VOLUMES 194–196
FEBRUARY II 1994

ISSN 0921-4526

PHYSICA B

CONDENSED MATTER

Proceedings of the
20th International Conference
on Low Temperature Physics

LT-20

held in Eugene, Oregon, USA
4–11 August 1993

Part II – Contributed Papers

Guest Editor:
Russell J. Donnelly

PHYBE3 194–196 (1994) 1359–2424

NORTH-HOLLAND

Physica B 194–196 (1994) 453–454
North-Holland

PHYSICA B

First Raman spectroscopy observation of magnetic field induced structural phase transition in $TmPO_4$ crystals.

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Raman spectra measurements of the $TmPO_4$ crystal under external magnetic fields up to 10T are presented. At temperatures lower than 20K the magnetic field induced structural transition theoretically predicted earlier is observed. The appearance of a new finite frequency Raman mode in the spectra at the critical magnetic field is the main characteristic of the phase transition manifestation.

1. INTRODUCTION

New phenomena of the stimulated cooperative Jahn-Teller effect resulting in external magnetic field induced structural phase transitions in pure, diluted [1] and mixed [2] Jahn-Teller crystals were predicted earlier. This was experimentally observed approximately a year ago by means of magnetostriction measurements in strong impulse magnetic fields which induced a structural phase transition in $TmPO_4$ crystals at 4.2K [3]. In this report we present data of the first experimental observation of the magnetic field induced structural transitions by means of Raman spectral measurements of the $TmPO_4$ crystals.

2. STIMULATED COOPERATIVE JAHN-TELLER EFFECT

The cooperative Jahn-Teller (JT) effect results from the electron-electron correlations mediated through virtual phonon exchange. Intersite interaction leads to a structural phase transition if the JT molecular field caused by electron-phonon interaction is bigger than the effective energy gap between the orbitally non-degenerate ground and the excited electronic states. An external magnetic field can influence the outcome in two ways [4]. It can reduce or smear out the phase transitions depending upon the symmetry of the magnetic field and of the spontaneous strain. Such effects have been observed , for example, in $TbVO_4$ [5]. Alternatively in situations where the electron-phonon interaction is not large enough to drive a JT phase transition, the magnetic field can reconstruct the electronic spectrum decreasing the effective energy gap between vibronically mixed states. This decrease can result in a magnetic field induced phase transition. This phenomenon was predicted earlier [1,2] and named the stimulated cooperative Jahn-Teller (SCJT) effect.

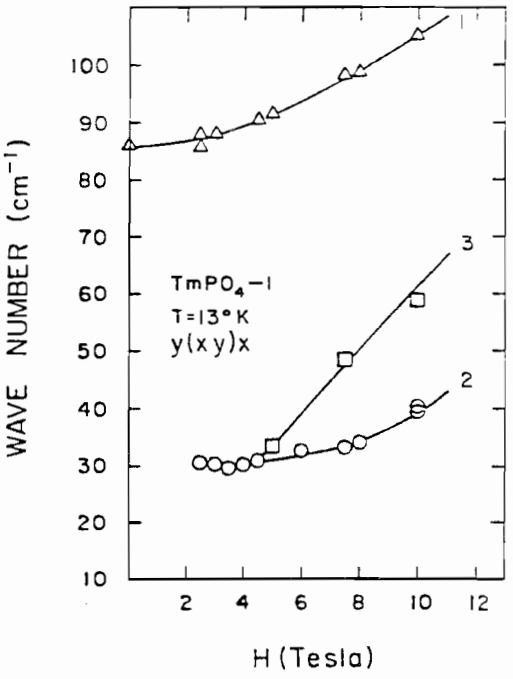
3. RESULTS

The low lying electronic excitation energies of the tetragonal $TmPO_4$ crystals were measured at T_1=13K, T_2=19K and T_3=26K as a function of the magnetic field from 0 - 10T using Raman scattering techniques [6]. The magnetic field was oriented along a [100] (not easy axis) direction.
At T=13K and T=19K a new electronic transition was observed when the magnetic field had reached the critical values. The results of the measurement at T_1 are presented in the first figure.The feature of interest is the appearance of a new mode

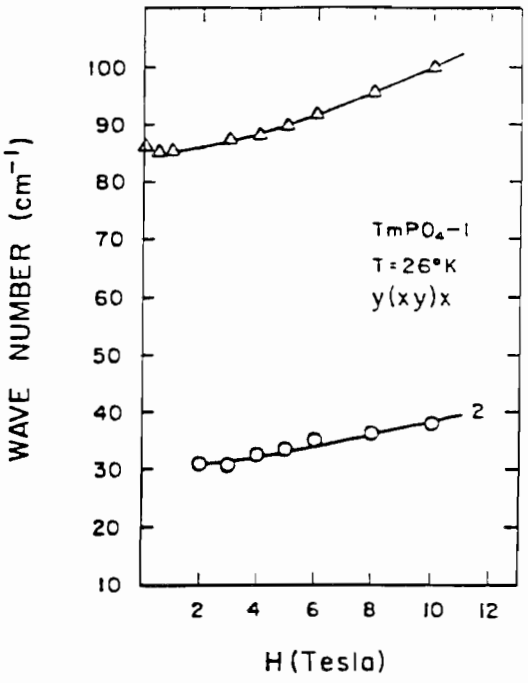
SSDI 0921-4526(93)E0806-R

labeled 3 at a finite frequency when the magnetic field value is equal or larger than the critical field.

At T=13K this mode appears at the critical magnetic field $H_{crit.}$=5.2T at the frequency E_3=32cm-1. We believe mode 3 is indicative of a SCJT phase transition and is associated with the transition from the ground singlet electronic state to the upper component of the first excited doublet, that has been split by the magnetic field. The transition is forbidden at $T>T_c$ when the spontaneous strain U_{B2g}=0. Mode 1 corresponds to the transition between the ground and first excited singlets and is present even at H=0. Mode 2 is connected with the mixing of the ground singlet and lowest component of the doublet, its appearance as can be seen from the figures is independent on the temperature.



The critical values of the magnetic field increase with the temperature [7]. At T=26K (second figure) no phase transition is observed as the applied magnetic field used was below $H_{crit.}$



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