

on talented scientists working in small groups who understand and address the problem in its entirety. A commitment of larger resources aimed at increasing the likelihood of success often works in the opposite direction since the effort to be effectively managed is broken up into a series of interrelated tasks which are readily managed through PERT charts.

4. Recent attempts by the US Government to stimulate industrial R&D by building up efforts in universities and/or the National Laboratories does not address the critical problems confronting much of US industry. There is absolutely no substitute for industrial scientists who are effectively interfaced with the needs of the particular industry and its markets and have available to them the latest techniques to probe and address the future needs in materials.
5. It is essential that universities undertake to train high quality materials scientists skilled in synthesis and processing of new materials. It should be obvious that the development of new materials tailored to specific needs or equally as important the unexpected discovery of materials with unique properties lies primarily within the purview of the clever scientists trained effectively in synthesis and processing of materials.

Finally, let me return to the question posed in the opening of this article; namely, can one organize an advanced

materials R&D effort which addresses the opportunities of the 21st century? I would argue that for industry, the primary emphasis should be directed at fulfilling the perceived materials needs of the next two to five years as discussed in conclusion "1". Such an approach will not only impact the company's immediate needs, but hopefully would have sufficient latitude to permit for unexpected breakthroughs which address longer range opportunities. With respect to planning and implementing a materials R&D effort for the 21st century, past experience shows that it is extremely difficult to identify needs of the next five to seven years let alone 10–15 years from now. Thus, one must think in terms of national or even international programs to establish an infrastructure which 1) attracts talented young people into the science programs of our universities, 2) provides for continuity of high risk research in all sectors of the R&D establishment, and 3) places responsibility for commitment of funds and resources by the government and industry in the hands of technically astute leaders. In this way one can greatly increase the potential for successful development of advanced materials critical to high technology industries of the 21st century.

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Magneto-Optical Recording and Data Storage Materials**

Rare-Earth Transition-Metal
Alloys
Thermomagnetic Writing
Lorentz Microscopy

By Frans J. A. M. Greidanus* and Stefan Klahn

Amorphous rare-earth (RE) transition-metal (TM) alloys are used for magneto-optical (MO) recording a rapidly developing technology, which combines the possibility of achieving high bit densities with practically unlimited erasability and rewritability. During the last years new insights, relating material properties to recording performance, have

been obtained. New experimental techniques, such as the observation of magnetic contrast in the electron microscope, have made a major contribution to the understanding of domain formation processes. The RE-TM alloys have been most successful in recording applications until now. In these materials the RE-TM composition determines both the compensation and Curie temperatures and has a strong impact on the recording characteristics. Improvements in deposition techniques and the application of dielectric layers resulted in carrier-to-noise ratios of 61 dB. Despite major improvements, problems related to corrosion and structural relaxation, which lead to long-term instabilities, have not yet been solved completely. Another important topic is the so-called direct-overwrite problem, which will be discussed in relation to the material properties.

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1. Introduction

In the past ten years the development of optical recording technology has led to products for various applications, such as the Compact Disc video and audio systems and the write-once DOR (Digital Optical Recording) system, which are now on the market. The availability of relatively cheap and mass-produced solid-state lasers made these developments possible. The driving forces behind these developments were the considerable advantages of optical recording over other storage techniques. Readout is done by a laser focussed through a substrate on the storage layer, providing a non-contact technique insensitive to dust and fine scratches on the substrate. A second advantage is the possibility of achieving high bit densities (10^8 bits cm^{-2}). This value compares favorably with other existing technologies, as can be seen in Figure 1.

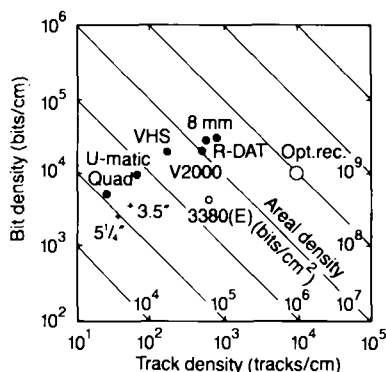


Fig. 1. Areal density for various recording systems. ●: helical scan, +: flexible disc, ○: rigid disc (courtesy of S. B. Luitjens).

In the history of optical recording three phases can be distinguished: 1) The oldest type of optical-storage systems are the replicated discs, used in the Compact Disc video and audio system. These are designed for the distribution of prerecorded information. 2) The second class is formed by write-once media. Here the writing process is based on the absorption of energy from a focussed laser beam, modulated according to the information to be recorded. The available commercial systems are based on the formation of holes in a reflective layer. 3) The latest class of optical systems, which is at present the subject of intensive research and development, employs media which can be erased and rewritten innumerable times. Here several different physical principles are being followed. Well known is the use of a reversible crystalline-to-amorphous phase transition in the recording film to store the information. A second method is based on a thermo-magnetic effect to write, and a magneto-optical (MO) effect to read out the information. In this review article we will focus our attention on the materials for this latter technique.

The principles behind MO recording have been known for years. However, it was not until the mid sixties that a

record and playback system employing a laser beam was suggested. Writing of domains in a magnetic material by heating it locally above the Curie temperature was suggested in 1958 by Mayer,^[1] long before an optical memory was envisioned. Mayer employed a focussed electron beam or the point of a heated needle. The first observation of an MO effect is even older and dates from 1846^[2] when Faraday observed that polarized light passed through a fluid in a strong magnetic field changes its polarization state. Ever since this phenomenon has been designated as the Faraday effect when it refers to the transmitted beam, whereas it is called the Kerr effect when it refers to the reflected beam.

The first proposal for employing a laser both for writing (high power) and reading (low power) dates from 1965, when Chang et al.^[3] discussed a memory based on $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ (GdIG). Since then several other materials, such as MnBi, EuO, CoPt and CoFe_2O_4 have been suggested and investigated with respect to their applications as an MO memory layer. The most important development, however, was started by Chaudhari et al.^[4] in 1973, with the observation that amorphous GdCo-layers possessing perpendicular anisotropy are suitable for MO recording. This discovery was the start of a series of exploratory studies in the rare-earth (RE) transition-metal (TM) binary and ternary alloy system with regard to their potential for MO data storage. These studies led, e.g., to the systems GdTbFe and TbFeCo which are the most suitable materials known today. To a large extent this is due to a steady increase in the signal-to-noise properties over the years as can be seen in Figure 2.

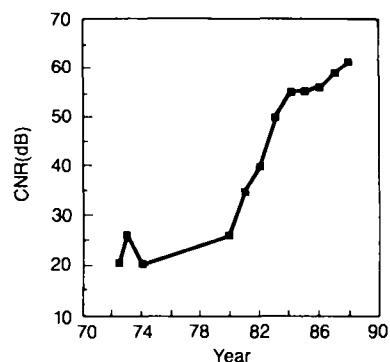


Fig. 2. Maximum reported carrier-to-noise ratio for MO recording (measured for a domain length of $1.5 \mu\text{m}$ at 1 MHz and 30 kHz bandwidth) versus year of publication.

2. Magneto-Optical Recording

The principle of optical readout of domains in a perpendicular MO layer is shown in Figure 3a. The change of the polarization direction when linearly polarized light is reflected from a magnetic surface is shown here. Phenomenologically, MO effects can be described by assigning to

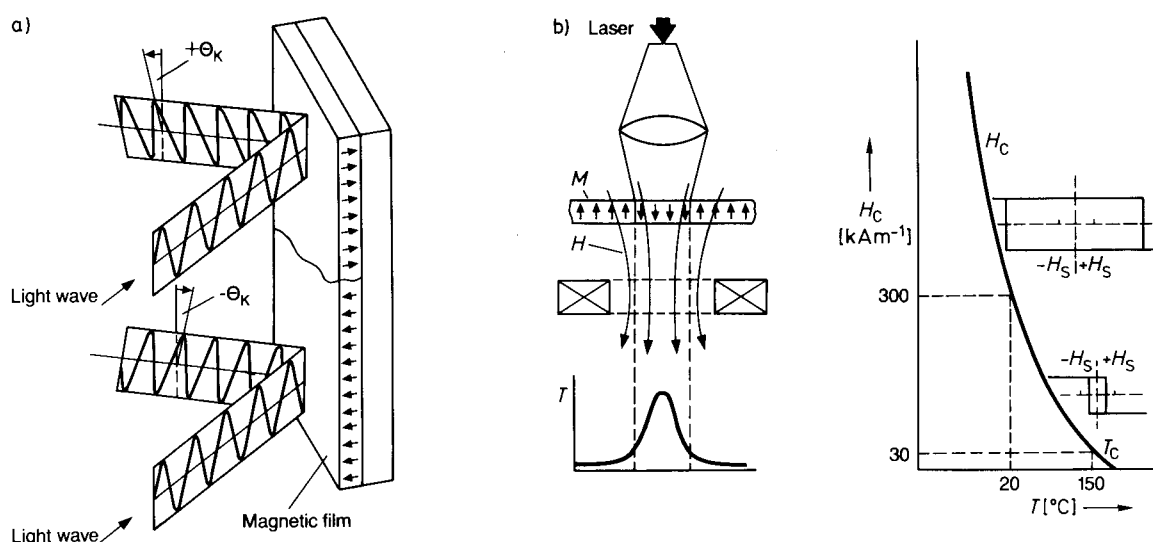


Fig. 3. a) Principle of MO reading. b) Principle of thermomagnetic writing.

the medium a dielectric tensor with non-zero off-diagonal elements. This is equivalent to introducing two different complex indices of refraction for right- and left-handed circularly polarized light. Microscopically these effects find their origin in the spin-orbit interaction, which couples the spin moment and the motion of the electron. The optical properties are influenced by the electron motion. The sign of the angle through which the polarization is rotated depends on the direction of the magnetization vector with respect to the propagation direction of the light. For practical materials the magnitude of the Kerr rotation is fairly small (typically $0.2\text{--}0.3^{\circ}$). Consequently the signals arising from the modulation of the polarization state of the reflected light are very small. Nevertheless, they can be detected with a good carrier-to-noise ratio (CNR) if sources of noise in the readout can be made sufficiently small.

Several schemes for thermomagnetic writing exist. With Curie point writing, the MO layer is heated above its Curie temperature and the magnetization is reversed in an applied magnetic field. A second method can be employed in materials in which the coercive field is a decreasing function of temperature. Upon heating, the magnetization can be reversed in an applied field larger than the coercive field of the layer at the elevated temperature, but smaller than the coercive fields in the remaining non-heated area of the film. In this way a domain can be written thermally, as illustrated in Figure 3b. This method is referred to as threshold writing when applied to ferromagnetic films and as compensation-point writing when applied to ferrimagnetic films. In general domains of the order of the diffraction limited diameter of the laser spot, which is about $1\text{ }\mu\text{m}$, can be written, although it is possible to write substantially smaller domains, as will be discussed in Section 5.

The discs employed for MO recording consist of a glass or polycarbonate substrate coated with a photo-polymerized lacquer with pregrooves. On top of this a stack of optical layers containing the MO layer is deposited. Information can be written in a disc and read out with a special recorder using polarizing optics.^[5]

3. Magneto-Optical Materials

In addition to a large Kerr rotation there are several other requirements a material has to satisfy in order to be suitable as a recording medium for thermomagnetic writing with optical readout. These requirements are:

- 1) Perpendicular magnetic anisotropy
- 2) High coercivity at room temperature
- 3) High magneto-optical figure of merit, $R\theta_K^2$, where θ_K is the Kerr rotation and R the reflectivity of the MO film
- 4) Switching characteristics matched to the available laser power ($T_c < 600\text{ K}$) and the available magnetic field strength ($< 50\text{ kA m}^{-1}$)
- 5) Low media and write noise
- 6) Low deposition temperatures and high deposition rates
- 7) Long-term stability

The classes of materials which enable all these requirements to be fulfilled at the same time, are limited. The best candidates at present are the ferrimagnetic amorphous RE-TM alloys. Several compositions, mostly containing Gd and/or Tb as the RE atom and Fe and/or Co as the TM atom, were suggested and have been studied in the past. Among these, GdTFe and TbFeCo are probably the most suitable materials. In the remaining part of this section we

will discuss how the requirements 1–7 can be fulfilled in the system $(\text{Gd,Tb,Dy})_x(\text{Fe,Co})_{100-x}$ with $(15 < x < 30)$.

1) Perpendicular anisotropy can be achieved by proper deposition conditions and composition. The origin of the perpendicular anisotropy in amorphous alloys is an anisotropic short-range ordering process during deposition. Mechanisms of magnetic anisotropy are magnetostriction, bond orientational anisotropy and single-ion (spin-orbit coupling) anisotropy.^[6] Typical values for the anisotropy constant involved are $K_u = 10^4 \text{ J m}^{-3}$ for Gd based alloys and $K_u = 10^5 \text{ J m}^{-3}$ for Tb and Dy based alloys.

2) The absolute value of the magnetization of $\text{Gd}_{24}\text{Tb}_{1}\text{Fe}_{75}$ is shown in Figure 4 as a representative example. The RE and TM sublattices have opposite magnetization directions and the temperature dependence of the "sublattice" magnetizations differs as well. Because of this the resulting total magnetization becomes zero at a temperature which is called the compensation temperature, T_{comp} . At this temperature the coercive field, which is about proportional to $1/M_s$, diverges. The Kerr rotation is caused chiefly by the Fe sublattice and has a non-zero value at T_{comp} . By adjusting the position of T_{comp} a few tens of degrees below or above T_{ambient} , which can be achieved by choosing the proper composition, very high coercivity values, implying high domain stability, can be achieved.

3) The Kerr rotation for the RE-TM is only moderate, with typical values of $0.2\text{--}0.3^\circ$. However, the shot noise limited CNR measured during a recording experiment is not proportional to the Kerr rotation angle, θ_k , but to the MO figure of merit $R\theta_k^2$. By depositing a suitable optical enhancement layer, it is possible to increase the Kerr rotation at the expense of the reflectivity and thus increase the figure of merit. In this way a satisfactory CNR can be obtained.

4) The power necessary for switching the magnetization is determined both by the thermal properties of the MO layer stack and substrate and by the Curie temperature of the MO layer. As an example, T_c versus composition, x , is shown for the four binary systems GdFe, GdCo, TbFe and TbCo in Figure 5.^[7] It can be seen that by a proper choice of composition, T_c can be adjusted. However, other requirements, such as a proper positioning of T_{comp} , should be achieved as well. Therefore it is necessary to use ternary systems in which T_c is adjustable by the Fe/Co ratio. In practical systems T_c is between 450 K and 500 K and T_{comp} is determined by the RE-TM ratio.

5) As the RE-TM-alloys are amorphous no crystal-grain boundaries are present. Therefore very low media-noise levels are achieved. By choosing proper RE-TM compositions, the write noise caused by imperfectly written domain patterns can be reduced to the level of the media noise.

6) The amorphous RE-TM alloys can conveniently be deposited with vapor deposition and sputtering techniques. In mass production lines, magnetron sputtering is preferred as it enables high deposition rates and does not necessitate substrate cooling.

The structure, and thus the magnetic properties, such as coercivity and anisotropy, as well as the stability against oxidation of the films, is very much dependent on the preparation parameters.^[8] RE-TM films prepared with magnetron sputtering at low argon pressures (0.6 Pa) and short substrate-target distances (60 mm) are very dense and smooth, whereas films prepared at high argon pressures (2.5 Pa) and large substrate-target distances (120 mm) exhibit a porous microstructure with voids and a pronounced columnar structure. The films deposited under the latter conditions are much less stable against oxidation and show reduced anisotropy constants compared to the former. The target technology of RE-TM targets is a current is-

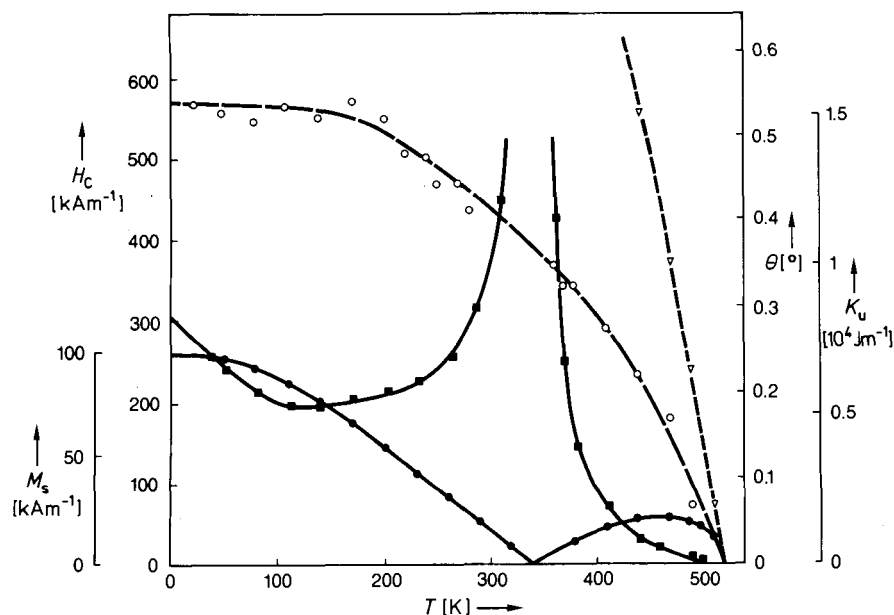


Fig. 4. Magnetic properties of a 50 nm thick $\text{Gd}_{24}\text{Tb}_1\text{Fe}_{75}$ layer versus temperature. The layer was protected with a 30 nm thick aluminum layer. ●: Saturation magnetization M_s , ■: Coercivity H_c , ○: Kerr rotation θ_k and ▽: Anisotropy K_u . The Kerr rotation is enhanced by the Al mirror.

sue. One problem is the brittleness of the RE-TM alloys, another the homogeneity of the deposited films. The magnetic properties of the films are very dependent on the composition, e.g. the compensation temperatures change with 40 K to 100 K per atom percent RE. Therefore MO discs should have a homogeneity better than 0.5 atom percent, which requires homogeneous targets with almost equal distributions of the sputtered atoms. The distribution of the sputtered atoms depends on the preparation technique of the target, e.g. casting of an alloy or hot pressing of powders.

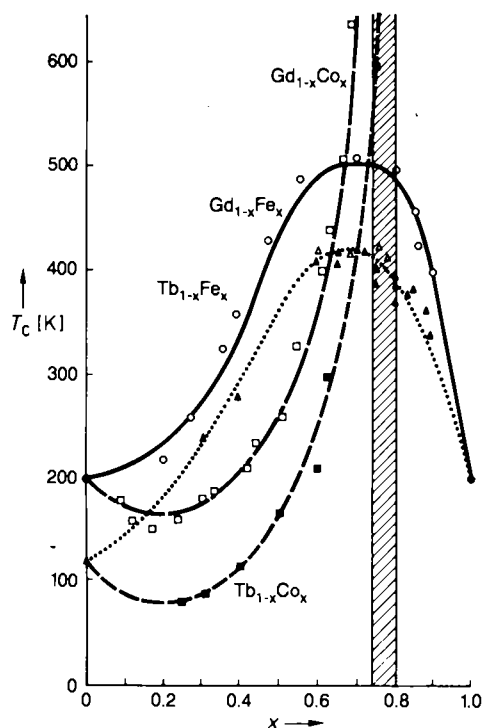


Fig. 5. Curie temperature versus composition for amorphous binary alloys (for references see P. Hansen [7], Fig. 2). The shaded area indicates the range of compensation temperatures in the vicinity of room temperature of these alloys.

7) One of the most serious disadvantages of the amorphous RE-TM alloys is their susceptibility to corrosion and oxidation.^[9] Therefore long-term stability can only be achieved in a suitable multilayer structure in which the MO layer is covered by protection/passivation layers. Such a structure is shown in the inset of Figure 6. From the substrate side the MO layer is protected by means of a dielectric layer, which can be e.g. AlN or Si₃N₄. Apart from acting as a protection layer, this layer also serves as the optical enhancement layer discussed under 3). On the back side the MO layer is protected by means of a metallic layer, e.g. Al, which also serves as a reflector. Aging experiments on such a structure are also shown in Figure 6. In general several mechanisms contribute to the degradation of MO-layers: oxidation, corrosion, structural relaxa-

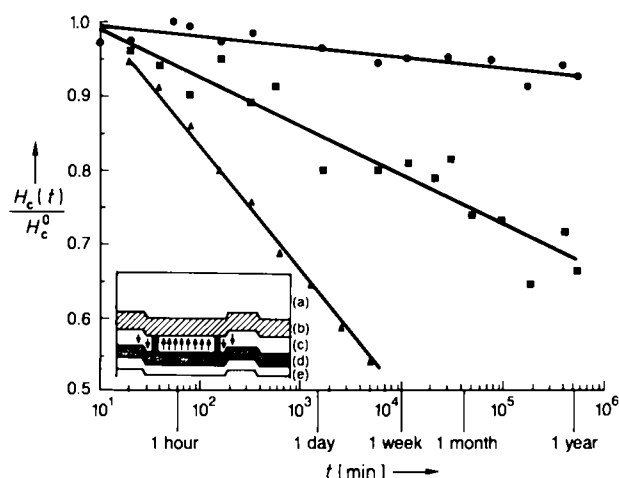


Fig. 6. Long-term stability of a GdTbFe MO layer. Measurements were performed at 80°C. ●: Structural relaxation, ▲: Oxidation of bare film, ■: Trilayer in dry atmosphere. The inset shows a typical trilayer stack. (a) substrate, (b) dielectric enhancement layer, (c) MO layer, (d) metal reflector, (e) organic protection layer.

tion and crystallization. Degradation affects the following magnetic properties: magnetic anisotropy, saturation magnetization, coercive field, and compensation temperature.

4. Thermomagnetic Writing

It is of vital importance to have insight into the physics of the thermomagnetic writing process in amorphous RE-TM layers, since irregularities in the written domains cause the so-called write noise, which is often the main noise contribution in practice. For this reason, several recent theoretical papers dealing with the thermomagnetic writing process have appeared.^[10-12] Several models have been worked out, starting from computer simulations,^[10] or applying a "bubble-like" phenomenological description.^[11, 12] In the latter case the stability of the domain is given by the following equation:

$$F = \frac{\partial}{\partial r} (E_w + E_H + E_D) = F_w + F_H + F_D,$$

where E_w is the domain-wall energy, E_H the magnetic-field energy, E_D the demagnetizing energy, F_w , F_H and F_D the corresponding forces and r the domain radius. The resulting domain diameter can now be calculated by equating the total force, as given above, to the coercive force F_c . Of course, the proper temperature dependences of the magnetic properties, at the end of the laser pulse, have to be taken into account in the calculation.

Experimental observations of the dynamics of domain formation processes have been scarce hitherto.^[13] However, very advanced techniques to study laser-written domains, using Lorentz microscopy, have recently become

available.^[14, 15] An example is shown in Figure 7. In this case the MO layer was deposited on a specially prepared silicon wafer disc, with 100 μm by 100 μm windows with 10 nm thick Si_3N_4 membranes. On top of this Si_3N_4 layer the MO layer was deposited and protected with a thin Al/SiO_2 layer. On this MO disc domains were written with a laser beam under various conditions. After recording, the silicon wafer was cut into pieces each containing a single window. These specimens were investigated in the electron microscope, using a special operation mode, called Lorentz microscopy. In this way it was possible to visualize magnetic contrast and to obtain images of the laser-written domains.

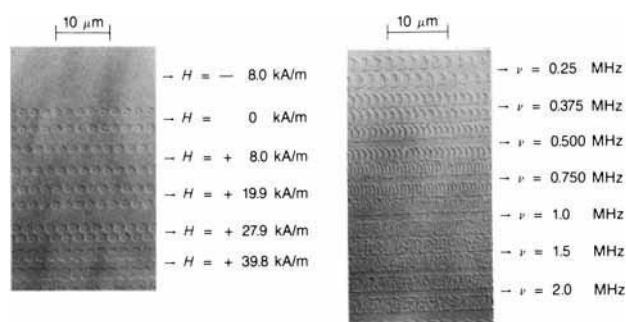


Fig. 7. Lorentz microscopy images of thermomagnetically written domains. Left: Dual tracks, written with laser modulation for various applied fields. Right: Dual tracks, written with magnetic-field modulation for various frequencies.

5. Direct Overwrite

The usual way to write data on an MO disc is to heat the MO layer locally in a fixed applied field. The information to be recorded is supplied by modulating the laser beam. The main drawback of this method is that information already recorded cannot be overwritten. Whole sectors or tracks have to be erased before new information can be recorded. Several solutions to this problem have been suggested. Shieh et al.^[16] suggested a method for a direct-overwrite technique, utilizing the demagnetizing field of the MO layer itself. The use of exchange-coupled magnetic double layers may also offer interesting possibilities in this respect.^[17] The most obvious and direct way to solve the direct-overwrite problem is to modulate the applied field.^[18] Using this method, the MO layer is heated continuously by the laser, while the applied magnetic field is switched with a high frequency and modulated according to the information to be recorded. In this way old information can be overwritten, just as in conventional magnetic recording.

By switching the applied field while rotating the disc, overlapping domains of different polarity are written, leaving only a crescent. Because the dimensions of this crescent in the tangential track direction are only determined

by the disc velocity and switching frequency of the applied field, domains much smaller than the laser spot can be written. In Figure 7 it can be seen that it is feasible to write domains as small as 0.25 μm in the writing direction in a $\text{Gd}_{15}\text{Tb}_{6.5}\text{Fe}_{78.5}$ layer. Apart from the direct overwrite feature and the possibility of obtaining very high recording densities, magnetic-field modulation offers two other advantages in MO recording. First the quality of the coded signals is improved, as the laser is on continuously and consequently there is no influence of a thermal history when writing a domain. Secondly, influences of compositional variations over the disc surface on the length of written domains, which is apparent in the case of laser modulation, are less critical when writing with magnetic-field modulation. This is due to the fact that the domain length in this case is only determined by the switching frequency of the applied field and the disc velocity.

The use of magnetic-field-modulation methods in MO recording requires the use of special magnetic heads which are capable of generating relatively high fields at large distances, which can be switched at high frequencies. It has recently been shown^[19] that it is feasible to design heads which meet all the requirements.

6. Future Materials

Although it is possible to employ amorphous RE-TM compounds for MO recording there is a continuous search for materials with improved properties. The focus is mainly on an increased MO figure of merit and improved chemical stability. Another important development is the future availability of compact solid-state lasers operating at shorter wavelengths than 800 nm. A continuous shift towards blue wavelengths is expected in the future.

Giant MO effects have recently been found in uranium compounds at low temperatures.^[20] It has not yet been possible to find room temperature ferromagnets with such high figures of merit, albeit that materials with higher Kerr rotations than the RE-TM materials are known. The material with the highest Kerr rotation at room temperature known today is the Heusler alloy PtMnSb discovered at our laboratories.^[21] Despite several attempts, it has not yet been possible to induce perpendicular anisotropy in this material, which is essential for its applications. In PtMnSb the high Kerr rotation value is found at 720 nm. This could become a disadvantage in the future when recording at shorter wavelengths is expected.

Magnetic oxides, such as ferrites and garnets, show unsurpassed chemical stability. This can be combined with high MO figures of merit as has been shown e.g. by the work of Martens et al.^[22] on Co-ferrite. One of the problems with the oxides is the presence of recording noise. CNR values comparable to those obtained for the RE-TM alloys have not been reported yet. Another drawback for the application of magnetic oxides is the necessity of using

high temperature deposition techniques. Nevertheless research is being continued at several laboratories and progress may be expected.

Very interesting MO effects have been found in metallic, artificially layered structures.^[23,24] This is a new field but the possibility for "atomic engineering" in these materials may provide yet unknown and unexpected phenomena.

7. Summary and Outlook

In this article we have discussed materials for MO recording and their application in rewritable recording systems. After a long period of intensive research the RE-TM alloys can be adjusted to satisfy all the requirements for suitable MO materials. This has been made possible by new insights in the physics of the recording processes, combined with advanced preparation techniques. The requirement of direct overwrite, which is expected to be essential for practical applications, will have an impact on further developments in the RE-TM alloy system. Lorentz microscopy pictures show that extremely small domains can be written in a controlled way in these materials allowing for very high recording densities. Unfortunately the resolution during readout is limited by the spatial extent of the laser spot which is about 1 μm at the present wavelength of 800 nm.

The main disadvantage of the present materials is their moderate MO performance and chemical instability. Although some practical solutions to these problems have been found, these drawbacks are the driving force in the search for completely new classes of materials. Magnetic oxides and metallic multilayers offer some interesting

prospects here. All these efforts are worth pursuing, because MO recording may develop as the future recording technology, combining the advantages of both magnetic and optical recording.

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Research News

A Novel Monolithic Thin-Film Electroluminescent Device with Extrinsic Memory

Flat display panels are a part of many portable consumer products. Their visual performance is inferior to that of cathodic ray tubes: the luminance of liquid crystal displays (LCDs), the leading flat display technology, is very low, and, in the conventional line-by-line addressing mode, their contrast decreases with increasing panel size. Electroluminescent (EL) display devices are an interesting alternative to LCDs. Figure 1 shows the structure of a

standard thin-film(TF) EL device. The active ZnS-Mn layer is sandwiched between two dielectric Y_2O_3 layers.^[1,2] Electroluminescence is excited by an alternating field of some 10^6 V cm^{-1} , applied between the metallic and the transparent electrode.

In principle the visual performance of emissive display panels is independent of screen complexity and addressing mode. In practice, the average luminance decreases with