1 Introduction
Multiferroics, materials comprising two or more ferroic properties, such as ferromagnetism, ferroelectricity or ferroelasticity in the same phase, are very promising systems for future applications in computing or sensing. These materials would allow to design new digital storage devices thereby combining the advantages of long lived magnetic storage with easy accessibility and robustness of electronic storage technology. Still, materials combining both properties to be useful for applications are scarce. To better adapt multiferroic properties, thin film growth introduces new parameters such as strain, allowing the tuning of materials properties. For a targeted growth control optical emission spectroscopy and time resolved imaging is utilized to investigate the plasma during the growth by pulsed laser deposition.

2 Materials and Methods
Pulsed laser deposition (PLD) is a very powerful tool to evaporate in vacuum or in a background atmosphere a materials, either metal or ceramic, thereby creating a plasma. The species from this plasma cloud are condensing on a substrate and in that way forming a film. The material LuMnO$_3$ with an orthorhombic structure is a promising multiferroic, however the orthorhombic structure is metastable and therefore not easy to synthesize as bulk to study its electrical and magnetic properties. The preparation as a thin film is one way to obtain high quality material with the correct crystalline structure but the preparation as a thin film is not easy to accomplish. In order to investigate the PLD deposition of LuMnO$_3$, in particular the properties of laser induced Lu plasma species from from three different Lu-containing targets were studied: A metal Lu disc target, a ceramic Lu$_2$O$_3$ disc target, and a cylindrical ceramic target of hexagonal-LuMnO$_3$. For the ablation of these targets, a KrF excimer laser ($\lambda = 248$ nm) was used at a repetition rate of 8 Hz with a laser fluence of $\sim 3.5$ J/cm$^2$. A 9 x 9 mm$^2$ mask was imaged onto the target resulting in approx. 1.7 mm$^2$ spot size of the laser beam. The background pressure was varied from vacuum ($1 \times 10^{-8}$ mbar) to $1.5 \times 10^{-3}$ and $5 \times 10^{-2}$ mbar using N$_2$O and O$_2$ background gases.

To study the plasma properties, plasma imaging was performed using an Andor Solis software and a USB iStar camera – DH334T-18F-03 equipped with an acousto-optical tuneable filter (AOTF, Brimrose VA210 .40 .65 H) for the wavelength range of 400 - 650 nm. The wavelength resolution is $\sim 0.8$ nm for the short wavelengths, and up to 2.1 nm around 650 nm. The settings for gating, frame repetition and delay times are mentioned in the respective parts of the discussion. To image the entire plasma (4 cm) including the target and the mass spectrometer nozzle onto the CCD a Nikkor 28 - 300 mm lens was used at a distance of 550 mm to the plasma set to $\sim 200$ mm focal length. These time and space resolved optical emission studies where complemented by plasma mass spectroscopy measurements to determine the plasma composition and the respective kinetic energy of these species. Both, the mass spectrometry and the plasma imaging were triggered from the same photodiode placed in the beam path and used simultaneously to assure consistency of the results as illustrated in Fig. 1.

The emitted plasma radiation was measured as a function of time, space and wavelength. Changes in relative intensities between Lu and LuO lines over time show a distinct behaviour when comparing vacuum conditions with an oxidizing background gas. While the initial expansion of plasma species for all background conditions is similar, the behaviour changes at later times when the interaction between species from the target and the background become dominant. In an oxidizing background gas the relative intensities increase favouring the oxidized species which agrees well with observations from mass spectrometry.
3 Plasma imaging

In plasma imaging, the entire plasma is pictured with a specific filter selecting ideally a single wavelength with a gate width of 10 μs to capture the integrated intensity for the time the plasma exists. In Fig. 2 the emissions from the Lu target at 499 nm (a) (Lu I) and 518.5 nm (b) (LuO) are shown and compared to an optical photograph (c). From these measurements, the integrated spatial expansion of these species can be studied. Whereas excited Lu species seem to reach the mass spectrometer respective substrate position, the intensity for excited LuO is severely diminished. This is confirmed by the mass spectrometry measurements where the number of LuO arriving at the substrate position is approx. 100 times smaller than Lu.

Introducing N₂O as a reactive background gas to enhance the creation of metal oxide species needed to form orthorhombic LuMnO₃, we observe as a function of the background pressure an increase in the number of metal oxide species due to chemical reactions taking place in the plasma cloud. At a pressure of 5x10⁻² mbar N₂O, the distribution of excited Lu and LuO species in the plasma is very different compared to the ablation in vacuum. In Fig. 3a) the distribution of Lu I (red) is shown with a fall-off of the excitation intensity already halfway between the target and the mass spectrometer position. Contrary to the decrease of the excited Lu species the excitation from LuO becomes stronger with increasing distance from the target for the following reason. Due to a much reduced mean free path of these plasma species at this pressure the number of collisions increases and hence the chemical activity thereby creating more LuO species further away from the target. Due to a much reduced mean free path of these plasma species at this pressure the number of collisions increases and hence the chemical activity thereby creating more LuO species further away from the target. These collisions also contribute to a re-excitation of LuO and enhance the lifetime of these excited species which can be observed as a half-moon shaped distribution around the area covered by Lu I.

Integrating the intensities from Fig. 4 shows that from vacuum to the intermediate pressure, an increase of the intensities for both Lu I and LuO is observed with a small increase in the LuO/Lu I ratios. At the highest pressure a strong increase of the LuO intensities is observed after ~400 ns with a maximum around 1200 –1300 ns at which the plasma reaches the mass...
spectrometer, agreeing well with the observations of species distribution in the plasma. At the highest pressure, the oxidation at the shockwave becomes the dominant factor and leads to a strong oxidation of the metal Lu species to LuO and higher oxides, as confirmed by mass spectrometry.

Figure 4: Plasma images for Lu I (top row of each block) and LuO (bottom row of each block) at different delay times for three different background conditions: a) vacuum, b) $1.5 \times 10^{-3} \text{ mbar} \text{N}_2\text{O}$, and c) $5 \times 10^{-2} \text{ mbar} \text{N}_2\text{O}$. All measurements performed on the Lu target.

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