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STATISTICAL ANALYSIS OF THE ORIENTATION OF SINTERED $SmCo_5$ MAGNETS*

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ABSTRACT

A statistical model is developed to describe the orientation of particle easy axes with respect to the alignment direction in sintered $SmCo_5$ magnets. The model can be used to describe the three-dimensional orientation distributions measured by Swift et al.[1] using x-ray techniques, and with appropriate modification, the two-dimensional orientation distributions measured metallographically by Martin.[2]

The distribution is described by a single parameter β, analogous to the standard deviation of a Gaussian distribution. For a given β, the model predicts the ratio of remanence to saturation and also the shape of the magnetization curve measured in decreasing fields when the field is applied perpendicular to the alignment axis. The effective anisotropy and the value of β can be determined by comparison of experimental and calculated curves.

INTRODUCTION

In a previous paper,[3] we have reported a method based on a comparison of measured hard-axis magnetization curves with computed curves, to determine the anisotropy and the degree of particle misalignment in sintered $SmCo_5$ permanent magnets. This paper will develop this method from an assumed distribution of particle orientations to the calculation of hard axis magnetization curves in decreasing fields, and extend the method to predict other magnetic quantities.

PARTICLE DISTRIBUTION

The orientation of a single particle is described in Fig. 1. The distribution of particle easy axes is assumed to be normal in terms of $\emptyset$ and uniformly distributed in θ. In spherical coordinates, the distribution is of the form

$$f(\emptyset,\theta)dA=k\exp(-\emptyset^2/\beta^2)\sin\emptyset d\emptyset d\theta \qquad (1)$$

where $dA=\sin\emptyset d\emptyset d\theta$ is an element of spherical area. This describes a distribution with a maximum at $\emptyset=0$, decreasing exponentially with $\emptyset$ as $\emptyset$ departs from 0 in any direction from the axis of orientation. The quantity β measures the degree of particle misorientation; small β means good alignment. The function $f(\emptyset,\theta)$ is defined for the region $0\le\emptyset\le\pi/2$ and $0\le\theta\le 2\pi$, and k is the normalization constant which is determined by the condition

$$\int_A f(\emptyset,\theta)dA=1. \qquad (2)$$

If $\beta<\pi/2$, which is true for the cases of interest, the upper limit of $\pi/2$ on $\emptyset$ can be replaced by infinity. Then the integral in equation (2) or any integral of the form

$$I(m)=\int_0^\infty\int_0^{2\pi} f(\emptyset,\theta)\,\emptyset^m\,\sin\emptyset d\emptyset d\theta,\quad m=\text{integer} \qquad (3)$$

can be evaluated by expanding $\sin\emptyset$ as a Taylor series. The solution can then be written as

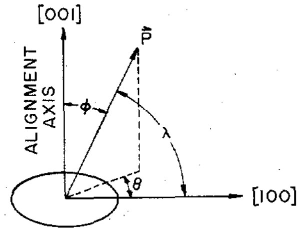

Fig. 1. Definition of angles used to describe particle orientations.

$$I(m)=\pi k\sum_{n=1}^{\infty}\frac{(-1)^{n-1}\beta^{2n+m}\Gamma(n+m/2)}{(2n-1)!} \qquad (4)$$

where Γ is the gamma function.

The normalization condition expressed in (2) can now be written as,

$$I(0)=\pi k\sum_{n=1}^{\infty}(-1)^{n-1}\beta^{2n}\Gamma(n)/(2n-1)!=1, \qquad (5)$$

so that k can be written as a series;

$$k=1/\pi\,(\beta^2-\beta^4/6+\beta^6/60-\beta^8/840+\ldots). \qquad (6)$$

The result in (5) was originally given by Legendre,[4] using a different method. For our calculation, the first four terms of the series will be used. Clearly, I (1) and I (2) represent the first and second moments of $f(\emptyset,\theta)$ for $\emptyset$, so that the mean, μ, and the variance, σ^2, are given by

$$\mu=I(1)=\pi k\sum_{n=1}^{\infty}(-1)^{n-1}\beta^{2n-1}\Gamma(n+\tfrac{1}{2})/(2n-1)!$$
$$=\frac{\sqrt{\pi}}{2}\,\frac{\beta(1-\beta^2/4+\beta^4/32-\beta^6/384+\ldots)}{(1-\beta^2/6+\beta^4/60-\beta^6/840+\ldots)} \qquad (7)$$

and

$$\sigma^2=I(2)-I(1)^2=\frac{\beta^2(1-\beta^2/3+\beta^4/20-\beta^6/210+\ldots)}{(1-\beta^1/6+\beta^4/60-\beta^6/840+\ldots)}$$
$$-\frac{\pi\beta^2}{4}\,\frac{(1-\beta^2/4+\beta^4/32-\beta^6/384+\ldots)^2}{(1-\beta^2/6+\beta^4/60-\beta^6/840+\ldots)^2} \qquad (8)$$

For small β, $\mu=\sqrt{\pi}\beta/2$ and $\sigma^2=\beta^2(1-\pi/4)$. This agrees with the results obtained for a Rayleigh distribution,[5,6] which describes a normal distribution about a point in a plane as opposed to the case treated here which is a distribution about a point on the surface of a sphere.

<u>Comparison of the assumed distribution with experiment.</u>

Using the Schulz x-ray method, Swift et al.[1] have measured the distribution of (0001) planes in sintered $SmCo_5$ magnets. The volume fraction of (0001) plane orientations measured on their best aligned sample is plotted in Fig. 2. Since the volume fraction at each orientation was measured, the histogram should represent $f(\emptyset,\theta)$ for a certain value of β. If the distribution measured by Swift et al. is the same as the distribution given in equation (1), then a plot of $\ln(f(\emptyset,\theta))$ vs $\emptyset^2$ will be linear and have a slope of $1/\beta^2$. For this sample, the value of β obtained in this way was 0.38. However, a histogram calculated for $\beta=0.43$ agreed better with the experimental distribution, as shown in Fig. 2a. Even better agreement is obtained by assuming a fraction 0.26 of the particles are distributed completely at random with the remaining 0.74 fraction aligned with $\beta=0.34$. This distribution is compared with the measured data in Fig. 2b, and the agreement is excellent.

D. L. Martin[2] has reported a method of metal-

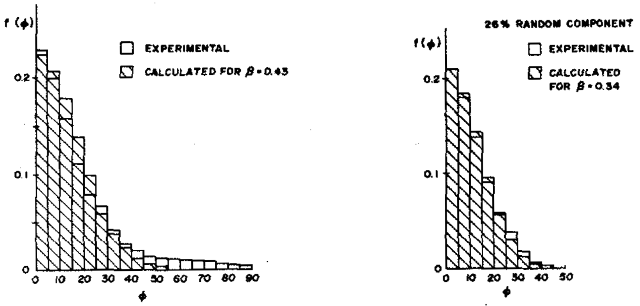

Fig. 2. Distribution of particle axes in a sintered $SmCo_5$ magnet measured by Swift et al.[1] a (left), The measured histogram and a calculated histogram for $\beta = 0.43$. b (right), The histogram with an assumed random fraction of 0.26 removed and a calculated histogram for $\beta = 0.34$.

lographic analysis to determine the texture of $SmCo_5$. On heat treating a sintered sample for 10 days at 1025 K, $SmCo_5$ undergoes a eutectoid decomposition, $SmCo_5 \rightarrow Sm_2Co_7 + Sm_2Co_{17}$. Lamellae precipitate preferentially on the basal plane of the hexagonal $SmCo_5$, as shown in Martin's micrograph, Fig. 3. A grid is placed over the micrograph, and an angle of misorientation ϕ' is measured at each point on the grid. (The angle of misorientation measured by Martin was actually $\phi'+\pi/2$ and is redefined here to simplify the comparison with the model). Fig. 4 shows a histogram of measured angles measured by Martin for $\phi' \geq 0$.

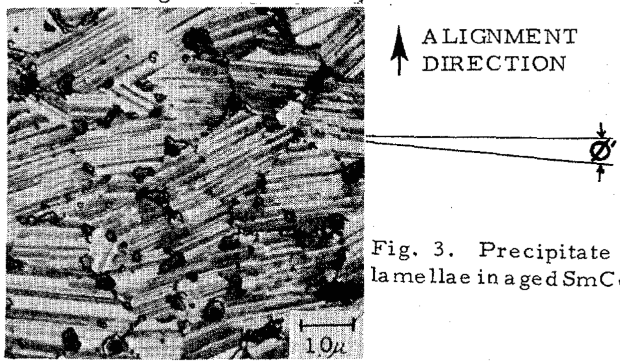

Fig. 3. Precipitate lamellae in aged $SmCo_5$.[2]

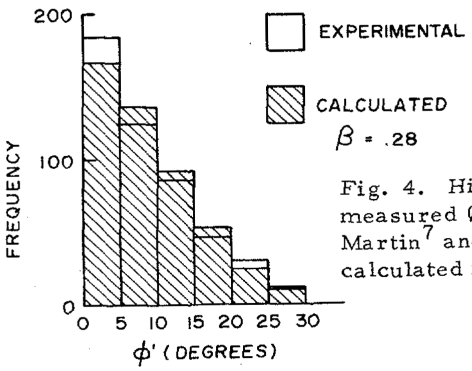

Fig. 4. Histogram of measured ϕ' from Martin[7] and a histogram calculated for $\beta = 0.28$.

The angle ϕ', however, is not equal to the angle ϕ defined in Fig. 1. The angle ϕ' is the projection of ϕ onto the plane of the metallographic section of the sample. The geometric relationship between ϕ' and ϕ is shown in Fig. 5. Mathematically, the relationship between ϕ' and ϕ is shown by the following argument.

If $\vec{P}$ and $\vec{P}'$ are taken to be unit vectors, their components in cartesian coordinates are,

$\vec{P} = [\cos\theta\sin\phi, \sin\theta\sin\phi, \cos\phi]$ and

$\vec{P}' = [\sin\phi', 0, \cos\phi']$

If α is defined as the angle between $\vec{P}$ and $\vec{P}'$, then

$$\cos\alpha = \vec{P}\cdot\vec{P}' = \cos\theta\sin\phi\sin\phi' + \cos\phi\cos\phi'$$

The law of spherical angles gives

$$\cos\phi = \cos\alpha\cos\phi' = \cos\theta\sin\phi\sin\phi'\cos\phi' + \cos\phi\cos^2\phi'$$

Dividing by $\cos\phi$, we have

$$1 = \cos\theta\tan\phi\sin\phi'\cos\phi' + \cos^2\phi'$$

$$\tan\phi' = \cos\theta\tan\phi. \qquad (9)$$

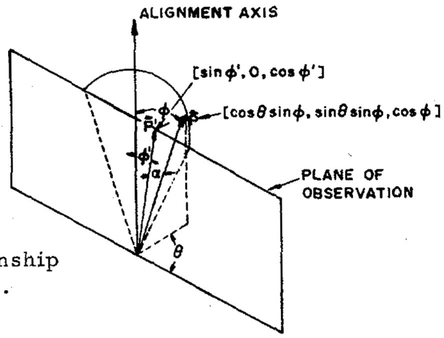

Fig. 5. Relationship between ϕ and ϕ'.

An assumed distribution obeying (1) for a fixed β can be converted to a predicted projected two-dimensional distribution by using equation (9). Fig. 4 shows a histogram calculated for $\beta = 0.28$ superimposed on Martin's measured histogram; the agreement is good. It seems unnecessary in this case to assume a random component.

<u>Calculation of magnetization curves.</u>

To calculate the magnetization of an assembly of single domain particles whose distribution about an alignment axis is given by (1), we consider the torque exerted by an applied field $\vec{H}$ on the particles whose orientation is in an element of spherical area dA. This torque is exactly balanced by the crystal anisotropy torque acting to hold the magnetization along the local easy axis $\vec{P}$. Thus

$$\vec{H} \times \vec{M}_s = dE_k/d\gamma \qquad (10)$$

where γ is defined in Fig. 6a for the case when $\vec{H}$ is applied perpendicular to the alignment axis and in Fig. 6b for the case when $\vec{H}$ is applied parallel to the alignment axis. Since $E_k = K_1\sin^2\gamma + K_2\sin^4\gamma$ for a hexagonal crystal, equation (10) can be expressed as

$$HM_s\sin(\lambda-\gamma) = K_1\sin2\gamma + 2K_2\sin2\gamma\sin^2\gamma$$
$$= K_1\sin2\gamma(1+\frac{2K}{K_1}2\sin^2\gamma) \qquad (11)$$

for the perpendicular field case and

$$HM_s\sin(\phi-\gamma) = K_1\sin2\gamma + 2K_2\sin2\gamma\sin^2\gamma$$
$$= K_1\sin2\gamma(1+\frac{2K}{K_1}2\sin^2\gamma) \qquad (12)$$

for the parallel field case. For any combination of H, ϕ, θ, K_1, and K_2/K_1, equations (11) or (12) can be used to determine γ for the particles whose orientation is in dA.

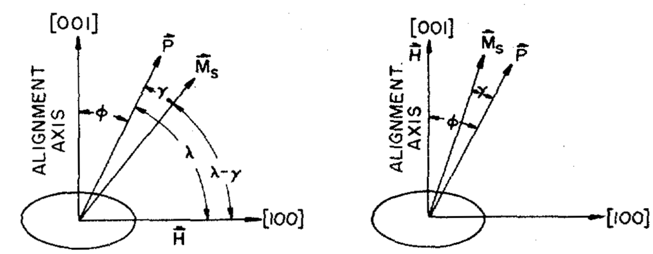

Fig. 6. Definition of angles in an applied field. a (left), field parallel to hard axis. b (right), field parallel to easy axis.

To obtain the resultant magnetization of the entire sample, it is necessary to sum the individual contributions of each unit area dA. Thus, $M_\perp/M_s$ can be found by evaluating the integral

$$M_\perp/M_S = \int_0^{2\pi}\int_0^{\pi/2} \cos(\lambda-\gamma)\, f(\phi,\theta)\sin\phi\, d\phi\, d\theta \qquad (13)$$

Similarly, $M_\parallel/M_s$ is given by

$$M_\parallel/M_S = \int_0^{2\pi}\int_0^{\pi/2} \cos(\phi-\gamma)\, f(\phi,\theta)\sin\phi\, d\phi\, d\theta \qquad (14)$$

In practice, the integrals (13) and (14) can be solved using the computer to generate $M_\perp/M_s$ or $M_\parallel/M_s$ vs $h(=HM_s/(2K_1+4K_2))$ curves, for selected values of β. The results of calculations based on (13) are given in Table I for $K_2=0$. Table II gives the results of calculations based on (14) for the special case when $H=0$, $\gamma=0$, for which $M_\parallel/M_s=M_r/M_s$, the ratio of remanence to saturation, or remanence ratio (sometimes called the alignment factor).

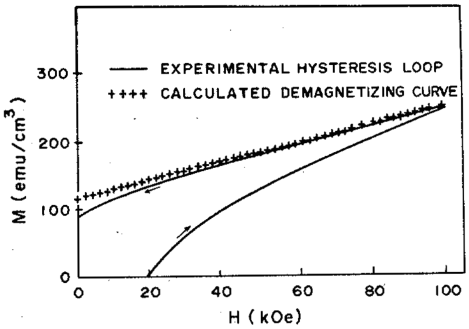

Fig. 7. An experimental hard axis hysteresis loop for $SmCo_5$ compared to a calculated demagnetizing curve. The calculated curve is for $K_1 = 2.6 \times 10^8$ ergs/cm^3, $K_2 = 0$, and $\beta = 0.25$.

TABLE I

$M_\perp/M_s$ vs h for several values of β.

h	β=0.05	β=0.10	β=0.15	β=0.20	β=0.25	β=0.30	β=0.35	β=0.40	β=0.45
0.0	0.028	0.056	0.084	0.111	0.138	0.164	0.190	0.214	0.238
0.1	0.127	0.154	0.181	0.207	0.233	0.258	0.281	0.304	0.326
0.2	0.226	0.251	0.276	0.301	0.324	0.347	0.368	0.389	0.409
0.3	0.323	0.347	0.369	0.391	0.412	0.432	0.451	0.470	0.487
0.4	0.420	0.440	0.460	0.478	0.496	0.513	0.530	0.546	0.560
0.5	0.516	0.532	0.547	0.562	0.576	0.590	0.604	0.617	0.629
0.6	0.610	0.620	0.631	0.642	0.652	0.663	0.673	0.683	0.692
0.7	0.702	0.706	0.711	0.717	0.723	0.730	0.737	0.744	0.751
0.8	0.790	0.787	0.786	0.786	0.789	0.792	0.795	0.799	0.804
0.9	0.874	0.861	0.853	0.849	0.847	0.846	0.847	0.848	0.850
1.0	0.944	0.923	0.910	0.901	0.895	0.892	0.890	0.889	0.889
1.2	0.990	0.976	0.964	0.956	0.949	0.949	0.940	0.938	0.936
1.4	0.996	0.990	0.982	0.976	0.971	0.967	0.963	0.961	0.959
1.6	0.998	0.994	0.990	0.985	0.982	0.978	0.976	0.973	0.972
1.8	0.999	0.996	0.993	0.990	0.987	0.985	0.983	0.981	0.979
2.0	0.999	0.998	0.995	0.993	0.991	0.989	0.987	0.986	0.984

TABLE II

β	M_r/M_s	β	M_r/M_s	β	M_r/M_s	β	M_r/M_s
.05	.998	.15	.989	.25	.969	.35	.941
.06	.998	.16	.987	.26	.967	.36	.938
.07	.997	.17	.986	.27	.964	.37	.934
.08	.996	.18	.984	.28	.962	.38	.931
.09	.996	.19	.982	.29	.959	.39	.928
.1	.995	.2	.980	.3	.956	.4	.924
.11	.994	.21	.978	.31	.953	.41	.920
.12	.992	.22	.976	.32	.950	.42	.917
.13	.991	.23	.974	.33	.947	.43	.913
.14	.990	.24	.972	.34	.944	.44	.909

Experimental hard axis magnetization curves have been compared to the calculated magnetization curves in Table I to find the combination of K_1 and β which gives the best agreement between experimental and calculated curves.[3] There is excellent agreement between calculated and experimental curves, as shown in Fig. 7, in decreasing applied fields from 100 to 20 kOe. It is in this region where the magnetization is expected to change by rotation as assumed in the calculation.

CONCLUSIONS

We have presented a mathematical model for the orientation of individual particles in an aligned compact. For the specific case of sintered $SmCo_5$ magnets, the model agrees with x-ray, metallographic, and magnetic measurements on $SmCo_5$. The model has been used to determine the anisotropy constant and the degree of misorientation from hard axis magnetization curves of $SmCo_5$ and can be used to predict other magnetic parameters such as M_r/M_s and (BH)max.

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