HIGH TEMPERATURE THERMAL EXPANSION OF RMn$_2$
INTERMETALLIC COMPOUNDS WITH HEAVY RARE EARTH ELEMENTS

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Abstract. The temperature variation of the thermal expansion coefficient, $\alpha$, of cubic and hexagonal Laves phase compounds RMn$_2$ (R = Gd + Lu and Y) was measured in a wide temperature range 5-900 K by the X-ray diffraction and dilatometric methods. The value of $\alpha$ is substantially enhanced, up to $55 \times 10^{-6}$ K$^{-1}$, in the paramagnetic region. At high temperatures, 600-800 K, $\alpha$ passes over a maximum and decreases with a further increase of temperature. Some change in $\alpha(T)$ depending on the type of the rare earth ion was discovered suggesting the instability of the Mn electronic structure related to the strong temperature dependence of the mean squared amplitude of spin fluctuations.

1. Introduction

Thermal expansion is one of the most important parameters widely used when discussing the features of 3d-magnetism in intermetallic compounds and metal alloys. In the magnetically ordered state the magnetovolume effect (volume magnetostriction) is proportional to the squared 3d moment. Hence, its magnitude, temperature and field dependencies can characterize the magnetic behavior of the 3d-electron system [1, 2]. The magnetic contribution to the thermal expansion is considered arising due to the strong dependence of the interatomic distances on the d-band width, which leads to a large positive magnetovolume effect, $\Delta V/V > 10^{-3}$ [3]. In the ordered state the magnetic part of the thermal expansion coefficient $\alpha_{\text{mag}}$ is proportional to the spontaneous magnetization of the d-electron system. In strongly correlated 3d-electron systems, where fluctuating local atomic moments persist above $T_C$, the importance of the magnetovolume effect in studying the temperature dependence of the spin-fluctuation

(SF) amplitude in the paramagnetic temperature region was emphasized by many authors [4 - 7]. The temperature variation of the mean squared amplitude of SF, \( \langle m_{sf}^2 \rangle \), leads to a temperature dependent contribution to the thermal expansion coefficient in the paramagnetic temperature range [4, 6]. According to the SF theory, at moderate temperatures \( \langle m_{sf}^2 \rangle \) is proportional to \( T \) and tends to saturate at higher temperatures; this region is determined by a characteristic SF temperature \( T_{sf} \). With increasing temperature \( \alpha_{mag} \) is temperature independent and then at higher temperatures asymptotically reduces to zero.

This concept was used to account for the giant thermal expansion observed in the Mn-rich pseudobinary alloys \( Y(Mn_{1-x}Al_x)Z \) and to derive qualitatively the temperature variation of the mean squared SF amplitude in the undiluted compound \( YMn_2 \), which was found to be in agreement with the inelastic neutron scattering experiments [8, 9]. The value of \( \alpha_{mag} \) in \( YMn_2 \) was found comparable to the lattice contribution \( \alpha_{lat} \) in the paramagnetic temperature range. Recently strongly enhanced thermal expansion coefficients, up to \( 55 \times 10^{-6} \text{ K}^{-1} \), were observed in the paramagnetic region in other binary Y-Mn intermetallic compounds (\( YMn_{12} \), \( Y_6Mn_{23} \) and the \( Y_6(Mn_{1-x}Fe_x)_{22} \) pseudobinary system), too [7, 10]. This value is significantly larger than \( \alpha \) observed in other well-known spin fluctuation systems, such as \( YCo_2 \), \( Pd \), \( FeSi \), \( MnSi \), etc., where \( \alpha \) does not exceed \( (18 \pm 20) \times 10^{-6} \text{ K}^{-1} \) [11, 2, 5]. This makes it possible to follow the anomalies of \( \alpha(T) \) caused by the SF contribution.

The anomalously high thermal expansion observed in the binary Y-Mn intermetallics was attributed to the instability of the 3d-electron configuration of Mn ions [7]. The strong temperature dependence of \( \alpha \) can be treated as if the mean ionic radius of Mn is temperature dependent providing a new contribution to the thermal expansion coefficient due to the temperature variation of the Mn ionic radius (this situation could resemble the intermediate valence case observed in some 4f-compounds being its “itinerant” analog). The thermal expansion data show that the SF excitations in the Mn intermetallics have essentially different temperature dependence compared to the “normal” temperature induced ones predicted by the conventional SF theory [7].

The above suggestion is to be checked by experiments in other Mn compounds showing enhanced thermal expansion. The aim of the present work is to study the thermal expansion coefficient in the \( RMn_2 \) intermetallics with heavy rare earths, RE. The \( RMn_2 \) compounds crystallize in two different structures, MgCu_2-type (with RE= Gd - Ho, and Y) or MgZn_2-type (with Ho - Lu), cubic and hexagonal type structures, respectively [12]. The systematic study through this series permits to find out the influence of different RE metals and crystal structure on the thermal expansion in the paramagnetic high temperature range.

2. Experimental Part and Discussion

Polycrystalline samples of \( YMn_2 \) and \( RMn_2 \) with \( R= \) Gd, Tb, Dy, Er, Tm and Lu were prepared by induction melting under argon gas atmosphere and subsequently