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A Theoretical Model for the Non-linear Susceptibility of the Antiferromagnetic Systems

Bekir ÖZÇELİK, Kerim KIYMAÇ, Ahmet EKİCİBİL
Çukurova Üniversitesi, Fen Edebiyat Fakültesi,
Fizik Bölümü, 01330 Adana-TURKEY

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Abstract

In this study, we have developed a general theoretical model, based upon the simple mean field model of Néel, for the non-linear response of an antiferromagnetic system. The results indicate that the odd order derivatives, $(d^{2n+1}m_a)_0= (-d^{2n+1}m_b)_0$, where n=1,2,3......, will diverge and the even order derivatives, $(d^{2n}m_a)_0= (d^{2n}m_b)_0$ will vanish due to the symmetry of two sublattices, "a" and "b", forming the antiferromagnet. This model also supports our experimental results performed on two antiferromagnetic samples, namely, $Cs_2MnCl_4.2H_2O$ and $MnCl_2.4H_2O$ [1].

Key Words: Antiferromagnetism, AC – Susceptibility

1. Introduction

One of the signatures of a true phase transition is the observation of a divergence in the nonlinear susceptibility in the vicinity of the so-called critical temperature. In spin glasses, this divergency obeys the power law $\varepsilon^{-\gamma}$ at the freezing temperature T_f, where $\varepsilon = (T-T_f)/ T_f$ is the reduced temperature and γ is the critical exponent [2]. However, for ferromagnets and antiferromagnets the mean field theory predicts the relation $\chi_3 \alpha$ $-\chi_1^4$ in the paramagnetic region [3,4]. For ferromagnets the linear susceptibility χ_1 diverges at T_c , as does the third harmonic χ_3 [5]. For antiferromagnets, χ_1 is finite at T_N and hence χ_3 is also finite [1]. The latter result, relating to the magnitude of the nonlinear susceptibility in an antiferromagnet , can also be obtained by using the available simple mean field theory, namely the theory of Néel: above the Néel temperature T_N the two sublattices are completely equivalent and the theory simplifies to a calculation analogous to the ferromagnetic mean field theory [1,5]. However, below T_N the calculations are considerably more complicated but lead essentially to the same result as that found for $T>T_N$.

In this work, we have developed a general theoretical model, based upon a simple mean-field model of Néel, for the nonlinear response of an antiferromagnetic system, for temperatures above as well as below the critical temperature T_N. The result of this theoretical model agrees with our experimental observations appearing in [1].

2. Theoretical Model

Néel postulated two different internal molecular fields, acting on the individual spins arranged in two different sublattices, a and b. The fields acting on the spins in a and b sublattices can be given as

$$\begin{aligned} &h_a = h + \alpha m_a + \beta m_b, \\ &\text{and} \\ &h_b = h + \beta m_a + \alpha m_b, \end{aligned} \tag{1}$$

respectively. Here, h represents the external field, and the other two contributions are due to the two sublattices; the coefficients α and β represent the contributions from spins in the same sublattice and from those in the other sublattice, respectively. Furthermore, m indicates the reduced magnetisation defined as $m = M/M_0$ [5].

We now have the following expressions for an antiferromagnet with two sublattices:

$$\begin{aligned} m_a = B_a(h_a) = B_a(h + \alpha m_a + \beta m_b) = b_1 h_a + b_3 h_a^3 + ... \\ m_b = B_b(h_b) = B_b(h + \beta m_a + \alpha m_b) = b_1 h_b + b_3 h_b^3 + ..., \end{aligned} \tag{2}$$

where the factor g $\mu_b S/kT$ is absorbed into the definition of the Brillouin function B. In other words, the temperature dependence of m_a and m_b is now contained in the coefficients b_1, b_3 ... etc. of the expansion.

It is easy to show that the sum and difference of the sublattice magnetisations can be written as:

$$\begin{aligned} &m_a + m_b = \{2h + (\alpha + \beta)(m_a + m_b)\}\Gamma_+(h_a, h_b) \\ &\text{and} \\ &m_a - m_b = (\alpha - \beta)(m_a - m_b)\Gamma_-(h_a, h_b) \end{aligned} \tag{3}$$

in which

$$\begin{aligned} \Gamma_+ \quad &= b_1 + b_3(h_a^2 - h_a h_b + h_b^2) + b_5(h_a^4 - h_a^3 h_b + h_a^2 h_b^2 - h_a h_b^3 + h_b^4)... \\ \text{and} \\ \Gamma_- \quad &= b_1 + b_3(h_a^2 + h_a h_b + h_b^2) + b_5(h_a + h_a^3 h_b + h_a^2 h_b^2 + h_a h_b^3 + h_b^4)... \end{aligned} \tag{4}$$

By arranging Eq. 3 one gets

$$(m_a + m_b)\{1 - (\alpha + \beta)\Gamma_+\} = 2h\Gamma_+ \tag{5a}$$

and

$$(m_a - m_b)\{1 - (\alpha - \beta)\Gamma_-\} = 0. \tag{5b}$$

It is seen from Eq. 5b that the condition for the difference of the sublattice magnetisations being different from zero, i.e. for m_a–m_b $\neq 0$, is clearly 1- (α - β) Γ_- = 0, for all values of the applied field h. However, for h = 0 one finds from Eq (5a) that (m_a + m_b) = 0, excepts for 1- (α + β) Γ_+ = 0, implying m_a = m_b = 0, and thus Γ_-= Γ_+ = b_1, for all temperatures down to T_N given by 1- (α - β)b_1 = 0.

Now we can discuss the derivatives of m_a and m_b in an antiferromagnet. For the sake of clarity, we will use the notation $d^n m$ for $d^n m/dh^n$, and B' and B'', etc. for the derivatives of B with respect to the arguments of it.

The first derivative of m_a and m_b given by Eq. 2 is :

$$\begin{aligned} dm_a &= B'_a dh_a \\ dm_b &= B'_b dh_b. \end{aligned} \tag{6}$$

Taking the first derivatives of Eqs. 1 with respect to h and substituting the results into Eq. 6 one can obtain

$$\begin{aligned} (1-\alpha B'_a)dm_a - \beta B'_a dm_b &= B'_a; \\ -\beta B'_b dm_a + (1-\alpha B'_b)dm_b &= B'_b. \end{aligned} \tag{7}$$

If we take h = 0, then the derivatives B'_a and B'_b will be $(B'_a\ )_o$= $(B'_b\ )_o$ = B'_o for all the values of T, where $(B'_a\ )_o$ and $(B_b\ )_o$ are even functions of $(h_a)_o$ and $(h_b)_o$.

However, for T $\geq$ T_N, we know that B'_o= b_1 but for T $\leq$ T_N;

$$B'_o = b_1 + 3b_3(h_a)_o^2 + 5b_5(h_a)_o^4 + ... = \Gamma_+^o. \tag{8}$$

Now Eq. 7 becomes

$$\begin{aligned} (1-\alpha B'_o)(dm_a)_o - \beta B'_o(dm_b)_o &= B'_o \\ -\beta B'_o(dm_a)_o + (1-\alpha B'_o)(dm_b)_o &= B'_o. \end{aligned} \tag{9}$$

At temperatures above T_N (T $\geq$ T_N) the magnetisation of two sublattices is the same, then the solutions of Eq. 9 give

$$(dm_a)_o = (dm_b)_o = \frac{B'_o}{1-(\alpha+\beta)B'_o}. \tag{10}$$

For T< T_N the two equations are dependent, as 1 – (α - β)Γ_-^o = 0. One then finds from Eq. 9

$$(dm_a)_o - (dm_b)_o = \frac{B'_o}{\Gamma_-^o}\{(dm_a)_o - (dm_b)_o\}. \tag{11}$$

However, since (B'_o / Γ_-^o) < 1 for T < T_N , Eq. 11 implies that $(dm_a)_o$=$(dm_b)_o$, just as for the case of T > T_N ; i.e.,

$$(dm_a)_o = (dm_b)_o = \frac{B'_o}{1-(\alpha+\beta)B'_o}. \tag{12}$$

Therefore comparison of Eq. 10 and 12 implies that there is no jump or divergence at T_N, a situation compatible with experimental observations.

Taking the first and second derivatives of Eqs. 1 and inserting them into the second derivatives of Eqs. 2, one obtains

$$\begin{aligned}(1-\alpha B'_a)d^2m_a - \beta B'_a d^2m_b = B'_a(dh_a)^2,\\ -\beta B'_b d^2m_a + (1-\alpha B'_b)d^2m_b = B'_b(dh_b)^2.\end{aligned} \tag{13}$$

If h = 0, for T >T_N one can obtain $(B'_a)_o = (B'_b)_o = B'_o$ and $(B'_a)_o = (B'_b)_o = 0$, since $h_a = h_b = 0$. Hence, under these conditions the solutions of Eqs. 13 are $(d^2m_a)_o = (d^2m_b)_o = 0$.

On the other hand, for T< T_N, $(B'_a)_o$=(B'_b) = B'_o and

$(B'_a)_o$= - (B'_b) = B'_o= $6b_3(h_a)_o + 20b_5(h_a)^3_o$ +.... , since at these temperatures h_a = - h_b.

Therefore, for all T

$$(d^2m_a)_o = -(d^2m_b)_o = \frac{B'_o(dh_a)^2_o}{1-(\alpha-\beta)B'_o}. \tag{14}$$

This equation is zero for T > T_N and is not zero for T < T_N. The results for T < T_N is interesting, even though $d^2(m_a + m_b)_o = 0$. Since 1 – (α - β)Γ^o_- =0 and $B_o = \Gamma^o_+$, for T < T_N , Eq. 14 can be written as

$$(d^2m_a)_o = -(d^2m_b)_o = \frac{(\alpha+\beta)^2}{(\alpha-\beta)}\frac{3}{(h_a)_o}\{1+\frac{4b_5}{3b_3}(h_a)^2_o + ...\}(dm_a)^2_o. \tag{15}$$

From Eq. 15 it can be immediately seen that the second derivative of the sublattice magnetisation is inversely proportional to $(h_a)_o$. Therefore, $(d^2m_a)_o$= - $(d^2m_b)_o$ goes to minus infinity as $(h_a)_o \to 0$, in the vicinity of T $\uparrow$ T_N. This implies that the second order sublattice susceptibility diverge $(m_a)^{-1}_o$, or $(\chi^a_2)_{h=0}$ α $(-\varepsilon)^{-1/2}$. The implication of this divergent behaviour probably leads to divergence of the total susceptibility $(\chi^a_2 + \chi^b_2)_o = (\chi_2)_o$ when the symmetry of the two sublattice is broken, as for instance, in a randomly diluted antiferromagnet. Also, short range order correlations near T_N probably will lead to relatively strong second harmonic response.

Following the same procedure for the second harmonic derivation one can get a result for the third harmonic response of an antiferromagnet as

$$(d^3m_a)_o = (d^3m_b)_o = \frac{6b_3}{(b_1)^4}(dm_a)^4_o \quad \text{for } T > T_N \tag{16}$$

and,

$$(d^3m_a)_o = (d^3m_b)_o \approx \frac{60b_3 + 312b_5(h_a)^2_o + ...}{(b_1)^4}(dm_a)^4_o \quad \text{for } T < T_N. \tag{17}$$

The result for T > T_N Eq. 16 is the same as for the ferromagnetic case [5] and can be expected from the complete equivalence of the two sublattices. The result for T< T_N

shows the same proportionality to $(\mathrm{dm}_a)_o^4$ as for T > T_N but differs by a factor of 10, for small $(h_a)_o$. This discontinuous jump at T=T_N leads one to expect much stronger effects in the higher derivatives, although one should keep in mind that a discontinuity as a function of T does not necessarily lead to a discontinuity as a function of h.

Without giving a full calculation, we can inspect the following expressions for the fourth and fifth derivatives:

$$(1-\alpha B_o')(d^4m_a)_o - \beta B_o'(d^4m_b)_o = \{B_o''''(dh_a)_o^4 + 6B_o'''(d^2h_a)_o(dh_a)_o^2 + 4B_o''(d^3h_a)_o(dh_a)_o + 3B_o''(d^2h_a)_o^2\}$$

and

$$-\beta B_o'(d^4m_a)_o + (1-\alpha B_o')(d^4m_b)_o = -\{B_o''''(dh_a)_o^4 + 6B_o'''(d^2h_a)_o(dh_a)_o^2 + 4B_o''(d^3h_a)_o(dh_a)_o + 3B_o''(d^2h_a)_o^2\}.$$

One can conclude that the last terms on the right-hand sides will lead to a stronger divergence in the fourth derivative. Therefore the result for the total magnetisation, $d^4(m_a+m_b)$ will vanish due to the symmetry of two sublattices in a perfect antiferromagnet.

On the other hand for the fifth harmonics:

$$(1-\alpha B_o')(d^5m_a)_o - \beta B_o'(d^5m_b)_o = \{B_o'''''(dh_a)_o^5 + 10B_o'''(d^3h_a)_o(dh_a)_o^2\} \{10B_o''''(d^2h_a)_o(dh_a)_o^3 + 15B_o''(d^2h_a)^2(dh_a) + 10B_o''(d^2h_a)_o(d^3h_a)_o + 5B_o''(d^4h_a)_o(dh_a)_o\}$$

and

$$-\beta B_o'(d^5m_a)_o + (1-\alpha B_o')(d^5m_b)_o = \{B_o'''''(dh_a)_o^5 + 10B_o'''(d^3h_a)_o(dh_a)_o^2\} \{10B_o''''(d^2h_a)_o(dh_a)_o^3 + 15B_o''(d^2h_a)^2(dh_a)_o + 10B_o''(d^2h_a)_o(d^3h_a)_o + 5B_o''(d^4h_a)_o(dh_a)_o\}$$

Again the terms in the last brackets on the right hand sides of the expressions will show a strong divergencies for T = T_N, caused by the factors $(d^2h_a)_o$ and $(d^4h_a)_o$!

3. Conclusion

As a result for all odd order derivatives $(d^{2n+1}m_a)_o = (d^{2n+1}m_b)_o$, according to this model the total, 5^{th} and higher order susceptibilities will diverge for an antiferromagnet. It is to be expected, however that the prefactor for the divergent terms will be proportional to some (high) power of $(\mathrm{dm}_a\ )_o$ or to the linear susceptibility which of course is rather small.

On the other hand, the even order derivatives will vanish due to the symmetry of the two sublattices in a perfect antiferromagnet. But short range ordered clusters, just above T_N will, in general, not be symmetrical for these sublattices. Due to the critical speeding up of the relaxation time in antiferromagnets, this will lead to a strongly enhanced response in all derivatives.

We should emphasise here that our meanfield theory results appearing in this article support our measurements performed on two standard insulating antiferromagnetic compounds, $MnCl_2.4H_2O$ and $Cs_2MnCl_4.2H_2O$, published elsewhere [1]. In a perfect antiferromagnet the molecular fields of the different sites exactly cancel each other, therefore, the external field cannot couple to the magnetisation. However if antiferromagnet is diluted, this argument does not hold and a diverging nonlinear susceptibility appears at TN in an external field.

The third harmonics can be easily measured [1], and compared with our theory. However, it is further highly desirable to measure higher order susceptibilities, such as the fifth harmonic χ_5 , but one has to note that it will be very difficult to separate the higher order divergent terms of χ_n in the measured response χ_n [1,5].

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