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The dipole–dipole contribution to the magnetic propagator in the $REBa_2Cu_3O_{7-\delta}$ compounds

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Abstract. The dipole–dipole interaction is calculated for antiferromagnetic configurations found in the $REBa_2Cu_3O_{7-\delta}$ compounds, as a function of the spacing between ab planes. Crossover from three-dimensional to two-dimensional behaviour is demonstrated for both the ground state energy and the coefficients of the quadratic terms in the propagator.

1. Introduction

A large number of experimental studies have demonstrated the existence of bulk superconductivity in the $REBa_2Cu_3O_7$ layered compounds (RE = rare earth). Recently there has been considerable interest in the magnetic ordering of the rare earth ions at low temperatures. Table 1 summarizes some of the available experimental results for the superconducting and related non-superconducting compounds. With the notable exception of Pr the superconducting transition temperature is little affected by the particular RE and is typically in the range 92-95 K for the fully oxygenated compounds indicating that the coupling between the rare earths ions and the superconducting charge carriers must be, in some sense, weak. Antiferromagnetic ordering of the rare earth ions typically occurs at a Néel temperature O(1 K) (again a notable exception is Pr). Neutron scattering experiments have determined the antiferromagnetic structure in $ErBa_2Cu_3O_7$ (figure 1(a)) and neutron scattering experiments and specific heat measurements provide strong evidence that the magnetic transition is in the universality class of the two-dimensional Ising model [8, 17, 24]. The two-dimensional nature of the transition is easily understood if the magnetic interaction is assumed to consist only of the exchange interaction since the lattice spacing in the c direction is approximately three times that in the b and a directions. However at ordering temperatures O(1 K) we expect a significant contribution from the magnetic dipole–dipole interaction which is inherently long ranged in nature.

The nature of the ordering is less clear in the other rare earth compounds. As an example, specific heat measurements on superconducting $REBa_2Cu_3O_{7-\delta}$ with RE = Gd, Dy, Sm or Nd [3, 7, 8, 17, 25] appear to be well fitted by the solution of the two-dimensional Ising model. Neutron scattering experiments on these compounds indicate that the tendency to order three-dimensionally is much stronger than in Er and have a spin configuration below T_N which is unstable if only dipole–dipole forces are present.

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Table 1. Summary of experimental results. The letters (a), (b), (c) and (d) indicate the magnetic configuration where known: (a) The AAA configuration; (b) AFA configuration; (c) AAF configuration; (d) AFF configuration.

Rare earth	Compound	T_N	β	T_c
Pr	$PrBa_2Cu_3O_7$ [1]	17 K (a)		
Nd	$NdBa_2Cu_3O_7$ [2]	<0.5 K		≈90 K
	$NdBa_2Cu_3O_{6.9}$ [3]	0.5 K (a)	0.37±0.13	92 K
	$NdBa_2Cu_3O_{6.5}$ [3]	1.7 K		—
	$NdBa_2Cu_3O_{6.86}$ [4]	0.551±0.01 K (a)	0.553±0.18	88 K
Sm	$SmBa_2Cu_3O_7$ [2]	0.6 K		≈90 K
	$SmBa_2Cu_3O_{6.9}$ [3]	0.61 K		92 K
Gd	$GdBa_2Cu_3O_{\approx 6.7}$ [5]	2.22 K		90 K
	$GdBa_2Cu_3O_{7-\delta}$ [6]	2.6 K		90 K
	$GdBa_2Cu_3O_7$ [2]	2.2 K		≈90 K
	$GdBa_2Cu_3O_{7-\delta}$ [7]	2.24 K		93 K
	$GdBa_2Cu_3O_{7-y}$ [8]	2.21 K		≈92 K
	$GdBa_2Cu_3O_7$ [9]	2.23 K		94.8 K
	$GdBa_2Cu_3O_{7-\delta}$ [10]	2.24 K		94 K
	$GdBa_2Cu_3O_{7-\delta}$ [11]	2.24 K		96±1 K
	$GdBa_2Cu_3O_7$ [12]	2.22±0.07(a)	0.15	85.9 K
	$GdBa_2Cu_3O_{7-\delta}$ [13]	2.2 K		93 K
	$GdBa_2Cu_3O_{7-\delta}$ [13]	2.2 K		—
	$GdBa_2Cu_3O_{6.5}$ [14]	2.2 K (c)		40 K
	$GdBa_2Cu_3O_7$ [15]	2.25 K	0.16±0.01	93.5 K
	$GdBa_2Cu_3O_{<6.5}$ [15]	2.20 K	0.11±0.02	<1.8 K
	$GdBa_2Cu_3O_{7-\delta}$ [11]	2.24 K		96 K
	$GdBa_2Cu_3O_{6.14}$ [16]	2.27 K	0.33±0.03	—
	$GdBa_2Cu_3O_{7-\delta}$ [17]	2.29 K		91.1 K
	$GdBa_2Cu_3O_{7-\delta}$ [17]	2.23 K		—
Dy	$DyBa_2Cu_3O_7$ [20]	1.0±0.05 K (a)		95 K
	$DyBa_2Cu_3O_7$ [2]	0.92 K		≈90 K
	$DyBa_2Cu_3O_{7-\delta}$ [11]	0.8 K		92 ±1 K
	$DyBa_2Cu_3O_{7-\delta}$ [21]	1.51 K		92 K
	$DyBa_2Cu_3O_{7-\delta}$ [21]	0.95 K		—
	$DyBa_2Cu_3O_{7-\delta}$ [22]	0.95 K		92–93 K
	$DyBa_2Cu_3O_{6.95}$ [23]	0.90 K (a)		91 K
	$DyBa_2Cu_3O_{7-\delta}$ [17]	0.88 K		91.8 K
	$DyBa_2Cu_3O_{7-\delta}$ [17]	0.82 K		—
Ho	$HoBa_2Cu_3O_{7-\delta}$ [11]	<0.3 K		93 ±2 K
	$HoBa_2Cu_3O_{7-\delta}$ [22]	0.17 K		92–93 K
	$HoBa_2Cu_3O_{6.8}$ [23]	≈0. 14 K		93.5 K
Er	$ErBa_2Cu_3O_x$ [11]	0.6 K		92 ±2 K
	$ErBa_2Cu_3O_{7-y}$ [8]	0.60 K		≈92 K
	$ErBa_2Cu_3O_7$ [24]	0.618 K (b)	0.122±0.004	92.8 K
	$ErBa_2Cu_3O_{7-\delta}$ [22]	0.59 K		92–93 K
	$ErBa_2Cu_3O_{7-\delta}$ [17]	0.62 K		92.5 K
	$ErBa_2Cu_3O_{7-\delta}$ [17]	0.61 K		—
	$ErBa_2Cu_3O_{7-\delta}$ [18]	0.5 K (d)		88 K
	$ErBa_2Cu_3O_{7-\delta}$ [19]	0.55 ± 0.02 K		88 K
Yb	$YbBa_2Cu_3O_{7-\delta}$ [11]	—		91±1 K

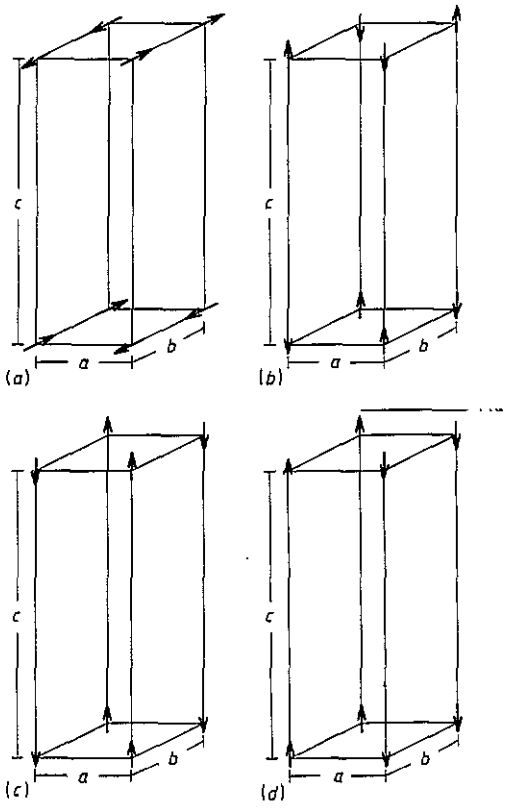

Figure 1. Schematic representations of (*a*) AFA, (*b*) AAA, (c) AAF and (*d*) AFF referred to in the text.

This apparent ambiguity in the determination of the effective dimensionality of the magnetic interactions manifests itself also in the critical exponent β associated with the order parameter. From table 1 we see that the measured values of the critical exponent β range from from 0.553 ± 0.18 K, characteristic of a Landau model, in the case of Nd [4] to a value of 0.11 ± 0.02 K, characteristic of a 2D Ising model, in the case of Gd [15]. In view of this a precise knowledge of the role of the long ranged dipole–dipole interaction and its dependence on the lattice parameters is desirable. We present a detailed calculation of the (Fourier transform of the) dipole–dipole contribution to the magnetic Hamiltonian.

In section 2 we briefly describe how the antiferromagnetic dipole lattice sum may be performed efficiently by means of the Jacobi theta functions. Section 3 summarizes the numerical results. The consequences for the layered rare earth compounds are discussed in section 4.

2. Dipolar lattice sums

For a particular magnetic configuration the interaction energy can be written in terms of the magnetic order parameter $\sigma(q)$ as

$$E = \frac{(g\mu_B)^2 s(s+1)}{2} n \sum_q \sigma^\alpha(-q)\sigma^\beta(q)A^{\alpha\beta}(q) \qquad (1)$$

(n = the number density of magnetic ions) where the definitions of the order parameter and the resultant propagator $A^{\alpha\beta}(q)$ depend upon the magnetic configuration under consideration. (It may be shown that a screening of the dipole–dipole interaction occurs due to the presence of the superconducting state. This effect is discussed in detail elsewhere [26]. Since the effect is expected to be very small for antiferromagnetic systems we neglect it here.)

It has previously been noted that the lattice sum required to evaluate the propagator for a ferromagnetic configuration may be evaluated efficiently by expressing it in terms of the Jacobi theta function [27]

$$\Theta_3(z;X) = \sum_{n=-\infty}^{\infty} \mathrm{e}^{\pi(2nz-n^2X)}. \tag{2}$$

For the ferromagnetic system on an orthorhombic lattice

$$\boldsymbol{\sigma}(\boldsymbol{q}) = \sum_n \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_n}\boldsymbol{S}(\boldsymbol{R}_n) \tag{3}$$

$$\begin{aligned} A^{\alpha\beta}(\boldsymbol{q}) &= -\lim_{|\boldsymbol{x}|\to 0}\frac{\partial^2}{\partial x_\alpha \partial x_\beta}\sum_n \frac{\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_n}}{|\boldsymbol{R}_n-\boldsymbol{x}|} \\ &= -\frac{1}{\Gamma(1/2)}\int_0^\infty \frac{\mathrm{d}y}{\sqrt{y}}\mathrm{e}^{-x^2y}\left(1-\prod_j \Theta_3\left(\frac{\mathrm{i}q_ja_j}{2\pi}+\frac{x_ja_jy}{\pi};\frac{a_j^2y}{\pi}\right)\right). \end{aligned} \tag{4}$$

In the region $y \to 0$ the sum is made rapidly convergent by use of the Jacobi imaginary transform

$$\Theta_3(z;X) = \frac{\mathrm{e}^{\pi z^2/X}}{\sqrt{X}}\Theta_3\left(\frac{z}{\mathrm{i}X};\frac{1}{X}\right). \tag{5}$$

In principle the lattice sum may be performed for an antiferromagnetic system by breaking the lattice into sublattices such that the spin arrangement is ferromagnetic on each sublattice. The method described above is then generalized, as described in [27] for non-orthorhombic ferromagnetic lattices, and the sum performed for each sublattice. However a more efficient procedure is to express the antiferromagnetic sum in terms of the Jacobi theta function of the fourth kind

$$\Theta_4(z;X) = \sum_{n=-\infty}^{\infty}(-1)^n\mathrm{e}^{\pi(2nz-n^2X)}. \tag{6}$$

For the antiferromagnetic system (AAA), shown in figure 1(b), in which spins alternate in sign along each lattice axis (for example, as in $GdBa_2Cu_3O_7$)

$$\boldsymbol{\sigma}(\boldsymbol{q}) = \sum_n (-1)^{(n_1+n_2+n_3)}\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_n}\boldsymbol{S}(\boldsymbol{R}_n) \tag{7}$$

$$\begin{aligned} A^{\alpha\beta}(\boldsymbol{q}) &= -\lim_{|\boldsymbol{x}|\to 0}\frac{\partial^2}{\partial x_\alpha \partial x_\beta}\sum_n (-1)^{(n_1+n_2+n_3)}\frac{\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_n}}{|\boldsymbol{R}_n-\boldsymbol{x}|} \\ &= -\frac{1}{\Gamma(1/2)}\int_0^\infty \frac{\mathrm{d}y}{\sqrt{y}}\mathrm{e}^{-x^2y}\left(1-\prod_j \Theta_4\left(\frac{\mathrm{i}q_ja_j}{2\pi}+\frac{x_ja_jy}{\pi};\frac{a_j^2y}{\pi}\right)\right). \end{aligned} \tag{8}$$

The sum is again made rapidly convergent in the small-y part of the integration range by using the appropriate Jacobi imaginary transform

$$\Theta_4(z;X) = \frac{e^{\pi z^2/X}}{\sqrt{X}}\Theta_2\left(\frac{z}{iX};\frac{1}{X}\right) \tag{9}$$

where $\Theta_2(z,X)$ is defined in the following manner

$$\Theta_2(z;X) = \sum_{n=-\infty}^{\infty} e^{\pi((2n-1)z-(n+1/2)^2X)}. \tag{10}$$

Besides the increased numerical efficiency, since only one lattice sum must be performed, this method is especially useful for the detailed study of the structure of the propagator (such as explicitly calculating the non-analytic terms). Other antiferromagnetic arrangements may also be treated by a single sum by an appropriate combination of the θ_3- and θ_4-functions. For example in the $ErBa_2Cu_3O_7$ system spins alternate in sign along the a and c axis but are uniform along the b axis (figure 1(a)), for this system (AFA)

$$\sigma(q) = \sum_n (-1)^{(n_1+n_3)} e^{iq\cdot R_n} S(R_n) \tag{11}$$

$$\begin{aligned} A^{\alpha\beta}(q) &= -\lim_{|x|\to 0}\frac{\partial^2}{\partial x_\alpha \partial x_\beta}\sum_n (-1)^{(n_1+n_3)}\frac{e^{iq\cdot R_n}}{|R_n - x|} \\ &= -\frac{1}{\Gamma(1/2)}\int_0^\infty \frac{dy}{\sqrt{y}} e^{-x^2 y}\left(1 - \Theta_4\left(\frac{iq_1a_1}{2\pi} + \frac{x_1a_1y}{\pi};\frac{a_1^2y}{\pi}\right)\right. \\ &\quad \left.\times \Theta_3\left(\frac{iq_2a_2}{2\pi} + \frac{x_2a_2y}{\pi};\frac{a_2^2y}{\pi}\right)\Theta_4\left(\frac{iq_3a_3}{2\pi} + \frac{x_3a_3y}{\pi};\frac{a_3^2y}{\pi}\right)\right). \end{aligned} \tag{12}$$

Corresponding results may be obtained for the (AAF) and (AFF) configuration shown in figure 1(c) and 1(d) respectively.

3. Magnetic propagator

The magnetic dipole–dipole interaction energy for each configuration may be readily calculated from the expressions in section 2 in the limit $|q| \to 0$. (Ground state energies for some antiferromagnetic arrangements have previously been calculated by Felsteiner [28] and our results are consistent with those.)

In the case of the antiferromagnetic configurations (e.g. AAA, AFA, AAF and AFF), shown in figure 1, the propagator $A^{\alpha\beta}(q)$ may be expanded in terms of the momentum as

$$A^{\alpha\beta}(q) = -\delta_{\alpha,\beta}\left(E_0^\alpha - \sum_i D_i^\alpha q_i^2\right) + D_1^{\alpha\beta}(1-\delta_{\alpha,\beta})q_\alpha q_\beta. \tag{13}$$

In the above expansion the alternating sign in the expressions (equations (6) and (11)) for the propagator $A^{\alpha\beta}(q)$, whose origin lies in the alternating nature of the antiferromagnetic spin configuration, results in a analytic expansion for the propagator

around $|q| = 0$ despite the long range character of the dipolar interaction. The coefficients in the expansion may then be obtained by taking appropriate derivatives of the expressions in section 2. Results are tabulated in table 2 for two systems of interest. Note the close agreement between the calculated value of the lowest energy E_2 and the observed transition temperature in the case of $ErBa_2Cu_3O_7$.

The dependance of the propagator on the spacing between the ab planes is best illustrated by the curves presented in figures 2 and 3, in which the the coefficients appearing in the above expansion of the propagator are plotted as a function of the ratio $r = c/a$ for the (AAA) and the (AFA) configurations respectively. In both figures the results for the corresponding 2D square lattice are shown for comparison. Note that the coefficients in the propagator are very close to the 2D results for $r = c/a > 2.5$, thus indicating a rapid crossover from three-dimensional to two-dimensional behaviour as a function of the spacing between the ab planes.

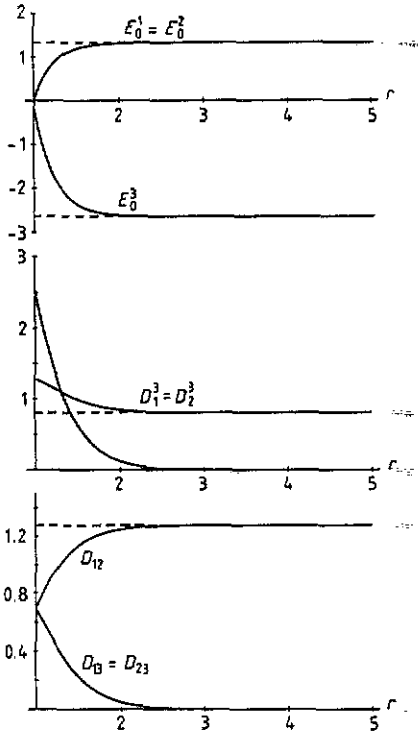

Figure 2. Graph showing the dependence of the coefficients given in the expansion of the propagator as a function of the ratio a/b for the AAA configuration with the spins aligned perpendicular to the ab plane.

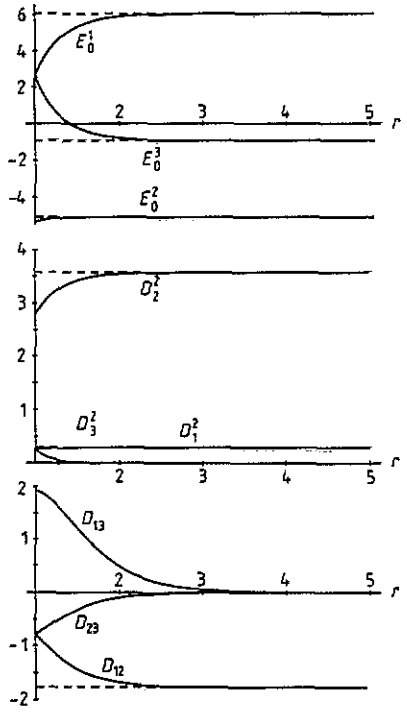

Figure 3. Graph showing the dependence of the coefficients given in the expansion of the propagator as a function of the ratio a/b for the AFA configuration with the spins aligned along the b axis.

4. Conclusions

Existing techniques developed to study ferromagnetic dipolar systems may be readily extended to the case of antiferromagentic systems. This allows the dipolar contribution

to the magnetic propagator to be computed efficiently with a high degree of precision. Typical results are tabulated in table 2.

Table 2. Estimates of the coefficents in the propagator in K. Notation is as in equation (13). The wavevector q is in units of inverse lattice spacing in the a direction. For $GdBa_2Cu_3O_7$ the free ion value of the saturated magnetic moment $p\mu_B$ with $p = g\sqrt{s(s+1)}(g = 2)$ is used. For $ErBa_2Cu_3O_7$ the experimentally determined value [18] of $p = 4.8$ is used.

Coefficent	$GdBa_2Cu_3O_7$	$ErBa_2Cu_3O_7$
E^1	0.445 53	0.749 70
E^2	0.445 53	−0.627 18
E^3	−0.891 06	−0.122 52
D_1^1	−0.974 53	−0.300 22
D_2^1	0.702 10	−0.328 99
D_3^1	0.000 46	0.007 19
D_1^2	0.702 10	0.037 96
D_2^2	−0.974 53	0.440 87
D_3^2	0.000 46	0.000 00
D_1^3	0.272 43	0.262 26
D_2^3	0.272 43	−0.111 88
D_3^3	−0.000 93	−0.007 19
D^{12}	0.430 07	−0.217 87
D^{13}	0.000 39	0.006 43
D^{23}	0.000 39	−0.000 76

In contrast to the situation that pertains in the case of a dipolar ferromagnetic system the momentum dependence of the propagator in the dipolar antiferromagnetic case does not contain a non-analytic term at $q \to 0$ [29]. This implies that the critical properties will be characteristic of the same universality class as a system with short range interactions only.

Results presented in section 3 show that in the case of the tetragonal lattice the propagator closely approximates the propagator for the corresponding 2D lattice when the ratio $r = a/c > 2.5$ for both spin configurations considered. This is somewhat surprising given the long range character of the dipolar interaction and differs substantially from the situation that pertains in the ferromagnetic case. In particular it implies that the layered systems studied experimentally will be in the universality class of the two-dimensional Ising model (except in a very small range of reduced temperature) despite the presence of dipolar interactions. This is completely consistent with experimental studies on $ErBa_2Cu_3O_7$ [24]. (The modification to the ground state energy arising from the screening of the dipolar interaction due to the presence of the superconducting state has been calculated elsewhere [26] and shown to be negligable in the antiferromagnetic case. We therefore do not expect it to modify our conclusions here.) The range of reduced temperature in which the crossover to three-dimensional behaviour will be seen may be estimated from the difference in the ground state energy of the physical system and the ground state energy of the two dimensional system. From this we expect that crossover to three-dimensional behaviour will only occur in a reduced temperature range $\Delta t \leq 0.03$.

As mentioned in the introduction, the effective dimension of the remaining compounds is less clear. Specific heat data for $GdBa_2Cu_3O_7$, $DyBa_2Cu_3O_7$, $NdBa_2Cu_3O_7$

and $SmBa_2Cu_3O_7$ [17,7,8,3,25] appear to be described by the solution of a two-dimensional Ising model. However neutron diffraction experiments appear to indicate a much stronger three dimensional ordering in these systems than in the corresponding Er compound (e.g. [16,20]). Moreover, as the data presented in table 1 indicate, the measured values for the critical exponent for the order parameter yield a range of values that encompass both 2D and 3D critical exponents.

Despite the apparent lack of a consistent picture with regard to the effective dimensionality of these systems, the above results, together with the fact that the (AAA) state found in these compounds is unstable with respect to the (AFA) state for a system with dipole–dipole interactions only, suggest that a further mechanism contributes to the magnetic Hamiltonian. In order to obtain a consistent picture it would appear that there exists some interplane (and possibly intraplane) exchange coupling in addition to the dipolar interaction. The precise nature of the mechanism giving rise to an interplane coupling is however far from obvious and while a superexchange mechanism via the oxygen atoms [15,7] is a possible candidate, there is no experimental evidence to substantiate such a hypothesis. Indeed it is difficult to discern any systematic behaviour either in the value of the Néel temperature or in the nature of the specific heat in the vicinity of the magnetic transition from existing experimental studies on the effect of the oxygen content on the magnetic properties of the rare earth ions [15,22,3,17,7].

The questions surrounding the interpretation of the experimental data concerning the nature and critical properties of the magnetic order parameter and the role of the oxygen content may arise from the fact that the systems of interest exhibit a crossover behaviour from 2D to 3D behaviour (and possibly Ising-like to XY or Heisenberg as has been suggested elsewhere [17]). The range of the crossover would obviously depend critically on the strength and nature of the interplane coupling. This implies that the determination of the critical properties of such systems from the experimental data would depend crucially on the temperature range used in the analyses and on the sample preparation, since this, one presumes, would modify the strength of the interplane coupling.

The picture that emerges regarding the magnetic properties of the $REBa_2Cu_3O_{7-\delta}$ compounds is far from clear. Based on the work presented here and the experimental studies cited in table 1 it is apparent that while the dipolar interaction is important it cannot account for the three-dimensional characteristics observed in many of these compounds. Careful determinations of the other critical exponents, in addition to β, would help to establish the extent to which these compounds can be regarded as effectively two-dimensional or three-dimensional.

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