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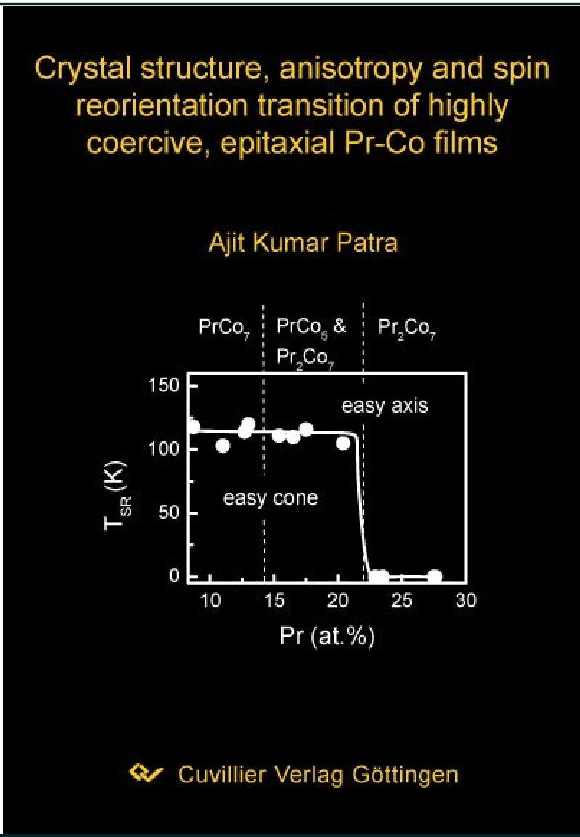
Crystal structure, anisotropy and spin reorientation transition of highly coercive, epitaxial Pr-Co films



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Chapter 1

Fundamentals

Magnetocrystalline anisotropy (MCA) of a RE-TM compound is the basic ingredient for the high coercivity of a modern hard magnetic material and its origin is thus both of fundamental and technological interest. Only a full understanding and analysis of MCA allows to tailor the properties of magnetic materials. This chapter discusses the phenomenology of anisotropy, spin reorientation, and its physical origin and analysis. Moreover, it includes different approaches to determine the anisotropy constants from the measurement and a brief literature survey on the intrinsic and extrinsic properties of different Pr-Co phases, prepared as a bulk or thin film magnet.

1.1 Phenomenological description of anisotropy and hysteresis

In magnetic materials, directional dependence of magnetic properties is known as magnetic anisotropy, which is an essential property of permanent magnets. Different types of magnetic anisotropy are:

- Magnetocrystalline anisotropy (depends on crystal structure)
- Shape anisotropy (depends on shape of the grains and the whole sample)
- Magnetoelastic anisotropy (depends on applied or residual stresses)
- Induced anisotropy (depends on process treatment, e.g. field annealing)

Magnetic anisotropy strongly affects the shape of the hysteresis loops and controls the coercivity and remanence. In the case of RE-TM, MCA is the dominant form of anisotropy and it will be discussed in more detail in this work. Presence of MCA leads to easy and hard directions of magnetization. In easy directions it is easier to magnetize the material compared to the hard directions. As an example, Fig. 1.1 shows the hysteresis loops of a strongly anisotropic magnet with field applied along the easy and hard directions.

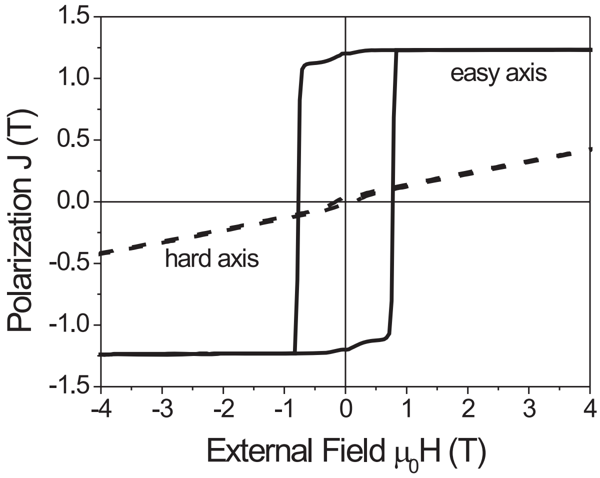


Figure 1.1: Magnetic hysteresis loop of a magnet measured with field applied along the easy and hard axis.

MCA is the energy required to deflect the magnetic moment from the easy to the hard directions. For a hexagonal crystal of $CaCu_5$ type the anisotropy energy density can be written in a general form as:

$$E_a = K_0 + K_1 \sin^2\theta + K_2 \sin^4\theta + (K_3 + K_3' \cos 6\phi)\sin^6\theta \quad (1.1)$$

where K_0, K_1, K_2, K_3 and K_3' are anisotropy constants, θ is the angle between the magnetization vector and the c-axis, and ϕ is the angle between the magnetization component in the basal plane and the a-axis (Fig. 1.2). K_0 is independent of angle and is usually ignored. Higher order terms like K_3 and K_3' are very small compared to K_1 and K_2 and are often not considered. In most cases, it is sufficient to consider K_1 and K_2, and their magnitude and sign determine the preferred direction of the magnetization vector for given conditions, by bringing the anisotropy energy to a

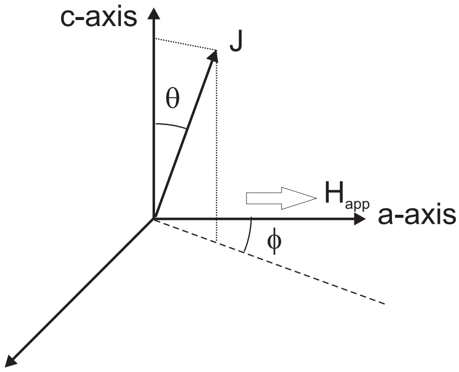


Figure 1.2: Direction of the polarization vector relative to the c-axis and the a-axis defining the angles θ and ϕ.

Table 1.1: Different categories of easy magnetization direction.

Conditions	E_{min} for	Category
$K_1 > 0$, $K_1{+}K_2 > 0$	$\theta = 0°$	easy axis
$K_1 < 0$, $K_1{+}K_2 < 0$	$\theta = 90°$	easy plane
$K_1 < 0$, $2K_2 > -K_1$	$\theta = \sin^{-1}\sqrt{\frac{-K_1}{2K_2}}$	easy cone

minimum. Different cases are shown in Table 1.1 and the corresponding energy profile as a function of θ is depicted in Fig. 1.3.

In case of $K_1 > 0$, and $K_1{+}K_2 > 0$, the anisotropy energy is minimum for $\theta = 0$ and 180°, thus the magnetization vector lies preferably along (two possible directions of) the c-axis. This case is called an easy axis configuration. On the other hand, for $K_1 < 0$, and $K_1{+}K_2 < 0$, the minimum occurs for $\theta = 90°$, which corresponds to the magnetization vector in the basal plane. As K_3' is ignored in this schematic representation, no additional orientation within the basal plane is preferred, so that the whole plane has equally low energy. Thus, this case is called easy plane anisotropy. In the case of $K_1 < 0$, and $2K_2 > -K_1$, the minimum in the anisotropy energy is observed for $0° < \theta < 90°$; the magnetization vector lies at a certain angle ($< 90°$) with respect to the c-axis. This situation corresponds to an easy cone behavior (Table 1.1).

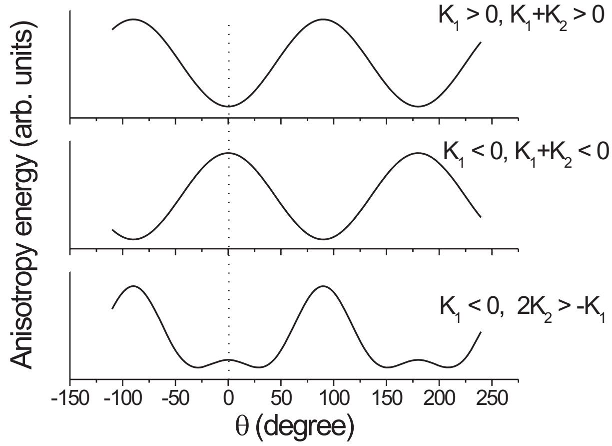
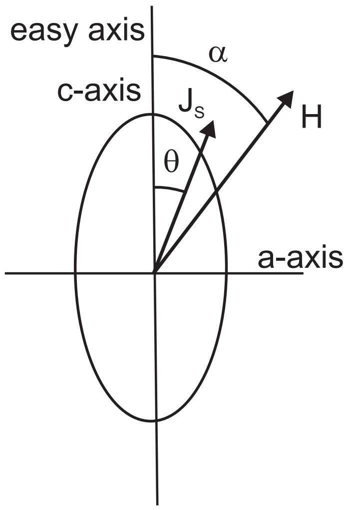


Figure 1.3: Variation of the anisotropy energy as a function of θ.

In an applied magnetic field, the total energy density for a hexagonal crystal can be written as the sum of the anisotropy term[1](see eqn. 1.1) and Zeeman term:

$$E_{tot} = K_1 \sin^2 \theta + K_2 \sin^4 \theta - H J_S \cos(\alpha - \theta) \tag{1.2}$$

Here, H is the magnitude of the external field, J_S is the saturation polarization



at a given temperature and α is the angle between $\overrightarrow{H}$ and the c-axis. As shown in Fig. 1.3, the minimum in the anisotropy energy is observed at $\theta = 0$ and $180°$ for

[1]Here, higher order terms are neglected.

$K_1 > 0$, $K_1+K_2 > 0$ (T = 300 K). Moreover, on application of the external field parallel to the c-axis ($\alpha = 0$), the energy minimum always lies at 0° and thus the polarization ($J = J_S\cos\theta$) always reaches the saturation (see Fig. 1.4 (●)).

When the field H is applied along the hard axis (⊥ to the c-axis), eqn. 1.2 can be written as:

$$E_{tot} = K_1 \sin^2\theta + K_2 \sin^4\theta - HJ_S \sin\theta \tag{1.3}$$

Magnetization along a hard axis proceeds via the rotation of the magnetization vector from the easy axis to the hard axis. Thus, the polarization along the hard axis ($J^{\perp} = J_S\sin\theta$) increases linearly with the field, and saturation in the polarization is achieved only at a field value of the anisotropy field (see Fig. 1.4 (×)). The preferred direction of the magnetization vector can be obtained by minimizing eqn. 1.3.

$$\frac{dE_{tot}}{d\theta} = 2K_1 \sin\theta\cos\theta + 4K_2 \sin^3\theta\cos\theta - HJ_S\cos\theta = 0, \tag{1.4}$$

which leads to:

$$H = \frac{2K_1\sin\theta + 4K_2\sin^3\theta}{J_S} \tag{1.5}$$

The anisotropy field H_A is the field value required to align the magnetization perpendicular to the easy axis, i.e., $\theta = 90°$. From eqn. 1.5 thus follows:

$$H_A = \frac{2K_1 + 4K_2}{J_S} \tag{1.6}$$

Experimentally measured values of the anisotropy field, H_A, are obtained as the

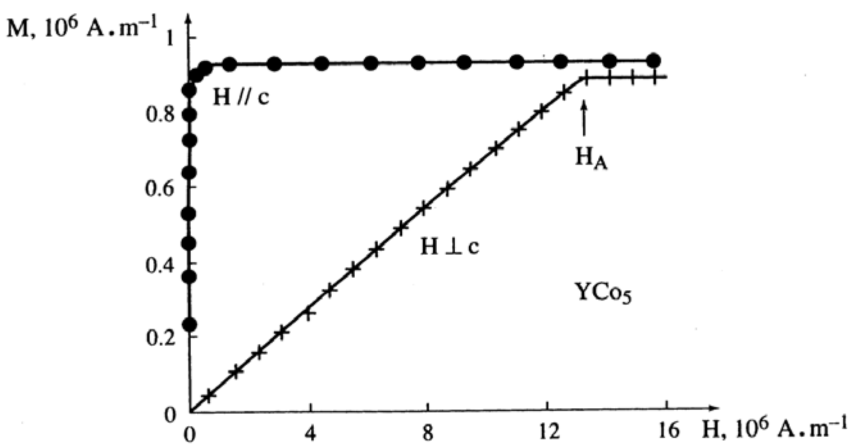


Figure 1.4: Magnetization measurements of YCo_5 [Ala81, dTdL05].

intersection of the easy axis and hard axis measurement curve with the field parallel and perpendicular to the easy axis, respectively[2](see Fig. 1.4).

When K_2 is negligible, measurement of H_A allows to determine K_1. However in many materials, K_2 plays a significant role (definitely at low temperatures for most cases). Then the measurement of H_A is not sufficient to determine K_1 and K_2 individually. The most common approaches for determination of anisotropy constants are:

- Sucksmith and Thompson approach (ST approach)
- Angular dependent magnetization measurements

Sucksmith and Thompson approach:

The Sucksmith and Thompson approach [Suc54] considers the magnetization curve of a uniaxial magnet measured in a direction perpendicular to the easy axis and is based on the relation:

$$\frac{H}{J} = \frac{2K_1}{J_S^2} + \frac{4K_2}{J_S^4} J^2 \tag{1.7}$$

which can be derived from eqn. 1.5 by substituting $\sin\theta = J/J_S$, where $J(H)$ is the polarization along the hard axis. When H/J is plotted versus J^2, the anisotropy constant K_1 and K_2 are derived from the vertical intercept and slope of the straight line, respectively. Equation 1.7 considers the perfect alignment of the grains, so substantial error arises due to misalignment of the grains (such as in the case of a powdered and textured sample, and even in the case of epitaxial samples with low texture quality) in determination of K_1 and K_2. For such a case, the modified ST approach proposed by Ram and Gaunt [Ram83] is used, which takes into account the misalignment of the grains (texture spreading). In the modified ST approach, $H/\gamma(J - J_R)$ is plotted versus $\gamma^2(J - J_R)^2$, where J_R is the remanence polarization in the hard direction and $\gamma = (J_S - J_R)/J_S$ is the alignment factor. Shown in Fig. 1.5 is a modified ST plot for a $Nd_2Fe_{14}B$ thin film.

The ST approach is quite useful in determination of anisotropy constants. However, this approach is restricted to K_1 and K_2. In some materials, K_3 plays a dominant role and the complete magnetization curve can not be understood without

[2]In case of lack of experimental facility for high field measurements, extrapolated intersection of the easy and hard axis curves yield an estimated value of H_A.