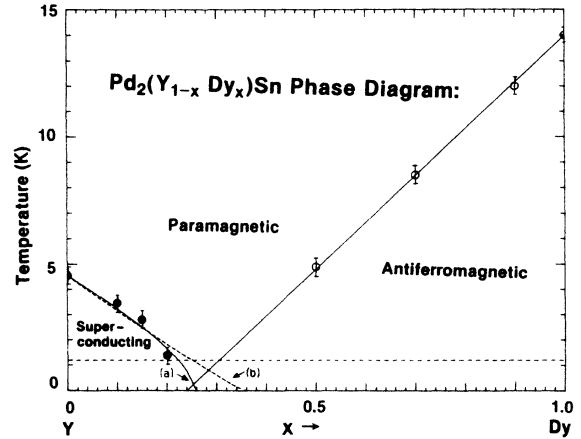


FIG. 1. The superconducting and magnetic phase diagram of the Heusler alloy system  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  as determined from ac susceptibility measurements. On the superconducting side, the curve labeled (a) is obtained from the Abrikosov-Gorkov (AG) type of analysis with the neglect of crystalline electric field (CEF) effects and with  $N(0)J_{sf}^2=1.23 \times 10^{-4}$  eV, curve (b) is obtained from AG-type analysis with the inclusion of CEF effects and with  $N(0)J_{sf}^2=2.29 \times 10^{-4}$  eV. The dashed line at 1.2 K parallel to the temperature axis represents the lower limit down to which ac susceptibility measurements have been carried out on all these samples.

the plot of molar susceptibility ( $\chi_M$ ) versus temperature ( $T$ ) and inverse molar susceptibility  $\chi_M^{-1}$  versus temperature. Peaks in  $\chi-T$  indicative of the antiferromagnetic ordering are observed. The Néel temperatures obtained from the (dc) susceptibility measurements are in very good agreement with those obtained from the ac susceptibility measurements. In the paramagnetic state, the susceptibility follows Curie-Weiss behavior [ $\chi=C/(T-\Theta_p)$ ] with effective paramagnetic moments close to the  $\text{Dy}^{3+}$  free-ion value and with negative  $\Theta_p$  values which are comparable to the respective Néel temperatures.

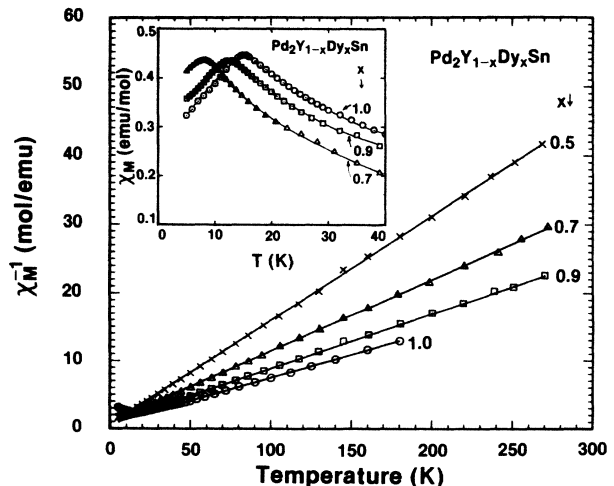


FIG. 2. Inverse molar susceptibility ( $\chi_M^{-1}$ ) of  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  compounds versus temperature,  $x \geq 0.5$ . Inset shows the variation of  $\chi_M$  vs  $T$ .

### B. Depression of the superconducting critical temperatures

The theory of the depression of  $T_c$  by dilute magnetic impurities has been worked out by Abrikosov and Gor'kov (AG).<sup>9</sup> The exchange interaction between the rare-earth spins  $\mathbf{S}$  and the conduction electrons spins  $\mathbf{s}$  given by the Hamiltonian

$$\mathcal{H} = -2J_{sf}\mathbf{S}\cdot\mathbf{s} \quad (1)$$

is responsible for the additional breaking of Cooper pairs by acting differently on the two spin directions. According to the AG theory, in the presence of randomly oriented magnetic impurities, the superconducting transition temperature, to second order in  $J_{sf}$ , is by the expression

$$\ln(T_c/T_{c0}) = \psi(\frac{1}{2}) - \psi[\frac{1}{2} + (2\pi T_c \tau_s)^{-1}], \quad (2)$$

with

$$\tau_s^{-1} = 2\pi n_I N(0)J_{sf}^2(g_J - 1)^2 J(J+1), \quad (3)$$

where  $n_I$  is the concentration of magnetic impurities,  $T_{c0}$  is the superconducting transition temperature in the absence of magnetic impurities,  $g_J$  is the Landé  $g$  factor and  $J$  the total angular momentum of impurity ion,  $N(0)$  is the density of states at the fermi level per atom per spin direction, and  $\psi$  is the digamma function. Details of the analysis of the  $T_c$  data as a function of  $x$  in  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  using Eq. (2) have been reported elsewhere.<sup>14</sup> If the crystalline electric field effects are neglected, the curve labeled (a) in Fig. 1 is obtained and the analysis yields a value of  $N(0)J_{sf}^2 = 1.23 \times 10^{-4}$  eV. Inclusion of the crystalline electric field effects in the manner suggested by Keller and Fulde<sup>15</sup> results in curve (b) in Fig. 1 with the value of  $N(0)J_{sf}^2 = 2.29 \times 10^{-4}$  eV. Since band-structure calculations on these compounds have not been performed so far, the value of the density of states is not known, and, therefore, it is not possible, at present, to obtain a realistic value of  $J_{sf}$  from the analysis of  $T_c$  data. However, as we shall see later, analysis of the critical-field data can lead to a good estimate of the exchange constant without the knowledge of the density of states. The small value of  $N(0)J_{sf}^2$  permits superconductivity to persist even when several percent of magnetic atoms are present in the lattice, as in  $\text{RMO}_6\text{S}_8$  and  $\text{RRh}_4\text{B}_4$  compounds. In the limit of low concentrations Eq. (2) has a linear asymptotic form<sup>16</sup>

$$\frac{T_c}{T_{c0}} - 1 = -0.691 \left[ \frac{n}{n_{cr}} \right], \quad (4)$$

where  $n_{cr}$  is the critical concentration of magnetic atoms for the complete suppression of superconductivity. For the  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  system, the critical concentration from Eq. (4) corresponds to  $x = 0.3$  while AG-type analysis with and without the CEF gives critical concentrations of  $x = 0.35$  and  $0.26$ , respectively.

The antiferromagnetic transition of  $\text{Pd}_2\text{DySn}$  also drops on dilution with Y in a linear way and extrapolates to zero around  $x = 0.3$ . The rare-earth  $4f$  spins interact via indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which weakens as the Dy atoms are moved away by dilution with Y. Under these conditions, superconductivi-

ty and magnetic order are not seen to coexist in any composition in the phase diagram and this rules out the possibility of finding a magnetic superconductor in this pseudoternary system, except in a very narrow range of  $x$ . An estimate of  $N(0)J_{sf}^2$  can also be obtained from the magnetic transition temperature which in the mean-field approximation is given by<sup>17</sup>

$$k_B T_M = zj^2(g_J - 1)^2 J(J+1), \quad (5)$$

where  $z$  is the average near-neighbor magnetic atom to another magnetic atom and  $j^2 \sim N(0)J_{sf}^2$ . For the Heusler alloy system  $z = 12$  and  $T_M = 15$  K for  $\text{Pd}_2\text{DySn}$  (not withstanding the structural transformation) yields a value of  $N(0)J_{sf}^2 = 0.15 \times 10^{-4}$  eV, in fair agreement with that obtained from the suppression of  $T_c$  data considering all the approximations involved.

### C. Field-induced reentrant behavior in $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$ compounds

When small amounts of Dy are substituted for Y in  $\text{Pd}_2\text{YSn}$ , the system exhibits reentrant behavior under externally applied fields as shown in Figs. 3 and 4. In Fig. 3, ac susceptibility ( $\chi_{ac}$ ) is plotted as a function of temperature for various values of applied fields. It is observed that as the temperature is lowered a transition to the superconducting state takes place first. For some values of the applied fields, as the temperature is lowered further, a transition to the normal state is observed marked by the increase in  $\chi_{ac}$ . Similar behavior is observed from the resistivity measurements (Fig. 4) where again for some values of the applied field transition to the superconducting state is seen first and then, at still lower temperatures, a transition back to the normal state is ob-

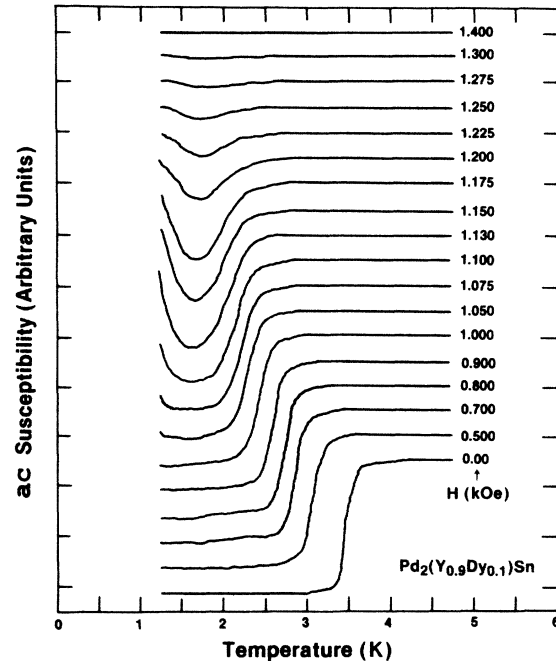


FIG. 3. Plot of ac susceptibility ( $\chi_{ac}$ ) versus temperature in  $\text{Pd}_2\text{Y}_{0.9}\text{Dy}_{0.1}\text{Sn}$  showing the field-induced reentrant behavior.

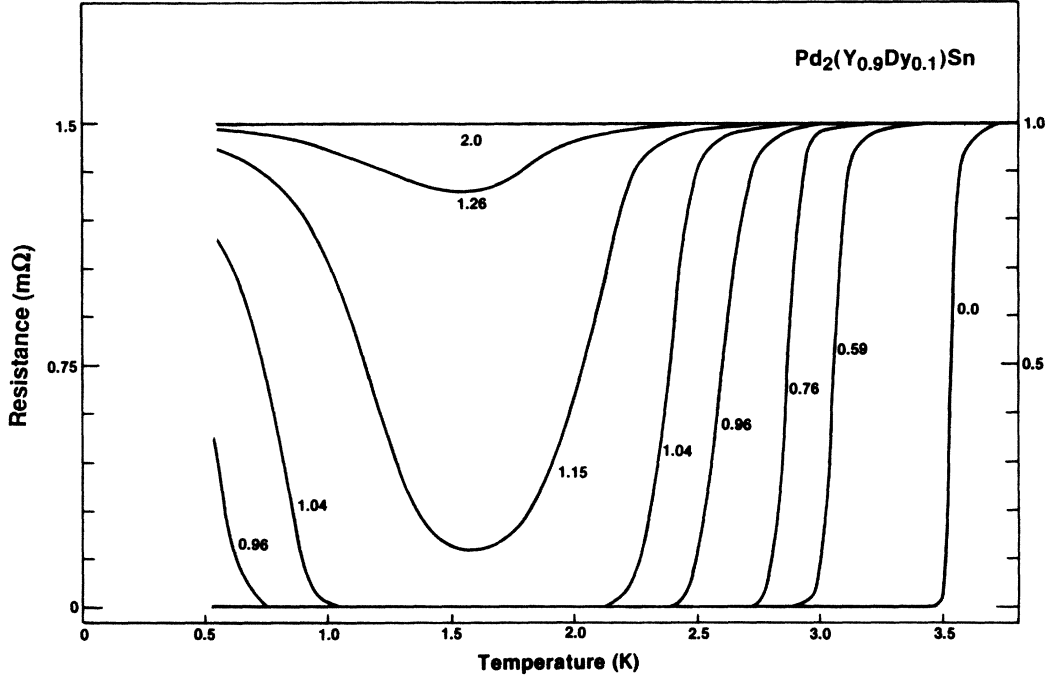


FIG. 4. Variation of resistance versus temperature in various applied fields for  $\text{Pd}_2\text{Y}_{0.9}\text{Dy}_{0.1}\text{Sn}$  showing the transition into a reentrant state.

served. The reentrant behavior arises because of the fact that, as the applied field is increased or the temperature is lowered, the rare-earth magnetization (that of Dy in this case) increases very rapidly and leads to additional pair breaking, resulting in a transition to the normal state. The reentrant behavior also implies that the critical field versus temperature curve will exhibit a bell-shaped curve with two  $T_c$ 's at some fields. This is discussed and analyzed in detail in the next section.

#### D. Upper critical fields in $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$ compounds

The temperature dependence of the upper critical field [ $H_{c2}(T)$ ] in  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  compounds obtained by the skin penetration depth technique is shown in Fig. 5. The  $H_{c2}(T)$  of  $\text{Pd}_2\text{YSn}$  behaves in a normal fashion like in any type-II superconductor. We have analyzed the  $H_{c2}(T)$  data of  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  ( $x=0.0, 0.1, \text{ and } 0.15$ ) compounds on the basis of the Wertheimer-Helfand-Hohenberg (WHH) theory modified by Fischer to take into account the additional pair-breaking effects due to paramagnetic impurities. The crystalline electric-field effects on the rare-earth magnetization have also been included. In the limit of  $\lambda_{s.o.} \ll \lambda_{tr}$  and  $\lambda_{tr} \gg 1$ ,  $H_{c2}$  is given by the following implicit equation

$$\ln Z = \left(\frac{1}{2} + i\alpha\right)\psi \left[ X + \frac{i\gamma Z}{2} \right] + \left(\frac{1}{2} - i\alpha\right)\psi \left[ X - \frac{i\gamma Z}{2} \right] - \psi\left(\frac{1}{2}\right), \quad (6)$$

with  $Z = T_{c0}/T$  and

$$\alpha = \frac{\lambda_{s.o.} - \lambda_m}{4\pi}, \quad (7a)$$

$$X = \frac{1}{2} + \frac{z}{2} \left[ 0.281 \frac{H + 4\pi M}{H_{c2}^*(0)} + \lambda_m + \frac{1}{2}(\lambda_{s.o.} - \lambda_m) \right], \quad (7b)$$

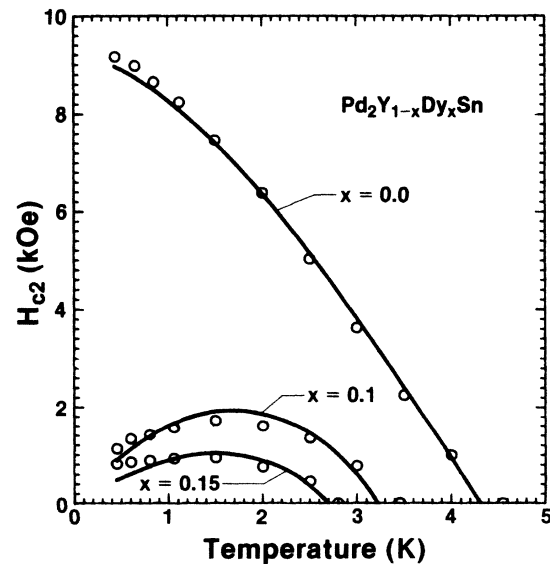


FIG. 5. Temperature dependence of the upper critical field [ $H_{c2}(T)$ ] in  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  ( $x=0.0, 0.1, 0.15$ ) compounds. The solid lines are the fit to the data based on the WHH theory including pair-breaking effects due to magnetic ions.

$$\gamma = \left[ \frac{(0.281\alpha)}{H_{c2}^*(0)} (H + H_J)^2 - \frac{1}{4} (\lambda_{s.o.} - \lambda_m)^2 \right]^2, \quad (7c)$$

where  $T_{c0}$  is the superconducting transition temperature in the absence of magnetic impurities, e.g., that of  $\text{Pd}_2\text{YSn}$  in this case,  $H_{c2}^*(0)$  is the orbital critical field,  $M$  is the magnetization of the rare-earth ions,  $\lambda_m$  is the magnetic pair-breaking term arising from the presence of magnetic rare-earth ions,  $H_J$  is the exchange field acting on the conduction electrons arising from the rare-earth ions, and  $\alpha$  is the Maki parameter, which describes the relative importance of spin paramagnetic and orbital effects in the absence of spin orbit scattering. In terms of the dirty-limit, orbital-critical-field  $H_{c2}^*(T=1)$  and the Chandrasekhar-Clogston limit  $H_p(0)$  one has

$$\alpha = \frac{\sqrt{2}H_{c2}^*(0)}{H_p}(0). \quad (8)$$

The Maki parameter can also be obtained from the slope  $dH_{c2}/dT$  (in kOe/K) by

$$\alpha = -0.0528 \left. \frac{dH_{c2}}{dT_c} \right|_{T_c}. \quad (9)$$

The exchange field  $H_J$  acting on the conduction electrons arises from the exchange interaction given by Eq. (1). In the molecular field approximation,  $H_J$  may be written as

$$\begin{aligned} H_J(H, T) &= - \frac{2xJ_{sf}\langle S_z \rangle_{av}}{g_e\mu_B} \\ &= \frac{2xJ_{sf}(g_J - 1)}{N\mu_B^2 g_J g_e} M(H, T), \end{aligned} \quad (10)$$

where  $x$  is the fraction of magnetic rare-earth ions,  $g_e \approx 2$  and  $\langle \rangle_{av}$  denotes the thermal and quantum-mechanical average over all the states. As usual,  $\langle S_z \rangle_{av}$  has been replaced by  $(g_J - 1)\langle J_z \rangle_{av}$  which in turn is proportional to the rare-earth magnetization  $M$ .

We first analyze the  $H_{c2}(T)$  data in  $\text{Pd}_2\text{YSn}$  in which Y is nonmagnetic. In this case all the terms involving quantities pertaining to the rare-earth, e.g.,  $M$ ,  $H_J$ , and  $\lambda_m$  are not present and Eq. (2) reduces to the simpler form given, for instance, in Ref. 18. The analysis involves  $T_{c0}$ , the Maki parameter  $\alpha$ , and the spin-orbit term  $\lambda_{s.o.}$ . The solid line in Fig. 5 shows one of the fits to the data obtained using  $T_{c0}^* = 4.32$  K,  $\alpha = 1.63$ , and  $\lambda_{s.o.} = 0.5$ , where  $T_{c0}^*$  is the  $T_c$  obtained from the fitting rather than the actual  $T_{c0}$ . In our analysis,  $T_c$  at  $H = 0$  was made a parameter and the  $T_{c0}^*$  thus obtained may better represent the property of the sample. As one can see from the figure, the data points near  $T_{c0}$  deviate from the theoretical fit and have a positive curvature. This is probably due to a small compositional inhomogeneity in the sample. Only a lower limit could be put on  $\lambda_{s.o.}$  as the fits are less sensitive to this parameter.

To see the origin of the bell-shaped curve for the critical-field temperature curve, one may use the simplified expression for  $H_{c2}(T)$  in the presence of magnetic impurities for  $\lambda_{s.o.} \gg 1$  as<sup>18</sup>

$$\begin{aligned} H_{c2}(T) &= H_{c2}^*(T) - 0.022 \left[ \frac{\alpha}{\lambda_{s.o.} T_{c0}} \right]^* \\ &\times [H_{c2}(T) + H_J(H, T)]^2, \end{aligned} \quad (11)$$

where  $H_J$  is given by Eq. (10). If  $H_J \gg H_{c2}$ , then the last term in Eq. (11) is  $H_J^2 = M(H, T)^2$ . As the temperature is lowered, the magnetization starts to build up rapidly. The paramagnetic reduction of  $H_{c2}$  becomes very important and the critical field passes through a maximum before decreasing towards lower temperatures giving rise to the bell-shaped curve.

We have used the complete Eq. (6) [rather than the simplified Eq. (11)] to analyze the temperature dependence of the critical field in the pseudoternary compounds containing Dy. The quantities  $T_{c0}$ , Maki parameter  $\alpha$ , and the spin-orbit coupling constant were taken from the fit of  $H_{c2}(T)$  data in  $\text{Pd}_2\text{YSn}$ . The magnetic scattering term  $\lambda_m$  is such that it reproduced the zero-field  $T_c$  suppression of  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  relative to that of  $\text{Pd}_2\text{YSn}$ . The effects due to crystalline electric fields acting on the  $\text{Dy}^{3+}$  ion were included by calculating the magnetization and the exchange field  $H_J$  in the presence of CEF splitting of the  $\text{Dy}^{3+}$  ion. The rare-earth ion  $R$  in  $\text{Pd}_2\text{RSn}$  occupies a site of cubic symmetry. The cubic CEF Hamiltonian is characterized by the parameters  $B_4^0$  and  $B_6^0$ , the strength of the fourth and the sixth degree terms. These CEF parameters have not been directly established for  $\text{Dy}^{3+}$  in  $\text{Pd}_2\text{DySn}$ . However, as a first approximation, we obtain them by scaling the corresponding values from  $\text{Pd}_2\text{TmSn}$  and  $\text{Pd}_2\text{YbSn}$ , as in the earlier work.<sup>14</sup> The values so obtained are  $B_4^0 = -0.61$  ( $10^{-2}$  K) and  $B_6^0 = 0.38$  ( $10^{-4}$  K), which should be regarded as tentative. The ground state corresponding to this combination is the  $\Gamma_7$  doublet which is isotropic in its magnetic behavior. The critical fields were calculated by solving the implicit Eq. (1). The magnetization and  $H_J$  values are required at the critical field, which itself is to be determined. Therefore, not only Eq. (6) is to be solved implicitly, we need to calculate  $M$  and  $H_J$  also in a self-consistent manner. The only unknown parameter involved in the fit is  $J_{sf}$ . Figure 5 shows one of the fits obtained to the  $H_{c2}(T)$  data for Dy containing samples. Note that since  $H_{c2} \ll H_J$ , the sign of  $J_{sf}$  cannot be determined. The exchange constant  $J_{sf}$  obtained from this fit is  $|0.04|$  eV. To some extent, the value of  $J_{sf}$  depends on the choice of  $\lambda_{s.o.}$  and, therefore, some uncertainty is created because  $\lambda_{s.o.}$  cannot be fixed exactly. Some uncertainty also arises because of the lack of the knowledge of exact CEF parameters. If the CEF effects are completely neglected, then for the same choice of remaining parameters, one can fit the  $H_{c2}(T)$  data with  $J_{sf} = |0.016|$  eV which may be regarded as the lower limit. Using  $J_{sf} = |0.04|$  eV and the  $N(0)J_{sf}^2$  values from the analysis of the depression of  $T_c$  data, we estimate the value of  $N(0)$  to fall in the range 0.08–0.14 states/(eV atom spin direction).

#### IV. CONCLUSIONS

In conclusion, the magnetic and superconducting phase diagram of the pseudoternary system  $\text{Pd}_2\text{Y}_{1-x}\text{Dy}_x\text{Sn}$  has

been investigated. On the Y-rich side, superconductivity is suppressed rapidly and the critical Dy concentration for the complete disappearance of superconductivity corresponds to about  $x = 0.3$ . On the Dy-rich side, the Néel temperature decreases linearly with the replacement of Dy by Y. Superconductivity and magnetic ordering may coexist only in an extremely narrow region of the phase diagram. Superconducting samples containing Dy show reentrant behavior under externally applied fields. The upper critical fields exhibit a bell-shaped curve in Dy con-

taining samples. Detailed analysis of the upper critical fields fully account for the bell-shaped dependence from which estimates of the exchange interaction constant  $J_{sf}$  are made.

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