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Properties of amorphous rare earth-transition metal thin films relevant to thermomagnetic recording

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Abstract. — Properties of amorphous RE-(Fe, Co) thin films relevant to thermomagnetic recording are reviewed. Attention is paid to the writing-, reading- and erasure process. The advantages and disadvantages of the amorphous materials are considered. Experimental data on the writing process are given.

1. Introduction. — The last five years much attention has been paid to amorphous metallic materials. Here an important group of alloys are the rare earth (RE)-transition metal (TM) thin films, which can be prepared both by vapour deposition and by sputtering. These methods of preparation are relatively simple because no special measures with respect to the substrate temperature are required; moreover the materials can be prepared within wide composition ranges. The interest in amorphous RE-TM films is stimulated by the fact that some of them possess magnetic properties that are very suitable for applications. An application suggested some time ago is that as a magnetic bubble memory [1]. Here a basic requirement is the presence of magnetic anisotropy perpendicular to the film plane combined with a minute coercive force \(H_c\). A well-known material showing these properties is amorphous Gd-Co, possibly modified by other elements like Mo or Cu [2]. Some amorphous RE-TM films with perpendicular anisotropy show high \(H_c\)-values. It has been demonstrated that certain compositions of amorphous Gd-Co with relatively high \(H_c\) are suitable for thermomagnetic recording [3]. Recently [4] it has been proved that also some Fe based amorphous films are applicable in this respect.

Thermomagnetic recording is a method of (generally) digital information storage that is achieved by means of local (laser) heating of a magnetic thin film. Written bits can be read by virtue of the existence of magneto optical effects e.g. Faraday- or Kerr-rotation. In this paper we will treat the specific requirements put to the materials in this application and answer the question to which extent they can be met in amorphous RE-TM films.

This paper is organized as follows. In the next section a brief review is given of the principles of thermomagnetic recording, including some remarks on the reading process and on the possibility of erasing written information. In addition we sum up the most important material requirements. In section 3 the amorphous RE-TM materials are introduced; their properties are summarized. In section 4 the requirements for thermomagnetic recording will be discussed with respect to these materials. A number of recording experiments are reported in section 5. Finally the advantages and disadvantages of amorphous relative to crystalline metallic materials [5, 6, 7] in magnetic recording are discussed in section 6.

2. Thermomagnetic recording. — There exist a number of ways to realize thermomagnetic writing [8, 9]. The more common ones are those that make use of the decrease of \(H_c\) with temperature in the vicinity of the Curie temperature \(T_C\) or the magnetic compensation temperature \(T_{comp}\). We will refer to these techniques as \(T_c\)-writing and \(T_{comp}\)-writing respectively. In both methods it is essential that one deals with ferro- or ferrimagnetic material in the shape of a thin film. It is required to have a reasonably high \(H_c\) in addition to the presence of magnetic anisotropy normal to the film plane.

In the case of \(T_c\)-writing [10], the pre-magnetized film is locally heated to above \(T_c\) and thereupon cooled to the ambient temperature in a magnetic field directed opposite to the original magnetization. The field consists of the magnetic closure flux, if necessary combined with an applied external field. The formed domain has the shape of a cylinder with the axis perpendicular to the film plane. Provided that \(H_c\) is sufficiently large it is stable, even when the applied field is removed. When \(T_c\)-writing is applied both ferro- and ferrimagnetic materials can be used.

In ferrimagnetic materials the two sublattice (or when the material is amorphous; sub-network) magne-
tizations can cancel each other at a certain temperature. At this temperature \( T_{\text{comp}} \) the material attains an extremely high value of \( H_c \). At temperatures sufficiently different from \( T_{\text{comp}} \), \( H_c \) becomes relatively low and switching of the magnetization direction becomes possible. \( T_{\text{comp}} \) writing is based on this (steep) decrease of \( H_c \). The film is operated at or near \( T_{\text{comp}} \); it is locally heated to a lower \( H_c \) state so that in the heated area the magnetization can be reversed by means of the applied field. In this case the applied magnetic switching field is essential, in contradistinction to \( T_c \)-writing where the magnetic closure flux may be sufficiently large to switch the magnetization without external field.

For reading purposes it is important that the films have a uniaxial anisotropy with the axis normal to the film plane, because in this situation Faraday- and Kerr-effect are maximal [11]. Because we consider metallic materials, in which the optical absorption coefficient \( (z) \) is large, the polar Kerr effect is the most attractive property for the reading process.

An important and inherent feature of thermomagnetic recording is the erasure of the written information with the aid of an applied field, possibly combined with locally heating of the film.

### 3. Amorphous RE-TM films.

For some years it is known that films of RE-TM can be prepared in the amorphous state both by vapour deposition [12] and by sputtering techniques [13]. It is possible, even on substrates at ambient temperature, to vary the composition of the films within wide boundaries, without loss of the amorphous structure. This is shown in figure 1 for a number of Co- and Fe-alloys. Within the frame work of this paper it is important to note that every composition within the shaded areas in figure 1 can be prepared in the amorphous state. This allows tailoring of the properties of the films in relation to their application.

A disadvantage of the amorphous materials is the tendency to change their structure irreversibly at somewhat elevated temperatures [14] such as to lower the free energy of the system [15]. At still higher temperatures the amorphous structure crystallizes into one or more crystalline phases, which as a rule have properties that differ completely from those of the amorphous phase. In the present materials crystallization sets in at about 250 °C and is completed at about 400 °C.

Because thermomagnetic recording is a technique in which some shortperiod increase of temperature is inevitable, the gradual structure change of amorphous materials can be rather inconvenient.


Starting with the demand for perpendicular magnetic anisotropy it is remarkable that only a very small number of the known amorphous metallic films [16] shows this property. In figure 2 we review experimental information on amorphous RE-TM films possessing such magnetic anisotropy. From this figure it is obvious that sometimes there exists a big difference between vapour deposited and sputtered films. The main reason is the incorporation of gas atoms in the film during preparation [17, 18, 19]. All materials showing perpendicular anisotropy belong to the group of alloys containing Fe or Co and one of the heavy RE elements (Gd, Tb, Dy, Ho, Er). Moreover all compositions lie in the vicinity of the compensation compositions (see figure 3). Perpendicular anisotropy implies that the uniaxial anisotropy field, directed normal to the film plane \( (2 K_u / M_s) \) exceeds the shape anisotropy \( (4 \pi M_s^2) \), i.e.

\[
K_u > 2 \pi M_s^2,
\]

where \( K_u \) is the anisotropy energy and \( M_s \) the saturation magnetization. This situation can be realized easily in compositions near the magnetic compen-

---

**Fig. 1.** — Composition ranges over which RE\(_{1-x}\)TM\(_x\) thin films may be prepared in the amorphous state by vapour deposition. Boundaries indicated by a dashed line are not amorphous-crystalline boundaries but no further experimental data are available in those cases. The data of Nd-Fe and Nd-Co are taken from reference [41].

**Fig. 2.** — Amorphous RE\(_{1-x}\)(Fe, Co) films showing magnetic anisotropy perpendicular to the film plane. The data are taken both from literature and from own experiments. For vapour deposited Dy-Co and sputtered Tb-Co films no data were available. Under special circumstances [17, 23] it is possible to prepare vapour deposited amorphous Gd-Co films having perpendicular magnetic anisotropy.
The origin of this anisotropy is not yet completely understood, but a number of contributing factors is known, i.e. pair ordering of the TM-atoms [20], anisotropic microstructure [21, 22], stress [23] and magnetoelastic coupling [24].

In figure 3 values of $T_c$ and $T_{comp}$ are plotted. From this it is obvious that for the Fe-based materials $T_{comp}$ varies drastically with composition. To a lesser extent this holds for both $T_c$ and $T_{comp}$ of Co-based alloys. From a practical point of view this steep composition dependence of the critical temperatures is undesirable in view of reproducibility and homogeneity requirements.

There are two other requirements for $T_c$-writing. In amorphous materials $T_c$ must be smaller than the temperature at which crystallization sets in and secondly $T_c$ must exceed room temperature by a proper amount. In the case of $T_{comp}$-writing one demands, in order to facilitate the writing process, $T_{comp}$ to be about room temperature. However, from bit density or domain size arguments it follows that $T_{comp}$ must below room temperature [3].

The other basic requirement is a fairly high $H_c$ in order to stabilize the written domains against collapse or growth when the applied field is removed. The coercive force of a film depends strongly upon details of the preparation [17, 18], and moreover upon the thickness ($d$) of the film, to which $H_c$ is about inversely proportional at the thicknesses under consideration. In ferrimagnetic materials $H_c$ is high when $T \approx T_{comp}$. In figure 4 we give temperature- and composition dependence of $H_c$ for some ferrimagnetic films of interest. In table I some $H_c$-values from literature have been collected.
Table I. — Coercive field at room temp. of some amorphous RE$_{1-x}$TM$_x$ alloy films (t.w. = this work).

<table>
<thead>
<tr>
<th>RE</th>
<th>TM</th>
<th>$x$ (at %)</th>
<th>Prep. method</th>
<th>$H_c$ (Oe)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gd</td>
<td>Fe</td>
<td>0.74-0.88</td>
<td>sputt.</td>
<td>200-10</td>
<td>[34]</td>
</tr>
<tr>
<td>Gd</td>
<td>Fe</td>
<td>0.75-0.81</td>
<td>evap.</td>
<td>75-3</td>
<td>[35]</td>
</tr>
<tr>
<td>Tb</td>
<td>Fe</td>
<td>0.61-0.91</td>
<td>sputt.</td>
<td>5 500-150</td>
<td>[36; t.w.]</td>
</tr>
<tr>
<td>Dy</td>
<td>Fe</td>
<td>0.73-0.85</td>
<td>sputt.</td>
<td>10-800</td>
<td>[37]</td>
</tr>
<tr>
<td>Ho</td>
<td>Co</td>
<td>0.65</td>
<td>evap.</td>
<td>200-800</td>
<td>[25]</td>
</tr>
<tr>
<td>Gd</td>
<td>Co</td>
<td>0.77</td>
<td>sputt.</td>
<td>0.5-2</td>
<td>[1]</td>
</tr>
</tbody>
</table>

For reading purposes a square hysteresis loop is favoured. The reading process occurs in zero magnetic field and a square hysteresis loop assures the largest possible magneto optical effect.

Amorphous materials in which $T_{\text{comp}}$-writing has been reported are Ho-Co [25] and Gd-Co [26]. The information about $T_x$-writing is much more extensive. Literature data together with results of our own experiments are collected in table II.

Another group of requirements is that related to optical and thermal properties. In the application under consideration the material will always have the shape of a thin film. The optimum thickness of the film follows from energy considerations. In order to make an efficient use of the available laser energy one likes the product of $x$ and film thickness to have values in the range of 1 to 5. In metals $x$ is of the order of $3 \times 10^5$/cm. This means that the optimal thickness is of the order of 0.1 $\mu$m.

Concerning the reading process we have to realize that in metallic materials besides $x$ also the reflectivity is relatively high. This implies that for reading it is obvious to use the Kerr effect [27]. Only for very thin films (thickness smaller than a few hundred Å's) the Faraday effect can be utilized. For RE-TM films the two effects are quite small as can be concluded from table III, in which also a value for the figure of merit of transmission $(2 F/\alpha)$ is given.

It has been suggested that the Kerr rotation ($\theta_k$) is mainly due to the magnetic moments of the TM part of the alloy [28-30]. Hence it is not expected in the amorphous metals considered, that the rotation can be much enlarged. As a matter of fact the value of $\theta_k$ can be increased drastically by employing an anti-reflection coating [31, 32]. However, the signal-to-noise ratio is proportional to $R \theta_k^2$ ($R$ is the reflectivity), so the increase of $\theta_k$ is partially undone by the decrease of $R$ [8]. The improvements achieved by coating layers are mainly found in a reduction of the medium surface noise.

In thermomagnetic recording another material property of interest is the thermal conductivity. The thermal diffusivity partly determines the amount of energy required to heating the material (locally)

Table II. — Thermomagnetic writing data in some amorphous RE$_{1-x}$TM$_x$ thin films (t.w. = this work).

<table>
<thead>
<tr>
<th>RE</th>
<th>TM</th>
<th>$x$ (at %)</th>
<th>Prep. method</th>
<th>$T_c$ or $T_{\text{comp}}$ (°C)</th>
<th>Writing energy (nJ)</th>
<th>Pulse length (µs)</th>
<th>$H_{\text{app.}}$ (Oe)</th>
<th>Domain diam. (µm)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ho</td>
<td>Co</td>
<td>0.65</td>
<td>evap.</td>
<td>$T_{\text{comp}} = 46$°C</td>
<td>5-10 $\times 10^3$</td>
<td>0.5/µm$^2$</td>
<td>10$^{-3}$</td>
<td>30-200</td>
<td>2.5-17</td>
</tr>
<tr>
<td>Gd</td>
<td>Co</td>
<td>—</td>
<td>sputt.</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Tb</td>
<td>Fe</td>
<td>0.738</td>
<td>sputt.</td>
<td>$T_c = 150$°C</td>
<td>50</td>
<td>10</td>
<td>50</td>
<td>3</td>
<td>[4]</td>
</tr>
<tr>
<td>Dy</td>
<td>Fe</td>
<td>0.742</td>
<td>sputt.</td>
<td>$T_c = 60$°C</td>
<td>20</td>
<td>10</td>
<td>50</td>
<td>3</td>
<td>[4]</td>
</tr>
<tr>
<td>Gd</td>
<td>Fe</td>
<td>0.79</td>
<td>evap.</td>
<td>$T_c = 210$°C</td>
<td>3.5</td>
<td>1</td>
<td>70</td>
<td>1.4</td>
<td>t.w.</td>
</tr>
<tr>
<td>Gd</td>
<td>Fe</td>
<td>0.74</td>
<td>evap.</td>
<td>$T_c = 230$°C</td>
<td>3.5</td>
<td>1</td>
<td>70</td>
<td>1.4</td>
<td>t.w.</td>
</tr>
<tr>
<td>Tb</td>
<td>Fe</td>
<td>0.80</td>
<td>evap.</td>
<td>$T_c = 150$°C</td>
<td>2.8</td>
<td>1</td>
<td>200</td>
<td>2.6</td>
<td>t.w.</td>
</tr>
<tr>
<td>Tb</td>
<td>Fe</td>
<td>0.72</td>
<td>evap.</td>
<td>$T_c = 150$°C</td>
<td>2.8</td>
<td>1</td>
<td>200</td>
<td>2.7</td>
<td>t.w.</td>
</tr>
<tr>
<td>Dy</td>
<td>Fe</td>
<td>0.78</td>
<td>evap.</td>
<td>$T_c = 60$°C</td>
<td>0.9</td>
<td>1</td>
<td>60</td>
<td>2.2</td>
<td>t.w.</td>
</tr>
</tbody>
</table>

Table III. — Some magneto optical data of amorphous RE$_{1-x}$TM$_x$ thin films. For comparison some crystalline materials are included (t.w. = this work).

<table>
<thead>
<tr>
<th>RE</th>
<th>TM</th>
<th>$x$ (at %)</th>
<th>Prep. method</th>
<th>$\theta_F$ ($^\circ$/cm)</th>
<th>$\theta_k$ ($^\circ$)</th>
<th>$\lambda$ (cm$^{-1}$)</th>
<th>$\lambda$ (nm)</th>
<th>$2 F/\alpha$ ($^\circ$)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gd</td>
<td>Co</td>
<td>—</td>
<td>sputt.</td>
<td>1.8 $\times 10^5$</td>
<td>—</td>
<td>8 $\times 10^5$</td>
<td>450-820</td>
<td>0.45</td>
<td>[26]</td>
</tr>
<tr>
<td>Tb</td>
<td>Fe</td>
<td>—</td>
<td>evap.</td>
<td>1.3 $\times 10^5$</td>
<td>—</td>
<td>6.6 $\times 10^5$</td>
<td>633</td>
<td>0.39</td>
<td>[38]</td>
</tr>
<tr>
<td>Gd</td>
<td>Fe</td>
<td>0.77</td>
<td>evap.</td>
<td>1.5 $\times 10^5$</td>
<td>0.25</td>
<td>7.5 $\times 10^5$</td>
<td>633</td>
<td>0.40</td>
<td>t.w.</td>
</tr>
<tr>
<td>Gd</td>
<td>Fe (+ 3 % Ag)</td>
<td>0.80</td>
<td>evap.</td>
<td>3.1 $\times 10^5$</td>
<td>—</td>
<td>—</td>
<td>633</td>
<td>1.0</td>
<td>[39]</td>
</tr>
<tr>
<td>MnBi(crystr)</td>
<td>—</td>
<td>0.40</td>
<td>—</td>
<td>5 $\times 10^5$</td>
<td>—</td>
<td>633</td>
<td>4</td>
<td>[40]</td>
<td></td>
</tr>
<tr>
<td>MnBi(crystr)</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>500</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>[28]</td>
<td></td>
</tr>
</tbody>
</table>
above $T_c$ or $T_{comp}$ and it influences the size of the written domain. In amorphous metals the thermal conductivity is low relative to crystalline metals. This has to do with the very small mean free path of the electrons in the amorphous state which is of the order of one interatomic distance [33].

5. Some recording experiments. — In this section some results of thermomagnetic writing experiments restricted to $T_c$-writing, will be shown. Figure 5 shows the written domains in Gd$_{0.23}$Fe$_{0.77}$ as observed by the polar Kerr effect. We used a focussed Ar laser (530 nm). With decreasing power the domains get smaller dimensions. This is plotted in figure 6. From figure 3 it is clear that there exists the possibility to choose $T_c$ at will between room temperature and 230 °C. The lower part of table II shows the minimum power required to write an oppositely directed domain for some amorphous RE-TM films with varying $T_c$-value. It is obvious that in the case of $T_c$-writing the value of $T_c$ is the main factor in determining the writing power.

Fig. 6. — Domain diameter of written domains versus laser power in a 1500 Å thick Gd$_{0.23}$Fe$_{0.77}$ amorphous film.

Specific advantages of amorphous RE-(Fe, Co) storage materials relative to other metallic thermomagnetic recording materials (like MnBi) are their relatively simple method of preparation, the absence of crystal structure transformations, the absence of grain boundaries (which implies an improvement in signal-to-noise ratio) and a low thermal conductivity. The disadvantages of these materials are of two kinds: the possibility to irreversible changes of the amorphous structure and to crystallization at relatively low temperatures and a rather small value of the magneto optical effects. More information about these two factors would be of great interest.

6. Conclusions. — Some amorphous RE-Fe and RE-Co films possess properties which make them attractive for (erasable) thermomagnetic recording applications. Writing-powers as low as 0.4 nJ/bit (about 1 μm$^2$) are observed. In the writing procedure it is not in all cases necessary to apply an external field but in many cases the presence of a field during writing enhances the magnitude of the reading signal.

Fig. 5. — Magnetic domains written in amorphous Gd$_{0.23}$Fe$_{0.77}$ in a thermomagnetic way with the aid of an Ar laser. The pulse length is 1 μs.

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