Photothermal investigation of local and depth dependent magnetic properties

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Abstract: To achieve a spatially resolved measurement of magnetic properties two different photothermal approaches are used which rely on heat dissipated by magnetic resonance absorption or thermal modulation of the magnetic properties, respectively. The heat produced by modulated microwave absorption is detected by the classical photothermal methods such as photoacoustic effect and mirage effect. Examples comprise depth resolution of the magnetization of layered tapes and visualisation of magnetic excitations in ferrites. The second photothermal technique relies on the local modulation of magnetic properties by a thermal wave generated with an intensity modulated laser beam incident on the sample. This technique has a higher spatial resolution and sensitivity and has been used to characterize lateral magnetic properties of multilayers and spintronic media. To extend the lateral resolution of the ferromagnetic resonance detection into the nm-range techniques have been developed which are based on the detection of the modulated thermal microwave response by the thermal probe of an atomic force microscope (AFM) or by detection the thermal expansion of the magnetic sample in the course of the resonant microwave absorption with an AFM or tunnelling microscope. These thermal near field based techniques in ferromagnetic resonance have been successfully applied to image magnetic inhomogeneities around nano-structures and to measure the ferromagnetic resonance from magnetic nano-dots.

1. Introduction

Photothermal methods have proven to be very suited to investigate spatial variation of physical properties due to the evanescent behaviour of thermal waves induced by periodic heating of the sample. To achieve a spatially resolved measurement of magnetic properties two different photothermal approaches had been developed and successfully applied in the last two decades. One technique makes use of the heat dissipated in the material in the course of the absorption of microwaves. The second technique relies on the local modulation of the high frequency susceptibility of the magnetic sample by a thermal wave generated with an intensity modulated laser beam incident on the sample.

The detection of the thermal response of ferromagnetic resonance of Fe and Ni foils was first reported by two Brasilian groups [1,2]. As compared to bolometric detection of the dc-thermal response of microwave resonance absorption, which had already been observed in a paramagnetic solid one decade earlier [3] the photothermal approach offers the possibility of magnetic depth profiling. In the following the methods for the measurement of the thermal response had been extended comprising techniques such as mirage effect and thermo-reflection. The photothermally modulated resonance technique had been introduced by Orth et al. 1988 [4]. The much larger dynamic range with modulation frequencies ranging up to several MHz
enables depth resolutions down to one micron which is required e.g. to resolve depth dependence of magnetic tracks on magnetic tapes. Nevertheless, the further decrease of the size of magnetic storage devices demanded techniques with resolution on nm-scale which could finally be achieved by the application of scanning near field microscope with temperature sensitive tip [5]. The highest lateral resolution of a few nm was obtained when a scanning tunnelling microscope was used to measure the thermal expansion of the nano-magnets in the course of heating by the ferromagnetic resonance [6].

In this report some major features of the different photothermal detection methods will be summarized and their impact for magnetic characterization will be demonstrated by experimental results. In a first part some typical examples for the local and depth resolution on mm- and µm-scale are described. The second part is devoted to the more actual results obtained with the combination of FMR with scanning thermal near field microscopy.

2. Microwave thermal response

The experimental set-up for the detection of the thermal response is based on a conventional microwave resonance apparatus which is modified to allow the measurement of the temperature rise of sample surface in course of the microwave heating in resonance. For more details see e.g. the review by J. Pelzl and U. Netzelmann [7]. In all experiments described in this report the measurements have been conducted at the fixed microwave frequency of 9.2 GHz (X-band). Different to the conventional field modulation the microwaves are intensity modulated by a pin-diode modulator which is built in behind the microwave source of the X-band spectrometer. The microwave cavity with the sample is connected to this source by a wave guide circulator set-up to avoid microwave losses as much as possible. The resonance condition is achieved by changing the external magnetic field until the uniform precession frequency of the magnetic moments of the sample in the external plus internal magnetic fields is equal to the microwave frequency. The absorption of microwave in resonance leads to a heating of the sample at those positions on the sample which meet the resonance condition. In the first experiments published in the eighties and nineties of the last century the released heat flux or the temperature increase of the sample was detected by the photoacoustic effect and the mirage. For this purpose the cavity inside the magnet was modified.

2.1 Microwave resonance detected by the photoacoustic effect

To measure the photoacoustic response of the heat dissipated after absorption of microwave in resonance essentially two main constructions have been used. In the first approach the sample is placed into a short-ended waveguide, which is connected by an air channel to the microphone [3,7]. The second approach uses the quartz-tube of the conventional EPR-spectrometer. The sample is deposited into this tube and then positioned in the middle of the X-band cavity. To measure the photoacoustic response, the end of the quartz-tube is closed by a microphone [7,8]. This design allows the simultaneous measurement of the conventional signal and of the photoacoustic response.

The photoacoustically detected microwave resonance offers no lateral resolution but by changing the modulation frequency of the microwave input different depths can be distinguished. This is demonstrated impressively by the resolution of the magnetic sandwich structure of a ferrochrome-magnetic tape [9]. The tape consists of Fe₃O₄-layer on the polyester support with a CrO₂ layer on the top. The resonance curves as a function of the external magnetic field are shown at the bottom. The photoacoustic response of the sandwich band clearly reveals the CrO₂ part at high modulation frequencies and the superposition of both magnetic resonances at low modulation frequencies (Fig.1). Al-
though the CrO$_2$ layer was very thin (2.2 µm) it could be resolved at audio frequencies because of the very low thermal diffusivity value of the magnetic tape materials.

2.2 Microwave resonance detected by the mirage effect

The mirage effect offers a lateral resolution of about 50 µm for the measurement of the surface temperature. Fig. 2 shows a laterally resolved FMR spectrum of magnetostatic modes in an Yttrium Iron Garnet (YIG) slab [10]. To achieve access to the sample in the cavity with the probe laser beam the cavity was supplied with two openings attached by chimneys to avoid Microwave losses. The magnetic field was applied perpendicular to laser beam and the perpendicular deflection of the laser beam was measured at each point along z in equally separated steps of 50µm as a function of the magnetic field. The FMR-image clearly reveals the standing magnetostatic modes with heat maxima at the points of excitations. The spatial resolution allows a detailed analysis of the collective magnetic excitations [11]. Also non-linear effects become directly accessibly with a local information. When the microwave power is increased the maximum in the center of the standing wave with the five maxima gains dramatically on intensity which could be identified as a result of a four magnon scattering process [12].

3. Thermal modulation of the microwave resonance

Magnetic properties such as the magnetization and the magnetic anisotropy are temperature dependent. Therefore, heating any position of the magnetic sample e.g. by an intensity modulated laser beam leads to a periodically oscillating magnetic property. As the laser heating is depth dependent via the modulation frequency a depth sensitivity of the magnetic property can be achieved. To measure the thermal modulation of the magnetic property two main approaches had been developed: the photothermally modulated read back and the photothermally modulated ferromagnetic resonance.

3.1 Photothermally modulated read-back

This technique relies on the Faraday’s law of induction. The thermal modulation of the magnitude and of the orientation of the magnetization causes a time dependent magnetic flux within an electromagnetic circuit. Applying this principle to the recorded magnetic pattern on magnetic tapes the recorder head can be used as detector [13,14]. The tape is fixed on the magnetic head and the magnetic tape is heated with a intensity modulated laser beam focussed on the tape in the head gap area. The voltage induced in the head is detected synchronously with the modulation frequency either as a function of the modulation frequency or as a function of the laser spot on the tape. Experiments were performed on ferrochrome tapes which were homogeneously magnetized or a sound track had been recorded with same head on the tape. Although the thermal wave amplitude decreases with the inverse power of the modulation frequency the signal of the photothermally modulated read-back increased by two orders of magnitude in the frequency range from 10 Hz to 100 kHz [13]. This frequency dependence is a result of the competition of the law of induction and the thermal wave behaviour offering a means for high spatial resolution studies with this technique.

3.2 Photothermally modulated ferromagnetic resonance (PM-FMR)

Among the photothermal microwave techniques the photothermally modulated ferromagnetic resonance (PM-FMR) is of outstanding importance because of its high spatial resolution (µm-scale) and its sensitiv-
ity, which is comparable or sometimes higher than that of the conventional detection. The sample is exposed to a continuous microwave and an intensity modulated laser beam, which is focussed to a small spot on the sample surface through a hole in the cavity. The PM-FMR signal is given by the modulated part of the conventional FMR diode signal of the spectrometer [7,15].

Laterally resolved magnetic images can be obtained by recording the PM-FMR signal at constant modulation frequency and constant external field as a function of the light beam position on the sample. The first PM-FMR image was obtained from a magnetic trace recorded on a CrO₂ magnetic tape [15]. Subsequent interesting applications comprise the imaging of magnetostatic modes on YIG spheres [10,16], laterally resolved observation of the collapse of the magnetostatic modes in a YIG slab near the ferri-magnetic phase transition [17] and thickness dependence of the magnetic anisotropy of thin iron films [18]. To measure the thickness dependence for the same preparation conditions a wedge sample of epitaxial iron was prepared and the PM-FMR was measured at different positions of the wedge. Fig. 3 shows the variation of the two resonance fields of iron as a function of the thickness of the iron layer at three different temperatures. 180 K (rhomb), 300 K (circles) and 380 K (squares). The Ag-coated epitaxial iron film, which was grown on a Ag buffer film with a thickness varying between 2.5 nm and 12 nm, displays thickness dependent resonance fields below 7.5 nm thickness which could be accounted for an growth induced uniaxial anisotropy [17,19,20].

More recently PM-FMR has been used to control the lateral variation of the anisotropy of magnetic films in spintronic devices such as Fe-films on HEMT structures of GaAs [21].

4. Thermal scanning near field techniques in ferromagnetic resonance

The more and more decreasing sizes of magnetic storage media require spatial resolution in the nm-range. The PM-FMR - although very successful in imaging small magnetic structures - is limited in resolution by the diffraction limit of the heating laser light. To extend the resolution limit thermal near field techniques have been combined with ferromagnetic resonance. Fig. 4 shows the principle set-up of these approaches.

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**Fig. 3.** Resonance fields of a iron wedge as a function of the film thickness at three different temperatures.

**Fig. 4:** Set-up of the ferromagnetic resonance detection by a scanning thermal microscope (SThM-FMR) and by the thermo-elastic response with a scanning tunneling microscope (SThEM-FMR).
The sample is positioned in a cavity and as in the methods described in chapter 2 the sample is heated periodically by the intensity modulated microwave. The thermal response of the microwave heating is detected locally by temperature sensitive tip of a atomic force microscope (SThM-FMR, left in Fig. 4) or the local perpendicular thermal expansion of the sample surface is measured by scanning tunnelling microscope (SThEM-FMR, right in Fig. 4) [6].

In both experiments the sample is placed at the upper wall of the cavity and the probes are in contact with the sample surface through a hole drawn into the upper wall. First SThM-FMR measurements have been conducted on a epitaxial iron film which exhibited lateral inhomogeneities due to a partial oxidation of iron [5]. Different types of oxidation could be distinguished. The lateral resolution achievable with the thermally modulated ferromagnetic resonance measurements was about 200 nm for the investigated iron film. In the near-field limit of thermal microscopy the spatial resolution is independent of the modulation frequency and solely determined by the size of the tip (see chapter 2 of ref. 22). For this reason a modulation frequency in a rather low frequency range between 1 kHz to 10 kHz could be used. In subsequent studies the local resonances of permalloy stripes have been investigated [22,23]. Finite size effects in magnetic resonance have been observed in Co-stripes [22,24]. Fig. 5 shows the amplitude and phase images of the SThM-FMR signal from the edge of the stripes which had the sizes 100µm x 2.5 µm x 0.01µm. The scanned area shows a small cross section of the stripe. The edges are excited more the the middle. The phase image indicates that the whole cross section acts as heat source as expected for backward volume modes with small k-values.

Higher lateral resolutions down to 10 nm could be achieved with the SThEM-FMR technique, which had been applied to single Ni-dots [22, 6]. The sample was an electrolytically grown Ni-film on (111) Au on a mica substrate. The shape distribution of the dots has a maximum at a height of 260 nm and at a diameter of 150 nm (Fig.6 left). The experimental set-up was composed of a microwave synthesizer in combination with a TWT-microwave amplifier capable of 35 W modulated microwave output. To measure the FMR of a single Ni-dot an STM probe was positioned on the top of the specific dot. Passing through the resonance by changing the external field the control voltage of the z-piezo was varied to maintain the tunnelling current constant. The variation of the control voltage as a function of the magnetic field is a measure of the thermo-elastic expansion of the dot due the heating of the dissipated microwave power in resonance. Figure 6 shows on the right the SThEM-FMR spectra from two different dots. The left line spectrum is obtained from a broad Ni-dot of 100 nm height and 150 nm diameter. The right line spectrum results from a column like dot 180 nm tall with a diameter of about 70 nm. This SThEM-FMR spectrum is shifted about 30 mT to higher resonance field compared to the full line spectrum.
This shift is due to the change in shape anisotropy from ellipsoid to cylindrical approximation. Both spectra show a comparable line shape. The sharpness of the lines is surprising but still slightly larger as the measured intrinsic damping of the sample. Both resonance line positions are well included in the conventionally measured integral FMR linewidth of about 80 mT.

5. Conclusion

Photothermal detection offers depth and lateral resolution of magnetic properties. Two approaches for photothermal detection of magnetic properties have been applied: Measurement of heat dissipation during resonance absorption of microwaves by magnetic moments using photoacoustic and mirage effect and photothermal modulation by a focused laser beam of magnetic properties such as the magnetization and anisotropy field. The response of the thermal wave impact is detected by conventional microwave diodes (PM-FMR) or by a recording head (PM-Readback). The depth and lateral resolution of all mentioned methods is restricted by the thermal wave or optical limit to the µm-range. Direct measurement of dissipated heat or of thermal expansion in the course of the microwave resonance absorption offers local resolution of 10 nm – 100 nm.

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