With an MgO sample containing  $\sim\!10^{17}$  spins, the inverted state persisted for about 2.5 milliseconds at 4.2°K. Amplification was observed with a gain of 20 db at 125 microseconds after inversion, falling to 3 db at 720 microseconds.

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U. S. Air Force. <sup>1</sup> The first attempt of this kind was reported by Comprusson, Honig, and Townes [Compt. rend. **242**, 2451 (1956)], using doped silicon.

<sup>2</sup> R. A. Weeks, J. Appl. Phys. 27, 1376 (1956). <sup>3</sup> Wertz, Auzins, Weeks, and Silsbee, Phys. Rev. 107, 1535 (1957)

<sup>(1957)</sup>: <sup>4</sup> F. Bloch, Phys. Rev. 70, 460 (1946).
 <sup>5</sup> N. Bloembergen and R. V. Pound, Phys. Rev. 95, 8 (1954).
 <sup>6</sup> P. F. Chester and D. I. Bolef, Proc. Inst. Radio Engrs. 45, 1287 (1957); D. I. Bolef and P. F. Chester, Trans. Inst. Radio

<sup>12</sup> Ford and Technique **MTT-6**, 47 (1958). <sup>7</sup> Feher, Gordon, Buehler, Gere, and Thurmond, Phys. Rev. 109, 221 (1958).

## Energy Levels of an Asymmetric Rotor

## J. G. BAKER

Physical Chemistry Department, University of Cambridge, Cambridge, England (Received January 20, 1958)

ICROWAVE spectroscopy is at present seriously

M handicapped by the lack of sufficiently extensive tables of the rotational energy of a rigid rotor. This is commonly described either by the  $E_{\tau}$ ,  $\kappa$  rotation of King, Hainer, and Cross<sup>1</sup> or by a series expansion<sup>2</sup> of a quantity w in powers of the Wang asymmetry parameter, b. The available tables<sup>2</sup> give  $E_{\tau}$  at 0.01 intervals of  $\kappa$  up to J=12. More recent compilations<sup>3,4</sup> give the coefficients in the series expansion up to J=40, but this converges prohibitively slowly when  $|\kappa| < 0.9$  and the K value is small.

Work in progress here on bent triatomic molecules requires accurate energies for J > 12 and large asymmetries. A satisfactory technique has been developed by use of the method of Golden.<sup>5</sup> This originally required an exceptionally good tabulation of characteristic values M(s) of the Mathieu equation of parameter s. The new method instead uses the energy tables<sup>2</sup> for low J to obtain the energy levels of higher J by a "stepup" process, to be described.

Golden gives the result:

$$w = M(4\theta) - 2\theta + \alpha \theta' + \beta \theta'^2 + \cdots,$$

where

$$\theta = \frac{1}{2} |b| \left[ J(J+1) - 1 - \frac{1}{2J(J+1)} \right],$$
  
$$\theta' = \frac{1}{2} |b| \left[ 1 + \frac{1}{2J(J+1)} \right],$$

the terms in  $\alpha$  and  $\beta$  being small corrections.

The stepup process consists of increasing J while simultaneously decreasing the asymmetry, b, so that  $\theta$ remains constant. The quantity w then also remains constant, apart from small variations in  $\theta'$ . The appropriate value of w for the lower J and larger asymmetry is found from the tables,<sup>2</sup> and corrected for higher J by using the values of  $\alpha$  and  $\beta$  listed by Golden.<sup>5</sup> In practice it was necessary to develop more complicated relations in the  $E_{\tau}$ ,  $\kappa$  rotation, but this does not affect the rapidity and convenience of the process.

Table I compares tabulated values of  $E_{\tau}$  for the level  $12_{12,0}$  with values calculated by the above method and by the series expansion up to  $b^6$ .

There is good agreement for asymmetries so large that the series oscillates violently, though agreement

TABLE I. Comparison of values of  $E_{\tau}$  for the level  $12_{12,0}$ .

· · · ·	κ=0.90	κ=0.80
Tabulated value	151.111552	148.695733
Stepup from 11 <sub>11.0</sub>	151.111558	148.695724
Stepup from $6_{60}$	151.111397	148.694102
Stepup from $3_{30}$	151.109561	148.572802
Stepup from $2_{20}$	151.068250	
Series expansion	151.923200	230.240702

becomes less good as the asymmetry increases, making the method valueless for  $|\kappa| < 0.5$ . It is also inaccurate when K is comparable with J, but the series expansion is then satisfactory. The value of the technique is that it gives satisfactory energies where other methods fail, and could yield even better results by use of these as starting points for an iteration solution of the secular determinant. Much tedious computation can also be saved by the recently suggested use of energy moments.<sup>6</sup>

I am grateful to the Department of Scientific and Industrial Research for a Studentship supporting this work and to Dr. T. M. Sugden for helpful discussions.

 <sup>1</sup> King, Hainer, and Cross, J. Chem. Phys. 11, 27 (1943).
 <sup>2</sup> C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill Book Company, Inc., New York, 1955).
 <sup>3</sup> R. H. Schwendeman, J. Chem. Phys. 27, 986 (1957).
 <sup>4</sup> S. R. Polo, Can. J. Phys. 35, 880 (1957).
 <sup>5</sup> S. Golden, J. Chem. Phys. 16, 78 (1948).
 <sup>6</sup> L. C. Brown and P. M. Parker, J. Chem. Phys. 27, 1109 (1957). (1957).

## Specific Heat of He<sup>3</sup> below 1°K

D. F. BREWER,\* A. K. SREEDHAR, H. C. KRAMERS,† and J. G. Daunt

Department of Physics and Astronomy, Ohio State University, Columbus, Ohio

(Received February 5, 1958)

HE first measurements of the specific heat of liquid He<sup>3</sup> under its saturated pressure by deVries and Daunt<sup>1</sup> covered the temperature range 0.5°K to 2.3°K. Subsequent measurements by Roberts and



FIG. 1. Molar specific heat of liquid He<sup>3</sup> at a pressure of 12–15 cm Hg.  $\bigcirc$  present results. Previous measurements at the saturated vapor pressure are shown as:  $\triangle$  deVries and Daunt<sup>1</sup>;  $\bullet$  Roberts and Sydoriak<sup>2</sup>;  $\times$  Abraham, Osborne, and Weinstock <sup>3</sup>

Sydoriak<sup>2</sup> extended the range of observation down to  $0.37^{\circ}$ K and those by Osborne, Abraham, and Weinstock<sup>3</sup> down to  $0.23^{\circ}$ K. The later measurements, carried out with greater volumes of He<sup>3</sup>, were of higher accuracy. All the data were consistent and showed that no marked anomaly, such as a  $\lambda$ -point, occurred in the range of temperature studied. They also showed the curious result that between about  $0.3^{\circ}$ K and  $1.0^{\circ}$ K the specific heat varied little with temperature, and no unambiguous extrapolation of the curve could be made to  $T=0^{\circ}$ K. It was considered important therefore to extend the observations to yet lower temperatures, and measurements down to  $0.085^{\circ}$ K are reported herewith.

An adiabatic calorimeter of volume 0.5 cm<sup>3</sup> was used which was cooled below 1°K by a paramagnetic salt, the thermal connection being made through a superconducting Pb thermal valve which could be opened and closed magnetically, as described elsewhere.<sup>4</sup> A 100ohm manganin heater was used and temperatures were determined by susceptibility measurements of a cerium magnesium nitrate specimen thermally attached to the calorimeter. This salt was chosen since it accurately follows Curie's law in the temperature range covered,<sup>5</sup> and has a negligible heat capacity. Details of the apparatus and method of measurement are deferred to a later publication.

The results of our measurements of the specific heat of pure<sup>6</sup> He<sup>3</sup> under a pressure of 12 to 15 cm Hg are

shown in Fig. 1, which also gives the data from previous observations made at the saturated vapor pressure.<sup>1-3</sup> It is estimated that the smooth curve drawn through our data is accurate within  $\pm 2\%$ . No corrections have been made for the differences between  $C_p$  and  $C_v$ , which are negligible in this temperature range.

It will be seen from Fig. 1 that the smooth curve falls close to the previous results down to 0.25°K and that below this temperature it continues to diminish smoothly with decreasing temperature down to 0.085°K. The curve appears to allow a linear extrapolation to 0°K to be made with a slope of 3.75 cal/mole-deg<sup>2</sup>. The justification for this linear extrapolation is reinforced by appeal to the entropy diagram of Fig. 2, which has been calculated by using the linear extrapolation and the measured specific heat. At 0.5°K we find  $S_{\text{lig}} = 1.39$  $\pm 0.03$  cal/mole-deg. Since the difference between the specific heat under 12 to 15 cm Hg and under the saturated vapor pressure is small in the range of our measurements, it is permissible to compare this figure with the value of 1.44 cal/mole-deg calculated for  $S_{\text{lig}}$ at saturated vapor pressure from the most recent vapor pressure data.7

We feel that the experimental results reported here, together with the magnetic susceptibility data of Fairbank *et al.*<sup>8</sup> and the recent theoretical work of Brueckner and Gammel,<sup>9</sup> now provide sufficient evidence to give a high degree of certainty to the linear extrapolation that we have made, particularly in view of the close agreement (within about 2%) between the experimental and theoretical values of the linear term. Rigorously there always remains the experimental possibility of a transition, possibly accompanied by the appearance of superfluidity, at temperatures lower than those of our measurements; but we feel that the data presented here make this possibility now unlikely.



FIG. 2. Molar entropy of liquid He<sup>3</sup> at a pressure of 12-15 cm Hg, calculated from the present specific heat measurements by using a linear extrapolation from 0.085°K to 0°K.

It is generally agreed  $^{10}$  that the condition leading to superfluidity in He<sup>4</sup> is the absence of low-lying energy states apart from the collective phonon excitations. It follows from the linear Fermi-type specific heat that in He<sup>3</sup> an abundance of low single-particle excitations exists down to 0°K, thus preventing the occurrence of superfluidity, a conclusion inherent in the original statistical approach of London.<sup>11</sup>

The marked deviation from an ideal Fermi function above about 0.2°K indicates a more complex character for the excitations at these higher temperatures. It is anticipated that further insight into the nature of the excitations will result from measurements on the liquid under pressure, which are now in progress. Further discussion is therefore postponed.

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On leave of absence from the Kamerlingh-Onnes Laboratory, Leiden University, Leiden, Netherlands. <sup>1</sup>G. de Vries and J. G. Daunt, Phys. Rev. 92, 1572 (1953);

93, 631 (1954). <sup>2</sup> T. R. Roberts and S. G. Sydoriak, Phys. Rev. 93, 1418 (1954);

98, 1672 (1955)

<sup>6</sup>Osborne, Abraham, and Weinstock, Phys. Rev. 94, 202 (1954); Abraham, Osborne, and Weinstock, Phys. Rev. 98, 551 (1955). We are grateful to Dr. Osborne for providing us with a table of their results.

<sup>4</sup>C. V. Heer and J. G. Daunt, Phys. Rev. **76**, 854 (1949); J. G. Daunt, Proc. Phys. Soc. (London) **B70**, 641 (1957). <sup>5</sup>J. M. Daniels and F. N. H. Robinson, Phil. Mag. **44**, 630

(1953).

<sup>6</sup> The He<sup>3</sup> was kindly lent to us by the Stable Isotopes Division of the U. S. Atomic Energy Commission. <sup>7</sup> T. R. Roberts and S. G. Sydoriak, Phys. Rev. 98, 1672

(1955)

<sup>(1955).</sup>
<sup>8</sup> Fairbank, Ard, and Walters, Phys. Rev. **95**, 556 (1954);
<sup>W.</sup> M. Fairbank and G. K. Walters, Symposium on He<sup>3</sup>, Ohio State University, August, 1957 (unpublished).
<sup>9</sup> K. A. Brueckner and J. L. Gammel, Symposium on He<sup>3</sup>, Ohio State University, August, 1957 (unpublished).
<sup>10</sup> See, for example, R. P. Feynman, *Progress in Low-Temperature Physics* (North-Holland Publishing Company, Amsterdam, 1955), Vol. 1, p. 17.
<sup>11</sup> F. London, Nature **163**, 694 (1949), For other references see

<sup>11</sup> F. London, Nature **163**, 694 (1949). For other references see J. G. Daunt, *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1952), Vol. 1, p. 209.

## Electron Nuclear Double Resonance of Neodymium\*

DONALD HALFORD, CLYDE A. HUTCHISON, JR., AND Peter M. Llewellyn

Enrico Fermi Institute for Nuclear Studies and Department of Chemistry, University of Chicago, Chicago, Illinois (Received February 5, 1958)

HE method of electron nuclear double resonance has recently been employed by Feher<sup>1</sup> for investigation of some solid state problems. In this letter we discuss the application of the method to an investigation of Nd<sup>+3</sup> in LaCl<sub>3</sub> crystals.

Both  $Nd^{143}$  and  $Nd^{145}$  have nuclear spin 7/2. The lowest crystal field state of  $Nd^{+3}(f^3, {}^4I_{9/2})$  in LaCl<sub>3</sub> is a Kramers doublet described approximately by<sup>2</sup>

$$0.941 \pm 7/2 \langle -0.339 \pm 5/2 \rangle.$$

The energies of the hyperfine levels are therefore given by<sup>3</sup>

$$W(M,m) = -\frac{1}{4}hcA + hcP\{M^{2} + m^{2} + 2Mm - 5\}$$
  

$$\pm \frac{1}{2}\{[(M+m)hc(A-2P) + g_{||}\beta H]^{2} + (hcB)^{2}[16 - (M+m)^{2}]\}^{\frac{1}{2}} - g_{N}'\beta Hm, \quad (1)$$

for the case in which H is parallel to the c axis of the