

of a new transport channel. It is proposed that, in addition to being incorporated as a positive defect with a singly coordinated chalcogen as a compensating center, some fraction of the thallium atoms introduces a shallow trapping level approximately 0.1 eV above the hole hopping states, N_h . While at lower concentrations the shallow traps do not contribute to the measured transit time, direct charge displacement through these traps will begin to dominate the intrinsic transport channel if their concentrations exceed the hopping density $N_h \approx 10^{19} \text{ cm}^{-3}$. This density is in numerical agreement with the observed transition at $\sim(1-5) \times 10^{19} \text{ Tl/cm}^3$ (Fig. 1). The changes of the transport properties (field dependence and activation energy of t_T and transient current shape) further support the argument that a new mechanism begins to dominate transport for $N_{T1} \approx 5 \times 10^{19} \text{ Tl/cm}^3$.

In summary, we have shown that thallium added to $a\text{-As}_2\text{Se}_3$ in concentrations $10^{17}\text{-}10^{19} \text{ cm}^{-3}$ significantly reduces transient hole transport but that it has no effect on photoluminescence and photoinduced ESR. Although our results can be explained in terms of the specific defect chemistry originally proposed by MDS and modified by KAF which invokes the existence of close defect pairs, several difficulties remain with that interpretation. We have proposed an alternative explanation which is generally consistent with the

proposed defect models but which, in our opinion, may provide a more satisfactory explanation.

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Excitations in a Random Ferromagnetic-Antiferromagnetic Alloy

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A study is presented of elementary excitations of an unfrustrated system which can exhibit frozen magnetism without average long-range order. In three dimensions long-wavelength excitations are quasipropagating with linear dispersion and quadratic damping in accord with hydrodynamic speculations and computer studies, but high-energy modes may be localized. In lower dimensions the modes are overdamped even in a linearized equation-of-motion approximation.

There is growing interest in the low-temperature elementary excitations of alloys with strong quenched ferromagnetic-antiferromagnetic exchange disorder; such systems can exhibit frozen (or quasifrozen) magnetism without average long-range order. Spin-glass systems¹ with frustrated² exchange are complicated by the difficulty of microscopically specifying the classical ground or low-lying metastable states, necessari-

tating poorly controlled³ approximations in analytic studies of their excitations. There exist, however, unfrustrated models which can exhibit frozen magnetism without long-range order but have microscopically specifiable classical ground states. In this Letter I consider the excitations of the simplest such model with nontrivial dynamics, the Heisenberg-Mattis⁴ model. Its classical thermodynamics is trivial, but its excitation

spectrum shows several features which may be common to spin-glass systems. It is hoped that this study and its extensions will help guide studies of the frustrated systems.

The model is characterized by the Hamiltonian

$$H = -\sum J_{ij} \xi_i \xi_j \vec{S}_i \cdot \vec{S}_j, \quad (1)$$

where the ξ_i are independent random parameters taking the values ± 1 .⁵ The concentration of $\xi = -1$ is denoted by c . In the absence of an applied field the classical Néel ground state is simply given by $\langle S_i^\alpha \rangle = \xi_i S$. For $c = \frac{1}{2}$ it exhibits no long-range

order. Although the classical thermodynamics is trivial under the transformation $\vec{\tau}_i = \xi_i \vec{S}_i$, the dynamics is nontrivial at the Heisenberg level since this transformation does not conserve the spin commutation relations.

Here I consider the dynamics only within the unrenormalized normal-mode approximation in which the spin fluctuations are expanded to harmonic order about the classical ground state. The novel disorder element in the excitations is clearly manifest in the effective noninteracting-boson spin-deviation Hamiltonian resulting from Holstein-Primakoff transformations,

$$H = \sum \epsilon_i a_i^\dagger a_i + \sum t_{ij} [\lambda_{ij} a_i^\dagger a_j + \frac{1}{2}(1 - \lambda_{ij})(a_i^\dagger a_j^\dagger + a_i a_j)], \quad (2)$$

where ϵ and t are now constants but $\lambda_{ij} [= \frac{1}{2}(1 + \xi_i \xi_j)]$ is randomly 1 or 0. Equation (2) is clearly diagonalizable in principle, but the interesting questions are (i) the density of states of the resultant modes, (ii) their coupling to a laboratory-frame probe, and (iii) the existence of localized solutions. In this Letter I concentrate on (i) and (ii) in the low-energy limit, but work is in progress on (iii) and will be reported shortly.

A scattering experiment in a collinear Heisenberg spin system measures the quantity⁶

$$S(\vec{q}, \omega) = \lim_{\delta \rightarrow 0} \text{Im} \Delta(\vec{q}, \omega + i\delta), \quad (3)$$

where

$$\Delta(\vec{q}, E) = N^{-1} \sum_{ij} \Delta_{ij}(E) \exp[i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)] = \sum_j \langle \Delta_{ij}(E) \rangle_\xi \exp[i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)] \quad (4)$$

$$\Delta_{ij}(E) = \langle \langle S_i^+; S_j^- \rangle \rangle_E + \langle \langle S_i^-; S_j^+ \rangle \rangle_E; \quad (5)$$

Zubarev Green's-function notation is employed and $\langle \rangle_\xi$ refers to a spatial disorder average. $\Delta_{ij}(E)$ turns out to be particularly advantageous for perturbative analysis. Linearizing the equations of motion for the Green's-functions $\langle \langle S^+; S^- \rangle \rangle$, $\langle \langle S^-; S^+ \rangle \rangle$, and substituting the Néel values for $\langle S_i^\alpha \rangle$, yields after some algebra the self-consistent equations for Δ ,

$$\Delta_{ij} = \Delta_{ij}^0 + \sum_{pl} \Delta_{il}^0 V_{pl} \Delta_{lj}, \quad (6)$$

where

$$V_{pl} = -\frac{1}{4} J_{pl} (\xi_p \xi_l - \varphi), \quad (7)$$

Δ_{ij}^0 is the Fourier transform of

$$\Delta^0(\vec{q}, E) = \frac{4S^2 [J(0) - J(\vec{q})]}{\{E^2 - S^2 [J(0) - J(\vec{q})]\} [J(0) - \varphi J(\vec{q})]}, \quad (8)$$

and φ is arbitrary. Iterating, averaging, and resumming (6) yields

$$[\langle \Delta(\vec{q}, E) \rangle_\xi]^{-1} = [\Delta^0(\vec{q}, E)]^{-1} - \Sigma(\vec{q}, E), \quad (9)$$

where $\Sigma(\vec{q}, E)$ is the infinite sum of terms of the form $\langle V \Delta^0 \dots V \rangle_\xi$ which are irreducible with respect to the ξ averages.

The perturbation analysis simplifies near the two limits of the relevant concentration range $c = 0$ to $\frac{1}{2}$. In this Letter I concentrate on these regimes and on the limit of small (\vec{q}, E) .

In the low-concentration regime the convenient choice for φ is unity so that to order c for any (\vec{q}, E) the self-energy is c times the sum of the irreducible terms of the form $V \Delta^0 \dots V$ for which each V has a common vertex. The series is readily summed for any finite-ranged J_{ij} but for simplicity I re-

strict discussion to nearest-neighbor interaction, yielding

$$\begin{aligned} \Sigma(\vec{q}, E) = & c \{ z J^2 \sum_{\delta, \delta'} \Delta_{\delta\delta'}^0 + z J^2 \Delta_{00}^0 + 2(1 + z J \Delta_{0\delta}^0 / 4) \sum_{\delta} \exp(i\vec{q} \cdot \vec{\delta}) + (J^2 \Delta_{00}^0 / 4) \sum_{\delta, \delta'} \exp[i\vec{q} \cdot (\vec{\delta} - \vec{\delta}')] \} \\ & \times \{ (1 + z J \Delta_{0\delta}^0 / 4)^2 - z J^2 \Delta_{00}^0 \sum_{\delta, \delta'} \Delta_{\delta\delta'}^0 / 16 \}^{-1} + O(c^2). \end{aligned} \quad (10)$$

The δ 's label nearest neighbors and z is the lattice coordination number. $S(\vec{q}, \omega)$ follows immediately to order c for any (\vec{q}, E) , but I report here explicitly only for long wavelengths—for small q the dominant feature of $S(\vec{q}, \omega)$ is well-defined peaks at $\omega(q) = \pm \lambda(1 + 2c)q^2$, where λ is the stiffness of the pure ferromagnet; the widths of the peaks are proportional to cq^{d+2} , where d is the lattice dimensionality. Note that in contradistinction to a diluted ferromagnet the addition of antiferromagnetically coupled impurities stiffens the spin waves. An analogous analysis can be applied to the case of ferromagnetic impurities in an antiferromagnet.

Let us now turn to the less *a priori* predictable case of the high concentration limit $c = \frac{1}{2}$. In this case the convenient choice of φ is zero, so that Δ^0 is different from that used above. A simplifying feature in this case is the fact that the average of any odd power of ξ is zero while any even power is 1. Additional simplification ensues for small (\vec{q}, E) since it may be shown that the dominant long-wavelength behavior of $S(\vec{q}, \omega)$ is given by retaining in Σ only the irreducible parts of the series $V_{ii} \Delta_{ii}^0 V_{ii} + V_{ii} \Delta_{ii} V_{im} \Delta_{mm}^0 V_{mi}$, etc. To dominant order in (q, E) the series can be summed competely. For dimension $d \geq 3$, $S(\vec{q}, \omega)$ at long wavelengths has peaks whose widths are less than the means, indicating quasipropagation. The peaks are located at

$$\begin{aligned} \omega(\vec{q}) = & \pm S \{ [J(0) - J(\vec{q})] J(0) / I \}^{1/2} \\ = & \pm (z J \lambda / I)^{1/2} S q + \dots, \end{aligned} \quad (11)$$

where I is the standard lattice sum,

$$I = N^{-1} \sum_{\vec{q}} [1 - J(\vec{q}) / J(0)]^{-1}, \quad (12)$$

which for cubic lattices takes the following values⁷: simple cubic, $I = 1.5164$; bcc, $I = 1.3932$; fcc, $I = 1.3447$. In three dimensions the corresponding half width is given by

$$\Gamma(q) = \frac{3}{4} \lambda q^2 \frac{\pi}{I^{3/2}} \left(\frac{J(0)}{\lambda q_m^3} \right)^{3/2} S, \quad (13)$$

where q_m is the Debye radius. For a simple-cubic lattice this reduces to

$$\Gamma(q) = 0.0792 \omega(q)^2 J S. \quad (14)$$

Note that the q dependences of (11) and (13) are the same as predicted in a recent hydrodynamic

analysis of a spin-glass⁸; that analysis must, however, be modified somewhat in the present case—for example, the collinearity of the spin structure leads to two modes in place of the three predicted for an isotropic system. The concept of a nonzero spin stiffness is thus vindicated in this case. The origin of the q^2 damping is, however, different in the two studies.

One can also compare the analytic results of this work with computer results of Ching, Leung, and Huber⁹ for the simple-cubic Heisenberg-Mattis model at the point $(\pi/6)(1, 1, 1)$, the only low q value considered by those authors. Equation (11) predicts that the position of the peak of $S(\vec{q}, \omega)$ will be reduced by 0.812 compared with the value with I replaced by unity. From (14) the width is predicted to be of order $J S / 4$. Both these predictions are in good accord with the computer study.

From the q linearity of the long-wavelength dispersion relation (11) it follows that, as for the pure isotropic antiferromagnet, the zero-point spin deviations will be small compared to S for large S and the consequences of linearizing the equations of motion will be qualitatively correct.

In less than three dimensions, it is known that there is no stable cooperative order. Nevertheless, the linear analysis of (6) yields a one-body problem which is of interest in its own right even in lower dimensions, being nontrivial through the disorder. Without further analysis one cannot predict its relevance to true one-dimensional random magnets but note that experiments on one-dimensional pure antiferromagnets with large spins¹⁰ S show excitation spectra remarkably like those given by the analogous analysis of the pure system,¹¹ the des Cloiseaux-Pearson modes¹² being more relevant to low S .¹³ I report only for $d = 1$. For small c the general results reported above still apply. For $c = \frac{1}{2}$, however, quite a different situation ensues, since lattice sums such as (12) diverge. I find as the dominant small- (q, ω) behavior of $S(q, \omega)$ the following:

$$S(q, \omega) = 4 J S^2 q^2 \omega^{-1/2} \left[\frac{\nu q^2}{(\omega^{3/2} - \nu q^2)^2 + \nu^2 q^4} \right], \quad (15)$$

where $\nu = (\sqrt{2} J S)^{3/2}$. Thus, in this case we find that the energies of the peak and the width of

$S(q, \omega)$ are comparable, indicating strong damping. Note further that both the energy and the width tend to zero as $q \rightarrow 0$, as expected from Goldstone's theorem.

In dimensionality $d > 1$ the corresponding non-separable random-signed bond problem is frustrated and even the classical ground-state problem is nontrivial. However, for $d = 1$ and nearest-neighbor interactions frustration is absent for this model also and the above analysis can be extended. I therefore consider the model characterized by

$$H = -J \sum_i \eta_i \vec{S}_i \cdot \vec{S}_{i+1} \quad (16)$$

with the η_i taking randomly the values ± 1 with probability $1 - c, c$. With suitable correlations between η 's and ξ 's, particular manifestations of (1) and (16) are identical but for random and independent distributions only the cases with $c = \frac{1}{2}$ are equivalent. For small c one can again calculate $S(q, \omega)$ exactly to order c . For small (q, ω) I find that dominantly¹⁴

$$S(q, \omega) = (4S^2 J q^2 \omega^{-1/2}) \left[\frac{\alpha q^2}{(\omega^{3/2} - \alpha q^2)^2 + \alpha^2 q^4} \right], \quad (17)$$

with $\alpha = c(JS)^{3/2}$. Thus we see that in this case the damping is strong even in the low-concentration limit and laboratory-frame modes are non-propagating. The physical origin of the difference between the one-dimensional random-site and random-bond problems is to be found in the fact that in the former case the system maintains an overall ferromagnetism except for the particular case $c = \frac{1}{2}$, while in the latter any finite concentration of negative bonds removes long-range order in the classical ground state.

The densities of states follow from integrating $S(\vec{q}, \omega)$ over \vec{q} . Several extensions of this work can be envisaged. Within the linearized equation-of-motion approximation one can mention the investigation of the localization of high-energy ex-

citations (to be reported), the development of a coherent-potential approximation to extend the range of this study of $S(q, \omega)$, and the extension of the model to allow for noncollinearity of the spins. In low dimensions, or small S , extension beyond the linear approximation should be considered. Finally, it is to be hoped this study can throw light on the more complicated problem of spin-glasses.

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