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Classical Heisenberg Magnet in Two Dimensions*

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The equilibrium properties of a square planar array of classical spins with near-neighbor Heisenberg interactions have been examined for arrays of up to 2025 spins with and without periodic boundary conditions. Equilibrium values of the root-mean-square magnetization $M_{\rm rms}$ were obtained by Monte Carlo calculations. Sample spin arrays whose energy agreed with the ensemble average at a given temperature were taken as characteristic of that temperature and were employed to obtain instantaneous correlation functions. The results are less clear than those reported previously for three-dimensional systems: The Monte Carlo calculations converged more slowly because of the lower connectivity of the lattice, and, unlike the three-dimensional case, the short-range order has a range as large, or larger, than the largest sample dimensions. The results are consistent with the observation of Mermin and Wagner that the system does not order ferromagnetically at finite temperatures, and lend some credence to the conjecture of Stanley and Kaplan on the existence of a special ordered state possessing "long-ranged short-range order."

I. INTRODUCTION

In an earlier paper,[1] we have presented results of extensive computer studies of a three-dimensional classical Heisenberg ferromagnet. That paper reported Monte Carlo calculations of the magnetization and the static spin correlations at various temperatures in various sizes of arrays. Also given were the time-displaced spin-correlation functions, found by numerically integrating the classical equations of motion for the entire system of spins. The same computer methods are readily extended to arrays of other dimensionality. The static properties of a two-dimensional array of three-dimensional spins are of interest because of the fact that a ferromagnetic state does not exist in near-neighbor coupled two-dimensional Heisenberg systems and because of associated questions concerning the existence of a nonferromagnetic state which nevertheless possesses some kind of long-range order.[2,3] Consequently, we have made Monte Carlo calculations of such two-dimensional systems of a variety of sizes and at a variety of temperatures.

In the next two sections, we review the method of calculation and summarize the results. A fuller discussion of the Monte Carlo procedure is given in I.

II. MODEL AND METHOD OF CALCULATION

Let each site n of a simple square lattice be occupied by a spin $\vec{S}_n$, where $\vec{S}_n$ is a three-dimensional vector of unit length. Let the energy of the system

be

$$E = -\tfrac{1}{2}J\sum_{n,m}\vec{S}_n\cdot\vec{S}_m \quad , \tag{1}$$

where the summation is over pairs of nearest neighbors only. We consider finite lattices of rectangular outline containing N sites. Two types of boundary conditions have been used: periodic boundary conditions and isolated boundary conditions, for which the only terms in (1) are for pairs inside the array. In the work reported, N has been assigned various values from 225 to 2025.

The Monte Carlo method[4] is employed as described in I. In effect a random walk, biased with a certain weighting function, is carried out in the configuration space of the spin system. The weighting function depends on a Boltzmann factor involving the energy E of the point in configuration space and the temperature, which plays the role of an arbitrary parameter of the calculation. After enough steps have been taken, averages over all the steps of any property of the system approximate the true canonical average of that property at the chosen temperature and converge to the average as the number of steps approaches infinity. A different Monte Carlo series is carried out for each temperature of interest. Temperature is defined in energy units where J of Eq. (1) is set equal to 1.

The averages were found to converge more slowly in two-dimensional systems than in three-dimensional systems; the resulting demands on computer time prevented use of arrays as large as some of those in I. In general, several Monte Carlo series would be run for a given temperature with starting points, in configuration space, characteristic of temperatures below, at, and above the temperature in question. This provided better average quantities than single longer samplings.

The magnetization of the system is defined as

$$\vec{M} = \sum_{n=1}^{N}\vec{S}_n \quad . \tag{2}$$

The root mean square of this quanity $M_{\rm rms}$ was calculated, as were the first moments of $M_{\rm rms}$, and the average of the absolute value of $\vec{M}$, denoted by $\langle|M|\rangle$. Also calculated were the two kinds of instantaneous pair correlation functions, longitudinal and transverse.

The longitudinal correlation function is

$$\langle S_{\parallel}(r)S_{\parallel}(r+\Delta)\rangle = \langle S_n^z S_m^z\rangle \quad , \tag{3}$$

where S_n^z is the z component of $\vec{S}_n$, z is in the direction of $\vec{M}$, and site n is at r and site m is at $r+\Delta r$.

Similarly, the transverse correlation is

$$\langle\vec{S}_{\perp}(r)\cdot\vec{S}_{\perp}(r+\Delta r)\rangle = \tfrac{1}{2}\langle S_n^x S_m^x + S_n^y S_m^y\rangle \quad . \tag{4}$$

The correlation functions were evaluated for arrays whose $M_{\rm rms}$ and energy coincided with the average values obtained from the set of Monte Carlo series for the temperature in question. To test, correlation functions were obtained for several arrays (appropriate to a particular temperature). The general features of the correlation functions were, by this criterion, quite reproducible. A better, but much more costly, procedure would have been to take running averages of the correlation functions in the course of the Monte Carlo samplings. This was not done.

III. RESULTS

The root-mean-square magnetization, found from the Monte Carlo investigations of isolated two-dimensional square arrays, is plotted against temperature in Fig. 1. That for the square arrays, but with periodic boundary conditions, appears in Fig. 2 (the dashed curves are the results for isolated arrays of 225 and 2025 spins repeated from Fig. 1). The inflection points correspond to the ordering temperatures of the arrays in question. The first moments of the $M_{\rm rms}$, of Figs. 1 and 2, appear in Figs. 3 and 4, respectively. The data of Figs. 1 – 4 are normalized with respect to M_0, the magnetization associated with ferromagnetic order at $T=0$. These moment data were used to help place the inflection points; the positions of the maxima in the moment data suggest that the inflection points are almost independent of array size for the isolated arrays, while shifting to significantly lower temperature with increasing array size for the periodic arrays. There is considerable uncertainty as to the exact positions of the inflection points. For example, Fig. 4 suggests that it lies in the region $0.5<T<0.7$ for the 2025 spin array. We have placed the transition at $T\sim0.6$ in Fig. 2, but we will shortly see results suggesting that it lies at a higher temperature. Each point on Figs. 3 and 4 is the result of a Monte Carlo calculation and the scatter in the points is symptomatic of the poor convergence of the Monte Carlo procedure for these two-dimensional arrays.

The vertical hatching in Figs. 1 and 2 is the Monte Carlo data on which the 2025 spin magnetization curves are based. The dashed regions indicate where at least one Monte Carlo run placed the magnetization, and the solid-lined regions indicate where the preponderance of the Monte Carlo results lie. (As was discussed in Sec. II, a number of Monte Carlo runs were obtained at most temperatures.) The Monte Carlo samplings employed here are almost two orders of magnitude larger than those used for the simple cubic three-dimensional array of 8192 spins reported in I, and the scatter is still an order of magnitude worse.

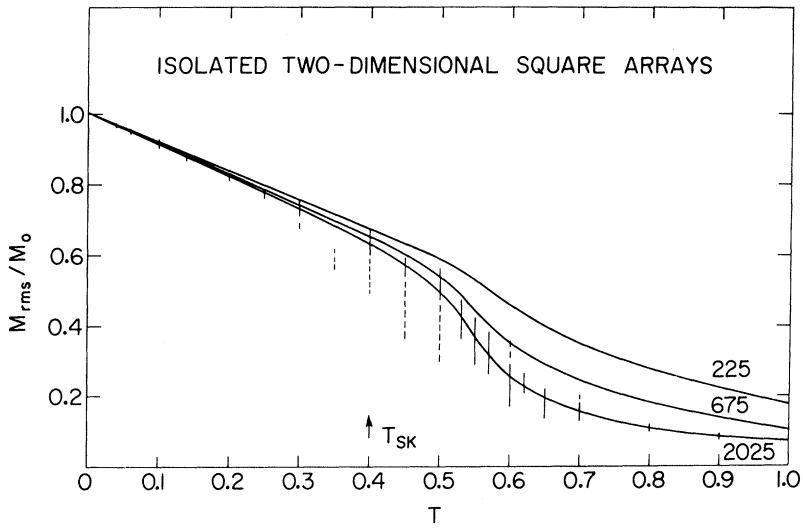

FIG. 1. Root-mean-square magnetization M_{rms} versus temperature from Monte Carlo calculations for various sizes of isolated two-dimensional square lattices of spins. T_{SK} is the temperature at which Stanley and Kaplan found a singularity in the susceptibility. M_{rms} is normalized with respect to the magnetization at $T=0$, i.e., M_0.

There are several factors contributing to the poor convergence. First, the convergence of such Monte Carlo samplings is known to be dependent on the connectivity of the particles, in this case, the spins. The connectivity of the near-neighbor coupled square lattice, with each spin having four near neighbors, is much weaker than that of the simple cubic lattice considered in I. For this reason alone, the Monte Carlo convergence should be poorer here. Second, the short-range order of the planar system appears to be as long, or longer ranged, than the dimensions of the largest arrays we have dealt with. This creates fluctuations in the sampling, hence poor Monte Carlo convergence. Similar effects, arising from critical fluctuations at temperatures in the vicinity of T_c, were encountered for the simple cubic lattice in I.

The tendency of M_{rms} for the periodic array to lie higher and to have a more rapidly moving inflection point, moving with changing array size, is reasonable. The M_{rms} lies higher because the spins on the edge of the sample see the exchange field of the neighboring array (s), whereas those of the isolated array have fewer near neighbors, hence feel weaker exchange fields. The more rapid change in shape and shift in the inflection point follows from the tendency of samples with periodic boundary conditions to converge more rapidly than isolated arrays to the thermodynamic limit (i.e., the limit of infinite sample size). The two samplings will, of course, coincide in this limit.

It should be noted that M_{rms} samples *both* long- and short-range order. The isolated array data

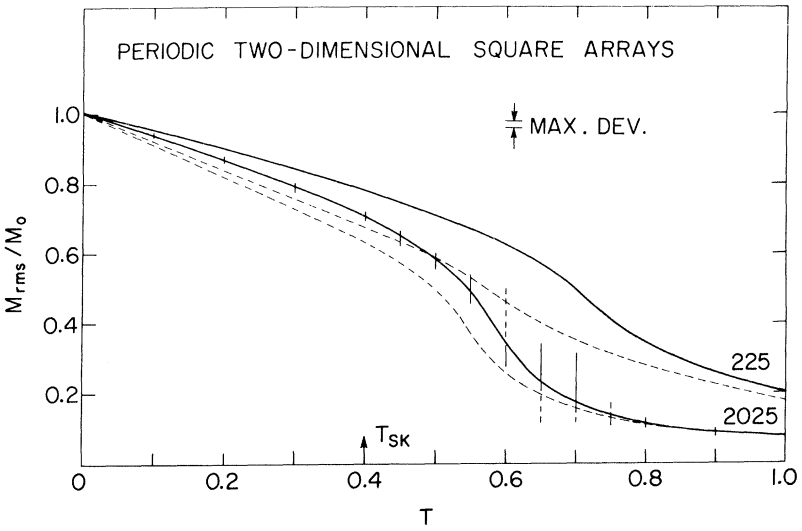

FIG. 2. Root-mean-square magnetization versus temperature from Monte Carlo calculations for various sizes of two-dimensional square arrays of spins with periodic boundary conditions. The dashed curves indicate the behavior for isolated arrays (from FIG. 1) of the same size. Max. Dev. provides a measure of the maximum deviation between the average of the magnitude of the magnetization and the root-mean-square value. (The maximum occurs at an inflection point.) M_{rms} is normalized with respect to the magnetization at $T=0$.

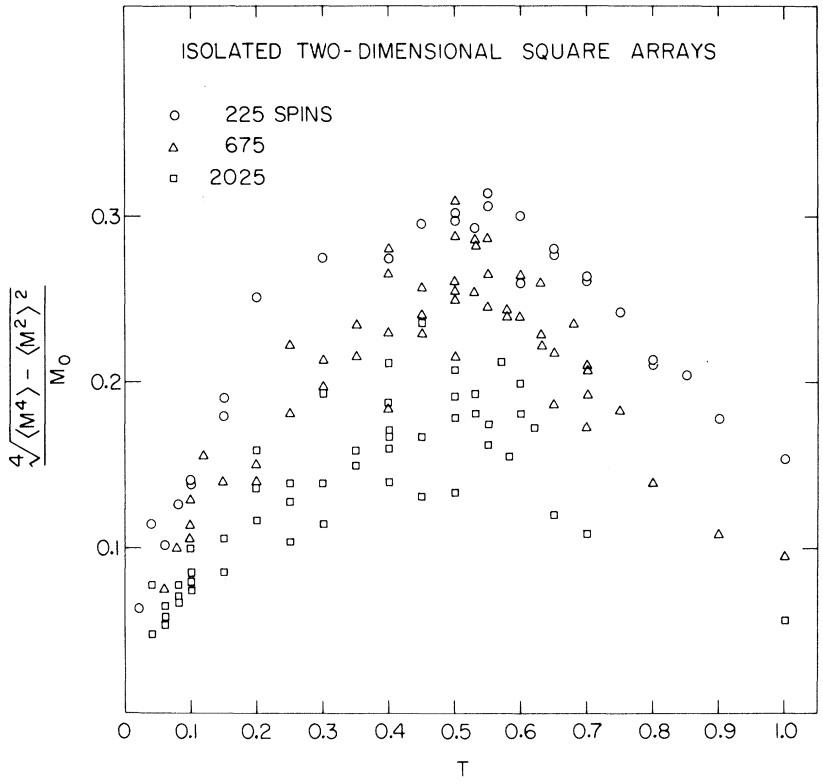

FIG. 3. First moment of M_{rms}, normalized with respect to M_0, for the isolated square planar arrays. Each point is the result of a Monte Carlo run.

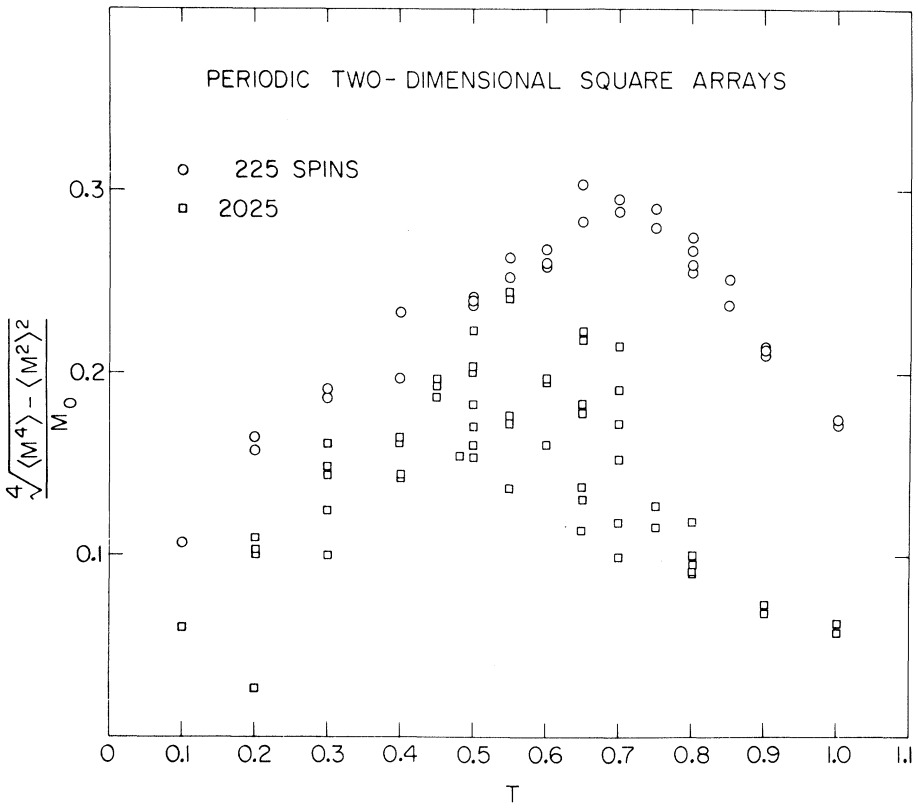

FIG. 4. First moment of M_{rms}, normalized with respect to M_0, for the square planar arrays with periodic boundary conditions.

of Fig. 1 suggest that much larger "laboratory size" arrays, of say 10 000 ×10 000 spins, would display nonzero $M_{\rm rms}$ curves. We believe that such behavior, as well as that of Figs. 1 and 2, arises from short-range order. This implies short-range order of extraordinary long range as compared with three-dimensional systems. This, taken with the attendant poor Monte Carlo convergence, makes it impossible to use our results to assay the properties of two-dimensional arrays in the thermodynamic limit. Mermin and Wagner concluded that the spontaneous magnetization (i.e., long-range order) vanishes at all finite temperatures, by considering the system with an external magnetic field, applied taking the limit $N \to \infty$, and then allowing the external field to vanish. Our results are compatible with Mermin and Wagner's conclusion. Stanley and Kaplan's high-temperature expansion of the susceptibility yielded a singularity at the temperature marked $T_{\rm SK}$ in Figs. 1 and 2. This would seem to imply some sort of magnetic ordering at that temperature, but given Mermin and Wagner's results, it is not conventional long-range order. Our $M_{\rm rms}$ results, with their suggestion of extraordinary short-range order, are compatible with Stanley and Kaplan's conclusions. Note that our inflection points lie at higher temperatures than $T_{\rm SK}$, and would decline for larger arrays.

Monte Carlo samplings of $\langle |M| \rangle$ as well as $M_{\rm rms}$ were taken for the periodic boundary spin arrays. While the samplings of $M_{\rm rms}$ and $\langle |M| \rangle$ displayed considerable scatter from run to run, the two averages tracked each faithfully and quite accurate quantitative statements can be made concerning their relative behavior. $\langle |M| \rangle$ lies lower than $M_{\rm rms}$, as it should. The two averages are in close agreement (approaching the width of the plotted lines in the figure) at the lowest and highest temperatures shown. The largest deviations occur in the vicinity of the inflection points. For the 2025 spin array, this deviation equalled the interval marked "Max. Dev." on the figure. One might hope that $\langle |M| \rangle$ would supply some insight, when compared with $M_{\rm rms}$, into the respective roles of long- and short-range order. This is not the case, since the apparent range of the short-range order is at least equal to sample dimensions, causing $\langle |M| \rangle$ as well as $M_{\rm rms}$ to be dominated by short-range effects.

The instantaneous pair correlation functions, $\langle \vec{S}_\perp(r) \cdot \vec{S}_\perp(r+\Delta r) \rangle$ and $\langle S_\parallel(r) S_\parallel(r+\Delta r) \rangle - (M/M_0)^2$ are plotted for $\langle 10 \rangle$ directions in the periodic 2025 spin array in Fig. 5. The plots have been drawn such that the self-correlation functions ($\Delta r = 0$) have common amplitude. The values of the self-correlation functions, i.e., the amplitudes of the pair correlation plots are shown in Fig. 6. We believe

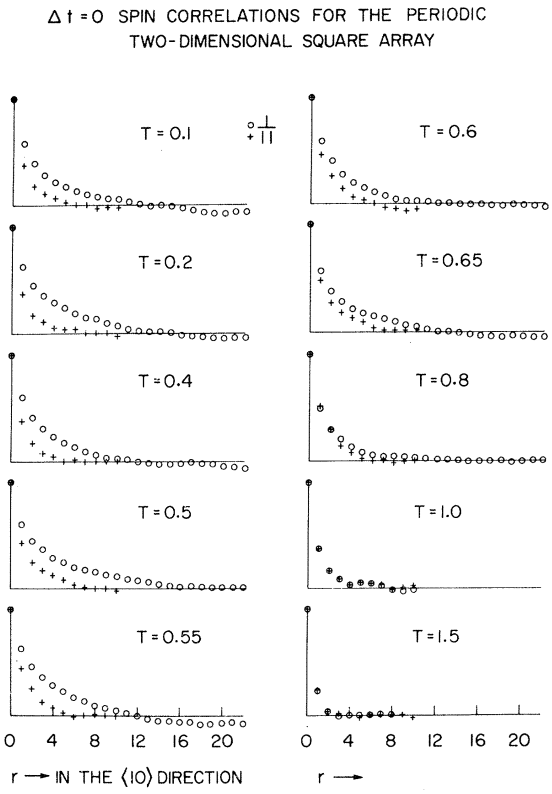

FIG. 5. Instantaneous spin correlations (normalized to the self-correlation functions) between various neighbors along ⟨10⟩ lines, for various temperatures. The results were obtained for the 45×45 (i.e., 2025) spin array with periodic boundary conditions. Crosses denote longitudinal, the circles transverse, correlation functions.

these results fail to give the full measure of the short-range order appropriate to the two-dimensional square lattice in the thermodynamic limit. This is suggested by the fact that these functions are sensitive to varying sample size and to the boundary conditions. Such a failure is consistent with our experience with the simple cubic lattice, in I. Some insight into the two-dimensional lattice can nevertheless be gained from the figures.

The correlation functions for the two-dimensional array fall off much more slowly than their three-dimensional counterparts. For example, the ⟨30⟩ transverse correlation function, at temperatures below the inflection point, is about one-third the self-correlation function (and two-thirds the near-neighbor) while the ⟨300⟩ transverse function in the simple cubic lattice is about one-fifteenth the self- (and one-quarter the near-neighbor) correlation function at temperatures below T_c. Some measure of an ordering temperature can be gained by

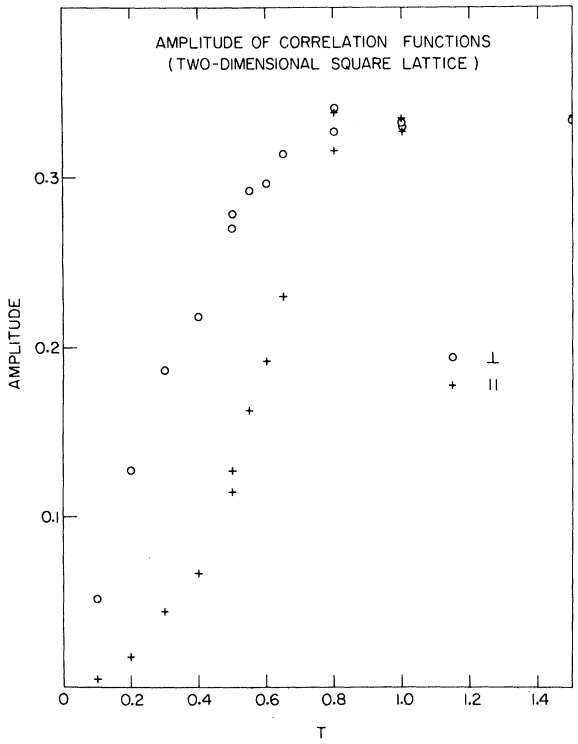

FIG. 6. Amplitudes of the instantaneous self-correlation functions (i.e., of the plots in Fig. 5) as a function of temperature. Longitudinal correlations are denoted by crosses, transverse correlations by circles.

inspecting where the self-correlation functions equal $\frac{1}{3}$ (implying no long-range order) in Fig. 6 or where the transverse and longitudinal functions first take on identical shape in Fig. 3. This would suggest an ordering temperature between 0.70 and 0.75, a rather higher value than is suggested by the moment data for the 2025 spin array in Fig. 4. (Such a measure always places the inflection point equal to or higher than the moment samplings of Figs. 3 and 4.)

The temperature dependence of the correlation functions is very similar to that found for the three-dimensional system, though there are suggestions of a few subtle differences. The transverse and longitudinal correlation functions are identical above the ordering temperature and become shorter ranged as the temperature increases. Consistent with most theories of three-dimensional systems, the transverse function is of fixed shape (though not amplitude) below the ordering temperature, while the longitudinal function collapses, i.e., the longitudinal order goes into the *long*-range term with decreasing temperature. The collapse of the longitudinal function seems somewhat slower than was our experience with the simple cubic lattice over the temperature range considered (i.e., down to $\sim\frac{1}{2}$ the ordering temperature). Also, there is a hint that the transverse function starts contracting before the longitudinal function has come up to it as one approaches and goes through T_c from below. If true, this is inconsistent with what occurs in three-dimensional systems. The temperature dependence of the self-correlation functions seen in Fig. 6 also differs from that for the simple cubic lattice. For the latter, the transverse function is quite faithfully proportional to T below T_c. Here it appears proportional to a lower power of T.

IV. CONCLUSIONS

The conclusions to be drawn from the present study are less definite than those for the three-dimensional system. This is because of the slower convergence of the Monte Carlo procedure in two dimensions, as is discussed below. The calculations indicate, for our finite samples, a nonzero root-mean-square magnetization below an apparent transition temperature. This is consistent with the theorem of Mermin and Wagner[2] on the absence of long-range order[5] at finite temperatures, and gives some credence to the conjecture of Stanley and Kaplan[3] of the existence of an ordered state with "long-ranged short-range order," i.e., a state for which $\langle \vec{S}_0 \cdot \vec{S}_r \rangle \to 0$ as $r \to \infty$ while $\sum_r \langle \vec{S}_0 \cdot \vec{S}_r \rangle$ diverges. It is not possible to argue definitively from our results that such a state exists because of the difficulties in the convergence of the Monte Carlo calculations and because of the question of the range of the short-range order relative to array size. Nevertheless, on balance, the evidence indicates that some sort of phase transition does occur in the two-dimensional Heisenberg system. Further experiments, of the type done here, which might sort this matter out require substantially larger, faster, and cheaper computing facilities.

ACKNOWLEDGMENT

We wish to thank Miss Estarose Wolfson for her assistance with the computer programming.

*Work supported by the U. S. Atomic Energy Commission.

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[5]We note that two-dimensional systems are expected to have long-range order once an anisotropic term of sufficient size is present, in addition to the Heisenberg term, in the Hamiltonian. Such terms are almost inevitably present in real two-dimensional systems for there will exist spin dipolar forces which do not cancel out (because of the two-dimensional character of the lattice). These terms will generally be large enough to have this effect.

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Intercell Corrections for Ionic Motion in Displacement Ferroelectrics

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(Received 17 July 1969)

A set of microscopic parameters determined for $LiTaO_3$ in an earlier paper by comparing dielectric properties with the results of a statistical calculation are analyzed in terms of a point-charge-plus-electronic-polarizability model for the system. The results indicate the existence of very strong intercell correlations for ionic motion to temperatures considerably in excess of the Curie point T_C. These correlations are weak functions of temperatures to the extent that they do not produce deviations from effective-field (e.g., Curie-Weiss) behavior near T_C. Neglect of correlation effects leads to many inconsistencies and indicates that the use of random-phase Lorentz fields in statistical theories for displacement ferroelectrics is qualitatively unsound. Spontaneous polarization for $LiTaO_3$ is found to be 45% ionic, 55% electronic; the tantalum-oxygen bond is very markedly covalent, with estimated ionic charges of $+0.8$ and -0.6 electronic units, respectively.

I. INTRODUCTION

In an earlier series of papers, [1] a statistical theory for displacement ferroelectrics was developed and applied in some detail for the salt lithium tantalate $LiTaO_3$. The statistical approximation adopted to describe equilibrium bulk dielectric properties was to replace all primitive cells except one by their ensemble averages, thereby including long-range (intercell) interactions only in a Hartree or effective-field approximation. In this way it was possible to write an effective system Hamiltonian in terms of the normal vibrational modes of a single cell. For some displacement ferroelectrics, only a single transverse optic mode of lattice vibration is grossly temperature dependent (the "soft" mode); $LiTaO_3$ and $LiNbO_3$ are good examples.[2] For these systems, the effective Hamiltonian contains sufficiently few parameters for them to be comfortably overdetermined by comparison of bulk ferroelectric properties with the results of the statistical theory.

In principle, by choosing a model for the microscopic system, the Hamiltonian parameters (which are microscopic quantities) could be determined from first principles. In practice, for most of the parameters involved, such a calculation at the present time is not very meaningful, and in the lithium tantalate work,[1] we adopted the "spin-Hamiltonian" philosophy from magnetism, treating the parameters simply as numbers to be determined by comparing theory with experiment. The problem was overdetermined to a considerable degree, and the self-consistency of the results was surprisingly good, even with completely temperature-independent parameters. Initially, this was taken to imply that the random-phase approximation (RPA) for intercell interactions was adequate in this context and that, consequently, correlation effects were not a major factor.

However, certain Hamiltonian parameters are fairly easily interpreted in terms of more fundamental microscopic properties of the lattice and constituent ions and, when an effort was made[1] to translate the "best-fit" parameter values into more familiar microscopic concepts, some baffling results were obtained: Lorentz fields more than an order of magnitude smaller than expectation and a Lorentz-field electronic contribution to polarization of opposite sign to the ionic term. In the present paper, we shall show that these, and other inconsistencies which follow, disappear completely when an allowance is made for intercell correlations between the ionic motions of neighboring primitive cells of the lattice.

The way in which the numerous pieces of the interpretational jigsaw fall neatly into place with-