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THERMAL EQUILIBRIUM NUCLEAR ORIENTATION<sup>1, 2, 3</sup>

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THERMAL EQUILIBRIUM NUCLEAR ORIENTATION<sup>1, 2, 3</sup>

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March 12, 1966

THERMAL EQUILIBRIUM NUCLEAR ORIENTATION<sup>1,2,3</sup>

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## 1. INTRODUCTION

The purpose of this brief review is to describe in general terms the advances in technique in the area of low temperature nuclear orientation, for systems of nuclei in thermal equilibrium with the lattice in which they are embedded. Thus this article is complementary to C. D. Jeffries' review "Dynamic Orientation of Nuclei", in Volume 14 (1), dealing with cases in which nuclei were oriented dynamically by irradiation of the system (usually at microwave frequencies). The scope of this review is further specifically restricted to include only systems below 5°K, and for which nuclear orientation is detected by microscopic measurement of phenomena associated with

<sup>1</sup>The literature survey for this article was completed in March, 1966.

<sup>2</sup>Some abbreviations used herein: CMN [ $\text{Ce}_2\text{Mg}_3(\text{NO}_3)_{12} \cdot 24\text{H}_2\text{O}$ ]; EPR (electron paramagnetic resonance); NES [ $\text{Nd}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ ]; NMR (nuclear magnetic resonance).

<sup>3</sup>The preparation of this article was supported by the United States Atomic Energy Commission.

individual nuclei, i.e., by counting techniques. Thus low temperature heat capacity and NMR experiments are excluded.

The theory of the mechanisms and description of nuclear orientation is in a general sense a closed subject. Although new orientation mechanisms of great practical importance are still being developed, they do not involve in themselves any really new physics. In fact the theoretical description of the orientation process is essentially an application of angular momentum theory. A comprehensive discussion of this theoretical formulation has been given by Blin-Stoyle and Grace (2), with references to the original literature. A brief account is given below.

The externally-observable properties of an assembly of identical nuclei are isotropic if the hamiltonian for each nucleus is rotationally invariant or if the hamiltonians for the individual nuclei are anisotropic but rotationally uncorrelated. Application of an anisotropic interaction to the system in such a way that the principle axes of the individual hamiltonians are parallel will tend to orient the nuclei. A particularly familiar case is provided by NMR, in which nuclei are oriented along the direction of

the static external field  $\vec{H}$  by an interaction of the form  $\mathcal{H} = -\vec{\mu} \cdot \vec{H}$ . Here  $\vec{\mu}$  is the nuclear magnetic dipole moment.

Nuclear orientation along a symmetry axis may be described by the orientation parameters  $B_\nu$ , defined by  $B_\nu = \cdot(2I+1)^{1/2} \sum_M (-)^{I-M} C(II\nu; M-M) W(M)$ . Here  $C(II\nu; M-M)$  is a Clebsch-Gordan coefficient and  $W(M)$  is the population of the substate  $|IM\rangle$ , which may or may not be an eigenstate of the hamiltonian (if not, the eigenstates  $|i\rangle$  may be expanded in the basis set  $|M\rangle$ ,  $|i\rangle = a_{iM} |M\rangle$ , and  $W(M) = \sum_i |a_{iM}|^2 W(i)$ ). Note that the above definition is related by a Clebsch-Gordan identity to the one given by Blin-Stoyle and Grace. The statistical tensors  $B_\nu$  are just linear combinations of moments of the nuclear spin along the symmetry axis.

It is useful to distinguish between alignment and polarization. Aligned nuclei have a direction of orientation only; polarized nuclei have in addition a sense of orientation. The properties of aligned nuclei may be anisotropic, that is, may vary with even powers of the angle  $\theta$  from the symmetry axis; polarized nuclei may have asymmetric properties, varying also with odd powers of  $\theta$ . The value of  $B_0$  is 1 at all temperatures. For  $\nu > 0$  the high temperature limit of  $B_\nu$  is 0 for all  $\nu$ . At lower temperatures the

$B_v$  are sensitive to the form of the spin hamiltonian. The low-temperature limits also depend on  $\mathcal{H}$ . They may be finite (positive or negative) or zero. The maximum value of  $v$  is  $2I$ . The tensor rank of observables is often further restricted by angular momentum triangle conditions and by the symmetry of  $\mathcal{H}$ .

An outline of this article is given below for convenience.

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## 2. RECENT MAJOR ADVANCES IN TECHNIQUE

Nuclear orientation is widely recognized as a method that has made substantial contributions to our understanding of basic physics. At the same time it has rightfully been regarded as somewhat qualitative. Among nuclear physicists it also has the (now quite undeserved) reputation of being applicable to only a few elements. In this section the major technical advances of the last few years, application to new elements, accurate thermometry below  $0.1^{\circ}\text{K}$ , and high-resolution spectroscopy, are discussed in turn. First, however, a brief description of the experimental aspects of a nuclear orientation problem is given below.

### 2.1 The Experimental Arrangement

To orient the vast majority of nuclei very low temperatures, of the order of  $10^{-2}^{\circ}\text{K}$ , are necessary. Such temperatures can be obtained only by adiabatic demagnetization of paramagnetic salts. An axis of quantization and anisotropic hyperfine interaction are also required. In the apparatus shown in Figure 1, all three of these conditions are met by incorporating the radioactive nuclei to be oriented in lattice sites of a single crystal of NES. This

apparatus was constructed by R. B. Frankel to do the first conversion-electron anisotropy experiment, using  $Ce^{137m}$ . It has been used to study angular distributions of conversion electrons, alpha particles, and  $\gamma$  rays from several isotopes.

The outer liquid nitrogen dewar is omitted from Figure 1 for clarity.

The liquid helium dewar has a pyrex pipe flange at the top. This flange is sealed by an O-ring to one of the nine outlets of a vacuum manifold made of 8-inch pipe. The manifold is supported on concrete piers that go into the ground below the building and are only weakly coupled to the floor, to minimize vibrational heating of the demagnetized sample. A 1250 cubic ft/min Roots pump evacuates the manifold and lowers the vapor pressure of liquid helium in the dewar. Temperatures as low as  $0.96^{\circ}K$  are obtained even in large dewars. An iron-core electromagnet mounted on tracks is used to magnetize the salt. On demagnetization it is rolled away and a counter table, holding several  $\gamma$ -ray detectors, is brought up.

The experimental chamber is made of pyrex glass. It has a Housekeeper copper-to-glass seal at the top, and the whole chamber is attached by soft solder to a stainless steel tube that leads to a vacuum pump. Within twelve

hours after assembly a pressure of  $10^{-6}$  torr is obtainable in the cooled experimental chamber. Heat exchange is achieved during magnetization by admitting helium gas to the experimental chamber, at a pressure of  $10^{-2}$  torr. The pressure may be lowered to  $10^{-6}$  torr again in 15 minutes. With a clean and vibration-free system, the heat leak into the sample is sufficiently low to allow the sample to remain below the helium bath temperature for many hours, or even days.

The very poor thermal conductivity of the sample, together with inhomogeneous heating, allows temperature gradients to arise rather rapidly, so data can be taken for only a few minutes after demagnetization if one is interested in studying the temperature dependence of orientation. If angular distributions are to be correlated with the entropy of the salt, it is useful to take several counts and to extrapolate the data back to the moment of demagnetization. Mutual inductance coils around the experimental may be used to determine the susceptibility, and hence the magnetic temperature, of the salt.

Figure 2 shows more details of the apparatus. The sample is suspended on a 2-mm glass rod framework. It is protected from heat leaks down the rod by

a chromium potassium sulfate-glycerin slurry in a glass cup on the rod. This slurry cools to  $\sim 0.01^{\circ}\text{K}$  when the demagnetization is done, and it has a relatively large heat capacity at this temperature. A pill of compressed manganous ammonium sulfate is attached further up the rod. Its large surface area and low temperature (it cools to  $0.14^{\circ}\text{K}$ ) allow it to adsorb residual helium exchange gas.

Particle counters of silicon or germanium are placed inside the chamber, as shown in Figure 2, at  $0^{\circ}$  and  $90^{\circ}$  from the trigonal crystal axis. For experiments involving charged particles the activity was evaporated onto a small spot on the crystal surface. Data were usually taken at  $0^{\circ}$  and  $90^{\circ}$  from the quantization axis because angular distributions had the form

$$W(\theta) = 1 + C_2 P_2(\cos \theta) + C_4 P_4(\cos \theta).$$

Here the  $C_2$  and  $C_4$  factors are functionally dependent on hyperfine structure constants, temperature, and various nuclear parameters. They are discussed in Section 3.4. The factors  $P_2(\cos \theta)$  and  $P_4(\cos \theta)$  are Legendre polynomials. Both  $C_2$  and  $C_4$  may be determined by counting with counters at  $\theta = 0^{\circ}$  and  $90^{\circ}$  from the quantization axis, with the nuclei oriented, then counting once again, for normalization, after the sample has been warmed to  $1^{\circ}\text{K}$  by the introduction of exchange gas.

## 2.2. Application to New Elements

Until about 1960 only the hfs alignment and hfs polarization methods had seen wide application. These methods are applicable only to elements which form paramagnetic ions, and were further practically restricted to those elements that could be grown isomorphously into certain hydrated crystals that could also serve as magnetic cooling salts. Only a few elements met all of these requirements: Mn, Co, several rare earths, and Np. The hfs methods have been extended to include most paramagnetic ions by the realization that trivalent transplutonium ions ( ${}_{95}\text{Am}^{3+}$  through  ${}_{100}\text{Fm}^{3+}$ ) could be incorporated

into rare-earth sites in ethylsulfate crystals (3) and by the observation (4,5) that nuclei of "impurity" or solute paramagnetic ions could be oriented by the hfs polarization techniques. It should thus be possible to orient nuclei in the ions  $\text{Fe}^{3+}$ ,  $\text{V}^{2+}$ , and in complexes of  $\text{Ru(III)}$ ,  $\text{Mo(V)}$ ,  $\text{Mo(III)}$ ,  $\text{Re(IV)}$ ,  $\text{Ir(IV)}$ ,  $\text{Ag}^{2+}$ , and  $\text{Au}^{2+}$ , for example, simply by contact-cooling these complexes, either in a frozen solution (glass) or in "brine holes" in a single crystal of CMN, and applying a polarizing field of a few hundred gauss. While  $g\beta H \gg kT$  is the requirement for saturation in free ions ( $g$  = electronic  $g$  factor,  $\beta$  = Bohr magneton), one must also overcome the (random) ligand field effects in order to polarize ions in dissolved complexes.

Early attempts to exploit the electric quadrupole method (6) below  $1^\circ\text{K}$  were unsuccessful, owing to long (nuclear) spin-lattice relaxation times, or insufficiently low temperatures. In paramagnetic copper paraiodobenzene-sulfonate  $\text{I}^{131}$  nuclei were aligned down to  $0.03^\circ\text{K}$  (7), and quadrupole alignment has been performed on nuclei of La, Eu, Gd (also paramagnetic), Tm, Lu, Am, and  $\text{Ba}^{137\text{m}}$ , at temperatures down to  $0.002^\circ\text{K}$  in some cases (8-14). The discovery of large antishielding factors in heavy elements (8,9,15) suggests

that any heavy ion that can be grown into regular lattice sites (for example  $\text{Bi}^{3+}$  or  $\text{Ac}^{3+}$  in CMN) can be oriented by this method.

The advent of high-field superconducting magnets makes the "brute force" technique more attractive. Fields of  $10^5$  Oe and temperatures in the 10 mdeg range are available with present-day techniques. This combination is sufficient to polarize rather completely nuclei with very large magnetic moments. To align even these nuclei to saturation, however, larger values of  $H/T$  are required. It seems far more fruitful for most laboratories to attempt to decrease  $T$  than to increase  $H$ . One way in which this might be accomplished would be to employ as a coolant CMN, which becomes antiferromagnetic somewhat below about 2 mdeg (16). Starting from a sufficiently high magnetizing field, and with reversible cooling, it should be possible to extract most of the magnetic entropy from even a polycrystalline sample of CMN. A sufficiently large sample would stay below 2 mdeg for several hours, long enough to cool nuclear samples in good thermal contact. "Brute force" polarization has recently been used on  $\text{He}^3$  (17), and on  $\text{Li}^6$ ,  $\text{Li}^7$ ,  $\text{Be}^9$ ,  $\text{Be}^{10}$ ,  $\text{Sc}^{45}$ , and  $\text{V}^{51}$  (18,19,20). Hopefully further development of ways to increase  $H/T$  will lead to wider application of this method.

Nuclei have been oriented in the ferromagnets Co (21,22), Eu (23,24), Gd (23,25), Tb (26), Ho (27), and EuS (24), and in the antiferromagnets  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  (28),  $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$  (29),  $\text{MnBr}_2 \cdot 4\text{H}_2\text{O}$  (29), and  $\text{MnSiF}_6 \cdot 6\text{H}_2\text{O}$  (30), as well as in Tutton salts (31). This type of orientation has special merit for producing oriented targets, but its applicability is obviously limited.

The most universal method by far at this time is the orientation of nuclei of atoms dissolved in ferromagnets. This method stems from the discovery by B. N. Samoilov, et al. (32) that such solute nuclei experience very large induced hyperfine magnetic fields. Thus it is necessary only to cool a ferromagnetic specimen containing radioactive solute nuclei to  $\sim 0.01^\circ\text{K}$  and to polarize it in a field of  $\sim 10^3$  gauss. Nuclei of at least fifteen elements dissolved in iron have been oriented in this way, and the method is applicable to  $\sim 30$  more. The great importance of this method has led to attempts at systematic characterization of the induced hyperfine fields for solutes of various atomic numbers (33,34,35). In Table I we give a compilation of the known hyperfine fields at solutes in Fe, Co, and Ni, together with estimated fields for cases where the fields are as yet unmeasured. These fields are discussed further in Section 4.



Table I. Hyperfine fields at atoms in Fe, Co, and Ni lattices,  
in kilogauss.<sup>a, b, c</sup>

Host Solute	<u>Fe</u>	<u>Co</u>	<u>Ni</u>	<u>Reference</u>
Al	55	32		36
Sc	(+)58	(<50)	(<50)	37
(Ti)	(<50)	(<50)	(<50)	
V	-87	(-)48	(-)7	36, 38
Cr	<100	(-100)		39
Mn	-225.5(5) 270(15)	140 130(15)	-325 -316(10)	38, 40, 41 42
Fe	-339	-329	283	43, 44
Co	-286	-215	-120	43, 44, 45, 46
Ni	235	189	75	43
Cu	-213	(-)158	-47	43, 47
(Zn)	(-150)	(-100)	(-50)	
Ga	(-)110(3)	(-)62(1.5)	(<50)	35, 48
(Ge)	(+100)	(<+100)	(<+50)	
As	(+)339.1	(+200)	(+100)	37
(Se)	(+600)			
(Br)	(+1000)			
Y	+286(5)			35, 47
Zr	(-200)	(-)90(2)	(<50)	48
Nb	(-)262.4	(-100)	(-)39	36, 37
Mo	(-)256(5)	(-)150(3)	(-100)	35, 48
(Tc)	(-400)	(-250)	(-150)	
Ru	-505(20)	(-350)	-222(5)	33, 49

Table I. Continued.

Host Solute	<u>Fe</u>	<u>Co</u>	<u>Ni</u>	<u>Reference</u>
Rh	(-)545(8)	(-)410(10)	(-250)	35
Pd	(-)600(10)	(-)430(10)	(-300)	35, 48
Ag	-282(20)	(-200)	-87(8)	50
Cd	(-)348(10)	(-200)	-65.3(1.6)	51
In	-295(10) 288(4)	(-150)	-42(5)	52 48
Sn	-81(4)	-20.5(15)	+18.5(5)	53
Sb	+235	(+)187(2)	+88(5)	35, 37, 47, 48, 60
Te	+620(20)	(+)550(50)	+195(10)	34
I	(+)1130(40)	(+1000)		54
Nd	+3100(600)			55
Sm	+1400(160)			55
Eu	+640(170)			55
Gd	-200(50)			55
Dy	-2600(352)			
Er	-1640(190)			55
Yb	-720(240)			55
Ta	-630(10)		(-)90(15)	35, 60
W	760(80) 430(100)	(-500)	(-90)	56
Re	-526(12)	(-)442(10)	(-)88(5)	35, 48, 57, 60
Os	(-)1145(25) 1400	(-800)	(-)250(30)	35, 48 58
Ir <sup>d</sup>	-1400(40) 1380	(-)965(25)	(-350)	35, 48, 60 59
Pt	-1390	(-)830(20)	(-)337(5)	48, 58, 60

Table I. Continued.

Host	<u>Fe</u>	<u>Co</u>	<u>Ni</u>	<u>Reference</u>
Solute				
Au	-1290(30) 1350(50)	-900	-310	61,62 63
Hg	-980(180)	(-700)		64
(Tl)	(-600)			
Pb	-200(30)			65

<sup>a</sup> Errors limits are denoted parenthetically after values, in units of last figure.

<sup>b</sup> In some cases author has chosen best value or values. Fields given are lowest temperature values available.

<sup>c</sup> Parentheses around values themselves or signs denote estimates by author, based on systematics of known fields.

<sup>d</sup> Errors for Ir do not include the ~17% error in the ground-state magnetic moment of Ir<sup>193</sup>.

Table II. Nuclei oriented by thermal equilibrium methods.

<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>	<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>
He <sup>3</sup>	1	17	Cu <sup>65</sup>	1	132
Li <sup>6</sup>	1	18	Y <sup>90</sup>	2	47
Li <sup>7</sup>	1	19	Ru <sup>103</sup>	2	133
Be <sup>9</sup>	1	19	Ag <sup>104</sup>	2	134
B <sup>10</sup>	1	19	Ag <sup>110m</sup>	2	134
Sc <sup>45</sup>	1	20	Ag <sup>110</sup>	2	134
Sc <sup>46</sup>	2	67	In <sup>114m</sup>	2	52,135,136
V <sup>48</sup>	2	68,69	In <sup>114</sup>	2	52,137
V <sup>51</sup>	1	20	In <sup>115</sup>	1,2	165,175,202
Cr <sup>51</sup>	3	4,70	Sb <sup>121</sup>	2	60
Mn <sup>52</sup>	3	71,73-78	Sb <sup>122</sup>	2	47,135,136
Mn <sup>54</sup>	2,3,5	14,28-30,69,77,79-85	Sb <sup>123</sup>	2	60
Mn <sup>55</sup>	3	86,87	Sb <sup>125</sup>	2	138,139
Mn <sup>56</sup>	3	88-90	I <sup>131</sup>	4	7
Co <sup>55</sup>	3	90-92	Ba <sup>137m</sup>	4	14
Co <sup>56</sup>	3	93-97	La <sup>140</sup>	4	10
Co <sup>57</sup>	3	98-100	Ce <sup>137m</sup>	3	140-145
Co <sup>58</sup>	3	72,95,96,101-107	Ce <sup>137</sup>	3	140,141,145
Co <sup>59</sup>	3,5	14,108	Ce <sup>139</sup>	3	141,146-148
Co <sup>60</sup>	2,3,5	14,21,22,67,81,102,103,109-131	Ce <sup>141</sup>	3	100,141,146-152
Cu <sup>63</sup>	1	132	Ce <sup>143</sup>	3	141,153
Cu <sup>64</sup>	2	47	Pr <sup>139</sup>	3	154
			Pr <sup>142</sup>	3	155,156

Table II. Continued

<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>	<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>
Pr <sup>143</sup>	3	90	Tb <sup>160</sup>	3	169-173
Nd <sup>143</sup>	1	60	Dy <sup>155</sup>	3	174
Nd <sup>145</sup>	1	60	Dy <sup>157</sup>	3	174
Nd <sup>147</sup>	3	147,149,157,158	Ho <sup>162</sup>	3	154
Pm <sup>143</sup>	3	159,160	Ho <sup>165</sup>	3,5	27,175-182
Pm <sup>144</sup>	3	143,160,161	Ho <sup>166m</sup>	3	171,172,183,184
Pm <sup>148m</sup>	3	160	Tm <sup>168</sup>	4	185
Pm <sup>148</sup>	3	160	Tm <sup>170</sup>	4	99
Pm <sup>149</sup>	3	162	Yb <sup>175</sup>	3	186
Pm <sup>151</sup>	3	162	Lu <sup>172</sup>	4	185
Sm <sup>145</sup>	3	163	Lu <sup>177m</sup>	4	10
Sm <sup>149</sup>	3	164,166	Lu <sup>177</sup>	4	9,10
Sm <sup>153</sup>	3	163	Ta <sup>181</sup>	1,2	60
Eu <sup>151</sup>	5	23,24	W <sup>187</sup>	2	133
Eu <sup>152</sup>	4	8,10	Re <sup>185</sup>	2	60
Eu <sup>153</sup>	5	23,24	Re <sup>186</sup>	2	137,187-189
Eu <sup>154</sup>	4	8,9,10	Re <sup>187</sup>	2	60
Gd <sup>155</sup>	5	23,25	Ir <sup>191</sup>	2	60
Gd <sup>157</sup>	5	23,25	Ir <sup>191m</sup>	2	190
Gd <sup>159</sup>	4	9,10	Ir <sup>192</sup>	2	137,187,191,192
Tb <sup>156</sup>	3	167	Ir <sup>193</sup>	2	60
Tb <sup>158</sup>	3	168	Pt <sup>195</sup>	2	60
Tb <sup>159</sup>	5	26	Au <sup>195</sup>	2	63



Table II. Continued.

<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>	<u>Nucleus</u>	<u>Method<sup>a</sup></u>	<u>Reference</u>
Au <sup>198</sup>	2	32,63, 66, 135, 136, 193, 194	Am <sup>241</sup>	4	203
Au <sup>199</sup>	2	63, 187	Cf <sup>249</sup>	3, 4	3
U <sup>233</sup>	4	195-197	Es <sup>253</sup>	3	3, 145
U <sup>235</sup>	4	196-198	Es <sup>254</sup>	3	204
Np <sup>237</sup>	3, 4	196, 197, 199, 200	Fm <sup>255</sup>	3	205
Np <sup>239</sup>	3, 4	201			

<sup>a</sup>Methods.

1. Brute Force
2. Universal
3. Hfs alignment or polarization
4. Electric quadrupole
5. Ferromagnet or antiferromagnet

fact the situation was even worse, because the published temperature scales for CMN and NES contained systematic errors (Section 4).

The above problems may be avoided by using the assembly of oriented nuclei as a thermometer. If anisotropic distributions of  $\gamma$  rays are detected, this technique is termed "gamma-ray thermometry". Its basis is: (1) If the form of the nuclear spin hamiltonian  $\mathcal{H}_N$  and the numerical values of its constants are known, then the eigenstates  $\psi_i = \sum_M C_{iM} |M\rangle$  and eigenvalues  $E_i$  can be worked out exactly. Here  $M$  is the magnetic quantum number of the nuclear spin  $I$  ( $M=I, I-1, \dots, -I$ ). The orientation axis is usually most conveniently taken as the axis of quantization. (2) For a given  $\mathcal{H}_N$ , each  $B_k$  is a single-valued function of the thermodynamic temperature  $T$  of the nuclear spin system. The converse is not necessarily true (although if  $\mathcal{H}_N$  is known this is irrelevant). For several simple forms of  $\mathcal{H}_N$  the converse is in fact true (i.e., each value of  $B_k$  implies a unique  $T$ ). (3) Measurement of the  $\gamma$ -ray angular distribution yields the parameters  $B_k$ , thus  $T$ . To avoid ambiguity in cases for which  $T$  may be a multi-valued function of a given  $B_k$ , the results may be ordered according to the magnetic entropy of the cooling salt, which

is monotonically related to T. As a check of the procedure we note that each  $B_k$  yields T separately.

Wheatley, Griffing, <sup>and Hill</sup> made an early application of  $\gamma$ -ray thermometry in 1955 (103). It was applied to the "universal" method by the Oxford group (66), where it has found extensive application. Recently it has been used at Berkeley to redetermine the temperature scales of CMN (143) and NES (144).

A note of caution is in order here. In using any kind of thermometry it is essential that appreciable thermal gradients should not be allowed to develop in the sample. Gradients lead, in this case, to a distribution of values of  $B_k$ , and the observed average  $\bar{B}_k$  is weighted in an unknown manner. Worse, the observed combination  $\bar{B}_2$  and  $\bar{B}_4$  (for example) need not be a combination allowed by  $\mathcal{H}$  for a system at thermal equilibrium. The derived temperature would be not only incorrect, but meaningless. Efforts to compensate for inhomogeneities by comparing two isotopes, one with known, the other with unknown, hfs constants, implicitly without actually <sup>maintaining a homogeneous</sup> temperature are doomed, because implicit in such experiments is the assumption that the two isotopes <sup>exactly</sup> have the same hfs constants. This procedure has led to erroneous values for magnetic moments in several cases.



## 2.4 High Resolution Spectroscopy

Inapplicability of high-resolution methods has long been a major impediment to the development of nuclear orientation as a tool for nuclear spectroscopy. This barrier has recently been removed with the advent of high resolution semiconductor detectors. Lithium-drifted germanium detectors have been applied at Berkeley to orientation experiments involving the isotopes  $Ce^{137m}$ ,  $Ho^{166m}$ ,  $Tb^{160}$ ,  $Sb^{125}$ ,  $In^{114m}$ ,  $Nd^{147}$ ,  $Lu^{177m}$ , and  $Co^{57}$ , with resolution ranging from 2 to 6 keV. Long counting times, of the order of 1 hour, are necessary to compile good statistical accuracy with Ge(Li) detectors because of their low efficiency, but the high inherent gain stability of these detectors and associated transistorized circuitry makes such long counting periods feasible. In fact it has been possible to study the anisotropies of several  $\gamma$  rays that had gone undiscovered using NaI(Tl) counters. As an illustration of the power of Ge(Li) detectors in nuclear orientation, Figure 3 shows a  $\gamma$ -ray spectrum, at intermediate resolution, of the 700-850 keV region in the decay of  $Ho^{166m}$ , contrasted with the NaI(Tl) spectrum. Angular distributions of 5  $\gamma$  rays from  $Ho^{166m}$  aligned in NES, together with their spin designations, are also shown (206).

Electron spectroscopy has also benefitted from semiconductor devices.

Walter et al. (207) reported in 1961 that the intrinsic germanium detectors that were used for alpha counting at low temperatures were also sensitive to beta particles. Navarro (208) found that conversion-electron groups could be resolved on similar detectors, and Frankel et al. first observed anisotropic angular distributions of conversion electrons from oriented nuclei using such detectors (142). Chin et al. (209) showed that intrinsic silicon counters could give resolutions as high as 8.3 keV for conversion electrons at low temperatures.

At this time it seems that with careful consideration of the sources of line broadening it should be possible to observe conversion electron lines at low temperatures with resolution comparable to that available with Ge(Li)  $\gamma$ -ray detectors, i.e., about 1 keV. Good separation of the M conversion electron group, and even resolution of the  $L_{III}$  subshell, should be possible in the heavier elements. Quantitative study of the energy-dependence of beta anisotropy and asymmetry is also within reach.

## 2.5. Summary

The conclusions of this section may be summarized in three parts:

1. Nuclear orientation is a technique of very wide applicability.

Nuclei of 43 elements have already been oriented, and it is clear that this list can be extended much further with existing techniques.

2. The development of semiconductor detectors has brought the advantages of high-resolution spectroscopy to nuclear orientation, making it possible to study individual transitions independently.

3. The development of reliable thermometry below  $0.1^{\circ}\text{K}$  makes nuclear orientation a more attractive method for determining hfs constants. With careful attention to sources of systematic error it is possible to attain 1% accuracy, rather than the earlier ~10%, in favorable cases. Statistical uncertainty alone can be made as small as 0.1% with some effort, but it is not usually feasible to reduce systematic errors (background, sample inhomogeneity, etc.) below 1%. The questions of accuracy and reliability are encountered again in Sections 3.7 and 4.1. Gamma-ray thermometry should also prove valuable in other investigations at very low temperatures (see Section 4.5).

### 3. NUCLEAR PHENOMENA

In this section and in Section 4 are discussed the general results that have emerged from recent nuclear orientation research. No attempt is made to catalog detailed numerical results. Rather we discuss, with illustration by example, the impact of nuclear orientation on nuclear physics.

#### 3.1 New Fundamental Experiments

In this section are discussed experiments that are qualitatively new.

These fall into three groups.

A. Conversion Electron Distributions. Shirley and Stone, Frankel, et al. (142) found that conversion electrons accompanying the 255-keV  $11/2^-(M4)3/2^+$  isomeric transition in  $Ce^{137}$  show a large spatial anisotropy when the parent  $Ce^{137m}$  nuclei are oriented in NES. The observed angular distributions of conversion electrons and  $\gamma$  rays accompanying an isomeric transition in oriented nuclei may be written

$$W_e(\theta) = 1 + g_2 b_2 B_2 F_2 P_2(\cos \theta) + g_4 b_4 B_4 F_4 P_4(\cos \theta), + \dots$$

$$W(\theta) = 1 + g_2' B_2 F_2 P_2(\cos \theta) + g_4' B_4 F_4 P_4(\cos \theta) + \dots$$

Here the  $g_v, g_v'$  are (known) solid angle factors for the detectors and  $b_v$  are "particle parameters" that relate the electron and  $\gamma$  ray distributions.

The orientation parameters  $B_\nu$  are defined above, in the Introduction. The angular correlation coefficients  $F_\nu$  are discussed in more detail in Section 3.4.

By combining data for  $\theta = 0$  and  $\pi/2$ , these equations will yield  $b_2$  and  $b_4$  for each resolved electron group. In the  $Ce^{137m}$  experiment,  $b_4$  was small and  $b_2$ , but not  $b_4$ , was derived from the data. The value for the K electron line was  $b_2(K) = +1.061 \pm 0.018$ , in excellent agreement with the theoretically predicted value  $+1.055$ . The result for the L+M group,  $b_2(L+M) = +1.059 \pm 0.020$ , could not be compared with theory, but since the experiment a theoretical value of  $b_2(L) = +1.059$  has been calculated by B. Deutch (203).

The parity sensitivity of the  $b_2$  parameters, as well as their independence of the initial- and final-state spins, make their measurement a powerful tool in nuclear spectroscopy. From a complete knowledge of the angular distributions of conversion electrons and  $\gamma$  rays from oriented nuclei it is possible to determine all the spins and multipolarities, including parities, in the daughter nucleus.

B. Anisotropy of Reactions with Deformed Nuclei. Ambler, / Fuller, and Marshak (181)

used bremsstrahlung to study the  $\gamma, n$  reaction on  $Ho^{165}$  nuclei oriented in holmium ethylsulfate. The experiment was designed to test the assumption that the structure in the "giant dipole resonance" arises from prolate nuclear deformation, with dipole excitation along the major axis accounting for the

lower energy component, and (twofold degenerate) excitation along the minor axes giving the higher-energy component. The experimental results are consistent with this model, yielding an interaction that is ~85% "deformed" and ~15% scalar.

Richardson and Tamara

Marshak, / (179) found an anisotropy in the total cross-section

for 14-MeV neutrons on aligned  $\text{Ho}^{165}$  nuclei. Although the analysis of the experiment is complicated, its essence is simple. In Fig. 4 are shown the two extreme orientations of prolately-deformed ellipsoids with respect to a

beam direction. In the classical limit, and when the approximation that the nucleus does not rotate during the time that it interacts with the neutron is valid, the experiment consists essentially of determining the relative

areas of the shadows cast by the nucleus in these two positions. This area ratio has the limiting value of  $b/a$ , the axial ratio, which is

related to the nuclear deformation,  $\delta$ . In the experiment of Marshak et al.

the anisotropy is consistent with a deformation  $\delta = .3$ .

C. Upper Limit for Spin-Spin Forces. Miller, Tamara, and Marshak  
Wagner, / (178) studied the

transmission of 350-keV polarized neutrons through polarized  $\text{Ho}^{165}$  nuclei.

They concluded that if there is a term in the optical-model potential of the

form  $-V_{ss}(\vec{\sigma} \cdot \vec{I})$  then it must be of magnitude  $-130 < V_{ss} < +280$  keV, an order

of magnitude smaller than spin-orbit forces.

### 3.2 Alpha Decay

Study of alpha decay from oriented nuclei allows the determination of the amplitudes of the various L waves in a transition. For a favored transition, in which the initial and final states are the "same" Nilsson state, only even L waves (S, D, G, ... corresponding to  $L = 0, 2, 4, \dots$ ) are allowed. Their intensities for the highest-energy ( $\Delta I=0$ ) transition in a rotational band go approximately as S:D:G  $\sim 1:0.1:0.01$ . The S wave itself is of course isotropic, and the D wave has a low intensity, but the S, D interference term leads to a very large anisotropy in the angular distribution of alpha particles. The physical meaning of these two partial waves being "in" or "out of" phase is that the angular distribution will go as  $1 + |A_2|P_2(\cos \theta)$  or as  $1 - |A_2|P_2(\cos \theta)$ , i.e., that alphas will preferentially be emitted from the poles or from the equator of prolately deformed nuclei. Hill and Wheeler (210) first pointed out in 1953 that preferential polar emission is expected because of easier tunnelling through the lower Coulomb barrier (Fig. 5). The relative phase of the D and G waves reflects more details of the nuclear shape.

The Oak Ridge group observed alpha anisotropies from oriented nuclei, in  $U^{233}$ ,  $U^{235}$ , and  $Np^{237}$ . For  $Np^{237}$  they showed explicitly that  $\alpha$  particles.



were emitted preferentially in the plane in which the nuclei were oriented, hence from the tips of prolately-deformed  $\text{Np}^{237}$  nuclei (200).

More recently, taking advantage of the fact that nuclei of trivalent transuranium elements could be oriented in rare-earth ethylsulfate crystals, alpha particle anisotropies have been studied in Berkeley from oriented  ${}_{95}\text{Am}^{241}$  (203),  ${}_{98}\text{Cf}^{249}$  (3),  ${}_{99}\text{Es}^{253}$  (3,<sup>145</sup>), and  ${}_{100}\text{Fm}^{255}$  (205) nuclei. The electronic properties of the lowest crystal-field states of these ions have not yet been studied by other experimental methods, but may be assumed to be the same as those of the lowest states in the corresponding rare-earth ions. This assumption was borne out by the observed temperature dependences of the orientation parameters for the four ions, as discussed in Section 4.4. In each case the S and D waves were found to be in phase in the alpha decays of these nuclei. In addition Frankel (145) has found a large  $P_4(\cos \theta)$  term in the  $\text{Es}^{253}$  decay. The anisotropy found by Frankel is shown in Fig. 6.

### 3.3 Beta Decay

Parity nonconservation has now been observed in the beta decay of perhaps ten elements, but very little low temperature nuclear orientation work has been reported that is

quantitative enough to provide definitive values of beta-decay matrix elements. Groups at the Bureau of Standards (151), in Czechoslovakia (188,189) and in Russia (194) have made some progress in this area. Especially encouraging is the development of semiconductor detectors. Determination of matrix elements in first-forbidden beta decay is especially difficult because of their multiplicity and because of the insensitivity of measured quantities to the matrix elements. When serious efforts are made to study these matrix elements, nuclear orientation <sup>should</sup> prove one of the valuable complementary techniques for doing so.

### 3.4 Spin and Electromagnetic Multipolarity Determinations

Nuclear orientation is unique among the tools available for studying nuclear structure in radioactive decay in that it allows the direct measurement, for each  $\gamma$  ray, of a quantity proportional to a single angular correlation coefficient,  $F_{\nu}$ , including the sign. The proportionality constant can also be directly determined or calculated in most cases. All of this is done by singles counting, for a few minutes, on an assembly or oriented nuclei. Most (or all) of the interpretation is based firmly on the conservation of angular momentum.

Let us consider a cascade of transitions from an oriented nuclear state 1, proceeding through states 2, 3, 4, etc. We may devote the (possibly mixed) multipolarity of the radiation connecting states 1 and 2 as  $L_1, L_1', L_1''$ .

In a typical experiment one may measure for each  $\gamma$  ray of multiplicities  $L_n, L_n'$  connecting initial and final states of spins  $I_n$  and  $I_{n+1}$  the quantities  $C_2$  and  $C_4$  in the angular distribution function

$$W(\theta) = 1 + C_2 P_2(\cos \theta) + C_4 P_4(\cos \theta)$$

Each  $C_\nu$  has the form  $B_\nu U_\nu F_\nu$ . If the orientation is nearly complete, the values of  $B_\nu$  are known and may be divided out to yield  $U_\nu F_\nu$ . The  $U_\nu$  functions, accounting for reorientation during preceding, unobserved, transitions, just correct the  $B_\nu$  from the values reached in the parent state being oriented to the  $B_\nu$  actually experienced by the state emitting the observed radiation (2). If the spins and multiplicities of preceding transitions are known, the  $U_\nu$  can be calculated exactly. For a preceding transition of a single multipolarity  $L_k$ , we have

$$U_\nu^k(I_k, I_{k+1}, L_k) = (2I_k + 1)^{1/2} (2I_{k+1} + 1)^{1/2} (-1)^{I_k + I_{k+1} - L_k} W(I_k, I_k, I_{k+1}, I_{k+1}; \nu L_k),$$

where  $W$  is a Racah coefficient. If the transition is of mixed multiplicities

$L_k, L_k', L_k'', \dots$  of fractional intensities  $f_k, f_k', f_k'', \dots$ , the above expression becomes

$$U_{\nu}^k = f_k U_{\nu}^k(L_k) + f_k' U_{\nu}^k(L_k') + f_k'' U_{\nu}^k(L_k'') + \dots$$

The overall  $U_{\nu}$  for a series of unobserved transitions in cascade is simply the product of the individual  $U_{\nu}^i$ 's. For beta decay  $L$  is the tensor rank of the interaction. Thus for a first-forbidden  $\Delta I = 0$  transition, with  $I \geq 1$ ,  $L$  has the possible values 0, 1, and 2. For transitions involving high spin and low multipolarities, the  $U_{\nu}$ 's are close to 1, and it is often possible to estimate the  $U_{\nu}$ 's quite accurately with very little knowledge about preceding transitions (160). Usually there is enough information, then, to evaluate the products  $B_{\nu} U_{\nu}$ , and thus the angular correlation coefficients  $F_{\nu}$ , to within a few percent.

These  $F_{\nu}$  depend explicitly on the spins and multipolarities in the observed transition. They are defined by

$$F_{\nu}^n(I_{n+1} I_n L) = (-1)^{I_{n+1} - I_n - 1} (2I_n + 1)^{1/2} (2L + 1)^{1/2} C(LL_{\nu}; 1-1) \\ \times W(I_n I_n LL, \nu I_{n+1}) ,$$

where  $C$  is a Clebsch-Gordan coefficient. If the observed transition has mixed multipolarities  $L_n$  and  $L_n'$ , of relative intensities  $1:\delta^2$ , then  $F_{\nu}^n$  becomes

$$F_{\nu}^n(I_{n+1} I_n)' = [F_{\nu}^n(I_{n+1} I_n L) + \delta^2 F_{\nu}^n(I_{n+1} I_n L') \\ + 2\delta F_{\nu}^n(I_{n+1} I_n LL')] / [1 + \delta^2] .$$

Here  $F_{\nu}^n(I_{n+1} I_n LL')$  is the angular correlation coefficient for mixed multipolarities. It is usually tabulated together with  $F_{\nu}^n(I_{n+1} I_n L)$ . In most cases these coefficients constitute enough data to determine spin and multipolarities uniquely. If not, or if a check is desired on the assignments, angular correlation measurements can serve a complementary

function. It should be noted that there are two values of  $F_\nu$  (for each  $\nu$ ) associated with a  $\gamma$ -ray transition, one for the transition "first" and one for it "last" in a cascade (158). For a mixed transition of (two) multipolarities there are two values, differing only in sign, for the amplitude mixing ratio  $\delta$ . This was first pointed out for an actual case by M. A. Grace (211).

The value of nuclear orientation in determining spins and multipolarity mixing ratios is best illustrated by citing some cases in which it has been used for this purpose:  $\text{Pm}^{147}$  (3 spins, 6 ratios),  $\text{Sm}^{148}$  (4 spins),  $\text{Fe}^{55}$  (3 spins, 1 ratio)  $\text{Cd}^{110}$  (4 spins, 2 ratios),  $\text{Te}^{125}$  (5 spins, 2 ratios). With the increasing emphasis in nuclear structure studies on spin and multipolarity determinations, nuclear orientation can be a very valuable tool in the nuclear spectroscopy laboratory.

### 3.5 Particle Parameters and Relative Parities

Both particle parameters for conversion electrons and conversion coefficients themselves are parity-sensitive. With the developments discussed in Section 2 it is now feasible to detect all the electron and  $\gamma$  radiation associated with a decaying isotope using one thin-window Ge(Li) detector. If

the source is an assembly of oriented nuclei, one can determine, using two detectors and only singles counting, the complete  $\gamma$  and electron spectrum, the coefficients  $F_\gamma$  for each  $\gamma$  ray and conversion electron group, and all the conversion coefficients. If a large degree of orientation is achieved, these data are sufficient to determine the spins of all populated levels and multipolarities of all transitions in the daughter nucleus. Only one conversion-electron anisotropy measurement has been reported to date, but it seems clear that the advantages of measuring conversion-electron parameters will lead to an expansion of this technique.

### 3.6 Neutron Transmission Experiments

Transmission of polarized neutrons through polarized targets provides a very clean method for determining the spins of neutron resonances. In s-wave capture with nuclei of spin  $I$ , only states of spins  $I \pm \frac{1}{2}$  are excited. The sign and magnitude of the change in transmittance at a resonance allows a decision between these two possibilities. Sailor et al. and Stolovy have exploited and developed this method to determine the spins of many resonances. A particularly impressive measurement by Alfimenkov et al. (182) at Dubna should also

be mentioned. These workers used a dynamically-polarized proton target as their neutron polarizer and reported spins for eleven resonances up to 54 eV in polarized holmium nuclei.

The use of hyperfine fields in ferromagnets to polarize nuclei has quite naturally led the Sailor and Stolovy groups into the study of these fields, and they have been able to make significant contributions in this area, partly because their method yields the sign of the hyperfine field directly.

In the future we may expect this branch of nuclear orientation to employ Ge(Li) detectors to study the angular distribution of  $\gamma$  rays emitted in the decay of states produced in neutron capture. Such studies would yield directly the spins of states thus populated, and the multipolarities of connecting transitions (212).

### 3.7 Nuclear Moment Determinations

Table III gives the nuclear magnetic moments that have been determined by nuclear orientation to date. Those that the author believes to be erroneous because of systematic errors are excluded. Values that are in some doubt because of uncertainties in temperature scales are marked with an asterisk.



Preliminary estimates indicate that the corrections to these values will not be drastic. The values of moments determined by other techniques are included for comparison.

#### 4. SOLID-STATE PHENOMENA

The power of nuclear orientation in studying the solid state has been fully realized in recent years. Among the qualitative discoveries of this method are the existence of hyperfine fields in metals (21, 22), the induced hyperfine fields at solute nuclei in ferromagnets (32), and the large anti-shielding factors in rare earth ions (8,9). So substantial has been the progress since 1960 that we find it convenient to review recent activity under five distinct headings.

##### 4.1 Hyperfine Magnetic Fields

Nuclear orientation makes its most valuable contributions in areas where it is uniquely applicable, in this case to systems that are extremely dilute in the species whose field is to be studied. Two types of system that have received considerable attention lately are solutions of nominally magnetic atoms (Mn, Co) in "nonmagnetic" metals and solutions of nominally "nonmagnetic" atoms in ferromagnets. The first type has been studied intensively by the Oxford group, who find evidence for local moments for solutions of Co in Pd, with abrupt changes in the effective field on addition of small amounts of stable Co (213).

The subject of induced hyperfine fields at nuclei of solutes in ferromagnets is taking shape, at least insofar as the low-temperature limits are concerned. Before discussing this subject, it is interesting to note that the accuracy obtained in determining hyperfine fields by nuclear orientation has been confirmed in several cases by other measurements based on spectroscopic techniques. Of the five cases in Table I for which such comparison was possible, Mn in Fe, Mn in Co, Mn in Ni, Sb in Fe, and Au in Fe, agreement was excellent in four. The quoted accuracy for the orientation results was 3-4% in three of these cases.

Induced hyperfine fields in iron, cobalt, and nickel were given in Table I. They are plotted in a way that emphasizes trends in Fig. 7. It is useful in planning orientation experiments to be cognizant of the observed trends, as they can help in selecting an optimum host. We note first that the fields tend to be negative, especially in the upper half of the d shells. They tend to be large for solute atoms of large atomic number. This led Shirley and Westenbarger to suggest (33) that conduction electron polarization is one important mechanism contributing to the fields. The peaking of these fields in the d shells suggests in addition core polarization for these

In Figure 8 the phenomenon of core-polarization is illustrated. cases (33). / Later measurements, especially on Rh and Pd, have borne this

out very nicely, and we may with considerable confidence expect that these latter two solutes have negative hyperfine fields in Fe, Co, and Ni.

Beyond the d shells the fields tend to swing through zero and become positive. This result is best established for the 4d-5p series in Fe. The origin of the positive sign is not established. It could arise from core polarization by spins of the p electrons or it might arise from a change from antiferromagnetic to ferromagnetic effective coupling of the 3d spins of the host and the outer s electrons of the solute, perhaps as a result of increased ionic size. Because of this sign change in the induced fields in the p shell, it is not possible to predict with any confidence the magnitude or even the signs to be expected for the alkalis and alkaline earths.

Finally spin-exchange polarization should lead to large hyperfine fields in rare-earth solutes in Fe, Co, and Ni, by orienting the angular momentum  $\vec{J}$  of the rare earth ion core, which should be a good quantum number since the 4f electrons are shielded from the crystal field. The fields should have opposite signs in the light and heavy rare earths (33), because they arise mostly from the orbital angular momentum  $\vec{L}$ , and  $J = L + S$  in the light (heavy) earths (Eu and Gd are exceptions). Boehm, Hagemann, and

Winther have confirmed this supposition (55), though they find that the effective relative polarization of 4f and 3d spins is antiparallel rather than parallel.

They also find that the magnitudes of the fields are substantially below the free ions values. Probably crystal-field effects can account for this result.

#### 4.2. Quadrupole Shielding and Antishielding

From alignment experiments in NES Judd et al. (8) discovered large negative antishielding factors in rare-earth ions. Theoretical values subsequently appeared that have now been combined with measured quadrupole coupling constants (11) to derive the parameters  $(1 - \sigma_2)$  that describe the shielding of the quadrupole crystal-field component  $V_2^0 = (1 - \sigma_2) A_2^0 \langle r^2 \rangle_{4f}$ . Here  $A_2^0$  is the angular component of the crystal-field electrostatic potential arising from charges external to the rare-earth ion that transforms as the spherical harmonic  $Y_2^0(\theta, \phi)$ . The rare-earth 4f electrons, which are responsible for the optical properties of the salt, are shielded by the outer electrons of the rare earth ion, reducing  $A_2^0$  by the factor  $(1 - \sigma_2)$ . The expectation value of the second radial moment of a single rare-earth 4f electron is denoted  $\langle r^2 \rangle_{4f}$ . It has been found that  $1 - \sigma_2 \cong 0.5$  for most of the rare-earths, but that this factor varies

systematically in the series. This result is expected for a term of relatively high symmetry, because the excited configurations of the outer electrons can couple to form levels that have nonzero matrix elements of  $A_2^0$  in many ways. For  $n > 2$ ,  $(1 - \sigma_n)$  is close to unity.

#### 4.3 Form of the Spin Hamiltonian

The temperature dependence of nuclear orientation, as well as its response to external and dipolar fields, can elucidate the form of the spin hamiltonian applicable to the lowest electronic crystal-field level. Johnson et al. (170) and later Lovejoy and Shirley (167) were able to detect both dipolar interaction and quadrupole coupling terms for  $Tb^{3+}$  in NES in this way. Chapman et al. (162)

discovered "pseudoquadrupole" coupling for  $\text{Pm}^{3+}$  in CMN from the temperature dependence of the  $\gamma$  ray anisotropy. et al. (175) Sailor/ found both magnetic and quadrupole hfs in Ho metal.

Alpha-particle anisotropies from  $\text{Am}^{3+}$ ,  $\text{Cf}^{3+}$ ,  $\text{Es}^{3+}$ , and  $\text{Fm}^{3+}$  nuclei oriented in NES have helped to establish, through their temperature dependences, that the lowest crystal-field states of these ions are like those of the corresponding rare-earth ions in NES. For  $\text{Am}^{3+}(5f^6 \ ^7F_0)$  the lowest crystal-field state is a singlet, and the antishielding crystal-field gradient leads to orientation along the trigonal axis, as in  $\text{Eu}^{3+}$ . The  $\text{Cf}^{3+}$  ion is analogous to  $\text{Dy}^{3+}$ , and an  $\text{AS}_z \text{I}_z$  interaction is probably mainly responsible for orientation, although quadrupole coupling also enhances it. Like  $\text{Ho}^{3+}(4f^{10})$ ,  $\text{Es}^{3+}(5f^{10})$  has a very large  $\text{AS}_z \text{I}_z$  interaction. In  $\text{Fm}^{3+}(5f^{11})$  as in  $\text{Er}^{3+}(4f^{11})$  the effective hfs hamiltonian has the form  $\text{AS}_z \text{I}_z + \text{B}(\text{S}_x \text{I}_x + \text{S}_y \text{I}_y)$ , with  $\text{B} > \text{A}$ .

#### 4.4 Time-Dependent Phenomena

For most orientation experiments in which  $\gamma$ -ray anisotropies are observed, the assumption/made that no reorientation occurs prior to emission of the observed  $\gamma$  ray save that due to angular momentum coupling (i.e., the redistribution

among magnetic substates of the "final" state in a preceding, unobserved transition that is implied by vector coupling of the spin of the initial state and the multipolarity of the transition). The / <sup>effect of this coupling</sup> may be calculated exactly, appearing finally as a "U<sub>v</sub>" coefficient. Of course, the above assumption is valid only when the lifetimes of those states that follow the original (oriented) parent but precede the observed transition are very short. If a state of lifetime  $\sim 10^{-10}$  sec occurs, the orientation may be perturbed in this state, thereby changing the angular distribution of the observed  $\gamma$  ray.

Reorientation effects may be subdivided in several ways. We shall distinguish between two essentially distinct phenomena. The first is reorientation that arises even in very short-lived states because of the inability of a daughter atom to achieve its equilibrium electron configuration rapidly enough following beta decay. Large time-dependent fields can cause substantial attenuation/in such cases. This effect should be large in dielectrics and absent in ferromagnetic metals, where exchange polarization keeps the hamiltonian diagonal and conduction electrons provide an efficient reducing atmosphere. R. C. Sapp/<sup>(100)</sup> has found attenuation in the 136.4-keV 4 nsec state of Fe<sup>57</sup> in dielectrics, following decay of oriented Co<sup>57</sup>. N. J. Stone finds



large attenuation in salts, but none in iron metal (153). Blok, Kaplan, and Shirley have found considerable attenuation for the 61-keV, 3 nsec state of  $\text{Pm}^{145}$  in CMN (163), and Barrett and Shirley (214) find substantial (~60%) but not complete attenuation of the 2.4 nsec, 91-keV state in  $\text{Pm}^{147}$  in CMN. Ambler, Hudson, and Temmer (147,149) reported earlier large attenuation in this state.

The second type of reorientation is simply spin-lattice relaxation, which is very long for many situations at very low temperatures. Here the experimental picture is even emptier. For  $\text{Ir}^{191m}$  in iron Cameron et al. (190) found an upper limit of ~1 sec for reorientation below  $0.1^\circ\text{K}$ , having observed complete reorientation in the 4.9 sec isomer. Blok and Shirley (10) set an upper limit of ~10 sec on the relaxation time of the ~1 sec isomer of  $\text{Hf}^{177}$  in NES. Lubbers et al. (14) have reported a slow approach to equilibrium in orienting  $\text{Co}^{60}$  in a Tutton salt. It would be interesting to make dynamic studies of faster relaxation processes.

#### 4.5 Temperature-Scale Determination

This topic was discussed in a general way in Section 2.2. Specific applications have been made to re-work the temperature scales of the two salts

Table III. Magnetic moments determined by  
equilibrium nuclear orientation.

Nucleus	Moment, mm <sup>a,b</sup>	Reference <sup>c</sup>	Nucleus	Moment, mm <sup>a,b</sup>	Reference <sup>c</sup>
V <sup>48</sup>	1.63(10)	68	Pm <sup>148</sup>	1.82(19) <sup>c,d</sup> 2.07(21)	160 215
Co <sup>55</sup>	4.3(3)	92	Pm <sup>149</sup>	3.3(5) <sup>c,d</sup>	160,162
Co <sup>56</sup>	2.8(9) 3.803(7) <sup>b</sup>	93 c	Sm <sup>145</sup>	1.3(3)	163
Co <sup>58</sup>	3.5(3) 3.996(11) <sup>b</sup>	101 c	Tb <sup>156</sup>	1.41(18) <sup>c,d</sup>	167
Co <sup>60</sup>	3.5(5) 3.754(8) <sup>b</sup>	101 c	Tb <sup>160</sup>	1.56(25) <sup>c,d</sup>	169
Ag <sup>110m</sup>	+2.9(13) 3.55(4) <sup>b</sup>	134 50	Dy <sup>155</sup>	0.21(5) <sup>d</sup>	174
Sb <sup>125</sup>	2.8(2)	139,37	Dy <sup>157</sup>	0.32(2) <sup>d</sup>	174
Ce <sup>137m</sup>	0.69(3)	144	Ho <sup>166m</sup>	3.9(5) <sup>c,d</sup>	172
Ce <sup>137</sup>	0.90(15) <sup>d</sup>	141	Ir <sup>191m</sup>	6.3(15)	190
Ce <sup>139</sup>	0.95(20) <sup>d</sup> 0.85(15) <sup>d</sup>	141 148	Ir <sup>192</sup>	1.9(4)	192
Pm <sup>143</sup>	3.75(50) <sup>c,d</sup>	160	Au <sup>195</sup>	0.13(4)	63
Pm <sup>144</sup>	1.7(2) <sup>c,d</sup>	143	Es <sup>253</sup>	4.9(10)	3
Pm <sup>148m</sup>	1.80(18) <sup>c,d</sup>	160			

<sup>a</sup> Where no sign is given, only the magnitude is known. Error limits are given parenthetically after values.

<sup>b</sup> Moment determined spectroscopically.

<sup>c</sup> Many values have been corrected, subsequent to their original publication. See I. Lindgren, in Appendix 1 of "Perturbed Angular Correlations", ed. by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland, 1964).

<sup>d</sup> Subject to slight further correction because of temperature scale changes.

CMN and NES at the lowest temperatures. The new CMN scale extends the available absolute temperature range down to 1.9 mdeg. This region is becoming important in searches for superconductivity and superfluidity.

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FIGURE CAPTIONS

Fig. 1. Nuclear alignment apparatus used at Berkeley for charged-particle anisotropy studies.

Fig. 2. Experimental chamber for apparatus shown in Fig. 1.

Fig. 3. Illustration of the advantages of Ge(Li) detectors in nuclear orientation ( $184,206$ ). At left are shown NaI(Tl) (dashed curve) and Ge(Li) (solid curve)  $\gamma$ -ray spectra in the 700-850 keV region of the decay of  $\text{Ho}^{166m}$ . At right are shown angular distributions of five peaks resolved by Ge(Li) detector, when  $\text{Ho}^{166m}$  is oriented in NES.

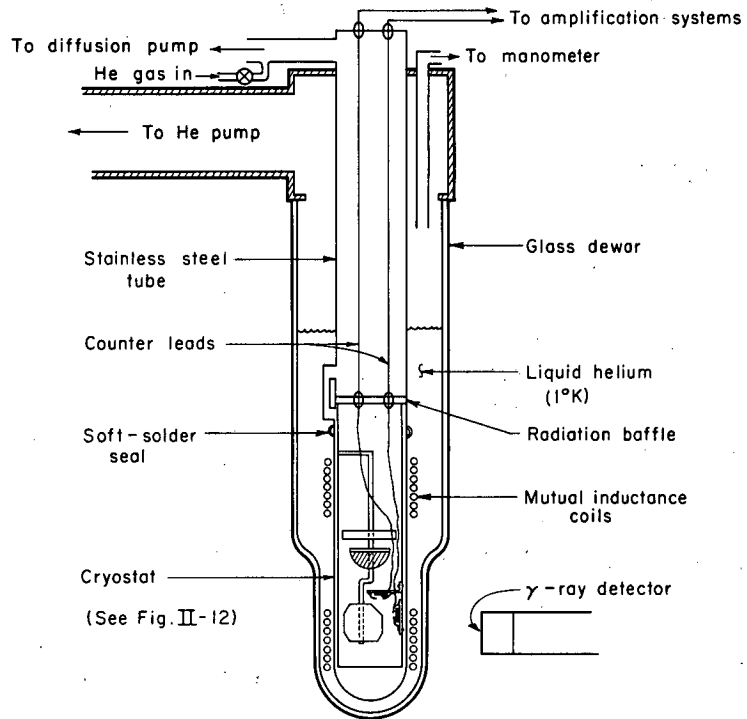
Fig. 4. Simplified representation of the Marshak-Richardson-Tamura experiments on total cross section of oriented  $\text{Ho}^{166}$  nuclei for 14 MeV neutrons. This experiment measures the deformation of the nuclear potential.

Fig. 5. Representation of the total (nuclear plus Coulomb) potential of a prolately deformed nucleus, of major (minor) radius  $r_a$  ( $r_b$ ), showing that alpha particles emitted from polar regions must penetrate a barrier that is both lower and thinner than at equator.

Fig. 6. Temperature dependence of angular distribution of alpha particles emitted from  $\text{Es}^{253}$  oriented in NES, from Ref. 145.

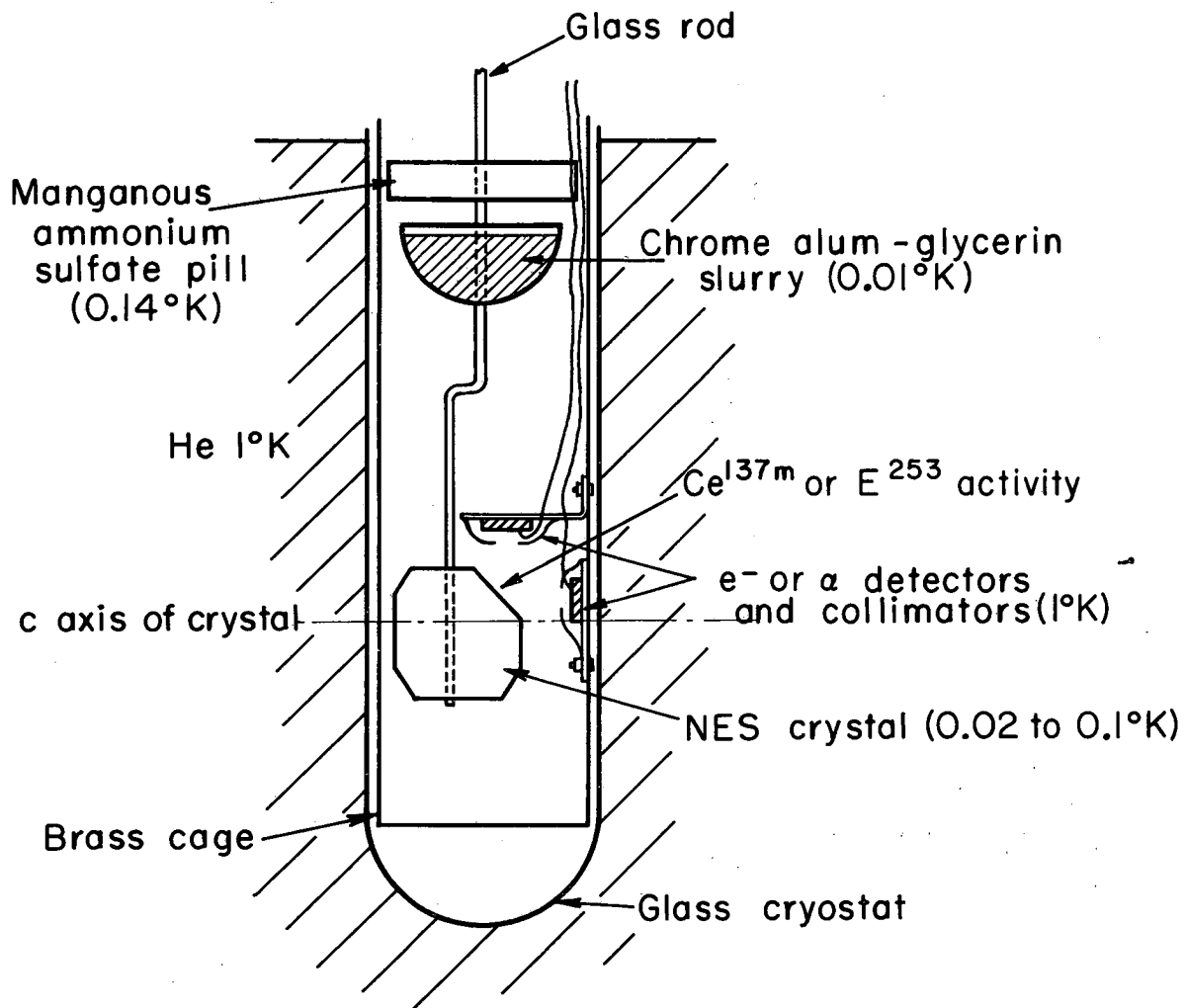
Fig. 7. Hyperfine magnetic fields for various solutes in Fe, Co, and Ni lattices (denoted by circles, triangles, and squares, respectively), plotted to emphasize regularities, especially maxima in 3d, 4d, and 5d groups. Filled figures are cases for which sign of field is measured: for open figures sign is surmised by author.

Fig. 8. Illustration of the origins of hyperfine fields from core polarization of inner s electrons and from conduction-electron polarization of 4s electrons. An external magnetic field is applied, polarizing the "magnetic" 3d electrons down. By ferromagnetic exchange interaction these 3d electrons polarize the 2s core electrons and 4s conduction electrons. This may be regarded as distortion of the radial wave functions of these electrons, with the spin up substate being attracted toward the 3d function. The result is a net "up" spin density for the 2s shell, and a net "down" spin density for the 4s band, at the nucleus. Electron-nucleus contact interaction gives an effective field antiparallel to the spin density in the nucleus, or antiparallel to the applied field (i.e., negative) for the core polarization term, which is often dominant.



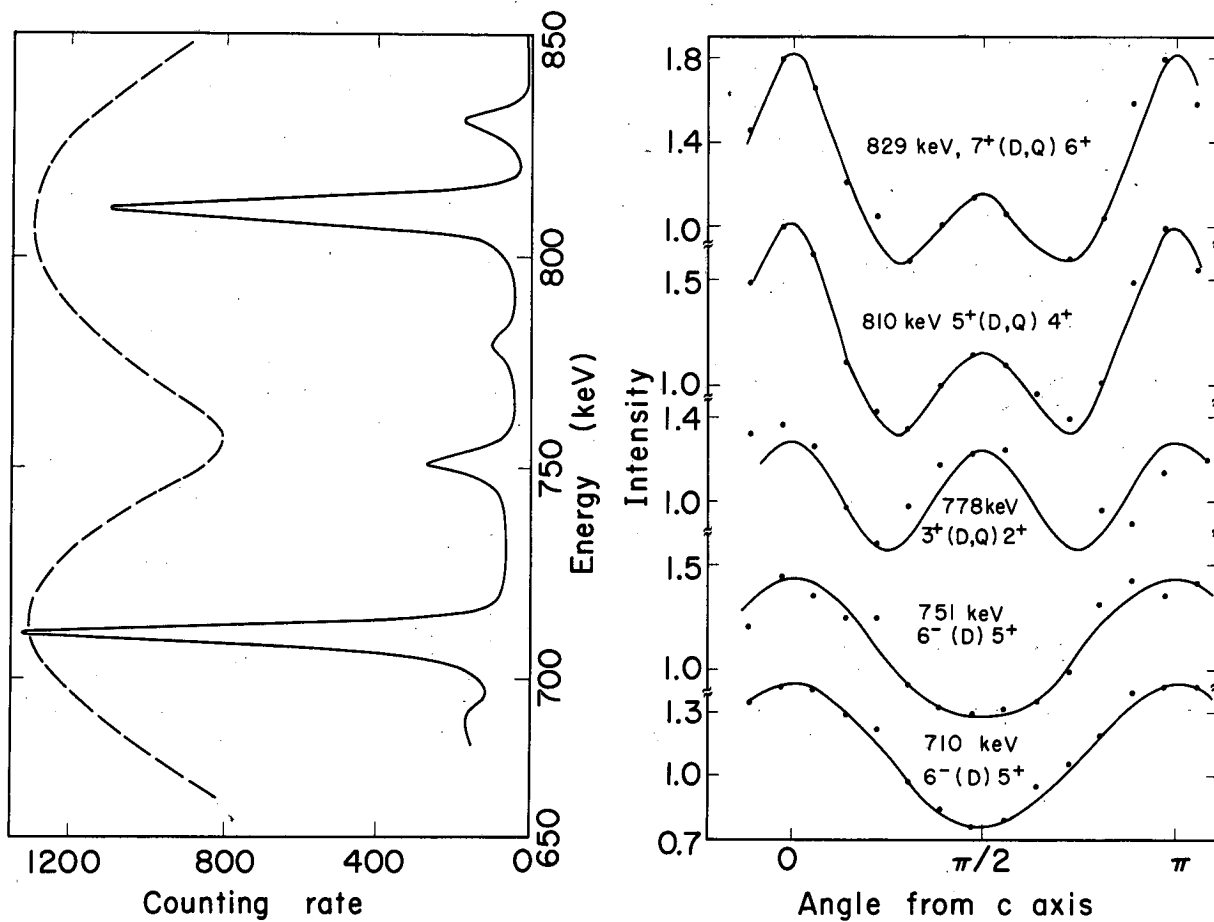
MU-35180

Fig. 1



MUB-2999

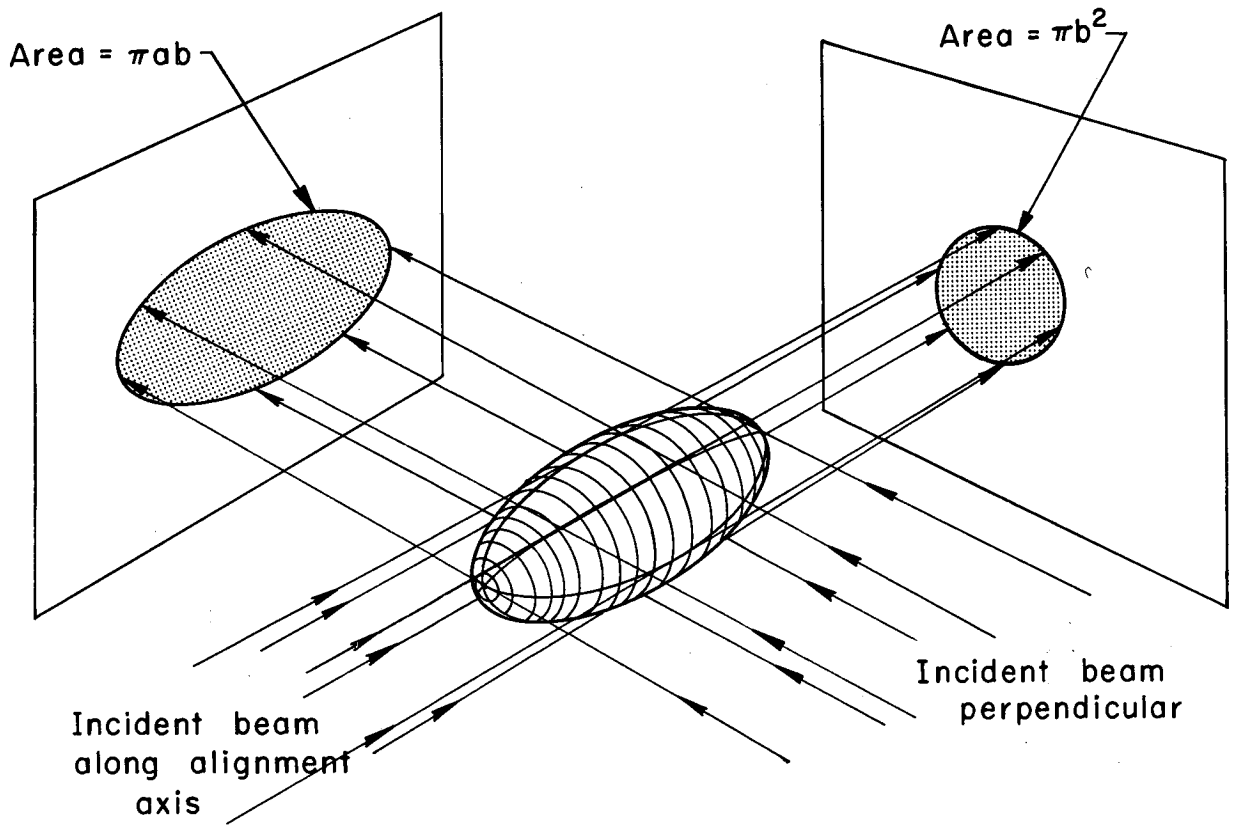
Fig. 2



MUB-40081

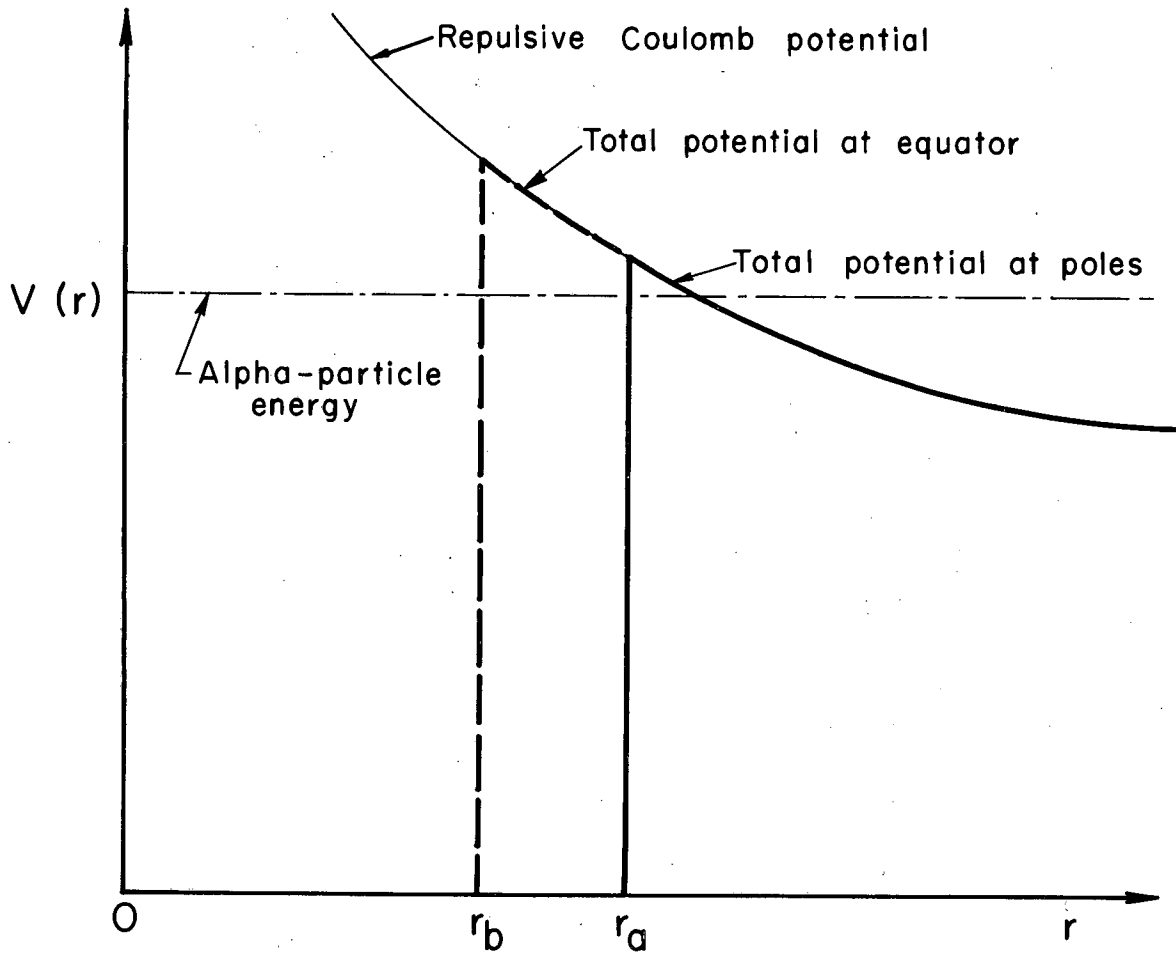
Fig. 3





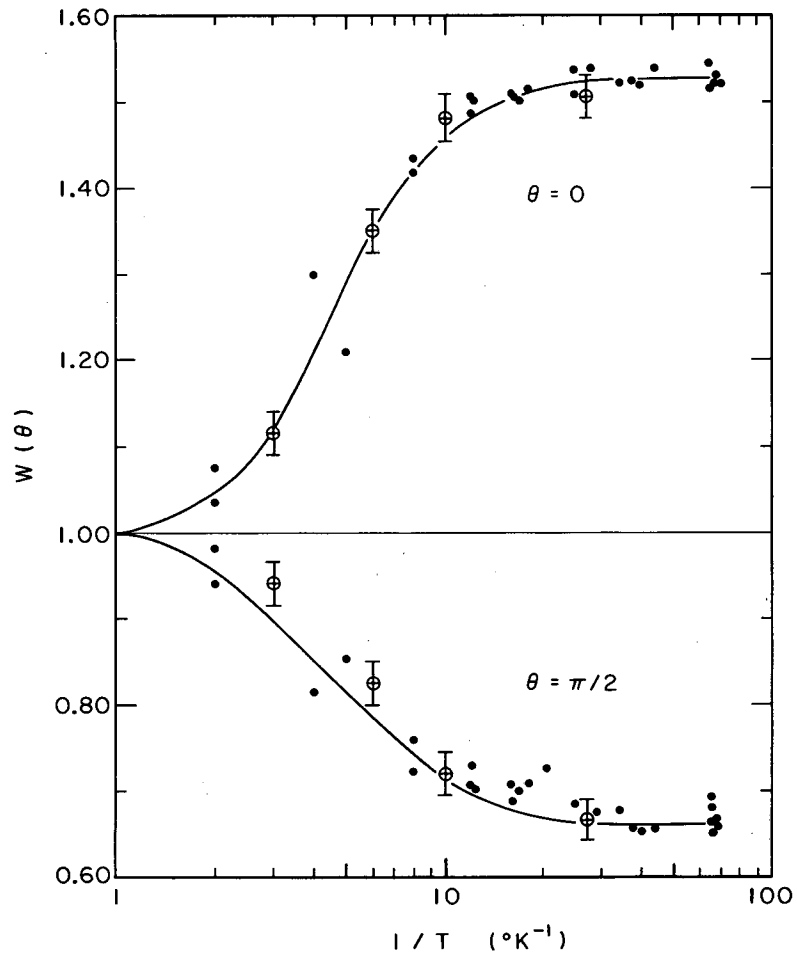
MUB-10079

- Fig. 4 -



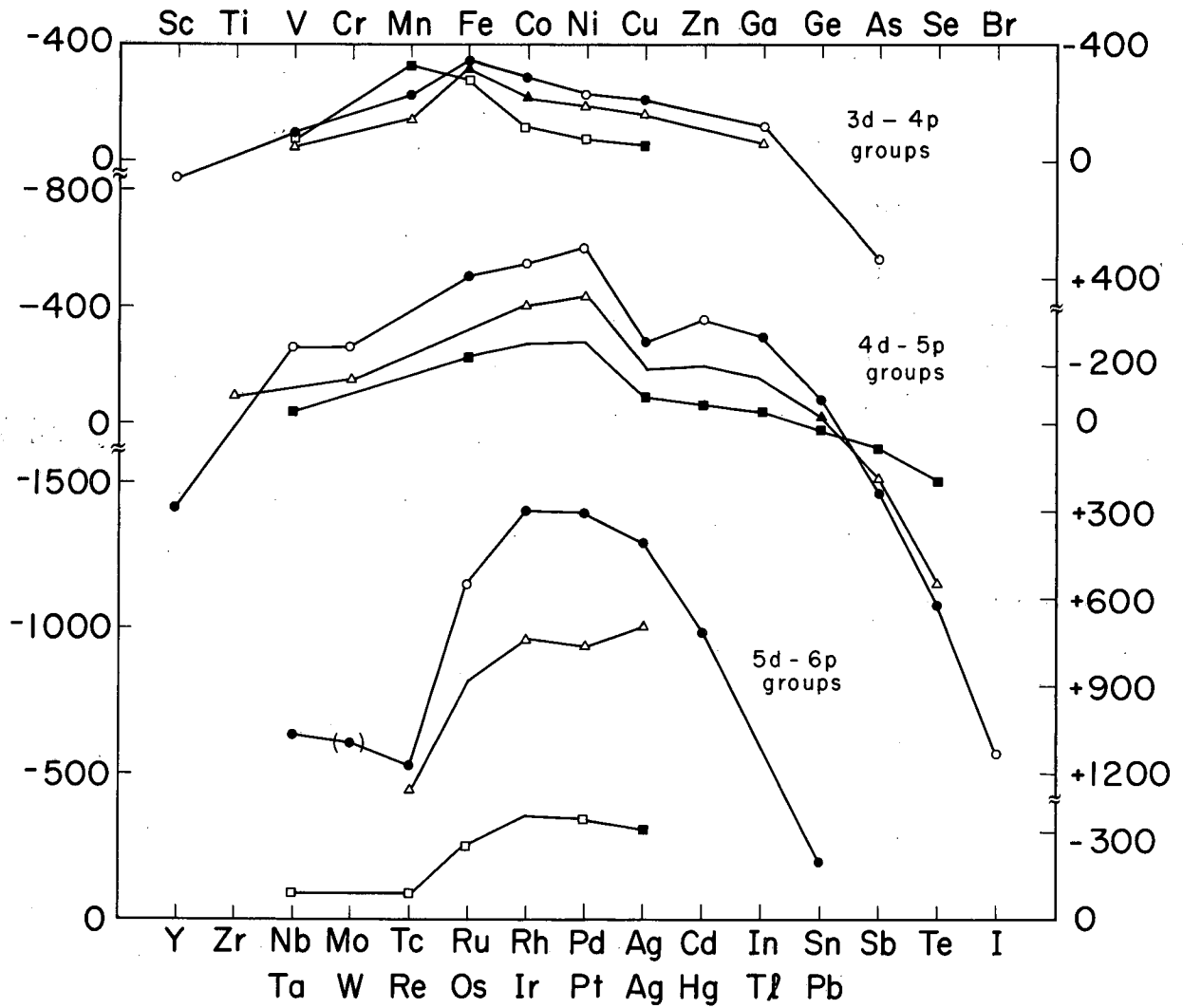
MUB-10078

Fig. 5



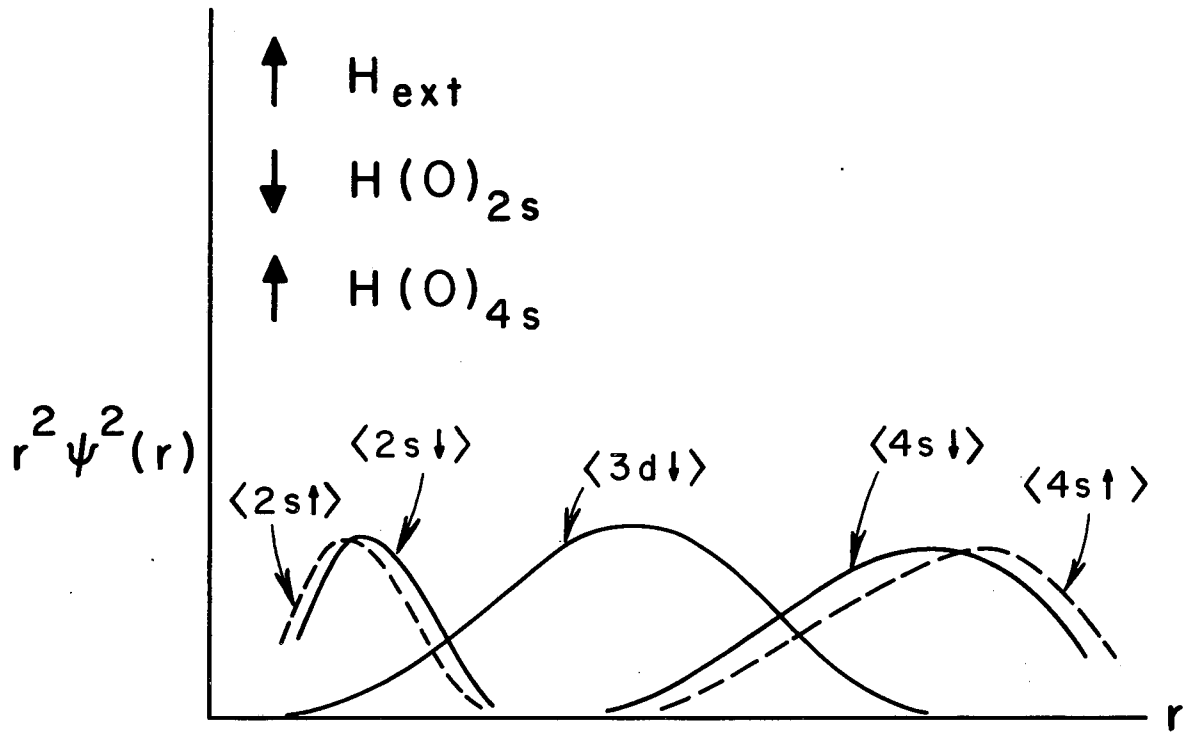
MU-35175

Fig. 6



MUB-10080

Fig. 7



MUB-10635

Fig. 8

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