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Critical behavior study of the spin ordering transition in RVO_3 (R = Ce, Pr, Nd, Sm, Gd, Er) by means of *ac* photopyroelectric calorimetry

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ABSTRACT

The thermal diffusivity of RVO_3 single crystals (R = Ce, Pr, Nd, Sm, Gd, and Er) has been measured with an *ac* photopyroelectric calorimetry in the region in which the G-type orbital ordering and C-type spin ordering take place. Detailed measurements in the close neighbourhood of the spin ordering temperature have allowed to extract the critical parameter α and the critical ratio A^+/A^- for this transition. While the samples containing Ce, Nd, Sm and Er belong to the 3D-XY universality class (showing that the spins have an easy plane anisotropy), the sample with Gd, which is known to present a clear easy axis, belongs to the 3D-Ising class. Finally, $PrVO_3$ shows an effective isotropic behavior, as the critical parameters found agree with the 3D-Heisenberg class.

1. Introduction

RVO_3 family of vanadium oxides (R being a rare earth ion from La to Lu and Y) has been extensively studied along the last twenty years because of the very interesting magnetic and electronic properties they present due to spin-charge-orbital coupling. Everything started with the study of anomalous diamagnetism in $LaVO_3$ [1] followed by the discovery of a magnetization reversal phenomena in several compounds of the family [2–4] and the more recent finding of multiferroic properties [5]. A wide variety of studies on the crystallographic and magnetic properties have been developed for samples with different rare earth ions showing the similarities and, specially, the variety of the interactions and their couplings depending on the particular ion.

These orthovanadates have in common with manganites that they are both correlated electron systems, experiencing a Jahn-Teller interaction which is weaker in the first group (the orbital-active electrons belong to the t_{2g} orbital in this case while in manganites it is e_g). The relationship among spin, orbital and lattice degrees of freedom is more subtle and complex in the vanadates [6,7]. All members of the series RVO_3 crystallize in the *Pbnm* space group at room temperature and experience, on lowering the temperature, first an orbital ordering transition together with a lattice distorsion induced by the collective Jahn-Teller coupling (which leads to a monoclinic phase) and, below, a spin-ordering one, both of them continuous, with the exception of $LaVO_3$ where the spin ordering precedes the orbital ordering and for which the latter has a first order character [4,8,9]. A particular case is $CeVO_3$, where, depending on the particular crystal used for the study, both situations can appear [8–11]. Different studies have made it clear that the orbital ordering is of the G-type while the spin ordering is of the C-type, which means that the V^{3+} spins are antiferromagnetically ordered in the *ab* plane while ferromagnetically aligned along the *c* axis. From $DyVO_3$ to $LuVO_3$ there is still another transition at a much lower temperature which is a concomitant orbital and spin ordering transition, with a first order character. The new spin ordering is now G-type with an antiferromagnetic coupling between the V^{3+} spins in all directions while the new orbital ordering is C-type [9,12,13].

Besides the general picture presented above for all RVO_3, more detailed studies have been undertaken for particular members of the family, showing that there are important differences in the details among them, starting with the position of the different transitions, which is clearly shown in Fig. 1 from Ref. [9]. In particular, the temperature of the first spin ordering transition is reduced as the atomic radius of the rare earth ion is decreased, possibly due to the increase of the exchange interaction, while the

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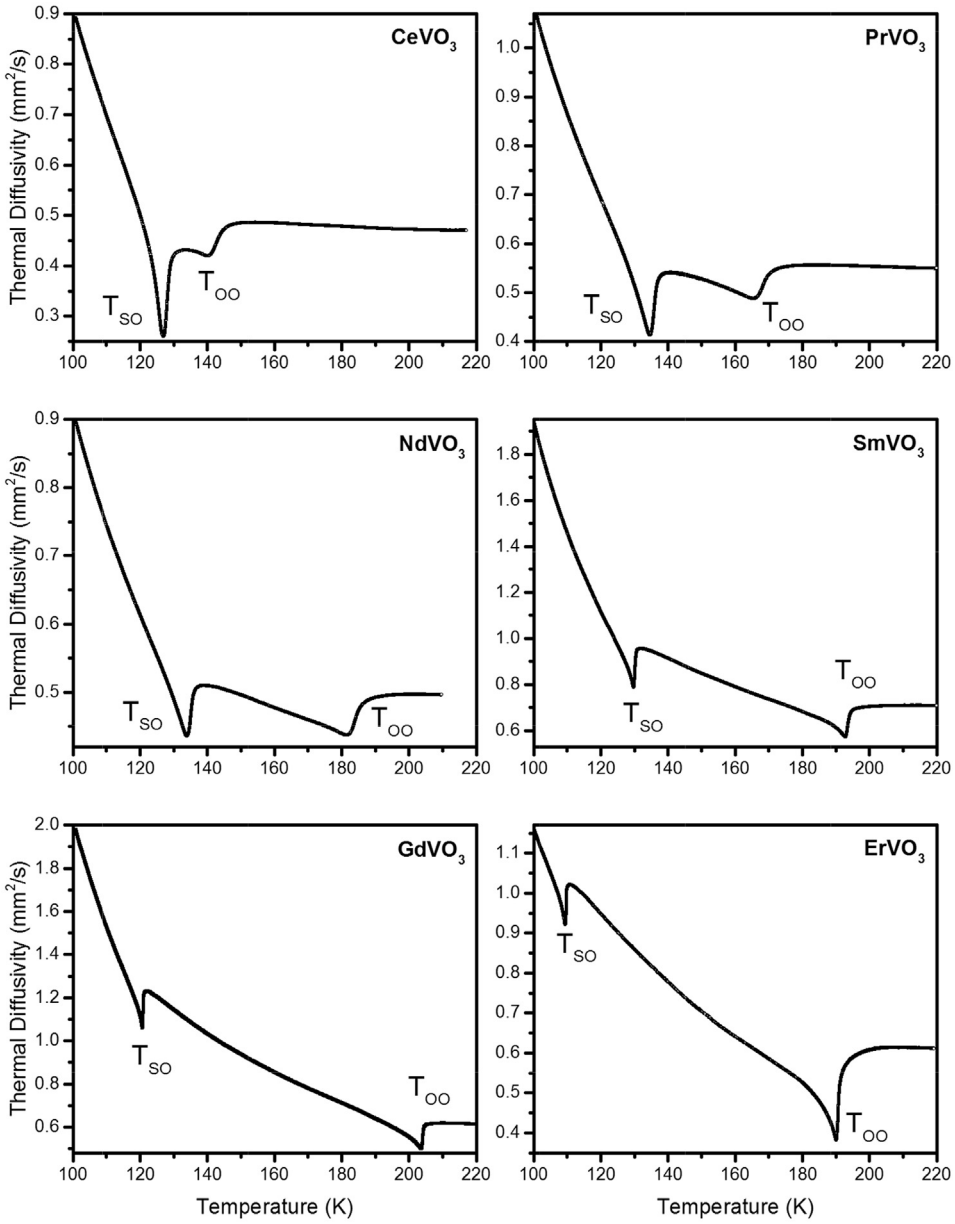

Fig. 1. Thermal diffusivity as a function of temperature for RVO_3 (R = Ce, Pr, Nd, Sm, Gd, and Er) showing the orbital ordering transition at T_{OO} and the spin ordering one at T_{SO}.

higher orbital ordering transition temperature reaches a maximum for an intermediate value of that radius, which has been suggested to be due to the competition between the increase of the orbital exchange interaction and the suppression of the Jahn-Teller instability [15]. Other differences arise in the particular direction in which the V^{3+} spins are placed within the C-type spin ordered phase, which has not been studied for all ions yet. Literature results point to differences between the two big groups (La to Tb, Dy to Lu) [16], some cantings have been proposed [12,17–19] and in some cases particular easy axes have been found for $SmVO_3$ [20] or $GdVO_3$ [21], while PrVO3 has been found to behave as a disordered antiferromagnet with random fields [22].

The aim of this work is to shed some light on this last issue: how spins are displayed in the C-type spin ordered state of RVO_3 (R = Ce, Pr, Nd, Sm, Gd, Er). An interesting support can be brought from the critical behavior theory and the universality classes theorized within the framework of renormalization group theory. In the critical region around a second order magnetic transition, several physical magnitudes behave critically after the following equations, where t is a reduced temperature $t = (T\text{-}T_C)/T_C$ and T_C the temperature of the transition [23]:

-specific heat $c_p(T) \sim A^{\pm} |t|^{-\alpha}$ (A^- for $T < T_C$, A^+ for $T > T_C$), (1)

-spontaneous magnetization $M_S(T) \sim |t|^{\beta}$ ($T < T_C$), (2)

-inverse of initial susceptibility $\chi_0^{-1}(T) \sim |t|^{\gamma}$ ($T > T_C$), (3)

just to cite some of them. Different sets of values of the exponents (α, β, γ) correspond to different models (universality classes) which have been theoretically developed after a certain expression of the Hamiltonian describing the physical system. Table 1 contains the most relevant universality classes for magnetic systems, for which the values of the exponents have been found by different methods