


<p style="text-align: center;"><b>Deliverable Report</b>  <b>Author: Nilesh Awari, Michael Gensch</b>  <b>Date : 30.09.2017</b></p>						
Deliverable	Deliverable name (Short name)	WP No.	Lead participant	Type	Diss. Level	Date
D3.2	Field dispersion of resonance > 500 GHz	3	HZDR	Report	Public	M9

<b>Progress beyond state-of-the-art:</b>	
<b>Field dispersion of resonance in Mn<sub>3-x</sub>Ga Heusler alloys</b>	
Partner	Contribution
TCD	Film optimisation and growth; Structural characterisation; Magnetisation measurements; Hall measurements
HZDR	Characterisation of the field dispersion relation in THz emission experiments in the new magnetization dynamics endstation at the TELBE laboratory.

In order to achieve the end goal of the project the initial proof that a ferrimagnetic film can emit THz radiation at distance must be proven. For this, several films based on the tetragonally distorted Mn<sub>3-x</sub>Ga family were to be grown by sputtering at TCD. In a second step HZDR is supposed to characterize these films by means of laser-driven THz emission and characterize the dispersion of the magnetic resonances in external magnetic fields. The initial THz emission spectroscopy measurements performed within a collaboration of TCD and HZDR and published as N. Awari et al in 2016 [1] only allowed to employ out-of-plane fields up to 400 mT. Hence, an upgrade of the experimental set-up for THz emission spectroscopy in the TELBE laboratory was successfully performed which involved the implementation of a 10 T split-coil magnet. Geometrical constraints of the split-coil magnet such as small size optical viewports and comparatively large distances between these viewports and the sample lead to considerable transport losses of the emitted THz pulses, reducing the achievable sensitivity in these measurements. The developed new end station (see figure 1) is unique and the performed measurements constitute the world's first observation of narrow-band spintronic THz emission in magnetic fields up to 10 T, to our knowledge. A publication of the results is underway [5].

### Sample growth

A series of Mn<sub>3-x</sub>Ga on 10 x 10 mm<sup>2</sup> double-side polished MgO single crystal substrates with 0 < x < 1 were grown by TCD partner. The growth conditions had previously been determined [2-4]. Supporting measurements were done prior to shipping include X-ray diffraction and reflectivity as well as SQUID magnetometry. These samples were sent to partner HZDR for characterization by transient THz emission spectroscopy. Magnetometry measurements in different geometries were also carried out at HZDR.

### Characterisation

HZDR characterized a new series of films from TCD in 2017 and observed that none of these films exhibited THz emission of similar strength as from the original sample series for up to now unclear reasons. For that reason, HZDR decided to utilize the original sample series from 2016 to perform the field dispersion measurements.

After characterization in moderate magnetic fields of up to 400 mT three samples of different stoichiometry were identified, that exhibited strong THz emission at different THz frequencies (these results have been published [1] and were also shown in deliverable 3.1.). As was shown, the films stoichiometry induces a change in anisotropy and thus allows shifting the THz emission between 0.21 THz ( $\text{Mn}_2\text{Ga}$ ) and 0.35 THz ( $\text{Mn}_3\text{Ga}$ ). These same films were then introduced into the new end station and the field dispersion was measured. As described above, the sensitivity of the THz emission spectroscopy in the split-coil magnet is reduced. Likely for this reason, THz emission from the  $\text{Mn}_3\text{Ga}$  sample, which has earlier been determined to be the weakest of the three samples, could not be observed. The experimental study was hence performed on the  $\text{Mn}_2\text{Ga}$  and  $\text{Mn}_{2.5}\text{Ga}$  only.

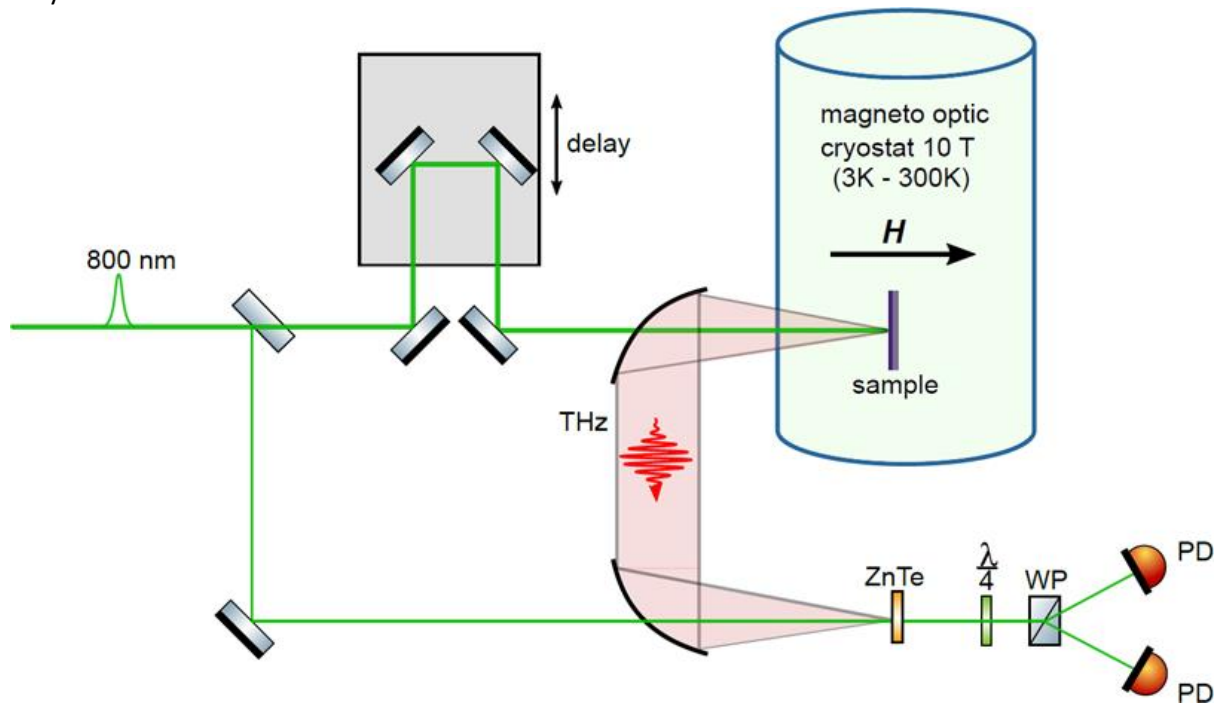


Figure 1: Upgraded experimental set-up to allow for sensitive measurements of THz emission spectra in high magnetic fields and at low temperatures.

Figure 2 shows the derived results. As the out of plane field increases the resonant frequency increases linearly with the slope of 27.8 GHz/T and exceeds 0.5 THz for  $\text{Mn}_{2.5}\text{Ga}$  values thereby achieving the goal of this deliverable. The linear slope corresponds to what is expected from a material with uniaxial anisotropy with the external field applied along the easy axis [5], and shows that we are clearly probing the low-frequency ferromagnetic-like mode [6]."

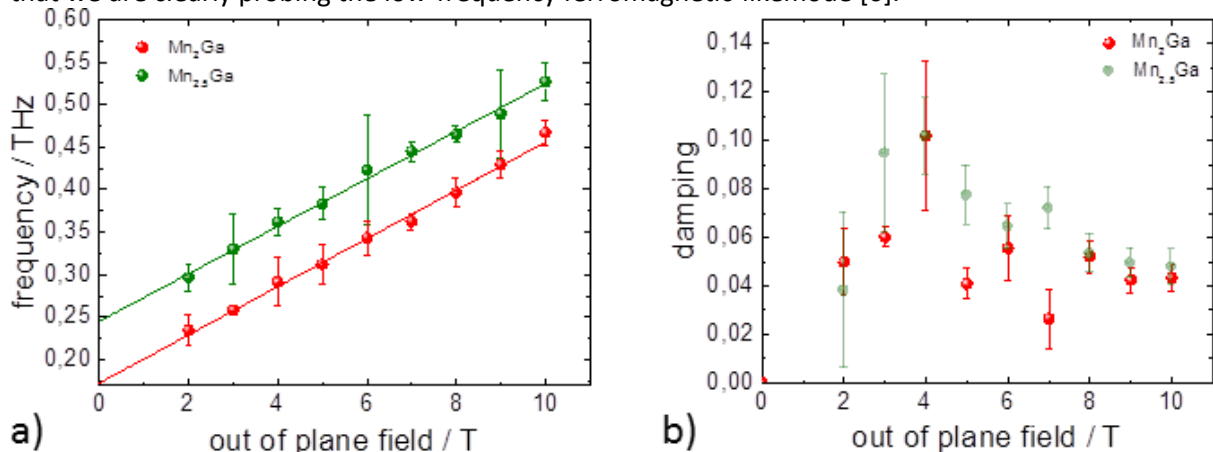


Figure 2: (a) Observed field dispersion of the resonance frequencies for  $\text{Mn}_2\text{Ga}$  (red) and  $\text{Mn}_{2.5}\text{Ga}$  (green). The frequency increases linearly with the field and reaches 0.52 THz for the  $\text{Mn}_{2.5}\text{Ga}$  sample. The temperature in these measurements was 240 K. (b) Observed changes of the damping as a function as the out of plane magnetic field.

## Risk Management

### HZDR

The established end station now enables to measure field dispersion in samples that exhibit a sufficiently large THz emission. The developed set-up in its current form is suitable to fulfil all foreseen characterizations of thin films within the TRANSPIRE project. The set-up nevertheless shall be further improved.

### TCD:

The existing series of samples has been sufficient to achieve all required deliverables. However for future work new films need to be grown and hence the observed the differing sample properties between runs needs to be thoroughly investigated.

## Outlook

Currently further measurements are performed that utilize the new capabilities of the THz emission spectroscopy set-up to investigate the THz emission characteristics of  $\text{Mn}_{3-x}\text{Ga}$ , including damping (see figure 2b), in the now available full parameter space of 0 – 10 T fields and 3 – 300 K temperatures. Measurements at moderate fields but to temperatures of up to 500 K are also under way and shall help to understand the magnon-phonon interactions in these films.

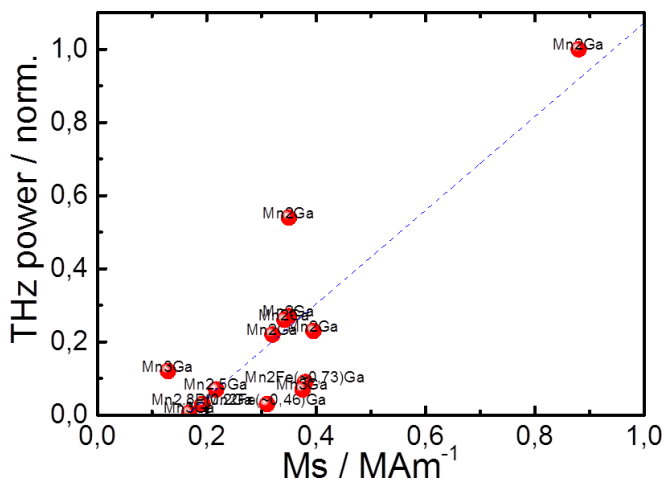


Figure 3: Dependence of the THz power in laser-driven THz emission on the  $M_s$ .

Discussions are ongoing on how more optimal THz emission properties can be achieved from optimized sample geometries and sample stoichiometries. For increasing the emitted THz amplitude, stacks of closely spaced  $\text{Mn}_{3-x}\text{Ga}$  films would be one option that shall be tested. According to the superradiance principle a stack of e.g. 10 nanofilms would yield a factor of 100 more power. Figure 3 shows a survey of THz emission intensities of more

than 10 samples with different saturation magnetization determined by magnetometry. The THz intensity seems to be directly proportional to the value of  $M_s$ , indicating that THz emission intensity can also be increased by optimizing  $M_s$  at a given stoichiometry.

## Conclusion

The field dispersion was successfully measured up to values beyond 500 GHz. The linear dependence of the resonance frequency on the out-of-plane magnetic field confirms the ferromagnetic nature of the resonance.

## References

- [1] N. Awari, et al., Appl. Phys. Lett. 109, 032403 (2016)
- [2] H Kurt, K Rode, M Venkatesan, P Stamenov, JMD Coey, Physical Review B, 83 020405 (2011)
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- [5] in preparation.
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