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Magnon Localization in Antiferromagnets*

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A criterion is developed for the Anderson localization of the one-magnon excitation in a substitutionally disordered two-sublayer Heisenberg antiferromagnet. Assuming nearest-neighbor exchange interactions, application is made to $K(\text{Mn}_{1-c}\text{Co}_c)\text{F}_3$ near zero temperature. Taking, for simplicity, $J^{\text{Mn-Co}} = (J^{\text{Mn}}J^{\text{Co}})^{1/2}$, a critical condition for localization is found for all impurity concentrations, yielding good agreement with recent experiments.

Recently, Buyers *et al.*¹ presented evidence for localized spin excitation modes in the substitutionally disordered antiferromagnets, $K(\text{Mn}_{1-c}\text{Co}_c)\text{F}_3$ and $(\text{Mn}_{1-c}\text{Co}_c)\text{F}_2$. They presented an empirical formula for the relation between the critical energy for localization, and the impurity concentration. They remarked that, "Although the general method of Anderson should in principle be applicable to any disordered system, it is not clear how to extend the formalism ... to spin waves in an antiferromagnet." This Letter extends Anderson's formalism² to that problem. We find a different form for the localization criterion than that written down by Buyers *et al.* Our result is in agreement with their data.

We begin with a Hamiltonian appropriate to a substitutionally disordered two-layer sublattice antiferromagnet:

$$H = 2 \sum_{mn} J_{mn} \vec{S}_m \cdot \vec{S}_n - 2\mu_B H_A \sum_m S_{mz} + 2\mu_B H_B \sum_n S_{nz}. \quad (1)$$

Here, m and n represent the lattice vectors on sublayers A and B , respectively; and J_{mn} , \vec{S}_m , and H_A are the nearest-neighbor exchange interaction, spin vector, and anisotropy field, respectively³ (the g factor is taken equal to 2). The Hamiltonian is second quantized using the linearized Holstein-Primakoff transformation. Apart from a constant term,

$$H = \sum_m E_m A_m^\dagger A_m + \sum_n E_n B_n^\dagger B_n + \sum_{mn} V_{mn} (B_m A_m + A_m^\dagger B_m^\dagger), \quad (2)$$

where the boson operator A_m^\dagger (B_n^\dagger) or A_m (B_n) creates or destroys a spin deviation at site m (n), re-

spectively. The random quantities are defined by

$$E_m = 2\sum_n J_{mn} S_n + 2\mu_B H_A(c) S_m, \quad E_n = 2\sum_m J_{mn} S_m + 2\mu_B H_A(c) S_n, \quad V_{mn} = 2J_{mn} (S_m S_n)^{1/2}.$$

E_m and E_n are the Ising energies required to create a spin deviation at sites m and n , and are determined by the cluster composition of nearest neighbors. A concentration dependence of the anisotropy field is explicitly allowed for.

To study the behavior of the magnetic excitation we create a spin deviation at site m_0 at time $t=0$ and compute the probability amplitude that it will propagate to site m at time t . This is found from the Green's function defined by

$$G_{mm_0}(t) = -i \langle g | \theta(t) e^{iHt} A_m e^{-iHt} A_{m_0}^\dagger | g \rangle, \quad (3)$$

where $\theta(t)$ is the unit step function and $|g\rangle$ is the ground state. We also define an auxiliary Green's function

$$G_{nm_0}^{(+)}(t) = -i \langle g | \theta(t) e^{iHt} B_n^\dagger e^{-iHt} A_{m_0}^\dagger | g \rangle. \quad (4)$$

We are particularly interested in $G_{m_0 m_0}(t=\infty)$ and define a delocalization (localization) of the excitation by the requirement that $G_{m_0 m_0}(t=\infty) = 0$ ($\neq 0$) in accordance with Anderson's criterion. These Green's functions satisfy coupled equations of motion,

$$\begin{aligned} i(d/dt)G_{mm_0}(t) &= \delta(t)\delta_{mm_0} + E_m G_{mm_0}(t) + \sum_n V_{mn} G_{nm_0}^{(+)}(t), \\ i(d/dt)G_{nm_0}^{(+)}(t) &= -E_n G_{nm_0}^{(+)}(t) - \sum_m V_{mn} G_{mm_0}(t). \end{aligned} \quad (5)$$

We have assumed for simplicity that $\langle g | A_m^\dagger, A_{m_0} | g \rangle = \delta_{mm_0}$ (Kronecker δ) and $\langle g | B_n^\dagger A_{m_0} | g \rangle = 0$, though in fact the antiferromagnetic ground state is not completely aligned. This simplification is not essential, neglecting only a small "background effect" unimportant for our purpose, but it materially simplifies our analysis. With the use of a Fourier transformation,

$$G_{mm_0}(t) = (2\pi)^{-1} \int_{-\infty}^{\infty} G_{mm_0}(E) e^{-iEt} dE, \quad G_{nm_0}^{(+)}(t) = (2\pi)^{-1} \int_{-\infty}^{\infty} G_{nm_0}^{(+)}(E) e^{-iEt} dE, \quad \text{Im} E = 0^+, \quad (6)$$

(5) leads to the recurrence relation

$$(E - E_m)G_{mm_0}(E) = \delta_{mm_0} - \sum_{m'} \sum_n \frac{V_{mn} V_{nm'}}{E + E_n} G_{m'm_0}(E). \quad (7)$$

For a pure crystal, (7) is readily solved in reciprocal-lattice space, leading to the well-known magnon spectrum $E_{\vec{k}} = [E_m^2 - (zV_{\vec{k}})^2]^{1/2}$, where $V = 2JS$, $E_m = 2zJS + 2\mu_B H_A S$, z is the coordination number, and $\gamma_{\vec{k}} = z^{-1} \sum_{\vec{\delta}} \exp(i\vec{k} \cdot \vec{\delta})$ with $\vec{\delta}$ the nearest-neighbor position vector. By iteration it follows from (7) that

$$G_{m_0 m_0}(E) = [E - E_{m_0} - V_c(E)]^{-1}, \quad (8)$$

where the Anderson series $V_c(E)$ is given by

$$\begin{aligned} V_c(E) &= - \sum_n \frac{V_{m_0 n} V_{n m_0}}{E + E_n} + \sum_{m_1}' \sum_{n_1} \sum_{n_2} \frac{V_{m_0 n_1} (-1) V_{n_1 m_1}}{E + E_{n_1}} \\ &\quad \times \left[\frac{V_{m_1 n_2} (-1) V_{n_2 m_0}}{(E - E_{m_1})(E + E_{n_2})} + \sum_{m_2}' \sum_{n_3} \frac{V_{m_1 n_2} (-1) V_{n_2 m_2} V_{m_2 n_3} (-1) V_{n_3 m_0}}{(E - E_{m_1})(E + E_{n_2})(E - E_{m_2})(E + E_{n_3})} + \dots \right], \end{aligned} \quad (9)$$

with \sum' meaning that the summation excludes the m_0 site. The most important idea of the Anderson theory is that, if the above series converges at an energy point E given by the zero of the real part of the denominator of (8), then $V_c(E)$ will have a vanishing imaginary part and the corresponding perturbed wave function will be localized around the site m_0 . However, $V_c(E)$ is a stochastic variable and one has to study this quantity in a probability sense. We consider a path of "length" $2L+1$ [i.e., the $(2L+1)$ th order term in (9) with $2L+1$ running indices]. The major difficulty of treating this quantity lies in the fact that the series may "visit" a same site repeatedly, thus causing serious correlation. To avoid this difficulty Anderson² uses Watson's⁴ multiple scattering theory and resums the series in such a way that no repetition of the same site in the path occurs. The price one pays is the introduc-

tion of an additive term (defined as the correlation energy) in each of the energy denominators. This complicates the problem greatly, although the effect of the additive term becomes small, according to Anderson, if the ratio (η) of the transfer matrix element to the fluctuation of the random energy levels (in our case $\eta \approx z^{-1}$) is small. Therefore we study the following quantity, a typical term representing a self-avoiding path in the resummed series (9),

$$T_{2L} = \left| \frac{V_{m_1 n_1} V_{n_1 m_2} V_{m_2 n_2} \cdots V_{n_L m_{L+1}}}{(E - E_{m_1})(E + E_{n_1})(E - E_{m_2}) \cdots (E + E_{n_L})} \right|, \quad (10)$$

where $2L \gg 1$ and correlation energies are neglected. The probability distribution function of T_{2L} is most conveniently found by introducing a quantity $X = \ln(T_{2L})$ and computing, $\Lambda(p)$ defined by

$$\Lambda(p) = \int_{-\infty}^{\infty} P(X) e^{ipX} dX, \quad (11)$$

where $P(X)$ is a normalized probability distribution function of X . We introduce a simplification of the problem at this point by taking $V^{\text{Mn-Co}} = (V^{\text{Mn}} V^{\text{Co}})^{1/2}$. This approximation was also made by Svensson *et al.*⁵ for real exchange interactions and greatly simplifies the mathematics. In (11), $\Lambda(p)$ is just the average of the quantity e^{ipX} . This average can be computed with the above approximation to arbitrary accuracy in the limit $L \rightarrow \infty$ for an arbitrary concentration (c) of Co ions accounting for all the possible occurrences of the cluster composition of the nearest neighbors. As a result, the inversion of (11) yields

$$P(X) = \sigma^{-1} (2\pi L)^{-1/2} \exp[-(X - X_0)^2 / 2L\sigma^2]. \quad (12)$$

Here

$$X_0 = -L[(1-c)\langle \epsilon_{\alpha}^-(\text{Mn}) + \epsilon_{\alpha}^+(\text{Mn}) \rangle + c\langle \epsilon_{\alpha}^-(\text{Co}) + \epsilon_{\alpha}^+(\text{Co}) \rangle],$$

$$\sigma^2 = (1-c)\{[\Delta \epsilon_{\alpha}^-(\text{Mn})]^2 + [\Delta \epsilon_{\alpha}^+(\text{Mn})]^2\} + c\{[\Delta \epsilon_{\alpha}^-(\text{Co})]^2 + [\Delta \epsilon_{\alpha}^+(\text{Co})]^2\},$$

with

$$\epsilon_{\alpha}^{\pm}(\text{Mn}) = \ln|E/V_{\text{Mn}} \pm E_{\alpha}(\text{Mn})/V_{\text{Mn}}|, \quad \epsilon_{\alpha}^{\pm}(\text{Co}) = \ln|E/V_{\text{Co}} \pm E_{\alpha}(\text{Co})/V_{\text{Co}}|,$$

where

$$E_{\alpha}(\text{Mn}) = 2J^{\text{Mn}} S^{\text{Mn}}(z - \alpha) + 2J^{\text{Mn-Co}} \alpha + 2\mu_B H_A(c) S^{\text{Mn}}, \quad E_{\alpha}(\text{Co}) = 2J^{\text{Co}} S^{\text{Co}} \alpha + 2J^{\text{Co-Mn}}(z - \alpha) + 2\mu_B H_A(c) S^{\text{Co}}.$$

The E_{α} terms are the Ising energies at manganese and cobalt sites with α nearest cobalt neighbors, occurring with a probability $p_{\alpha} = [z!/\alpha!(z-\alpha)!](1-c)^z c^{\alpha}$. The angular brackets mean averaging over the cluster composition (α) of the nearest neighbors, and $[\Delta \epsilon_{\alpha}]^2$ means the square of the standard deviation. The "sharp" Gaussian distribution (12) is simply a consequence of the central-limit theorem which holds here because of the finite number of discrete energy levels. Therefore (12) is expected to hold for a dominant number of possible paths (z^{2L}), although it is derived for a total of K^{2L} self-avoiding paths (K is the connective constant in the percolation theory). According to (12) almost all of the T_{2L} will lie in the range $T_{2L} = \exp(x_0) \exp(\pm \zeta \sigma \sqrt{L})$ with a probability approaching unity for $\zeta \gg 1$ (say $\zeta = L^{1/4}$). This means that we can treat the quantity

$$S_{2L} \equiv \sum_{\pm} z^{2L} T_{2L}$$

as a random-walk problem with a unit step given by T_{2L} for a total of z^{2L} steps. As is well known, one obtains a "sharp" Gaussian distribution centered at $S_{2L} = (\frac{1}{2} - q)z^{2L}T_{2L}$ with a "small" width of order of $z^L T_{2L}$. The quantity q includes the effect of a small deviation from equal probabilities for both negative and positive signs, and is given by $q = \frac{1}{2} + O(L^{-z-2})$. Thus we find the Anderson series will always converge if $(L^{-z-2}z^{2L}T_{2L})^{1/L} < 1$, and diverge otherwise. Using the fact that $[L^{-z-2} \exp(\pm \zeta \sigma \sqrt{L})]^{1/L} = 1$ for $L = \infty$, one obtains a critical condition for the convergence of the series:

$$\sum_{\alpha=0}^z P_{\alpha} \left\{ (1-c) \ln \left| \frac{E^2 - E_{\alpha}(\text{Mn})^2}{(zV_{\text{Mn}})^2} \right| + c \ln \left| \frac{E^2 - E_{\alpha}(\text{Co})^2}{(zV_{\text{Co}})^2} \right| \right\} > 0. \quad (13a)$$

This is our central result: the critical condition for the localization. We note that for the energy

FIG. 1. Regions of localized and propagating states according to (13) for $K(Mn_{1-c}Co_c)F_3$. The parameters used are $z=6$, $S^{Mn}=5/2$, $J^{Mn}=0.0792$, $2\mu_B H_A(c=0) \approx 0$ (in THz) (Ref. 7), and $S^{Co}=1/2$, $J^{Co}=1.17$, $2\mu_B H_A(c=1)=0.07$ (in THz) (Ref. 8). A linear concentration dependence is assumed for $H_A(c)$. The dashed lines suggest a rapid drop to the band tops (indicated by the arrows).

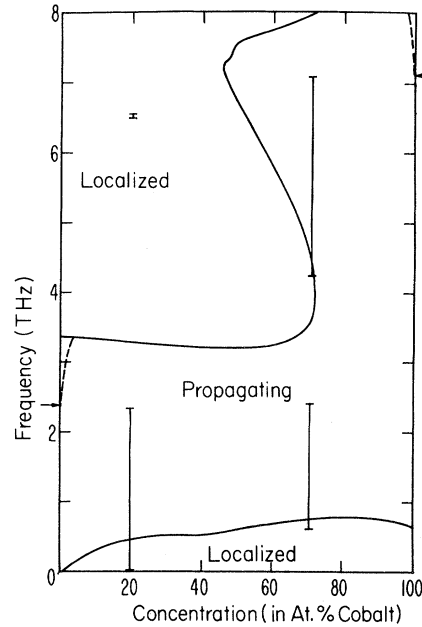
range where E is (i) smaller than the minimum of $E_\alpha(Mn)$ and $E_\alpha'(Co)$, or (ii) larger than the maximum of $E_\alpha(Mn)$ and $E_\alpha'(Co)$, [it turns out that $E_\alpha(Mn)^{min} < E_\alpha'(Co)^{min}$ and $E_\alpha(Mn)^{max} < E_\alpha'(Co)^{max}$], there is no randomness of the sign for the summand of S_{2L} . However, it is readily seen that the same criterion for convergence as (13a) is obtained in either case. For the latter case, however, the series will possess no imaginary part even in the divergent energy range because of the alternating sign in the Anderson series, thus always leading to a localization. This gives an additional criterion for localization,

$$E > \max\{E_\alpha(Co)\} = \max\{2J^{Co}S^{Co}\alpha + 2J^{Co-Mn}(z - \alpha) + 2\mu_B H(c)S^{Co}\}. \quad (13b)$$

Similar situations occur at $c=0$ ($c=1$) for the energy range $E_{\alpha=0}(Mn) < E [E_{\alpha=z}(Co) < E]$. In this case there is no randomness of the sign because of lack of cobalt (manganese) ions [see (9)], and the sign in the series will alternate leading to localization. This simply means one is at the top of the pure band. We note that (13a) is similar to the result of Ziman⁶ for an electronic localization in the binary alloy. Our result (13a) also approaches the empirical formula of Buyers *et al.*,¹ if we ignore the effect of the cluster composition, only keeping $E(Mn)$ and $E(Co)$ as distinctive. In Fig. 1 we exhibit the critical E -vs- c curve for given values^{7,8} of parameters for $K(Mn_{1-c}Co_c)F_3$. For the reasons already discussed, the curves should drop rapidly to the band tops (as indicated by the dashed lines) in the pure limits ($c=0$, $c=1$). It is likely to be rapid because randomness of sign in (9) will be introduced even with slight disorder. {We expect that the disorder will prevail in the concentration range $(1 - c^z)^{\xi L}$, $[1 - (1 - c)^z]^{\xi L} \ll 1$ (ξ is of order unity), where all kinds of cluster compositions are available along the path}. We also show the recent experimental data of Buyers *et al.*,¹ by vertical line segments. The theory predicts localized and propagating upper branches, respectively, for $K(Mn_{0.8}Co_{0.2})F_3$ and $K(Mn_{0.29}Co_{0.72})F_3$, in agreement with their experiments. However, for the lower branches of these the theory predicts localized states toward the bottom as shown, whereas the above authors claim all of them are propagating. The figure suggests that the modes are not localized beyond the cobalt concentration of about 70% for the upper branch.

We expect that the model will be particularly applicable to a system with perovskite structure where the nearest-neighbor exchange interaction is dominant. This method can also be applied to other (e.g., ferromagnetic) magnetic systems.

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High-Temperature Superconductivity in Bridge and Layer Compounds

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A microscopic model is presented for the structure and superconductivity of $A_x T_{1+y} C_2$ compounds, where A is a monovalent atom or radical, T is an even-valence transition metal, C is a chalcogenide, and the parameters x and y lie in the ranges $0.1 < x \lesssim 0.3$, $0 \leq y \lesssim 0.5$. The model explains why the maximum superconducting transition temperature T_S^{max} is higher than 12°K in bridge compounds with $y = 0.1$, although the maximum value of T_S observed in related layer compounds with $y = 0.0$ is about 6°K.

Interest in layer compounds as possible superconductive materials was stimulated by the work of Wilson and Yoffe (WY)¹ on the transition-metal dichalcogenides TC_2 . When these materials are pure and the even-valence transition metal T belongs to column IVb or VIb, they form sandwich structures which are small-gap semiconductors. In analogy with Cohen's many-valley superconductivity² in GeTe, WY suggested¹ that the TC_2 semiconductors might become superconductive if properly doped, and that an easy way to achieve high carrier densities was through intercalation of monovalent atoms or radicals. Several column-Vb (odd-valence) transition-metal dichalcogenides were already known to be metallic and superconducting, and these have recently been intercalated³ with the result that those with T_S near 1°K in the pure state may exhibit increased values of T_S up to 3°K when intercalated, while those with T_S near 6°K in the pure state actually show a decrease of T_S with intercalation.

The compound $Ti_{1+y}S_2$ ($0.1 \leq y \leq 0.5$) does not have a sandwich structure. The Ti atoms are distributed alternately between filled and partially occupied sheets, tending as y approaches 0.5 to the Ti_3S_4 structure, where the metal layers are alternately filled and half filled.⁴ The y or supernumary Ti atoms act as bridges connecting filled Ti sheets, and make a contribution to the density of states at the Fermi energy intrinsically different from that of intercalated donors. Barz *et al.*⁵ have discovered that the value of T_S in intercalated $Ti_{1.1}S_2$ is about 10 to 13°K, or about twice as great as T_S^{max} in the intercalated $T(Vb)C_2$ family.³ This suggests that the $A_x T_{1+y} C_2$

compounds with $y \gtrsim 0.1$ be described as intercalated bridge compounds in contrast to $A_x TC_2$ compounds, which may be described simply as intercalation compounds. As we shall see, the experimental evidence⁵ indicates that the role played by the intercalated A atoms in affecting T_S is incidental in the bridge compounds.

A semiempirical model of the energy bands of TC_2 compounds has recently been presented.⁶ The analysis suggests that stoichiometric TiS_2 would have an energy gap of about 1 eV separating a filled Ti 3d-S 3p valence band from an empty Ti 4s conduction band. These two bands have a high, approximately two-dimensional, density of states, as shown in Fig. 1(a). When intercalated donors in sufficient density are added to the material, a low, broad, three-dimensional s-p band is expected to fill the energy gap of the host semiconductor. Because the intercalated ions

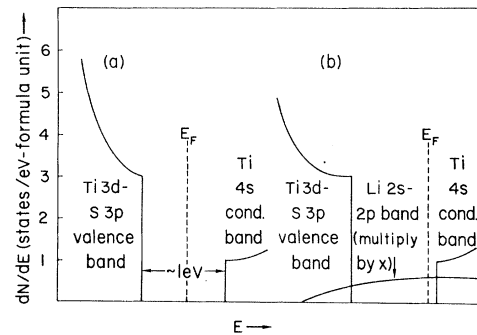


FIG. 1. Density of electronic states near E_F in stoichiometric TiS_2 . (a) Without intercalation TiS_2 is a small-gap sandwich semiconductor. (b) $Li_x TiS_2$ ($x \sim \frac{1}{2}$) is a metal with a low density of states near E_F .