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## MAGNETISM AND FERROELECTRICITY

# Simulation of the Magnetic Properties of Manganese Oxide $Pb_3Mn_7O_{15}$

**S. S. Aplesnin[a, b, *] and A. I. Moskvin[a]**

[a] *Reshetnev Siberian State Aerospace University, Krasnoyarsk, 660014 Russia*
**e-mail: apl@iph.krasn.ru*
[b] *Kirensky Institute of Physics, Siberian Branch, Russian Academy of Sciences, Akademgorodok, Krasnoyarsk, 660036 Russia*

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**Abstract**—The magnetic susceptibility, heat capacity, and spin–spin correlation functions of manganese oxide $Pb_3Mn_7O_{15}$ are calculated by the Monte Carlo method. Two critical temperatures are determined: $T_1 \approx 20$ K, above which a modulated structure along the hexagonal axis is formed, and $T_2 \approx 70$ K, at which the long-range magnetic order disappears. The antiferromagnetic exchange interaction constant in a hexagonal plane is estimated to be $J_1 \sim 7$ K, and the antiferromagnetic and ferromagnetic exchange interaction constants between hexagonal planes are calculated to be $J_2 \sim 3$ K and $K \sim 50$ K, respectively.

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## 1. INTRODUCTION

Magnetic systems with frustrated exchange interactions [1], which are formed in stochastic magnets [2] and chemically ordered compounds due to the lattice topology [3], possess high degeneracies and unusual magnetic properties [4]. This fact is reflected in the rich variety of phases and phase transitions [5] and explains their high sensitivity to various perturbations. To such compounds belongs $Pb_3Mn_7O_{15}$ [6] with competing ferromagnetic and antiferromagnetic exchange interactions.

X-ray studies of the crystal [6] showed that it crystallizes in the hexagonal system (space group $P6_3/mcm$) with unit cell parameters $a = 10.0287(4)$ Å and $c = 13.61376(6)$ Å. The unit cell contains four formula units. The crystal possesses a pronounced layered structure, which is reflected in its physical properties. Each manganese ion is in an octahedral environment consisting of oxygen ions. The $Mn^{3+}$ and $Mn^{4+}$ ions are in a ratio of 4 : 3 and occupy four nonequivalent positions in the crystal (Fig. 1a). The lead ions are located between the layers and occupy nonequivalent positions Pb1 and Pb2. The magnetic structure of this compound has two hexagonal close-packed (hcp) unit cells bound to each other by ferromagnetic exchange interactions (Fig. 1b).

Experimental data [6] indicate that there are several characteristic temperature ranges in which the crystal is in different magnetic states. In the range from ~200 K to high temperatures, the magnetization exhibits a behavior characteristic of the paramagnetic state. At temperatures below $T \sim 160$ K, the temperature dependence of the magnetization has a low broad peak associated with the appearance of a short-range magnetic order. At $T_2 \approx 70$ K, a sharp narrow peak is clearly observed [6] indicating the appearance of a long-range magnetic order. Below 70 K, the $Pb_3Mn_7O_{15}$ compound is in an uncompensated antiferromagnetic state with weak ferromagnetism, with the spontaneous magnetic moment (equal to 0.06 $\mu_B$ per formula unit at 2 K) lying in the basal plane of the crystal. Another magnetic phase transition is observed at $T_1 \sim 20$ K, and the magnetic susceptibility measured for samples cooled in a field $H = 500$ Oe and in the absence of a magnetic field exhibits thermal hysteresis below 45 K [6].

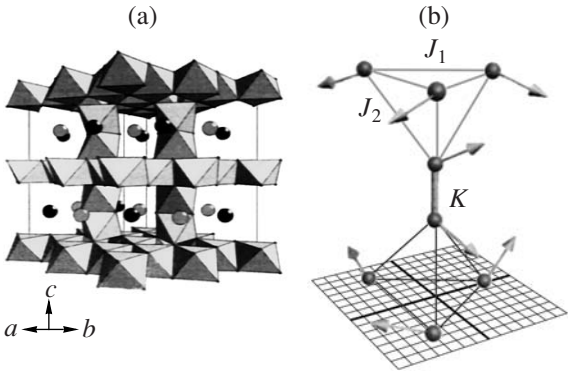


**Fig. 1.** (a) Crystallographic structure of $Pb_3Mn_7O_{15}$ (schematic): Mn cations are positioned at the centers of oxygen octahedra, and Pb ions are shown by bright (position Pb1) and dark (position Pb2) circles. (b) Magnetic structure with three types of exchange.

cally through the hcp planes. This conclusion is confirmed by the calculated spin–spin correlations (Fig. 3) along the $z$ axis perpendicular to the hexagonal plane. With increasing temperature, the ferromagnetic ordering of spins is destroyed and a modulated structure arises. The spin–spin correlation function changes sign at a distance $r = 3$ at $T = T_1$. The correlation between spins along a direction in the lattice basal plane remains positive and disappears at the Néel temperature. The temperature dependence of the heat capacity (Fig. 2b) has a pronounced maximum at $T = T_N$. From comparing the $T_1/T_N$ ratio calculated by the Monte Carlo method with experimental values $T_1/T_2 \approx 0.3$ and $T_N/JS^2 = 2.5$, the exchange constants can be estimated to be $J_1 \sim 7$ K, $J_2 \sim 3$ K, and $K \sim 50$ K.

## 3. CONCLUSIONS

Thus, the low-temperature phase transition in $Pb_3Mn_7O_{15}$ is associated with the formation of a modulated structure along the [001] direction at $T > 20$ K. Below this temperature, the pairs of spins are ordered ferromagnetically and a spontaneous magnetic moment arises. The maximum in the heat capacity corresponds to the disappearance of the long-range magnetic order. The effective exchange constants in $Pb_3Mn_7O_{15}$ have been found.

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