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DETERMINATION OF THE DEGREE OF CRYSTALLITES ORIENTATION IN PERMANENT MAGNETS BY X-RAY SCATTERING AND MAGNETIC MEASUREMENTS

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Résumé - Les aimants permanents sont essentiellement des composés ferromagnétiques et uniaxiaux. Certains aimants sont élaborés en frittant des poudres préalablement orientées et comprimées sous champ. La valeur de l'induction rémanente B_r dépend directement du degré d'alignement des cristallites et nous présentons dans cet article deux méthodes quantitatives différentes pour le déterminer.L'analyse aux rayons X permet de comparer les intensités des réflexions de Bragg pour les différentes valeurs de l'angle entre une direction [hkl] et l'axe cristallographique principal. L'analyse magnétique permet de comparer les courbes d'aimantation expérimentales et celles calculées, pour différentes distributions statistiques des orientations des grains. Les deux méthodes ont été appliquées à des aimants Nd-Fe-B et $SmCo_5$. Elles révèlent que la désorientation des particules dans ces systèmes conduit à une réduction d'environ 10 % de l'induction rémanente.

Abstract - Permanent magnet compounds are basically ferromagnets with uniaxial anisotropy. In different cases, the magnets are elaborated by sintering of powders previously oriented and compressed under field. The value of the remanent induction B_r is directly dependent on the degree of crystallites alignment, and we present in this paper two different quantitative methods to determine it. X-ray analysis involves the comparison of the intensities of Bragg reflections, for different values of the angle of a direction [hkl] with respect to the unique crystallographic axis of the compound. Magnetic analysis involves the comparison between experimental magnetization curves and calculated ones for different statistical distributions of crystallites orientations. Both methods were applied to NdFeB and $SmCo_5$ magnets. They reveal that the misorientation of particles in these systems leads to a reduction of the remanent induction by about 10 %.

I - INTRODUCTION

Permanent magnet compounds are basically ferromagnets with uniaxial anisotropy. The coercivity of the magnets, closely related to this anisotropy, develops in materials which are inhomogeneous at a microscopic scale. Such a property is in particular obtained in ferrites and rare earth-transition metal magnets, by sintering of powders previously oriented and compressed under field. The value of the remanent induction B_r is directly dependent on the degree of crystallites alignment. A quick method to determine an average value of their degree of alignment consists in measuring the remanent induction B_r of the magnet with respect to the spontaneous induction of the compound /1/. We present in this paper a more quantitative analysis of the statistical distribution of grains orientation, which was obtained by X-ray analysis and magnetostatic measurements, on NdFeB and $SmCo_5$ magnets.

II - X-RAY DETERMINATION OF THE DEGREE OF GRAIN ALIGNMENT

For a reflection hkl, Bragg scattering occurs if the direction [hkl] is along the scattering vector $\vec{K}$, defined as the bissectrice of the angle between the direction of the incident beam and that of the scattered beam. In a polycrystalline sample, in

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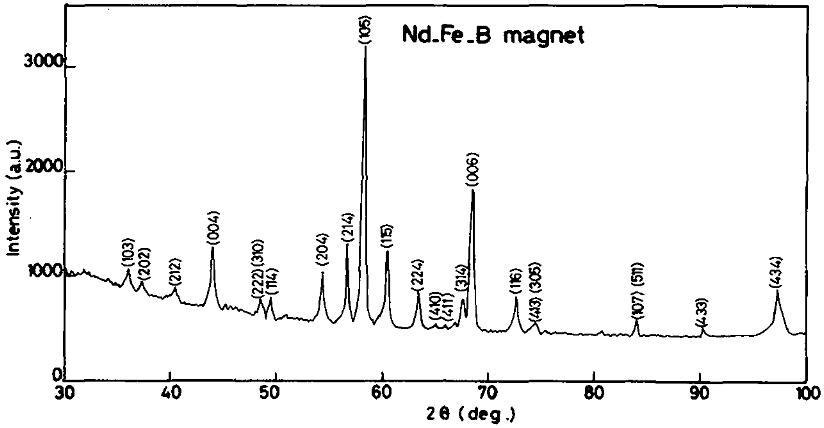

Fig. 1 - X-ray diffractogram of a Nd-Fe-B magnet obtained with the Co radiation (λ_{Co} = 1.7903 Å).

which crystallites orientations are perfectly at random, the probability for any direction [hkl] to be along $\vec{K}$ is constant. In the case of a single crystal, it is necessary to adjust the crystal orientation in order to align [hkl] along $\vec{K}$. Permanent magnets elaborated by sintering of powders, can be considered as an assembly of single crystals, for which the crystallographic $\vec{c}$-axes, i.e. the directions [001] of the reciprocal space, are distributed within a certain solid angle. If the axis of the solid angle, z, is aligned along $\vec{K}$, the probability $p(\omega_{//})$ for crystallographic $\vec{c}$-axes to be along a given direction at an angle $\omega_{//}$ with respect to z is simply equal to the probability for a direction [hkl] at an angle $\omega_{hkl} = \omega_{//}$, from [001] to be along $\vec{K}$. Thus the comparison of the intensity of a reflection [hkl], I^{magnet}_{hkl}, to that observed in a polycrystalline sample, I^{poly}_{hkl}, is a measurement of the proportion of crystallites along any direction at $\omega_{//}$ from z.

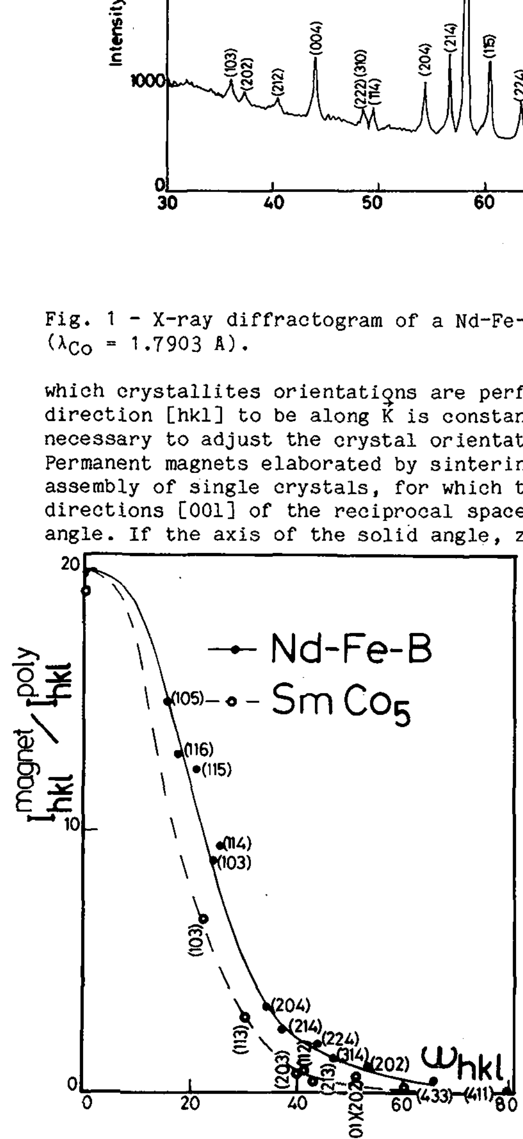

Fig. 2 - $I^{magnet}_{hkl}/I^{poly}_{hkl}$ as function of the angle ω_{hkl} between directions [hkl] and [001].

We used this method to determine the degree of grain alignment in Nd-Fe-B and $SmCo_5$* magnets. X-ray diffractograms were recorded on slices of Nd-Fe-B permanent magnets, cut at different thicknesses perpendicularly to z. The results obtained show that the degree of grain alignment is a constant over the volume of the magnet, except on a very thin skin of about 0.1 mm at the surface. Typical X-ray pattern is presented in figure 1. Comparison to the intensities measured on a non-oriented powder /2,3/ leads to the determination of $I^{magnet}_{hkl}/I^{poly}_{hkl}$ which is plotted as a function of ω_{hkl} in figure 2. As shown in figure 3, the decrease of $I^{magnet}_{hkl}/I^{poly}_{hkl}$

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*the sample of $SmCo_5$ was kindly provided to us by "Aimants UGIMAG S.A."

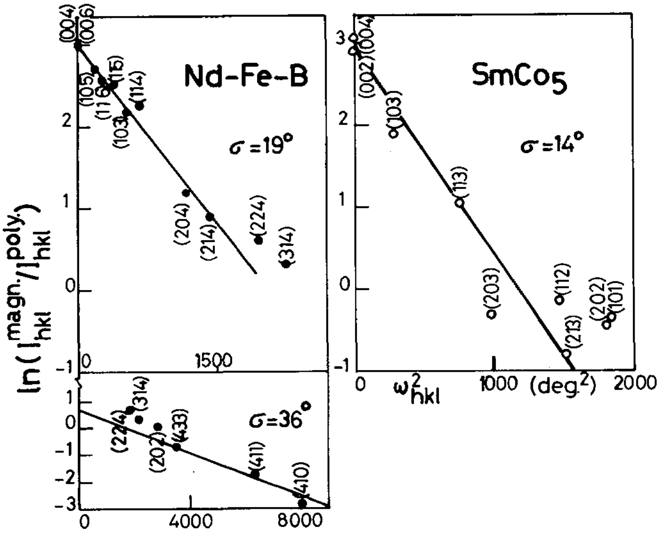

Fig. 3 - ω^2_{hkl} dependence of the logarithm of normalized Bragg reflections in Nd-Fe-B and $SmCo_5$ magnets.

with ω is approximately a gaussian $\exp(-\omega^2_{//}/2\sigma^2)$ with $\sigma = 18.7°$. This would mean that 99 % of crystallites make an angle smaller than 55° with respect to the $\vec{z}$-axis of the magnet. However, the observation of weak reflections with $\omega_{//} > 45°$ reveals an additional small contribution of another gaussian with $\sigma' = 36°$, corresponding to a much broader distribution of crystallites orientation (Figure 3). The proportion of such crystallites is approximately 15 % of the total number of crystallites. This property can be understood by considering that a proportion of crystallites obtained after grinding of the bulk ingots are not single crystals. Such crystallites are not efficiently oriented in the field applied during the compression.

The same analysis was performed for a $SmCo_5$ magnet. The results are presented in figures 2 and 3. Very similar results are obtained. The main gaussian describing the statistical distribution of grains orientation is determined by $\sigma = 14°$. The accuracy of the results is not sufficient to reveal a possible additional contribution from another gaussian describing a broader distribution of grains orientation. Such a contribution is in any case much weaker than in NdFeB, and suggests therefore that, in $SmCo_5$ magnets, almost all grains can be considered as single crystals.

III - MAGNETIC DETERMINATION OF THE DEGREE OF GRAINS ALIGNMENT

The field dependence of the magnetization of an uniaxial single-crystalline compound has been calculated by Néel et al /4/ for any angle between the applied field and the easy $\vec{c}$-axis. The first magnetization curve can be divided into two parts associated with different physical processes. In low fields, the crystal is decomposed into several domains, and the magnetization measured along the field H is

$$m(\phi, H) = H \times M_s[\cos^2\phi/H_D + \sin^2\phi/(H_D + H_A)] \qquad (1)$$

where M_s is the spontaneous magnetization, H_D the demagnetizing field for saturated magnetization and H_A the anisotropy field. The magnetization follows this law until a threshold field, $H_s(\phi)$, for which the crystal becomes single domain. Above H_s, the angle Θ between the magnetization and the easy axis is given by :

$$(H_A/2H)\sin 2\Theta + \sin(\Theta - \phi) = 0 \quad (2)$$

and the magnetization measured along the field is :

$$m(\phi, H) = M_s \cos(\phi - \Theta) \qquad (3)$$

The calculated recoil magnetization curve, assuming that the coercivity impedes the nucleation of reverse domains, corresponds to the second process alone.

As described in the previous section, the permanent magnets considered here are formed of independent crystallites whose easy axes are distributed about a preferred direction z. When the field is applied along z, crystallites which make the same angle $\omega_{//}$ with respect to z, i.e. with respect to the field, exhibit identical magnetization law. The number of crystallites between $\omega_{//}$ and $\omega_{//} + d\omega_{//}$ is obtained by integrating over α around the $\vec{z}$-axis :

$$N_{//}(\omega_{//})d\omega_{//} = \int_{\alpha=0}^{2\pi} \sin\omega_{//}\; p(\omega)_{//}\; d\omega_{//}\; d\alpha = 2\,\pi \sin\omega_{//}\; p(\omega_{//})\; d\omega_{//} \qquad (4)$$

where $p(\omega_{//})$ is the statistical distribution of crystallites. When the field is applied perpendicular to z, the crystallites behaving identically make the same angle $\omega_{\perp}$ with respect to the field. The number of crystallites between $\omega_{\perp}$ and $\omega_{\perp} + d\omega_{\perp}$ is :

$$N_{\perp}(\omega_{\perp})\; d\omega_{\perp} = \int_{\alpha=0}^{2\pi} \sin\omega_{\perp}\; p(\omega_{//})\; d\omega_{\perp}\; d\alpha \qquad (5)$$

with $\omega_{//}$ depending on α through :

$$\cos\omega_{\perp} = \sin\omega_{//} \times \cos\alpha \qquad (6)$$

In Both cases, the calculated magnetization of the magnet is therefore :

$$M(H) = \frac{\int_0^{\pi/2} N_{(\perp)}\omega_{(\perp)} \times m(\omega_{(\perp)},H)\; d\omega_{(\perp)}}{\int_0^{\pi/2} N_{(\perp)}\omega_{(\perp)} d\omega_{(\perp)}} \qquad (7)$$

where $m(\omega, H)$ is deduced from relations (1) to (3) with $\phi = \omega_{//}$ or $\omega_{\perp}$ depending whether the field is along or perpendicular to H.

Subsequently, this method was used to deduce the degree of grains orientation from magnetic measurements. The field dependence of the magnetization for a Nd-Fe-B magnet, under fields applied respectively along z and perpendicular to z, are presented in figure 4. Satisfactory agreement between calculated first magnetization curves and experimental results can be obtained if considering a unique gaussian distribution for $p(\omega_{//})$. However a better fit is obtained with the superposition of two gaussians with $\sigma = 17°$ and $\sigma' = 28°$, and respective weight 90 % and 10 % (Figure 4). The differences between experimental and calculated magnetization curves in intermediate fields can be attributed to the inhomogeneities of the demagnetizing field in the cubic specimen studied, as verified by measuring an Fe sample of identical shape.

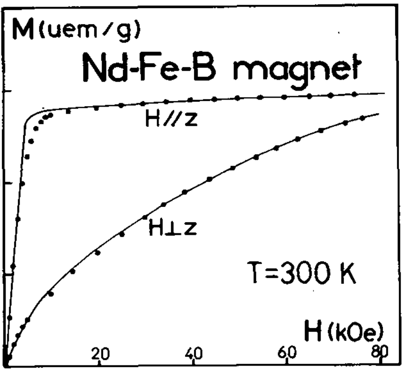

Fig. 4 - Experimental (•••) and calculated (——) magnetization curves of a NdFeB magnet. Calculation parameters : $M_s = 150$ e.m.u./g, $H_A = 73$ kOe.

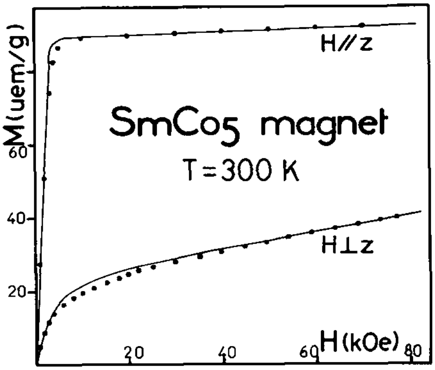

Fig. 5 - Experimental (•••) and calculated (——) magnetization curves of a $SmCo_5$ magnet. Calculation parameters : $M_s = 98$ e.m.u./g, $H_A = 350$ kOe.

A same analysis for $SmCo_5$ magnet is presented in figure 5. The calculated curves were obtained for a main (95 %) gaussian with $\sigma = 15°$, and a weak additional contribution (5 %) with $\sigma = 30°$.

IV - CONCLUSION

Crystallites orientation in permanent magnets was determined by two different methods involving X-ray and magnetic analysis respectively. Consistent results were obtained. The reduction of the remanent induction B_r, due to misalignment of the grains, reaches about 10 % in both Nd-Fe-B and $SmCo_5$ magnets. Comparing both methods, X-ray analysis is more quantitative, but not as easy to realize as magnetic analysis. It is worth recalling that the usual way to determine grains orientation involves a direct measurement of B_r. The result may be affected by a possible decrease of the induction resulting from domain nucleation in grains of small coercivity. The measurement of the first magnetization curve considered in this study, is obviously not affected by such a phenomenon. Finally, a straight generalization of this method would permit a more quantitative analysis of magnetic properties in uniaxial systems where single crystals are not available /3,5/.

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