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MAGNETIC PROPERTIES OF ACTINIDE SYSTEMS

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ABSTRACT

In the six years since the last Durham Conference a large number of experiments have been performed on actinide systems. The early expectations that the proximity of the 5f band to the Fermi level would lead to complex magnetic (or almost magnetic) behaviour were correct. Well documented examples of spin-fluctuation systems, itinerant and localized magnetic systems now exist. Rather than attempt a complete survey of the experimental results, this review will focus on three systems of major importance ; (a) the elements, (b) Laves phase compounds, and (c) NaCl-type compounds. In (a) the main advances have come in theories to explain the unusual resistivities of the elements and in the positive identification that curium metal is antiferromagnetic with $T_N = 52^\circ\text{K}$. In (b) experiments on materials such as UAl_2 have shown "idealized" spin fluctuation behaviour whereas in the Np compounds simple arguments related to the Np-Np spacing lead to at least a qualitative understanding of the magnetic behaviour. In AmFe_2 we may have detected the first example of a mixed valence state, $\text{Am}^{2+}-\text{Am}^{3+}$. Finally a number of "second generation" experiments on actinide rock salt compounds will be reviewed. Contrary to earlier ideas, not all these systems can be understood with a localized model. This is especially true of the uranium compounds with small lattice spacings like UN, US and probably UC. On the other hand, for USb and compounds with transuranium elements the localized model will probably apply, e.g. PuP and AmSb. The advent of single crystals in some of these systems gives hope that a number of important and exciting experiments will be performed in the near future.

Elements

Because of their complex crystallographic phases and almost magnetic character, the actinide elements have been of major interest over the years. Speculation that both uranium and plutonium ordered magnetically appeared in the literature, but was usually based on rather indirect evidence from resistivity or susceptibility measurements. We now believe that none of the actinide elements lighter than curium is magnetic. The unusual resistivity behavior shown in Fig. 1 has been extensively discussed (Brodsky et al. 1974, Fradin 1976).

At low temperatures the theory of spin fluctuations can account for the T^2 temperature dependence in the metals and in a number of other nearly magnetic actinide systems (Arko et al. 1972). In these, the $5f$ electrons form a relatively narrow conduction band with little localized character. The magnetic moments do not have lifetimes long enough to form a stable magnetic configuration, but are able to scatter the conduction electrons and thus contribute to the resistivity. At higher temperatures the spin-fluctuation (paramagnon) contributions are still important, Jullien et al. (1974), but cannot by themselves reconcile the temperature dependence of the resistivity with the almost temperature independent susceptibility. An alternative explanation deals with the effect of sharp structure in the electronic density of states. Fradin (1974) and Fradin et al. (1975) have shown how these effects contribute to the resistivity of almost magnetic materials such as Pd and Pt.

For curium, magnetization measurements of Kanellakopoulos suggest an antiferromagnetic transition, $T_N = 52^\circ\text{K}$, and Fournier et al. (1977) has verified this by neutron diffraction. Unfortunately, this first neutron experiment did not provide a solution to the antiferro structure of the metal, so that another

one needs to be attempted. The isotope used, ^{244}Cm is a strong α and neutron emitter ($t_{1/2} \sim 17$ yrs) so these experiments are not easy.

Laves-phase systems

The magnetic properties of a large number of actinide Laves phases (C-15 crystal structure) have been studied over the last few years. This structure is not only extremely common in intermetallic compounds but the close packing, and especially the nearest-neighbor actinide separation, indicates that direct overlap of the $5f$ wave functions may occur. Some years ago, Hill (1971) suggested that the occurrence of magnetic ordering in actinide intermetallics could be correlated with a critical actinide-actinide separation. Although a number of important exceptions exist, the neptunium Laves phases provide strong support for this concept. Some of the magnetic properties of these systems are given in Table I. Initial experiments on these compounds, Aldred et al. (1974), suggested that as the actinide-actinide separation d_{An} is decreased, the magnetism changes from local-moment (NpAl_2) to itinerant behavior (NpOs_2 and NpIr_2) and finally to paramagnetism with no long-range order (NpRu_2).

High-field magnetization, Mössbauer, and neutron-diffraction experiments, Aldred et al. (1975) and (1976), show quite clearly the difference between NpAl_2 , a localized $5f$ system, and NpOs_2 , an itinerant ferromagnet. Specific-heat measurements (Brodsky & Trainor, 1977) have also demonstrated the itinerant nature of the magnetism in NpOs_2 . However, this concept in which the magnetic order is lost as the interatomic distance decreases and the narrow $5f$ levels are broadened by f - f interaction is certainly an oversimplification. Perhaps the most interesting exceptions are the compounds UAl_2 and PuAl_2 . From d_{An} one might expect localized behavior, as is found for NpAl_2 , but this is not the case. Both have been the subject of extensive investigations, Trainor et al. 1975, Brodsky and Trainor, 1977, Fradin (1976). In UAl_2 the predictions of

spin-fluctuation theory appear to be well satisfied and the resistivity, magnetization, and specific heat behavior are well understood. In PuAl_2 the situation is certainly more complex and the very sharp rise in the resistivity at low temperatures is still not understood.

Returning now to the ordered compounds listed in Table I, there are a number of interesting points. (1) In NpAl_2 a discrepancy exists between the moments obtained by neutron and Mössbauer measurements ($1.50 \pm 0.05 \mu_B/\text{Np atom}$) and the magnetization value of $1.21 \pm 0.02 \mu_B/\text{Np}$. It seems likely that it is due to polarization of the $7s-6d$ electrons. The first two techniques measure the localized moment (gJ) whereas the magnetization measures the total moment $\bar{\mu}_0 = gJ - 2s$, where $2s$ is the conduction-electron polarization (cep). As we shall see, this effect also occurs in a number of NaCl ferromagnets. Unfortunately because of the large anisotropy in NpFe_2 and PuFe_2 the values of $\bar{\mu}$ are unreliable and similar comparisons cannot be made. (2) In PuFe_2 the magnetic form factor is almost independent of scattering angle and this shows unambiguously that the electronic state is Pu^{3+} , $5f^5$. This interesting property of the cross section from f^5 systems is further discussed in connection with PuP below. (3) In AmFe_2 a small moment of $-0.4 \mu_B$ is found on the Am site, the negative sign indicates it is antiparallel to the large iron moment. If the Am ion is trivalent, the ground state ($f^6: {}^7F_0$) is nonmagnetic and the energy gap between the 7F_0 and excited 7F_1 state is 2700 cm^{-1} , so that the moment induced by the internal exchange field on the Am^{3+} ion should be essentially zero. However, any ions with the Am^{2+} configuration have a ground state ($f^7: {}^8S_{7/2}$) that readily supports a magnetic moment. Further evidence to support this hypothesis of a mixed-valence situation comes from the sign of the Am moment. As discussed by Taylor (1971) in the lanthanide-iron Laves phases, the transition metal moment is coupled antiparallel to the lanthanide (or actinide) spin component S . Thus, for $J = L - S$ ions Laves phase compounds are ferromagnetic (as is found for UFe_2 , NpFe_2 , and PuFe_2), whereas for $J = L + S$ ions, the lanthanide and $3d$ moments are aligned antiparallel (ferrimagnetic). Inasmuch as the f^7 state

has J parallel to S , the ferrimagnetic arrangement in AmFe_2 is compatible with a partial occupancy of the $\text{Am}^{2+}:f^7$ state, the time and space-averaged configuration being $0.95 \underline{5f^6} + 0.05 \underline{5f^7}$.

NaCl type compounds

The NaCl type compounds have been the most extensively studied of any actinide systems and we present data on these systems in Table II. For those entries without references see Lam & Aldred (1974) and Lander & Mueller (1974b). For the uranium compounds see Kuznietz (1971) and Grunzweig-Genossar et al. (1968). Despite this wealth of information the main question as to their electronic structure remains unresolved. Attempts, for example, to determine the ionicity from the lattice parameter, which is a reliable indicator for the corresponding lanthanide compounds, does not succeed (Charvillat et al. 1976). All compounds are metallic with resistivities ranging between 100 and 2000 $\mu\Omega$ cm.

As expected, the curium compounds have a large effective moment and probably have a $\underline{f^7}$ configuration. A study of the anisotropy, which should be very small, and the saturated moments ($\sim 7\mu_B$) would confirm this ground state. The americium compounds exhibit a tendency to have non-magnetic ground states, which is compatible with $J = 0$. In plutonium compounds trivalent Pu^{3+} has a $\underline{5f^5}$ configuration that, in the Russell-Saunders approximation, has a small value of J ($=5/2$) due to the cancellation of a large orbital term ($L=5$) with the antiparallel spin ($S=5/2$). As a result of this partial cancellation of L and S the elastic neutron scattering cross section has an unusual shape, with maximum not at $K = 0$, (\vec{K} , the scattering vector, is given by $|\vec{K}| = 4\pi\sin\theta/\lambda$ where θ is the scattering angle and λ the neutron wavelength) but at some finite value. This is illustrated in Fig. 2 which shows results obtained by Lander & Lam (1976) for PuP. Another interesting point of this study is that the saturation magnetization value is $0.42 \mu_B$ (Lam et al., 1969) whereas the neutron experiments,

which sense the localized $5f$ moment only, find $0.77 \mu_B$. The discrepancy of $-0.35 \mu_B$ may be a consequence of the polarization of the $6d - 7s$ electrons. This effect is also found in $NpAl_2$ and US. In neptunium compounds the magnetic behavior is rather complex, although the excellent correlation between the saturated moment as determined by neutron diffraction and the hyperfine field measured in Mössbauer experiments strongly suggests that $Np^{3+}:5f^4$ is the correct configuration (Dunlap & Lander 1974). In uranium compounds, on the other hand, the task of specifying the f^n configuration is much more difficult ; and it may be more appropriate to consider a band structure approach. By attempting to characterize the $5f$ electrons as localized we are, of course, assuming that the $5f$ bands have a narrow energy distribution in momentum space and that their energies lie well below E_F . Extensive band structure calculations, Freeman & Koelling (1974) & Davis (1974), show that this is not true, particularly for the lighter actinides. The presence of f electrons near E_F contributes to the peak in the density of states, which is compatible with the high values for the electronic specific heats of UN, UP and US.

The task of elucidating the electronic structure and understanding these compounds from a microscopic point of view remains a formidable task. We are now moving from an era of straight forward magnetic (and lattice) properties to one of "second-generation" experiments. It seems worthwhile to mention some of these without attempting a comprehensive treatment.

Advanced Experiments

(1) Lattice distortions.

Lander & Mueller (1974a) first drew attention to the fact that the actinide antiferromagnets seem to be cubic below T_N , whereas the ferromagnets all show large distortions at T_C . The reasons for this correlation remain obscure ;

it is certainly quite different from the lanthanide series, and probably reflects a strong anisotropic spin-lattice interaction. An interesting idea is that the "internal distortion" found in UO_2 (Faber & Lander 1976) may occur in other systems. Such an idea awaits comprehensive phonon studies with single crystals.

(2) Elastic constants and phonons.

The above considerations are clearly related to the lattice properties and the few elastic constant measurements on these systems show a number of interesting anomalies. Data are now available for UN (du Plessis and van Doorn, 1977) and US (du Plessis, private communication). Phonon studies of both polycrystalline, Wedgwood (1974), and UN single crystal samples, Buyers et al. 1976, show very strong optical modes with almost no dispersion. These features of the phonon spectrum are difficult to relate to the magnetic behavior, but the large vibrations of the anion sublattice will certainly add a dynamic term to the crystal-field potential.

(3) Magnetic anisotropy.

Single-crystal measurements on US (Tillwick and du Plessis 1976) have demonstrated the extremely large anisotropy in this material and estimate an anisotropy field $H_A \sim 1000$ KOe. Similar measurements are now in progress on UTe (Vogt, private communication) and also demonstrate strong anisotropy. Such effects can only be associated with a localized moment and its associated orbital moment. These measurements illustrate the importance of the magnetoelastic term in the general Hamiltonian, and this makes the absence of a spontaneous distortion at T_N in the antiferromagnets all the more puzzling.

(4) Critical effects.

Since actinide systems exhibit considerable anisotropy, the detailed behavior near the ordering temperature is of particular interest. Tillwick and du Plessis (1976) have studied the magnetization of US single crystals and find

values of the critical exponents in approximate agreement with theory. Recently we have performed critical neutron scattering experiments on a single crystal of USb and shown the strong anisotropy in the magnetic correlations of this cubic system. Briefly, the experiment consists of measuring the intensity of the diffuse scattering near the Bragg point ($q = 0$) of the system. The spatial dependence of the elastic cross section is then given by

$$(d\sigma/d\Omega) \propto \chi^{\alpha\beta}(\vec{q})$$

where $\chi^{\alpha\beta}(\vec{q})$ is the wavelength dependent susceptibility and $\alpha\beta$ are cartesian coordinates. A simple model for the susceptibility is the Ornstein-Zernicke form

$$\chi(q) \propto \frac{1}{K_1^2 + q^2}$$

where K_1 is an inverse correlation range such that the static distribution function, or the time-averaged correlation of spin components, is

$$G(R) = \langle \Delta S_m^\alpha \cdot \Delta S_n^\alpha \rangle \propto e^{-K_1 R/R}$$

where R is in real space. In Fig. 3 we show scans for USb just above the ordering temperature $T_N = 241.2^\circ\text{K}$. The magnetic structure of USb is type I, in which ferromagnetic (001) planes are stacked in the simple alternating sequence $+-+-$ etc. with the spin direction parallel to $|001|$. The absence of critical scattering at the point (001), see Fig. 3(c), implies negligible transverse XY spin correlations so the system is truly Ising in its behavior. Furthermore, scans around the (110) point, which may be identified with longitudinal ZZ fluctuations, show considerable anisotropy (the two K_1 values differ by about a factor of 5). If one is considering the correlation within the (001) plane, these are long-ranged and thus have a small K_1 , as given in Fig. 3(a),

whereas the interplanar correlations are much weaker and have a larger K_p , as can be seen in Fig. 3 (b). Further details are beyond the scope of this article, but this behavior is quite unusual in a cubic compound and certainly more experiments need to be performed.

(5) Magnetic Excitations and Crystal Fields.

Of all the questions raised in ordered actinide intermetallic systems perhaps this is the most intriguing. A detailed documentation has been given by Lander (1977) and will not be repeated here. The essential point is that despite a number of experiments - those of Wedgwood (1974) being the most comprehensive - no indication of well-defined crystal-field levels have been found by neutron inelastic scattering. Perhaps even more surprising was the failure to observe spin-waves in UN single crystals (Buyers et al. 1976). Recent experiments on single crystals of USb at the Institute Laue-Langevin in Grenoble have found some evidence of magnetic excitations in this system and these investigations are to continue.

(6) Dilute systems.

Since the magnetic interactions in the actinide systems are strong (ordering temperatures often exceed 100°K) one way to reduce these effects is by dilution with thorium. Such methods have been very successful in separating the crystal-field (electrostatic) and magnetic interaction potentials in lanthanide systems, since only the latter should change on dilution. De Novion & Costa (1972) have studied the UN-UC system and Danan et al. (1976) have reported extensive studies on the US-ThS system. In this latter system they are unable to find evidence to support a localized $5f^2$ configuration and favour a band description.

(7) Magnetization densities.

The neutron elastic magnetic cross-section is sensitive to both the radial and angular parts of the $5f$ wave function (Freeman et al., 1976). The radial part is of interest because it is related to the extent of localization of the $5f$ electrons ; see, for example, Aldred et al. (1976). The angular part is related to the precise shape of the magnetization density in the solid and is thus able to distinguish between different ground states. Let us consider the simple case of prolate and oblate distributions as shown in Fig. 4. The effective magnetic form factor may be written

$$f'(\vec{K}) = \langle j_0 \rangle + c_2 \langle j_2 \rangle + c_4 \langle j_4 \rangle + c_6 \langle j_6 \rangle$$

where the c_i coefficients are functions of the spherical angles θ and ϕ , but not of the scattering vector \vec{K} . The functions $\langle j_i(K) \rangle$ are smoothly varying functions of K and represent the radial distribution of the $5f$ electrons, Freeman et al. (1976). If we focus on the asphericity of magnetization density and consider only terms in the dipole and quadrupole moments, we can define a quantity

$$\Delta f = f'(\vec{K}_1) - f'(\vec{K}_2) = \{C_2(\theta_1) - C_2(\theta)\} \langle j_2(K) \rangle$$

where $|\vec{K}_1| = |\vec{K}_2| = K$ and $\theta_1 < \theta_2$. Such experiments have defined the ground state wave function of USb (Lander et al. 1976). Somewhat surprisingly the wave function appropriate to the $U^{3+}: 5f^3$ ion is not that expected from the free-ion configuration, nor is it that found for the analogous $4f^3$ compound NdSb. Similar experiments on US by Wedgwood (1972) are more difficult to interpret because the experimental values of Δf are very small, Freeman et al. (1976).

(8) de Haas van Alphen effect.

Measurements of the dHvA effect give important information on the wave functions in the crystal. Unfortunately, these experiments require almost perfect single crystals, but an experiment has been successfully completed on the system URh₃ (Arko et al. 1975). This compound is paramagnetic with strong

hybridization between the $5f6d$ states on the uranium atom and the rhodium $4d$ electrons. This technique is especially useful in that to interpret the experimental data requires input from band theory, thus providing close collaboration between experiment and theory.

(9) X-ray photo-emission spectroscopy

The central question in much actinide research relates to the position of the $5f$ electrons with respect to the Fermi edge. If these are close together then we expect interaction with the $6d-7s$ conduction electrons, and a band description of the total electron system to be more appropriate ; whereas if they are well below E_F , like the $4f$ electrons in lanthanide systems, then we expect localized electron behavior. In principle, XPS measurements can answer this question directly as Veal & Lam (1974) have shown for the oxide systems. However, this technique is very sensitive to the top few surface layers of the material and the high reactivity of actinides presents a preparation problem. Attempts are presently being made to prepare thin film samples in situ by evaporation techniques and to improve the instrumental resolution so that these questions can be answered.

Conclusion

In this paper I have attempted, firstly, to review the progress in our understanding of the magnetic properties of some actinide systems since Durham 1971. At that Conference Merwyn Brodsky was able to cover almost all active actinide research efforts. This is no longer possible in the time available, which shows the growing activity in this field. Secondly, I have attempted to outline some so-called "second generation" efforts in actinide research. These experiments are very stringent in their sample requirements, for example, six out of the nine need single crystals, so it is not surprising that few experiments have been performed in the past and those only on uranium

compounds. Extending these to other actinide systems presents a formidable challenge. Fortunately, the rewards of these experimental studies have been sufficient that strong motivation for such efforts exists in a number of laboratories.

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TABLE I

Summary of magnetic properties of some actinide Laves-phase compounds.

d_{An} is the actinide-actinide interatomic distance, of those that order $NpIr_2$ is antiferromagnetic, the others ferro ; $\bar{\mu}_0$ is the total moment deduced from magnetization experiments ; the individual moments determined by neutron diffraction are μ_{An}^N and μ_{Fe}^N .

Compound	d_{An} (Å)	order temp (°K)	$\bar{\mu}_0$	μ_{An}^N	μ_{Fe}^N
UAl ₂	3.38	-			
NpRu ₂	3.23	-			
NpIr ₂	3.25	7.5	~0.6		
NpOs ₂	3.26	7.5	0.44	0.25	-
NpAl ₂	3.37	57	1.21	1.50	-
PuAl ₂	3.39	-			
UFe ₂	3.06	~160	1.11	0.06	0.59
NpFe ₂	3.09	~500	~2.7	1.09	1.35
PuFe ₂	3.11	~600	~2.3	0.45	1.47
AmFe ₂	3.16	~600	3.13	-0.4	1.7

TABLE II. Properties of actinide NaCl compounds. TIP-temperature independent paramagnetism, AF-antiferromagnet, F-ferromagnet, μ_{eff} is the effective moment in paramagnetic state, μ_{sat} the saturated moment, the quantity $(1-c/a)$ refers to a lattice distortion associated with ordering.

Compound	a_0 (Å)	Magnetism	Order temp (°K)	$\mu_{\text{eff.}}$ (μ_B)	μ_{sat} (μ_B)	Easy Axis	$(1-c/a)$ ($\times 10^4$)	f^n	Ref.
UC	4.96	TIP							
UN	4.89	AFI	53	3.1	0.75	<100>	-6		a
UP	5.59	AFI	125 23	3.2	1.7 1.9	<100>	<5	3?	
UAs	5.78	AFI IA	127 63	3.4	1.9 2.2	<100>	<5	3	a
USb	6.20	AFI	241	~3.8	2.8	<100>	<2	3	b
US	5.49	F	178	2.3	1.7	<111>	+105	2?	c
USE	5.74	F	160	~2.4	2.0	<111>	+81		
UTe	6.16	F	104	~2.8	2.2	<111>			
NpC	5.00	AFI F	310 220	3.4	2.1	<100> <111>	<5 +23	4?	d
NpN	4.90	F	87	2.4	1.4	<111>	-52	4	e
NpP	5.62	AF 3+, 3-	130	2.8	1.8 2.3	<100>	-42	4	e
NpAs	5.84	AF 4+, 4- AFI	175 142	~2.6	2.5	<100>	-8 <3		d, e
NpSb	6.25	AFI	207	2.3	2.5	<100>	<15	4	e
NpS	5.53	AFI1	20	2.2	0.9	<100>	<5	4?	d

TABLE II continued

Compound	a_0 (Å)	Magnetism	Order temp (°K)	$\mu_{\text{eff.}}$ (μ_B)	μ_{sat} (μ_B)	Easy Axis	(1-c/a)	f^n	Ref.
PuC	4.98	AFI	~100	~1	0.8	<100>		5	
PuN	4.91	AF	13	1.1				5	
PuP	5.66	F	126	1.1	0.77	<100>	-31	5	f
PuSb	6.24	F	85	1.0	0.6			5	
PuS	5.54	TIP						6	
AmN	5.00	TIP ?						6	g
AmSb	6.24	TIP						6	
CmN	5.04	F	109	7.0				7	g
CmAs	5.90	F	88	6.6				7	g

Ref. : a - Marples et al. (1975), b - Lander et al. (1976), c - Wedgwood (1972), d - Lander and Mueller (1974),

e - Aldred et al. (1975), f - Lander and Lam (1976), g - Kanellakopoulos et al. (1976).

Figure Captions

- Fig. 1 Electrical resistivity of pure actinide metals (Brodsky et al., 1974). The points are based on a model calculation for Np (Fradin 1976).
- Fig. 2 Experimental points for the magnetic cross section from ferromagnetic ^{242}PuP at 4.2 K. The solid and dashed curves are the best fits to the data with $5f^5$ and $5f^4$ configurations, respectively. The arrow on the ordinate axis gives the total moment as determined by magnetization measurements. The insert shows the relativistic Dirac-Fock values for $\langle j_0 \rangle$ and $\langle j_2 \rangle$.
- Fig. 3 (a) and (b) are reciprocal lattice scans in different directions through the (110) point of a USb single crystal just above the ordering temperature. These scans illustrate the anisotropy of the critical scattering. The FWHM of the instrumental resolution function is the same for both scans. In (c) is shown a scan through the (010) point, at which the transverse critical fluctuations should be seen, but these are negligible.
- Fig. 4 Schematic representation of magnetization densities and magnetic form factors for oblate and prolate densities. Δf is defined in the text.

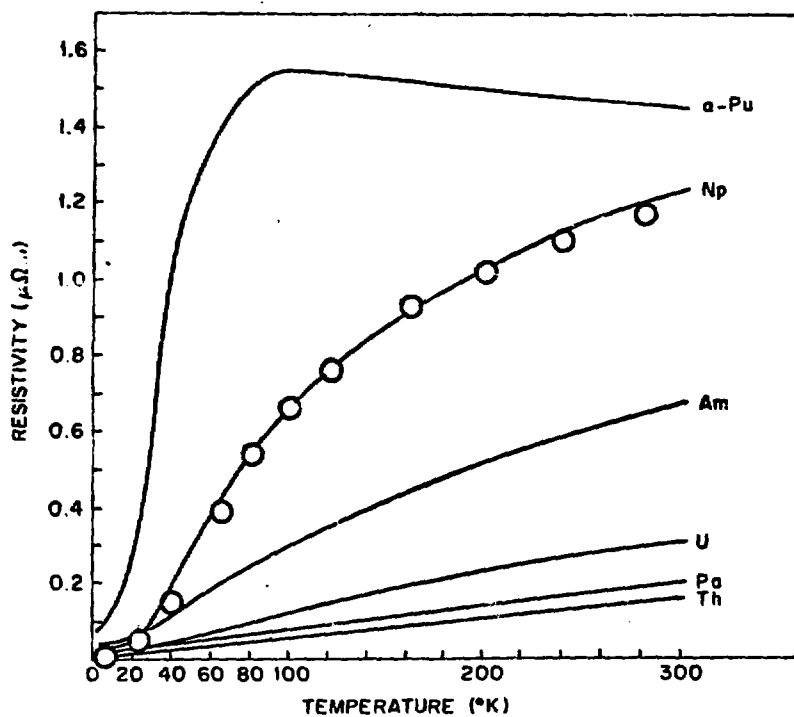


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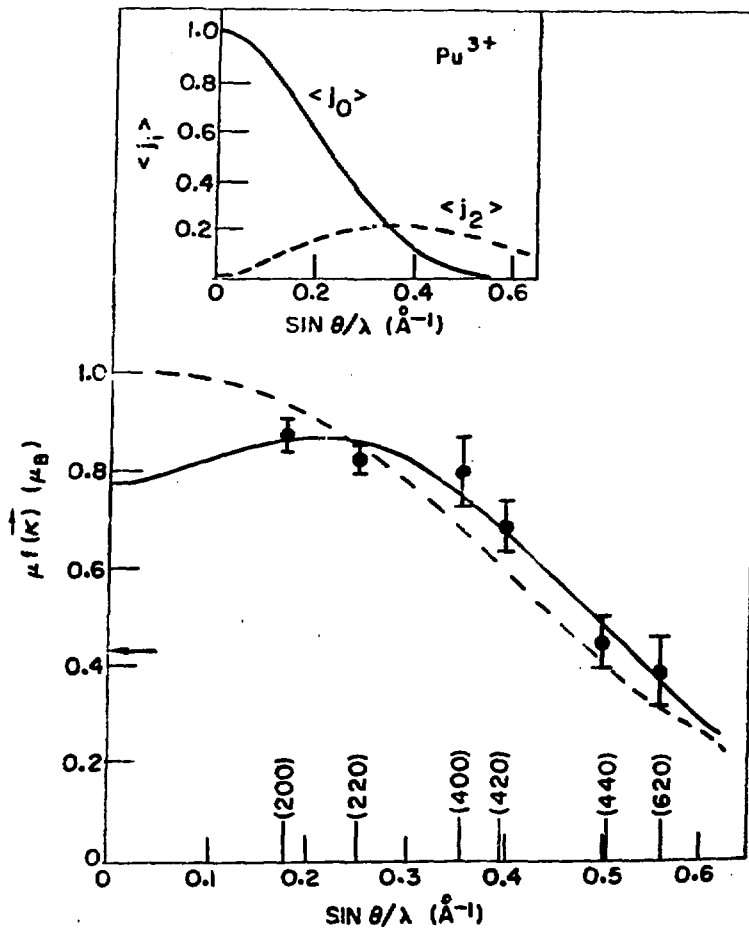


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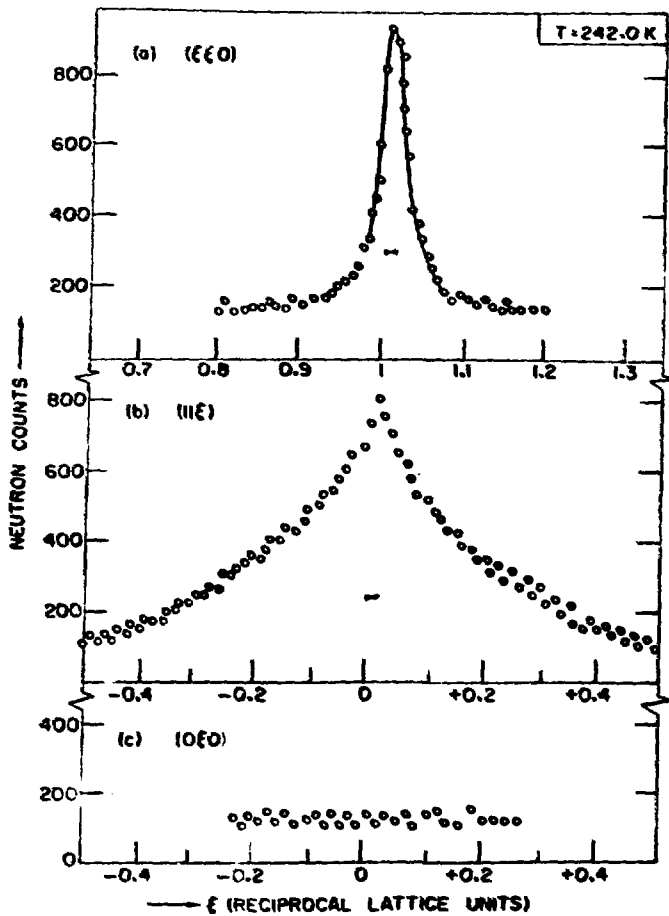
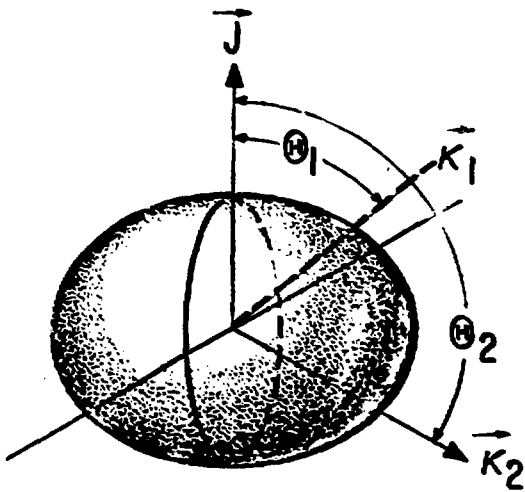
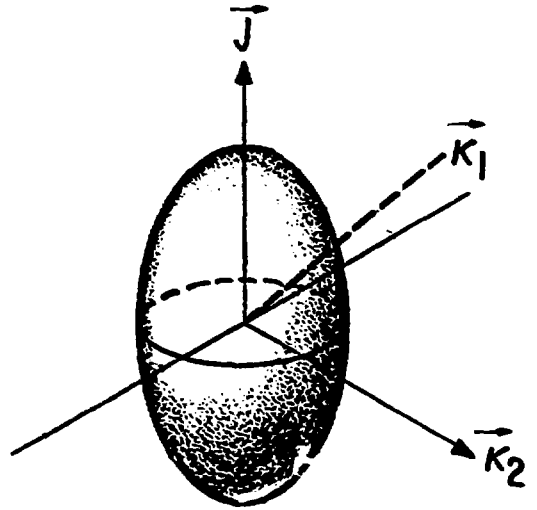


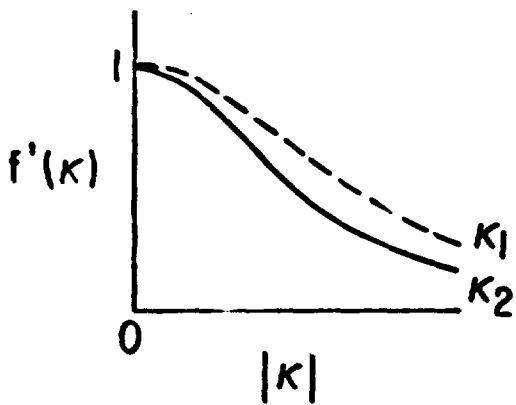
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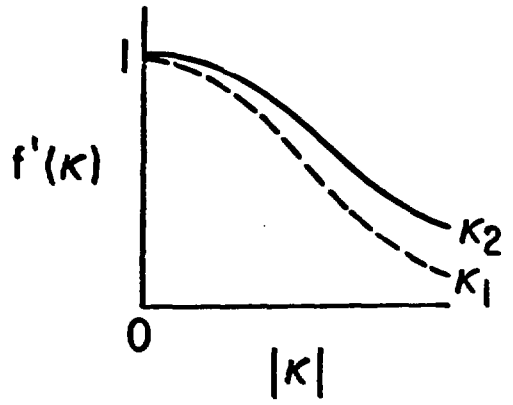
OBLATE



PROLATE



$$\Delta f > 0$$



$$\Delta f < 0$$

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