Brose[14] from the Stark effect studies which perforce covered a different range of discharge conditions. It has further been shown that such a general linear distribution of field, if analyzed in terms of a proper variation of ion drift velocity with E/p and using appropriate values of the second Townsend coefficient, enables a calculation of α, the ionization per unit length along the axis. While accuracy of the data at the cathode do not permit a positive statement, the nature of the inherent errors strongly suggest that the α will be low near the cathode and rise as the negative glow is approached. This yields the type of ionization across the dark space indicated by Morton[4] and Johnson[5] to be probable in this E/p and p range.

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[14] E. Brose, Ann. phys. **58**, 731 (1919).

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Antiferromagnetic Resonance above the Curie Temperature*

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The role of short-range order in effecting the disappearance of absorption as the Curie temperature is approached is discussed. The prediction of a simple theory of the effect agrees very well with experimental results from MnO. Consideration is also given to the relation of resonance experiments and studies made by neutron diffraction.

I.

THE first overt experimental study of magnetic resonance in an antiferromagnetic substance was made on Cr_2O_3 and gave the surprising result that as the material was cooled below its Curie temperature T_c the absorption suddenly and almost completely disappeared.[1] A curve showing the temperature dependence of the magnitude of the absorption at resonance (the "peak height" h) is a convenient way of partially presenting the data. A series of such curves giving h as a function of T/T_c for several materials is given in Fig. 1 which is due to Maxwell and McGuire.[2] The extinction of the resonance near T_c is a general feature of all these curves. The decline is less abrupt, however, for the compounds MnO, MnS, and MnTe, and the relative temperature range involved becomes smaller as one goes down the column of the periodic table containing the elements of negative valence. Because of the theory reviewed below and the difficulty of preparing the samples, it is felt that the residual absorption at T_c is due to impurities. Therefore, this residuum will have been subtracted from the data discussed below, and additional corrections will have been made by dividing each reading by a factor which approximately accounts for the temperature variation of the properties of the apparatus. These questions are discussed in more detail by Maxwell and McGuire.[2]

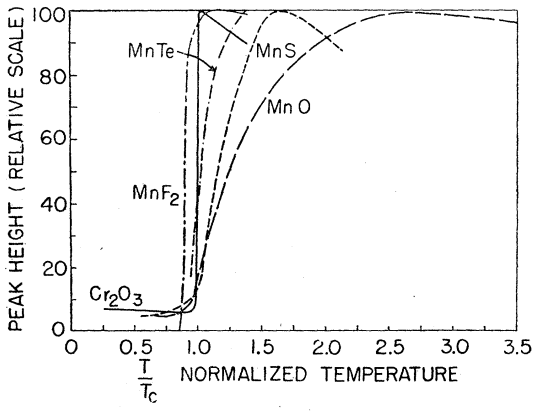

FIG. 1. Peak height of resonance absorption *vs* temperature for several antiferromagnetic materials.

Present views on the vanishing of the absorption are due to Nagamiya and Kittel[3] and ascribe it to a combined effect of anisotropy and exchange. Because of the increasing anisotropy as T falls below T_c, the actual resonance frequency will no longer coincide with the applied microwave frequency and, in fact, generally

* Supported in part by the ONR.

[1] Trounson, Bleil, Wangsness, and Maxwell, Phys. Rev. **79**, 542 (1950).

[2] All data except MnF_2 from L. R. Maxwell and T. R. McGuire (to be published in Revs. Modern Phys.). MnF_2 results due to C. A. Hutchison, Jr. (private communication to L. R. Maxwell).

[3] T. Nagamiya, Prog. Theoret. Phys. **6**, 350 (1951); C. Kittel, Phys. Rev. **82**, 565 (1951). See also: F. Keffer and C. Kittel, Phys. Rev. **85**, 329 (1952); R. K. Wangsness, Phys. Rev. **86**, 146 (1952); J. Ubbink, Phys. Rev. **86**, 567 (1952).

increases rapidly to the millimeter wave region. The basis of the derivation of the resonance conditions is the division of the magnetic atoms into sublattices whose resultant magnetizations are then discussed by using the classical equations of motion. Although this treatment is commonly used in the theories of antiferromagnetism, its dynamical validity is questionable near and above the Curie temperature since a meaningful division into such sublattices requires the existence of appreciable long-range order in the magnetic lattice.

For most materials of interest, T_c is so large that the difficulty of working in the required frequency region has prevented the observation of the resonance predicted for the antiferromagnetic state, although an actual antiferromagnetic resonance has apparently been observed in $CuCl_2 \cdot 2H_2O$ for which T_c is only a few degrees K.[4] Thus, it is desirable that we be able to learn as much as possible from those experiments which can be conveniently performed and which do yield results, namely, those done in the paramagnetic region above T_c.

The peak height for a paramagnetic material can be expected to be proportional to the total resultant moment and hence to the susceptibility χ.[5] That this is not the only effect which must be considered is most clearly shown by the contrast between the curve for MnO in Fig. 1 and the steady increase of χ to its maximum as T decreases to T_c.[6]

A clue to another feature which may affect the dependence of h on T is provided by neutron diffraction results. At room temperature (which is about 2.5 T_c) a residual coherence was observed in the pattern from MnO which indicated the existence of short-range order.[7] It is noteworthy that the decline in h of MnO begins at about this point and, at the same time, is the most gradual of any of those shown in Fig. 1. Accordingly, it seems worth while to attempt to assess the influence of short-range magnetic order upon the resonance absorption.

II.

The concept of the average short-range order, or order of neighbors, is familiar in the theory of alloys and gives a measure of the effectiveness of interactions in producing a nonrandom distribution of neighbors about a central atom.[8] For AB type alloys, the short-range order parameter σ is defined as the fractional excess of unlike over like nearest neighbor pairs. Above the Curie temperature where long-range order is absent, σ is still different from zero and increases with decreasing temperature until a critical value σ_c is reached. Below this temperature, there is a marked change in the dependence of σ upon T, with a concomitant appearance of long-range order.

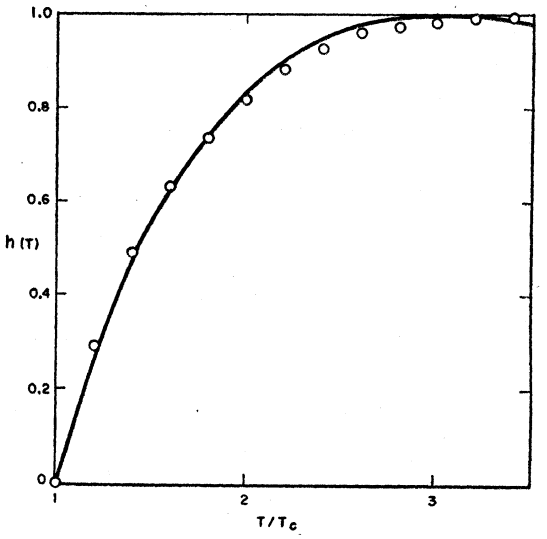

FIG. 2. Peak height *vs* temperature for MnO. Solid curve was calculated from Eq. (1). Open circles are experimental values.

The discussion of order in binary alloys is similar to that appropriate to the Ising model which, however, does not take into account the vectorial character of angular momentum which is requisite for a dynamical theory of resonance. Moreover, it is not likely that antiferromagnetic ordering can be adequately described by a single parameter; for example, one may need to consider the ordering within the shells of more remote neighbors. We shall, however, assume a description in terms of σ, defined as the average fractional excess of antiparallel over parallel nearest neighbor pairs, to be sufficiently accurate for our present purposes.

One simple possibility which suggests itself is that at temperatures not too far above T_c, for which the average short-range order is less than the critical value, varying local conditions may result in actual values of the short-range order in various separate regions which are greater than σ_c. Correspondingly, one would expect the material in these regions to be essentially antiferromagnetic, although the sample as a whole could not be said to be so. The disparity between the resonance frequencies of these regions and the applied frequency would then be so great, as discussed above, that they would effectively be excluded from the material for which the normal resonance could be obtained. The result is that the total absorption above T_c would be less than that anticipated from the usual arguments; this agrees qualitatively with Fig. 1.

We already expect h to be proportional to the susceptibility $\chi(T)$. Recognition of the possible effect of ordering now leads us to expect h to be further proportional to a function of σ, $w(\sigma)$, such that $w(\sigma_c)=0$ and $w(0)=1$. A definite dependence of h upon σ can be obtained by expanding $w(\sigma)$ about σ_c; in first approxi-

[4] Poulis, van den Handel, Ubbink, Poulis, and Gorter, Phys. Rev. **82**, 552 (1951).
[5] M. H. L. Pryce and K. W. H. Stevens, Proc. Phys. Soc. (London) **A63**, 36 (1950).
[6] H. Bizette, Ann. phys. **1**, 295 (1946).
[7] Shull, Strauser, and Wollan, Phys. Rev. **83**, 333 (1951).
[8] F. C. Nix and W. Shockley, Revs. Modern Phys. **10**, 1 (1938).

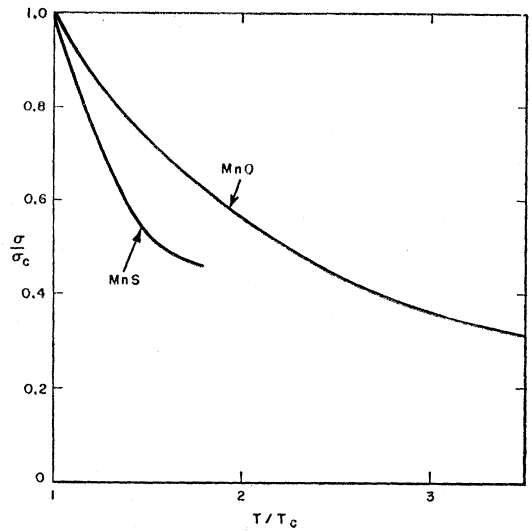

FIG. 3. Relative short-range order *vs* temperature. MnO curve is that used for Fig. 2. MnS curve from resonance and susceptibility measurements.

mation, this results in h being proportional to $(\sigma_c-\sigma)$. Combining these considerations, we can write an expression for h in the following convenient form

$$h(T)=P[1-(\sigma/\sigma_c)][\chi(T)/\chi(T_c)]. \quad (1)$$

For now, we shall treat P as a normalization constant for the purpose of comparison with experiment; later, we shall briefly consider the factors one can expect to be involved in P.

The quantities $\chi(T)/\chi(T_c)$ and $\sigma(T)$ have been calculated by Li[9] for a simple cubic lattice of moments of spin $\frac{1}{2}$. Keeping in mind these limitations on their applicability, we can use his values to evaluate (1); the result is shown together with the experimental data from MnO in Fig. 2. The agreement between the two curves in this case is surprisingly good. It should be noted that no experimental determination of theoretical parameters has been made, nor has the curve given by (1) been otherwise adjusted except for normalization.

III.

It seems safe to conclude from Fig. 2 that the effects of antiferromagnetic interactions upon the resonance are noticeable at temperatures for which the material would ordinarily be described as paramagnetic. The temperature range over which Eq. (1) is useful is also much greater than might be anticipated from its derivation.

There are several possible reasons for the slight discrepancies in the two curves other than those discussed by Li concerning the validity of his calculations of σ and χ. Li's values of σ were obtained for zero applied field, and if an external field were present, σ could be expected to be decreased because of the tendency toward greater parallelism of the spins. This would in turn increase the values of h given by (1); for the initial regions, this is the correct direction to improve the agreement between the curves. If the residual absorption is of paramagnetic origin, its value would decrease at higher temperatures rather than remaining constant as was assumed in treating the experimental data. This would increase the agreement at higher temperatures by raising the experimental values. Although P was treated as a constant, a correct expression for it should involve the relaxation times of the system. These in turn will depend upon the temperature due to the effects of thermal agitation, and their value may well be influenced by the existing average short-range order. Other cases may require an expansion of w beyond the first two terms, resulting in a direct dependence of P upon σ.

If we assume the general validity of (1), it is then possible to invert our procedure and use the results of resonance experiments and susceptibility measurements to learn something about the short-range order in other materials. We have thus the possibility of a fruitful relation between the work on neutron diffraction and that on antiferromagnetic resonance; for example, the MnS curve in Fig. 1 would then indicate that short-range ordering effects are not as pronounced as in MnO for comparable values of T/T_c. Equation (1), unfortunately, enables us to obtain only the ratio σ/σ_c rather than the absolute value of σ; this ratio has been calculated for MnS by using the experimental values[6] of χ and is shown in Fig. 3 with the theoretical values which were previously used for MnO. These curves confirm the qualitative expectations of the relative values of the short-range order; the experimental data do not suffice to extend the curve to larger values of T/T_c in order to see if the value of the ratio for MnS does become greater than that ascribed to MnO at sufficiently great temperatures as the trend of the curve suggests.

I wish to thank Dr. L. R. Maxwell, Dr. T. R. McGuire, and Dr. J. S. Smart for many helpful discussions of this work.

[9] Y. Y. Li, Phys. Rev. 84, 721 (1951).