series

$$I_{z2}\propto\sum_{m=0}^{\infty}\int_0^{\pi}d\psi_0\int_{-\psi_0}^{\psi_0}d\psi$$
$$\times\int_a^b d\theta\{\cos[\psi+\theta+2(m+1)(\pi-\psi_0)]$$
$$-\cos\psi\}\exp[-\theta/\omega_c\bar{\tau}],\quad\text{(B7)}$$

where $a=\psi_0-\psi+2m\psi_0$ and $b=\psi_0-\psi+2(m+1)\psi_0$. Changing variable,

$$\theta'=\theta-(\psi_0-\psi)-2m\psi_0,$$

one can sum the series as a binomial expansion of $[1-\exp(-2\psi_0/\omega_c\bar{\tau})]$. One finally obtains

$$I_{z2}\propto\int_0^{\pi}d\psi_0\int_0^{\psi_0}d\psi\frac{\cosh(\psi/\omega_c\bar{\tau})}{\sinh(\psi_0/\omega_c\bar{\tau})}$$
$$\times\int_0^{2\psi_0}d\theta'[\cos(\theta'-\psi_0)-\cos\psi]\exp[-\theta'\omega_c\bar{\tau}].\quad\text{(B8)}$$

The result in the form of Eq. (B8) may be shown to be identically zero because contributions to I_{z2} at the surface are 180° out of phase to contributions at distances of order r_c.

PHYSICAL REVIEW VOLUME 138, NUMBER 6A 14 JUNE 1965

Magnetization Distribution in a Palladium-Rich FePd Alloy*

WALTER C. PHILLIPS†
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received 19 October 1964; revised manuscript received 1 February 1965)

A ferromagnetic $Fe_{0.013}Pd_{0.987}$ single crystal was examined with the polarized-beam neutron-diffraction technique in a study of the distribution of the localized magnetization in the alloy. Intensities of all nineteen Bragg reflections out to $\sin\theta/\lambda=0.90$ Å^{-1} were measured at 4.2°K in a field of 14 kOe, yielding the magnetic form factor averaged over all atoms. These data are fitted to a linear combination of calculated 3*d* and 4*d* free-atom form factors, resulting in a moment of $0.050\pm0.006\ \mu_B$ of 3*d*-like moment and $0.088\pm0.008\ \mu_B$ of 4*d*-like moment per average atom. A Fourier inversion of the magnetic scattering amplitudes emphasizes the aspherical shape of the unpaired-electron distribution. The over-all E_g/T_{2g} ratio is 0.39 ± 0.02. The measured saturation magnetization of this alloy is $0.114\pm0.004\ \mu_B$ per atom at 4.2°K, which is considerably smaller than the total moment of $0.138\ \mu_B$ seen by neutron diffraction. This discrepancy suggests a negative conduction-electron polarization of $-0.024\pm0.011\ \mu_B$ per atom. The temperature dependence of the magnetic scattering amplitude and the saturation magnetization indicate that the conduction-electron polarization disappears near the Curie temperature, which is about 55°K. In addition, these data suggest that the 3*d* moment on an Fe atom and the 4*d* moments on surrounding Pd atoms are strongly coupled, although the range of the Pd polarization is not determined. The total *d* moment associated with the moment cluster around each impurity site is $10.7\pm0.6\ \mu_B$.

INTRODUCTION AND THEORY

MANY alloys in which a 3*d* atom impurity is present in a 4*d* atom matrix exhibit interesting magnetic properties.[1–6] Dilute solutions of Fe in Pd are ferromagnetic at suitably low temperatures for all compositions which have been studied (except for an extremely dilute sample[7]) and apparently ferromagnetism will exist at fractional-percent Fe concentrations. An FePd alloy was chosen for this study because of the relatively high Curie temperature and large moment per Fe atom reported earlier.[3,5–7] (The measured Curie temperature is 55°K and the saturation magnetization is $8.8\pm0.5\ \mu_B$ per Fe atom in our sample.) From these moment values it is evident that not only the Fe atoms contribute to the ferromagnetic moment, but each Fe impurity polarizes some Pd atoms, resulting in an enhanced moment per Fe atom. The large paramagnetic susceptibility[8] of pure Pd shows that the 4*d* bands are not filled, and apparently a ferromagnetic impurity can trigger the Pd matrix into a ferromagnet.

---

* This work was supported by a research grant from the U. S. Atomic Energy Commission. It is based on a thesis submitted to the Department of Physics at the Massachusetts Institute of Technology in May, 1964 in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

† Present address: Argonne National Laboratory, Argonne, Illinois.

[1] M. Fallot, Ann. Phys. (Paris) **10**, 291 (1938).

[2] D. Gerstenberg, Ann. Physik **2**, 236 (1958).

[3] J. Crangle, Phil. Mag. **5**, 335 (1960).

[4] R. M. Bozorth, P. A. Wolff, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. **122**, 1157 (1961).

[5] R. M. Bozorth, D. D. Davis, and J. H. Wernick, J. Phys. Soc. Japan **17**, Suppl. BI, 112 (1962).

[6] A. M. Clogston, B. T. Matthias, M. Peter, H. J. Williams, E. Corenzwit, and R. C. Sherwood, Phys. Rev. **125**, 541 (1962).

[7] P. P. Craig, D. E. Nagle, W. A. Steyert, and R. D. Taylor, Phys. Rev. Letters **9**, 12 (1962).

[8] D. W. Budworth, F. E. Hoare, and J. Preston, Proc. Roy. Soc. **A257**, 250 (1960).

With the polarized-beam technique[9] we have been able to study the distribution of localized magnetization in an FePd single crystal with composition 1.29 at.% Fe. In this alloy the average Fe-Fe distance is 6.2 Å, corresponding to an average fifth-nearest-neighbor separation. The magnetic form factor and the magnitudes of the 3*d*, 4*d*, and conduction-electron moments were determined from the measured magnetic scattering amplitudes and the saturation magnetization.

The magnetic contribution to the coherent elastic scattering depends upon the time average, over all spin configurations of the ferromagnet, of the atomic spin and orbital-momentum components perpendicular to the scattering vector. The magnetic scattering amplitude (in centimeters) for an atom in a random solid solution can be written

$$p(\mathbf{k})=\frac{e^2\mu_N}{2mc^2}n_B f(\mathbf{k})=0.2695\times10^{-12}\times\int\rho(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}. \quad (1)$$

k is the scattering vector, n_B is the saturation moment per average atom in Bohr magnetons, μ_N is the neutron magnetic moment, and e^2/mc^2 is the classical electron radius. The form factor $f(\mathbf{k})$ is normalized to unity at $k=0$ and the integration is over the volume occupied by one atom. $\rho(\mathbf{r})$ is the magnetic moment density in Bohr magnetons per unit volume and is characteristic mainly of the distribution of unpaired electron spin. There is a contribution to $\rho(\mathbf{r})$ from unquenched orbital motion; the relative magnitudes of the orbital and spin moments can be deduced from the magnetomechanical ratio.

The direction of magnetization in the crystal was perpendicular to the scattering plane. The diffracted intensity at the center of each Bragg reflection was measured for the two states of beam polarization, parallel and antiparallel to the direction of crystal magnetization. From the ratio of these intensities (the polarization ratio), $p(\mathbf{k})$ was obtained directly:

$$R(\mathbf{k})=\left(1+\frac{\langle p(\mathbf{k})\rangle}{\langle b\rangle}\right)^2\bigg/\left(1-\frac{\langle p(\mathbf{k})\rangle}{\langle b\rangle}\right)^2, \quad (2)$$

where b is the nuclear coherent scattering amplitude and the averages are over all atoms.

$p(\mathbf{k})$ can be determined for the values of **k** at which the Bragg condition is satisfied. A Fourier inversion of these data yields the time-average value of the localized magnetic-moment distribution on an average atom. The symmetry displayed in the Fourier map is characteristic of this averaged moment density and does not reveal effects of magnetic clusters because the coherent scattering depends only on the long-range periodicity of the lattice.

The polarized-beam technique permits an accurate determination of the form factor for *d* electrons and this can be compared directly to form factors calculated from free-atom wave functions. The use of free-atom functions to describe transition-metal unpaired spin distributions appears to be justified on theoretical grounds.[10] Neutron-diffraction experiments have revealed that free-atom wave functions do indeed characterize the 3*d* unpaired spin distributions in Fe and Co and the 4*d* distribution of Rh in an FeRh alloy.[11] The form factors determined experimentally for Fe by Shull and Yamada[12] and for hexagonal Co by Moon[13] are in surprisingly good agreement with form factors obtained from spin-polarized free-atom wave functions calculated by Watson and Freeman.[14] The excellent agreement was partially brought about by adding to the calculated 3*d* moment density a constant negative moment (which Shull and Yamada assign to the conduction electrons). In fitting a calculated form factor to the FePd-alloy data we also adopted free-atom functions and allowed a constant polarization because this procedure 1) was successful in matching the Fe and Co data, and 2) seems less arbitrary than modifying the calculated free-atom functions to obtain agreement with experiment.

The form factor is not a smooth function of **k** if the moment density is aspherical. Weiss and Freeman have treated the problem of *d* electrons which have their orbital degeneracy removed by the crystalline field.[15] For cubic symmetry the five *d* states split into a triply degenerate T_{2g} state and a doubly degenerate E_g state. In a pure metal or an ordered alloy the relative populations of the *d* substates occupied by the unpaired electrons can be accurately determined from the measured $f(\mathbf{k})$.[11–13] For the disordered FePd alloy a unique determination of the symmetry of both the 3*d* and 4*d* distributions was not possible and only the over-all *d*-substate population was obtained.

CRYSTAL

An FePd single crystal was grown from the melt by the Bridgman technique in an rf furnace using 99.99+% pure Pd. To aid in eliminating any Fe clusters, the crystal ingot was homogenized by annealing at approximately 1000°C for 50 h, 1100°C for 100 h, and 1200°C

---

[9] R. Nathans, C. G. Shull, G. Shirane, and A. Andresen, J. Phys. Chem. Solids **10**, 138 (1959).

[10] C. Herring, J. Appl. Phys. **31**, 3S (1960).

[11] G. Shirane, R. Nathans, and C. W. Chen, Phys. Rev. **134**, A1547 (1964).

[12] C. G. Shull and Y. Yamada, J. Phys. Soc. Japan **17**, Suppl. BIII, 1 (1962).

[13] R. M. Moon, Lincoln Laboratory, Massachusetts Institute of Technology, Technical Report No. 312, 1963 (unpublished); Phys. Rev. **136**, A195 (1964).

[14] R. E. Watson and A. J. Freeman, Phys. Rev. **123**, 2027 (1961).

[15] R. J. Weiss and A. J. Freeman, J. Phys. Chem. Solids **10**, 147 (1959).

for 50 h. All of the neutron diffraction data were obtained with a flat disc approximately 3/8 in. in diameter and 0.021 in. in thickness cut from the ingot. The composition of this slice, determined by a subsequent chemical analysis, was 1.29±0.02 at.% Fe. For alloys with less than 25 at.% Fe, FePd is a solid solution with a face-centered cubic Bravais lattice.

ANALYSIS OF THE RESULTS OBTAINED AT 4.2°K

The crystal was mounted in a transfer-tube type of cryostat which was positioned in the gap of an electromagnet capable of producing a field of from 0 to 14 kOe at the sample. The crystal temperature could be varied in this system by changing the flow rate of the liquid helium or helium vapor and could be controlled to $\pm\frac{1}{2}$°K or better from liquid-helium to liquid-nitrogen temperature.

Only small corrections to the measured intensities for instrumental effects and extinction were called for because the polarization of the beam was high (>99%), the crystal mosaic width was very broad, and the measured polarization ratios were nearly unity. Several azimuthal sweeps were made and these revealed that simultaneous reflections were not contributing significantly to the measured intensities.[16] The Pd and Fe nuclear scattering amplitudes were taken as $b_{\mathrm{Pd}}=0.60\pm0.02\times10^{-12}$ cm,[17] $b_{\mathrm{Fe}}=0.96\times10^{-12}$ cm, and the primary wavelength was 1.05 Å.

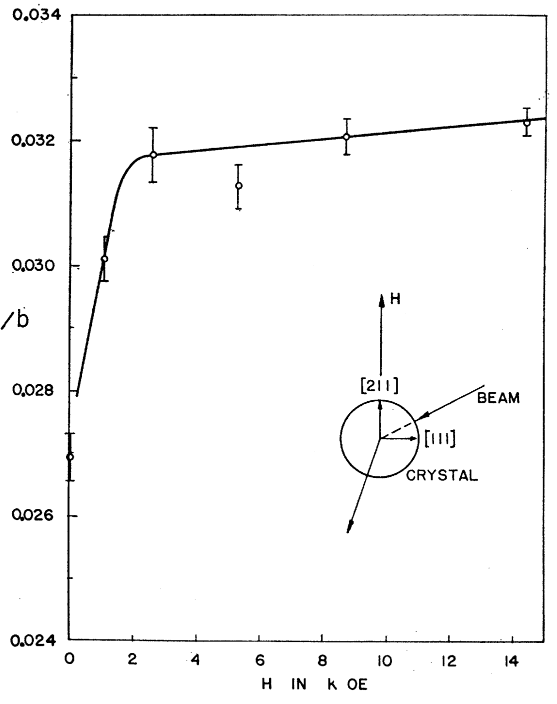

FIG. 1. Scattering amplitude ratio versus magnetic field for the (111) reflection at 4.2°K. The scattering vector and the direction of magnetization are in the plane of the crystal slice.

The polarization ratio of the (111) reflection was measured as a function of magnetic field at 4.2°K. The resulting scattering amplitude ratio shown in Fig. 1 indicates that in the [211] direction the crystal was magnetically saturated at 2.5 kOe. The increase in p/b with field can be accounted for by a paramagnetic susceptibility of the Pd matrix of $\chi_M=9\times10^{-4}$ emu per mole. The molar susceptibility of pure Pd is 7.8×10^{-4} emu per mole at 20°K.[8]

The polarization ratios of the first 19 Bragg reflections were measured in a 14-kOe field. The measured and corrected polarization ratios and the corresponding scattering amplitude ratios are given in Table I. The magnetic scattering amplitude at $k=0$ was obtained from the measured saturation magnetization, which was 0.114±0.004 μ_B per atom (8.8±0.5 μ_B per Fe atom) at 4.2°K.[18] Values for the 3d, 4d, and conduction electron moments per atom were determined from these data using the following procedure.

TABLE I. The measured and corrected polarization ratios and the corresponding scattering-amplitude ratios for the 4.2°K data.

$(\sin\theta)/\lambda$ (Å^{-1})	$h\ k\ l$	Measured polarization ratio	Corrected polarization ratio	Scattering-amplitude ratio $\langle p\rangle/\langle b\rangle$	Calculated scattering-amplitude ratio[a]
0.223	1 1 1	1.1342±0.0010	1.1395	0.0326±0.0004	0.0328
0.258	2 0 0	1.1069±0.0015	1.1109	0.0263±0.0005	0.0261
0.365	2 2 0	1.0507±0.0014	1.0522	0.0127±0.0005	0.0128
0.427	3 1 1	1.0261±0.0017	1.0269	0.0066±0.0005	0.0069
0.446	2 2 2	1.0273±0.0019	1.0281	0.0069±0.0005	0.0073
0.516	4 0 0	1.0065±0.0013	1.0067	0.0017±0.0004	0.0009
0.562	3 3 1	1.0111±0.0019	1.0114	0.0028±0.0005	0.0025
0.577	4 2 0	1.0042±0.0016	1.0043	0.0011±0.0004	0.0009
0.632	4 2 2	1.0068±0.0018	1.0070	0.0017±0.0005	0.0011
0.670	5 1 1	0.9929±0.0017	0.9927	−0.0018±0.0004	−0.0009
0.670	3 3 3	1.0096±0.0016	1.0099	0.0025±0.0004	0.0016
0.729	4 4 0	1.0001±0.0022	1.0001	0.0000±0.0005	0.0006
0.764	5 3 1	1.0004±0.0017	1.0004	0.0001±0.0004	0.0003
0.774	6 0 0	0.9952±0.0019	0.9949	−0.0013±0.0005	−0.0015
0.774	4 4 2	1.0036±0.0024	1.0037	0.0009±0.0006	0.0010
0.815	6 2 0	1.0005±0.0023	1.0005	0.0001±0.0006	−0.0006
0.845	5 3 3	1.0055±0.0020	1.0058	0.0014±0.0005	0.0009
0.855	6 2 2	0.9960±0.0023	0.9959	−0.0010±0.0006	0.0000
0.894	4 4 4	0.9976±0.0025	0.9976	−0.0006±0.0006	0.0013

[a] From Eq. (8) with $\mu_{\mathrm{Fe}}=0.048$, $\mu_{\mathrm{Pd}}=0.090$, $g_{\mathrm{Fe}}=2.08$, $g_{\mathrm{Pd}}=2.10$, and the Watson and Freeman form factors.

From Eq. (1)

$$\langle p(\mathbf{k})\rangle=\frac{e^2\mu_N}{2mc^2}[n_{\mathrm{Fe}}f_{\mathrm{Fe}}(\mathbf{k})e^{-M_D(k)}+n_{\mathrm{Pd}}f_{\mathrm{Pd}}(\mathbf{k})], \tag{3}$$

where n_{Fe} and n_{Pd} are the total moments per average atom associated with Fe and Pd atoms respectively, $f_{\mathrm{Fe}}(\mathbf{k})$ and $f_{\mathrm{Pd}}(\mathbf{k})$ are the corresponding normalized form factors and M_D is the difference between the Debye-Waller factors for the Fe and Pd atoms. Assuming the magnetic moment arises from three sources, each form factor

$$f(\mathbf{k})=\frac{2}{g}f_{\mathrm{spin}}(\mathbf{k})+\frac{g-2}{g}f_{\mathrm{orbit}}(\mathbf{k})+f_{\mathrm{core}}(\mathbf{k}), \tag{4}$$

---

[16] R. M. Moon and C. G. Shull, Acta Cryst. 17, 805 (1964).

[17] J. W. Cable (private communication).

[18] The magnetization was kindly measured for us by S. Foner of the National Magnet Laboratory, Massachusetts Institute of Technology.

where $f_{\rm spin}$ is a normalized spin-density form factor, $f_{\rm orbit}$ is a normalized orbital form factor, $f_{\rm core}$ is a core-polarization form factor and is very small compared to $f_{\rm spin}$ or $f_{\rm orbit}$ and g is a spectroscopic splitting factor. We introduce a constant spin density, represented by a $\delta(k)$, and write the normalized spin-density form factor

$$f_{\rm spin}(\mathbf{k})=(1+\alpha)f_d(\mathbf{k})-\alpha\delta(k)\,. \tag{5}$$

Since the form factor associated with $4s$ electrons falls essentially to zero before the first Bragg reflection, we ascribe this uniform spin density to conduction electrons, as Shull and Yamada[12] have done in analyzing their results for Fe. The constant α determines the magnitude and direction of the conduction electron polarization. For d electrons in a cubic field[15]

$$f_d(\mathbf{k})=\langle j_0\rangle+(\tfrac{5}{2}\gamma-1)A_{hkl}\langle j_4\rangle\,. \tag{6}$$

$\langle j_0\rangle$ characterizes the spherical part of the spin distribution, $\langle j_4\rangle$ the aspherical part, γ is the fraction of unpaired d electrons in the E_g state ($\gamma=0.40$ for spherical symmetry) and

$$A_{hkl}=\frac{h^4+k^4+l^4-3(h^2k^2+h^2l^2+k^2l^2)}{(h^2+k^2+l^2)^2}\,. \tag{7}$$

Equation (3) now becomes

$$\begin{aligned}\langle p(\mathbf{k})\rangle=\frac{e^2\mu_N}{2mc^2}\Bigg[&(1+\alpha_{\rm Fe})n_{\rm Fe}e^{-M_D}\frac{2}{g_{\rm Fe}}\\ &\times(\langle j_0\rangle_{\rm Fe}+(\tfrac{5}{2}\gamma_{\rm Fe}-1)A_{hkl}\langle j_4\rangle_{\rm Fe})\\ &+(1+\alpha_{\rm Pd})n_{\rm Pd}\frac{2}{g_{\rm Pd}}\\ &\times(\langle j_0\rangle_{\rm Pd}+(\tfrac{5}{2}\gamma_{\rm Pd}-1)A_{hkl}\langle j_4\rangle_{\rm Pd})\\ &+n_{\rm Fe}e^{-M_D}\left(\frac{g_{\rm Fe}-2}{g_{\rm Fe}}f_{\rm orbit}+f_{\rm core}\right)_{\rm Fe}\\ &+n_{\rm Pd}\left(\frac{g_{\rm Pd}-2}{g_{\rm Pd}}f_{\rm orbit}+f_{\rm core}\right)_{\rm Pd}\\ &-\left(\frac{2n_{\rm Fe}\alpha_{\rm Fe}}{g_{\rm Fe}}+\frac{2n_{\rm Pd}\alpha_{\rm Pd}}{g_{\rm Pd}}\right)\delta(k)\Bigg]\,.\end{aligned} \tag{8}$$

This expression was used to evaluate $\gamma_{\rm Fe}$ and $\gamma_{\rm Pd}$ and the moments per average atom associated with Fe $3d$ electrons, Pd $4d$ electrons, and conduction electrons

$$\begin{aligned}\mu_{\rm Fe}&=(1+\alpha_{\rm Fe})n_{\rm Fe}\\ \mu_{\rm Pd}&=(1+\alpha_{\rm Pd})n_{\rm Pd}\\ \mu_0&=-(\alpha_{\rm Fe}n_{\rm Fe}+\alpha_{\rm Pd}n_{\rm Pd})\,.\end{aligned} \tag{9}$$

All of the calculated form factors used are free-atom form factors due to Watson and Freeman: $\langle j_0\rangle_{\rm Fe}$ from a spin-polarized Hartree-Fock calculation[19] for a $3d^64s^2$ configuration, $\langle j_4\rangle_{\rm Fe}$ and $f_{\rm orbit}{}^{\rm Fe}$ from a restricted Hartree-Fock calculation[20] for Fe^{+2}, and $\langle j_0\rangle_{\rm Pd}$, $\langle j_4\rangle_{\rm Pd}$ and $f_{\rm orbit}{}^{\rm Pd}$ from a restricted Hartree-Fock calculation[21] for Pd^{+2}. $f_{\rm core}{}^{\rm Fe}$ was estimated and $f_{\rm core}{}^{\rm Pd}$ neglected.

The g values for this alloy are not known. Consequently, several pairs of values were assumed ($2.00\leqslant g_{\rm Fe}\leqslant 2.08$ and $2.00\leqslant g_{\rm Pd}\leqslant 2.19$) and $\mu_{\rm Fe}$, $\mu_{\rm Pd}$, and μ_0 determined for each pair from Eq. (8). The following results were obtained: the $3d$ moment $=0.050\pm0.006$ μ_B per atom (3.9 ± 0.5 μ_B per Fe atom), the $4d$ moment $=0.088\pm0.008$ μ_B per atom (6.9 ± 0.7 μ_B per Fe atom), the conduction-electron moment $=-0.024\pm0.011$ μ_B per atom (-1.9 ± 0.8 μ_B per Fe atom). It should be emphasized that these derived moment values depend strongly on the shapes of the calculated $3d$ and $4d$ form factors. Scattering-amplitude ratios calculated using Eq. (8) are listed in Table I. Figure 2 shows the measured form factor and a calculated spherical form factor. Using only spin form factors in Eq. (8) ($g_{\rm Fe}=g_{\rm Pd}=2.00$), the $3d$, $4d$, and conduction-electron moments per Fe atom are 4.3 μ_B, 6.5 μ_B, and -1.9 μ_B, respectively. The effect of including some orbital contribution in the fit is to decrease the $3d$ moment per atom (and thus increase the $4d$ moment)

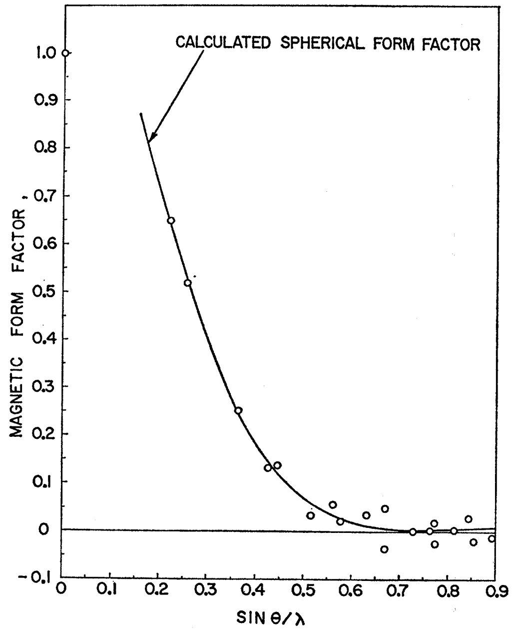

FIG. 2. Measured magnetic form factor and a calculated spherical d form factor. The measured form factor is normalized at $k=0$ by the saturation-magnetization value. The spherical form factor is calculated from Eq. (8) using Watson and Freeman form factors (Refs. 19, 20, and 21) and $\mu_{\rm Fe}=0.048$, $\mu_{\rm Pd}=0.090$, $g_{\rm Fe}=2.08$, $g_{\rm Pd}=2.10$.

---

[19] R. E. Watson and A. J. Freeman (unpublished), from calculations given in Ref. 14.

[20] R. E. Watson and A. J. Freeman, Acta Cryst. 14, 27 (1961) and unpublished calculations.

[21] A. J. Freeman and R. E. Watson (unpublished).

because the orbital-moment distribution is more contracted than the spin distribution and therefore, as a function of increasing k, the orbital form factor falls off more slowly than the spin form factor. Thus for $g_{\text{Fe}}=2.08$ and $g_{\text{Pd}}=2.10$, the $3d$, $4d$, and conduction moments per Fe atom are 3.7 μ_B, 7.0 μ_B, and -1.9 μ_B, respectively. For any of the g values used in the data fits, the $3d$ moment per Fe atom is larger than the values previously reported for Fe atoms in metals. Shull and Yamada found a $3d$ moment of 2.39 μ_B in pure iron.[12] Cable *et al.* measured 3.0 μ_B per Fe atom in $\text{Fe}_{0.03}\text{Pd}_{0.97}$ and $\text{Fe}_{0.07}\text{Pd}_{0.93}$ in a neutron diffuse scattering experiment.[22]

In principle it is possible to obtain values for both γ_{Fe} and γ_{Pd} from Eq. (8). However, our data did not permit the determination of the asymmetry parameters separately because $\langle j_4\rangle_{\text{Fe}}$ and $\langle j_4\rangle_{\text{Pd}}$ are nearly equal, except for the first few Bragg reflections. The data are consistent with any γ pair having a total E_g/T_{2g} ratio of 0.39 ± 0.02 and a Pd E_g/T_{2g} ratio $\leqslant 0.67$. Thus $(28\pm1)\%$ of the localized moment has E_g symmetry, $(72\pm1)\%$ T_{2g} symmetry.

A three-dimensional Fourier inversion was made[23] using the 20 magnetic scattering amplitudes determined by neutron diffraction and, for the point at $k=0$, by the saturation magnetization. Figure 3 shows the resulting average moment density in $\mu_B/\text{Å}^3$ in the (100) plane ($\frac{1}{4}$ of the base of the fcc unit cell is shown). Figure 4 shows the moment density in the (110) plane. In both figures T_{2g} symmetry is evident near the lattice site occupied by an average atom and the negative moment density associated with the conduction electrons appears in regions far from the lattice site. Termination errors do not significantly alter the features of these maps.

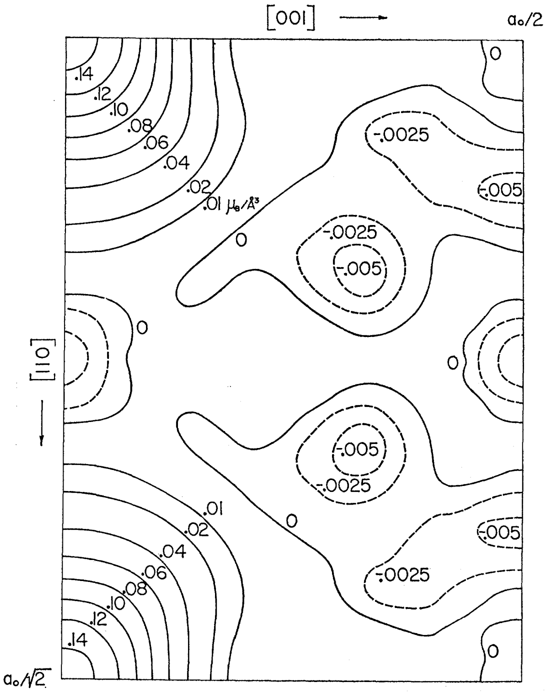

FIG. 4. Magnetic-moment density in Bohr magnetons per Å^3 in the (110) plane.

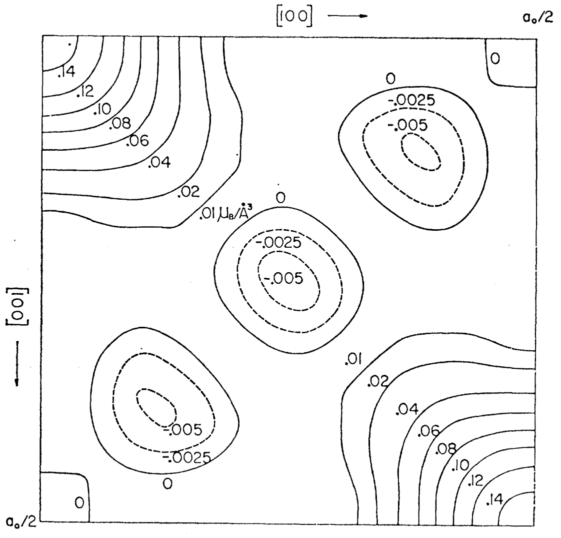

FIG. 3. Magnetic-moment density in Bohr magnetons per Å^3 on the cube face.

TEMPERATURE DEPENDENCE OF THE MAGNETIZATION

The polarization ratio of the (111) Bragg reflection was measured at temperatures between 4 and 114°K in a 14 kOe applied field, and the saturation magnetization was measured from 4 to 200°K in a 17 kOe field.[18] In analyzing the scattering data we have assumed the form factor is temperature-independent over the range of temperatures investigated. This assumption implies that the ratio of $3d$ to $4d$ moment is constant, since the s conduction-electron form factor is negligible at the (111) position. Thus the magnetic contribution to the coherent scattering is proportional to the d moment per atom which is parallel to the applied field, while the saturation magnetization is proportional to the total aligned moment per atom. The d moment per atom is given by

$$\mu_{\text{Fe}}(T)+\mu_{\text{Pd}}(T)=\mu_{\text{d}}(T)=0.138\frac{P_T(111)}{P_{4.2°}(111)}\mu_B, \tag{10}$$

and the total moment per atom by

$$\mu_t(T)=0.114\frac{M(T)}{M(4.2°)}\mu_B, \tag{11}$$

using the values for the d moment (0.138 μ_B per atom)

---

[22] J. W. Cable, E. O. Wollan, and W. C. Kohler, J. Appl. Phys. 34, 1189 (1963).

[23] W. G. Sly, D. P. Shoemaker, and J. H. Van den Hende, ERFR2 Two and Three Dimensional Chrystallographic Fourier Summation Program, SHARE 1344 (unpublished).

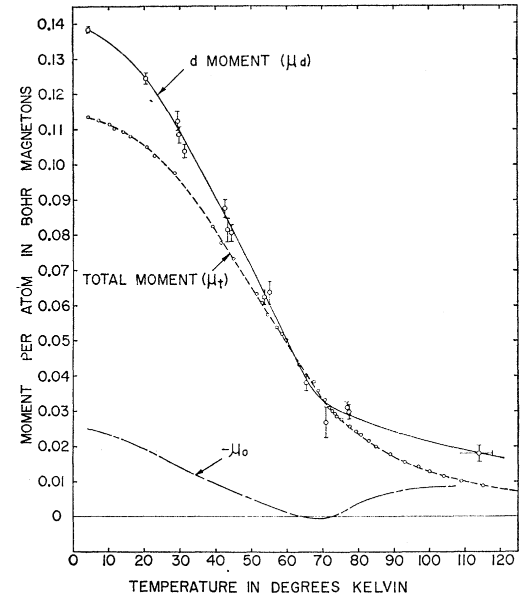

FIG. 5. Temperature dependence of the total moment per atom μ_t (measured in a 17 kOe applied field), the d moment per atom μ_d (measured in a 14 kOe applied field), and the magnitude of the conduction-electron moment per atom, $-\mu_0=\mu_d-\mu_t$.

and saturation magnetization (0.114 μ_B per atom) determined at 4.2°K. $P_T(111)$ is the (111) magnetic scattering amplitude (measured in a 14 kOe field) and $M(T)$ is the saturation magnetization (measured in a 17 kOe field). The measured values of $\mu_d(T)$ are shown in Fig. 5.

In order to examine the coupling between the Pd 4*d* moment and the 3*d* moment localized at impurity sites, the temperature dependence of the total moment and of the *d* moment was calculated on the basis of a simple Weiss molecular field model.[24] A similar calculation was made by Craig *et al.*, who fit their Mössbauer data for the temperature dependence of the hyperfine field of Fe in a very dilute paramagnetic FePd sample to a Brillouin function model.[7] For the paramagnetic system, Freeman has shown that this simple procedure yields the total spin associated with a cluster of Fe and rigidly coupled Pd moments.[25]

Two parameters are involved in calculating the temperature dependence of the magnetization on the Weiss model: the Curie temperature T_c and the total spin J associated with each localized moment or moment cluster (we put $g=2$). Good agreement with the measured temperature dependence of μ_d and μ_t was obtained for $T_c=55\pm5$°K and $J=5.4\pm0.5$. This value for T_c is in agreement with the Curie temperature of 66°K measured by Crangle in a 1.25% Fe FePd sample.[3] The large J value indicates that in our 1.3% Fe alloy the Pd moments are strongly coupled to an Fe 3*d* moment, forming a moment cluster of 10.7±0.6 μ_B about an impurity site. Craig *et al.* observed similar behavior in the very dilute system, where they found $J=13/2\pm3/2$. Although the extent of the Pd polarization around an Fe impurity cannot be determined from our data, we can conclude that the Pd matrix is not uniformly polarized, but instead clusters of moment are present. Recently, Low[26] has found a long-range Pd polarization extending over many-neighbor distances in dilute ($\lesssim1\%$ Fe) alloys.

The magnitude of the difference between the total moment and the *d* moment,

$$\mu_0(T)=\mu_t(T)-\mu_d(T)\,, \qquad (12)$$

decreases as the temperature increases from 4°K to the Curie temperature (see Fig. 5). Again associating μ_0 with conduction-electron moment, we find the conduction-electron polarization disappears near the Curie temperature and is directed opposite to the *d* moment (and the external field) below and above T_c.

ACKNOWLEDGMENTS

I wish to thank Professor C. G. Shull for all the help he has given me, for suggesting this problem and for guiding the experiment. I am grateful to Dr. S. Foner for measuring the saturation magnetization of several specimens, to Dr. A. J. Freeman for making available the calculated Pd form factors before publication and to Dr. R. M. Moon for many useful discussions. The Fourier summations were calculated on the MIT Computation Center's 7094 computer.

---

[24] A. J. Dekker, *Solid State Physics* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1959), p. 466 ff.

[25] A. J. Freeman, Phys. Rev. **130**, 888 (1963).

[26] G. G. Low, in Proceedings of the International Conference on Magnetism, Nottingham, 1964 (unpublished).