Monte Carlo study of two-dimensional Ising dipolar antiferromagnets as a model for rare-earth ordering in the *R*-Ba-Cu-O compounds (*R*=rare earth)

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The two-dimensional Ising model with dipolar interactions is studied using Monte Carlo simulation. In the case of a pure dipolar interaction, estimates are obtained for the reduced transition temperature, $\theta_n = 2a^3 k_B T_n/\mu_{\rm eff}^2$ for two particular spin alignments in which the spins are aligned parallel to and perpendicular to the plane, respectively. A comparison with measurements made on $R\mathrm{Ba_2Cu_3O_{7-\delta}}$ (R=Nd, Dy, and Er) and $R\mathrm{Ba_2Cu_4O_8}$ (R=Dy and Er) show that the calculated Néel temperature falls consistently below that observed experimentally. The model is generalized to include an isotropic intraplanar nearest-neighbor exchange interaction and the Néel temperature calculated as a function of the strength of the exchange coupling for configurations in which the spins are aligned perpendicular to the plane. From this estimates of the exchange coupling are obtained for some of these materials. The results obtained from these calculations confirm that the long-range character of the dipolar interaction does not affect the universality class of the two-dimensional Ising antiferromagnet. Extensions of the present analysis are discussed.

I. INTRODUCTION

One of the more remarkable features of the class of high-temperature superconductors based on the Y-Ba-Cu-O compounds is the fact that the substitution of the Y ion by several of the rare-earth ions has little measurable effect on the superconducting transition temperature.[1–3] Moreover the appearance of a magnetic ordering of the rare-earth ions at low temperatures in certain of the *R*-Ba-Cu-O compounds means that a number of these belong to the class of materials, generally referred to as magnetic superconductors, in which superconducting and magnetic order coexist.[4–9] Other such materials exhibit a number of interesting phenomena associated with the interplay of the superconductivity and the magnetic interactions. While the coexistence of superconductivity with long-range magnetic order is undoubtably the most interesting feature of these rare-earth compounds, the magnetic properties of these intriguing compounds are proving to be interesting in their own right. In particular the highly anisotropic nature of these compounds combined with the various stable phases and oxygen stoichiometries that can be fabricated means that such materials provide an ideal opportunity in which to study magnetism in layered systems. It is also worth noting that recent experimental work suggests that the changes in the magnetic properties with oxygen content are closely correlated to the decrease in the superconducting transition temperature and the ultimate suppression of the superconductivity[10,11] with decreasing oxygen content.

There is a considerable wealth of experimental evidence that attests to the two-dimensional (2D) character of these compounds. A number of recent neutron-scattering experiments exhibit two-dimensional critical scattering close to the Néel temperature,[11–14] while in the case of $\mathrm{ErBa_2Cu_3O_7}$, the antiferromagnetic order parameter shows remarkable agreement with the result obtained from the 2D ising model.[12,13] In addition specific-heat measurements, for several of these compounds, are well described in terms of the two-dimensional Ising model.[15–17] While this may not be so surprising given the layered nature of this class of compounds, it is clear that the situation that pertains is far from straightforward. Not only does a certain amount of controversy surround several of the reported measurements and the conclusions drawn from them,[11,18] but low-temperature neutron scattering clearly exhibits three-dimensional magnetic order in some,[4,6,8,9,11–14,17,19] but apparently not all, of these compounds.[11,14] The questions surrounding the effective

dimensionality of these compounds is made more interesting by the fact that little is known about the mechanism giving rise to the interplanar coupling between the rare-earth ions.

In addition to the uncertainties surrounding the nature and origin of the interplanar coupling between the rare-earth ions a number of unresolved questions also persist with regard to the nature of the intraplanar coupling. Early work on these compounds revealed that the magnetic transition temperature scaled with the DeGennes factor $(g_J - 1)^2 J(J+1)$.[1,2] This leads one to suppose that the exchange interaction of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type is the dominant interaction in these compounds. However as Maple *et al.* have pointed out[2] this rather naive interpretation is misleading since it assumes that crystalline electric-field (CEF) effects are negligable, which they are not, and does not account for the data of Dunlap *et al.* on $GdBa_2Cu_3O_x$,[20] which yields a virtually identical transition temperature for both the orthorhombic (superconducting) and tetragonal (semiconducting) phases. Moreover calculations show that the dipolar interaction contributes significantly to the magnetic energy of the ground state in these compounds.[21,22]

One aspect of these compounds about which there is little controversy is the importance of the crystalline electric fields (CEF's) in determining the magnetic properties of the compounds. Both inelastic-neutron-scattering[10,23–28] and specific-heat[1,3,29–31] experiments show that the degeneracy of the rare-earth ions associated with the rotational invariance of the free ion is removed by the crystalline electric fields, with the result, in the case of the Kramers ions, that the ground state of the rare-earth ions is a doublet several meV below the first excited state. An exception to this is of course Gd, which has an orbital S-wave ground state and which, specific heat experiments indicate, retains the eightfold degeneracy of the free-ion ground state.[1,3,29] While the nature of the ground state of the individual rare-earth ions in these compounds is well established, the nature of the transition and, by implication, the character of the magnetic ground state appears to be strongly dependent on the oxygen content. Experiments on Nd (Ref. 17), Sm (Ref. 17), Dy (Ref. 29) and Er (Refs. 11, 29, and 30) all show a substantial rounding of the transition in the change from the orthorhombic (superconducting) phase to the tetragonal (insulating) phase. This has led several authors to postulate that the transition from orthorhombic to tetragonal induces a crossover from two-dimensional Ising-like behavior to spin-$\frac{1}{2}$ X-Y type behavior.[29,30] This is not an unreasonable assertion and may well be related to the change in the CEF ground state that arises as a consequence of the increased symmetry of the $4f$ wave function of the rare-earth ions in the tetragonal phase.[30] The fact that in Gd the magnetic transition is almost identical in both the orthorhombic and tetragonal phase[20] lends weight to this argument regarding the importance of CEF effects in accounting for this difference.

Much of the difficulty in trying to arrive at a systematic understanding of these compounds undoubtably stems from the fact that the magnetic interactions in these compounds are sufficiently complex that they are not easily accounted for within the framework of a single tractable theoretical model. While a comparison with existing theoretical studies provide some insight into certain aspects of the behavior of these compounds the limitations of the models used as the basis for these studies should be kept in mind. The effect of the dipolar interaction provides a case in point. While it is without question a fact that the dipolar interaction contributes significantly to the magnetic properties of these componds at low temperatures, little quantative work has been done on the magnetic properties of dipolar systems. For example, few reliable estimates, beyond those provided by mean-field theory, of the transition temperature and phase behavior of dipolar spin systems in two dimensions exist. Moreover studies that have been done indicate that the presence of the dipolar interaction can modify the magnetic properties in some very subtle ways that, not surprisingly, differ significantly from the predictions provided by mean-field studies.[32,33] In the present work we examine by means of Monte Carlo simulation the properties of a two-dimensional spin-$\frac{1}{2}$ Ising model on a square lattice. We consider the case in which the easy axis of magnetization is aligned perpendicular to the plane as well as the case in which the easy axis is aligned parallel to the plane. The interaction between the spins is assumed to consist of both a dipolar interaction and a nearest-neighbor exchange interaction. While such a model falls far short of accounting for the many facets exhibited by the R-Ba-Cu-O compounds it is nevertheless of obvious relevance in the interpretation of existing experimental studies. The results obtained from the analysis of such a model should serve to provide some bounds on the strength of the interplanar coupling as well as providing the basis for a more realistic modeling of these systems.

It is well known that the long-range character of the dipolar interaction presents a problem in the Monte Carlo simulation[34] which is, by necessity, carried out on a small finite system. In the context of the present work, earlier studies by Kretschmer and Binder[33] suggest that finite-size effects may best be treated by considering an infinite sytem of spins but restricting the allowed spin configurations to those which satisfy periodic boundary conditions. Imposing such a restriction allows us to specify the state in terms of a finite number of variables, namely the orientation of the spins in a single cell consisting of a finite number of spins, while the energy of each configuration may be calculated in terms of an effective interaction between the spins in this cell. This is discussed in more detail in Sec. II, in which we consider the case of a pure dipolar interaction (i.e., no exchange interaction).

Section III summarizes the results of the simulation studies obtained from Sec. II and provides a comparison with the experimental results. In Sec. IV the model is generalized to include an intraplanar exchange interaction and its effect on the transition temperature is examined. Section V contains a discussion of the results obtained in the present work and examines their impli-

cations. Possible generalizations of the model employed in the present analysis are considered.

II. THE HAMILTONIAN

In this section we consider the case of a two-dimensional spin-$\frac{1}{2}$ Ising model in which we include only the dipolar interaction between the spins. In the following section we will consider the more general case that includes both the dipolar interaction as well as a nearest-neighbor exchange interaction. The analysis of a spin system that includes a long-range interaction, such as the dipole-dipole interaction, by Monte Carlo simulation, is complicated by difficulties associated with the mapping of the system onto a lattice of finite extent.[33,34] In comparable systems that involve only a short-range interaction between the spins, finite-size problems may be overcome by applying suitable boundary conditions (such as cyclic boundary conditions) and then extrapolating, by finite-size analysis, to infinite systems. The extension of such techniques to the case of dipolar systems is complicated by the long-range character of the interaction. A comparison of various schemes by Kretschmer and Binder,[33] has shown that finite-size effects may best be treated by considering an infinite system of spins but restricting the allowed configurations to those whose spins satisfy the periodic boundary condition. We define the magnetic moment at the nth lattice site as

$$\boldsymbol{\mu}_n = \mu_{\text{eff}} \mathbf{S}_n \,, \tag{1}$$

where $\mathbf{S}_n$ is a unit vector (i.e., $|\mathbf{S}_n| = 1$) defining the orientation of the magnetic moment and μ_{eff} defines the effective magnitude of the magnetic moment. The periodic nature of the allowed spin configurations requires that

$$\mathbf{S}_n^\alpha = \mathbf{S}_{n'}^\alpha, \quad \text{where} \quad \mathbf{r}_{n'} = \mathbf{r}_n + \mathbf{G}, \tag{2}$$

where $\mathbf{r}_n$ denotes the position of the nth lattice site and the vector $\mathbf{G}$ satisfies the requirement that

$$\mathbf{G} = aL\,(g_x, g_y) \quad \text{for} \quad \left.\begin{matrix} g_x \\ g_y \end{matrix}\right\} \in \{0, \pm 1, \pm 2, \ldots\}\,, \tag{3}$$

where a denotes the lattice spacing (we have assumed a square lattice, $a = b$) and L is an integer characterizing the periodicity of the allowed configurations. The restriction imposed by the periodic boundary conditions given in Eq. (2) means that the state of the system may now be specified in terms of a finite number of variables, namely the L^2 spins in a $L \times L$ unit cell, allowing the analysis of the system by Monte Carlo simulation. The result for the infinite system is then obtained, using conventional finite-size scaling techniques, by extrapolation. This prescription will be followed in the work reported here.

The Hamiltonian for a two-dimensional spin system that interacts solely through the magnetic dipolar interaction may be written as

$$\mathcal{H} = \frac{\mu_{\text{eff}}^2}{2} \sum_{\substack{n \neq m \\ \alpha,\beta}} S_n^\alpha S_m^\beta \lim_{|\mathbf{r}| \to 0} \frac{\partial}{\partial x_\alpha} \frac{\partial}{\partial x_\beta} \frac{1}{|\mathbf{R}_{nm} - \mathbf{r}|}, \tag{4}$$

where α and β label the spin components and $\mathbf{R}_{nm} = \mathbf{r}_n - \mathbf{r}_m$.

In order to exploit the periodic character of the allowed spin configurations and hence reduce the infinite summation in the Hamiltonian given in Eq. (4) to one over a finite number of lattice sites, we introduce the reduced vector $\mathbf{r}_{nm}$ as

$$\mathbf{R}_{nm} = \mathbf{r}_{nm} + \mathbf{G}\,, \tag{5}$$

where $\mathbf{G}$ is chosen such that $\mathbf{r}_{nm}$ lies in the first unit cell. A schematic representation of an allowed spin configuration is given in Fig. 1.

The Hamiltonian may then be written as

$$\mathcal{H} = \frac{\mu_{\text{eff}}^2}{2} \sum_{\mathbf{G}} \sum_{\substack{n \neq m \\ \alpha,\beta}}^{L^2} S_n^\alpha S_m^\beta \lim_{|\mathbf{r}| \to 0} \frac{\partial}{\partial x_\alpha} \frac{\partial}{\partial x_\beta} \frac{1}{|\mathbf{G} + \mathbf{r}_{nm} - \mathbf{r}|}$$

$$+ (\mu_{\text{eff}})^2 \sum_{\mathbf{G} \neq 0} \sum_{n,\alpha}^{L^2} (S_n^\alpha)^2 \lim_{|\mathbf{r}| \to 0} \left(\frac{\partial}{\partial x_\alpha} \right)^2 \frac{1}{|\mathbf{G} + \mathbf{r}_n - \mathbf{r}|}\,. \tag{6}$$

The periodic nature of the allowed spin configurations allow us to rewrite the above expression as

$$\mathcal{H} = \frac{\mu_{\text{eff}}^2}{2} \sum_{\substack{n \neq m \\ \alpha,\beta}}^{L^2} S_n^\alpha S_m^\beta \lim_{|\mathbf{r}| \to 0} \frac{\partial}{\partial x_\alpha} \frac{\partial}{\partial x_\beta} \sum_{\mathbf{G}} \frac{1}{|\mathbf{G} + \mathbf{r}_{nm} - \mathbf{r}|}$$

$$+ \mu_{\text{eff}}^2 \sum_{n,\alpha}^{L^2} (S_n^\alpha)^2 \lim_{|\mathbf{r}| \to 0} \left(\frac{\partial}{\partial x_\alpha} \right)^2 \sum_{\mathbf{G} \neq 0} \frac{1}{|\mathbf{G} + \mathbf{r}_n - \mathbf{r}|}$$

$$= \left(\frac{N}{L^2} \right) \frac{\mu_{\text{eff}}^2}{2a^3} \left[Ca^3 + \sum_{\substack{n \neq m \\ \alpha,\beta}}^{L^2} S_n^\alpha \mathcal{W}^{\alpha\beta}(\mathbf{r}_{nm}) S_m^\beta \right]. \tag{7}$$

Thus we see that by restricting the allowed configurations to those that satisfy the periodic boundary condition given by Eq. (2), the Hamiltonian may be expressed as a sum over a finite number of sites which involve the effective interaction $\mathcal{W}^{\alpha\beta}(\mathbf{r}_{nm})$, which is defined as

$$\mathcal{W}^{\alpha\beta}(\mathbf{r}_{nm}) = a^3 \lim_{|\mathbf{r}|\to 0} \frac{\partial}{\partial x_\alpha}\frac{\partial}{\partial x_\beta}\sum_{\mathbf{G}}\frac{1}{|\mathbf{G}+\mathbf{r}_{nm}-r|}$$

$$= \lim_{\mathbf{x}\to 0} L^{-1}\frac{\partial}{\partial x_\alpha}\frac{\partial}{\partial x_\beta}\left[\sum_{G}\frac{\mathrm{erfc}\,(\eta\,|(\mathbf{x}-\mathbf{x}_{nm})/L+\mathbf{G}|)}{|(\mathbf{x}-\mathbf{x}_{nm})/L+\mathbf{G}|}+\sum_{\mathbf{G}\neq 0}\frac{\exp\left[2\pi i\mathbf{G}\cdot(\mathbf{x}-\mathbf{x}_{nm})/L\right]}{\mathbf{G}}\mathrm{erfc}\left(\frac{\pi\mathbf{G}}{\eta}\right)\right]. \tag{8}$$

The constant C, which appears in Eq. (7), arises from the interaction of a spin in the first unit cell with the spins at equivalent lattice sites and is given by

$$C = a^3\sum_{n,\alpha}\lim_{|\mathbf{r}|\to 0}\left(\frac{\partial}{\partial x_\alpha}\right)^2\sum_{\mathbf{G}\neq 0}\frac{1}{|\mathbf{G}+\mathbf{r}_n-\mathbf{r}|}. \tag{9}$$

The Hamiltonian given by Eq. (4) contains no information regarding the nature of the allowed spin states. In a magnetic solid the $2J+1$ degeneracy of the electronic states forming the $4f$ shell, associated with the rotational symmetry of the free ion, is typically removed by the perturbing effects of the crystalline electric fields (CEF's). In the case of Nd, Sm, Dy, and Er specific-heat data[1,3,29–31] together with inelastic neutron scattering[10,23–28] indicate that the ground state of these ions is a Kramers doublet several meV below the first excited multiplet with a magnetic moment whose magnitude and axis of orientation are determined by the wave function of the CEF ground state. Confining our attention to compounds comprising only those particular ions, we therefore restrict the orientation of the spin variable in the Hamiltonian defined in Eq. (7), to lie along the easy axis of magnetization, defined by the CEF ground state and assume that the magnitude of the magnetic moment μ_{eff} is equal to the ground-state magnetic moment of the rare-earth ion. Our description of these systems then reduces to a spin-$\frac{1}{2}$ Ising system, in which the orientation axis and the magnitude of the magnetic moment are determined from experimental data. For the purposes of the present analysis, two distinct spin alignments are considered, which we refer to as the in-plane alignment and the out-of-plane alignment. In the case of the out-of-plane alignment the spins are assumed to orient in the z direction, parallel to the c axis. This describes the nature of the magnetic ordering that is observed in the case of $DyBa_2Cu_3O_7$,[6,7] $DyBa_2Cu_4O_8$,[14] $NdBa_2Cu_3O_7$,[17,19] and $GdBa_2Cu_3O_7$.[8,35] In the case of the in-plane alignment the spins are assumed to orient in the y direction parallel to the b direction. This describes the various magnetic orderings that have been observed in both $ErBa_2Cu_3O_7$ and $ErBa_2Cu_4O_8$,[14] although it should be noted that the nature of the ordering between the planes, in the case of the $ErBa_2Cu_3O_7$ (Refs. 4, 9, 11–13, and 18) and $GdBa_2Cu_3O_7$, appears to be sample dependent, a fact that is attributed to the weak interplanar coupling in these compounds.

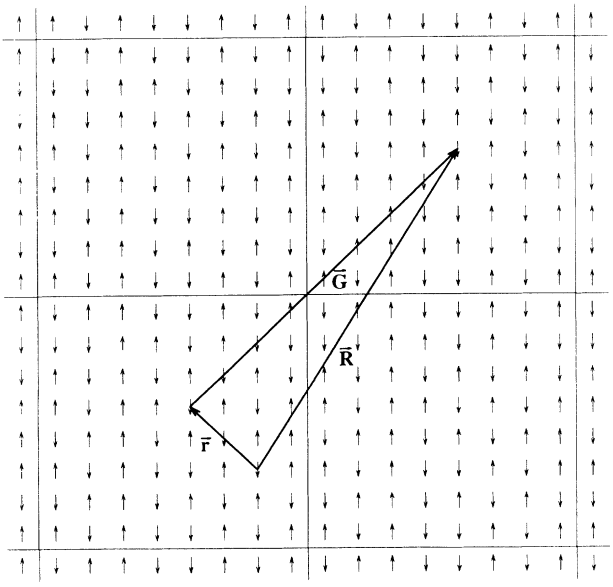

FIG. 1. Diagram showing a particular spin configuration that satisfies the periodicity requirement imposed by Eq. (2) with the vectors $\mathbf{r}_{nm}$ and $\mathbf{G}$ defined by Eq. (5).

In the present calculation, the effective interaction matrix $\mathcal{W}^{\alpha\beta}(\mathbf{r}_{nm})$, defined by Eq. (8), is calculated, for both the in-plane spin alignment ($\alpha=\beta=2$) and the out-of-plane spin alignment ($\alpha=\beta=3$) (using the symbolic manipulation program MATHEMATICA) prior to running the simulation and is stored in a "look up" table for use during the simulation. The reduced temperature scale θ used in the simulations is related to the temperature T in kelvins through the relation

$$\theta = \frac{2a^3k_BT}{\mu_{\mathrm{eff}}^2}. \tag{10}$$

Also we have neglected the small difference between the lattice spacing along the a and b axis and assumed a square lattice.

III. DISCUSSION OF RESULTS

Simulations were carried out on various size lattices, from $N=4\times 4$ up to $N=64\times 64$, and for various lengths of time. The normal Monte Carlo simulation on the larger of the lattices comprised 10000 initialization steps and typically 10^6–10^7 Monte Carlo steps. Normally an initial configuration was chosen from a previous run at a lower temperature (to prevent "freezing in" of high-temperature domain-wall formations),[36] although low-temperature simulations were initialized to

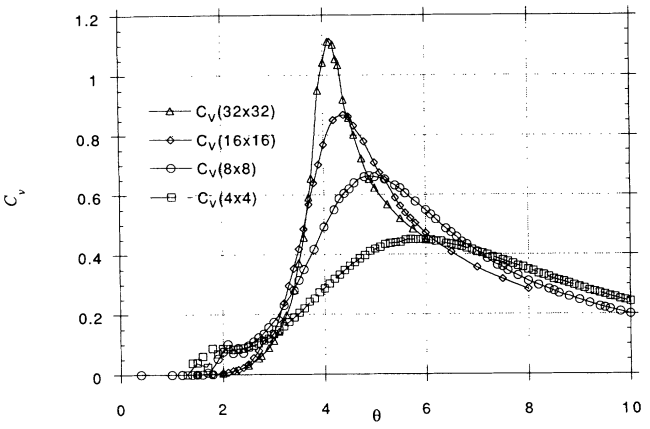

FIG. 2. Specific heat as a function of temperature for several values of L (in-plane spin configuration).

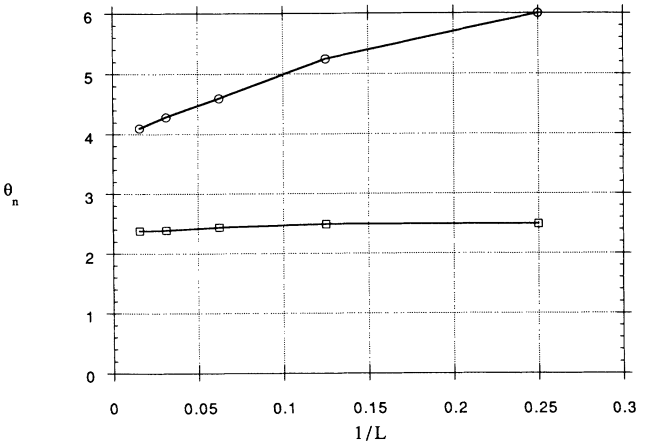

FIG. 4. Estimates of the Néel temperature as a function of L^{-1} for in-plane (○) and out-of-plane (□) spin configurations.

the groundstate with a larger number of initial steps to allow the system to reach equilibrium.

The results of the Monte Carlo simulations for the magnetic specific heat are shown graphically in Figs. 2 and 3 for the in-plane and the out-of-plane spin alignments, respectively, for several values of lattice size L. In both cases the resultant ground-state spin configuration corresponded to what is observed experimentally and gave a ground-state energy that was in excellent agreement with those obtained previously.[22]

Using the peak of the specific heat as the estimate of the Néel temperature $\theta_n(L)$ in the reduced temperature scale , we find the critical exponent $\nu \approx 1$ for both cases, where ν is defined by $\theta_n(L) \sim L^{-\nu}$. Values of θ_n calculated for different values of L are plotted in Fig. 4, for both the in-plane and the out-of-plane spin configurations. Extrapolation of our results to $L = \infty$ puts θ_n at 3.9 ± 0.1 for in-plane ordering and $\theta_n = 2.39 \pm 0.05$ for out-of-plane ordering. In order to make a comparison with experimental data values $2a^3 k_B T_n/\mu_b^2$ are plotted against μ_{eff}^2 in Figs. 5 and 6 for samples, for which both μ_{eff} and T_n are available. The figures correspond to the in-plane (Fig. 5) and out-of-plane (Fig. 6) spin alignments, respectively. The results obtained from the Monte Carlo simulations are represented by the solid line. From the comparison given in Figs. 5 and 6 we see that the results obtained from the Monte Carlo simulations are consistently lower than the experimental values. This implies that there must be some other interaction, besides the dipolar interaction, contributing to the magnetic ordering.

Finally we comment on the critical indices we obtain for this model. Using the corresponding susceptibility and order parameter data together with the relations,

$$M \sim (-t)^{\beta}(t \to 0^-) \sim L^{\frac{-\beta}{\nu}}(t = 0), \tag{11}$$

$$\Psi \sim (t)^{-\gamma}(t \to 0^+) \sim L^{\frac{\gamma}{\nu}}(t = 0), \tag{12}$$

the exponents are found to be $\beta/\nu = 0.2 \pm 0.1$, $\gamma/\nu = 1.9 \pm 0.1$ and $\beta/\nu = 0.14 \pm 0.10$ and $\gamma/\nu = 1.8 \pm 0.1$, for in-plane and out-of-plane spin alignments, respectively. The lack of precision is, in part, due to the curvature in the plots resulting from the small system sizes. Nonetheless, these numbers are in agreement with the expected result

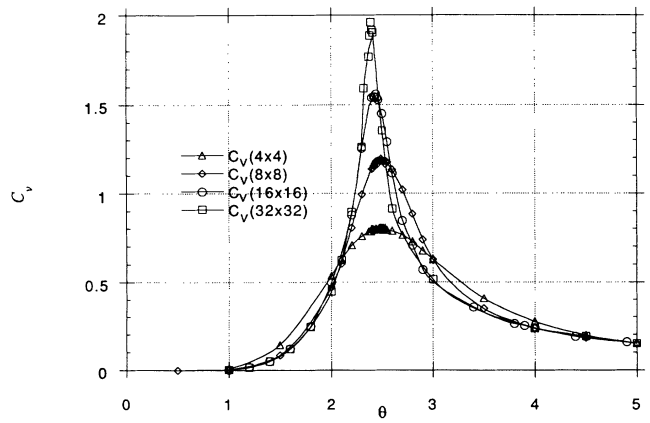

FIG. 3. Specific heat as a function of temperature for several values of L (out-of-plane spin configuration).

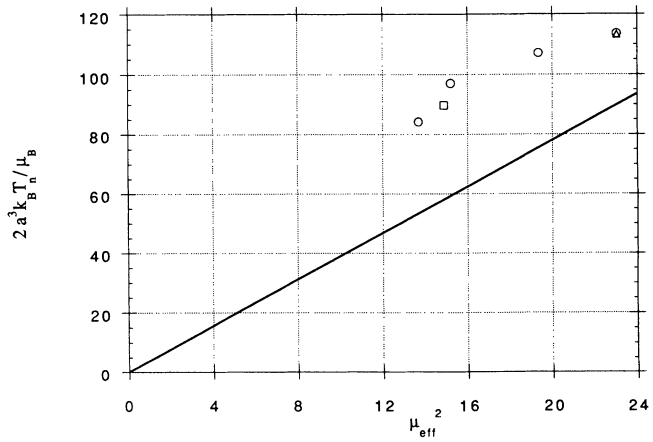

FIG. 5. Comparison of experimental Néel temperature with result from pure dipolar interaction (in plane). Data points are (△) $ErBa_2Cu_3O_7$ (Ref. 12), (○) $ErBa_2Cu_3O_x$ (Ref. 11), and (□) $ErBa_2Cu_4O_8$ (Ref. 14).

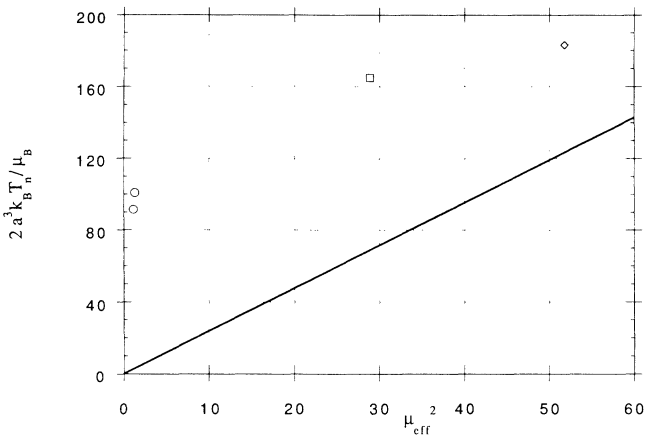

FIG. 6. Comparison of experiment and result from pure dipolar interaction (out of plane). Data points are (◇) $DyBa_2Cu_3O_7$ (Ref. 6), (□) $DyBa_2Cu_4O_8$ (Ref. 14), and (○) $NdBa_2Cu_3O_7$ (Refs. 17 and 19).

that the pure dipole system is in the universality class of the two-dimensional Ising model.[22,37]

IV. EFFECTS OF INTRAPLANAR EXCHANGE

The fact that the values of the Néel temperature obtained from the preceding analysis fall consistently below the observed values in all cases for which a quantative comparison is possible, suggests that some form of exchange interaction between the rare-earth ions contributes to the magnetic interaction. While this has been the source of some speculation[38] little is known about the origin and character of this interaction. This said, however, given the highly anisotropic nature of these compounds, it is undoubtably the case that both the nature and magnitude of the exchange interaction between rare-earth ions located in the same crystalographic plane (intraplanar coupling) will differ qualitatively from that between ions located in successive planes (interplanar coupling).

Given that the different crystal structures that have been observed in these compounds ($R Ba_2Cu_3O_7$, $R Ba_2Cu_4O_8$, and $R Ba_2Cu_4O_7$) differ primarily in the stacking of the planes containg the rare-earth sites, it might be hoped that a comparison of the various magnetic properties would permit the identification of the intraplanar and interplanar contributions to the magnetic interaction between the rare-earth ions. While such studies have been conducted the magnetic properties, such as the Néel temperature and the saturation moment $\mu_{\rm eff}$, determining the results do not appear to admit a systematic interpretation. One point concerning the stuctural differences between the rare-earth lattice in the $R Ba_2Cu_3O_7$ compounds and in the $R Ba_2Cu_4O_8$ compounds is the fact that in the case of $R Ba_2Cu_4O_8$ succesive planes of rare-earth ions are displaced half a lattice length along the b axis relative to the corresponding planes in the $R Ba_2Cu_3O_7$ compounds. Consequently the rare-earth ions are no longer stacked in rows along the c direction. In the case of the out-of-plane spin alignments Lynn has argued, on the basis of crystallographic considerations, that this results in a cancellation of all interplanar interactions.[14] This is indeed borne out by the fact that as one lowers the temperature below the Néel temperature in $ErBa_2Cu_3O_7$, $ErBa_2Cu_4O_8$, and $DyBa_2Cu_3O_7$ one begins, as one must, to observe three-dimensional ordering. In $DyBa_2Cu_4O_8$, however, experiments are not able to detect any evidence of a crossover to 3D ordering.[14]

In order to gain some insight into the magnitude of the exchange interaction, within the context of the present model, the Hamiltonian given in Eq. (4) is generalized to include a nearest-neighbor exchange interaction.

$$\mathcal{H} = \frac{\mu_{\rm eff}^2}{2}\left[\sum_{\substack{n\neq m\\ \alpha,\beta}} S_n^\alpha S_m^\beta \lim_{|\mathbf{r}|\to 0} \frac{\partial}{\partial x_\alpha}\frac{\partial}{\partial x_\beta}\frac{1}{|\mathbf{R}_{nm}-\mathbf{r}|} + \frac{J}{a^3}\sum_{\substack{\rm NN\\ \alpha}} S_n^\alpha S_m^\alpha\right], \tag{13}$$

where $\sum_{\rm NN}$ denotes a sum over nearest neighbors and J characterizes the strength of the exchange coupling. The variation of the Néel temperature with the exchange parameter J is shown in Fig. 7 for a 16×16 lattice for the out-of-plane spin configuration. The relationship between the Néel temperature θ_n and the exchange parameter is well characterized in the range shown by the linear relationship

$$\theta_n = 2.4242 + 2.3523J\,. \tag{14}$$

Extending the range of the calculation to include larger (negative) values of the exchange coupling leads ultimately to the destabilization of the pure antiferromagnetic ground state. If the value of the exchange interaction is sufficiently large (and negative) then Monte Carlo simulations, in which the system is slowly quenched from a random initial configuration at high temperature, show that the ordered state is no longer the pure antiferromag-

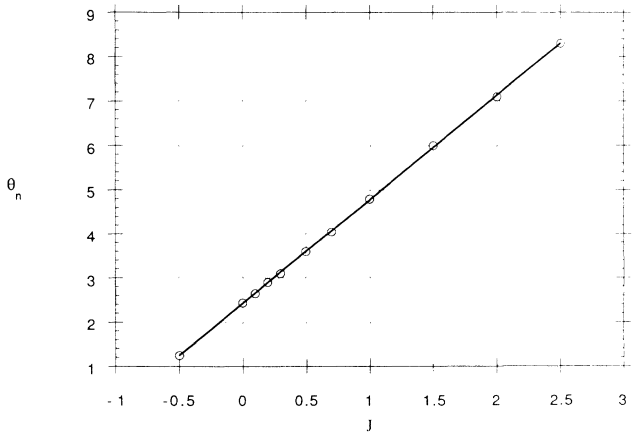

FIG. 7. The Néel temperature θ_n as a function of the exchange parameter J for out-of-plane configuration. The straight line is given by Eq. (14).

TABLE I. Estimates of the exchange coupling parameter J for several compounds.

	μ_{eff}	J	θ_n	$\mu_{\text{eff}}^2 J/2k_B a^3$ (K)	T_n (K)
$DyBa_2Cu_3O_7$[a]	$7.25\mu_B$	0.509	3.67	0.138	0.95
$DyBa_2Cu_4O_8$[b]	$5.3\ \mu_B$	1.703	6.44	0.251	0.95
$NdBa_2Cu_3O_7$[c]	$1.14\mu_B$	33.3	80.77	0.454	0.551

[a]Reference 6.
[b]Reference 14.
[c]Reference 19.

netic state but corresponds instead to the ferromagnetic state. The Monte Carlo simulations also reveal an interesting region intermediate between the ferromagnetic and pure antiferromagnetic phase, in which the spins order ferromagnetically along one axis and antiferromagnetically along the other axis at sufficiently low temperature. A comparison of the energy of this configuration, with the energy associated with the ferromagnetic and the pure antiferromagnetic configurations, reveals a range of values for the exchange coupling for which the energy of this antiferromagnetic ordering is less than both energy of the ferromagnetic and the pure antiferromagnetic states. Given the degree of symmetry with respect to the a and b axis inherent in the out-of-plane spin configuration this result is somewhat surprising and serves to illustrate the subtle phenomena that can arise as a consequence of the interplay between the long-range character of the dipolar interaction and the short-range exchange interaction. As a consequence of the complex phase behavior manifest by the variation of the strength of the exchange coupling J the temperature at which the peak in the transition temperature appears is no longer described by the simple formula given in Eq. (14) but shows instead a nonmonotonic behavior that delineates, at least approximately, the boundary between the various ordered phases and the paramagnetic phase.

Despite the complexities discussed above, the relation between the strength of the coupling and the Néel temperature given by Eq. (14) is nevertheless valid over a sufficient range of temperature that we can obtain an estimate of the strength of the exchange coupling required to reproduce the experimentally observed Néel temperatures, for the Dy and Nd compounds, all of which are observed to order with spins aligned perpendicular to the plane. However, it should be kept in mind that the following analysis assumes that the exchange interaction is both isotropic and restricted to ions located in the same plane. While these assumptions are possibly valid in the case of $DyBa_2Cu_4O_8$ their validity in the other compounds is less certain. Estimates of the exchange parameter J for several compounds are given in Table I.

A similar analysis for the in-plane spin configuration has also been carried out. Results indicate that the dipolar ground state is stable only for a particular range of coupling, with the pure antiferromagnetic or ferromagnetic state being stabilized with the inclusion of a sufficiently large positive or negative coupling constant, respectively. While this is hardly surprising calculations indicate that the magnitude of the coupling required to stabilize the ferromagnetic phase is relatively small, and as a consequence we are unable to reliably describe the dependence of the Néel temperature on the strength of the exchange coupling J in terms of a simple linear relationship similar to that given by Eq. (14) for the out-of-plane spin configuration in the domain of interest (i.e., $J \approx 0$). The problem is compounded by the fact that finite-size effects are stronger for the in-plane spin configurations than for the out-of-plane spin configurations, while the problem of stable domain-wall formations is found to be more prevalent in the in-plane spin configurations. Both of these problems, finite-size effects and domain-wall formation, make the studies for the in-plane spin configurations more time consuming and make it more difficult to obtain consistent results than in the corresponding studies for the out-of-plane spin configurations.

A more detailed discussion of the phase behavior of these systems and the possible relevance of the results obtained with regard to the $RBa_2Cu_3O_{7-\delta}$ and $RBa_2Cu_4O_8$ compounds will be deferred till later.

V. SUMMARY

In the following analysis we have obtained, by means of Monte Carlo simulation, a reasonably accurate estimate of the transition temperature of a two-dimensional Ising dipolar system for two distinct magnetic configurations in which the spins are aligned out of plane and in plane, respectively. A comparison of the results obtained from the analysis with results obtained for several of the Nd, Dy, and Er compounds show that, in all instances for which a comparison is possible, the observed transition temperature is higher than that predicted by theory. By generalizing the analysis, to include the effects of a nearest-neighbor exchange, estimates of the exchange interaction that yield the observed value of the transition temperature are obtained. The results show that the exchange and dipolar interactions are of comparable magnitude for the Nd and Dy compounds. This conclusion is, however, based on the assumption that the exchange interaction is isotropic and is restricted to ions located in the same plane. While these assumptions may be valid in the $DyBa_2Cu_4O_8$ compound, for crystallographic reasons, their validity in the other materials considered is less certain. One other important result of this study is the fact that, for the relevant range of parameters, it would appear that the magnetic properties of the two-dimensional antiferromagnetic Ising model are not qualitatively affected by the long-range character of the dipolar interaction, in particular our results confirm the conclusion that the presence of the dipolar interaction

does not change the universality class of the system.

There are two rather obvious extensions to the present analysis. In the first instance, while one can argue that the interplanar coupling between the rare-earth ions is in some sense weak, experiments clearly show the appearance of three-dimensional magnetic order in all but a few instances at sufficiently low temperatures. The extension of the present work to include the interplanar coupling in some manner would therefore be of obvious value. Such work is currently in progress. A second aspect of the present work that deserves closer scrutiny is the validity of the Ising model in describing these systems. While there is little doubt that in the case of the Kramers ions a spin $S = \frac{1}{2}$ model is appropriate in the temperature range of interest, the precise character of the CEF ground state does not, obviously, justify the assumption of an Ising model. It may therefore be of some value to consider the origin of the Ising-like behavior of these systems from within the context of a more basic model.

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