
*Incommensurate crystal phase in $KTm(MoO_4)_2$ compound

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Abstract

A differential magnetic susceptibility and an electron spin resonance are investigated in $KTm(MoO_4)_2$ single crystal at temperature 1.7 K at permanent magnetic fields up to 80 kOe. In the field range between 12 and 45 kOe a number of peculiarities of the differential susceptibility and the microwave absorption was found. It is assumed that these peculiarities are due to the sequence of alternating structural phase transition with commensurate and incommensurate structure ("devil's staircase") induced by external magnetic field.

Key words: phase transition, incommensurate phase, magnetic susceptibility, microwave absorption.

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Introduction

The investigation of low-dimensional systems, such as rare-earth double molybdates is of interest for a number of reasons. A strong anisotropy and competition of one-ion and different-ions anisotropy leads to peculiarities in energy spectrum of magnetic ion. A presence of considerable magnetic moment in rare-earth ion and rather small exchange interaction increase a role of dipole-dipole interaction. And finally in these low symmetrical magnetic crystals an essential lattice instability is noted as the result of electron and phonon subsystems interconnection and existence of degenerated or quasi-degenerated ground state of magnetic ion. As a result a variety of phase transitions is observed, which includes not only magnetic but structural (also of cooperative Jahn-Teller type) [1,2], ferroelectric and ferroelastic phase transitions. Phase transitions (among them structural ones) in these systems can be induced not only by the temperature but also by the magnetic field.

Recently [3] in studies of electron spin resonance and microwave absorption in single crystal $KTm(MoO_4)_2$ a low temperature structural phase transition induced by external magnetic field was found. At temperature 1.8 K when the external magnetic field is applied along a -axis of crystal (the smallest g -factor) under the forward and reverse variation of magnetic field, a drastic modification of the absorption line is observed at fields between 30 to 45 kOe. A considerable hysteresis (several kOe) is noted. It follows that observed peculiarities are due to a phase transition, evidently of the 1st type. The results were explained on the basis of theoretical models [4,5] of magnetic field induced structural phase transition in compounds with singlet ground state.

In this paper is more detail investigations of the peculiarities of this phase transition in $KTm(MoO_4)_2$ compound are presented.

Experimental results and discussions

$KTm(MoO_4)_2$ layered crystal belong to the family of double alkali rare-earth molybdates.

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$KTm(MoO_4)_2$ refers to the rhombic space group symmetry D_{2h}^{14} [6]. The lattice constant are: $a = 5.05 \text{ \AA}$, $b = 18.31 \text{ \AA}$, $c = 7.89 \text{ \AA}$. The unit cell contains four molecules. The rare-earth ions form chains along a -axis and the distance between ions is about 3.9 \AA . For Tm^{3+} in the ground state is 3H_6 with the lowest singlet level. But the first excited state derived from our

microwave absorption data is very close and is at the distance 2.3 cm^{-1} [7]. These two levels form quasi-doublet. The next state is spaced at 200 cm^{-1} .

The differential magnetic susceptibility dM/dH was measured at temperature 1.7 K for a magnetic field orientation along the a -axis of crystals (fig.1).

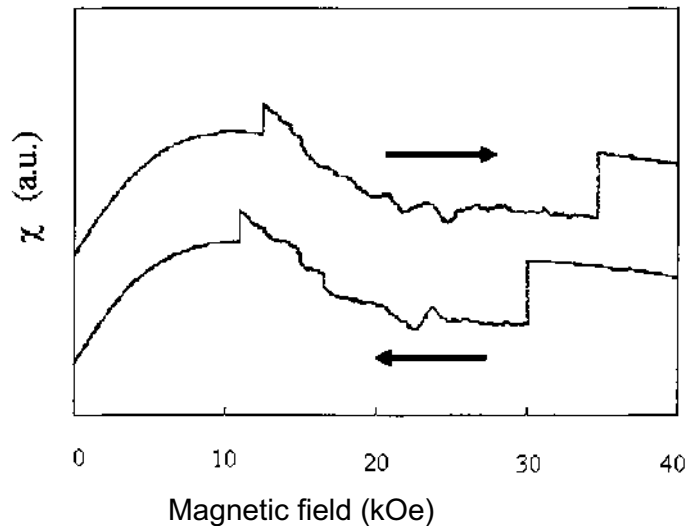


Fig.1. Field dependence of magnetic susceptibility of $KTm(MoO_4)_2$ at temperature 1.7 K and frequency 1 kHz. The arrows indicate a direction of permanent magnetic field variation

Jumps of dM/dH are observed at magnetic field of about 12 and 35 kOe. A remarkable hysteresis occurs in magnetic field, when passing forth and back. These anomalies all vanish as the temperature increases higher than 2 K. The jumps of susceptibility are caused by the first order phase transitions [3]. Strong instability of $dM/dH(H)$, which takes place in range between two phase transition is of special interest here.

More clearly these peculiarity are observed in microwave absorption spectra.

High frequency properties of $KTm(MoO_4)_2$ was studied at temperature 1.7 K at frequency 80 GHz at constant magnetic field, which changes up to 8 T. Two nonequivalent positions of Tm^{3+} were found rotated in plane ac in the angle $\pm 9^\circ$ with respect to a -axis with highly anisotropic ground level g -factor.

Figure 2 shows the absorption spectrum of the given crystals. The magnetic field is applied in direction close to a -axis. It is shown, that at small angles of declination from a -axis the sharp collapse of absorption line which is due to the first order phase transition [3], takes place on high field edge. This phenomena is accompanied by some stair like absorption process. The width of the stairs observed and the value of transition field depends strongly on defects in crystal, strains and rate of magnetic field deviation. An attention must be given to additional line at 25 kOe, which disappears at reverse field deviation. Origin of this line is not clear now.

Analysis of obtained experimental results leads to the conclusion that at magnetic fields in the 12-40 kOe a modulated superstructure of crystal lattice exists. This superstructure consists of the commensurate regions separated by very

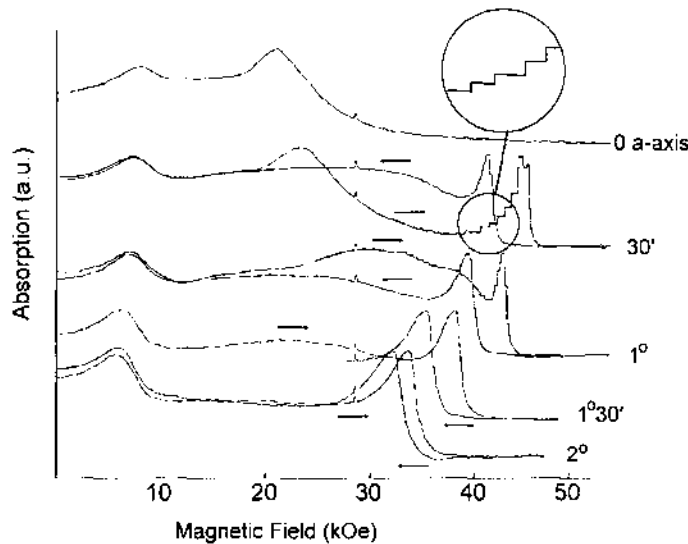


Fig.2. The angular dependencies of microwave absorption in $KTm(MoO_4)_2$ at 1.7 K, $\nu=80$ GHz. The arrows indicate a direction of permanent magnetic field variation.

narrow incommensurate regions (“devil’s staircase”). As it is shown by Djaloshinsky [8], commensurability of period of superstructure is energetically profitable. Therefore variation of period of incommensurate phase with temperature is jumplike, and incommensurate phase presents a temperature sequence of long periodic commensurate phases. In the given case of strong spin-lattice interaction an external magnetic field plays a role of temperature. The space symmetry of $KTm(MoO_4)_2$ allows for the existence of Lifshitz gradient invariant. Therefore incommensurate structure can arise as result of dynamic interrelation of electron excitation branch and acoustic phonon branch of crystal.

Here we present preliminary experimental results. The proposed explanation of the nature of observed peculiarities is only of descriptive character. Finally the question, if the modulated structure exists in the given case, can be solved only through direct experimental observation using X-rays or neutrons.

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