

Magnetic susceptibilities of antiferromagnetic Re^{4+} compounds

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The low-temperature (0–30 K) antiferromagnetic susceptibilities of hexachloro- and hexabromorhenates (K_2ReCl_6 and K_2ReBr_6) are explained by using correlated effective-field theory and considering XY symmetry of the exchange Hamiltonian. The theory gives a good account of the observed magnetic susceptibilities of these compounds and the sublattice magnetization of the K_2ReCl_6 compound. The nearest- and next-nearest-neighbor exchange integrals for these compounds are $J_1 = -1.32 \text{ cm}^{-1}$, $J_2 = 0.20 \text{ cm}^{-1}$ for K_2ReCl_6 and $J_1 = -1.82 \text{ cm}^{-1}$, $J_2 = 0.15 \text{ cm}^{-1}$ for K_2ReBr_6 .

I. INTRODUCTION

Hexachloro- and hexabromorhenates belong to a class of sublattices where the magnetic ions form an fcc lattice. These are antiferromagnetic below the Néel temperatures 12.3 and 15.3 K, respectively.^{1–3} Among theoretical methods which have been applied to antiferromagnetism we may mention the generalized molecular-field model,⁴ the self-consistent molecular-field theory⁵ and the Green's-function method.⁶ Of these the generalized molecular-field model is the simplest and predicts various types of ordering possible in an antiferromagnetic lattice. The main drawback of this method is that it is not possible to include crystal-field effects, whereas one can include the crystal-field effects in the self-consistent molecular-field method. But using the self-consistent molecular-field method it is difficult to estimate the individual exchange integrals (J_1, J_2), etc., which give rise to the molecular field. The Green's function method though powerful becomes unwieldy to treat fcc lattices. As there is a minimum of four sublattices, one gets four coupled equations for the Green's functions. General solution in the presence of an external magnetic field is possible in random-phase approximation⁷ (RPA). In contrast to these methods the correlated effective-field (CEF) theory,⁸ while retaining the simplicity of mean-field (MF) theories, is capable of taking into account crystal-field effects and correlations. In Sec. II we represent a summary of available results of K_2ReCl_6 and K_2ReBr_6 and conclusions can be obtained by the generalized molecular-field model. In Sec. III we give CEF theory as applied to the compounds under investigation and in Sec. IV we present our results and comment on them.

II. SUMMARY OF AVAILABLE RESULTS

The hexachloro- and hexabromorhenates belong to the space group $Fm\bar{3}m (O_h^5)$ and have the antiferromagnetic

type of lattice.⁹ The magnetic ions are distributed in an fcc arrangement. The lattice constant a_0 and the parameter u [a_0u is the $\text{Re}-\text{Cl}(\text{Br})$ distance] are 9.861 and 0.24 Å for K_2ReCl_6 and 10.445 and 0.25 Å for K_2ReBr_6 . The nearest-neighbor distance between the magnetic ions is 6.97 Å in K_2ReCl_6 and 7.39 Å in K_2ReBr_6 . K_2ReCl_6 undergoes structural changes¹⁰ at temperatures 76, 103, and 111 K. The structural phase transitions at 103 and 111 K show no hysteresis whereas the transition at 76 K clearly shows hysteresis. On the basis of this observation the two transitions at higher temperatures may be of second order but the transition at 76 K is certainly of first order. Though the overall symmetry (space group) is reduced at low temperature the immediate environment of Re^{4+} ions has essentially octahedral symmetry.¹¹

Since the ground electronic state is orbitally nondegenerate ($t_{2g}^3 e_g^1 A_2$), the cluster is stable against Jahn-Teller distortion. However, if one includes the spin-orbit interaction, the ground state becomes fourfold degenerate and it belongs to Γ_8 irreducible representation of the octahedral double group. This state does become Jahn-Teller sensitive, but weakly so. The neutron-diffraction experiments^{2,3} suggest that the magnetic ordering in the compounds K_2ReCl_6 and K_2ReBr_6 is of type I. The magnetic moments are ferromagnetically aligned in (001) planes and the magnetic moments in adjacent planes are oriented antiparallel to each other. The magnetic structure of the compounds is shown in Fig. 1. The transition temperatures from these experiments are (12.3 ± 0.5) and 15.3 K for K_2ReCl_6 and K_2ReBr_6 , respectively. The magnetic susceptibility data¹² show that the transitions occur at (12.4 ± 0.5) K and (15.3 ± 0.5) K, respectively, agreeing with neutron-diffraction experiments. The specific-heat measurements^{13,14} place these transitions at (11.9 ± 0.1) K and (15.2 ± 0.1) K, respectively. The specific-heat measurements also give evidence of second-order phase transitions at 76, 103, and 111 K for K_2ReCl_6 and 225 and 245 K for

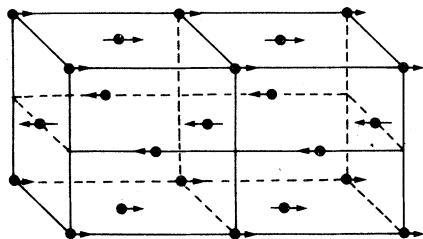


FIG. 1. Type I magnetic structure.

K_2ReBr_6 . Van Driel *et al.*¹⁵ have also observed the existence of a phase transition in K_2ReCl_6 at 111 K from the study of soft-mode behavior of nuclear quadrupole resonance (NQR) spectra. EPR measurements^{1,16} of the Re^{4+} ion in single crystals of K_2PtCl_6 give $g = 1.815 \pm 0.001$. The magnetic susceptibilities of K_2ReCl_6 and K_2ReBr_6 have been measured over the temperature range 80–300 K by several workers.^{17–20} A detailed theoretical calculation of the magnetic susceptibility in the paramagnetic phase has been done by Eisenstein.²¹ The optical-absorption spectra of Re^{4+} ion in dilute as well as concentrated systems have been recorded and analyzed^{11,21–25} by considering the crystal field of cubic symmetry and spin-orbit interaction.

ter Haar and Lines²⁶ have used a method due to Anderson²⁷ to study the stability of various antiferromagnetic patterns in fcc lattices. They have considered²⁶ third and fourth neighbor interactions and anisotropies in first and second nearest-neighbor interactions. They have derived expressions for the transition temperatures T_N and Θ in terms of exchange integrals. If we assume only nearest- and next-nearest-neighbor isotropic interactions, T_N and Θ , for type I order are given by⁴

$$\Theta = \frac{2S(S+1)}{3k} (12J_1 + 6J_2) ,$$

$$T_N = \frac{S(S+1)}{3k} (-8J_1 + 12J_2) .$$

Further, in order that type I order is stable with respect to the other types, the second neighbor interaction should be either zero or ferromagnetic.²⁸ As discussed in Ref. 26, anisotropies in the exchange integral also stabilize type I magnetic structure. Substituting the experimental values of Θ and T_N in the

above expressions [$\Theta = (-55 \pm 5)$ K for K_2ReCl_6 and $\Theta = (-76 \pm 10)$ K for K_2ReBr_6], we get $J_1 = -1.2$ cm^{-1} , $J_2 = -0.2$ cm^{-1} for K_2ReCl_6 and $J_1 = -1.6$ cm^{-1} , $J_2 = -0.4$ cm^{-1} for K_2ReBr_6 . J_2 values are small as expected but of wrong signs.

III. CEF THEORY: APPLICATION TO $S = 3/2$ SYSTEM

The correlated effective field theory was first proposed by Lines²⁹ in connection with lattice dynamics. Later he extended it to cover magnetic problems.⁸ In this and later works^{30–32} he showed the superiority of this method over the other effective field theories. The CEF theory has the simplicity of the molecular-field theories and is applicable to problems where the crystal-field energies, exchange energies, and thermal energies are all of the same order of magnitude. The correlations are determined by forcing a consistency with the fluctuation theorem.³³

In the CEF theory developed by Lines⁸ static spin correlations are taken into account. In this theory each spin S_j appearing in the equations of motion of a particular spin S_i is replaced by the sum of two terms as follows

$$S_j^\gamma \rightarrow \langle S_j^\gamma \rangle + A_{ij}^\gamma (S_i^\gamma - \langle S_i^\gamma \rangle), \quad (\gamma = x, y, z) ,$$

where $\langle S_j^\gamma \rangle$ represents the thermal average of S_j^γ and A_{ij}^γ is the temperature dependent static correlation parameter. Using the above replacement of the spin operators the effective Hamiltonian for the i th ion in the absence of applied magnetic field can be written as

$$H_i^0(\text{eff}) = H_i - \sum_{\gamma, J} J_{ij}^\gamma \alpha^\gamma (S_i^\gamma)^2 - 2 \sum_{\gamma, J} J_{ij}^\gamma S_i^\gamma (\langle S_j^\gamma \rangle - \alpha^\gamma \langle S_i^\gamma \rangle) , \quad (1)$$

where α^γ ($\gamma = x, y, z$) are parameters defined by

$$\sum_j A_{ij}^\gamma J_{ij}^\gamma = \alpha^\gamma \sum_j J_{ij}^\gamma .$$

In the above equation, the first term represents an effective anisotropy energy and the second term represents the interaction of i th spin with the correlated effective field. The static magnetic susceptibility is then given by⁸

$$\chi^\gamma(\bar{q}) = \frac{1}{kT} \left[\langle \langle \mu_i^\gamma; \mu_i^\gamma \rangle \rangle + \frac{2[J^\gamma(\bar{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle \mu_i^\gamma; S_i^\gamma \rangle \rangle^2}{kT - 2[J^\gamma(\bar{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle S_i^\gamma; S_i^\gamma \rangle \rangle} \right] , \quad (2)$$

where

$$J(\vec{q}) = \sum_{\text{NN,NNN}} J e^{i\vec{q} \cdot \vec{r}} \quad (3)$$

$$\vec{\mu}_i = (\kappa \vec{L}_i + 2\vec{S}_i) \mu_B \quad (4)$$

$$\langle \langle \vec{A} \cdot \vec{B} \rangle \rangle = \langle \vec{A} \cdot \vec{B} \rangle - \langle \vec{A} \rangle \langle \vec{B} \rangle \quad (5)$$

$$\langle \vec{A} \cdot \vec{B} \rangle = \sum_n \rho_n \left[A_{nn} B_{nn} + kT \sum_{m \neq n} \frac{A_{mn} B_{nm} + A_{nm} B_{mn}}{E_m - E_n} \right] \quad (6)$$

$$\langle \vec{A} \rangle = \sum_n \rho_n A_{nn} \quad (7)$$

ρ_n denotes the probability of occupation of the eigenstate $|n\rangle$ of the effective Hamiltonian (1). Now summing over (\vec{q}) in the first Brillouin zone, we

$$\alpha^\gamma = \sum_{\vec{q}} \frac{J^\gamma(\vec{q}) kT}{kT - 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle} / \sum_{\vec{q}} J^\gamma(0) \quad (9)$$

which is computationally more convenient than the expression used by Lines.⁸ Equation (9) is solved self-consistently through Eqs. (5)–(7).

For the compounds of interest $\text{Re}^{4+}(5d^3)$ has t_{2g}^3 strong field configuration and has 4A_2 ground state in octahedral field. This is a pure $S = \frac{3}{2}$ state and corresponds to Γ_8 after spin-orbit interaction. The first excited state is 2T_2 and it is situated at¹¹ 7060 cm^{-1} above the ground state. At low temperatures (0–30 K) the excited state does not contribute to magnetic properties as it is high above the ground state taking kT as a yardstick. Thus essentially we have a $S = \frac{3}{2}$ system and magnetic moment operator is given by

$$\mu_i^\gamma = g \mu_B S_i^\gamma \quad (10)$$

where g is the spectroscopic splitting factor. The susceptibility given by (2) then reduces to

$$\chi^\gamma(\vec{q}) = \frac{g^2 \mu_B^2 \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle}{kT - 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle} \quad (10)$$

$$H_i^0(\text{eff}) = H_i - (12J_1 + 6J_2) [\alpha^x (S_i^x)^2 + \alpha^y (S_i^y)^2] - (8J_1 + 12J_2) (\langle S_i^x \rangle_0 + \langle S_i^y \rangle_0) + (24J_1 + 12J_2) (\alpha^x \langle S_i^x \rangle_0 + \alpha^y \langle S_i^y \rangle_0) \quad (12)$$

where J_1 and J_2 are the isotropic nearest- and next-nearest-neighbor exchange constants and H_i is the Hamiltonian containing the free-ion term, interelectronic Coulomb and exchange interaction, spin-orbit, and crystal-field effects. For Heisenberg symmetry of exchange interaction, the effective Hamiltonian becomes

$$H_i^0(\text{eff}) = H_i - (12J_1 + 6J_2) [\alpha^x (S_i^x)^2 + \alpha^y (S_i^y)^2 + \alpha^z (S_i^z)^2] - (8J_1 + 12J_2) (\langle S_i^x \rangle_0 + \langle S_i^y \rangle_0) + (24J_1 + 12J_2) (\alpha^x \langle S_i^x \rangle_0 + \alpha^y \langle S_i^y \rangle_0) \quad (13)$$

have,

$$\sum_{\vec{q}} kT \chi^\gamma(\vec{q}) = N \langle \langle \mu_i^\gamma \cdot \mu_i^\gamma \rangle \rangle + \sum_{\vec{q}} u^\gamma(\vec{q}) \quad (8)$$

where

$$u^\gamma(\vec{q}) = \frac{2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle \mu_i^\gamma S_i^\gamma \rangle \rangle^2}{kT - 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)] \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle}$$

Again from fluctuation theorem we have

$$\sum_{\vec{q}} kT \chi^\gamma(\vec{q}) = N \langle \langle \mu_i^\gamma \cdot \mu_i^\gamma \rangle \rangle$$

Therefore, the condition $\sum_{\vec{q}} u^\gamma(\vec{q}) = 0$ has to be satisfied, and from this condition the correlation parameters are determined. Using this condition,

and the uniform molar susceptibility ($\vec{q} = 0$) is given by

$$\chi^\gamma = \frac{Ng^2 \mu_B^2 \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle}{kT - 2J^\gamma(0)(1 - \alpha^\gamma) \langle \langle S_i^\gamma S_i^\gamma \rangle \rangle} \quad (11)$$

The magnetic structure of the compounds under investigation is type I in which the moments are ferromagnetically aligned in XY plane and along Z direction the alignment alternates.³⁴ Thus each magnetic ion has eight nearest-neighbor spins parallel and four nearest-neighbor spins antiparallel. Out of six next nearest neighbors, all are parallel. This magnetic structure has been incorporated in the present calculation. With the consideration of XY as the symmetry of the exchange interaction, the zero-field effective Hamiltonian is written as

Here α 's are the correlation parameters to be determined self-consistently from the Eq. (9). In this equation, the sum over \vec{q} is converted into integration over the first Brillouin zone. In the calculation of susceptibility the experimental value^{1,16} of g ($g = 1.815$) is used. For XY symmetry of the Hamiltonian, $\alpha^x = \alpha^y \neq \alpha^z$ in both the paramagnetic and the ordered phases. As a result, $\chi^x = \chi^y \neq \chi^z$ in the above-mentioned phases. This anisotropy is due to XY symmetry of the Hamiltonian. For Heisenberg symmetry, of course, $\alpha^x = \alpha^y = \alpha^z$ and $\chi^x = \chi^y = \chi^z$ in the ordered phase but the correlation parameters and susceptibilities in different directions give identical results ($\alpha^x = \alpha^y = \alpha^z$, $\chi^x = \chi^y = \chi^z$) in the paramagnetic phase. The average susceptibility is calculated from

$$\chi_{av} = \frac{1}{3} (\chi^x + \chi^y + \chi^z) \quad (14)$$

The average susceptibility is required to compare the experimental powder susceptibility. The sublattice magnetization corresponds to either $\langle S_i^x \rangle_0$ or $\langle S_i^y \rangle_0$ as they are equal in the ordered phase and these thermal averages go to zero in the paramagnetic phase.

IV. RESULTS AND DISCUSSION

The magnetic structure of the compounds under investigation is shown in Fig. 1. As suggested by the neutron-diffraction experiment, magnetic moments lie in a plane perpendicular to one of the cubic axes. The moments in a plane have ferromagnetic alignment and those in adjacent planes are arranged antiferromagnetically. Introducing this type I magnetic structure in CEF calculation, the average susceptibilities are calculated for K_2ReCl_6 and K_2ReBr_6 using the experimental value of^{1,16} g ($g = 1.815$). The results are shown in Fig. 2. The results obtained by using XY symmetry of the exchange Hamiltonian are shown by full lines whereas those obtained by using Heisenberg symmetry of the exchange Hamiltonian

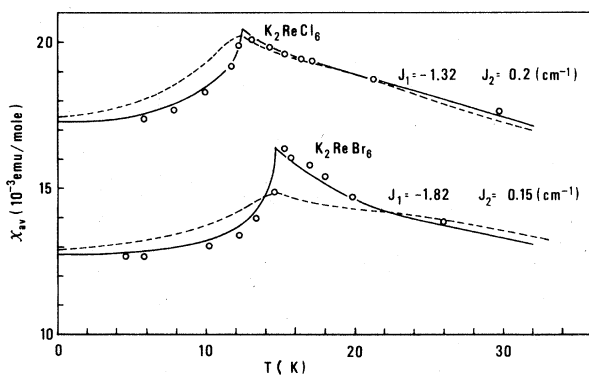


FIG. 2. Low-temperature magnetic susceptibilities.

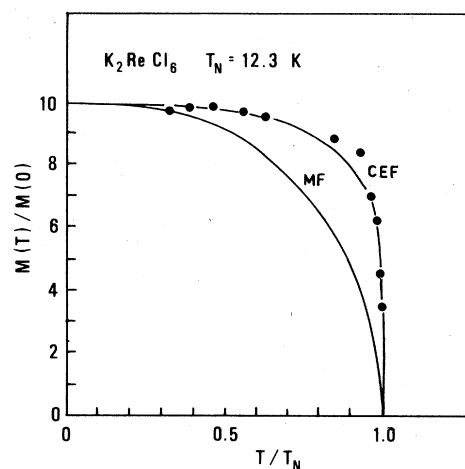


FIG. 3. Sublattice magnetization of K_2ReCl_6 .

are shown by broken lines. The parameters used in these calculations are $J_1 = -1.32 \text{ cm}^{-1}$, $J_2 = 0.20 \text{ cm}^{-1}$ for K_2ReCl_6 and $J_1 = -1.82 \text{ cm}^{-1}$, $J_2 = 0.15 \text{ cm}^{-1}$ for K_2ReBr_6 . The experimental results are shown by open circles. In Fig. 3 sublattice magnetization of K_2ReCl_6 is shown, where experimental results are shown by dots. The MF result is shown for comparison. The variation of correlation parameters with temperature is shown in Fig. 4 for a given set of parameters ($J_1 = -1.32 \text{ cm}^{-1}$, $J_2 = 0.20 \text{ cm}^{-1}$). The susceptibilities in different directions for the same set of parameters are shown in Fig. 5. The calculations of sublattice magnetization, correlation parameters and susceptibilities are referred to the Hamiltonian (12). The anisotropy due to XY symmetry of the exchange Hamiltonian is evident from Fig. 4 and Fig. 5.

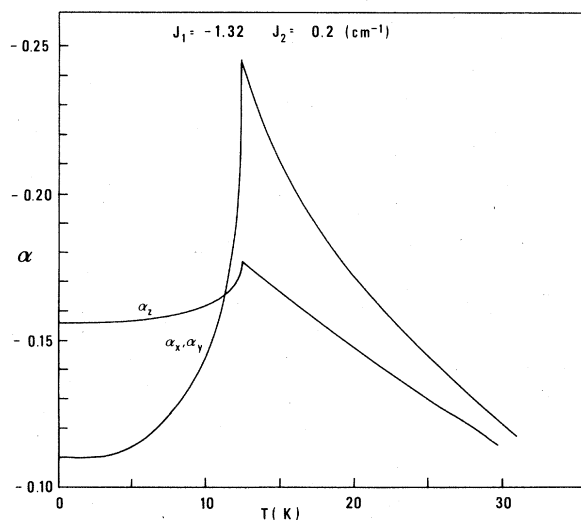


FIG. 4. Variation of correlation parameters with temperature.

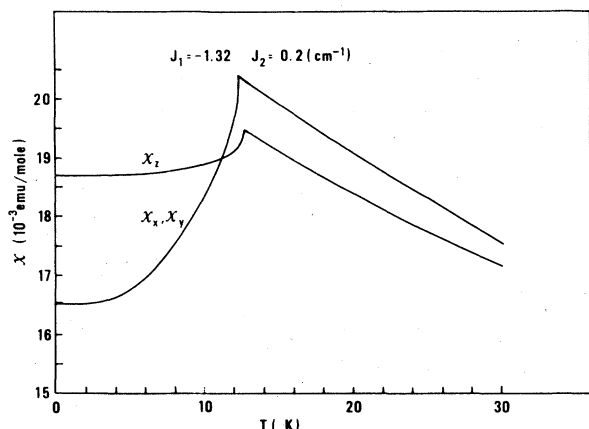


FIG. 5. Variation of susceptibilities with temperature.

It is evident from Fig. 2 that an excellent agreement between theoretical and experimental results is obtained if we assume XY symmetry of the exchange Hamiltonian. Figure 3 also shows a good agreement of experimental results of magnetization. By comparing the results of the present calculation with the calculation of MF theory shown in Fig. 3, it is evident that CEF theory is better than MF theory. This is because the fluctuation effect which is completely neglected in MF theory has been considered in CEF theory. Therefore, though MF explains the qualitative behavior of magnetization in the ordered phase, it fails to reproduce the experimental results near the transition temperature, where fluctuation becomes important. On the other hand, CEF theory explains the magnetic behavior near the critical temperature

better than does the MF theory. That the fluctuation becomes important and pronounced in the critical region is evident from Fig. 4. Since no single-crystal measurements have been done so far, the anisotropic susceptibilities shown in Fig. 5 cannot be compared. The powder susceptibilities are, therefore, compared with the average susceptibilities as shown in Fig. 2.

The nature of the order parameter as shown in Fig. 3 might indicate a first-order phase transition which has to be confirmed by hysteresis. This is in agreement with the remarks made by Benguigui³⁵ on CEF theory as a self-consistent phonon approximation^{36,37} and the generalized spherical model of Brout.³⁸ This type of first-order phase transition is called fluctuation-induced first-order phase transition introduced by Dzyaloshinskii, Brazovskii, and Kulkarenko^{39,40} and also by Bak, Mukamel, and Krinsky.⁴¹⁻⁴³ They come to the conclusion that the transition becomes first order when there is no stable fixed point in the renormalization-group calculation. Thus the CEF theory gives a good account of the magnetic behavior of the antiferromagnetic substances under investigation. For any realistic theory of antiferromagnetism in insulators, one needs to know the J integrals and a tractable model Hamiltonian. Towards this end one can work with this theory along with the other existing theories and show the CEF to be a good theory.

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