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Crossover in the Van Vleck paramagnet $TmPO_4$

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Abstract

Magnetic anomalies caused by crossing of the lowest-lying energy levels (crossover) of Tm^{3+} ion in the magnetic field along the tetragonal axis of a $TmPO_4$ single-crystal near 30 T are investigated experimentally and theoretically. Measurements of the differential magnetic susceptibility dM/dH by the compensated pick-up coil method with different rates of up- and down-sweep of the magnetic field from 2 K to about 19 K have allowed studying temperature variations of width and shape of the dM/dH peak and hysteresis phenomena associated with relaxation processes. The influence of the misorientation effect and hyperfine coupling on the character of anomalies are explored. Comparison of experimental and theoretical data suggests a cooling of a sample, accompanied by an increase of spin-lattice relaxation times, when approaching the crossover in pulsed regime.

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Keywords: Energy level crossings; Magnetocaloric effect; Van Vleck paramagnets; Enhanced nuclear magnets

1. Introduction

In rare-earth (RE) compounds the interaction of the lowest-lying energy levels of the RE ion in a magnetic field (their crossing or approaching, crossover) are accompanied at low temperatures by pronounced anomalies of magnetic and magnetoelastic characteristics. The character of the anomalies and magnetocaloric effect at pulsed fields strongly depends on the nature of the energy level interaction. Investigations of crossover effects provide valuable information on the crystal field and other essential interactions in the compound under study.

The compound with the tetragonal zircon structure, space group $I4_1/amd$, $TmPO_4$ is characterized by a peculiar character of energy spectrum of the RE ion and belongs to a group of the singlet, or Van Vleck, paramagnets. The crystal-field splits the ground $^3H_6$ multiplet of the non-Kramers ion Tm^{3+} in such a way that the

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ground state is a singlet, with a low-lying doublet at about $30\,cm^{-1}$. At liquid helium temperatures only the singlet state is populated, and, unlike the rather similar compounds $TmVO_4$ and $TmAsO_4$, which have a doublet as the ground state, no cooperative Jahn–Teller transition occurs.

$TmPO_4$ is expected to be a suitable material for studies of enhanced nuclear spin systems. NMR results [1] indicate that an applied magnetic field produces an effective field at the site of Tm nuclei enhanced by a factor $(1+K)\approx 77.9$ through hyperfine interactions. Although the enhancement factor is considerable, it was expected that the ordering temperature of nuclear magnetic moments may be very low, possibly in the μK range, because of the small nuclear moment of Tm nuclei $\mu = 0.2308\mu_N$ and the large lattice constants in the zircon compounds. Bleaney et al. [2] predicted that the ordering temperature for the ^{169}Tm nuclear spins in $TmPO_4$ should be about 6 μK. Thus, $TmPO_4$ offers the possibility of reaching μK temperatures by adiabatic demagnetization from initial fields of less than 1 T. Indeed, the minimum temperature reached in adiabatic demagnetization experiments [3] was about 0.4 mK. In paper [2] the enhanced nuclear magnetic resonance spectrum of ^{169}Tm $(I = 1/2)$ in $TmPO_4$ has been measured from 1.6 to 30 K. It turned out to be highly anisotropic, with $\gamma/2\pi = 11.34$ and 276 MHz/T parallel and perpendicular to the c-axis, respectively, at the lowest temperatures. Above 4 K the values of $\gamma/2\pi$ become temperature dependent as excited levels become populated. These data allow a precise measurement of the principal values of the electronic susceptibility of the Tm^{3+} ions. Temperature and magnetic field dependences of the spin-lattice relaxation time of $TmPO_4$ were measured in Refs. [1,3] in the temperature range from 20 mK to 2 K at weak magnetic fields (<1 T). The spin-lattice relaxation time of Tm nuclei in $TmPO_4$ was shown to be temperature, field and frequency dependent. It may be of the order of 1 s in the temperature range of some decimals of Kelvin. No information on the spin-lattice relaxation times at high magnetic fields is available.

It was predicted in our previous works (see e.g. Ref. [4]) that in $TmPO_4$ a crossing of the lowest energy levels occurs at the field along the tetragonal axis near 30 T which should be accompanied by pronounced anomalies in magnetic and magnetoelastic properties of the compound at low temperatures. Indeed, a relevant maximum of the differential susceptibility dM/dH was observed in our work [5] at $T = 5$ K. This paper is devoted to the detailed experimental and theoretical investigations of peculiarities of energy level crossing effects in $TmPO_4$.

2. Experimental results

The measurements of the differential magnetic susceptibility are performed on single-crystal samples of $TmPO_4$ from 2 K to about 19 K by the compensated pick-up coil method in pulsed magnetic fields generated by the single-turn coil technique [6]. A preliminary dM/dH curve at 5 K and details of experimental technique are given in Ref. [5]. For this series of measurements the experimental set-up for the megagauss generator was completed and improved, the measuring system for magnetization worked with reduced noise and a bath cryostat for temperatures down to 2 K was usable.

As an example the differential magnetic susceptibility dM/dH of $TmPO_4$ at 2 K in a magnetic field along the tetragonal axis [0 0 1] is presented in Fig. 1. We note that the H-axis is cut off, the maximum field value for this run is 112 T (see Table 1, where the data for various temperatures and different maximum values of the field are given). As is seen from Table 1 and Fig. 2, where the peaks for down-sweep at five various temperatures are collected, sharp maxima of dM/dH take place for all temperatures except the highest one, 18.5 K. They are connected with crossing of the lowest-lying energy levels of the Tm^{3+} ion. At $T_0 = 18.5$ K the $dM/dH(H)$ curve has no very distinct maximum, indicating quasi-absence of magnetization jump at this temperature. Since the magnetic anomaly occurs around 30 T, it is recorded four times in the experiment. When evaluating the data it is considered (see Table 1) that data from up- and down-sweep are generally more significant than those obtained at the reversed field direction. It is seen from Fig. 1 and

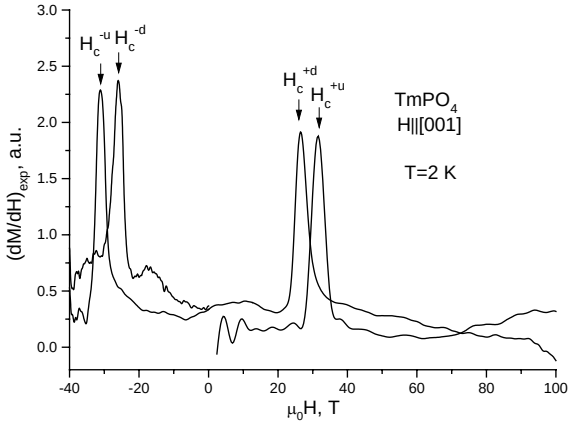

Fig. 1. Experimental differential magnetic susceptibility dM/dH of $TmPO_4$ for a magnetic field parallel to the tetragonal axis [0 0 1] at $T = 2$ K. H_c^{+u} and H_c^{+d} are the critical fields for the up- and down-sweep, respectively; H_c^{-u} and H_c^{-d} are the same for the reversed field direction.

Table 1
Experimental data for $TmPO_4$, H||[0 0 1]

T_0 (K)	H_{max} (T)	H_c^u (T)	H_c^d (T)	H_c (T)	ΔH_c (T)
2.2	39	31.0	30.7	30.8	0.3
	46	31.4	30.1	30.8	1.3
	55	31.8	29.1	30.4	2.7
	83	31.9	29.6	30.8	2.3
	112	31.5	26.5	29.0	5.0
2.2	55	31.8	29.1	30.4	2.7
4.2		31.6	30.0	30.8	1.6
8.9	58	31.6	30.2	30.9	1.4
14.3		31.3	30.5	30.9	0.8
16.3		31.4	30.5	31.0	0.9
18.5		31.3	30.9	31.1	0.4

Note: Maxima of dM/dH obtained at the initial temperatures T_0 occur at H_c^u and H_c^d in up- and down-sweep, respectively; $H_c = (H_c^u + H_c^d)/2$, and $\Delta H_c = H_c^u - H_c^d$, H_{max} is the maximum field of the pulse.

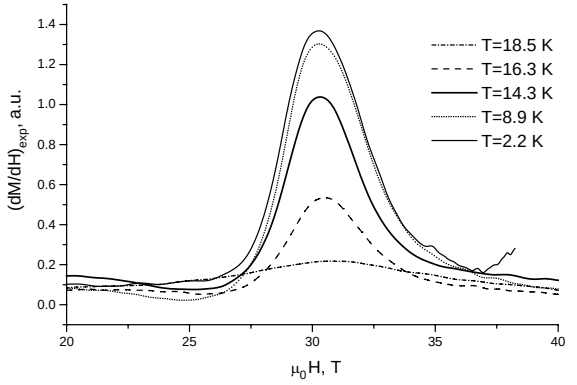

Fig. 2. Experimental differential magnetic susceptibility dM/dH of $TmPO_4$ for H||[0 0 1] at various initial temperatures. The peaks of dM/dH are shifted along the H-axis to coincide at $H = H_c$.

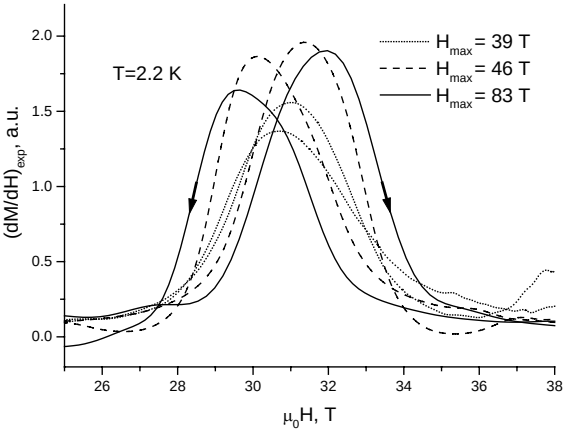

Fig. 3. Experimental differential magnetic susceptibility dM/dH of $TmPO_4$ for up- and down- sweep and various maximum fields H_{max} at $T = 2.2$ K.

Table 1 that hysteresis phenomena take place. The hysteresis width is quite different for different H_{max}, i.e. for different field sweep rates, the pulse duration for all runs is about 7 μs. Pronounced, about 5 T in width, hysteresis for $H_{max} = 112$ T (see Fig. 1) is definitely beyond the maximum error in a data/field correlation of 2 T (see Ref. [5]), and is caused by relaxation processes when magnetizing this rare-earth Van Vleck paramagnet and will be discussed in more detail in Section 3. This situation is displayed in Fig. 3, from which it is clear that the hysteresis is larger the higher field-sweep rate is. We note that for $H_{max} = 39$ T the hysteresis is very small and quasi-absent. The analysis of experimental data shows that the width of the dM/dH peaks do not depend on the maximum value of the field, and the peaks are practically of the same width independently of the temperature, it comprises about 3 T at a half-height.

The average low-temperature value of the crossover field H_c is 30.8 T. The experimental accuracy in this field range may be taken as 1.5 T. The run with $H_{max} = 112$ T is excluded from this estimation because of the highest field sweep rate and the maximum manifestation of relaxation processes.

3. Calculations and discussion

To calculate the Zeeman effect and magnetic characteristics, we use a Hamiltonian which incorporates the crystal-field Hamiltonian H_{CF} written in terms of the equivalent operators O_n^m, the Zeeman term H_Z and the hyperfine Hamiltonian H_N.

$$H = H_{CF} + H_Z + H_N, \quad (1)$$

$$H_{CF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_4^4 O_4^4 + B_6^4 O_6^4,$$

$$H_Z = g_J \mu_B \mathbf{HJ},$$

$$H_N = -\hbar\left[\gamma_{\|} H_z I_z + \gamma_{\perp}(H_x I_x + H_y I_y)\right].$$

Here B_n^m are the crystal-field parameters, g_J is the Lande factor, γ is the magnetogyric hyperfine tensor. The hyperfine Hamiltonian H_N is taken in the form of the spin-Hamiltonian because the values of hyperfine parameters $\gamma_{\|}$ and $\gamma_{\perp}$ in $TmPO_4$ used for our numerical calculations were found in Ref. [2] in this form. The crystal-field parameters B_n^m were deduced in Ref. [7] for the crystal-field Hamiltonian written in form (1) on the basis of the neutron spectroscopy data [8] and the crystal-field parameters deduced in Ref. [8] for a crystal-field Hamiltonian written in the irreducible tensor operators. They describe the level scheme of the low-lying levels [8] and the temperature variations of the initial susceptibilities of Refs. [7,8]. Moreover, they reproduce the crossover field H_c very well.

Calculation of the Zeeman effect for the field orientation along the tetragonal axis $H \parallel [0\,0\,1]$ shows crossings of the lowest levels near 30 T and in the vicinity of 500 T. These crossings manifest themselves as jumps in the appropriate magnetization curve at $T = 4.2$ K, as it is seen in Fig. 4 where

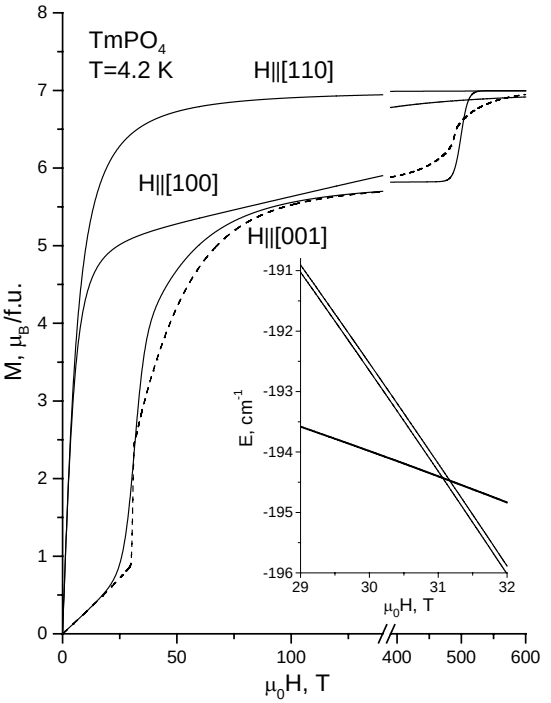

Fig. 4. Calculated isothermal (solid lines) and adiabatic (dashed line) magnetization curves of $TmPO_4$ at $T = 4.2$ K for the main crystallographic directions. A fragment of Zeeman effect at $H \| [0\,0\,1]$ for two lowest-lying levels with regard to the hyperfine coupling is displayed in the inset.

the magnetization curves for the main crystallographic directions are depicted. In the inset of Fig. 4 a pattern of Zeeman effect calculated with regard to the hyperfine coupling at $T = 4.2$ K is shown. Near 30 T the jump value is much larger than that for the field near 500 T. The same figure displays the adiabatic magnetization curves for the initial temperature $T_0 = 4.2$ K calculated when taking into account the magnetocaloric effect. It is seen that for the field direction with crossovers the adiabatic magnetization curve differs from the isothermal one.

Calculation of adiabatic magnetization curves is necessary to interpret the magnetization processes at pulsed fields with small pulse durations. As was shown in Ref. [9] on the example of the RE paramagnetic garnets, the magnetization process in RE paramagnets at pulsed fields can be considered as being close to adiabatic for the field sweep rates exceeding 10^3 T/s. In our

measurements the field sweep rate ($\geqslant 10^7$ T/s) exceeds greatly this value in the entire field region of interest. For the situation of $H_{max} = 39$ T the magnetization curves do not manifest hysteresis (Fig. 3), this provides evidence that the magnetization process is close to reversible adiabatic. We note that for all available H_{max}, i.e. for all field sweep rates, the peaks in $dM/dH(H)$ curves are very similar in width and shape (see Section 2), though for $H_{max} > 39$ T hysteresis phenomena manifest themselves. This gives us a possibility to interpret the experimental data for the peaks in $dM/dH(H)$ curves associated with crossover on the basis of calculations performed on the assumption of an adiabatic magnetization process.

To deduce a spectrum and wave functions of the Tm^{3+} ion, a numerical diagonalization of the Hamiltonian (Eq. (1)) is carried out for each specific field value from 0 to 500 T with a step size small enough to provide a conversion of the numerical procedure, and a "partial" magnetocaloric effect ΔT under the field change from H to $H + \Delta H$ is calculated:

$$\Delta T = -T\left(\frac{\partial M}{\partial T}\right)_H \frac{\Delta H}{C_H} \equiv -T\left(\frac{\partial S_{mag}}{\partial H}\right)_T \frac{\Delta H}{C_H}. \quad (2)$$

In Eq. (2) the total heat capacity C_H includes the lattice heat capacity $C_{lat} = 12\pi^4/5k_B v(T/T_D)^3$ (the Debye temperature for the zircon lattice being $T_D = 275$ K [10], $v = 6$) and the magnetic heat capacity C_{mag} calculated for each specific value of the field and temperature on the basis of the energy spectrum of the RE ion with regard to the hyperfine coupling.

As is seen from Eq. (2) a sign of the magnetocaloric effect is governed by the sign of the derivative $(\partial M/\partial T)_H$ or, in accordance with the general thermodynamic identity, by the sign of the change of the magnetic part of entropy with increasing field. In the case of increasing S_{mag}, cooling under magnetization is possible, which is not usual for paramagnetic systems. The magnetic contribution to the entropy, which is associated with the electronic and nuclear magnetic moments coupled by the hyperfine interaction and exposed to a magnetic field, is given by the expression (per one RE ion):

$$S_{mag} = k_B\left\{\ln\sum_i \exp(-E_i/T) + \frac{\langle E\rangle}{T}\right\}, \quad (3)$$

where E_i are the eigenvalues of Hamiltonian (1) and $\langle E\rangle$ is the mean energy. We discuss some properties of S_{mag} and C_{mag} in the isothermal regime.

The field dependence of the magnetic part of the entropy for two temperatures 0.3 and 0.6 K is shown in Fig. 5. It is seen that up to the fields rather close to H_c the entropy S_{mag} does not depend on the field, because the density of states for the ground singlet split by the hyperfine interaction does not depend on the field and the excited levels do not contribute to the entropy at these temperatures. When the crossing of the levels occurs the entropy reaches its maximum value. After the crossover, at $H > H_c$, the decrease of the

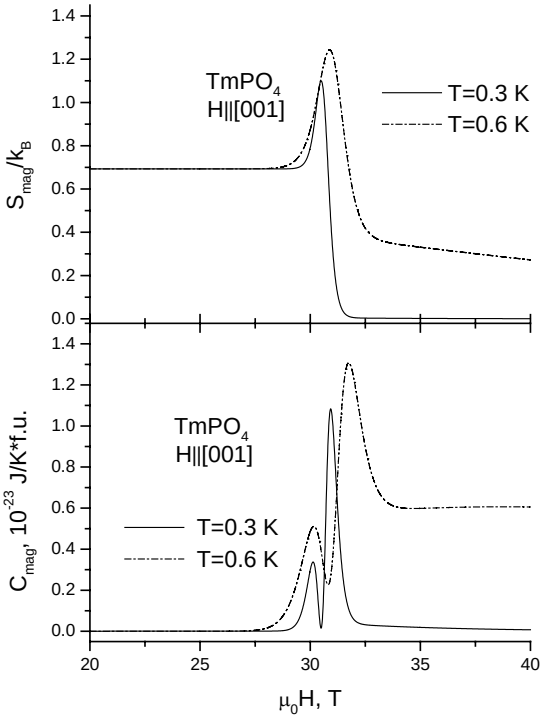

Fig. 5. Field dependences of the magnetic part of the entropy (upper part) and the magnetic heat capacity (lower part) of $TmPO_4$ (per formula unit) for $H||[0\,0\,1]$ and the temperatures $T = 0.3$ and 0.6 K.

density of states results in the decrease of the entropy. At $T = 0.3\,\mathrm{K}$ it practically vanishes because at this temperature even the low-lying sublevel of the ground (at $H > H_c$) level (see inset in Fig. 4) does not give any contribution to the entropy, in contrast to the situation at $T = 0.6\,\mathrm{K}$ when such a contribution exists. A larger maximum value of the entropy at 0.6 K is also due to the contribution of the upper hyperfine component of the level approaching from above.

Analysing in an analogous manner the field dependence of the magnetic heat capacity, which is displayed in Fig. 5 for the same temperatures 0.3 and 0.6 K, we can point out the different heights of maxima which are the Schottky-type anomalies and a greater heat capacity value after crossover at $T = 0.6\,\mathrm{K}$ than at 0.3 K. These features are due to different hyperfine splitting of the levels which act as a ground level prior to and after the crossover (see inset in Fig. 4).

Field dependences of the adiabatic susceptibility $\mathrm{d}M/\mathrm{d}H$ calculated for $H||[0\,0\,1]$ and various initial temperatures T_0 give the crossover field equal to 30.7 T, in a very good agreement with the experimental result of $(30.8 \pm 1.5)\,\mathrm{T}$. They show that the peaks of $\mathrm{d}M/\mathrm{d}H$ are very similar in width for different T_0, the case of $T_0 = 18.5\,\mathrm{K}$ excluded. This is in accordance with the experimental data.

If one restricts Hamiltonian (1) to the first two terms $H_{\mathrm{CF}} + H_{\mathrm{Z}}$, the calculations of the adiabatic magnetization process in the field strictly parallel to the tetragonal axis result in the overcooling in the crossover for the initial temperatures $T_0 < 18\,\mathrm{K}$. It means that such calculations are incorrect. A consideration of the nuclear Zeeman interaction,

$$H_{ZI} = -\gamma_{\mathrm{I}}\hbar\mathbf{H}\mathbf{I}, \tag{4}$$

(γ_I is the nuclear gyromagnetic ratio) enables one to take into account a nuclear heat capacity and shifts the overcooling problem to the very low initial temperatures ($< 0.1\,\mathrm{K}$). The inclusion of the hyperfine coupling leads to correct calculations of adiabatic magnetization process in Van Vleck paramagnets at any temperature interval.

As our calculations have shown, when tilting the magnetic field from the tetragonal axis the manifestation of the hyperfine coupling falls off, and for the misorientation angle $\vartheta \geqslant 1^\circ$ the hyperfine coupling becomes unessential. When analysing the experimental data for $\mathrm{d}M/\mathrm{d}H(H)$ at various T_0 in $TmPO_4$, an inevitable misorientation of the field should be considered and the misorientation effect should be calculated and discussed. Our investigations of the magnetic anomalies caused by the crossover in another Van Vleck paramagnet, $PrVO_4$, [11] have revealed that the misorientation effect is negligible in this compound. In contrast to $PrVO_4$, the misorientation effect in $TmPO_4$ should be very substantial, as it follows from the electronic structure of the Tm^{3+} ion in this compound. In $TmPO_4$, the level which is strongly mixed by a perpendicular to the tetragonal axis component of the field with the ground level, thus giving an essential contribution to the perpendicular susceptibility, is the first excited level which approaches to the ground level and crosses it in the crossover. In $PrVO_4$ a similar level is the second excited level ($\Delta \sim 84\,\mathrm{cm}^{-1}$) and it does not participate in the crossover.

Calculated magnetization curves $M(H)$ and magnetocaloric effect $T(H)$ are given in Fig. 6 for $\vartheta = 1^\circ$ and the initial temperatures T_0 available in experiment. This value of the misorientation angle is estimated from the analysis of our experimental data. For all initial temperatures, $T_0 = 18.5\,\mathrm{K}$ excluded, near the crossover a jump of magnetization takes place, its value being decreased with rising T_0 because of a more limited cooling of the crystal near crossover. The decrease in temperature when approaching the crossover is caused by the increase in the magnetic contribution to the entropy which occurs when the levels of the RE ion approach one to another, as it was already discussed. The calculated magnetocaloric effect refers to the magnetic subsystem of the compound under examination. Whether the temperature of the sample is diminished to the calculated values depends on the spin-lattice relaxation times, and the comparison of calculated and observed widths of the $\mathrm{d}M/\mathrm{d}H$ peak at low temperatures may provide some information about them.

To investigate the influence of the form of the hyperfine Hamiltonian on the results obtained we have performed calculations also with the use of

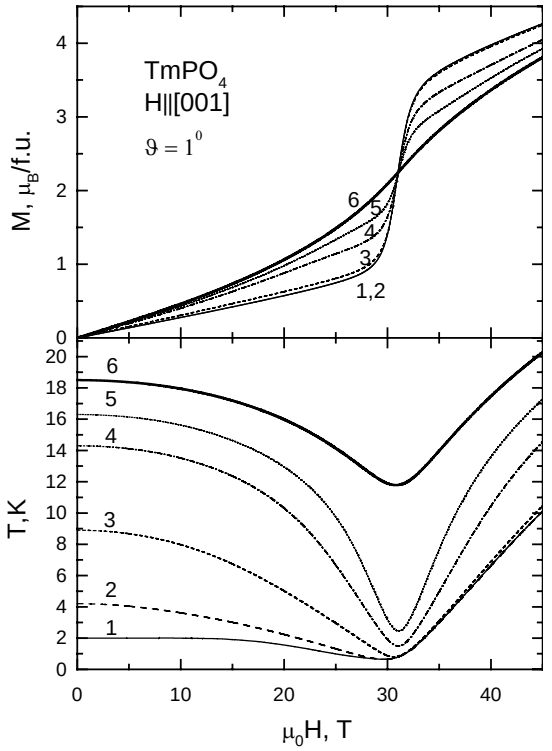

Fig. 6. Calculated adiabatic magnetization curves (upper part) and magnetocaloric effect (lower part) of $TmPO_4$ at the magnetic field **H** tilted by 1° with regard to the [0 0 1] direction for various initial temperatures T_0.

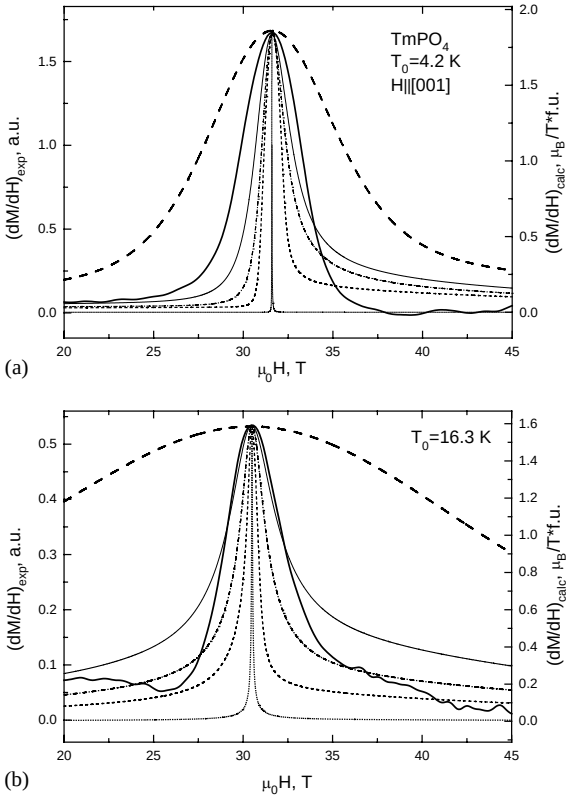

Fig. 7. Comparison of experimental differential magnetic susceptibility dM/dH (thick solid lines) with calculations for $TmPO_4$ at $T_0 = 4.2$ K (a) and 16.3 K (b). Thick dashed lines represent isothermal susceptibility multiplied by 5.4 for $T_0 = 4.2$ K and by 12.2 for 16.3 K. Calculated adiabatic susceptibility are given: by thin solid lines for the misorientation angle $\vartheta = 1°$, multiplied by 1.8 for $T_0 = 4.2$ K and by 3.1 for 16.3 K; by dash-dotted lines for $\vartheta = 0.5°$, multiplied by 1.2 for $T_0 = 4.2$ K and by 1.7 for 16.3; by thin dashed lines for $\vartheta = 0°$ and hyperfine parameters from Ref. [2] and by dotted lines for $\vartheta = 0°$ and free-ion hyperfine coupling, multiplied by 1/134 for 4.2 K and by 1/15 for 16.3 K.

the free-ion hyperfine Hamiltonian

$$H_{\rm hf} = A_{\rm J}\mathbf{JI}, \tag{5}$$

where for $^{169}Tm^{3+}$ $A_{\rm J} = -393.5$ MHz [12].

All calculated $dM/dH(H)$ curves together with experimental ones (shifted along the H-axis to coincide at $H = H_c$) are presented for the initial temperatures 4.2 K (Fig. 7a) and 16.3 K (Fig. 7b) taken as an example. As can be seen from Fig. 7, the peaks of the isothermal susceptibility calculated at the appropriate temperatures are wider than the experimental ones, which gives evidence for a cooling of the sample. The maxima of the susceptibility, calculated in the adiabatic regime for $H||[0\,0\,1]$ as described above, are too narrow (several times narrower than the experimental ones when calculated with the hyperfine parameters from Ref. [2] and still narrower when calculated for the free-ion hyperfine coupling). Let us discuss this point.

The reasoning given in Ref. [13] has shown that possible broadening caused by the measurement system is ruled out, the corresponding estimate of the relevant contribution gives ~0.4 T, i.e. much less than the actual width of peaks (~3 T).

Generally speaking, near the crossover a magnetic ordering can appear due to some pair interactions in the situation when a degenerate, or nearly degenerate, ground state arises.

A magnetic ordering is characterized by a deviation of the magnetic moment out of the field direction near H_c and hence by the broadening of the dM/dH peak. The magnetic-field-induced phase transitions of this sort in the vicinity of crossover were considered from the theoretical point of view in various systems with a singlet ground state and were observed in some d-compounds, e.g. in $Cu(NO_3)_2 \cdot 2.5H_20$ [14] and $CsFeCl_3$ [15] when measuring the magnetization curves. As our calculations have shown, magnetic ordering in a magnetic field near the crossover is of little probability in $TmPO_4$, because of small exchange and dipole interactions in this compound in particular. So, it cannot be a reason of the broadening of the dM/dH peaks.

A conceivable misorientation of the magnetic field with respect to the tetragonal crystalline axis results also in a widening of the dM/dH peaks. Indeed, as is seen from Fig. 7 the inclusion of misorientation in 0.5° improves the agreement between calculations and experiment. However, increasing the misorientation angle ϑ to 1° shows that the misorientation effect is not responsible in full measure for this disagreement. For $\vartheta = 1^\circ$ the calculated peaks become closer in width to experimental ones, but some distinctions in their configuration therewith appear. They are, for instance, the "wings" caused by perpendicular susceptibility which are clearly seen in the calculated for $\vartheta = 1^\circ$ $dM/dH(H)$ curve at $T_0 = 16.3$ K. The experimental data do not show the appropriate perpendicular susceptibility which should accompany the perpendicular to the tetragonal axis component of the field and which has an appreciable value for the misorientation angle of 1° because of the very high anisotropy of this Van Vleck compound.

In the experiment an equal misorientation angle was maintained for all initial temperatures. Determining it from the situation at $T_0 = 16.3$ K as $\leqslant 1^\circ$ we see that for lower temperatures (see Fig. 7a, the case $T_0 = 4.2$ K) the calculated on the assumption of an adiabatic magnetization process peak is narrower than that in experiment for any hyperfine parameters. Hence, the discrepancy between experimental and calculated peak widths manifests itself to a greater extent in the low-temperature curves with a cooling near crossover to the lower temperatures. This fact suggests a possibility of the increase of spin-lattice relaxation times when, in the vicinity of crossover, the temperature may decrease to some decimals of Kelvin. An increase of the spin-lattice relaxation times means that the actual magnetization process is not adiabatic. If the processes of spin-lattice relaxation at low temperatures and high fields are slow, this means the breaking of bonds between the magnetic subsystem and the lattice under the conditions of magnetization in pulsed fields of high sweep rate.

4. Conclusions

To conclude we emphasize that our measurements for various maximum field values at a given temperature have shown the dependence of the hysteresis width on a maximum field value, i.e. on a field sweep rate (the pulse duration is about 7 μs for all runs). The higher the field sweep rate is, the larger is the hysteresis (see Table 1 and Fig. 3). This fact gives evidence for the great role of relaxation processes in pulsed fields with large sweep rates and may also be ascribed to the increase of the spin-lattice relaxation times in the range of temperatures of the order of 0.1 K and high magnetic fields.

Our theoretical consideration of experimental data has allowed accounting for main data of magnetic anomalies in $TmPO_4$ connected with the crossover. Comparisons of experimental and calculated findings for the peaks of differential magnetic susceptibility dM/dH have provided a possibility of discussing the misorientation effect and an increase of the spin-lattice relaxation times in the low-temperature region in the vicinity of crossover.

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