Investigation of level crossing in the tetragonal paramagnet $YbPO_4$ in ultrahigh magnetic fields up to 400 T

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The crossing of the energy levels of Yb^{3+} ions in paramagnetic $YbPO_4$ in ultrahigh magnetic fields of up to 400 T, produced by an explosive method, is investigated experimentally and theoretically. A wide maximum is found in the differential susceptibility dM/dH in a field $H_c \approx 280$ T. This maximum is due to the crossing of the energy levels of the magnetic ions in the field. The magnetocalorimetric effect is calculated under the assumption that the magnetization process in the pulsed fields is adiabatic. The effect is nonmonotonic as a function of the field and is accompanied by a substantial cooling of the crystal near H_c. *© 1997 American Institute of Physics.*
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Crossing of the energy levels of magnetic ions in a field (crossover) and an associated jump in magnetization have been predicted theoretically[1] for the compound TmSb and have been observed experimentally in various rare-earth (RE) compounds (see, for example, Ref. 2) in both paramagnetic and ordered states. This effect occurs when the energy of the lower level of the crystal-field-split ground-state multiplet of the RE ion depends on the magnetic field much more weakly than the energy of one of the excited multiplets. For this reason, in sufficiently high fields these levels first draw together and then exchange places. Since in this case the more ''magnetic'' level becomes the ground-state level, crossover is accompanied by an abrupt increase in the magnetization M and a maximum in the differential susceptibility dM/dH.

It can be expected that level crossing effects will be quite numerous in the RE zircons RXO_4 (R is a rare-earth ion, X = As, P, V), since the quite low tetragonal symmetry of zircon (space group $D_{4h}^{19} = I4_1/amd$) gives a rich, weakly degenerate, spectrum of the RE ion and an appreciable magnetic anisotropy in directions parallel to and perpendicular to the tetragonal axis in the paramagnetic state. Since there are no nonequivalent positions for a RE ion in the zircon structure, these effects should be clearly

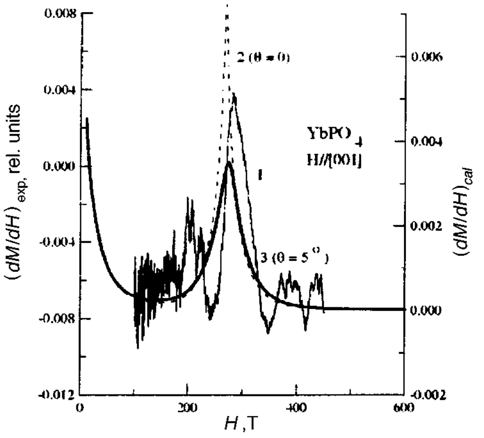

FIG. 1. Experimental (*1*) and computed (*2, 3*) dependences of the differential susceptibility dM/dH for $YbPO_4$ for magnetic field orientation near the tetragonal axis [001] (θ — angle of orientation).

observed even in macroscopic characteristics such as the magnetization. Of the entire series of RE zircons, crossover in comparatively weak magnetic fields (~ 10 T) has been investigated thus far only in paramagnetic $HoVO_4$.[3–5] The substantial progress made in producing high and ultrahigh magnetic fields makes level crossing effects now accessible for experimental investigation. In the present work we investigated level crossing in paramagnetic $YbPO_4$ both experimentally and theoretically.

The measurements were performed on a $YbPO_4$ single crystal at 4.2 K by an induction method in pulsed magnetic fields of up to 400 T, produced by an explosive method.[6] The rise time of the field in the pulse was equal to 15 μs. These are one-time measurements and the measuring coils and samples are destroyed after each pulse; it is impossible to compensate the signal from the magnetic field completely. Therefore the signal induced in the measuring coils can be written in the form

$$V_1(H) \sim \frac{dM}{dt} + K\frac{dH}{dt}, \tag{1}$$

where the first term corresponds to the signal from the sample and the second term corresponds to the coil decompensation signal. During the field pulse the signals V_1 and $V_2 \sim dH/dt$ from the measuring and "field" coils were recorded every 0.002 μs (approximately 8000 points). These data make it possible to calculate the curves $V_1(H)/V_2(H)$ (which, assuming that the decompensation signal is only a weak function of the field intensity, are proportional with proportionality constant K to the differential susceptibility of the sample $dM/dH = (dM/dt)/(dH/dt)$ and also to time-average the signal in order to decrease the high-frequency fluctuations of the background.

Figure 1 displays the experimental and theoretical curves of dM/dH for a $YbPO_4$ single crystal for a magnetic field oriented along the tetragonal axis [001]. The wide maximum of the susceptibility at $H_c \approx 280$ T is due to the crossing of the energy levels of the Yb^{3+} ion. The large width of the maximum is apparently due to the change in the temperature of the sample as a result of the magnetocaloric effect accompanying magnetization in a pulsed field. Since the spin–lattice relaxation time in RE ionic compounds is very short ($\sim 10^{-9}$ s; see, for example, Ref. 7) compared with the duration of the field pulse, the electronic subsystem and lattice are in equilibrium during the measurements and the magnetization process is close to adiabatic. In our view, the heat transfer between the sample and the surrounding medium is very weak, even in pulsed fields with duration ~ 10 ms, as is indicated by the fact that the magnetization curves with increasing and decreasing field are the same.

The Zeeman effect and the magnetic characteristics were calculated using a Hamiltonian that includes the crystal-field Hamiltonian, written in terms of the irreducible tensor operators C_q^k, and a Zeeman term:

$$H = B_0^2 C_0^2 + B_0^4 C_0^4 + B_0^6 C_0^6 + B_4^4 (C_4^4 + C_{-4}^4) + B_4^6 (C_4^6 + C_{-4}^6) + g_J \mu_B \mathbf{H} \cdot \mathbf{J}. \tag{2}$$

Here B_q^k are the crystal-field parameters and g_J is the Landé factor. The crystal-field parameters for the Yb^{3+} ion in the phosphate matrix are known only for the doped compounds $Yb:LuPO_4$ and $Yb:YPO_4$ (Ref. 8) (the parameters B_0^2 of these compounds are substantially different) and can, generally speaking, differ appreciably from the values for the concentrated $YbPO_4$ compound. The crossover field H_c is determined by the spectrum and wave functions of the ground-state multiplet $2F_{7/2}$ of the Yb^{3+} ion, which are formed by the crystal field. Our calculations of H_c with the crystal-field parameters for $Yb:YPO_4$ and $Yb:LiPO_4$ give values of 210 T and 180 T, respectively. Numerical analysis shows that the value of the crossover field is most sensitive to the parameter B_0^2. Solving the optimization problem for $YbPO_4$ taking account of all available information (from spectroscopy and ESR[8,9] as well as our magnetic measurements) gives crystal-field parameters which fall within their range of variation in the series of RE phosphates and yields the theoretical value $H_c = 270$ T. This value agrees well with the experimental value within the error limits associated with measurement of the field ($\pm 10\%$) and a possible disorientation of the sample of $\leqslant 3°$.

An interesting feature of crossover in $YbPO_4$ is that the ground-state level ($g_z^{\mathrm{gr}} \sim 2$) crosses the bottom level of the first excited doublet, for which the z-component of the g-tensor in the absence of a magnetic field is much smaller ($g_z^{\mathrm{ex1}} < 0.1$). In a field, however, the third excited doublet is strongly admixed to the bottom level of the first excited doublet of the state $|+7/2\rangle$, both states belonging to the same representation, and a large increase occurs in the g_z factor of the latter. For the ground-state doublet g_z does not change as strongly in a field because of the weak admixture of the second excited doublet to it. We note that for a field orientation strictly in the direction of the tetragonal axis there is no mixing of the wave functions of the ground-state and first excited doublets and for this reason the ‘‘true’’ level crossing should be observed. A misorientation of the field by even 3–5° causes components $|\pm 7/2\rangle$ to appear in the wave function of the ground state and in the interaction between the ground-state and first excited levels, which gives rise to a small gap (level repulsion) in the spectrum near the crossover fields. This results in broadening of the maximum in dM/dH and increases the crossover field.

In calculating the magnetic characteristics for fields from 0 to 400 T with step $\Delta H = 0.01$ T, the Hamiltonian (2) was diagonalized numerically in order to determine the

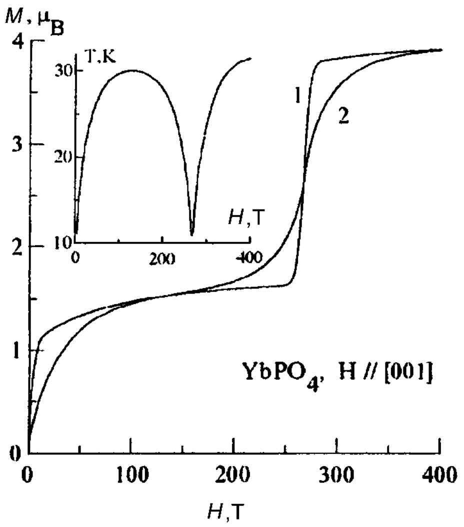

FIG. 2. Isothermal (*1*) and adiabatic (*2*) magnetization curves $M(H)$ for $YbPO_4$ for the initial temperature $T_0=4.2$ K and variation of the temperature $T(H)$ of the sample due to the magnetocaloric effect accompanying adiabatic magnetization.

spectrum and the wave functions of the Yb^{3+} ion, and the ''elementary'' magnetocaloric effect ΔT accompanying a change in the field from H to $H+\Delta H$ was calculated:

$$\Delta T=-(\partial M/\partial T)_H T\Delta H/C_H. \tag{3}$$

In this formula the total specific heat C_H of the crystal includes the lattice specific heat $C_{\text{lat}}\sim(T/T_D)^3$ (the Debye temperature for the phosphate lattice $T_D=275$ K[10]) and the magnetic specific heat C_{mag}, calculated for each value of the field and temperature on the basis of the spectrum of the RE ion. These data made it possible to calculate the isothermal and adiabatic magnetization of $YbPO_4$ and the temperature of the sample as a function of the magnetic field (Fig. 2). The latter function is nonmonotonic, i.e., at first the sample is heated by approximately 25 K and then it cools down by approximately 20 K in the region of the crossover fields. The sign of the ''elementary'' magnetocaloric effect is determined by the sign of the derivative $(\partial M/\partial T)_H$. For isothermal magnetization curves with jumps this derivative is positive as the crossover field is approached (heating smooths the jumps), which explains the cooling of the crystal near crossover.

Level-crossing investigations yield a great deal of information about the spectrum and wave functions of the RE ion, and ultimately their purpose is to investigate the crystal field. The good agreement between the computed and experimental data for $YbPO_4$ confirms that the magnetization process in an ultrahigh field is nearly adiabatic. Depending on the character of the levels participating in crossover, both heating and cooling of the sample can be observed near crossover. Cooling of the crystal gives anomalies of the magnetic characteristics that in a number of cases are even more pronounced than for an isothermal process in static fields. This makes it possible to investigate crossover effects by performing magnetic measurements in high and ultrahigh pulsed fields. We underscore that, as our calculations show, such effects can be expected for an entire series of compounds from the group of RE oxides with the zircon structure.

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