

NEW METHOD FOR THE DETERMINATION OF EXCHANGE INTERACTIONS

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The amplitude and the sign of exchange interactions in magnetic insulators are generally obtained by comparing the data of one (or more) experiment with the results of a statistical theory (molecular-field theory, Bethe-Peierls-Weiss theory, spin waves, \dots).¹ Let us present a new method, largely model independent, which gives in some cases the amplitudes of these interactions with a good accuracy.

We consider in the following a cubic insulator, with one kind of magnetic ion. We assume that the coupling is of the Heisenberg type, $-2J_{ij}\vec{S}_i \cdot \vec{S}_j$. The paramagnetic Curie temperature is given by

$$\theta_p = \{2S(S+1)/3k\} \sum_l z_l J_l \quad (1)$$

where J_l is the exchange interaction between l th-nearest neighbors and z_l the number of l th-nearest neighbors.

This relation is rigorous, but the results deduced from its application are generally not satisfactory because it is valid only in the limit of very high temperature, where the temperature-independent susceptibility terms² and the variation of the exchange integrals due to the thermal expansion and the lattice vibrations³ should be taken into account.

We limit ourselves to the case where only one kind of interaction, J , plays a role in the calculation of the volume anomaly at absolute zero and the magnetic-ordering temperature.

The expression for the magnetic-ordering temperature T_c , in terms of J , depends on the statistical model. In all models, however, the critical temperature T_c is directly proportional to the exchange constant J .¹ Therefore, we have

$$(d \ln T_c / d \ln V)_{T_{c,1}} = (d \ln |J| / d \ln V)_{T_{c,1}}, \quad (2)$$

where $T_{c,1}$ is the magnetic-ordering temperature at atmospheric pressure. From $K T = T_{c,1}$, the lattice compressibility at the temperature $T_{c,1}$ and from the variation of T_c with pressure, we can deduce

$$\begin{aligned} (d \ln T_c / d \ln V)_{T_{c,1}} \\ = -(1/K_{T_{c,1}}) (d \ln T_c / d P), \quad (3) \end{aligned}$$

where we have neglected the variations of the volume due to the thermal expansion. The magnetic energy per unit volume, at absolute zero, is given by

$$W_m(T=0^\circ\text{K}) = -zN_0 S^2 |J|_{(T=0^\circ\text{K})}, \quad (4)$$

where z is the number of effective neighbors and N_0 the number of magnetic ions per unit volume at 0°K . This relation is a consequence of the description of the fundamental magnetic state; it is rigorous in the molecular-field model, and is very nearly valid in the spin-wave model.⁴ The magnetic volume anomaly at absolute zero results from the minimization of the sum of the elastic and magnetoelastic energies.³

Experimentally, we can obtain the value of ΔV_0 , the difference between V_0 at 0°K and the volume V_0' which should be observed in the absence of any magnetoelastic interaction. We evaluate V_0' by extrapolating to absolute zero the empirical relation between the volume of the lattice and the temperature (for instance with a Debye-type law). The volume anomaly can be deduced from the variation of the magnetic energy with the volume, by the relation⁵

$$\Delta V_0 / V_0 = -K_0 (dW_m / dV)_{(T=0^\circ\text{K})}, \quad (5)$$

where K_0 is the isothermal lattice compressibility at 0°K , and W_m is given by expression (4). We assume that the relative variation $(1/J)dJ/dV$ and the lattice compressibility do not depend on temperature. Consequently, we deduce from the relations (2)-(5)

$$|J|_{(T=0^\circ\text{K})} = - \left(\frac{1}{zN_0 S^2} \right) \frac{\Delta V_0}{V_0} \left[\frac{1}{T_c} \frac{dT_c}{dp} \right]^{-1}. \quad (6)$$

Let us consider the magnetic materials MnO, MnS, and EuO, which have the NaCl-type crystal structure, and S-state magnetic ions. The ferromagnetic coupling between nearest-neighbor Eu^{++} ions is by far the largest one in EuO^6 ; for MnO and MnS, we assume that the volume anomaly and the magnetic-ordering temperature are proportional to the value of the antiferromagnetic-exchange interaction between the next-nearest-neighbor Mn^{++} ions.⁷ For MnO and MnS, $z=6$ and $S=\frac{5}{2}$; for EuO, $z=12$ and $S=\frac{7}{2}$. We present

Table I. Magnetic-ordering temperature, volume anomaly, number of magnetic ions per cm^3 at 0°K , variation of magnetic-ordering temperature with pressure, and values of exchange interactions obtained from present method, molecular-field, spin-wave, and Green's function theories for EuO, MnO, and MnS.

	T_c ($^\circ\text{K}$)	$\Delta V_0/V_0$ (10^{-3})	N_0 (10^{22} cm^{-3})	dT_c/dP ($10^{-3} \text{ }^\circ\text{K bar}^{-1}$)	$ J $ ($^\circ\text{K}$) calculated using present method	J ($^\circ\text{K}$) calculated using molecular-field theory	J ($^\circ\text{K}$) calculated using spin-wave theory	J ($^\circ\text{K}$) calculated using Green's-function theory
EuO	70	-2.4 ^a	2.94	0.37 ± 0.1 ^d	0.76 ± 0.2	0.65 ^f	0.750 ± 0.0025 ^h	
MnO	120	-3.8 ± 0.4 ^b	4.60	0.30 ± 0.02 ^e	6.4 ± 1.1	-3.5 ^g		-5.5 ⁱ
MnS	150	-4.75 ± 0.5 ^c	2.82	1.20 ± 0.02 ^c	4.1 ± 0.5	-4.4 ^g		-6.25 ^j

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in Table I the experimental values of $\Delta V_0/V_0$, N_0 , and dT_c/dp for these compounds and the values of the exchange interactions which are deduced from them. We also give the results obtained from the molecular-field theory, and the Green's-function random phase approximations, or the spin-wave theory.

For MnO the method proposed gives a result which is in much better agreement with that obtained using the random phase approximation than the one obtained using the molecular-field theory. For MnS the value obtained from our method is close to the molecular-field value. For EuO, our result is very close to the spin-wave theory value. Because of the relative inaccuracy of the determination of the various constants used, it is not possible to assert that these values are more precise than those given by the other methods. However, this treatment is more exact than the molecular-field theory

and more straightforward than spin-wave or Green's-function analysis.

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