Deliverable Report *Author : Ciarán Fowley, Karsten Rode, Alexandre Dimitriades* *Date : 30.06.2017*			TRANSPIRE			
Deliverable	**Deliverable name (Short name)**	**WP No.**	**Lead participant**	**Type**	**Diss. Level**	**Date**
D3.1	Resonant excitation in zero-field of an ferrimagnet film with a resonance frequency of order 0.5 THz	3	St12	Report	Public	M6

Progress beyond state-of-the-art: **Detection of tunable narrowband THz emission from magnetic thin films over a distance of several tens of centimetres, resonant excitation of a ferrimagnetic film at a center frequency of 0.36 THz**	
Partner	**Contribution**
TCD	Film optimisation and growth; Structural characterisation; Magnetisation measurements; Hall measurements
HZDR	Characterisation of THz emission, resonant selective excitation of the ferromagnetic mode
St12	Zero-field transmission and reflection measurements of ferrimagnetic films in the range of 0.11-0.17 THz

The deliverable was met to a large part with the publication of a paper from the consortium with authors only from the TCD and HZDR research groups.

Title: "Narrow-band tunable terahertz emission from ferromagnetic $Mn_{3-x}Ga$ thin films"

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In order to achieve the end goal of the project the initial proof that a ferrimangetic film can emit THz radiation at distance (in far field), must be proven. For this, several films based on the tetragonally distorted $Mn_{3-x}Ga$ family were to be grown by sputtering at TCD. HZDR is supposed to characterize the films by means of laser-driven THz emission (later on also spin-torque driven) and by resonant excitation by means of a novel superradiant narrow band THz sources called TELBE. St12 is supposed to develop a fast screening process for zero-field excitation using a Vector Network Analyzer (VNA) and compatible THz sources and utilize them to characterize further films produced by TCD. Due to technical problems described later in this report the latter part of the deliverable (fast screening of samples at St12) is delayed, without however affecting the achievement of the deliverable.

Sample growth

Two sets of samples were grown by the TCD partner:

i) a series of $Mn_{3-x}Ga$ on 10x10 mm^2 double-side polished MgO single crystal substrates with 0 < x < 1. The growth conditions had previously been determined [1-3], however due to slight changes in heater configurations and target yields a one week optimisation run was necessary to obtain samples of sufficiently high quality for reliable measurements of the resonant frequency. Supporting measurements, performed prior to shipping, include X-ray diffraction and reflectivity as well as SQUID magnetometry.

ii) A series of Mn_2Fe_xGa on 10x10 mm^2 double-side polished MgO single crystal substrates with 0.1 < *x* < 1.2. This second set was subjected to an intense set of characterisation techniques in order to determine the physical properties of this novel Heusler alloy. Part of this work was carried out under TRANSPIRE and is currently awaiting publication in Phys. Rev. B [4].

Set i) was sent to partner HZDR for THz emission spectroscopy while set ii) was sent to partner St12 for THz absorption/reflection measurements. Due to technical problems (see risk management section below), a third sample set (iii) of Mn_2Fe_xGa was grown on 25 x 25 mm^2 single-side polished MgO substrates. Due to late arrival at the St12 site, these samples are awaiting measurement, currently planned for July 2017.

It should be noted that the current setup at St12 has a spectral range limited to a maximum frequency of ~0.17 THz. The samples produced for them by TCD were adapted to sattisfy this requirement. Furthermore, the samples measured at HZDR were found to have a maximum resonant frequency of 0.36 THz, of the same order as the stated 0.5 THz aimed for in this deliverable. Using $Mn_{3-x}Ga$, we find that there is prospect of raising the resonant frequency by either substrate/seed compressive strain, or by doping with Ni. It has been shown that the inclusion of Ni allows for a reduction of the saturation magnetisation by a factor of three, thereby multiplying the theoretical resonant frequency by the same factor [5]. TCD will produce such samples during the first available growth run, tentatively scheduled for September 2017.

Characterisation

At HZDR these films were first successfully characterized by means of laser-driven THz emission which is based on the following principle: a femtosecond laser pulse causes a rapid demagnetisation of the films in a small applied field (few 100 mT), the magnetisation then undergoes coherent precession around the effective applied magnetic field consistent with the Kittel formalism for magnetic resonance. The precessing magnetic moments in the plane of the sample cause electromagnetic radiation at the same frequency to be emitted from the films. Electro-optic sampling was used to detect the emitted THz fields. It could thereby be established that the films stoichiometry allows us to shift the THz emission between 0.21 THz (Mn_2Ga) and 0.35 THz (Mn_3Ga). In order to demonstrate resonant excitation, where an oscillating magnetic field matching the precessional frequency is needed, the High-Field High-Repetition-Rate Terahertz facility @ ELBE (TELBE) was used. Here, the frequency of the excitation can be “dialled-in” and the films can be excited selectively. The selective excitation was verified/detected by transient Faraday measurements (0.36 THz with Mn_3Ga). To perform these measurements on ferrimagnetic thin films as intended, a novel, tuneable, spectrally dense THz source such as TELBE is required [6].

Figure 1 show the results of the femtosecond laser demagnetisation experiment, here one can clearly see that based on the chemical composition of the film the magnetic resonance frequency can be altered and covers a broad range from 0.21 THz to 0.35 THz. Due to the placement of the detector at a distance from the sample these data also show that the emitted THz radiation can be detected approximately one meter away which is a necessary requirement for the short-range communication envisioned in TRANSPIRE.

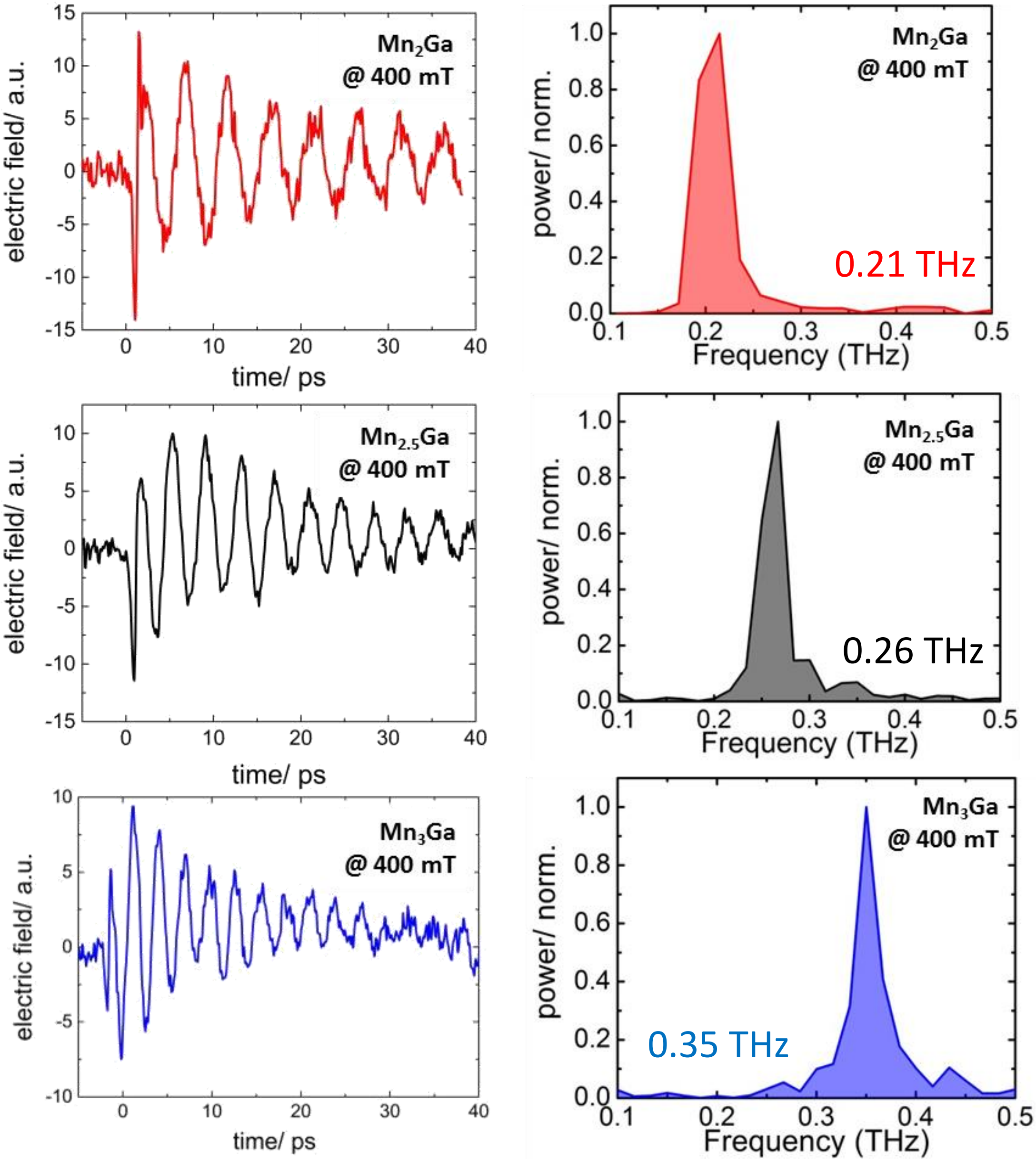

Figure 1: Left column shows the detected electric field component of the emitted THz radiation from Mn_2Ga, $Mn_{2.5}Ga$ and Mn_3Ga films. The right column is a fast-fourier transform of the same data which highlights any predominant frequencies in the spectra. It is clearly seen that MnGa films can emit THz radiation between 0.21 THz and 0.35 THz based on the relative content of manganese and gallium [7].

As mentioned above, the data obtained in figure 1 stems from laser-driven THz emission. In this case the excitation is applied off-resonance at many orders of magnitudes higher photon energies (800 nm / 1.4 eV). The films are subsequently heated and the observed frequencies may not represent those at thermal equilibrium. Selective resonant excitation by tunable, narrow-band, spectrally dense THz pulses from TELBE were hence also performed. In figure 2 the laser-driven THz emission for a Mn_3Ga film sample (0.35 THz) is compared to THz-driven selective excitation of the ferrimagnetic mode probed by transient Faraday rotation (0.36 THz). Frequencies obtained by both methods are in excellent agreement.

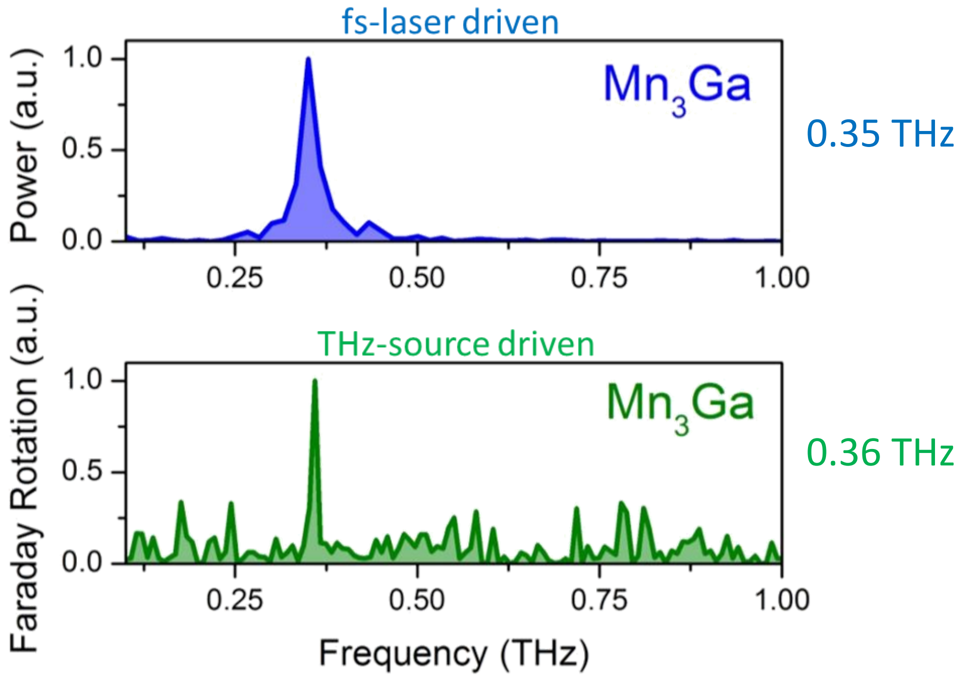

Figure 2: A comparison of emitted frequencies from femtosecond laser demagnetisation and those excited resonantly using the TELBE facility (modified [7]).

The data obtained in these films lead directly into other deliverables in the project where we will attempt to use spin-polarised currents rather than a femtosecond laser to emit THz radiation and use the resonant absorption of radiation to generate spin-currents in adjacent metals via spin-pumping to detect the THz radiation incident on the films.

The third part of this deliverable, the characterization of further films produced by TCD at ST12 is delayed due to sample exchange issues, requiring new samples to be optimised and grown which lead to considerable time delay (more detail is given in the in the risk management section).

Risk Management

The $Mn_{3-x}Ga$ series was successfully characterised by partner HZDR. The TCD and HZDR partners have been collaborating prior to TRANSPIRE and the effective sample exchange, including results from preliminary standard sample characterisation, between the two partners reflects this. Risk here is well managed. The first batch of samples that were sent by TCD to St12 could not be measured due to two main problems: firstly, the sample package had been opened in transit, presumably by Swiss customs, and the sample quality compromised (sample positions in dedicated boxes switched, surfaces scratched, etc); secondly, partner St12 realised that the sample surface (10 x 10 mm^2) was too small to be measured in the currently available setup. These points were raised during the May remote TRANSPIRE meeting and it was decided to make sample set iii) as mentioned above, and also to produce Teflon™ (PTFE) inserts for the setup in order to be able to measure smaller sample surfaces. Sample set iii) was produced at TCD, but reached St12 a day after the planned experiment and could therefore not be measured on time for this report due to equipment unavailability. They will however be measured in the near future (expected July 2017).
The risk at HZDR related to the ability to measure very thin films. The THz emission set-up and the end station at TELBE for selective THz excitation was modified and advanced in a way that allowed us to successfully characterize ferrimagnetic films of only few 10 nm thickness.

Outlook

The data obtained in this deliverable show that we can induce and detect tunable narrow band THz emission from coherently excited ferrimagnetic resonances in $Mn_{3-x}Ga$ films. The actual efficiency within these narrow THz lines is comparable to standard laser-driven THz emitters such as ZnTe [7]. This is a fundamental technological breakthrough as ferrimagnetic resonance is a general phenomenon, which should apply to other ferrimagnetic systems such as MnRuGa (MRG) as well as exchange coupled synthetic anti-ferromagnetic systems (NiFe/Ru/NiFe) which should allow reaching even higher THz frequency bands.

Future efforts will aim at detecting emission from resonances tuned to higher THz frequencies. It shall also be investigated if we can observe laser-driven THz emission from antiferromagnetic resonances which generally occur at higher THz frequencies. St12 will work on the characterization of the aforementioned sample set iii) with their zero-field reflection/transmission measurement setup during July 2017.

Conclusion

We have demonstrated that $Mn_{3-x}Ga$ films can emit tuneable narrow-band THz radiation over a distance of several tens of centimetres and they can be used to selectively detect radiation at the same frequency as their emission profile. The efficiency of the emission is comparable to that of common broad-band laser-based THz emitters such as ZnTe. They therefore show high promise to be implemented in spin-transfer-torque oscillators for the end-goal of the TRANSPIRE project.

References

[1] H Kurt, K Rode, M Venkatesan, P Stamenov, JMD Coey, Physical Review B, 83 020405 (2011)

[2] H Kurt, K Rode, M Venkatesan, P Stamenov, JMD Coey, physica status solidi (b) 248 2338 (2011)

[3] K Rode, N Baadji, D Betto, Y-C Lau, H Kurt, M Venkatesan, P Stamenov, S Sanvito, JMD Coey, E Fonda, E Otero, F Choueikani, P Ohresser, F Porcher, G André, Physical Review B 87 184429 (2013)

[4] D Betto, Y-C Lau, K Borisov, K Kummer, NB Brookes, P Stamenov, JMD Coey, K Rode, arXiv:1704.01326 (2017)

[5] G. Reiss, private communication

[6] B. Green, et al., Scientific Reports 6, 22256 (2016)

[7] N. Awari, et al., Appl. Phys. Lett. 109, 032403 (2016)