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Using non-linear magneto-optic techniques to characterise particulate recording media

D.M. NEWMAN[1,✉]
M.L. WEARS[1]
R. ATKINSON[2]
D. MCHUGH[2]

[1] Centre for Data Storage Materials, Coventry University, Coventry, CV1 5FB, UK
[2] School of Mathematics and Physics, The Queens University of Belfast, Belfast, BT7 1NN, UK

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ABSTRACT The concept of using comparison between magneto-optic measurements taken in the linear and second-harmonic reflected fields to address problems attending the development of ultra-high-density recording media as the limits of thermal stability are approached is presented. It is shown how magneto-optical measurements made routinely in the linear reflected field to access many of the bulk characteristics of such media may, by operating in the second-harmonic field, be fruitfully extended. To illustrate the techniques being explored, reference is made to initial measurements on a longitudinal recording medium under continuing development at Coventry. This medium, already shown to support recording at linear densities in excess of 100 kfci, is based on cobalt particles around 10 nm in diameter.

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1 Introduction

Surface or interfacial magnetism remains a topic of vital current interest, since the phenomenon whereby the surface and the bulk of the same material exhibit very different magnetic behaviour impacts crucially on the development of many artificial magnetic materials at the forefront of technological application. The recording industry, for instance, is striving to develop conventional longitudinal recording media capable of supporting storage densities that approach the theoretical limit. This is set by the onset of superparamagnetic behaviour as the particle size is reduced. An ideal medium might be constructed [1–3] from an array of magnetic nanoparticles 4 nm to 10 nm in diameter, depending on the material, dispersed in a suitable non-magnetic host. At this scale the surface to volume ratio of the particles is extreme. Experimental determination of the respective surface and bulk contributions to the overall magnetic behaviour of the dispersed system is clearly problematic but required to optimise media performance. The noise in granular media has for example been shown to arise predominantly from mutual interactions between grains or particles. Few analytic techniques exist that are appropriate to studying problems in surface magnetism, particularly practical problems associated with the development of recording media. Fortunately most materials of interest to the recording industry are centro-symmetric and produce a second-harmonic (SH) optical response only from their interfaces. In principle, therefore, optical and magneto-optical measurements made in the SH reflected fields can provide powerful surface-sensitive probes as the industry seeks to address problems of thermal stability in the wide variety of nano-scale media under development. These include multilayer compositionally modulated, patterned and particulate materials. In adopting this approach we are however cautioned by reports [4] that attribute enhancement of non-linear magneto-optic effects in granular Cu–Co films in part to a bulk contribution to the susceptibility arising from a lack of inversion symmetry within the cobalt nano-crystals.

Magneto-optical measurements made in the linear reflected fields have long been used to access many of the bulk characteristics of magnetic media such as anisotropy constants, blocking temperatures and the nature and degree of interparticle interactions. In this report we explore, using the particulate cobalt media developed in [3], how non-linear magneto-optic techniques may be employed in conjunction and comparison with more conventional measurements to address some of the problems raised above. Initial attempts to extend such measurements into the SH field are discussed.

2 The medium and its characterisation using conventional techniques

Preparation of the medium used in this study has been described in full previously [3]. Figure 1 shows a simplified view of the production process and allows the nature of the resulting medium to be appreciated. As shown, the thermodynamics of the in-vacua rapid thermal dissociation process favours the condensation of a near-mono-dispersed array of cobalt particles on to the glass substrate. This is studied and characterised as formed, but prior to recording it is necessarily protected with an overcoat of silicon nitride.

Magnetisation measurements taken conventionally using a vibrating sample magnetometer (VSM) yield values of the saturation magnetisation (I_s) in the range of 20% to 60% of that of bulk cobalt depending on the thickness of the precursor film. As a recording medium it has a moment–thickness product ($I_R \cdot t$) between 0.15 memu cm^{-2} 0.5 memu cm^{-2} and

✉ Fax: +44-24/7688-8372, E-mail: apx091@coventry.ac.uk

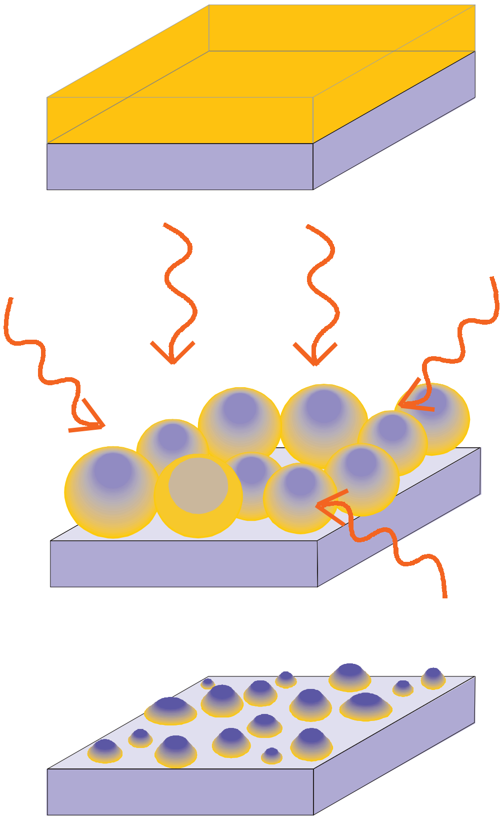

FIGURE 1 Production process of recording medium studied. Precursor film of cobalt oxide (*top*), rapid thermal dissociation of which (*centre*) produces a monolayer of near-mono-disperse cobalt particles (*bottom*)

supports linear recording at densities in excess of 100 kfci (Fig. 2).

Measurement of the temperature dependence of the medium's coercivity using the longitudinal Kerr effect (in the linear optical field) to access the magnetic properties of the samples in a vacuum cryostat allows determination of their blocking temperature (T_B) and hence the particles' magnetic volume [5]. This is found to correlate with the existence of spherical particles around 9 nm to 10 nm in diameter or a somewhat slightly larger diameter if, as is likely, they are actually nearer hemispherical in shape. Magneto-optic techniques are further used in conjunction with conventional magnetic measurements to obtain both isothermal remanent magnetisation I_R(H) and dc demagnetisation I_D(H) curves for the medium. Analysis of this data is used to obtain the nature and degree of interparticle interaction and to provide, by way of the switching-field distribution, an indication of the dispersion of particle sizes.

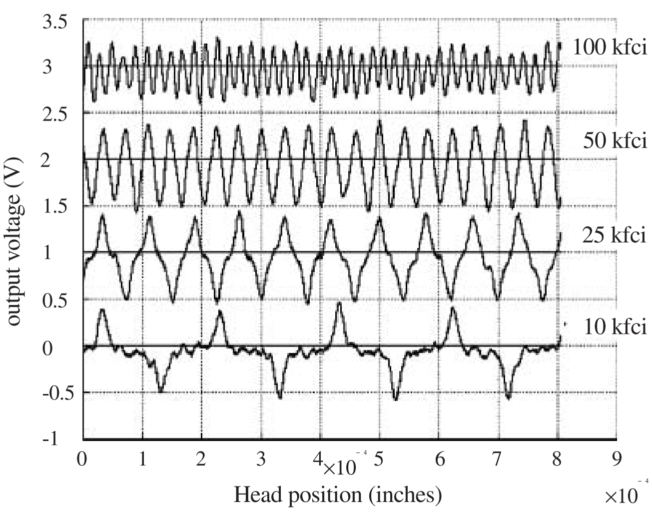

FIGURE 2 Low-velocity contact recording. (Courtesy of Manchester University)

3 Non-linear characterisation

Initial attempts to augment characterisation of this medium using non-linear optical and magneto-optical measurements are shown in Figs. 3 and 4. These measurements were taken using a Ti-sapphire ($\lambda = 800$ nm) laser

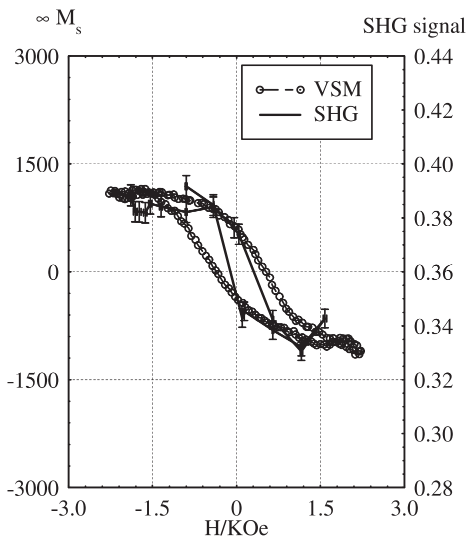

FIGURE 3 Hysteresis loop obtained in the transverse Kerr configuration using second-harmonic radiation compared with that obtained using conventional magnetic instrumentation (VSM)

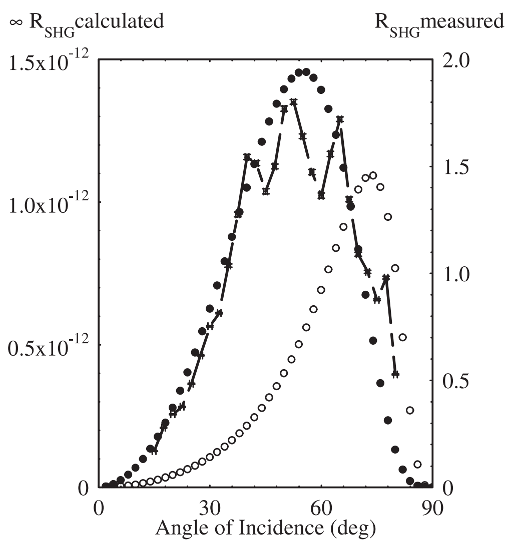

FIGURE 4 Reflected second-harmonic intensity as a function of angle of incidence: – – – experimental data; ●●●● and ○○○○ calculated (see text)

running at a repetition rate of 250 kHz and focussed to a 100-μm spot at the medium surface. The incident beam was *p*-polarised in both cases.

Figure 3 shows the hysteresis loop of the medium obtained when the transverse Kerr magneto-optic effect recorded in the reflected SH field is used to sense the magnetisation-reversal process It is compared with the loop obtained conventionally on a VSM.

Figure 4 shows the angular spectrum of SH radiation ($\lambda = 400$ nm) reflected specularly from this medium.

4 Discussion

The sum total of measurements on the medium under discussion prior to the non-linear studies indicates that it must be considered as a single layer of cobalt particles on a glass substrate with a mean interparticle separation of about twice their diameter. It is likely given the nature of the medium and the way it is produced that the particles' surfaces are oxidised to at least monolayer thickness. This was observed in previous work on particulate cobalt systems [6], when it was found necessary to assume the presence of an oxide monolayer in order to reconcile experimental optical measurements with theoretical effective-medium models. In the present case the differences in the loops recorded by the two techniques (Fig. 3) again provides indirect evidence for the presence of an oxide shell. Cobalt oxide is antiferromagnetic with a Neel temperature of about 20 °C, so that for both the measurements in Fig. 3 it would be very weakly antiferromagnetic. Similar behaviour from exchange-coupled ferromagnetic–antiferromagnetic interfaces has been reported recently by Banshchikov et al. [7], who also make instructive comparison to the much earlier work of Meiklejohn and Bean [8].

The loop obtained on the VSM represents the sum-total response of the predominantly cobalt medium. Conversely, that taken using the magneto-optic signal in the SH reflected field arises only from the interface between the cobalt core and its oxide shell and the oxide-free surface, with the dominant contribution being determined in part by the oxide thickness. Irrespective of any 'surface-magnetism' effect at the surface of the core, the results seen can be broadly understood if it is assumed that the weakly antiferromagnetic oxide layer is being 'driven' through exchange coupling to the cobalt cores' interface. Clearly, it would be instructive to repeat these measurements as a function of temperature when the behaviour of the oxide contribution would vary and should vanish at temperatures above 20 °C. Moreover, the presence and properties of the oxide layer must impact on the nature and degree of interparticle interactions that critically influence recording performance. Ideally, the dispersion of the particles should be such that interparticle interactions are minimised. As indicated above the $I_{\mathrm{R}}(\mathrm{H})$ and $I_{\mathrm{D}}(\mathrm{H})$ curves for the present samples have been measured using magneto-optic measurements in the linear reflected field as part of the basic characterisation process. Analysis of this data indicates that they consist of weakly interacting particles. (Note: The samples used for these initial non-linear studies were not optimised for recording but selected instead to be reversible in the fields available on that experiment.) The results of Fig. 3 and the inference that can already be drawn from them demonstrate that it would be equally worthwhile to also determine $I_{\mathrm{R}}(\mathrm{H})$ and $I_{\mathrm{D}}(\mathrm{H})$ curves using the non-linear magneto-optic response. Such experiments would however be extremely demanding of laser stability.

Consideration is now given to the results shown in Fig. 4. The experimental SH reflectance curve (– – –) is typical of the sample, although different areas do not show all the features so well-pronounced or they are slightly displaced. The regularity of the features observed on what would otherwise be a smooth curve is strongly indicative of their origin being a consequence of the medium structure. The two theoretical curves are calculated taking the free-electron model of Jha [9] as a starting point. The curve (○○○○) is for a continuous cobalt surface, whilst that designated (●●●●) is calculated with Jha's expressions modified by incorporating effective-medium techniques to allow for the structure, particle size, dispersion and oxide shell. This simple model however contains no mechanism that might describe the features observed experimentally, and unfortunately its sensitivity to input variables is too ambiguous to determine with any confidence whether or not they represent peaks or troughs on the curve. The consensus is that they are troughs. In similar measurements [10] on nickel gratings we observed both peaks and troughs and were able to successfully correlate all features with plasmon-resonance phenomena at both the fundamental and harmonic wavelengths. The present structure is more complex and no comparable features are observed in the angular spectrum of the linearly reflected field.

The problem of enhancement and scattering of SH radiation, including the role of various possible surface excitations in these processes, has been the subject of considerable interest for some while [11–16]. However, with the exception of the work [16] on yttrium iron garnet particles dispersed in organic layers, the rough surfaces considered in most other cases are of a continuous conducting nature. The results presented in Fig. 4 are, by contrast, for a smooth continuous dielectric (glass) surface bestrewn with metallic particles surrounded (probably) by an oxide shell. If excitations are stimulated on such a surface they will probably be localised in the vicinity of the particles. Enhanced production of second-harmonic radiation localised in the vicinity of defects has been observed by Zayats et al. [15] using near-field techniques. The mechanisms of localised enhancement have been studied by Boyd et al. [12] using a model that approximates a rough surface by a set of non-interacting hemispheroids randomly distributed in size and shape. They point out that any given wavelength will resonate with some surface features and that many different features can be near resonance. Unfortunately, consideration of this model leads only to the expectation of peaks on the smooth curve of second-harmonic production, not troughs as we believe we observe.

A mechanism is required that either resonantly re-absorbs SH radiation as found in [10] or which at certain angles of incidence diverts energy from the fundamental beam that would otherwise have gone to produce SH radiation. O'Donnell et al. [13] have studied the diffuse scattering of SH radiation from a fully characterised rough surface and observe strong resonant enhancement in both the forward- and back-scattered radiation as a consequence of plasmon excitation. This may offer an explanation of the results we observe, since the process will be competing with the generation of SH radiation in the specularly reflected beam that is all we currently record. Moreover the surfaces we study possess a structure that extends in both in-plane dimensions, whilst the surfaces studied by O'Donnell were fabricated specifically to exhibit only one-dimensional roughness in order that they might more closely approximate the structures widely assumed in theoretical calculations. The structure of the media considered here therefore supports the possibility of scattering processes at angles other than in the plane of incidence and this may account for the multiple features we observe. An alternative but more speculative possibility is that the troughs are a consequence of some resonant third-order process such as stimulated Raman scattering diverting energy from second-harmonic production in the specularly reflected beam. Positive identification of the mechanisms involved will eventually provide structural information, but further detailed measurements are required. It would be particularly interesting to study the magneto-optic response across the features in the SH reflectance spectrum.

5 Conclusion

Initial attempts to characterise real recording media using non-linear optical measurements have demonstrated that further development of these techniques is worthwhile.

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