Calorimetric Investigation of the Hyperfine Interactions in Metallic Nd, Sm, and Dy†

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The specific heats of metallic Nd, Sm, and Dy have been measured in the temperature range 0.025–0.4 K, where the nuclear (hyperfine) heat capacity predominates. The data are fitted to theoretical Schottky curves corresponding to a Hamiltonian of the form $H=a'I_z+P[I_z^2-\frac{1}{3}I(I+1)]$, where a' and P are the magnetic dipole and electric quadrupole interaction parameters, respectively. The Nd data are consistent with a two-sublattice model with the electronic moments on each sublattice appreciably less than the full saturation value. Both the Sm and Dy data fit Schottky anomalies corresponding to the full saturated electronic moment on each ion.

I. INTRODUCTION

THE hyperfine splitting of the rare-earth metals, of order 0.1 K, contributes a Schottky anomaly which dominates all other contributions to the specific heat. Since the nuclear spin, magnetic dipole moment, and, in some cases, electric quadrupole moment are known, calorimetric measurements near 0.1 K provide information concerning the hyperfine fields, the electronic magnetic ordering, etc. In the present paper, we report data from measurements on metallic Nd, Sm, and Dy. Their inclusion together is not based on any similarity as rare earths but rather because each metal has two stable isotopes which contribute to the nuclear heat capacity C_N. The analysis of the data becomes less certain in this case than for a single isotope since twice as many adjustable parameters are available.

The presence of large hyperfine fields in metallic Nd, Sm, and Dy has been detected in earlier heat capacity measurements made down to ~0.4 K,[1–8] where the high-temperature tail of the Schottky curve becomes evident. The hyperfine fields in metallic Dy have also been observed by nuclear magnetic resonance (NMR)[9] and Mössbauer[10] techniques at higher temperatures (1.4 and 20 K, respectively), and by neutron transmission at lower temperatures.[11] The latter two measurements could each detect only one isotope, Dy^{161} and Dy^{163}, respectively.

For the three rare earths of concern here, the hyperfine fields arise primarily from the electrons in the unfilled $4f$ shell of the host ion. As the $4f$ electrons are deeply buried within the electron cloud of the host ion the hyperfine fields produced are very large and, although the environment of the ion may indirectly affect the fields by the interaction of the host $4f$ shell with the crystal field and with neighboring ions through exchange, any direct influence of the environment should be negligible. The hyperfine Hamiltonian may therefore be written with considerable accuracy as[12]

$$H=a'I_z+P[I_z^2-\tfrac{1}{3}I(I+1)],$$

where I and I_z are the nuclear-spin quantum numbers, and a' and P are the magnetic dipole and electric quadrupole interaction parameters, respectively. Contributions from electrons other than $4f$ are considered to be included in a' and P. The off-diagonal terms in the quadrupole interaction are neglected as being too small to be observed in the present measurements.

If the hyperfine fields at all nuclear sites are the same, then the resulting Schottky specific-heat curve should be characterized by the two parameters a' and P. In the present situation, with two contributing isotopes, we must modify this statement to include four parameters a_1', a_2', P_1, P_2, since the magnitudes of the nuclear moments are reflected in a' and P.

In the rare earth metals, the electronic magnetic moment is not always the same at each lattice site, and hence the hyperfine fields are not the same for every nucleus. This fact complicates the analysis of calorimetric data, and it would be wise to review what is known about the magnetic order in Nd, Sm, and Dy before attempting an analysis.

The most precise data on the magnetic structure of metallic Nd have been obtained by neutron diffraction from a single crystal in the temperature range 1.6–

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20 K.[13] In Nd the double hexagonal-close-packed crystalline structure is such that alternate layers have different nearest-neighbor environments, either fcc or hexagonal. The neutron diffraction results for the most part were consistent with a model in which the hexagonal sites order in an antiferromagnetic arrangement at 19 K with a sinusoidal modulation of the moments within each layer. The wave vector of this modulation changes gradually with decreasing temperature until at 7.5 K the cubic sites order in a similar structure with a different modulation wave vector. The amplitude of the modulation at 1.6 K was found to be $(2.3\pm0.2)\mu_B$ (Bohr magnetons) for the hexagonal sites, and $(1.8\pm0.2)\mu_B$ for the cubic sites. These two magnetic transitions have also been observed in calorimetric and other measurements.[14–17] In brief, we do not expect the hyperfine fields to be the same at all nuclear sites in metallic Nd.

The situation is less clear in the case of Sm. An order-order transition at 14 K has been detected in various types of measurements.[15–21] At liquid-helium temperatures the metal is thought to be antiferromagnetic with moments parallel to the hexagonal axis.[21]

Neutron diffraction from a single crystal of Dy shows a ferromagnetic transition at 87 K below which the moments are parallel to the hexagonal axis.[22] The

TABLE I. Mass and impurity content of samples, heat-capacity contributions from electronic, lattice, and magnetic excitations, and ratios of nuclear moments used in the computations.

	Nd	Sm	Dy
Mass (g)	9.846	9.535	12.417
Mass (moles)	0.06825	0.06342	0.07641
Impurity (% weight)			
Al	0.015	···	0.04
B	0.0045	···	···
Ba	0.0045	···	0.01
C	0.025	0.02	0.008
Er	···	···	0.03
Eu	···	···	0.01
F	0.004	···	0.045
Fe	0.065	···	0.05
Gd	0.002	···	···
H	0.001	0.04	0.03
K	0.002	···	0.13
N	0.070	···	0.004
Na	0.050	···	···
Ni	0.0015	···	···
O	0.13	0.008	0.092
Pr	···	···	0.03
Ta	0.12	···	0.026
Th	···	···	0.02
Y	0.0015	···	0.09
Contributions to C–C_N (in J/K mole)			
T	0.0225	0.0121	0.0095
T^3	0.1257	0.0009	0.00022
$T^{3/2}$	···	···	0.0097
Ratios of nuclear moments			
a'_n/a'_{n+2}	+1.609	+1.213	−0.714
P_n/P_{n+2}	+1.894	−3.460	+0.943

TABLE II. Measured specific heat of neodymium metal in J/mole K.

T(K)	C	T(K)	C
Run I			
0.0307	1.498	0.1911	0.1795
0.0336	1.467	0.2085	0.1522
0.0375	1.416	0.2268	0.1311
0.0415	1.348	0.0602	1.005
0.0456	1.272	0.0663	0.9079
0.0499	1.194	0.0731	0.8087
0.0546	1.107	0.0928	0.5871
0.0599	1.016	0.1027	0.5036
0.0668	0.9022	0.1132	0.4322
0.0736	0.8038	0.1244	0.3718
0.0372	1.416	0.1375	0.3138
0.0413	1.351	0.1531	0.2610
0.0465	1.254	0.1713	0.2152
0.0520	1.154	0.1926	0.1749
0.0571	1.063	0.2187	0.1399
0.0628	0.9651	0.2486	0.1113
0.0692	0.8655	0.2809	0.0901
0.0763	0.7688	0.3140	0.0768
0.0843	0.6729	0.3628	0.0631
0.0930	0.5839	0.3920	0.0601
0.1025	0.5069	0.2247	0.1331
0.1128	0.4355	0.2556	0.1052
0.1231	0.3776	0.2815	0.0907
0.1344	0.3277	0.3153	0.0764
0.1475	0.2791	0.3244	0.0739
0.1623	0.2362	0.3423	0.0683
0.1776	0.2020	0.3676	0.0635
0.1753	0.2073		
Run II			
0.0264	1.521	0.3261	0.0739
0.0302	1.505	0.1312	0.3350
0.0340	1.464	0.1367	0.3134
0.0378	1.408	0.1429	0.2917
0.0417	1.342	0.1489	0.2770
0.0458	1.269	0.1579	0.2478
0.0501	1.192	0.1699	0.2187
0.0569	1.068	0.1790	0.1984
0.0925	0.6070	0.1937	0.1753
0.1298	0.3470	0.2094	0.1505
0.1416	0.2995	0.2296	0.1258
0.1709	0.2138	0.2419	0.1157
0.2103	0.1497	0.2812	0.0918
0.2356	0.1232	0.2983	0.0801
0.2612	0.1033	0.2998	0.0795
0.2877	0.0886	0.3309	0.0708

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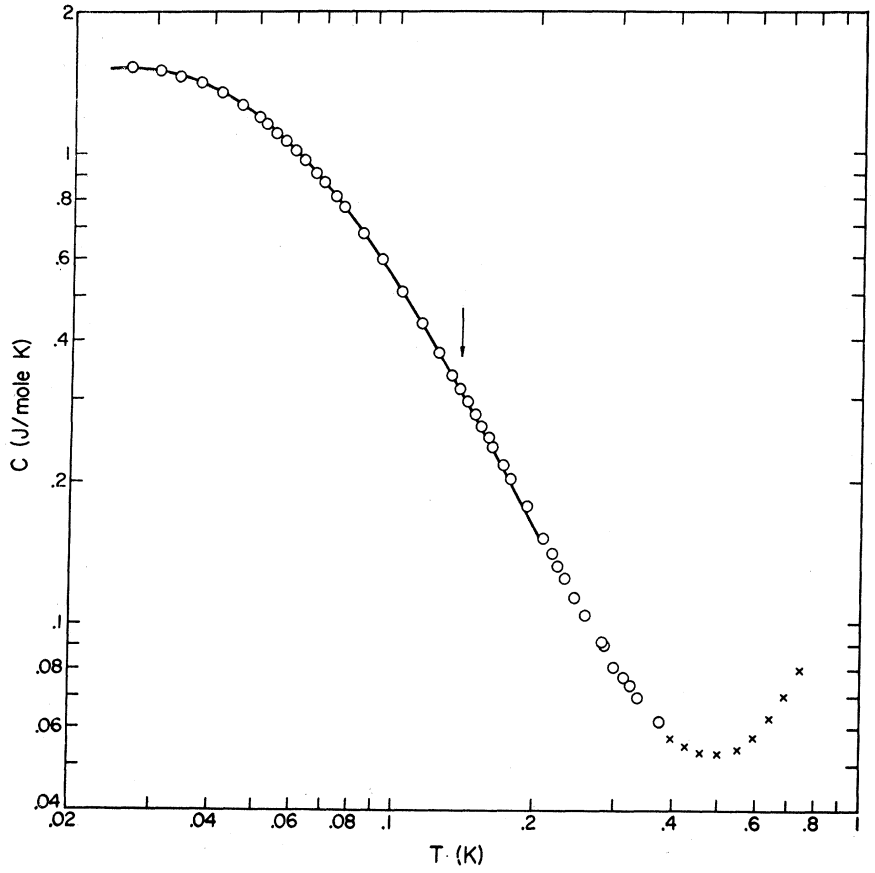

FIG. 1. Specific heat of Nd metal versus temperature. ○, present results; ×, Lounasmaa (Ref. 1). In each figure, the arrow indicates the temperature below which $C=C_N$ to within ~1%, and the solid curve represents our best fit to the nuclear contribution, C_N, as discussed in the text. For purposes of clarity, a few almost coincident points in these figures have been averaged.

magnetic moment, however, was found to be $\sim 9.5\mu_B$ as compared with the maximum value of $10.0\mu_B$ expected for the tripositive ion. Other measurements are consistent with ferromagnetic ordering.[23–30] We may therefore expect the hyperfine fields to be the same at all sites in metallic Dy.

II. EXPERIMENTAL

The calorimeter has been described in detail in an earlier publication.[31] The design was such that no problems with heat leak or thermal contact were encountered. Temperatures were measured with a magnetic thermometer (cerous magnesium nitrate) to a precision of $\pm 0.2\% \pm 10^{-4}$ K relative to the (1962) vapor pressure scale of liquid He^3. The over-all accuracy of the heat capacity determinations was ±1%. The heat capacity of the empty calorimeter was both measured and calculated, and represented a correction of 4% to the data for Sm or Nd at 0.4 K, less than 1% for Dy at 0.4 K. This correction rapidly became negligible towards lower temperatures.

The samples were each cut from larger pieces originally used in the calorimetric measurements of Lounasmaa.[1,4,8] The masses and results of spectrographic and chemical analyses are given in Table I.

III. RESULTS AND DISCUSSION

A. Neodymium

The results for two separate runs on metallic Nd are presented in Table II and Fig. 1. The data are in good agreement with the measurements of Lounasmaa which extended down to 0.4 K.[1] In general, the specific heats of the rare-earth metals can be written as the sum

$$C=C_N+C_E+C_L+C_M,$$

where C_E, C_L, and C_M are contributions from the electronic, lattice, and magnetic systems, respectively. These last three terms can be estimated from heat-capacity measurements made at higher tempera-

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tures,[1,15,32] and are summarized in Table I. Below $\sim$0.14 K, indicated by the arrow in Fig. 1, $C=C_N$ to within the experimental error of 1%.

To analyze the data we assume the Nd to have the normal isotopic composition of 12.20% Nd^{143} and 8.30% Nd^{145}.[33] The other stable isotopes being even-even nuclei have no spin and thus do not contribute to C_N. We also assume that the ratios of the interaction parameters for each isotope (a'_{143}/a'_{145}) and (P_{143}/P_{145}) are determined by the ratios of the corresponding nuclear magnetic dipole and electric quadrupole moments, respectively. These ratios, which are well known from other experiments,[34] are listed in Table I. This allows us to halve the number of independent parameters in the fitting process.

In the case of Nd, the analysis of the data is complicated by the complex magnetic structure of the metal. Nuclear diffraction measurements suggest that there are two magnetic sublattices, one comprising the hexagonal sites and the other the cubic sites, with a sinusoidal modulation of moments within each. Any analysis of the observed specific heat must take into account the fact that there are now four contributing Schottky anomalies arising from the two possible sites for each isotope, and any fit is rather uncertain in consequence. Nevertheless, the data cannot be fitted to a Hamiltonian representing such a modulated structure, this structure producing a much broader anomaly than we observe. On the other hand, if we assume a two sublattice model but with no modulation of moments on each site we obtain a good fit which gives (in Kelvin)

Sublattice I:

$$a'_{143}=-0.0366\pm0.0005, \quad P_{143}=-0.0005\pm0.0004,$$
$$a'_{145}=-0.0227\pm0.0003, \quad P_{145}=-0.0003\pm0.0002,$$

Sublattice II:

$$a'_{143}=-0.0251\pm0.0004, \quad P_{143}=-0.0005\pm0.0007,$$
$$a'_{145}=-0.0156\pm0.0002, \quad P_{145}=-0.0003\pm0.0004.$$

The uncertainties quoted are the variances of the parameters. The corresponding Schottky curve is plotted in Fig. 1. The sign of a' is determined from the known sign of the nuclear magnetic moment and the assumed electronic state of the host ion. The sign of P is determined directly from the specific-heat data.

Assuming Bleaney's theoretical values of a' [12] to be correct, our figures correspond to electronic moments of $2.58\mu_B$ and $1.77\mu_B$ on the two sublattices. These figures are close to the amplitudes of the sinusoidal modulation of moments suggested by neutron diffraction, namely, $(2.3\pm0.2)\mu_B$ and $(1.8\pm0.2)\mu_B$.[13] The agreement is especially interesting as we observe no such modulation as seen at 1.6 K but rather the same moment on all sites in the same sublattice. However, on the basis of this correspondence with the neutron diffraction findings, we have tentatively identified our sublattices I and II with the hexagonal and cubic sites, respectively.

It is possible that the modulated spin structure in Nd is not stable and that at sufficiently low temperatures the oscillation gradually becomes more "square-wave"-like and finally transforms to a series of antiphase domains within each basal layer with the same moment on each site. Similar behavior has been observed in Tm at higher temperatures by neutron diffraction.[35] The magnetic specific heat of Nd is very high in the liquid-helium range,[1] which also suggests that the electronic moments are still ordering at these temperatures. The fact that the moments on the two sublattices are considerably less than the free-ion value of $3.27\mu_B$ supports Bleaney's proposal[12] that in Nd the crystal-field effects should reduce $\langle J_Z\rangle$ below the maximum $\langle J_Z\rangle=J$, the different values on the two sites reflecting the differing symmetries of the corresponding crystal fields.

TABLE III. Measured specific heat of samarium metal in J/mole K.

T(K)	C	T(K)	C
0.0323	2.141	0.0453	1.803
0.0361	2.052	0.0504	1.656
0.0414	1.917	0.0558	1.506
0.0468	1.765	0.0618	1.354
0.0524	1.606	0.0686	1.196
0.0585	1.438	0.0761	1.041
0.0655	1.266	0.0847	0.8968
0.0734	1.095	0.0949	0.7537
0.0826	0.9287	0.1070	0.6220
0.0938	0.7699	0.1208	0.5068
0.1072	0.6218	0.1365	0.4114
0.1237	0.4891	0.1560	0.3243
0.1451	0.3685	0.1788	0.2521
0.1735	0.2667	0.2055	0.1956
0.1971	0.2114	0.2286	0.1608
0.2302	0.1587	0.2623	0.1240
0.2784	0.1120	0.3034	0.0930
0.3152	0.0884	0.3460	0.0757
0.3275	0.0825	0.0241	2.176
0.3700	0.0665	0.0286	2.165
0.4167	0.0539	0.0320	2.128
0.4159	0.0537	0.0348	2.075
0.0355	2.046	0.0288	2.173
0.0402	1.955		

B. Samarium

The results for metallic samarium are presented in Table III and Fig. 2. The low-temperature data of Lounasmaa[4] and of Trolliet[36] are also included in Fig. 2. The arrow indicates the temperature below which $C=C_N$ to within 1%. The terms C_E, C_L, and C_M,

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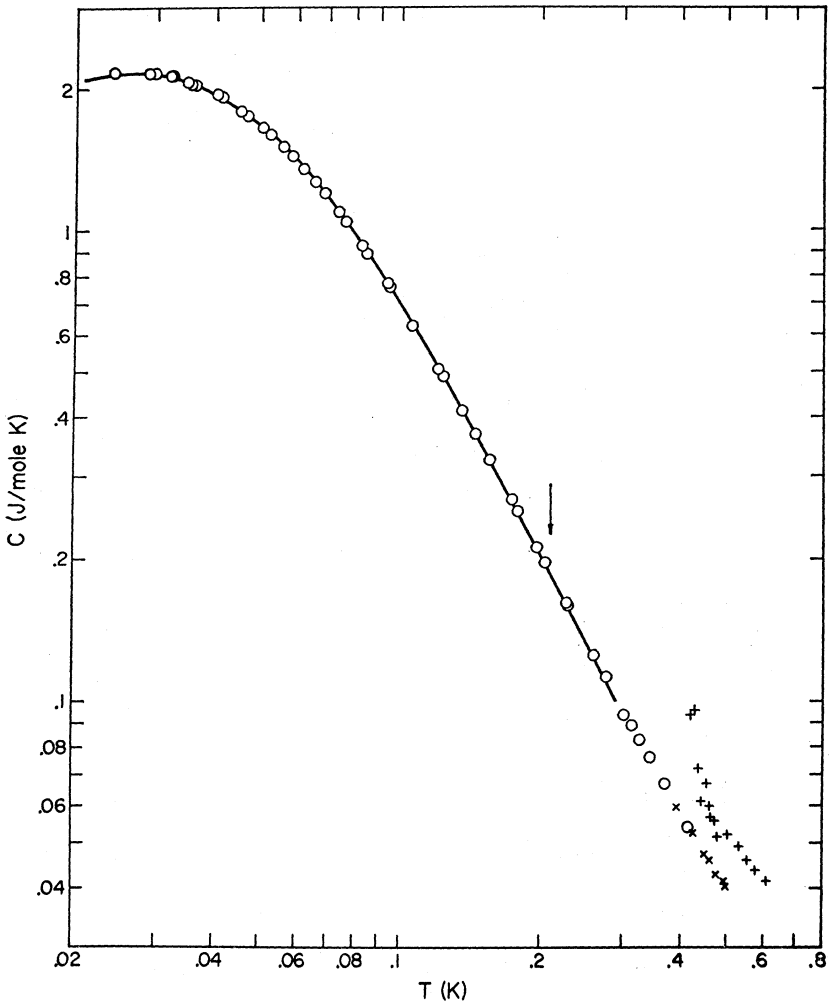

FIG. 2. Specific heat of Sm metal versus temperature. ○, present results; ×, Lounasmaa (Ref. 4); +, Trolliet (Ref. 36).

estimated from high-temperature measurements,[4,15,32] are given in Table I.

To analyze the data, we assume a normal isotopic abundance of 15.07% of Sm^{147} and 13.84% of Sm^{149},[33] other stable isotopes not contributing to C_N. We also assume that the ratios of the parameters (a'_{147}/a'_{149}) and (P_{147}/P_{149}) are determined by the ratios of the corresponding nuclear moments known from other experiments.[34] A two-parameter fit gives (in Kelvin)

$$a'_{147}=-0.0283\pm0.0008, \quad P_{147}=-0.0002\ \pm0.0003,$$
$$a'_{149}=-0.0233\pm0.0007, \quad P_{149}=+0.00005\pm0.00008,$$

and fits our data to within 1% as shown in Fig. 2. The values of a' are ~2% lower than Bleaney's calculated values.[12] As there are no other measurements of hyperfine interactions in Sm there is little we can say except that the excellent agreement between the theoretical and experimental values of a' indicates that the electronic moment is fully developed at all sites ($\langle J_z\rangle=J$). This is compatible with the simple antiferromagnetic structure which has been tentatively proposed for Sm as mentioned in Sec. I.

C. Dysprosium

The results for metallic Dy are presented in Table IV and Fig. 3. Figure 3 also includes the low-temperature results of Parks,[7] Lounasmaa,[8] Trolliet[36] and Dash *et al.*[5] The arrow indicates the temperature below which $C=C_N$ to within 1%. The terms C_E, C_L, and C_M, as summarized in Table I, have been estimated from high-temperature specific-heat measurements.[37–39]

To analyze the data we assume Dy to have the normal isotopic composition of 18.88% Dy^{161} and 24.97% Dy^{163}.[33] The other stable isotopes do not contribute to C_N. We also assume that the ratios of the interaction

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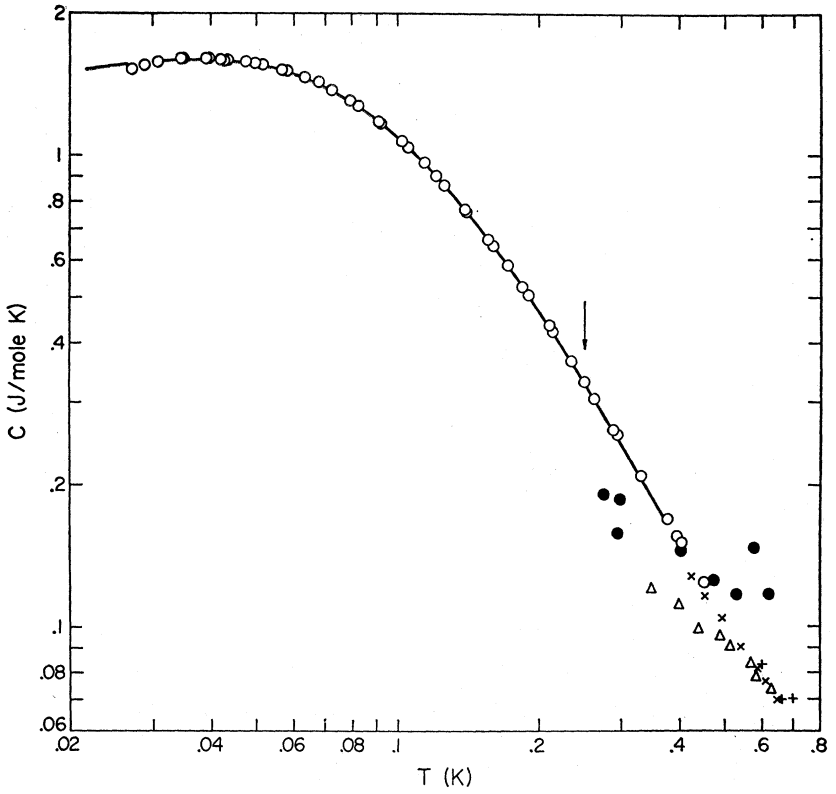

FIG. 3. Specific heat of Dy metal versus temperature. ○, present results; ●, Dash *et al.* (Ref. 5); △, Parks (Ref. 7); ×, Lounasmaa and Guenther (Ref. 8); +, Trolliet (Ref. 36).

parameters (a'_{161}/a'_{163}) and (P_{161}/P_{163}) are the same as the ratios of the corresponding nuclear moments.[34] A two-parameter fit yields (in Kelvin)

$$a'_{161}=-0.0396\pm0.0003, \quad P_{161}=+0.009\pm0.003,$$
$$a'_{163}=+0.0554\pm0.0004, \quad P_{163}=+0.010\pm0.004.$$

The corresponding Schottky curve is plotted in Fig. 3.

In the case of Dy, there are several determinations of the hyperfine parameters for comparison. NMR measurements disclosed a well-resolved spectrum,[9] but with one of the expected lines missing. Assuming the missing line to have been due to instrumental difficulties gave

$$a'_{161}=0.0399\text{K}, \quad P_{161}=0.0093\text{K},$$
$$a'_{163}=0.0559\text{K}, \quad P_{163}=0.0098\text{K},$$

with an accuracy of $\sim0.2\%$ in a' and $\sim1\%$ in P. A Mössbauer measurement[10] has been possible only with Dy^{161} yielding $a'_{161}=(0.0388\pm0.0014)$K, $P_{161}=(0.0077\pm0.0006)$K. Neutron transmission[11] has given $a'_{163}=(0.061\pm0.005)$K. Thus, the various measurements are in reasonable agreement. The accord between the calorimetric and NMR parameters is especially good. However, it is from the NMR measurement that the ratios (a'_{161}/a'_{163}) and (P_{161}/P_{163}) used in our calculation are taken and hence the agreement may be forced to some extent.

We note that the hyperfine parameters agree within experimental error with the values calculated by Bleaney.[12] This suggests that the electronic moment is fully developed $(\langle J_Z\rangle=J)$, contrary to the neutron diffraction result which indicates a moment $\sim5\%$ less than maximum.

TABLE IV. Measured specific heat of dysprosium metal in J/mole K.

T(K)	C	T(K)	C
0.0289	1.562	0.1402	0.7603
0.0351	1.613	0.1559	0.6657
0.0420	1.602	0.1719	0.5849
0.0497	1.572	0.1897	0.5066
0.0584	1.514	0.2103	0.4357
0.0680	1.427	0.2342	0.3673
0.0793	1.310	0.2623	0.3054
0.0913	1.181	0.2943	0.2556
0.1053	1.042	0.3325	0.2086
0.1211	0.9049	0.3781	0.1691
0.1391	0.7692	0.4041	0.1496
0.1598	0.6425	0.0272	1.533
0.1842	0.5269	0.0308	1.579
0.2140	0.4222	0.0347	1.614
0.2498	0.3302	0.0397	1.610
0.2888	0.2606	0.0437	1.600
0.3324	0.2048	0.0475	1.584
0.3774	0.1668	0.0518	1.557
0.0634	1.470	0.0568	1.524
0.0723	1.382	0.0350	1.609
0.0823	1.278	0.0390	1.612
0.0923	1.170	0.0432	1.598
0.1026	1.070	0.3971	0.1549
0.1138	0.9652	0.4513	0.1243
0.1263	0.8647	0.3958	0.1541

TABLE V. Summary of the hyperfine parameters for metallic Nd, Sm, and Dy as determined by various techniques. a' and P are given in Kelvin.

Method	Isotope	Crystal site	a'		P		Ref.
Theory	Nd^{143}		−0.0463		+0.000024		
(for $\langle J_z\rangle=J$)	Nd^{145}			−0.0288		+0.000012	12
Calorimetric	Nd^{143}	hexagonal	−0.0366		−0.0005		
		cubic	−0.0251		−0.0005		
	Nd^{145}	hexagonal		−0.0227		−0.0003	This
		cubic		−0.0156		−0.0003	work
Theory	Sm^{147}		−0.0290		−0.000078		
	Sm^{149}			−0.0238		+0.000023	12
Calorimetric	Sm^{147}		−0.0283		−0.0002		This
	Sm^{149}			−0.0233		+0.00005	work
Theory	Dy^{161}		−0.0394		+0.0072		
	Dy^{163}			+0.0547		+0.0086	12
NMR	Dy^{161}		0.0399		0.0093		
	Dy^{163}			0.0559		0.0098	9
Mössbauer	Dy^{161}		0.0388		0.0077		10
Neutron trans.	Dy^{163}			0.061			11
Calorimetric	Dy^{161}		−0.0396		+0.009		This
	Dy^{163}			+0.0554		+0.010	work

IV. CONCLUSIONS

Calorimetric measurements on the metallic rare earths have been notoriously irreproducible at higher temperatures, presumably as a result of impurities, and may also prove to be so in the present temperature range. However, in view of the fact that our data were independent of magnetic and thermal history and agree with NMR results where available (Dy,Tb),[31] we assume the heat capacity may properly be represented by the sum $C=C_N+C_E+C_L+C_M$, where C_E, C_L, and C_M have approximately the values quoted in Table I and hence may be ignored below ~0.2K.

Table V summarizes the various determinations of the hyperfine parameters for the three metals. Our data for Nd can be fitted best to a two-sublattice model with the electronic moments on the two nonequivalent sites both reduced considerably from the free-ion value by the crystal field. The data cannot be reconciled to the sinusoidal modulation of moments suggested by neutron diffraction at 1.6K.

For Sm there are no data with which to compare our results. However, the values of a' are in excellent agreement with theoretical predictions and indicate that the electronic moment is fully developed on all lattice sites.

For Dy our data are in good agreement with NMR and other measurements and with the theoretical predictions, which also indicates a fully developed electronic moment.

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