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ANOMALOUS THERMAL VARIATION OF THE COBALT ANISOTROPY AND LONGITUDINAL SPIN FLUCTUATIONS IN Y_2Co_7

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Abstract. – A single crystal magnetostatic study of the intermetallic compound Y_2Co_7 is presented, revealing the existence of extremes in the thermal variation of the anisotropy constants of the compound. These anomalies are understood as resulting from large longitudinal spin fluctuations which are gradually frozen by an applied field, and associated with metamagnetic instabilities of the cobalt magnetism occurring within a finite range of temperatures.

Intermetallic compounds of the Y-Co systems in the composition range from YCo_5 up to YCo_2 have the following related features: their crystallographic structures are derived from the YCo_5 one by the same type of ordered atom substitutions. When this substitution occurs for every three, two or one YCo_5 unit cell, the Y_2Co_7 , YCo_3 and YCo_2 phases are generated respectively [1]. Whereas the magnetism of the cobalt in YCo_5 keeps its character of well-established ferromagnetism, with magnetization and Curie temperature of the same order as in pure cobalt [2], an exchange enhanced Pauli paramagnetism is observed in YCo_2 [2], which undergoes a collective electron metamagnetism in high fields [3]. This collapse is to be associated with a corresponding change in the cobalt atoms coordination in yttrium. From YCo_5 to YCo_2 , this change is gradual and the cobalt atoms are shared out in alternating blocks of two kinds. The first block has the same arrangement as in YCo_5 for which the same well-established cobalt magnetism should be expected. In the second block, the cobalt atoms have almost the same surroundings as in YCo_2 and are then subject to magnetic instabilities. In order to find out whether such instabilities do occur, a single crystal magnetostatic study of the Y_2Co_7 compound has been undertaken.

The single crystal had been prepared by a Czochralski method, using high frequency induction technique, in a water cooled copper crucible, under an argon atmosphere. The ingot was checked using X-ray diffraction which revealed it to be single-phased. A crystal of 45 mg with a structure belonging to the space group $R\bar{3}m$ was cut from the ingot by spark erosion. Magnetization measurements were carried out in fields up to 15 T in the temperature range from 4.2 K to 420 K at the Service National des Champs Intenses in Grenoble.

The magnetic isotherms obtained at 4.2 K, 240 K and 400 K in fields applied parallel and perpendicular to the magnetization axis (trigonal axis of the structure) are reported in figure 1. In addition to large magnetocrystalline and magnetization anisotropies, several other peculiarities were observed and are described below.

Fig. 1. – Magnetic isotherms measured on the Y_2Co_7 single crystal at 4.2 K, 240 K and 400 K in fields applied respectively parallel and perpendicular to the trigonal axis of the structure.

When the applied field is parallel to the magnetization axis the superimposed differential susceptibility at 240 K exhibits a maximum around 8 T suggesting a smooth collective electron metamagnetic transition. On extrapolating the isotherms to zero fields we obtained the thermal variation of the spontaneous magnetization reported in figure 2. It may be seen that the spontaneous magnetization has a positive concave curvature at temperature above 320 K indicating the end of the process of metamagnetic collapse. The thermal variation of the magnetization measured at 14 T has also been drawn on the same figure. It exhibits the field stabilizing effect on the cobalt magnetism.

When the applied field is perpendicular to the magnetization axis a quasi-linear variation followed by a bending of the magnetization curves is observed on increasing field. This normally arises from the effects of higher order terms in the expansion of the anisotropy

Fig. 2. – Thermal variations of the spontaneous magnetization M_s and the magnetization M 14 T measured under an applied field of 14 T in Y_2Co_7 .

energy which, according to the cylindrical symmetry of the structure, can be written as:

$$E_{\text{ani}} = K_1(T) \sin^2 \theta + K_2(T) \sin^4 \theta,$$

θ is the angle between the magnetization vector and the magnetization axis and $K_1(T)$ and $K_2(T)$ are anisotropy constants respectively responsible for the afore-mentioned linearity and bending.

A visual comparison of the isotherms reported in figure 1 shows that on increasing temperature $K_1(T)$ will go to a minimum while $K_2(T)$ will have a maximum in the neighbourhood of 240 K (as indicated by the small range of field over which linearity is observed and the large curvature of the magnetic isotherm at this temperature). These anisotropy constants $K_1(T)$ and $K_2(T)$ have been quantitatively determined from the measured isotherms using the Sucksmith-Thompson method, taking account of the large observed magnetization anisotropy [4]. Their thermal variations, reported in figure 3, are strongly correlated, that is where $K_1(T)$ shows a decrease, $K_2(T)$ increases and conversely.

Fig. 3. - Thermal variations of the anisotropy constants in Y_2Co_7 .

Thus, while being in a stable magnetic state at low temperature cobalt magnetism in Y_2Co_7 undergoes magnetic instabilities on increasing temperature, leading to a lower magnetization state. This has been confirmed by a polarized neutron diffraction study [5] which also showed that the instabilities concern essentially those cobalt atoms which have almost the same surroundings as in YCo_2 whilst those in the YCo_5 environment remain magnetically stable. When the in-

stabilities occur, the high magnetization state is reinduced by applying a field which gives rise to a maximum in the superimposed differential susceptibility measured along the magnetization axis: a second-order metamagnetic transition takes place through a gradual field freezing of large longitudinal magnetic fluctuations. At higher temperatures the fluctuations are reduced, and the metamagnetic transition ends. The positive curvature in the thermal variation of the spontaneous magnetization observed above 320 K indicates that the exchange field acting on the cobalt of the YCo_2 blocks becomes weak.

Occurrence of large longitudinal spin fluctuations associated to magnetic instabilities in a finite range of temperatures and frozen under applied provides an understanding of the observed thermal variation of the anisotropy constants. These variation, instead of a monotonous decrease expected for a ferromagnet, exhibit extremes which are by no means ascribable to the effect of competing sublattice anisotropy effects. In fact when only thermal transverse magnetic fluctuations are taken into account (the modulus of the magnetization vector being constant [6]) monotonous decreases are predicted. On allowing the modulus to vary as it must in an itinerant system, the extra degree of freedom i.e. longitudinal fluctuations associated to the instabilities will further decrease the anisotropy energy. This explains the minimum of $K_1(T)$ at a temperature where the amplitude of the longitudinal fluctuations are the largest. On the other hand, the longitudinal fluctuations are frozen by the application of a field which enables the anisotropy energy to reach again the value that it should have in the absence of any magnetic instabilities. This implies a maximum for $K_2(T)$. Consequently the observed anomalies provide characteristic signatures for the instability of the cobalt magnetism in Y_2Co_7 .

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