our hypothesis about the constancy of μ is correct, there must be at least a small contribution from a term of the form (7) at any temperature above T_c, showing that the actual density has a more complicated structure than Suzuki proposed.

In summary, numerical investigations of the density of zeros on the Lee-Yang circle for a two-dimensional and a three-dimensional Ising model indicate few surprises for $T \leq T_c$, but suggest that above T_c the behavior near θ_G is dominated by a singularity with an exponent μ whose connection with other "critical" exponents is at present not clear. Also, $g(\theta)$ is not a monotone function of θ for T substantially greater than T_c, unlike the situation in the one-dimensional Ising and mean-field models. Finally, for the two-dimensional lattice there is strong evidence of some sort of singular behavior at $\theta = \pi$ for $T > T_c$.

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[8]For z near 1, M should vary as $(1-z)^{1/\delta}$, and the behavior of u can be deduced from Eq. (2).

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Character of Excitations in Substitutionally Disordered Antiferromagnets

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Magnetic excitations in several single crystals of the systems K(Co, Mn)F_3 and (Co, Mn)F_2 have been studied by neutron inelastic scattering. It is found that two branches of well-defined magnetic excitations, both of which exhibit propagating character, occur at certain compositions. The dispersion relation for two branches of propagating excitations cannot be adequately described by current theories, but an extension of Anderson's criterion for localization in disordered alloys gives approximate agreement with the observed character.

The excitations in randomly disordered systems may have either localized or nonlocalized character. Although Anderson[1] first investigated the conditions for localization in 1958, there is still considerable disagreement[2] about the detailed behavior. The theoretical work has dealt mainly with electron states; but, in principle, the problem is the same for a wide class of excitations and in particular for spin waves, which have the advantage that they can be studied directly by neutron inelastic scattering. We report here measurements on disordered antiferromagnets that for the first time show the existence of two branches of propagating excitations in a disordered quasibinary system. Current Green-function theories for antiferromagnets cannot describe the results, but the character of the excitations is reasonably well described by a localization criterion of the Anderson form.

The measurements were carried out by means of neutron inelastic scattering using a triple-axis crystal spectrometer controlled so that the wave-

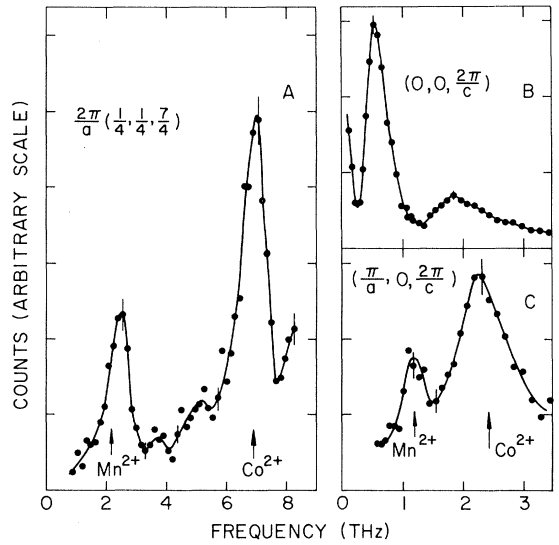

FIG. 1. Typical scattered-neutron distributions for (a) $KCo_{0.71}Mn_{0.29}F_3$ and (b),(c) $Co_{0.7}Mn_{0.3}F_2$. Each distribution is labeled with the value of the wave-vector transfer $\vec{Q}$ at which the measurement was carried out. Solid lines are simply a guide to the eye. Statistical counting errors are shown. Arrows indicate predictions of molecular-field theory (see text).

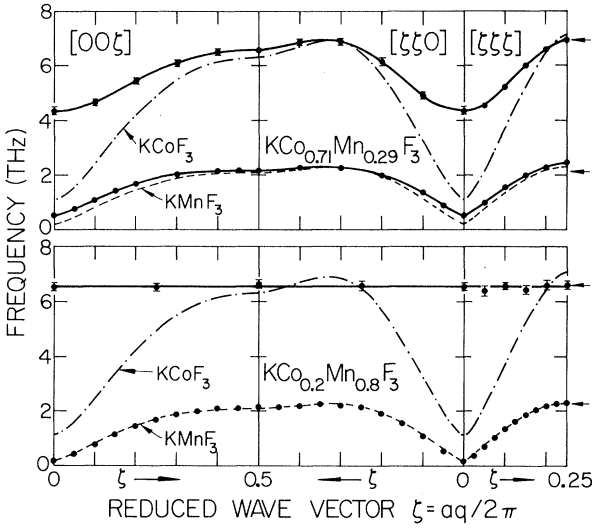

FIG. 2. The dispersion relations for magnetic excitations propagating along the three major symmetry directions in $KCo_{0.2}Mn_{0.8}F_3$, $KCo_{0.71}Mn_{0.29}F_3$, and the two pure materials. Solid curves are simply a guide to the eye. Arrows at the right indicate predictions of molecular-field theory.

vector transfer $\vec{Q}$ was held constant throughout each scan. Typical scattered-neutron distributions, each of which exhibits two well-defined inelastic peaks corresponding to magnetic excitations, are shown in Fig. 1. All peaks except the higher-frequency peaks for $Co_{0.7}Mn_{0.3}F_2$ have widths that are essentially determined by the experimental resolution. The latter peaks are much wider than the experimental resolution and often exhibit asymmetric line shapes with tails extending to higher frequencies. In addition, the total variation with wave vector of their assigned frequencies is only about half their average width [full width at half-maximum (FWHM)], and their integrated intensities only vary by a factor of ≈ 3 and are greatest near zone boundaries. In contrast, the intensities of the lower-frequency peaks vary by a factor of ≈ 20 and are greatest at magnetic reciprocal-lattice points.

The results for $KCo_{0.71}Mn_{0.29}F_3$ and $KCo_{0.2}Mn_{0.8}F_3$ are summarized in Fig. 2. For comparison, we also show the dispersion curves[3,4] for $KCoF_3$ and $KMnF_3$. When 20% of the manganese ions in $KMnF_3$ are replaced by cobalt ions, the perturbation of the "host" modes is small and a flat branch of excitations localized on the cobalt "impurities" appears at a frequency of 6.55 ± 0.15 THz. (For details see Svensson *et al.*[5]) However, when 29% of the cobalt ions in $KCoF_3$ are replaced by manganese ions, the perturbation of the "host" modes is extremely large (the zone-center frequency increases from 1.12 ± 0.06 to 4.36 ± 0.15 THz) and a branch of excitations having a marked frequency variation appears at lower frequencies. (For this material, low-concentration Gree-function theory predicts a flat lower branch at a frequency of 0.8 THz corresponding to excitations localized on the manganese ions.) Since both branches of excitations are observed to have intrinsic widths (inverse lifetimes) that are small compared with their frequency variation, both correspond to propagating excitations. The behavior is obviously quite different near opposite ends of the K(Co, Mn)F_3 system. $KCo_{0.2}Mn_{0.8}F_3$ exhibits the behavior (local impurity mode plus weakly perturbed band modes) typical of low impurity concentrations, but for $KCo_{0.71}Mn_{0.29}F_3$ the band modes are greatly perturbed and the "impurity" modes are propagating excitations.

The results for $Co_{0.7}Mn_{0.3}F_2$ are summarized in Fig. 3. For comparison we also show the dispersion curves[6-8] for CoF_2 and MnF_2. Note that both branches for $Co_{0.7}Mn_{0.3}F_2$ differ markedly from those for the pure materials. The lower branch corresponds to propagating excitations since the intrinsic widths of the excitations are small relative to the total frequency variation. The frequency variation for the upper branch is only about

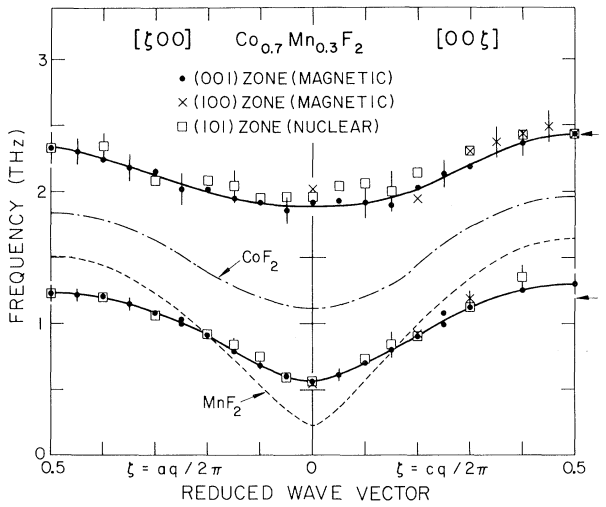

FIG. 3. The dispersion relations for magnetic excitations propagating along the a and c directions in $Co_{0.7}Mn_{0.3}F_2$, CoF_2, and MnF_2. Different symbols refer to measurements in Brillouin zones centered on different reciprocal-lattice points as indicated. Typical errors are shown. Solid curves are a guide to the eye for the results in (001) zone. Arrows are predictions of molecular-field theory.

half the width (FWHM) of the observed peaks (see Fig. 1). Because of this and the unusual intensity variation mentioned above, it is possible that the higher-frequency peaks in $Co_{0.7}Mn_{0.3}F_2$ correspond to scattering by several modes of excitations, some or all of which might be local modes having rather similar frequencies but different structure factors. Alternatively, there could simply be a single mode of excitation with appreciable damping and an unusual structure factor. The exact nature of the higher-frequency excitations in $Co_{0.7}Mn_{0.3}F_2$ is not unambiguously determined by the measurements. Possibly they correspond to the random structure of discrete poles recently discussed by Anderson.[2]

In contrast to the results for $Co_{0.7}Mn_{0.3}F_2$, earlier measurements[9] on $Co_{0.05}Mn_{0.95}F_2$ showed a lower branch negligibly different from that for pure MnF_2, and a flat upper branch at 3.57 ± 0.05 THz corresponding to excitations localized on the cobalt "impurities."

No detailed theory for the magnetic excitations in substitutionally disordered antiferromagnets containing large concentrations of impurities has yet been developed. However, some progress in understanding the above results may be made by applying molecular-field theory and low-concentration Green-function theory. Using molecular-field theory together with the known exchange constants and wave functions for the pure materials,[3,4,6,7] and taking $J_{Mn\text{-}Co}$ from the experimental local-mode frequencies,[5,9] the energies required to create a spin deviation on Mn^{2+} and Co^{2+} ions in the materials of interest can readily be calculated. The resulting Ising frequencies of course depend on the composition of the cluster of ions surrounding the reference ion. When the known probabilities of occurrence of the different clusters are plotted against the corresponding Ising frequencies, one obtains two well-separated peaks for each material. The lower-frequency peak corresponds to Mn^{2+} ions and the higher-frequency peak to Co^{2+} ions. The centers of gravity of these calculated peaks are indicated by arrows in Figs. 1–3. The agreement with the observed zone-boundary frequencies is reasonably good in view of the approximation involved and suggests that, at least at these wave vectors, the excitations of lower (higher) frequency are associated mainly with manganese (cobalt) ions. (In carrying out the calculations, we have allowed approximately for the change in the exchange mixing of the cobalt levels with cluster composition.)

A further result of the calculations is that the Ising frequencies are quite insensitive to cluster composition for the $K(Co, Mn)F_3$ system, but are critically dependent on cluster composition for the $(Co, Mn)F_2$ system. In particular, for $Co_{0.7}Mn_{0.3}F_2$, where several compositions occur with comparable probability, the calculated widths (FWHM) are 0.22 and 0.72 THz for the peaks of lower and higher frequency, respectively. When allowance is made for experimental resolution, these values are in good agreement with the observed widths at the zone boundary [Fig. 1(c)]. Molecular-field theory thus gives a simple explanation of the large width of the higher-frequency peaks for $Co_{0.7}Mn_{0.3}F_2$.

Green-function theories have the advantage that they give results for all wave vectors, but, unfortunately, they are strictly applicable only at very low concentrations of impurities where they give excellent descriptions.[8,10] This type of theory cannot give rise to two branches of propagating excitations. This has been confirmed by detailed calculations for the materials studied. As expected, the theory adequately describes the results when a localized excitation is present as in $Co_{0.05}Mn_{0.95}F_2$ and $KCo_{0.2}Mn_{0.8}F_3$, but for $KCo_{0.71}Mn_{0.29}F_3$ and $Co_{0.7}Mn_{0.3}F_2$ it fails completely to describe the propagating behavior of the lower branch.

In the absence of a detailed theory, it is inter-

esting to consider if one can even describe the character (localized or propagating) of the excitations in our specimens. Although the general method of Anderson[1,2] should in principle be applicable to any disordered system, it is not clear how to extend the formalism (which has been developed mainly for electron bands in disordered alloys) to spin waves in an antiferromagnet. We suggest that a plausible, although somewhat oversimplified, form of the localization condition is

$$\chi(\nu)=\frac{2|\nu-\nu_1|^c|\nu-\nu_2|^{1-c}}{cB_1+(1-c)B_2}, \qquad (1)$$

where c and $1-c$ are the respective concentrations of magnetic ions of types 1 and 2, and ν_1 and ν_2 are the centers of the spin-wave bands (of widths B_1 and B_2) in the pure materials. The criterion for localization at frequency ν is $\chi(\nu)>1$. Apart from spurious behavior near ν_1 or ν_2, Eq. (1) correctly predicts that, as $c\to 0$ or $c\to 1$, excitations will be localized if outside the "host" band.

For $Co_{0.05}Mn_{0.95}F_2$ and $KCo_{0.2}Mn_{0.8}F_3$, Eq. (1) predicts that the lower and upper branches should correspond, respectively, to propagating and localized excitations as observed (see Fig. 2 and Refs. 5 and 9). For $KCo_{0.71}Mn_{0.29}F_3$, it predicts nonlocalized excitations for $0.4\lesssim\nu\lesssim 6.0$ THz, and localized excitations otherwise. This agrees with the observed behavior (Fig. 2) except for part of the upper branch. For $Co_{0.7}Mn_{0.3}F_2$, Eq. (1) predicts propagating character when $0.75\lesssim\nu\lesssim 1.9$. This is correct for the lower branch except close to the zone center. The prediction that the upper branch corresponds to localized excitations cannot be checked since the character is not unambiguously determined by the measurements. From the general agreement between the calculated and observed character we conclude that a localization criterion similar to that proposed exists for magnetic excitations in substitutionally disordered antiferromagnets. These results lend support to the ideas of Anderson on localization in disordered systems, but these ideas can only be further confirmed by a rigorous extension to antiferromagnets of the Anderson type of theory.

The results presented in this Letter have demonstrated the occurrence of two branches of propagating magnetic excitations in disordered antiferromagnets. This behavior cannot be described even qualitatively by current Green-function theory, and emphasizes the need for a different type of theory for antiferromagnets containing large concentrations of impurities.

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