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Electrical and Magnetic Properties of Some Cubic Intermetallic Compounds of Plutonium with Ru, Rh, Ir, Pd, and Pt[†]

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The electrical resistivities and magnetic susceptibilities of the cubic intermetallic compounds of the form PuX_2 ($X = \text{Ru, Rh, Ir, Pt}$) and PuX_3 ($X = \text{Rh, Pd, Pt}$) have been measured from 2 to 300 °K. The PuX_3 compounds order antiferromagnetically with Néel temperatures T_N of 6.6, 24, and 40 °K, respectively. The resistivities of the PuX_3 compounds decrease rapidly below T_N , and there is also an abrupt change in the slope of the resistivity-versus-temperature curve at T_N in PuRh_3 .¹ PuPt_2 orders ferromagnetically at a Curie temperature of $T_C = (6.0 \pm 0.3)$ °K, and the resistivity falls dramatically below T_C . PuRh_2 has a temperature-dependent susceptibility but there is no evidence for magnetic order, while the susceptibilities of PuRu_2 and PuIr_2 are temperature independent. However, the resistivities of PuRh_2 , PuRu_2 , and PuIr_2 have anomalous negative curvature at high temperatures, although the slopes remain positive throughout the temperature range investigated. The results are discussed in terms of the interatomic distance between plutonium atoms and the localized-spin-fluctuation model.

I. INTRODUCTION

A wealth of interesting magnetic and transport behavior has been observed recently in the actinide elements and their intermetallic compounds,¹ and the unusual nature of the phenomena in these systems is intimately related to how the actinide 5f electrons respond to a given environment. The results of Nellis and Brodsky² on PuPd_3 and NpPd_3 and of Hill and Elliott³ on actinide-Pt compounds indicate that a study of actinide-transition-metal compounds should yield considerable information on the properties of actinide 5f electrons.

This paper presents the electrical resistivities and magnetic susceptibilities of the cubic intermetallic compounds of the form PuX_2 ($X = \text{Ru, Rh, Ir, Pt}$) and PuX_3 ($X = \text{Rh, Pd, Pt}$). These compounds were chosen because the crystal structures are relatively simple—the MgCu_2 cubic Laves phase and ordered fcc AuCu_3 structures, respectively⁴—and because the transition metals themselves are nonmagnetic. A preliminary account of the resistivity measurements has already been given.⁵ A study of the magnetic properties has also been reported,⁶ but new data on better samples of PuRh_2 and PuPt_3 are included in the present paper as a supplement to the previous investigation. The susceptibility measurements show that the PuX_3 compounds order antiferromagnetically, and this finding is supported by the resistivity results. The

PuX_2 compounds show a spectrum of magnetic behavior: PuRu_2 is strongly paramagnetic, PuRh_2 is nearly magnetic, and PuPt_2 is a weak ferromagnet. The results are discussed in terms of the interatomic distance between plutonium atoms and the localized-spin-fluctuation model.

II. EXPERIMENTAL PROCEDURE

The transition metals used in this study had purities which were typically ~99.995%. The high-purity plutonium was obtained from Los Alamos Scientific Laboratory and the impurities it contained were (in ppm by weight) Mg, 3; Al, 10; Si, 5; Ca, <3; Zn, <5; Am, 55 (on 2-18-71); Fe, 4; Ta, 11; Th, 9; W, 2.5; U, 25; N, 3; C, <10; H, 15; and all others, <2. From these starting materials, 1-g quantities of the intermetallic compounds were prepared by arc-melting appropriate amounts of the constituents in an argon-helium atmosphere. Resistivity samples with the approximate dimensions $1 \times 1 \times 7$ mm were machined out of the buttons with a spark-erosion apparatus, and magnetic-susceptibility samples were prepared from pieces of the remaining material. PuPt_3 , PuPd_3 , PuRh_3 , and PuRu_2 formed congruently from the melt,⁴ and consequently no further heat treatment was given. PuPt_2 , PuRh_2 , and PuIr_2 were given one-week anneals at 1100, 1250, and 1000 °C, respectively. Powder x-ray-diffraction techniques were used to confirm that the desired

compound had in fact been formed. Lattice parameters a_0 determined from the x-ray patterns are listed in Table I, and the values agree quite well with those reported in the literature.⁴ In addition, the absence of extraneous lines in the patterns indicated that with the exception of PuIr_2 , all of the samples were single phase. In the case of PuIr_2 , the intensities of the extraneous lines indicated that 5–10% of the sample was a second phase (possibly Pu_5Ir_4 or Pu_5Ir_3).

Electrical-resistivity measurements were made from 1.5 to 300 °K using the standard dc four-probe technique and an automated system described previously.⁷ With this system, data were recorded at either 20- or 48-sec intervals while the sample warmed at the rate of $\frac{1}{3}$ °K/min; the data were processed using a computer. The susceptibility measurements were made from 2.5 to 300 °K using the Faraday method and an experimental arrangement similar to one described previously.⁸ The susceptibilities $\chi = M/H$ of the PuX_3 compounds, PuRh_2 and PuPt_2 , were obtained by taking data at six fields up to 14.5 kOe, fitting the results to a two-term equation linear in $1/H$, and extrapolating to $1/H=0$. The susceptibilities of PuRu_2 and PuIr_2 were independent of the magnetic field up to 14.5 kOe.

III. RESULTS

A. Magnetic Susceptibility

An account of the magnetic properties has already been given.⁶ Since that time better samples of PuPt_3 and PuRh_2 have been prepared, and their susceptibilities are shown in Fig. 1. The peak in the curve for PuPt_3 is characteristic of antiferromagnetic order with a Néel temperature T_N of 40 °K. The results for PuRh_3 and PuPd_3 were similar, and values of T_N for these samples are listed in Table I. The earlier data for a two-phase sample of PuPt_3 also showed characteristic antiferromagnetic behavior. For reasons not understood at this time, the value of p_{eff} is the same for the two samples, but T_N is now 20 °K lower.

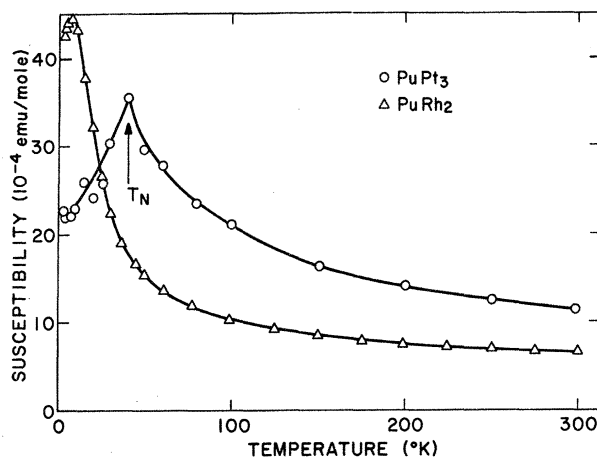


FIG. 1. Magnetic susceptibilities of PuPt_3 and PuRh_2 as a function of temperature.

Recent neutron-diffraction results⁹ confirm that PuPd_3 is a simple antiferromagnet with an ordered moment per plutonium atom at 4.2 °K of $(0.8 \pm 0.1)\mu_B$ and a magnetic lattice similar to that found in NpPd_3 ¹⁰ in which nearest-neighbor neptunium atoms have their moments oppositely directed. The susceptibility data in the paramagnetic region were analyzed by fitting the susceptibilities to an equation of the form $\chi = \chi_0 + C/(T - \Theta_p)$, where χ_0 is a temperature-independent contribution and $C/(T - \Theta_p)$ is the usual Curie-Weiss expression. The effective paramagnetic moment p_{eff} per formula unit (F.U.) was determined from the fitting constant C in the usual way, and values of χ_0 , Θ_p , and p_{eff} for the PuX_3 compounds are listed in Table I.

The susceptibility of PuRh_2 increases with decreasing temperature down to about 10 °K, where a small maximum develops. However, this behavior is not indicative of magnetic order, because the maximum is rounded and because there is no supporting evidence in the resistivity data. The maximum was not seen in the earlier sample, but powder x-ray-diffraction patterns indicated that it

TABLE I. Magnetic parameters of cubic plutonium compounds.

Compound	Ordering temperature (°K)	Θ_p (°K)	p_{eff} ($\mu_B/\text{F. U.}$)	χ_0 (10^{-4} emu/mole)	a_0 (Å)
PuRh_3	$T_N = 6.6$	-63	1.0	6.2	4.008 ± 0.001
PuPd_3	$T_N = 24$	-34	1.0	1.4	4.102 ± 0.001
PuPt_3	$T_N = 40$	-36	1.3	4.9	4.103 ± 0.002
PuRu_2	9.31	7.474 ± 0.001
PuRh_2	...	-49	0.88	3.7	7.488 ± 0.001
PuIr_2	5.75	7.531 ± 0.001
PuPt_2	$T_C = 6$	+6	0.89	2.4	7.652 ± 0.001

was a multicomponent system and not pure PuRh_2 . The other three PuX_2 compounds have magnetic properties which are in marked contrast to PuRh_2 .⁸ PuIr_2 and PuRu_2 have temperature-independent susceptibilities, and their values are listed under χ_0 in Table I. PuPt_2 orders ferromagnetically at a Curie temperature of $T_C = (6.0 \pm 0.3)^\circ\text{K}$ as indicated by plots of M^2 vs H/M , has a magnetization which does not saturate at 2.5°K in fields up to 14.5 kOe, and has a small spontaneous moment of $\sim 0.2 \mu_B/\text{F.U.}$ These results indicate that the magnetism in PuPt_2 is probably itinerant in nature. The high-temperature data for both PuRh_2 and PuPt_2 were also analyzed by the modified Curie-Weiss law, and the various fitting parameters are listed in Table I.

B. Resistivity

The electrical resistivities of the PuX_3 compounds are shown in Fig. 2 as a function of temperature. These materials are metallic in nature with room-temperature resistivities of 125, 90, and $54 \mu\Omega\text{cm}$ for PuPd_3 , PuPt_3 , and PuRh_3 , respectively. The resistivities show strong temperature dependences below T_N . There is also a pronounced change in the slope of the resistivity-versus-temperature curve at T_N in PuRh_3 , whereas the slope changes are more gradual at T_N in PuPd_3 and PuPt_3 . In the temperature range $7 < T < 13^\circ\text{K}$, the resistivity of PuRh_3 varies as $\rho = (16.8 + 0.181T/^\circ\text{K}) \mu\Omega\text{cm}$. Also, it should be noted that the PuPd_3 resistivity curve has a negative slope above 60°K . The dramatic decreases in the resistivities below T_N are most certainly related to a decrease in magnetic scattering of the conduction electrons. A measure of the magnetic scattering ρ_m above T_N was determined for PuRh_3 and

TABLE II. Values of the parameters associated with the resistivities of cubic plutonium compounds.

Compound	ρ_m ($\mu\Omega\text{cm}$)	ρ_0 ($\mu\Omega\text{cm}$)	A ($\mu\Omega\text{cm}/(^\circ\text{K})^2$)	B ($10^{-4} \mu\Omega\text{cm}/(^\circ\text{K})^3$)
PuRh_3	13.8	3.0
PuPd_3	131.3	19.7
PuPt_3	77.3	11.2
PuRu_2	...	10.40	...	5.27
PuRh_2	...	22.80	0.127	...
PuIr_2	...	17.05	...	3.27
PuPt_2	80.0	8.0

PuPt_3 by extrapolating the high-temperature data to 0°K , taking the resistivity value thus obtained, and subtracting the residual resistivity ρ_0 . Because of the negative slope at high temperatures in PuPd_3 , the procedure was modified by subtracting the residual resistivity from the value at the peak. The values obtained for ρ_m are listed in Table II.

The electrical resistivities of the PuX_2 compounds are shown in Fig. 3. These materials are also metallic in nature, with room-temperature resistivities of 117, 113, 112, and $80 \mu\Omega\text{cm}$ for PuRh_2 , PuPt_2 , PuRu_2 , and PuIr_2 , respectively. The resistivity of PuPt_2 shows a pronounced drop as the temperature is lowered through T_C , while the other PuX_2 compounds behave less dramatically. An estimate of the resistivity associated with the ordering in PuPt_2 was made in the manner described above, and the value $\rho_m = 82 \mu\Omega\text{cm}$ was obtained. The resistivity of PuRh_2 varies initially as T^2 , and the results below 12°K are described by the equation $\rho = \rho_0 + AT^2$, where ρ_0 is the residual resistivity and A is a fitting parameter. The values of ρ_0 and A are listed in Table II. The resistivities of PuRu_2 and PuIr_2 show temperature dependences of the form $\rho = \rho_0 + BT^3$ below 16 and 15

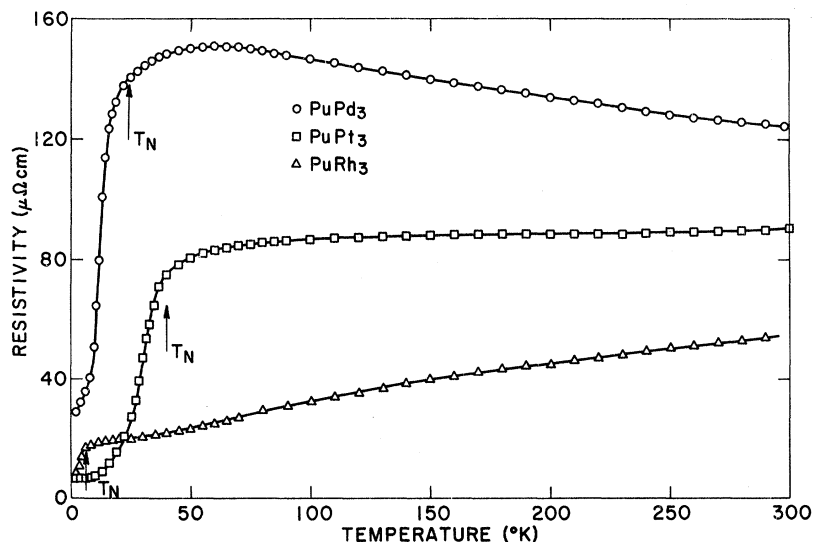


FIG. 2. Electrical resistivities of PuRh_3 , PuPd_3 , and PuPt_3 as a function of temperature.

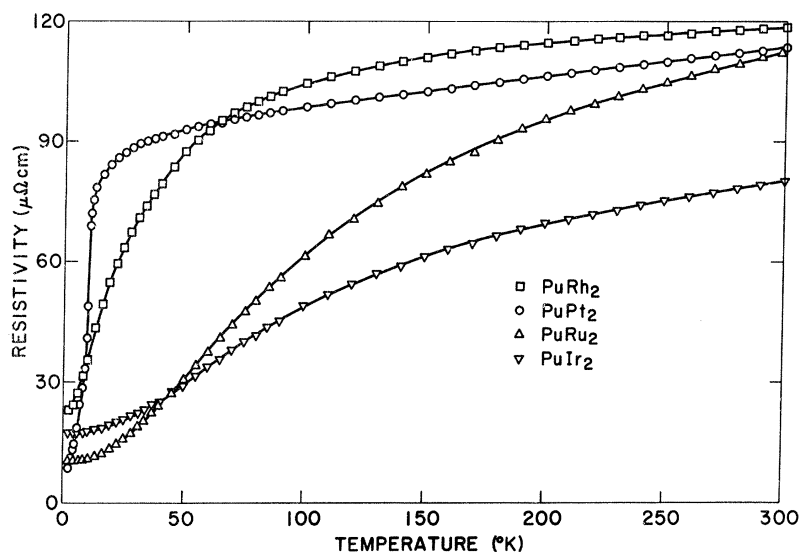


FIG. 3. Electrical resistivities of PuRh₂, PuPt₂, PuRu₂, and PuIr₂ as a function of temperature.

°K, respectively, and values of ρ_0 and B for both samples are also listed in Table II. In addition, the high-temperature resistivity curves of PuRh₂, PuRu₂, and PuIr₂ do not behave in a simple manner in that they show a tendency to bend over; i. e., they have negative curvature. However, the slopes of the curves do remain positive throughout the temperature range covered.

A note of caution should be sounded here with regard to the values of ρ_m and the temperature dependences cited above. ²³⁹Pu is radioactive and damages by self-irradiation. This damage has an effect on the low-temperature resistivity of plutonium,¹¹ and, furthermore, the resistivity caused by the damage does not obey Matthiessen's rule.¹² It is not known at this time what effect the self-irradiation damage has on the compounds discussed in this paper, but an attempt was made to minimize the effect of this phenomenon by measuring the resistivities and susceptibilities as soon as possible after fabrication or annealing.

IV. DISCUSSION

Recent band calculations on the actinides¹³ and on actinide compounds with the NaCl type of structure¹⁴ indicate that the actinide $5f$ electrons give rise to narrow f bands at or near the Fermi level which are strongly hybridized with the conduction band. This leads naturally to the idea that the degree of hybridization and the stability of the f band have a direct bearing on whether or not magnetism is observed in actinide systems. For example, plutonium does not order magnetically,¹⁵ whereas in actinide compounds with the NaCl type of structure, local-moment behavior is observed.¹⁶ These considerations correlate with Hill's qualitative argument¹⁷ concerning the relation between the in-

teratomic actinide distance and the appearance of magnetic order. In particular, Hill indicates that if the interatomic plutonium-plutonium distance $d_{\text{Pu-Pu}}$ is less than the critical value of $\sim 3.4 \text{ \AA}$, nonmagnetic behavior is observed because of f - f overlap (the average $d_{\text{Pu-Pu}}$ in α -Pu is $\sim 3.2 \text{ \AA}$), but if $d_{\text{Pu-Pu}}$ is greater than 3.4 \AA , long-range magnetic order can be expected. In the PuX₃ compounds studied here, $d_{\text{Pu-Pu}} = a_0$, the lattice parameter, and is about 4.1 \AA . In the PuX₂ compounds, $d_{\text{Pu-Pu}} = \frac{1}{2} a_0 3^{1/2}$ and is about 3.3 \AA . Consequently, on this simple basis, long-range magnetic order in the PuX₃ compounds is expected. The PuX₂ samples have a $d_{\text{Pu-Pu}}$ very close to the critical value, and here the situation is not as clear. However, it seems reasonable to assume that the tendency to exhibit magnetism is less strong in the PuX₂ compounds, and the conclusion that the magnetism observed in PuPt₂ has itinerant rather than local-moment character is in agreement with the arguments for f -band formation discussed in Refs. 13, 14, and 17. Since the value of $d_{\text{Pu-Pu}}$ is so close to the critical value, it is not surprising that the magnetic behavior of the PuX₂ compounds is very sensitive to the particular transition metal used.

The susceptibility measurements on the PuX₃ compounds indicate that the transition element has little effect on the magnetic order except to shift T_N . This is in contrast to the case for the PuX₂ compounds and probably results from the fact that $d_{\text{Pu-Pu}}$ is so much larger than the critical value. The susceptibility data also indicate that the f electrons are reasonably well localized in the PuX₃ compounds. The fits of the data to $\chi = \chi_0 + C/(T - \Theta_p)$ were quite good, and values of $p_{\text{eff}}/F.U.$ were all about $1 \mu_B/F.U.$ The latter result implies that the plutonium atoms are in the same electronic

configuration for all three samples; i. e., assuming that the plutonium atoms are trivalent and in the $5f^5$ configuration, application of Hund's rules yields paramagnetic and ordered moments of $0.85\mu_B$ and $0.72\mu_B$, respectively. The ordered moment is in good agreement with the neutron-diffraction results⁹ on PuPd_3 which indicate an ordered moment per F.U. of $(0.8 \pm 0.1)\mu_B$. However, since ρ_{eff} for PuPt_3 is slightly greater than ρ_{eff} for PuRh_3 and PuPd_3 , some of the moment may lie on the platinum atoms, or crystal field effects may be entering. Neutron-diffraction and Mössbauer data are needed to obtain information on the ordered moments and to resolve these questions. The resistivity results also indicate local-moment behavior in the PuX_3 compounds. The sharp resistivity drops below T_N are similar to the spin-disorder resistivities in the heavy rare-earth metals¹⁸ in which local moments are well formed. However, note the differences between the values of ρ_m in the PuX_3 compounds. If the plutonium atoms are in the same electronic configuration, then the coupling between the conduction electrons and the f electrons is sensitive to the nonactinide element.

There remains the problem of the negative slope of the resistivity-versus-temperature curve of PuPd_3 above 60°K . This behavior can be explained if the temperature dependence of the Fermi level is taken into account and the band structure of the material has special properties.¹⁹ However, the authors favor an alternative approach based on the localized-spin-fluctuation (LSF) model²⁰⁻²³ which has been successful in explaining the results on several actinide systems.^{1,15(a),24-27} Briefly, the model for intermetallic compounds²¹ predicts that the resistivity should drop to zero at $T=0^\circ\text{K}$ because in atomically ordered materials the f band hybridizes with the conduction band to generate a Fermi-surface structure. As T is increased from 0°K , the conduction electrons begin to scatter from spin fluctuations in the f band and the resistivity rises rapidly, initially as T^2 , and then goes into a region linear in T . However, because of the high concentration of actinide atoms, the scattering is quite strong and has a drastic effect on the mean free paths of the f electrons themselves. As a result, in the regime $T > T_{\text{sf}}$, where T_{sf} is the characteristic spin-fluctuation temperature, the scattering is self-limiting, and the resistivity will level off or even decrease with temperature. In this approach, the self-limiting aspect of the spin-fluctuation scattering accounts for the negative slope of the resistivity-versus-temperature curve of PuPd_3 , and could explain the negative curvature of the resistivity-versus-temperature curves of PuPt_3 and PuRh_3 at high temperatures as well. Further evidence that the LSF model applies to all

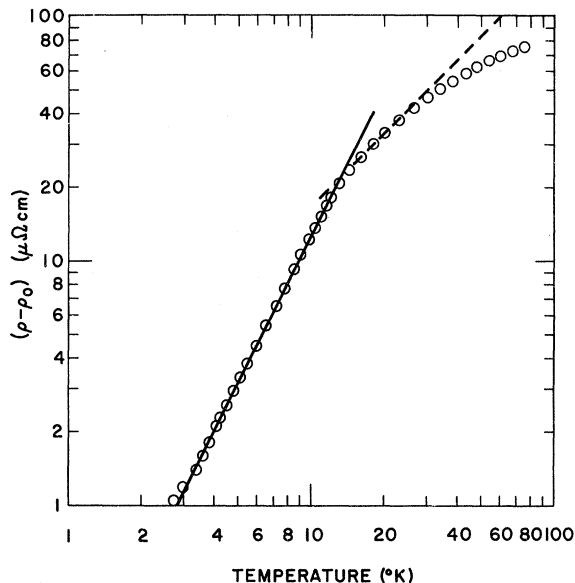


FIG. 4. Bilogarithmic plot of the resistivity of PuRh_2 as a function of temperature.

of the PuX_3 compounds is the linear variation with temperature of the resistivity of PuRh_3 above T_N and, more qualitatively, the rounded behavior of the resistivity-versus-temperature curves of PuPd_3 and PuPt_3 in the vicinity of T_N . Implicit in this interpretation is the suggestion of Buschow and van Daal²⁵ that $T_{\text{sf}} < T_N$ and that spin fluctuations are quenched in the magnetically ordered state below T_N .

In the PuX_2 compounds, PuRh_2 appears to be a good candidate for the LSF model. Figure 4 shows a bilogarithmic plot of the resistivity-versus-temperature curve for PuRh_2 , and the solid and dashed curves show the regions of T^2 and T behavior, respectively. Assuming that $T_{\text{sf}} \approx T_1$, the upper limit of the T^2 region of the resistivity, $T_{\text{sf}} \approx 12^\circ\text{K}$ for PuRh_2 . The Curie-Weiss susceptibility above 30°K is indicative of the magnetic behavior expected at temperatures above T_{sf} . The LSF model may also be applicable to PuPt_2 in the same way that it is for the PuX_3 compounds, and if this is the case, $T_{\text{sf}} < 6^\circ\text{K}$ for PuPt_2 . The large temperature-independent magnetic susceptibilities of PuRu_2 and PuIr_2 indicate a broad, hybridized $6d-5f$ band with little structure and a high density of states at the Fermi level. The T^3 temperature dependences of the resistivities of PuRu_2 and PuIr_2 are characteristic of the $s-d(f)$ interband scattering²⁶ that could be expected in such materials. The strong negative curvature of the resistivity-versus-temperature curves of PuRu_2 and PuIr_2 at high temperatures is similar to the behavior of other actinide metals and compounds. However,

the origin of this strong curvature is at present not understood. The absence of a contribution to the resistivity, which varies as T^2 , argues against the LSF model, and the magnetic susceptibilities do not vary with temperature in the manner that would be expected if the thermal smearing explanation were appropriate.

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Refraction of Thermal Neutrons by Shaped Magnetic Fields*

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The refractive bending of thermal neutrons by shaped magnetic fields has been studied experimentally and compared with that calculated from the field distribution. The small deflections, a few seconds arc, were measured on a double-crystal spectrometer utilizing matched perfect silicon crystals. Deflections were measured for a beam of polarized neutrons upon its polarization reversal for field regions of both prismatic and cylindrical configuration. The focusing and polarizing character of a magnetic lens have been observed.

I. INTRODUCTION

When slow neutrons carrying a magnetic moment μ are introduced into a region of space where

a magnetic field B is present, there occurs a Zeeman splitting of the potential energy of magnitude $\pm \mu B$, corresponding to the two quantized spin states of the neutron. This occurs at the expense