field.

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EFFECT OF ZERO-POINT SPIN DEVIATION ON ENERGY LEVELS OF MAGNETIC IMPURITIES IN ANTIFERROMAGNETS

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In a recent paper Johnson, Dietz, and Guggenheim[1] studied the infrared emission spectra of Ni^{++} as an impurity in the antiferromagnetic crystals MnF_2, $KMnF_3$, and $RbMnF_3$. The ground state of the Ni ion, $^3A_{2g}$, was found to split into three levels, due to the exchange field of the neighbor Mn ions. A small asymmetry in the splitting was observed. We report in this paper more accurate measurements of the asymmetry in the splitting of these levels (see Fig. 1) using unstrained single crystals with smaller concentration of Ni (below 10 parts per million of Mn) and with high resolution. This asymmetry is shown to arise from the effects of the zero-point spin deviation.

For a given crystal, the spacing between the three levels ($M_S = 1, 0, -1$) should be constant according to the molecular-field model, in approximate agreement with the experimental results. However, there are small differences, $\delta E = \Delta E_{1,0} - \Delta E_{0,-1}$, of around 10 cm^{-1} which are not explained within a simple molecular-field model. In order to explain those anomalies, we extend the molecular-field model, by taking into account the difference between the usual effective magnetic field and the actual exchange interaction with neighbors as a perturbation. The Hamiltonian of our system is

$$\mathcal{H} = \beta H \sum_i g_i S_{zi} - 2 \sum_{i \neq j} J_{ij}(S_{zi}S_{zj} + S_{xi}S_{xj} + S_{yi}S_{yj}) + \beta H_A\Big(\sum_i g_i S_{zi} - \sum_j g_j S_{zj}\Big),$$

where H_A is the anisotropy field. Usually the exchange interaction J_{ij} is negligible except when i and j are nearest neighbors. The molecular-field theory considers only the first term of the exchange interaction (neglecting the other two) and equates it to an effective magnetic field H_E:

$$\mathcal{H}_0 = \beta H \sum_i g_i S_{zi} + \beta \sum_i g_i H_{Ei} S_{zi} + \beta H_A\Big(\sum_i g_i S_{zi} - \sum_i g_j S_{zj}\Big).$$

Therefore, our perturbation is

$$\mathcal{H}' = -2 \sum_{i \neq j} J_{ij}(S_{xi}S_{xj} + S_{yi}S_{yj}) = -\sum_{i \neq j} J_{ij}(S_{+,i}S_{-,j} + S_{-,i}S_{+,j}).$$

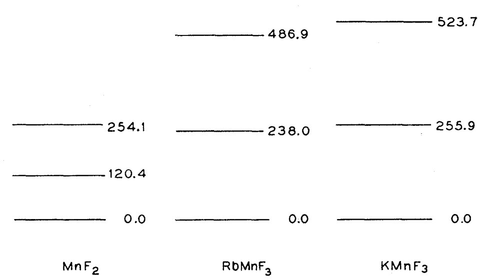

FIG. 1. Energy levels of Ni^{++} in different antiferromagnetic crystals at 4.2°K, in cm^{-1}. The emission lines from $^3T_2$ are about 1 cm^{-1} half-width, and the frequencies quoted above are obtained from splittings measured between the peaks of the lines. The estimated error in measuring the splittings is about 0.1 cm^{-1}.

This perturbation will mix excited states obtained by flipping the spins of the ions into the ground state.

Let us consider, for example, a simple antiferromagnetic body-centered cubic structure, which can be divided into sublattices, 1 and 2, in such a way that all the neighbors of an atom on sublattice 1 are on sublattice 2, and vice versa. The unperturbed ground state consists of all the spins on sublattice 1 aligned parallel along the z direction, and all the spins on sublattice 2 aligned along the $-z$ direction. Deviations from such a simple model for the ground state of an antiferromagnet have been considered theoretically in several papers,[2,3] but have not been observed experimentally.

We want to consider the energy levels of Ni as an impurity in an antiferromagnetic crystal. The unperturbed hamiltonian $\mathcal{H}_0$ will spit the three levels ($M_S=1,0,-1$) of the ground state ($^3A_{2g}$) of Ni symmetrically. However, $\mathcal{H}'$ will affect these states in a different way and, consequently, give a contribution to δE.

Assuming an antiferromagnetic interaction between the Ni ion and its Mn neighbors, the unperturbed ground state will consist of the spin of the Ni ion oriented, for instance, in the − direction, and the nearest Mn ion in the + direction. $\mathcal{H}'$ will mix the ground state with excited states coming from flipping simultaneously the spins of two neighboring ions by $\Delta M_S=+1$ and -1, respectively. The most important contribution to δE occurs when the flipping ions are the Ni and a first-neighbor Mn. For simplicity, let us consider only one Mn neighbor to the Ni ion, and write the basis functions as $|M_S(\mathrm{Ni}),M_S(\mathrm{Mn})\rangle$. The three unperturbed states of Ni are

$$|-1,+\tfrac{5}{2}\rangle;\quad |0,+\tfrac{5}{2}\rangle;\quad |+1,+\tfrac{5}{2}\rangle.$$

Now, $\mathcal{H}'$ will mix $|-1,+\frac{5}{2}\rangle$ with $|0,+\frac{3}{2}\rangle$, and $|0,+\frac{5}{2}\rangle$ with $|+1,+\frac{3}{2}\rangle$. However, it will not affect $|+1,+\frac{5}{2}\rangle$. Since the difference in energy between $|-1,+\frac{5}{2}\rangle$ and $|0,+\frac{3}{2}\rangle$ is

$$g_{\mathrm{Ni}}\beta H_{E,\mathrm{Ni}}+[(z-1)/z]g_{\mathrm{Mn}}\beta H_{E,\mathrm{Mn}},$$

and the same value for the difference between $|0,+\frac{5}{2}\rangle$ and $|+1,+\frac{3}{2}\rangle$, we obtain, in second-order perturbation and summing over all nearest neighbors,

$$\delta'E=\frac{10zJ_a^{\,2}}{g_{\mathrm{Ni}}\beta H_{E,\mathrm{Ni}}+[(z-1)/z]g_{\mathrm{Mn}}\beta H_{E,\mathrm{Mn}}}=\frac{-2zJ_a^{\,2}}{zJ_a+(z-1)J_b},$$

where J_a is the exchange integral between a Ni and a Mn ion, and J_b between two Mn ions, and z is the number of nearest neighbors.

Let us consider now contributions to δE coming from flipping simultaneously two Mn ions. Since we assume J_{ij} to be negligible except when i is a nearest neighbor (on the other sublattice) of j, one of the flipping Mn ions must be a nearest neighbor of the other flipping Mn ion. There will be a contribution if these ions have different effective fields for each of the orientations of the Ni spin. Therefore, these contributions will decrease strongly as we increase the distance between the pair of flipping Mn ions and the Ni. When one of the flipping Mn ions is a first neighbor to a Ni, we get, after summing over all possible pairs,

$$\delta''E=-\frac{50z(z-1)J_b}{10z-7}\left[1-\frac{1}{1-4J_a^{\,2}/(10z-7)^2J_b^{\,2}}\right].$$

Flipping spins on other Mn ions[4,5] will not contribute to δE in second order.

Values of J_b are obtained from Low et al.,[6] Windsor and Stevenson,[7] and Pickart, Collins, and Windsor.[8] We get J_a from the splitting of the lowest two energy levels, $\Delta E_{0,-1}$, of Ni in the antiferromagnets (Fig. 1), assuming the molecular-field model ($-3.01\ \mathrm{cm}^{-1}$ for MnF_2, -7.93 for $RbMnF_3$, and -8.53 for $KMnF_3$), since this splitting is not significantly affected by the transverse part of the exchange. Table I compares the theoretical values δE

Table I. Comparison of experimental and theoretical values for the asymmetry in the splitting of $^3A_2$ state of nickel ions in antiferromagnetic compounds.

	$\delta'E$ (cm^{-1})	$\delta E=\delta'E+\delta''E$	δE (Experimental)
MnF_2	12.5	12.3	13.3 ± 0.1
$RbMnF_3$	12.7	11.6	10.9 ± 0.1
$KMnF_3$	13.7	12.5	11.9 ± 0.1

$= \delta'E + \delta''E$ with the experimental ones. The small disagreement (less than 8%) may arise in part from the approximations[9] in obtaining J_a and from high-order interactions.[3] This result gives support to our extension of the molecular-field model for discussing deviations from the ground state consisting of antiparallel spins on different sublattices, and confirms the observed differences δE as experimental evidence for the existence of deviations from that ground state.

The spin deviations defined as $1-\langle S_z\rangle/S$ can be computed within this approximation. For the nickel ion in $KMnF_3$ and $RbMnF_3$ we get 4.1%, and in MnF_2, 2.7%. The spin deviation of the Mn ions having a nickel nearest neighbor is about the same as for Mn ions in the pure crystal. However, it has been shown that it is necessary to go to higher order perturbation theory to obtain accurate values of the spin deviation. For example, we compute a deviation of 1.3% for pure MnF_2 in second order, while Walker[10] obtains 1.7% in fourth order. The good agreement between our estimates of δE and the experimental values suggests that the convergence in computing the asymmetry of the energy splittings is faster than for the spin deviations.

In the case of MnF_2, part of the observed δE is due to crystalline effects (zero-field splitting) since the local symmetry of the Mn ions is not octahedral. Our estimated $\delta'E$ contains both the crystalline and the exchange contributions and was estimated by solving the eigenvalues of the spin Hamiltonian with an effective magnetic field, H_E, due to exchange interaction and tetragonal ($D = 4.05$ cm^{-1}) and rhombic ($|E| = 3.28$) parameters. These parameters were obtained by linear extrapolation as a function of the ratio of lattice parameters c/a, from the corresponding parameters[11] in ZnF_2 ($D = 4.19$ cm^{-1}, $|E| = 2.68$ cm^{-1}) and values we obtained by optical spectroscopy in MgF_2 ($D = 4.27$ cm^{-1}, $|E| = 2.33$ cm^{-1}). However, the resultant δE does not depend strongly on this extrapolation; for instance δE is increased by 0.3 cm^{-1} if we use the values of D and E corresponding to ZnF_2.

In conclusion, we have shown that an asymmetry in the splitting of the $^3A_2$ states of nickel impurities in antiferromagnetic compounds is produced by the transverse part of the exchange interaction, resulting in zero-point spin deviations of the ions. The effect is most sensitive to the deviations of the nickel and first-neighbor manganese ions. Although we have not measured the zero-point spin deviation of the pure, antiferromagnetic crystal, we have demonstrated that the formalism which predicts the disputed zero-point spin deviation of the pure crystal also correctly predicts the effect of the zero-point spin deviation on a nickel impurity.

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3^- CONTINUUM STATES OF O^{16} IN THE EIGENCHANNEL REACTION THEORY*

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The complete 3^- part of the S matrix for O^{16} has been computed in the one-particle, one-hole approximation. In the continuum states the isospin invariance is totally broken; analogous partial cross sections for protons and neutrons show large differences.

In a recent publication[1] a method has been described by which the eigenstates of the S matrix, i.e., the eigenchannels, can be directly computed. We have tried out this method in the case of the 3^- states of O^{16} in the one-particle, one-hole approximation. In this Letter we report briefly the results of this calculation. The details will be given elsewhere.

The essential points of the method are as follows: The eigenstates of the S matrix are standing waves in all experimental channels with a common phase shift, say $\delta^{(\beta)}$. There are as many eigenstates as there are open channels at this energy. We denote the amplitudes of the standing waves of an eigenstate of the S matrix in the experimental channel c by $V_c^{(\beta)}$. In terms of these quantities the S matrix is given by

$$S_{cc'} = \sum_\beta V_c^{(\beta)} \exp(2i\delta^{(\beta)}) V_{c'}^{(\beta)*}. \tag{1}$$

A knowledge of the $V^{(\beta)}$ and $\delta^{(\beta)}$ as functions of the energy thus allows the complete description of all one-particle reactions. For example, the total cross section then is (I = spin of target nucleus, s = spin of incident nucleon)

$$\sigma_{\text{tot}} = \frac{2\pi\lambda\!\!\!^{-2}}{(2I+1)(2S+1)} \times \sum_J (2J+1) \sum_c [1 - \operatorname{Re} S_{cc}^{[J]}], \tag{2}$$

where the summation over c is restricted to those channels which contain only the ground state of the target nucleus. We compute here only the term with $J = 3$. The form of the eigenchannel wave function in the asymptotic region, i.e., for $r_c \geq a$, is

$$\Psi^{(\beta)} = \sum_c V_c^{(\beta)} [\cos\delta^{(\beta)} F_c(k_c r_c) - \sin\delta^{(\beta)} G_c(k_c r_c)] \tilde{\psi}_c, \tag{3}$$

where the F and G are the regular and irregular radial functions of the continuum particle; for a neutron they are simply $j_l(k_c r_c)$ and $n_l(k_c r_c)$, respectively. The channel wave functions $\tilde{\psi}_c$ contain in addition to the wave function of the daughter nucleus (i.e., the hole state) the angular momentum part of the continuum particle.

The computation of the eigenchannels was done as follows: At a given energy, say E, the wave numbers k_c are known for all open channels from the binding energy and the spectrum of the bound states of the daughter nucleus. Assuming a phase shift, say δ, the logarithmic derivatives of the radial wave functions in all open channels are computed from (3) at $r_c = a$. Sets of single-particle wave functions for the different channels are now obtained for a real Saxon-Woods potential[2] using these logarithmic derivatives as the boundary conditions. Arbitrary boundary conditions can be used for the states appearing only in closed channels. An orthonormal set of particle-hole states is now constructed with these single-particle wave functions and the Hamiltonian is diagonalized in the space of these one-particle, one-hole (1p-1h) states. A zero-range force[3] with exchange was employed. The eigenvalues obtained are plotted as a function of δ in Fig. 1 for the case $E = 20$ MeV. The eigen-