# Mössbauer study of disordered, quenched Pd<sub>80</sub>Co<sub>20</sub>

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We report Mössbauer source experiments on disordered <sup>57</sup>CoPd<sub>80</sub>Co<sub>20</sub>, and ask if there is any evidence for weak ferromagnetic random anisotropy (RA) near the critical point. In making this search we are encouraged by the fact that Co has an unquenched orbital angular momentum, and that there is evidence of local-moment disalignment in small-angle neutron scattering. Spectra taken over the temperature range 89 < T < 507 K were well fitted by a continuous magnetic hyperfine field distribution which can be described by a modal field,  $H_{hf}^m(T)$ , and a full width at half maximum (FWHM)  $\Delta H_{hf}(T)$ .  $H_{hf}^m(T)$  was used to deduce  $\beta=0.38(3)$  for the range  $10^{-2} < 1-T/T_C < 2 \times 10^{-1}$ . Though insufficiently asymptotic for a strict test of theory, this value is in agreement with predictions for the random-exchange Heisenberg model.  $\Delta H_{hf}(T)$  is found to increase above 320 K, a behavior that can be described via a local-moment mean-field theory which assumes the <sup>57</sup>Fe Mössbauer site has a 17% FWHM distribution in the exchange constant and a 10% FWHM distribution in  $H_{hf}(0)$ . Neither the deduced critical behavior nor the observed hyperfine-field broadening requires the attribution of RA effects. The only evidence of the possible presence of RA in our experiments is the fact that cold working substantially broadens both  $T_C$  and the hyperfine-field distribution near  $T_C$ .

## I. INTRODUCTION

For quenched random magnetic disorder, the combination of random exchange (RE) and random anisotropy (RA) is predicted to either alter critical exponents or destroy the phase transition. In this paper we report on a critical phenomena study of concentrated, randomly disordered  $Pd_{1-x}Co_x$ , an alloy which may be a useful model for weak RA in ferromagnetic order.

If RA is present in  $Pd_{1-x}Co_x$ , we expect discernable differences with respect to the behavior of concentrated  $Pd_{1-x}Fe_x$ , an alloy for which RA is not expected. If RA is absent in  $Pd_{1-x}Co_x$ , we expect critical exponents characteristic of short-range random exchange, as observed for concentrated  $Pd_{1-x}Fe_x$ .

Our <sup>57</sup>Co Mössbauer source experiment utilizes a sample of  $Pd_{80}Co_{20}$  having a Curie point of  $T_C \approx 510$  K. At this temperature the diffusion jump rate is  $10^{-15}$  s<sup>-1</sup>,<sup>1</sup> thus assuring that the constituent atoms are *immobile* on the experimental time scale, and the chemical disorder is "quenched." Below  $T_C$  our Mössbauer spectra exhibit broadened, symmetric sextets indicating a distribution of hyperfine fields and no measurable electric quadrupole interaction. By fitting the reduced modal hyperfine field to the power law

$$h(T) \equiv H_{hf}^{m}(T) / H_{hf}^{m}(0) = B(1 - T / T_{C})^{\beta}, \qquad (1)$$

we find a value of  $\beta = 0.38(3)$ , consistent with the results for  $Pd_{1-x}Fe_x$  and inconsistent with the expected effects of RA. Transition rounding of  $\Delta T_C \approx 3$  K restricts reduced temperatures to  $t \ge 10^{-2}$ , so that effects of RA may be missed.

Independent of the observed critical behavior, we note substantial line broadening above 320 K which may be described via a distribution of the local exchange field without reference to RA. At the same time, we find the broadening above 320 K and  $\Delta T_C$  both *increase* with cold working, giving possible evidence for RA (see below). Unfortunately, current data and theory do not permit a distinction between these alternatives.

## II. BACKGROUND

Pd is known to have a high degree of magnetic polarizability. As a result, even though Pd is itself not magnetic at any temperature, its 3d magnetic alloys  $Pd_{1-x}Co_x$ ,  $Pd_{1-x}Fe_x$  and  $Pd_{1-x}Ni_x$  have ferromagnetic order for x as small as 0.1 at. %.<sup>2,3</sup> For  $x \le 1$  at. % each magnetic moment polarizes the neighboring Pd atoms, forming a "giant" moment of magnitude  $\mu_G \sim 10\mu_B$ , with  $dT_C/dx \sim 40$  K/at. %. For higher x values  $T_C(x)$  becomes nonlinear, and the average Pd moment saturates.<sup>4</sup>

Early neutron work suggested that the induced Pd moments saturate at low x, with polarization clouds ranging to 10 Å.<sup>5</sup> More recently it has been proposed that saturation occurs only at higher x, with a polarization range not much larger than the nearest-neighbor distance, and with ferromagnetism occurring through a percolation process between otherwise isolated polarization clouds.<sup>6</sup> In either case, the Pd moment appears to have a maximum value of  $\mu_{Pd}=0.4\mu_B$ , while  $\mu_{Co}$ ,  $\mu_{Fe}$ , and  $\mu_{Ni}$  are substantially larger.

In addition to the general interest of these alloys to magnetism, the critical behavior of  $Pd_{1-x}Fe_x$  has been the subject of several investigations focused on the balance between the short- and long-range magnetic interactions. These experiments have involved bulk<sup>7</sup> and Mössbauer studies<sup>8-10</sup> of both dilute and concentrated

	β	γ	δ	t <sub>min</sub>	t <sub>max</sub>	Reference
Theory:						
mean-field	0.5	1	3			
RE Heisenberg	0.365(3)	1.386(4)	4.80(4)			11
Dilute systems:						
$Pd_{98,6}Fe_{1,4}$	0.464	1.40	4.06	$1.48 \times 10^{-2}$	$9.4 \times 10^{-2}$	7
Pd <sub>96.9</sub> Fe <sub>3.1</sub>	0.428	1.37	4.24	$1.2 \times 10^{-2}$	$4.7 \times 10^{-2}$	7
Pd <sub>97.35</sub> Fe <sub>2.65</sub>	$\leq 1/2$			$3.3 \times 10^{-2}$	0.13	8
Concentrated systems:						
Pd <sub>92.5</sub> Fe <sub>7.5</sub>	0.42					9
$Pd_{86.7}Fe_{13.4}$	0.42					9
Pd <sub>3</sub> Fe	0.371(10)			$1 \times 10^{-2}$	0.15	10
Pd <sub>3</sub> Fe	0.364	1.32	4.61	$2.4 \times 10^{-3}$	$2.3 \times 10^{-2}$	7
PdFe	0.377(10)			$1 \times 10^{-2}$	0.15	10
Pd <sub>77.8</sub> Fe <sub>22.2</sub>	0.394(20)			$1 \times 10^{-2}$	0.15	10
This work:						
Pd <sub>80</sub> Co <sub>20</sub>	0.38(2)			$1 \times 10^{-2}$	0.2	this work <sup>a</sup>
	0.38(3)			$1 \times 10^{-2}$	0.2	this work <sup>b</sup>

TABLE I. Critical exponents of disordered PdFe (PdCo) systems.

<sup>a</sup>Result from free  $I_{25}/I_{16}$  parameter fitting (see text for details).

<sup>b</sup>Result from fixed  $I_{25}/I_{16}$  parameter fitting (see text for details).

samples, as summarized in Table I. With some exceptions,  $\beta$  approaches mean-field values in the dilute case, but reflects predictions for the RE Heisenberg model<sup>11</sup> in the concentrated case. This suggests that within the temperature range sampled the direct interaction between Fe moments is dominant for concentrated  $Pd_{1-x}Fe_x$ , whereas the interaction between Pd polarization clouds dominates the dilute case.

If there is a problem with this interpretation, it is that the approach to  $T_C$  in either case is insufficient to guarantee measurement of true asymptotic values of the critical exponents, as discussed by Suter and Hohenemser.<sup>12</sup> In the case of Mössbauer experiments, insufficient approach to  $T_C$  arises from substantial line broadening near  $T_C$ , thus making it difficult to extract the magnetic splitting. This line broadening, which extends over as much as  $0.5 < T/T_C < 1$ , appears to be of fundamental origin and has been explained in terms of a probability distribution of the local exchange coupling.<sup>13</sup> Hence, even for the most successful Mössbauer study<sup>10</sup>  $\Delta T_C \approx 5$  K, implying an accessible reduced-temperature range  $t \equiv 1 - T/T_C \ge 10^{-2}$ .

To date similar studies have not been undertaken for the case of  $Pd_{1-x}Co_x$ . Existing Mössbauer source studies of  ${}^{57}CoPd_{1-x}Co_x$  exhibit a saturation hyperfine field of ~310 kG independent of x (Ref. 14), line broadening as large or larger than that of  $Pd_{1-x}Fe_x$  (Ref. 15), and  $\Delta T_C \sim x^{1/2}$ .<sup>16</sup>

In contrast to Fe in  $Pd_{1-x}Fe_x$ , Co in  $Pd_{1-x}Co_x$  is known to have a strong orbital-moment component, as indicated both by a positive hyperfine field and magnetostriction.<sup>17-21</sup> As suggested by Mirebeau *et al.*,<sup>22</sup>  $Pd_{1-x}Co_x$  may therefore be a useful model of weak RA in ferromagnetic order. These authors have shown that cold work enhances local-moment disalignment seen by small-angle neutron scattering in  $Pd_{0.95}Co_{0.05}$ . This conclusion is supported by measurements of the crystalline anisotropy constant  $K_1$ , which is significantly different for annealed and rapidly quenched samples of  $Pd_{1-x}Co_x$ , with x = 0.27 and x = 0.30.<sup>23</sup> In addition, Dunlap and Dash<sup>16</sup> reported that annealing *narrows* and cold work broadens the distribution  $\Delta T_C$ . In the critical region a random alloy of  $Pd_{1-x}Co_x$  may therefore be affected not only by the balance between long- and short-range RE interactions, as in the case of  $Pd_{1-x}Fe_x$ , but independently by the effects of RA.

The behavior of d = 3 Hamiltonians with RE plus RA has been the subject of much recent theoretical work, and leads to predictions of altered critical exponents, and in some cases, destruction of the phase transition. For example, by adding RA to the RE Heisenberg Hamiltonian, Aharony and Pytte<sup>24</sup> find a phase with zero magnetization and infinite susceptibility, and Mukamel and Grinstein<sup>25</sup> suggest a new random fixed point with critical exponents distinct from the pure system. So far, little conclusive experimental work exists on this subject.<sup>26</sup>

## **III. SAMPLE AND APPARATUS**

Pd<sub>80</sub>Co<sub>20</sub> was prepared by arc-melting appropriate amounts of 99.99% pure Pd and 99.9% pure Co in an Ar atmosphere, followed by splat quenching to room temperature at a rate of ~  $10^4$  K/s. The sample thus formed was a ~ 50- $\mu$ m-thick disk. A 3-mm-diameter source was made by drying 3 mCi of carrier free <sup>57</sup>Co in 0.5M HCl on the surface of the sample, followed by a 40-h vacuum anneal at 1223 K and slow cooling.

X-ray diffraction analysis showed both the  $Pd_{80}Co_{20}$ sample and the annealed  ${}^{57}CoPd_{80}Co_{20}$  source had an fcc lattice structure with a lattice constant of 3.80 Å and no superlattice lines, indicating the samples were chemically random. All measurements were done with either an oven or a liquid-nitrogen Dewar with the sample in an oil-free vacuum. The oven<sup>27</sup> had a water-cooled vacuum jacket and a single-stage heating unit constructed of boron nitride, with the sample clamped between two BeO disks. The heater winding was made of Kanthal Al wire, and the temperature was measured and controlled via a Chromel-Constantan thermocouple. Temperature instability was generally better than 0.02 K over 24 h, with a temperature gradient across the sample  $\leq 0.1$  K.

Conventional constant-acceleration Mössbauer spectra were obtained with a single line  $K_4$ Fe(CN)<sub>6</sub>·3H<sub>2</sub>O absorber which had been previously used by Kobeissi.<sup>28</sup> Assuming a natural source linewidth, the absorber is calculated to produce a linewidth of 0.35 mm/s FWHM via resonant thickness broadening.<sup>29</sup> The observation of an experimental linewidth of 0.35(1) mm/s above  $T_C$  thus indicates that the source has no significant quadrupole broadening or other unaccountable linewidth effects.

The observed value of  $T_C = 513.0(15)$  K (see below) is in reasonable agreement with the phase diagram given by Hansen, <sup>30</sup> as well as the more recent analysis of Ododo,<sup>4</sup> according to which  $T_C \approx 500(5)$  K for  $Pd_{80}Co_{20}$ .

# IV. MÖSSBAUER DATA AND ITS ANALYSIS

To define the value of  $T_C$ , a thermal scan of the centroid-velocity transmission (CVT) was performed for the annealed sample in the temperature range 499 < T < 531 K using a constant-velocity Mössbauer drive. As shown in Fig. 1, this yielded  $\Delta T_C \approx 3$  K, considerably smaller than  $\Delta T_C \approx 20$  K seen in earlier scanning experiments by Dunlap and Dash.<sup>16</sup>



FIG. 1. Centroid-velocity transmission (CVT) as a function of temperature, showing  $\Delta T_C \approx 3$  K.

To obtain  $H_{hf}^m(T)$ , 30 Mössbauer spectra were collected over the range  $89 \le T \le 526$  K, with the absorber temperature held at 293 K. Representative spectra above and below  $T_C$  are given in Fig. 2, and indicate that the line broadening increases as  $T \rightarrow T_C$  and that individual lines show no resolved satellites in contrast to, for example,  $Fe_{96}Al_4$  (Ref. 31) and  $Fe_{12.5}V_{87.5}$ .

Based on dynamical models for isotropic spin fluctuations, <sup>33,34</sup> one expects the inner lines to broaden and collapse to a single line faster than the outer lines. This evidently does not occur in Fig. 2, and suggests the presence of a static field distribution, which should yield a linear relation between the line energy  $\Delta E_i$ , and the corresponding effective component linewidths  $\Gamma_i^{\text{eff}}$  <sup>35,36</sup> To demonstrate that this condition holds we fit selected spectra with  $\Gamma_i^{\text{eff}}$  free, and obtained results as shown in Fig.3. Hence we conclude that the observed broadening is of static origin.

To deduce the distribution of  $H_{\rm hf}$  we used a program due to Le Caër<sup>37</sup> designed to yield a continuous static distribution  $P(H_{\rm hf})$ , for each spectrum. All six lines were fixed at  $\Gamma$ =0.35 mm/s, the "natural" width for the thickness-broadened absorber, and the experimental linewidth was fitted by a  $P(H_{\rm hf})$  consisting of a 58-point histogram over  $0 \le H_{\rm hf} \le 350$  kG. The best values of the quadrupole splitting Q, the center shift  $\Delta$ , and the lineintensity ratio  $I_{25}/I_{16}$  were selected by varying each pa-



FIG. 2. Mössbauer spectra for five different temperatures, together with  $P(H_{\rm hf})$ . The FWHM of the field distribution  $\Delta H_{\rm hf}$ , is indicated.



FIG. 3. Illustration of the linear relation between the Mössbauer line energy  $\Delta E_i$  (relative to zero velocity) and the measured linewidths  $\Gamma_i^{\text{eff}}$  at three different temperatures. The linewidth above  $T_c$  is indicated by an open circle.

rameter independently. An alternative fitting method allowed freeing  $I_{25}/I_{16}$ .

As expected from the symmetry of the spectra and the lack of broadening above  $T_C$ , Q=0 provided the best fit. The best values of  $\Delta$  are expressed by the approximation  $\Delta = aT + b$  where  $a = 7.02(8) \times 10^4$  mm/s K<sup>-1</sup>, and b = -0.288(4) mm/s, in good agreement with theoretical expectations.<sup>28,38</sup> For the range  $T/T_C \leq 0.9$  the best value of the line-intensity ratio was  $I_{25}/I_{16} = 0.67(7)$ , indicating random domain alignment. For  $T/T_C > 0.9$ , however,  $I_{25}/I_{16}$  shows large fluctuations which we attribute to the ambiguity of the fitting.<sup>27</sup>

Figure 2 shows  $P(H_{\rm hf})$  for five typical spectra, with  $H_{\rm hf}^m(T)$  and  $\Delta H_{\rm hf}(T)$  summarized in Table II. We find

that  $H_{\rm hf}^m$  and  $\Delta H_{\rm hf}$  are insensitive to variations of  $\Delta$  and  $I_{25}/I_{16}$  over the range of the latter's fitting uncertainty.<sup>27</sup> To illustrate, Table II compares results for  $I_{25}/I_{16}$  free and fixed at  $I_{25}/I_{16}=0.67$ , respectively, indicating for  $H_{\rm hf}^m(T)$  a maximum difference  $\leq 4\%$ . This is too small to affect the analysis of critical behavior (see below).

Near  $T_C$  the spectra contain a region for which a single paramagnetic line coexists with a magnetically split spectrum. This permits an independent estimate of the range  $\Delta T_C$ , defined by the range of T from where P(0) first exceeds 0 to where  $H_{\rm hf}^m$  first reaches 0. This range corresponds to  $508 \le T \le 511$  K, implying  $\Delta T_C \approx 3$  K, as found via thermal scanning in Fig. 1. Because of the large concentration dependence of  $T_C$  (i.e.,  $dT_C/dx = 16$  K/at. %,) and the small estimated temperature gradient, the observed value of  $\Delta T_C \approx 3$  K may be attributed to statistical fluctuations in x over the sample.<sup>27</sup>

The values of  $H_{hf}^m(T)$  are found to fall on a magnetization curve without discernible discontinuities, as shown in Fig. 4. In contrast,  $\Delta h(T) \equiv \Delta H_{hf}(T) / \Delta H_{hf}(0)$  is anomalous: It is essentially constant for  $T/T_C \leq 0.6$ , but then increases as  $T \rightarrow T_C$ . Figure 5 contrasts this behavior to the more normal behavior of Fe<sub>87.5</sub>V<sub>12.5</sub>.<sup>32</sup>

## **V. EXPERIMENTAL RESULTS**

#### A. Critical behavior

To avoid the effect of  $\Delta T_C$ , fitting to Eq. (1) was limited to 400 < T < 507 K, i.e., a reduced-temperature range of  $10^{-2} \le t \le 2 \times 10^{-1}$ . The value  $H_{\rm hf}^m(0) = 317.8(14)$  kG was obtained by extrapolation using a Brillouin function. With values of  $H_{\rm hf}^m(T)$  given in Table II, a three-

TABLE II. Table of modal field and hyperfine-field broadening for  $Pd_{0.80}Co_{0.20}$ .  $\Delta H_{hf}$  is expressed as the full width at half maximum (FWHM). The errors of both  $\Delta H_{hf}$  and  $H_{hf}^m$  are 1.4 kG. The temperature error is 0.1 K.

Temperature	$\Delta H_1$	(kG)	$H_{\rm hf}^m$ (kG)		
(K)	$(I_{25}/I_{16} \text{ free})$	$(I_{25}/I_{16}=0.67)$	$(I_{25}/I_{16} \text{ free})$	$(I_{25}/I_{16}=0.67)$	
89.00	36.5	36.5	316.4	316.5	
199.14	35.0	35.0	301.0	301.0	
293.00	35.0	35.0	273.0	273.0	
321.75	35.2	35.3	261.2	261.2	
334.56	39.1	39.1	257.6	257.7	
349.23	40.7	40.7	250.6	251.4	
380.00	42.0	39.4	236.6	238.0	
423.03	44.8	47.7	207.2	207.2	
450.88	47.7	48.6	187.6	192.0	
466.99	53.1	55.3	162.4	160.2	
470.88	51.8	55.6	158.2	158.6	
474.00	50.5	53.1	151.2	151.0	
480.80	53.1	55.6	138.6	139.5	
490.91	57.5	58.8	121.8	123.9	
494.40	53.1	59.1	109.2	105.8	
495.27	50.5	52.4	107.8	103.6	
499.09	50.5	55.9	98.0	97.6	
503.07	54.7	56.9	89.6	90.9	
505.11	51.8	55.9	74.2	69.9	
506.45	50.5	50.5	53.2	67.0	



FIG. 4. Modal hyperfine field as a function of  $T/T_c$ .

parameter least-squares fit to Eq. (1) yielded

 $B = 1.30(5); T_C = 511.5(10) \text{ K}; \beta = 0.38(2)$ , (2a)

with  $I_{25}/I_{16}$  free, and

 $B = 1.31(9); T_C = 510.7(15) \text{ K}; \beta = 0.38(3)$ , (2b)

with  $I_{25}/I_{16}$  fixed. The minor difference between the two results indicates that the deduced critical behavior is not seriously affected by the details of fitting  $P(H_{\rm hf})$ . Figure 6 illustrates the quality of the power law fit in the form of a logarithmic plot for fixed  $I_{25}/I_{16}$ .

As for the best work on  $Pd_{1-x}Fe_x$ , because of  $T_C$ rounding, the deduced exponent must be considered an "effective" value since the data are outside the range  $t \le 10^{-2}$  which is normally considered the asymptotic region for d=3 systems.<sup>12</sup> Nevertheless, the result is consistent with  $\beta=0.365(3)$  predicted for the pure<sup>11</sup> as well as the RE Heisenberg model<sup>39</sup> in three dimensions. The result is, further, closely comparable to those found for



FIG. 5. Normalized hyperfine-field distribution width  $\Delta H_{\rm hf}(T)/\Delta H_{\rm hf}(0)$  as a function of  $T/T_C$ . The open circles are for <sup>57</sup>CoPd<sub>80</sub>Co<sub>20</sub>. The solid circles are for Fe<sub>87.5</sub>V<sub>12.5</sub>.



FIG. 6. Logarithmic plot of the reduced modal hyperfine field, h, as a function of the reduced temperature t. The solid line is a power law fit yielding an effective value of  $\beta = 0.38(3)$ .

concentrated  $Pd_{1-x}Fe_x$ . As in  $Pd_{1-x}Fe_x$ , the shortrange interaction between Co moments in  ${}^{57}CoPd_{80}Co_{20}$ appears to overwhelm any long-range interaction between Pd polarization clouds. There is, at the same time, no basis for altered critical behavior that might be attributable to RA effects.

## **B.** Field distribution

The question remains: Why does the field broadening  $\Delta H_{\rm hf}$  increase dramatically for  $T/T_C \ge 0.6$ ? Can this be evidence for RA effects? Before we can consider this possibility, we consider two other explanations of the broadening.

(1) The effect of  $T_c$  rounding. Consider that for an effective temperature inhomogeneity  $\Delta T$ , the field distribution is  $\Delta H_{\rm hf} = (\partial H_{\rm hf} / \partial T) \Delta T$ , where  $\partial H_{\rm hf} / \partial T$  is the numerical derivative of  $H_{\rm hf}(T)$ . Using  $\Delta T = 3$  K leads to  $\Delta H_{\rm hf}$  values an order of magnitude smaller than the observed effect shown in Fig. 5. Therefore, the observed  $\Delta H_{\rm hf}$  cannot be related to the critical-temperature distribution  $\Delta T_c \approx 3$  K.

(2) The effect of a distribution of exchange coupling. In other alloys the temperature dependence of  $\Delta H_{\rm hf}$  has been attributed to a distribution of the exchange coupling. <sup>13,36,40-42</sup> For magnetically dilute Pd<sub>99,6</sub>Fe<sub>0.4</sub>, Kitchens and Trousdale<sup>13</sup> did self-consistent calculations of the magnetic and exchange field distribution based on the magnetic interaction. In the present case we adopt a related model developed for concentrated NiFe (Refs. 36 and 42) using three assumptions: (1)  $H_{\rm hf}$  at the Fe nucleus is proportional to the local magnetization m(T) of the sample; (2)  $H_{\rm hf}(T)$  is given by a Brillouin function depending on the local exchange coupling  $\zeta$ ; and (3) the distribution  $\Delta H_{\rm hf}$  arises both from the spread in the exchange coupling  $\Delta \zeta$  and the spread in saturation hyperfine field  $\Delta H_{\rm hf}(0)$ . The first two assumptions imply<sup>40</sup>

$$H_{\rm hf}(T) = H_{\rm hf}(0)B_{S}\{[\zeta T_{C}/T][m(T)/m(0)]\}, \qquad (3)$$

where  $\zeta \equiv g\mu_B SH_{\rm ex,Fe}/kT_C$ ,  $H_{\rm ex,Fe}$  is the exchange field, and  $B_S$  is known as a "modified" Brillouin function. For a chemically disordered alloy  $H_{\rm hf}(0)$  and  $\zeta$  are assumed to have widths,  $\Delta H_{\rm hf}(0)$  and  $\Delta \zeta$ , respectively, with the result

$$\Delta H_{\rm hf}(T) = \Delta H_{\rm hf}(0) B_S(y) + H_{\rm hf}(0) B_S'(y) \Delta \zeta , \qquad (4)$$

where  $y = (\zeta T_C / T)[m(T)/m(0)]$  and the prime denotes differentiation with respect to  $\zeta$ . The first term is due to the variation in the saturation field at different sites, and has a monotonically *decreasing* temperature dependence. The second term is due to the spatial fluctuation in the exchange constant at different sites, and *increases* from zero at low temperatures to a maximum at high temperature, followed by a fast decrease near  $T_C$ .

Figure 7 shows a fit of Eq. (4) to our data using the experimental values of  $\Delta H_{\rm hf}(0)/H_{\rm hf}(0) \approx \Delta H_{\rm hf}(89)/H_{\rm hf}(89) = 0.10$ , the parameters  $S = \frac{1}{2}$ ,  $\zeta = 2.4$ ,  $\Delta \zeta = 0.4$ , and the assumption that m(T)/m(0) is given by a Brillouin function. Thus, the observed linewidth is described by a 10% FWHM distribution in  $H_{\rm hf}(0)$  and a 17% FWHM distribution in the exchange constant. Considering the simplicity of the assumptions and the fact that the calculated curve catches most of the features of the data, the fit is considered to be very good.

We therefore conclude that within the local-moment model assumed in Eq. (4), the observed field broadening may be described by a distribution of the RE interaction without reference to RA effects. At the same time, it must be recognized that the local-moment picture assumed in Eq. (4) may not be adequate and a consistent description of alloys requires considering significant magnetic contributions from itinerant electrons.<sup>43</sup>

#### C. Effects of cold working

In addition to the above results we have obtained thermal scans and field distributions near  $T_C$  for a source that has been cold rolled to increase the source foil area by ~25%. We find this results in  $\Delta T_C \approx 10$  K instead of 3 K, and an increase in  $\Delta H_{\rm hf}(T)$  by ~30% without a significant change in the mean value of  $T_C$ . Given the association of cold rolling with local-moment disalignment as measured by small-angle neutron scattering,<sup>22</sup> one may surmise that our  $T_C$  rounding and field broadening are, in part, attributable to the effects of RE plus RA, as predicted by Aharony and Pytte<sup>24</sup> and Mukamel and Grinstein.<sup>25</sup> Unfortunately we know of no way of making this surmise more quantitative.

### VI. SUMMARY AND CONCLUSION

We have reported a Mössbauer source experiment on disordered  ${}^{57}\text{CoPd}_{80}\text{Co}_{20}$ . Spectra taken over the temper-



FIG. 7. Comparison of the calculated  $\Delta H(T)/\Delta H(0)$  with the <sup>57</sup>CoPd<sub>80</sub>Co<sub>20</sub> data (closed circles). The calculated curve is characterized by  $S = \frac{1}{2}$ ,  $\zeta = 2.4$ ,  $\Delta \zeta = 0.4$ , and  $\Delta H_{hf}^{m}(0)/H_{hf}^{m}(0) = 0.10$ .

ature range 89 < T < 507 K were well fit by a continuous magnetic hyperfine field distribution described by a modal field  $H_{hf}^m(T)$  and a width  $\Delta H_{hf}(T)$ . We used  $H_{hf}^m(T)$  to deduce  $\beta = 0.38(3)$  for the range  $10^{-2} < t < 2 \times 10^{-1}$ , in agreement with predictions for the RE Heisenberg model. Hyperfine field broadening,  $\Delta H_{hf}(T)$ , is found to increase above 320 K. This behavior can be described by a local-moment mean-field model that assumes a 17% FWHM distribution in the exchange constant and a 10% FWHM distribution in the saturation field at the <sup>57</sup>Fe Mössbauer site.

Neither the deduced critical behavior nor the observed hyperfine-field broadening require the attribution of RA effects. The only possible evidence for the presence of RA is the fact that cold working substantially broadens  $T_c$  and the hyperfine-field distribution.

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