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Canted-to-canted singular points in ferrimagnetic polycrystalline magnetization curves

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ABSTRACT

The study of singularities in the magnetization curve of magnetic materials is since a long time a well known tool to gain insight into the material properties. Such singularities are usually due to discontinuous magnetization processes related to magnetic anisotropy, therefore an ideal investigation scheme would comprise magnetization measurements along the symmetry axis of a single crystal. As a matter of fact, one has to often deal with polycrystalline samples: in that case, however, the singularities can still be detected at the same positions as for the single crystal case. In this paper we focus on ferrimagnetic materials with uniaxial magnetic anisotropy. After recalling how theory allows us to calculate the critical fields at which singularities occur when the magnetic field is applied along a symmetry direction, we show that a new type of singular point not yet reported in the literature can exist. Such new singularity appears *only* in polycrystalline samples. We derive its analytical expression and we also show that, in spite of the need to overcome the exchange interaction, combinations of the material parameters can occur for which it falls inside experimentally accessible region.

1. Introduction

The magnetization curve of a magnetic material can show, in particular cases, points of discontinuity that are called *singular points*, or *singularities*, at determinate values of the applied magnetic field $\boldsymbol{H}$ that depend on the material and on the direction of $\boldsymbol{H}$ with respect to measured sample orientation. Such singularities can be either of first order, in the case of abrupt magnetization discontinuities, or of higher order, when the discontinuity becomes evident in the magnetization derivatives with respect to H, and are a very powerful tool to gain insight into the material properties. Well known examples of first order singularities are the First Order Magnetization Processes (FOMP) [1], arising from the occurrence of irreversible rotations of the magnetization vector between equivalent energy minima, that can be considered as *field induced* spin reorientation transitions (SRT). In those ferrimagnets where magnetic anisotropy and exchange are comparable, such as RE-intermetallic compounds, FOMPs are originated by their mutual competition. In other materials, such as transition metal oxides, the exchange interaction is so strong that they can be considered as rigidly collinear ferrimagnets and can be treated as *effective ferromagnets*: in this case the energy profile shape is due to magnetic anisotropy only and it turns out that high order terms of magnetic anisotropy are required in the energy expression to induce a FOMP. In addition to FOMPs, higher order singularities can be observed as well at particular orientations of the magnetic field with respect to the crystal axes [2].

The observation of singular points is a powerful tool to gain insight into material properties [3–9], widely employed since several decades and of topical interest thanks to the more and more increasing value of the maximum available magnetic field that has recently reached the value of 90 T [10]. Unfortunately, while single crystals are often hardly available in ordinary laboratory practice, singular points in polycrystalline samples are usually hidden by the average over the different grain orientations. Nonetheless one can still extract physical parameters from $M(H)$ by means of two special methods: the Singular Point Detection (SPD) [11] and the Free Powder Technique (FPT) [12–15]. In FPT the material is grinded into very fine grains that are completely free to rotate, so that they

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orient themselves with their magnetization vector along the applied magnetic field $\boldsymbol{H}$: this technique has proven particularly useful in the exchange coupling determination for 3*d* RE-TM intermetallics [14–18]. On the other hand, in SPD one measures $M(H)$ for a clamped polycrystalline sample and analyses the derivatives with respect to H: in this way, starting from a certain derivation order which depends on the crystal symmetry, one recovers the singularities otherwise hidden in $M(H)$. This a very effective and often the only method to measure singular points when single crystals are not available.

Usually, when dealing with single crystals, one detects singularities measuring along the crystal symmetry axes, whilst when dealing with polycrystals, one can however find again the same singularities by means of the SPD technique. In this paper we describe a new type of singular point, not yet reported in the literature, that adds to the other already known $M(H)$ singularities and has the characteristic to appear *only* in polycrystalline samples.

2. Theory

The magnetization curve $M(H)$ of a crystalline magnetic material is given, at a microscopic level, by essentially two factors acting on each single magnetic ion: the exchange interaction with neighbouring ions and the magnetocrystalline field. However, for most practical uses it is more convenient to describe the system in terms of macroscopic parameters associated with one, two, or more magnetic sublattices, each one formed by rigidly collinear spins below its own critical temperature. This can be done in the frame of an extended Stoner–Wohlfarth model [19] by restricting the analysis at temperature $T=0$ K and minimizing the system free energy written as the sum of Zeeman, magnetocrystalline and exchange energies:

$$E = E_Z + E_K + E_J. \tag{1}$$

When only two sublattices with uniaxial magnetic anisotropy are involved, Eq. (1) can be written as

$$\begin{aligned} E_Z(\theta_A, \theta_B) &= -(\boldsymbol{M_A} + \boldsymbol{M_B})\cdot\boldsymbol{H}, \\ E_K(\theta_A, \theta_B) &= \sum_i K_A^i \sin^{2i}(\theta_A - \alpha) + K_B^i \sin^{2i}(\theta_B - \alpha), \\ E_J(\theta_A, \theta_B) &= -J(\boldsymbol{M_A}\cdot\boldsymbol{M_B}), \end{aligned} \tag{2}$$

where $\boldsymbol{M_A}$ and $\boldsymbol{M_B}$ are the sublattice magnetization vectors, θ_A, θ_B and α are the angles between the applied magnetic field $\boldsymbol{H}$ and $\boldsymbol{M_A}$, $\boldsymbol{M_B}$ and the magnetic anisotropy axis respectively, as shown in Fig. 1, K_A^i and K_B^i are the sublattices anisotropy constants,

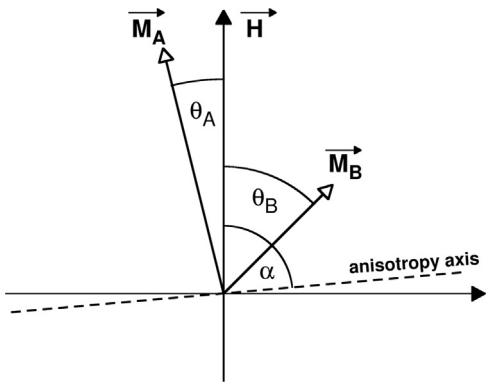

Fig. 1. Vector and angle definitions. The anisotropy axis can be either an easy or a hard magnetization axis.

and J is the exchange constant (negative for ferrimagnetic or antiferromagnetic interaction). We can assume $M_A > M_B$ without loss of generality, where M_A and M_B are the moduli of $\boldsymbol{M_A}$ and $\boldsymbol{M_B}$ respectively. The anisotropy axis can be either an easy or a hard magnetization axis, depending on the algebraic signs of the anisotropy constants, however the results for the case of hard axis (viz. easy magnetization plane) can be obtained from the easy axis results by means of the $K \Leftrightarrow R$ transformation [1]. The case of $K_A^i = 0$, pertinent to 3*d* RE-TM intermetallics where RE has negligible anisotropy compared to TM, has been exhaustively carried out by Kuz'min [22] considering K_B^i up to 4th order ($i \le 2$). On the contrary, here we take into account the anisotropy of both sublattices, and arrest the anisotropy contribution E_K to the second order terms. In this case we can drop the i superscripts and the $K \Leftrightarrow R$ transformation simply prescribes that substituting (K_A, K_B) with $(-K_A, -K_B)$ into the results of the easy axis case for a given α value, one obtains the results of the easy plane case for angle $\pi/2 - \alpha$ between $\boldsymbol{H}$ and the hard magnetization axis. We also assume that both sublattices have the same anisotropy axis, while we do not make any assumption regarding the algebraic signs of anisotropy constants, which can independently be either positive or negative.

The relationship between the macroscopic parameters appearing in Eq. (2) and microscopic physical quantities can be derived by the methods of mean field theory in a rather straightforward way, as shown in various textbooks and literature [14,20,21]. The issue of how to obtain the values of anisotropy and exchange constants from a magnetization curve is therefore of primary importance in magnetic materials science, for both theoretical and practical reasons. One way to gain informations about K_A, K_B and J is through the observation of singularities in the $M(H)$ curve that can occur at particular values of the applied magnetic field H which depend on K_A, K_B, J and α.

Such singularities can be well understood in the frame of the model described by Eq. (2). The possible system states can be classified into canted and collinear types, according to the mutual orientation of the $\boldsymbol{M_A}$ and $\boldsymbol{M_B}$ vectors. Among the collinear states, fall the two states having $\boldsymbol{M_A}$ and $\boldsymbol{M_B}$ along the same direction of $\boldsymbol{H}$, namely the *forced ferromagnetic* or Parallel Saturation (PS) and the *ferrimagnetic* or Antiparallel Saturation (AS), with parallel and antiparallel $\boldsymbol{M_A}$ and $\boldsymbol{M_B}$ respectively.

In general, when $\boldsymbol{H}$ is along a high-symmetry crystallographic direction, starting from $H=0$ one can have three singular points, either of first or higher order, corresponding to transitions from canted to AS, from AS to canted and from canted to PS states, at three critical fields H_{t1}, H_{t2} and H_{t3} respectively. Sometimes, depending on the material parameters and on the crystal orientation α, H_{t1} and/or H_{t2} may be missing: an example is displayed in Fig. 7a and d, where it can be noticed that H_{t1} does not appear when H is along the easy magnetization direction. A compact expression for such critical fields is obtained, when the singularity is of order higher than first, in Ref. [23] considering small deviations from the collinear state:

$$(2K_A + M_A H_t)^{-1} + (2K_B + M_B H_t)^{-1} + (JM_A M_B)^{-1} = 0 \tag{3}$$

For $\boldsymbol{H}$ along the anisotropy axis, H_{t3} is given by Eq. (3) as is, while H_{t1} and H_{t2} can be obtained after substituting M_B with $-M_B$. For $\boldsymbol{H}$ perpendicular to the anisotropy axis, H_{t1}, H_{t2} and H_{t3} are calculated in the same way after substituting (K_A, K_B) with $(-K_A, -K_B)$, in agreement with the $K \Leftrightarrow R$ transformation. Of special interest is the critical field H_{t3} corresponding to transition to the PS state perpendicular to the anisotropy axis: in that case H_{t3} can be identified as an effective anisotropy field H_S and is given by [24]