Magnetic properties of dipolar crystals with rhombohedral symmetry

E. Koutroumanou and A. K. Theophilou
Nuclear Research Center "Demokritos," Aghia Paraskevi Attikis, Greece
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This paper deals with the quantum-mechanical properties of rhombohedral crystals with dipole-dipole interactions, where the g-factor component along the trigonal axis z, $g_{\parallel}$, is small. The sensitivity of the above-noted properties to small changes in the lattice and magnetic parameters is examined. It is shown that the qualitative features of the ground state and the density of states do not change. The Niemeijer-Meijer method, which is based on the classical Luttinger-Tisza approach, is used. The ground-state energy and magnetization of cerous zinc nitrate in a homogeneous external magnetic field are calculated, and the results are compared with the available experimental data.

Physical properties of spin systems depend on the lattice parameters and the relation of the spins to the magnetic moments. It is thus of interest to examine which of the above properties are sensitive to these parameters and which depend only on some general features like, e.g., the crystal symmetry. It is also of interest to examine the sensitivity of the various theoretical results to the value of the parameters, as these parameters are subject to experimental errors.

Peverley and Meijer[1] and Wong, Dembinski, and Opechowski[2] have shown that the entropy, specific heat, and magnetic susceptibility of dipolar crystals above the critical temperature are not sensitive to small changes of the crystal parameters. These authors used the method of Van Vleck,[3] which however, is not applicable for temperatures below the transition point. In order to examine whether this trend persists at low temperatures, Meijer, Lockhart, and Niemeijer[4] treated the problem by using the classical Luttinger-Tisza method[5] and derived a positive answer.

Up to now most of the theoretical calculations on dipolar crystals are based on the classical Luttinger-Tisza method.[6–12] However, the quantum-mechanical treatment of the magnetic properties of these crystals may reveal some features absent in the classical treatment.[13,14] It may also lead to some discrepancies between the classical and the quantum-mechanical results.

In this paper we examine the case of rhombohedral crystals with dipole-dipole interactions. In particular, we deal with the cerous zinc nitrate (CZN) crystal, for which experimental information is available,[15,16] and its crystal structure is similar to cerous magnesium nitrate (CMN) studied in Refs. 17–19.

The method used for deriving our results is the Niemeijer-Meijer method, a detailed description of which

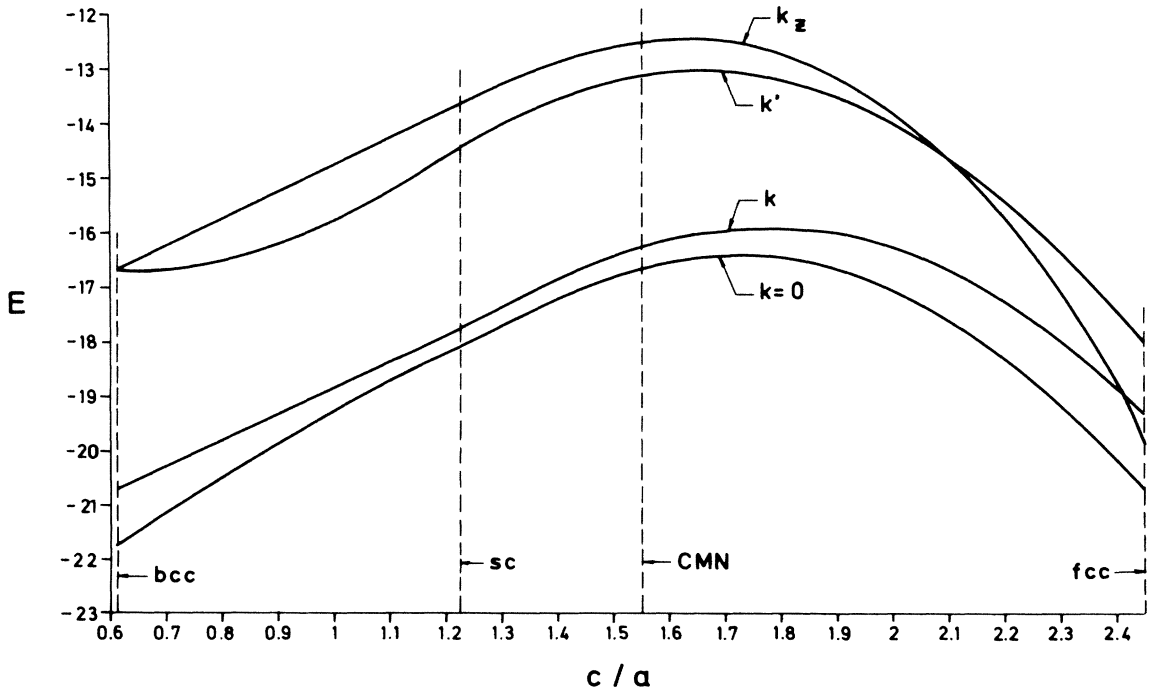

FIG. 1. Minimum energy for the various k representations vs c/a. By k and k' we denote the degenerate states of $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3$ and $\mathbf{k}_1', \mathbf{k}_2', \mathbf{k}_3'$ representations, respectively.

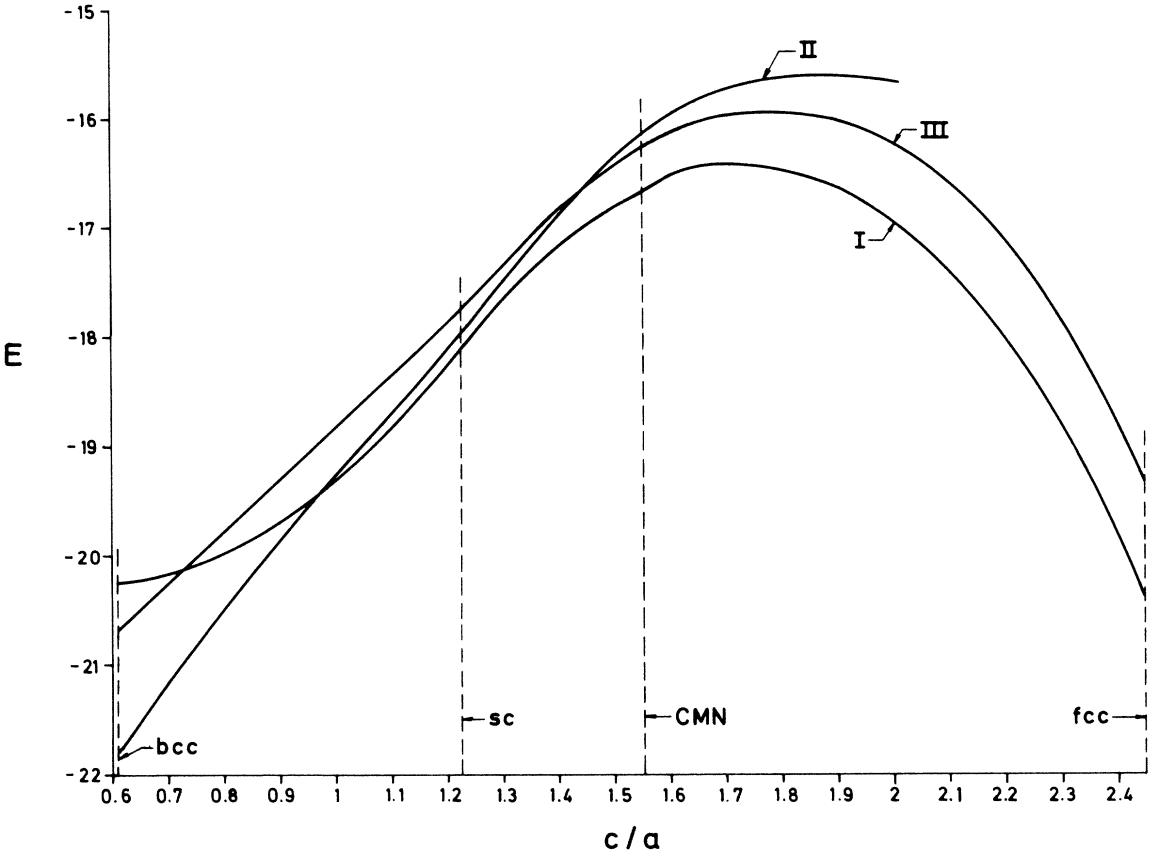

FIG. 2. Energy vs c/a for the three lower-lying states of CMN. Curve I is the energy of the doubly degenerate $\mathbf{k}=\mathbf{0}$ state, curve II corresponds to a nondegenerate $\mathbf{k}=\mathbf{0}$ state, and curve III is the threefold-degenerate energy level of the $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ states.

can be found in Refs. 14 and 17. For the calculation of the lattice sums, we used the method developed by Nijboer and DeWette.[20]

The crystal structure of rhombohedral crystals is determined by the length of the primitive lattice translation $a_{\rm rh}$ and its angle θ with the threefold symmetry axis z. The angle θ is connected with the parameters of the hexagonal cell by the relation $\tan\theta=\sqrt{3}(a/c)$, where c is the height of the hexagonal cell and a the radius of the hexagon. The effective Hamiltonian can be expressed in the form $H=H'/a_{\rm rh}^3$,

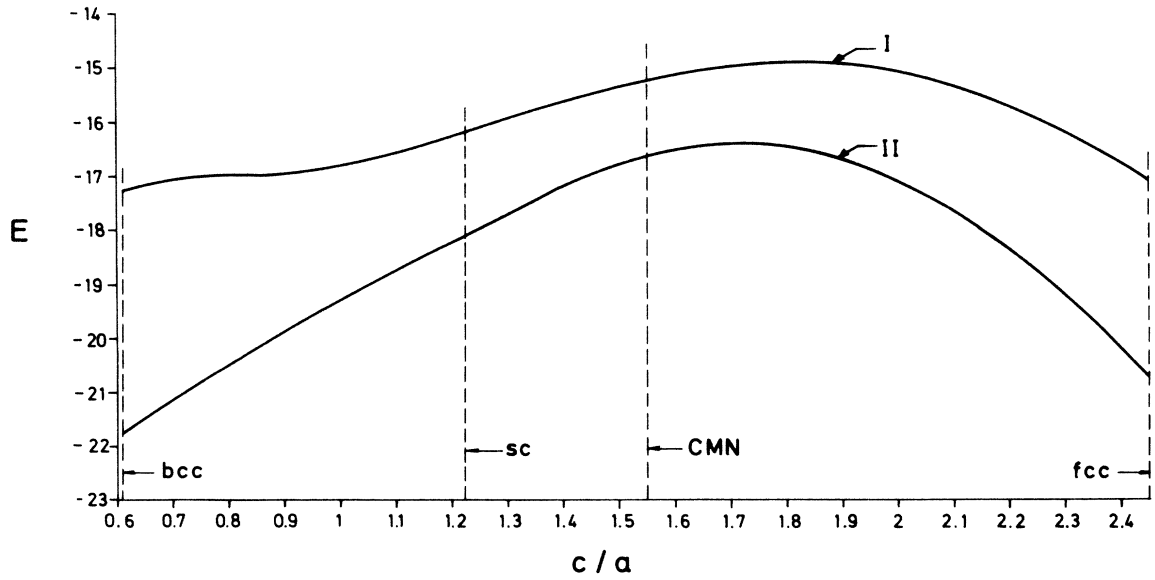

FIG. 3. Ground-state energy vs c/a as obtained by the classical Luttinger-Tisza method (I) and by the quantum Niemeijer-Meijer method (II).

where H' depends only on θ. For this reason, in our calculations we kept a_{rh} fixed ($a_{rh}=8.513$ Å) while we varied c/a.

In Fig. 1 we present the energies versus c/a of the lowest-energy eigenstates of each irreducible representation of the translation group. Although c/a varies from 0.61 to 2.45, the ground-state energy varies from -21.8 to -16.4 mK per 4 moles CMN, the maximum being at $c/a=1.7$. No appreciable change appears in the ground-state energy of CMN. In fact, for a 10% change of c/a, the ground-state energy varies only by 3%.

A more important feature is that an antiferromagnetic state with $\mathbf{k}=\mathbf{0}$ is always the lowest-energy state, and no crossing with lowest-energy states of other representations occurs as the value of the parameter varies from the bcc to the fcc lattice. Some crossing occurs between the $\mathbf{k}$ and the $\mathbf{k}_z$ lowest energies as the symmetry of the crystal approaches the fcc symmetry. There is also a crossing between $\mathbf{k}'$ and $\mathbf{k}_z$ lowest energies when $c/a \simeq 2.1$. At the bcc lattice the $\mathbf{k}'$ and $\mathbf{k}_z$ values coincide.

In Fig. 2 we plotted the energies of the lowest-energy states versus c/a irrespective of representation. The doubly degenerate $\mathbf{k}=\mathbf{0}$ state keeps to be the lowest-energy state in the region of $c/a \gtrsim 1.0$. The origin of this degeneracy is discussed in Ref. 16. On the left of this point the nondegenerate $\mathbf{k}=\mathbf{0}$ state has the lowest energy.

The classical ground-state energy lies above the quantum-mechanical one, while the minimum deviation occurs at about $c/a=1.6$ (see Fig. 3).

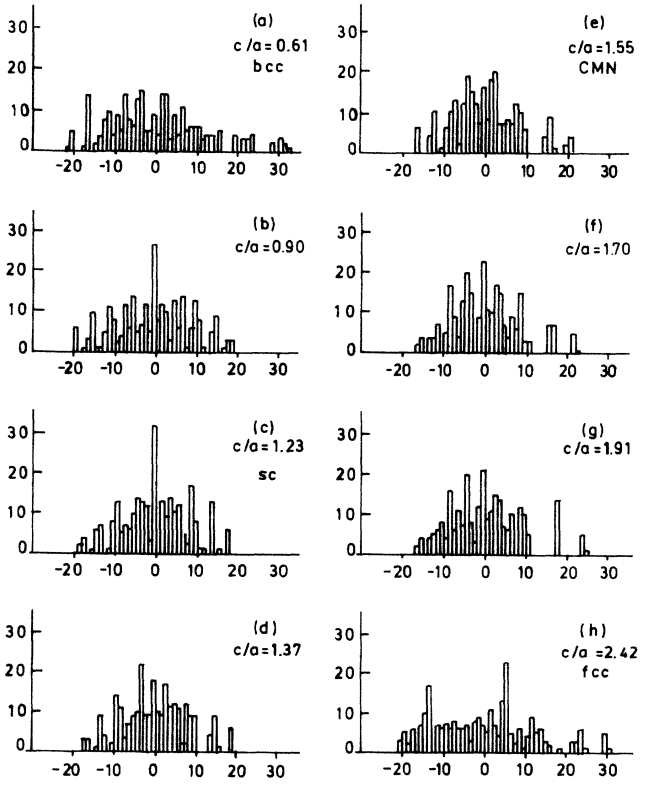

FIG. 4. Density of states vs energy for different values of the parameter c/a. Some values of c/a corresponding to lattices of particular interest are depicted in the following figures: (a) bcc lattice, (c) sc lattice, (e) CMN lattice, and (h) fcc lattice.

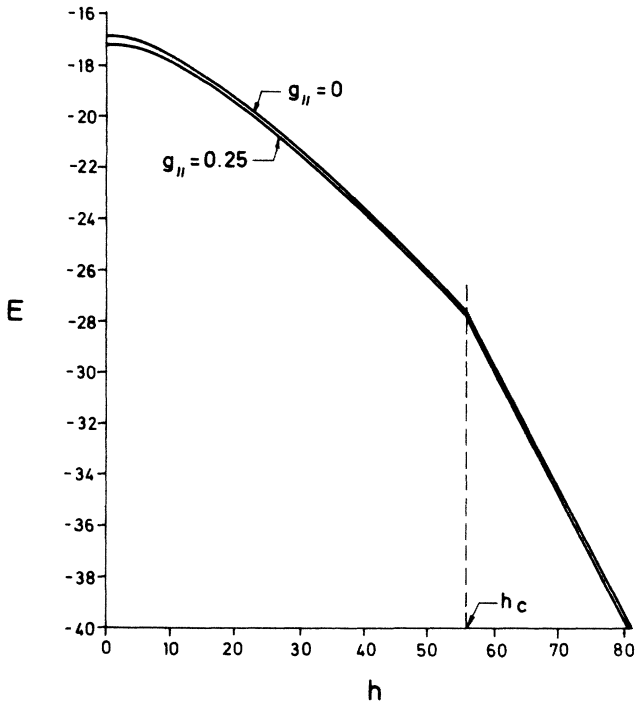

FIG. 5. Ground-state energy vs magnetic field parallel to the x axis for CZN.

The density of states (see Fig. 4) keeps the same features and apart from small details it does not change when c/a varies from 1.3 to 1.9. Energy gaps appear always on the left and the right ends of the spectrum, while the high density is around 0. The density of states presents the most compact form around the simple-cubic (sc) lattice, whereas in the neighborhood of the bcc and fcc lattices the histogram presents longer tails.

The crystal structure of cerous zinc nitrate is similar to that of CMN. The crystal parameters vary very slightly with respect to those of CMN:

$$a_{rh}=8.463 \text{ Å}, \quad \theta=47.69^\circ \text{ for CZN},$$

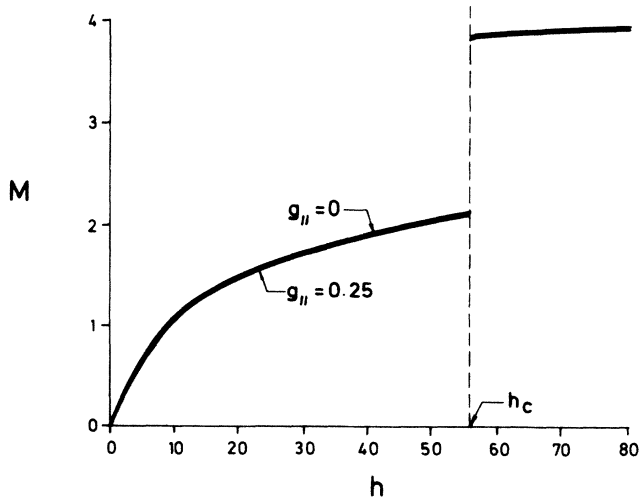

FIG. 6. Magnetic moment vs magnetic field for CZN.

and

$$a_{rh} = 8.513 \text{ Å}, \quad \theta = 48.16° \text{ for CMN} .$$

The g-factor components are $g_\perp = 1.82$ for CZN and 1.84 for CMN, while the value of $g_\parallel$ for CZN is estimated to be in the range between 0–0.25.

For $g_\parallel$ in the above range the ground state of CZN was found to be antiferromagnetic with energy values between -17.24 and -16.88 mK/8 gramions.

When an external magnetic field is applied to the crystal along the x axis, a phase transition occurs at 56 Oe. This value compares well with the experimentally measured value of 58.5 Oe.[15,16] The magnetic moment induced along the x direction, i.e., the direction the field is applied, presents a discontinuity at the critical field. This discontinuity occurs when the magnetization assumes about half the saturation value. Thus the main features are like those of CMN, the influence of $g_\parallel$ being very small (see Figs. 5 and 6).

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