about the validity of this interpretation, however, and the problem is to be pursued further.

(2) Bleaching an additively colored crystal narrows the resonance width from 45 gauss to 35 gauss peak-to-peak in the derivative and gives a center whose resonance saturates less readily than the "pure" F center resonance. The variation in saturation behavior cannot be interpreted in terms of a simple two-center model.

(3) The changes in the resonance behavior discussed here are not correlated with the presence of R, R', M, or N bands in the crystal. However, because of the small relative optical density of these bands, one cannot conclude from these results that the corresponding centers are nonmagnetic.

(4) The optical absorption associated with most of the resonance intensity is associated with the region near the F band throughout the changes induced by bleaching (or possibly with other bands of very low oscillator strength). This optical and magnetic absorption may be due either to F centers in altered surroundings or to a different defect produced by bleaching of the F center.

(5) It is suggested that the change in saturation behavior after long bleaching time is evidence of the clustering of the magnetic centers.

Note added in proof. Very similar results have been obtained independently by H. Gross and H. C. Wolf [Naturwissenschaften 8, 299 (1961)].

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Susceptibility of Gadolinium Iron Garnet below the Néel Point*

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Measurements of the spontaneous magnetization and the superposed paramagnetic susceptibility of gadolinium iron garnet have been repeated with a specimen of good stoichiometry, in the range of 2 to 300°K. It is concluded that the effective field which the Fe sublattices exert on the Gd ions causes paramagnetic saturation and consequently a reduction in susceptibility of the Gd sublattice. Good quantitative agreement is obtained by using molecular field theory without any adjustable constants, and in contrast to earlier interpretations, it is not found necessary to postulate appreciable antiferromagnetic interactions between the Gd ions.

INTRODUCTION

AMONG many other unusual properties, several of the rare-earth iron garnets are distinguished by relatively large variations of magnetic moment with applied field, at temperatures well below their Néel points. This effect was already observed in the early measurements of Pauthenet,[1] and ascribed by him as due to the weak nature of the exchange forces acting on the rare-earth ions, leaving them virtually free to orient in an applied magnetic field. This is consistent with the fact that the observed susceptibilities were all close to the free-ion values, varying with temperature approximately as $1/T$. In all cases small deviations from Curie's law were observed and these were interpreted by Pauthenet in terms of a weak antiferromagnetic interaction between the rare-earth ions themselves.

In this note we wish to draw attention to the fact that there is another, more important effect which will give rise to deviations from a simple Curie law, arising from ordinary paramagnetic saturation of the rare-earth moments in the exchange field produced by the iron sublattices.[2] The magnitude of this effect may be estimated from the observed value of the spontaneous magnetic moment, without the need of introducing any adjustable constants, and we have found that good agreement with experiment is obtained if it is assumed that the rare earth–rare earth interaction postulated by Pauthenet is in fact negligibly small. This conclusion is also in agreement with various susceptibility measurements in paramagnetic rare-earth gallium and rare-earth aluminum garnets.[3–6]

* Supported in part by the U. S. Air Force Cambridge Research Research Center, through European Office of the Research and Development Command.

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

[2] A brief discussion of this effect has previously been given by W. P. Wolf and J. H. Van Vleck, Phys. Rev. 118, 1490 (1960), and W. P. Wolf, *Reports on Progress in Physics* [The Physical Society, London (to be published)].

[3] J. Cohen and J. Ducloz, J. phys. radium 20, 402 (1959).

[4] R. Pauthenet, J. phys. radium 20, 388 (1959).

[5] M. Ball, G. Garton, M. J. M. Leask, and W. P. Wolf, *Pro-*

The simple effect which we consider will occur to some extent in all partially ordered systems, though often it may be masked by other complications. We have chosen gadolinium iron garnet (GdIG) for our analysis for two reasons. Firstly it is the only garnet containing magnetic rare earths (except EuIG) in which the effect of crystalline electric fields on the susceptibility may safely be neglected. Secondly, and basically for the same reason, GdIG has a very small magnetic anisotropy, so that it is possible to use polycrystalline samples for the magnetization measurements without introducing errors from magnetic "hardness." Using a polycrystalline sample is a considerable practical advantage if absolute measurements are required, since it is difficult to prepare large single crystals without some inclusions of flux or foreign phases.

MEASUREMENTS

The material used was a polycrystalline specimen prepared by Gilleo and Mitchell,[7] who took great care to achieve stoichiometry and uniformity. Magnetic moments were measured by using the pendulum magnetometer previously described,[8] in which the sensing elements were strain gauges deformed by motion of the pendulum, and the moment of the specimen was balanced by a current through a compensating coil.

As shown by Pauthenet, the magnetization per mole of $Gd_3Fe_5O_{12}$, σ, is of the form

$$\sigma=\sigma_s+\chi H_0,$$

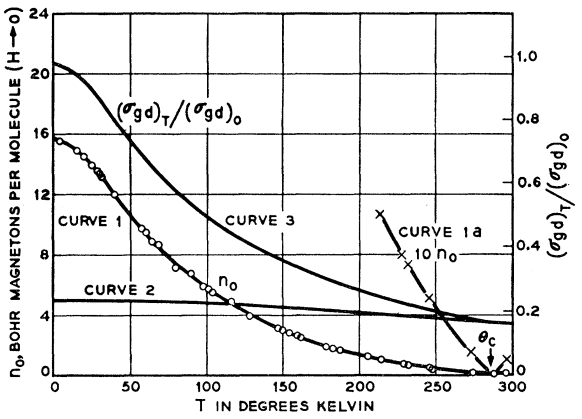

FIG. 1. Curve 1, spontaneous magnetization of $Gd_3Fe_5O_{12}$; curve 2, magnetization of $Y_3Fe_5O_{12}$, smoothed data by Pauthenet; curve 3, sum of curves 1 and 2, magnetization of rare-earth sublattice alone.

in which σ_s is the spontaneous moment arising from the vector sum of antiparallel Gd and Fe sublattice moments, and χ the paramagnetic susceptibility due almost entirely to the rare-earth ions. [In $Y_3Fe_5O_{12}$ (YIG), in which Y^{3+} is diamagnetic, χ is very small except in the vicinity of the Néel point, 560°K.[1]] In our experiments σ was determined at a given temperature by applying a field of $H_0=5000$ and adjusting the compensating current to balance the bridge containing the strain gauges. The field was then changed to 10 000 and the unbalance of the bridge noted. The unbalance was a direct measure of χ and was used also to derive the spontaneous magnetization σ_s from the measured σ. The measurements were repeated with the sample removed from the apparatus to take account of the slight moment of the pendulum.

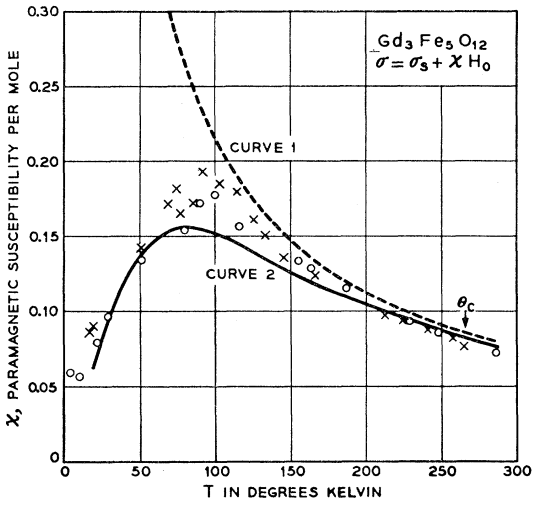

FIG. 2. Experimental paramagnetic susceptibilities of GdIG; crosses by Pauthenet, circles by present authors. Curve 1, Pauthenet's fitted law, $\chi=C/(T+8)$. Curve 2, our theory (see text).

The spontaneous moments are given in curve 1 of Fig. 1, in Bohr units per molecule n_0. Extrapolating to $T=0$ gives a value of n_0 about 1% below the theoretical 16,[1] a much smaller discrepancy than that observed by Pauthenet.[1] We take this as evidence that our sample was more nearly stoichiometric, or chemically purer. However at the lowest temperatures even in our sample there was some evidence of magnetic "hardness" and this almost certainly accounts for the low value of n_0. If we extrapolate "upwards," i.e., to $H_0=\infty$ assuming $\chi=0$ we find $n_0=16.0$. The compensation point θ_c shown in curve 1, and more plainly in curve 1a with expanded ordinates, occurs at 286°K, in good agreement with the temperature observed by Geschwind and Walker[9] in a single-crystal sample.

ceedings of the Seventh International Conference on Low-Temperature Physics (University of Toronto Press, Toronto, Canada, 1961), p. 128.

[6] M. Ball, G. Garton, M. J. M. Leask, D. Ryan, and W. P. Wolf, J. Appl. Phys. **32**, 267S (1961).

[7] As described by Gilleo and Geller, Phys. Rev. **110**, 73 (1958).

[8] R. M. Bozorth and D. E. Walsh, J. Phys. Chem. Solids **5**, 299 (1958).

[9] S. Geschwind and L. R. Walker, J. Appl. Phys. **30**, 163S (1959).

Values of χ as determined by Pauthenet (crosses) and ourselves (circles) are given in Fig. 2.

DISCUSSION

The origin of the intrinsic susceptibility of a partially saturated sublattice is illustrated in Fig. 3. In terms of the usual molecular field theory we may think of the sublattice magnetization σ_s at a temperature T as corresponding to a point on the Brillouin function appropriate to the angular momentum J of the magnetic ions:

$$(\sigma_s)_T/(\sigma_s)_{T=0}=B_J(x), \tag{1}$$

where

$$x=g\mu_B H_e/kT, \quad (\sigma_s)_{T=0}=NgJ\mu_B,$$

and H_e is the effective molecular field.

To estimate the change of moment $\delta\sigma$ induced by an applied magnetic field H_0, we must distinguish between two different cases: (i) in which H_e arises from interactions with other magnetic moments which are also

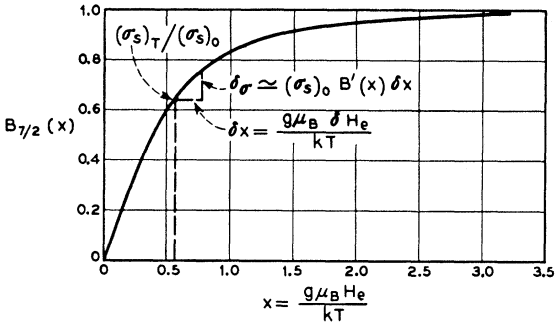

FIG. 3. Brillouin function for Gd^{3+}, and slope $\delta\sigma/\delta x$ from which differential susceptibility is derived.

not fully saturated, and (ii) in which H_e is due to some other sublattices which are already nearly saturated. In the first case the molecular field will itself change as the various sublattice magnetizations are changed by the applied field, while in the second case the interaction field remains constant. The total effective field is then simply increased (or decreased) by H_0. We shall only consider here the second case, since in the rare-earth garnets by far the biggest effects arise from the rare earth ions and their interactions with the Fe sublattices, which even at 300°K are over 70% saturated. In the region of the Néel point the other effect will also become important, and an additional contribution to the susceptibility is observed.

If, as in our case, $H_0 \ll H_e$ ($\sim 10^4$ gauss against 2×10^5 gauss), the change of magnetic moment is given by

$$\delta\sigma=(\sigma_s)_{T=0}B'(x)\delta x$$
$$=Ng^2\mu_B{}^2JB'(x)H_0/kT.$$

This corresponds to a differential susceptibility

$$\chi=\chi_c[3B'(x)/(J+1)]=\chi_c f(x), \tag{2}$$

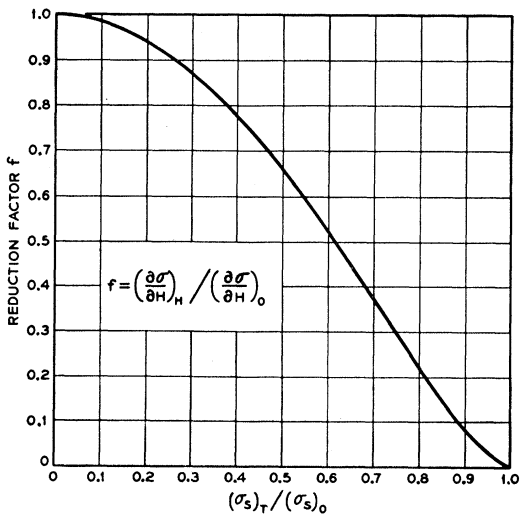

FIG. 4. Reduction factor for calculating susceptibility for various values of magnetization (see text).

where χ_c is the Curie law value. When $kT \gg g\mu_B H_0$, $B'(x)\to\frac{1}{3}(J+1)$ and $\chi\to\chi_c$, provided H_e remains constant as discussed above. At low temperatures σ_s tends towards its saturation value and $\chi\to 0$. The reduction factor f can be calculated explicitly in terms of H_e and T, but it is simpler to eliminate the parameter x between Eqs. (1) and (2) and obtain a direct relationship between the relative magnetization $(\sigma_s)_T/(\sigma_s)_{T=0}$ and the factor f. For the case of $J=\frac{7}{2}$ appropriate to Gd^{3+} this may be calculated by direct numerical differentiation of the corresponding Brillouin function given by Giauque *et al.*[10] The result is shown in Fig. 4. It may be seen that even when the sublattice is only 20% saturated the effective susceptibility is reduced by some 6% from its Curie law value.

For the case of GdIG only the Gd^{3+} sublattice contributes appreciably to the susceptibility at temperatures below 300°K, and the relative magnetization of this sublattice may be found very simply following the discussion by Pauthenet. He showed that the spontaneous moment σ_s arises from the vector sum of rare earth and Fe^{3+} moments, the iron sublattices behaving much as in yttrium iron garnet, in which the "rare earth" is diamagnetic. Thus at any temperature the moment of the rare-earth sublattice alone may be found by *adding* the observed value of σ_s to the corresponding value of σ_s in YIG. The result of this leads to curve 3 in Fig. 1, where we have used Pauthenet's smoothed values for YIG, and our measurements for GdIG. Using this curve together with Fig. 4 leads to the calculated curve 2 shown in Fig. 2.

The relatively crude procedure we have followed is open to several detailed objections. (i) It is clear that the molecular field method is not an ideal approximation

[10] W. F. Giauque, J. W. Stout, C. J. Egan, and C. W. Clark, J. Am. Chem. Soc. **63**, 403 (1941).

in a region where the sublattices approach saturation. For the Gd sublattice this does not happen until T is appreciably below 100°K, but the Fe sublattices are over 70% saturated even at 300°K. This may explain why there is practically no contribution at all for the Fe ions, even though our molecular field treatment would predict a small but noticeable effect. (ii) Even in our best sample the theoretical saturation moment was not attained in the helium range. This makes the calculation of relative magnetization at higher temperatures somewhat uncertain, but the effect on the susceptibility is generally negligible. (iii) The discrepancy between the theoretical and experimental $(\sigma_s)_{T=0}$ value may have been due to small impurities of other rare earths, which might have the effect of increasing the magnetic anisotropy at the lowest temperatures (as in the experiments of Dillon and Nielsen[11]). Too large a magnetic hardness in the polycrystalline sample would lead to uncertainty in both the extrapolated values of σ_s and the deduced values of susceptibility. (iv) In estimating the Gd sublattice magnetization we assumed, with Pauthenet, that there is no "back action" by the Gd ions on the Fe sublattices. This cannot be true exactly, and in the regions in which σ_s varies rapidly with temperature the effect of this on the calculated susceptibility may be appreciable. Inclusion of this effect might very well account for the systematic discrepancy in the region of 100°K.

[11] J. F. Dillon and J. W. Nielsen, Phys. Rev. **120**, 105 (1960).

CONCLUSION

We have made measurements of the intrinsic susceptibility of a sample of polycrystalline gadolinium iron garnet between 2° and 300°K. The results are generally in reasonable agreement with earlier measurements by Pauthenet on a sample presumably less nearly stoichiometric. At high temperatures the susceptibility approximately follows Curie's law for free Gd^{3+} ions, but there are significant deviations which become very large at low temperatures. It is shown that these deviations are quantitatively consistent with the effects to be expected from paramagnetic saturation. Interactions between the rare-earth ions appear to be much less important than previously supposed.

ACKNOWLEDGMENTS

We wish to thank Professor J. H. Van Vleck for helpful and stimulating discussions, and Mr. D. D. Davis for assistance with the measurements. We are indebted to Dr. M. A. Gilleo for lending us the specimen of $Gd_3Fe_5O_{12}$.

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Bloch Wall Excitation. Application to Nuclear Resonance in a Bloch Wall*

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The excitation spectrum of an assembly of electronic spins in a Bloch wall structure is studied, assuming a uniaxial anisotropy. The spectrum may be divided into two branches; one is a specific wall excitation and does not spread outside the wall, the other one is similar to the spin-wave excitation spectrum in a uniform ferromagnet. These calculations are used to study the properties of the nuclear magnetic resonances in a Bloch wall. The relaxation times are evaluated, taking into account the damping of the motion of the electronic spins and are compared with experimental values. The spin-spin coupling and the variation of the magnetization across the wall is also estimated.

I. INTRODUCTION

NUCLEAR magnetic resonances in several ferromagnetic substances have been observed recently.[1] The large amplitude of the signals is explained by assuming that the observed nuclei are within the Bloch wall.

The relaxation times T_1 and T_2 have also been measured using spin-echo techniques.[2] The theoretical estimation of relaxation times and spin-spin couplings requires a detailed knowledge of the motion of the electronic spins within the Bloch wall.

A complete solution for this problem may be found by assuming a uniaxial anisotropy and using a simplified demagnetizing field, then the relaxation times are estimated, taking into account the damping of the wall motion. An indirect interaction is also estimated.

* This research has been supported by the National Science Foundation.

† On leave from Centre d'Etudes Nucleaires de Saclay, Gif-sur-Yvette, Seine et Oise, France.

[1] A. C. Gossard and A. M. Portis, J. Appl. Phys. **31**, 205S (1960); A. C. Gossard, A. M. Portis, and W. J. Sandle, J. Phys. Chem. Solids, **17**, 341 (1961); C. Robert and J. M. Winter, Compt. rend. **250**, 3831 (1960); J. Hervé and P. Veillet, Compt. rend. **252**, 99 (1961).

[2] M. Weger, E. L. Hahn, and A. M. Portis, J. Appl. Phys. **32**, 124S (1961); C. Robert and J. M. Winter, Nuovo cimento, (to be published).