Angular distribution of magnetization in coated magnetic films

F. Habbal
Polaroid Corporation, Waltham, Massachusetts

W. T. Vetterling
Lyman Labortatory, Harvard University, Cambridge, Massachusetts

Recent Mössbauer studies of magnetic films have described the orientational distribution of spins in terms of a mean angle of orientation between the spin direction and the direction of γ-ray propagation. These studies have improperly interpreted the quantity \( \sin^2 \theta \), and have failed to account for the effects of saturation. We present an improved analysis which describes the spin orientational distribution in terms of an expansion in spherical harmonics. Coefficients in the expansion are determined from line intensities in the Zeeman-split spectra. A similar description, using the same distribution function, is applied to the angular dependence of the squareness of the hysteresis loop. Mössbauer results rely most heavily on the \( l = 2 \) coefficients in the expansion. The magnetization, however, depends also on coefficients describing the characteristics of individual particles. Examples of the application of these techniques to films with different distributions of acicular particles are given.

PACS numbers: 76.80. + y, 75.60.Jp

I. INTRODUCTION

An important parameter controlling the signal output level in conventional particulate magnetic media is the direction of the easy axis of magnetization of the particles with respect to the read head.\(^1\)\(^2\) For highly acicular particles, the easy axis is along the axis of the particles, and orienting the magnetic pigments in the direction of motion past the head is desirable. The ability to determine particle orientation is therefore important to the choice of applied fields during coating. Attempts to model particle orientational distributions have been made only with ad hoc trial functions.\(^3\)\(^4\)

A weakness of previous papers on particle orientation measurement with the Mössbauer effect has been an improper interpretation of the quantity \( \sin^2 \theta \) where \( \theta \) is the angle between the spin direction and the gamma ray propagation direction, and a failure to account for the effects of the saturation of the spectral lines.\(^5\) The result has been a number of faulty reports of inexplicably large values of “canting” angles.\(^6\)\(^7\) In this work we present a scheme for the proper study of spin-orientational properties of particulate magnetic films.

II. MöSSBAUER STUDIES

The Mössbauer spectrum for \(^{57}\)Fe in \( \gamma \)-Fe\(_2\)O\(_4\), consists of a symmetrical set of six absorption lines corresponding to the allowed dipole transitions between the magnetically split ground state and first excited state, having spins 1/2 and 3/2, respectively. The absorption coefficients \( \mu_n \) leading to the three different observed line intensities \( I_n \) are functions of the angle \( \theta \) between the gamma-ray path and the spin direction

\[
\begin{align*}
\mu_1 &\propto 1/(1 + \cos^2 \theta), \\
\mu_2 &\propto \sin^2 \theta, \\
\mu_3 &\propto 3/(1 + \cos^2 \theta).
\end{align*}
\]

The ratio of \( \mu_3 \) to \( \mu_1 \) or \( \mu_3 \) therefore determines the value of \( \cos^2 \theta \). For an ensemble of domains, an averaging must be performed. Furthermore, in considering the line amplitude ratio, \( I_2/I_1 \), which is involved in finding \( \langle \cos^2 \theta \rangle \), correction must be made for saturation due to absorber thickness. The effect of saturation on the intensity of the lines can be considered by writing the intensity of line “n” in the form:

\[
I_n = I_0 [1 - \exp(-\mu_n x)].
\]

For a random spin distribution, the \( \mu_n \) are in the ratio \( 3:2:1:2:3 \), and the line intensities assume these ratios for thin samples. For more general distributions, we find

\[
\langle \cos^2 \theta \rangle = (\delta - 1)/\delta + 1,
\]

where

\[
\delta = 4 \left[ \ln \left( -1/2 + \sqrt{\frac{I_2}{I_1} - \frac{3}{4}} \right) / \left[ \ln \left( 1 - \frac{I_2}{I_0} \right) \right] \right].
\]

As an example, we give in Table I measured values of \( \langle \cos^2 \theta \rangle \) for three samples—a \( \gamma \)-Fe\(_2\)O\(_4\) powder, and two samples of magnetic tape, called T60 and T65, with different degrees of orientation. Measurements were obtained with the gamma ray propagating perpendicularly to the plane of the tape. These values may be compared with the values of \( \langle \cos^2 \theta \rangle = 0.33 \) for completely disordered spins, 1.00 for spins parallel to the gamma-ray propagation, and zero for spins perpendicular to the gamma ray propagation. The value obtained for the powder sample is close to that of the randomly oriented spins, whereas sample T65 shows a small value of \( \langle \cos^2 \theta \rangle \) indicating that the particles are oriented preferentially in the plane of the tape. These conclusions are

<table>
<thead>
<tr>
<th>Sample</th>
<th>( I_2/I_1 )</th>
<th>( I_0 )</th>
<th>( I_2/I_0 )</th>
<th>( \langle \cos^2 \delta \rangle )</th>
<th>( \langle \cos^2 \theta \rangle )</th>
<th>( OR )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Powder</td>
<td>2.27</td>
<td>0.0115</td>
<td>0.46</td>
<td>2.06</td>
<td>0.35</td>
<td>1.05</td>
</tr>
<tr>
<td>T60</td>
<td>2.26</td>
<td>0.0831</td>
<td>0.61</td>
<td>1.35</td>
<td>0.14</td>
<td>1.80</td>
</tr>
<tr>
<td>T65</td>
<td>2.25</td>
<td>0.0051</td>
<td>0.67</td>
<td>1.16</td>
<td>0.07</td>
<td>2.24</td>
</tr>
</tbody>
</table>

consistent with the behavior of the orientation ratio 
\[ OR = S(\pi/2, 0)/S(0, \pi/2) \], where \( S \) is the squareness of the 
\( M(H) \) loop, defined as the ratio \( M_r/M_s \) between the remanent 
and saturation magnetizations. It should be noted that except for the extreme values of zero and one, a given value of \( \langle \cos^2 \theta \rangle \) is not associated with a unique array. The following 
analysis is needed to determine the full distribution function of the particles.

III. THE ORIENTATIONAL DISTRIBUTION FUNCTION

A general way of writing the spin orientational distribution function is in terms of spherical harmonics,

\[ P(\theta, \phi) = \sum_{l,m} A_{lm} Y_{lm}(\theta, \phi), \tag{5} \]

where \( \theta, \phi \) are angles of a given particle axis with respect to the 
laboratory frame. For convenience, the polar axis is chosen perpendicular to the film. We require a determination of the coefficients \( A_{lm} \). The distribution function must be real-valued and normalized, and may be taken as symmetric upon inversion. Consequently it is written

\[ P(\theta, \phi) = \frac{1}{4\pi} + a_{20} \sqrt{\frac{5}{16\pi}} (3 \cos^2 \theta - 1) \]
\[ + \sqrt{\frac{60}{32\pi}} \sin^2 \theta \text{Re} \{ (a_{22} + ib_{22}) e^{2i\phi} \}, \tag{6} \]

where we have defined \( A_{20} = a_{20} \) and \( A_{22} = a_{22} + ib_{22} \). In terms of the measurement we have described above, it follows that

\[ \langle Y_{20}(\theta, \phi) \rangle = A_{20} = \sqrt{\frac{5}{4\pi}} \frac{\delta - 2}{\delta + 1}. \tag{7} \]

The other unknown \( l = 2 \) coefficients can also be measured by determining \( \langle Y_{20} \rangle \) with the gamma ray propagating at different angles with respect to the film, and using the "addition theorem" to transform back to the initial coordinates,

\[ \langle Y_{20} \rangle_{\alpha} = \sqrt{\frac{4\pi}{5}} \sum_{m=-2}^{2} A_{2m} Y_{2m}(\beta, \alpha), \tag{8} \]

with \( \beta, \alpha \) being the Euler angles specifying the tilt and 
rotation of the film with respect to its original orientation 
perpendicular to the gamma ray path. Clearly, measurements of \( \langle Y_{20} \rangle_{\alpha} \) for several directions \( \beta, \alpha \) are sufficient to determine all of the \( l = 2 \) coefficients. As an example, Table II shows results for film T65 at four film orientations. Some of the Mössbauer spectra are shown in Fig. 1. Applying the formulas above, we find that

\[ a_{20} = -0.26, \quad a_{22} = 0.21 \pm 0.04, \quad b_{22} = 0.03, \]

and hence that, up to terms \( l = 2 \)

\[ P(\theta, \phi) = 0.16 - (0.25)\cos^2 \theta + (0.16)\sin^2 \theta \cos(2\phi). \]

This distribution function is highly planar and well oriented 
along the direction \( \phi = 0 \) (along which a field was applied 
during manufacture). Notice the slightly negative value of 
\( P(\theta, \phi) \) along \( \theta = 0 \), which is an artifact of the truncation of the harmonic expansion at \( l = 2 \).

IV. MAGNETIZATION STUDIES

In considering the squareness of the \( M(H) \) loop, we need to take into account the distribution of the particles, which is 
given by \( P(\theta, \phi) \) and the angular dependence of magnetic properties of the individual particles. In general, \( S \) can be expressed by the additional harmonic expansion

\[ S(\theta', \phi') = \sum_{l,m} C_{lm} Y_{lm}(\theta', \phi'). \tag{9} \]

[In the case of uniaxial particles, this expansion is of the function \( S(\theta', \phi') = |\cos \theta'|. \) For illustration, we consider only the two leading terms in the series, and we take the particles to be cylindrically symmetric, with \( \theta' \) measured 
with respect to the cylinder axis. If this axis is positioned at 
angles \( \alpha, \beta \) with respect to some laboratory fixed frame, then 
the squareness with respect to the angles \( \theta, \phi \) of the fixed 
frame are given by the addition theorem:

\[ S_{\alpha, \beta}(\theta, \phi) = C_{oo} Y_{00} \]
\[ + C_{20} \sum_{m=-2}^{2} \sqrt{\frac{4\pi}{5}} Y_{2m}(\theta, \phi) Y_{2m}^{*}(\beta, \alpha). \tag{10} \]

For a collection of particles with orientational distribution 
\( P(\alpha, \beta) \), the net squareness is

\[ S(\theta, \phi) = \int S_{\alpha, \beta}(\theta, \phi) P(\alpha, \beta) d\Omega_{\alpha, \beta}, \tag{11} \]

so that if we apply the expansion of \( P(\alpha, \beta) \) introduced earlier, we find

\[ S(\theta, \phi) = \frac{C_{oo}}{\sqrt{4\pi}} + C_{20} \sum_{m=-2}^{2} \sqrt{\frac{4\pi}{5}} A_{2m} Y_{2m}(\theta, \phi). \tag{12} \]

By studying squareness as a function of angle, we can
measure $C_{00}$ and the quantities $C_{20} A_{20}$. Comparison with Mössbauer results then allows the determination of $C_{20}$. Consequently, a combination of these two methods indicates the properties not only of the film, but also of its composite particles.

Figure 2 presents measurements of squareness for film T65 as a function of angle in the plane $xy$ of the tape, and the plane $yz$ perpendicular to it. The solid curves of Fig. 2 show $S_{xy}$ and $S_{yz}$ obtained from Eq. (12) and yield two independent determinations of $C_{20} A_{20}$ which were found to agree within 10%. Combining these parameters with the Mössbauer result, we obtain the values $c_{00} = 1.60$ and $c_{20} = 1.04$, leading to the following expression for the squareness of a typical particle:

$$S^2 = 0.12 + 0.98 \cos^2 \theta'. $$

Extensions of the methods described here to higher orders in $l$ and to particles of noncylindrical symmetry will be considered in future work.

ACKNOWLEDGMENTS

We are grateful to J. Averell for discussions, to B. Falabella for providing samples, and to R. Seeg for collecting magnetization data.