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Abstract

Heat capacity measurements of the iron garnets of Y, Gd, Er, Ho, and Yb between 1.4° and 20°K are presented. Below 5°K, the specific heat of YIG can be represented by the sum of a lattice term proportional to T^3 and the spin-wave contribution $2.15 \times 10^{-3} T^{3/2}$ joules/mole-deg. This last term agrees satisfactorily with that calculated from a spin-wave analysis, in which the exchange interaction coefficients were those derived from Pauthenet's magnetization data. The results of the magnetic specific heat of the rare earth ions could be interpreted in terms of a Weiss molecular field acting on these ions. For Gd^{3+} and Yb^{3+}, this field was found to be, respectively, about 3.0×10^5 and 1.5×10^5 oe below 20°K, in satisfactory agreement with that derived from Pauthenet's data.

Disciplines

Physics

Comments

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Specific Heat of Some Rare Earth Iron Garnets and YIG at Low Temperatures*

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Heat capacity measurements of the iron garnets of Y, Gd, Er, Ho, and Yb between 1.4° and 20°K are presented. Below 5°K, the specific heat of YIG can be represented by the sum of a lattice term proportional to T^3 and the spin-wave contribution $2.15\times10^{-3}T^{\frac{3}{2}}$ joules/mole-deg. This last term agrees satisfactorily with that calculated from a spin-wave analysis, in which the exchange interaction coefficients were those derived from Pauthenet's magnetization data. The results of the magnetic specific heat of the rare earth ions could be interpreted in terms of a Weiss molecular field acting on these ions. For Gd^{3+} and Yb^{3+}, this field was found to be, respectively, about 3.0×10^5 and 1.5×10^5 oe below 20°K, in satisfactory agreement with that derived from Pauthenet's data.

IN the past few years, the rare earth iron garnets (formula $5\ Fe_2O_3\cdot3\ M_2O_3$, where M is a rare earth or yttrium) have been the subject of numerous experimental investigations both by resonance and by magnetization measurements. Pauthenet[1,2] was able to interpret the magnetization results of several garnets by applying the Weiss molecular field theory to the following ferrimagnetic model; between the ions Fe^{3+}, situated on the octahedral sites 16*a* and tetrahedral sites 24*d*, there exists a strong interaction which aligns their moments in an antiparallel way and determines the Curie point of these garnets. The ions M of the sites 24*c* are magnetized principally by the molecular field produced by the resultant magnetization of the Fe^{3+} ions. Their magnetization tends to compensate that of the Fe^{3+} sublattices. There is, in addition, a smaller interaction between the ions M. For zero external applied field, the effective molecular field H_{eff} acting on the ions M is the difference of the fields from these two interactions. From Pauthenet's data, H_{eff} is of the order 10^5 oe, which corresponds to energy level splittings Δ of about 10-30 cm^{-1}, and one would expect to observe these splittings by an anomaly in the specific heat at sufficiently low temperatures. We therefore measured the specific heat of the iron garnets of Gd, Yb, Er, and Ho between 1.4° and 20°K in zero external magnetic field. We also investigated very pure (99.99%) yttrium iron garnet (YIG), where only the interactions between the Fe^{3+} ions are present. As these interactions are very strong and correspond, below 50°K, to a Weiss molecular field of the order of 4×10^6 oe, no such anomaly in the specific heat was expected.

The experimental results are presented in Fig. 1. For yttrium iron garnet, our results between 1.4° and 4°K could be represented to a good approximation by the expression,

$$C=2.15\times10^{-3}T^{\frac{3}{2}}+0.36\times10^{-3}T^3\ \text{joules/mole.}$$

The specific heat is appreciably smaller than that found by Edmonds and Petersen,[3] possibly because of some rare earth or orthoferrite impurities in their sample.[4] As the specific heat of the other rare earth garnets is much larger, the amount of impurities here is very critical. The lattice contribution in T^3 agreed satisfactorily with that calculated from velocity of sound measurements by McSkimin,[5] taking $V_t=3.87\times10^5$ cm/sec, and $V_l=7.17\times10^5$ cm/sec as transverse and longitudinal velocities, respectively. The term in $T^{\frac{3}{2}}$, which is the magnetic contribution to the specific heat, will be discussed below.

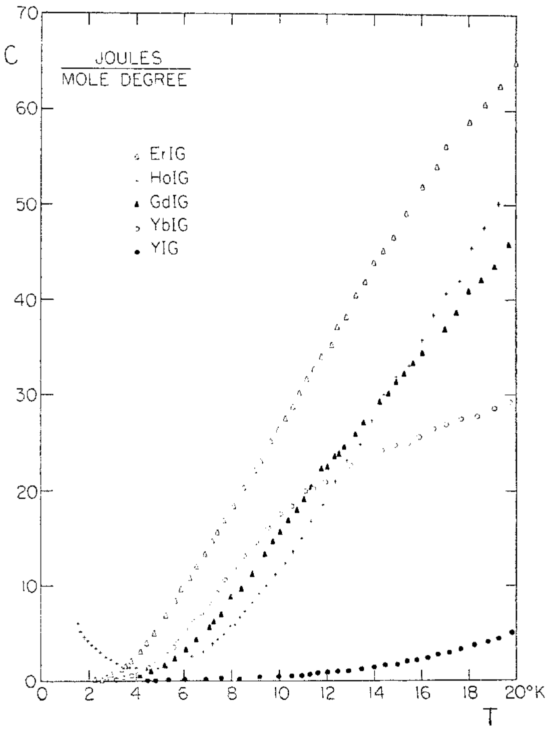

FIG. 1. Specific heat of the iron garnets of yttrium, gadolinium, holmium, erbium, and ytterbium. Most of the points below 5°K have been omitted.

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[1] R. Pauthenet, Ann. phys. **3**, 424 1958.

[2] R. Pauthenet, J. phys. radium **20**, 388 1959.

[3] D. T. Edmonds and R. G. Petersen, Phys. Rev. Letters **2**, p. 499 1959.

[4] Measurements on a YIG sample with about 10% orthoferrite impurity have shown a marked increase of the specific heat over that of the pure YIG sample.

[5] H. J. McSkimin (private communication).

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For the other rare earth iron garnets, we have assumed that, in first approximation, the specific heat can be written as $C_L+C_{RE}+C_{Fe}$, where C_L is the lattice contribution, C_{RE} is the magnetic specific heat of the rare earth "*c*" sublattice and C_{Fe} is that of the "*a*" and "*d*" iron sublattices. (C_L+C_{Fe}) was assumed to be about the same for all the rare earths except for YIG, where the lattice contribution should be somewhat smaller because of the smaller molecular weight. From our results, we have determined by successive approximations C_{RE} and (C_L+C_{Fe}) for gadolinium iron garnet, where the energy level disposition of Gd^{3+} is particularly simple. We then subtracted this value of (C_L+C_{Fe}) from the total specific heat of the other garnets, in order to find C_{RE} in each case.

For $Gd^{3+}(S=\frac{7}{2})$ which is in an S state, one finds that at these low temperatures each level is separated from the next one by the energy 28 cm^{-1} corresponding to an effective magnetic field $H_{eff}=3.00\times10^5$ oe. This is in satisfactory agreement with the value of 2.66×10^5 oe obtained from Pauthenet's results at 20°K.[1]

For Yb^{3+} $(J=\frac{7}{2},\ S=\frac{1}{2})$, the crystalline field theories by White and Andelin[6] and by Ayant and Thomas[7] show that at low temperatures only two levels are populated. The splitting between them was found to be about 24.4 cm^{-1}, corresponding to an effective field $H_{eff}=1.55\times10^5$ oe, if one assumes the magnetic moment of these levels to be $M_z=1.7$ Bohr magnetons.[7] This experimental value is to be compared to 10 cm^{-1}, as estimated by White and Andelin.[8]* The value of the splitting, deduced from Pauthenet's data, is about 26 cm^{-1} at 15°K.

For the rare earth ions Er^{3+} and Ho^{3+}, the energy level scheme is more involved,[6] and the interpretation of the results is being undertaken presently. The specific heat anomaly for Ho^{3+} near 1°K probably is due to the interaction between the electronic and the nuclear spins.

At low enough temperatures, when nearly all the magnetic ions are in their lowest energy state, the spin-wave theory should be followed rather than the molecular field approach. Calorimetric measurements can confirm this point because these theories give a very different temperature dependence of the specific heat (respectively, $T^{\frac{3}{2}}$ and $(\Delta/kT)^2e^{-\Delta/kT}$). Below 20°K, the spins in yttrium iron garnet almost have reached their zero-point alignment and a spin-wave calculation by one of us (A. B. H.) gives the energy-*versus*-*k* relation for the lowest acoustical mode as

$$\hbar\omega=1/16\{40J_{aa}-25J_{ad}+15J_{dd}\}a^2k^2,$$

where $J_{aa}-5.75$ cm^{-1}, $J_{ad}-24.2$ cm^{-1}, $J_{dd}-10.3$ cm^{-1} are the interaction coefficients derived by Pauthenet from his magnetization measurements,[1] $a=12$ A is the lattice constant for the garnet, and k is the usual wave vector. The spin-wave specific heat then is found to be $2.6\times10^{-3}T^{\frac{3}{2}}$ joules/mole in reasonable agreement with our experimental value. As one can see from this equation, small changes of the J's will affect drastically the energy spectrum and, hence, the spin-wave specific heat. The discrepancy with the experiment is, therefore, not astonishing. For Gd^{3+}, the specific heat below 3°K is systematically larger than that expected from the Weiss molecular field theory. This excess of specific heat probably is due partly to dipole-dipole interaction, although for an unmagnetized sample, the theoretical situation is unclear. The observed specific heat in this temperature range is about twice as large as predicted from a spin-wave analysis.

In conclusion, the specific heat results offer a valuable comparison with those obtained from magnetization measurements. At least for Yb^{3+} and Gd^{3+}, the Weiss molecular field theory is confirmed well by the experiments above about 3°K. Measurements on other garnets are in progress, and soon we will extend our experiments to lower and higher temperature ranges.

ACKNOWLEDGMENTS

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[6] R. L. White and J. P. Andelin, Phys. Rev. **115**, 1435 (1959).

[7] Y. Ayant and J. Thomas, Comptes rend. **248**, 387 1959.

[8] R. L. White (private communication).

* *Note added in proof.*—Recent optical measurements on YbIG by Wickersheim and White[9] showed the existence of inequivalent sites of the ytterbium ions with 2 different splittings of the two lowest levels, respectively 17.1 and 31.7 cm^{-1}. The specific heat calculated from these splittings still does not agree quantitatively with our data.

[9] K. A. Wickerman and R. L. White, Phys. Rev. Letters 4, 123 (1960).

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