Electric field strength-dependent accuracy of TiAlN thin film composition measurements by laser-assisted atom probe tomography

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Abstract
Accurate quantification of absolute concentrations represents a major challenge for atom probe tomography (APT) since the field evaporation process is affected significantly by the measurement parameters. In the present work we investigate systematically the effect of laser pulse parameters on the accuracy of laser-assisted APT for a TiAlN thin film previously quantified by ion beam analysis, combining Rutherford backscattering spectrometry and time-of-flight elastic recoil detection analysis. The electric field strength is estimated from the Al\(^{2+}/Al^{+}\) charge state ratio for all systematically varied measurement parameters. Subsequently, the absolute concentrations from laser-assisted APT are compared to ion beam analysis data. An increase of the electric field strength from approximately 25–28 V nm\(^{-1}\) improves the accuracy of absolute concentrations measured by laser-assisted APT from 11.4 to 4.1 at% for N, from 8.8 to 3.0 at% for Al and from 2.8 to 0.9 at% for Ti. Our data emphasize that the measurement accuracy of laser-assisted APT for TiAlN is governed by the electric field strength. It is shown that the smallest compositional discrepancies between ion beam analysis and APT are obtained for the maximum electric field strength of approximately 28 V nm\(^{-1}\) at 10 pJ laser pulse energy. This can be rationalized by considering the enhanced ionization of neutral fragments caused by the increased electric field strength.

1. Introduction

Atom probe tomography (APT) allows for spatially-resolved chemical composition analysis at the nanometer scale [1] and the basic principle has been first described in 1968 [2]. Application of a DC voltage on the order of several kV to a needle-shaped specimen with a tip radius on the order of tens of nanometers induces field evaporation [3] of atoms or molecules. The required material-dependent evaporation field is proportional to the ratio of voltage and tip radius. Evaporated species are subsequently ionized up to charge states of +4 due to the surface electric field [4, 5] and a fraction of the ionized species is detected. The field evaporation process can be controlled by applying voltage or laser pulses to the specimen in addition to the DC voltage and for each pulse the time-of-flight (ToF) is measured, while the flight path length is known. Thus, based on the conservation of potential and kinetic energy, the mass-to-charge state ratio can be determined and represents the chemical fingerprint of the detected ion. Spatial information is obtained by the detector hit position of the ion and the original position of the atom or molecule within the tip prior to field evaporation can be reconstructed by using projection algorithms. The subsequent removal of atom-by-atom enables three-dimensional tomographic quantitative probing. With technological progress, such as the advent of local extraction electrodes enabling e.g. detection of \(>10^9\) atoms within a single measurement [6] or femtosecond laser pulsing for characterization of poorly conductive materials [7], versatile commercial instruments have become available [8]. Nowadays APT is recognized as a powerful technique for the identification of local chemical composition variations within nanometer-sized features.
However, accurate quantification represents a major challenge for APT analysis since the obtained absolute concentrations are affected significantly by the measurement parameters. A prominent example is GaN which has been intensely studied by laser-assisted APT and shows significant deviations from the N/Ga = 1 stoichiometry [9–14]. Despite experimental differences of these investigations with respect to employed instruments and measurement parameters, the reported variations of the N/Ga ratio between 0.05 and 1.63 emphasize the measurement parameter-dependent accuracy or lack thereof. The electric field strength was identified to govern the absolute concentration values of GaN since Ga-deficiency was obtained at high electric field strength and associated with preferential evaporation [14], while N-deficiency was observed at low electric field strength and could be explained by the formation of neutral fragments upon dissociation of N$_2$-carrying molecular ions [13]. The stoichiometric composition N/Ga = 1 was obtained for the Ga$^{2+}$/Ga$^+$ ratio of approximately 0.04 which corresponds to an electric field strength of approximately 23.5 V nm$^{-1}$ [14]. In case of Al$_{0.44}$Ga$_{0.56}$N the highest N/(Al+Ga) ratio of 0.86 was obtained for the Ga$^{2+}$/Ga$^+$ charge state ratio of 0.95 [15] which corresponds to an electric field strength of approximately 24.9 V nm$^{-1}$ [13].

Since such drastic compositional variations are obtained by laser-assisted APT already for binary nitrides, it can be expected that the accuracy of APT for ternary nitrides is also significantly affected by the measurement parameters due to different evaporation fields of the individual constituents. We have recently compared the chemical composition of a TiAlN thin film by ion beam analysis and laser-assisted APT, varying the laser pulse energy ascending from 10 to 200 pJ within a single measurement: absolute N concentrations from APT were underestimated by at least 5.5 at% (20 pJ) and up to 8.2 at% (200 pJ) and absolute Al concentrations from APT were overestimated by at least 4.5 at% (20 pJ) and up to 6.2 at% (200 pJ), while absolute Ti concentration values were found to be in good agreement for both techniques with maximum deviations <2 at% [16]. Hence, the accuracy was on the order of 5–8 at%, while the precision was approximately 1 at%. Underestimation of N was explained by the formation of neutral fragments upon dissociation of molecular ions during the flight, where the electric field strength is insufficient to induce ionization of the neutral fragment [13] and the Al overestimation was suggested to be caused by preferential Al evaporation [16].

In the present work, the effect of systematic variations of APT measurement conditions on the chemical composition of a TiAlN thin film, previously analyzed by ion beam analysis, is investigated. Our data emphasize that the accuracy of composition measurements is governed by the electric field strength. Recommendations for improving the accuracy of laser-assisted APT for TiAlN by tuning the measurement parameters, and hence the evaporation conditions, are presented.

2. Experimental details

2.1. Materials

The entire experimental details regarding thin film synthesis and ion beam analysis were reported by to Baben et al [17] and a brief summary is provided here. TiAlN thin films were grown reactively by high power pulsed magnetron sputtering [18] on sapphire substrates at a temperature of 300 °C from a TiAl target. While the Ar flow of 200 sccm was supplied homogeneously, the N$_2$ flow of 9 sccm was introduced by a separate inlet, positioned at one end of the target. Thereby, TiAlN thin films with a N composition spread [19] were obtained and a close-to-stoichiometric thin film was identified using Rutherford backscattering spectrometry (RBS with 2 MeV He$^+$ ions) and ToF elastic recoil detection analysis (ERDA with 36 MeV 18O$^+$ ions). The combination of RBS and ToF-ERDA (referred to as RBS/ERDA in the following) resulted in a total measurement uncertainty of 2.5% relative deviation and the absolute composition is (Ti$_{0.255}$Al$_{0.235}$)(N$_{0.505}$O$_{0.005}$). Moreover, the film exhibits a single phase face-centered cubic crystal structure (space group Fm$3$m, NaCl prototype) [17] and a columnar growth morphology with column widths on the order of 100–200 nm [16].

2.2. Characterization

APT measurements were performed in a CAMECA local electrode atom probe (LEAP) 4000X HR equipped with a reflectron and the flight path length was 382 mm. An ultraviolet laser was used for pulsing and the detection rate was set to 5 ions out of 1000 applied pulses. The evaporation conditions were chosen in an effort to probe systematically the effects of laser pulse parameters on measured absolute concentrations as explained in the following:

- The effect of base temperature on the measurement accuracy was investigated by systematic variation from 20 to 40, 60, 80 and 100 K within individual measurements. For each temperature $10 \times 10^6$ ions were acquired. Laser pulse energy and frequency were set at 30 pJ and 250 kHz, respectively.
- The effect of laser pulse frequency on the measurement accuracy was investigated by a subsequent raise from 125 to 250, 333 and 500 kHz within a single measurement. For each frequency $4 \times 10^6$ ions were...
acquired, resulting in a total ion count of $16 \times 10^6$. Laser pulse energy and base temperature were set at 30 pJ and 60 K, respectively.

- The systematic reduction of the laser pulse energy from 200 to 100, 50, 40, 30, 20 and 10 pJ (descending) within a single measurement is compared to the previously published systematic increase from 10 to 200 pJ (ascending) [16]. For each energy $4 \times 10^6$ ions were acquired, resulting in a total ion count of $28 \times 10^6$. Laser pulse frequency and base temperature were set at 250 kHz and 60 K, respectively.

- An additional measurement series at constant laser pulse energy of 30 pJ was carried out and is compared to the descending laser pulse energy variation. The total ion count was $28 \times 10^6$ ions and data analysis in segments of $4 \times 10^6$ detected ions enables the comparison with the descending laser pulse energy variation. Laser pulse frequency and base temperature were set at 250 kHz and 60 K, respectively.

Data analysis was carried out with the IVAS 3.8.0 software package. The amount of multiple detection events was extracted by using the software EPOSA which was developed at the Institute of Physics (IA) of RWTH Aachen University. Ranging of the mass spectrum was intended to minimize the contribution of thermal tails. For each peak the mass-to-charge state ratio of the maximum intensity was identified (e.g. 13.494 Da for Al$^{2+}$) and compared to the peak onset from the background (e.g. 13.450 Da for Al$^{1+}$). The range was then defined symmetrically by the difference of these mass-to-charge state ratios (e.g. 13.450–13.538). Besides Ti, Al and N, impurities of O (approximately 1 at%) and traces of C, Ar and Ga (<0.1 at%) were identified. APT specimens were prepared by focused ion beam (FIB) with a FEI Helios Nanolab 660 dual-beam microscope according to a standard protocol [20]. Sample transfer from the FIB system to the LEAP took <3 min and during that time the specimens were exposed to atmosphere.

3. Results and discussion

3.1. Mass spectrum

Mass spectra from the constant 30 pJ laser pulse energy series are presented for ranges of $0–4 \times 10^6$ and $24–28 \times 10^6$ detected ions in figure 1 and the most abundant ionized species (Al$^{2+}$, N$^+$, Ti$^{2+}$, Al$^+$, N$_2^+$ and TiN$^{2+}$) are also provided as insets. The fraction of molecular ions accounts to approximately 30% for both mass spectra. Elemental concentrations were obtained by ranging of the peaks according to the protocol described in section 2.2.2, decomposition of molecular ions and integration of the peak areas for all of the in the following discussed chemical composition data.

3.2. Base temperature and laser pulse frequency

The chemical composition of the close-to-stoichiometric TiAlN film quantified by APT is shown in figure 2(a) for base temperatures of 20, 40, 60, 80 and 100 K and (b) for laser pulse frequencies of 125, 250, 333 and 500 kHz. The base temperature does not affect the accuracy significantly since Ti, Al and N concentrations are between 25.8 and 26.6, 27.9 and 28.9 as well as 43.6 and 44.3 at%, respectively. Thus, base temperature-induced maximum compositional variations are $\leq 1$ at% and within the instrument precision for TiAlN [16]. In comparison to RBS/ERDA it can be learned that depending on the base temperature absolute N concentrations from APT are underestimated by 6.2–6.9 at%, absolute Al concentrations are overestimated by 4.4–5.2 at% and Ti concentrations are for both techniques in good agreement with deviations of only 0.8–1.1 at%.

Similar results were obtained for laser pulse frequencies $\leq 333$ kHz, figure 2(b): Ti, Al and N concentrations are between 26.3 and 26.6, 27.9 and 28.1 as well as 44.1 and 44.8 at%, respectively. Hence, for laser pulse frequencies $\leq 333$ kHz absolute N concentrations from APT are underestimated by 5.7–6.4 at%, absolute Al concentrations are overestimated by 4.4–4.6 at% and Ti concentrations are for both techniques in good agreement with deviations of only 0.8–1.1 at%. However, changing the frequency from 333 to 500 kHz resulted in significant compositional changes as they are larger than the instrument precision for TiAlN [16]: Ti decreases from 26.3 to 23.8 at%, Al increases from 27.9 to 29.4 at% and N increases from 44.8 to 46.4 at%. These significant changes of absolute concentrations can be understood by considering the ToF of ionized species. For 500 kHz the laser pulse period corresponds to 2 $\mu$s, hence, the ionized species need to reach the detector within 2 $\mu$s before the following pulse is initiated. The highest detectable mass-to-charge state ratio at 500 kHz can be calculated based on energy conservation:

$$m \frac{q}{u} = \frac{2Ue}{\left(\frac{t}{l}\right)^2}$$

$m$ and $q$ denote mass and charge state, $U$, $t$ and $l$ correspond to voltage, ToF and flight path length and $e$ and $u$ are elementary charge and atomic mass unit. The voltage range of the measurement segment at 500 kHz was
6600–7000 V which means that the mass spectrum is cut off at 35–37 Da. Hence, ionized species with \( q/m > 35–37 \) Da (including \( TiN^+, Ar^+, AlO^+, TiO^+ \) and \( TiO^+ \), figure 1) can not be detected. Consequently, N is underestimated by 4.1 at% and Al is overestimated by 5.9 at% and Ti is underestimated by 1.7 at%.

Ti, Al and N concentrations measured for systematic variations of laser pulse energy (ascending from 10 to 20, 30, 40, 50, 100 and 200 pJ) \([16]\), base temperature (20, 40, 60, 80 and 100 K) and laser pulse frequency (125, 250 and 333 kHz) are compared to RBS/ERDA data in table 1. Minimum and maximum deviations represent the measurement accuracy of laser-assisted APT for TiAlN. Significant compositional deviations with respect to ion beam analysis data are evident and the largest compositional deviations are obtained for the laser pulse energy variation.

### 3.3. Laser pulse energy

The descending laser pulse energy variation from 200 to 10 pJ is compared to the ascending variation from 10 to 200 pJ \([16]\) in figure 3. Reduction of the laser pulse energy from 200 to 10 pJ results in a N concentration increase from 39.1 to 46.4 at%, an Al concentration decrease from 32.3 to 26.5 at% and a Ti concentration decrease from

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**Figure 1.** Mass spectra for constant laser pulse energy of 30 pJ showing the detected ions of \( 0 \times 10^6 \) (blue solid line) and \( 24 \times 10^6 \) (red dashed line). The insets provide an enlarged view of selected peaks from the mass spectrum. Detailed peak indexation is explained in the following as ‘Da - species’: \( 6.0 - C^2+; 7.0 - N^2+; 9.0 - Al^+; 12.0 - C^+; 13.5 - Al^2+; 14.0, 15.0 - N^+; 15.3, 15.7, 16.0, 16.3, 16.7 - Ti^+; 20.5 - AlN^2+; 21.0 - N^2; 21.5 - AlO^2+; 22.7 - AlN^3+; 23.0, 23.5, 24.0, 24.5, 25.0 - Ti^2+; 26.0 - Ti^3+; 27.0 - Al^2+; 28.0, 29.0 - N^2; 30.0, 30.5, 31.0, 31.5, 32.0 - TiN^2+; 36.6 - Ti_2N^3+; 40.0 - Ar^+; 43.0 - AlO^+; 48.0 - Ti^+; 62.0 - Ti^3+; 64.0 - TiO^+.

**Figure 2.** Chemical composition as a function of (a) base temperature and (b) laser pulse frequency. Ti, Al and N absolute concentrations are represented by squares, circles and diamonds, respectively. The colored regions indicate average concentrations including corresponding uncertainties quantified by combination of RBS/ERDA: \( 25.5 \pm 0.6 \) at% Ti (blue), \( 23.5 \pm 0.6 \) at% Al (red) and \( 50.5 \pm 1.3 \) at% N (green).
Table 1. Accuracy of laser-assisted APT for TiAlN, represented by the minimum and maximum deviations of measured absolute Ti, Al and N concentrations with respect to RBS/ERDA upon variations in laser pulse energy (ascending), base temperature and laser pulse frequency. The 500 kHz frequency data is excluded since ionized species with q/m > 35–37 Da can not be detected.

|                  | Laser pulse energy [16] | Base temperature | Laser pulse frequency |
|------------------|-------------------------|------------------|-----------------------|
|                  | Min. | Max.   | Min. | Max. | Min. | Max. |
| Ti               | +0.3 | +1.6   | +0.8 | +1.1 | +0.8 | +1.1 |
| Al               | +4.5 | +6.2   | +4.4 | +5.2 | +4.4 | +4.6 |
| N                | −5.5 | −8.2   | −6.2 | −6.9 | −5.7 | −6.4 |

Figure 3. Comparison of descending variation of laser pulse energy from 200 to 10 pJ (filled symbols) and ascending variation [16] of laser pulse energy from 10 to 200 pJ (open symbols). (a) Ti, Al and N concentrations as a function of the laser pulse energy. The colored regions indicate average concentrations including corresponding uncertainties quantified by combination of RBS/ERDA: 25.5 ± 0.6 at% Ti (blue), 23.5 ± 0.6 at% Al (red) and 50.5 ± 1.3 at% N (green). (b) Difference of APT concentration values x and RBS/ERDA data x_{RBS/ERDA}.

Figure 4. Constant laser pulse energy of 30 pJ. (a) Ti, Al and N concentrations as a function of the number of detected ions. The colored regions indicate average concentrations including corresponding uncertainties quantified by combination of RBS/ERDA: 25.5 ± 0.6 at% Ti (blue), 23.5 ± 0.6 at% Al (red) and 50.5 ± 1.3 at% N (green). (b) Difference of APT concentration values x and RBS/ERDA data x_{RBS/ERDA}.
number of detected ions since e.g. N is underestimated by 6.8 and 5.3 at% for the ranges of 0.28 to 26.4 at%,
from 29.4 to 27.6 at% and a Ti concentration increase from 25.7 to 26.3 at%,
further discussed in section 3.5. It should be noticed that the laser pulse energy was with 30 pJ identical for the
descending variation as well as constant laser pulse energy in the range of 16–26.2 at%, 27.7/28.0 at% and 45.0/45.0 at% (descending/
constant), figures 3(a) and 4(a). Maximum absolute concentration deviations ≤0.3 at% are significantly smaller
than the instrument precision of 1 at% [16].

3.4. Multiple detection events
Figure 5 shows (a) the amount of multiple detection events for the descending variation of laser pulse energy
from 200 to 10 pJ as well as (b) for the constant 30 pJ laser pulse energy. The occurrence of multiple detection
events can be understood by a strong correlation of ionized species in time and space since evaporation is
initiated locally in confined zones where the electric field is most intense [21]. Simultaneous arrival of ionized
species at the detector may not be monitored due to the dead time and dead zone of the detector [22], hence, it is
reasonable to expect that multiple detection events affect the accuracy of laser-assisted APT [21]. The reduction
of the laser pulse energy from 200 to 10 pJ results in an increase of multiple detection events from 27% to 53%, while e.g. the N concentration is increased from 39.2 to 46.7 at%. Hence, it appears unlikely that multiple detection events govern the evolution of absolute concentrations for the variation of laser pulse energy. Rather, the improved accuracy of absolute N concentrations from 11.3 to 3.8 at% appears to be related to the laser pulse energies for the ascending laser pulse energy variation [16].

In case of constant 30 pJ laser pulse energy, the fraction of multiple detection events is reduced from 51% to 49% with increasing number of detected ions, while the N concentration increases from 44.2 to 45.7 at%. This may be understood by the fact that the dissociation of molecular ions contributes to multiple detection events [23] and underestimation of N was explained by the formation of neutral fragments upon dissociation of molecular ions during the flight, where the electric field strength is insufficient to induce ionization of the neutral fragment [13]. Hence, a lower number of neutral fragments should be accompanied by a reduction in multiple events and an increase in N concentration.

3.5. Electric field strength

In the following, the deviation of the TiAlN chemical composition data determined by APT is compared to the ion beam analysis data by using the N/(Ti+Al) ratio. For each set of evaporation conditions probed here, as well as the data from [16], the electric field strength was estimated based on the measured Al$^{+}$/Al$^{+}$ charge state ratios [3] which are shown in figure 6(a). It can be learned that the accuracy of APT for TiAlN is governed by the electric field strength, figure 6(b). An increase from approximately 25–28 V nm$^{-1}$ improves the accuracy of laser-assisted APT from 11.4 to 4.1 at% for N, from 8.8 to 3.0 at% for Al and from 2.8 to 0.9 at% for Ti. This correlation is in agreement with field evaporation of GaN at low electric field strength [14] since dissociation of molecular ions during the flight leads to the formation of neutral fragments and the electric field is not sufficient to induce ionization thereof [13]. Evidence of dissociation is not observed in the present data since the LEAP 4000X HR is equipped with a reflectron which compensates energy discrepancies of particles with lower or higher energies than expected [23]. Nevertheless, it appears reasonable that dissociation of N-containing molecules takes place during evaporation of TiAlN and an increase of the electric field strength enhances ionization of these neutral fragments, thereby improving the accuracy of laser-assisted APT for TiAlN thin films.

Estimated electric field strengths are in the range of 27.3–27.6 V nm$^{-1}$ for variations of base temperature and laser pulse frequency as well as for the constant 30 pJ laser pulse energy. However, the variation of laser pulse energy results in an electric field strength range of 25.1–28.0 V nm$^{-1}$. As all of the measurements were carried out at constant detection rate, measurements at a larger pulse energy are conducted at lower DC voltage. Thus, decreasing the laser pulse energy from 200 to 10 pJ, while keeping the detection rate constant, is accomplished by increasing the DC voltage. Consequently, the magnitude of the electric field strength is enhanced by approximately 12%. The highest electric field strength of approximately 28 V nm$^{-1}$ was obtained for the 10 pJ laser pulse energy segment of the descending variation. Hence, it is evident that evaporation conditions of low laser pulse energy are favorable in order to maximize the electric field strength and, thereby, the measurement.

![Figure 6. Comparison of absolute TiAlN chemical compositions for all APT measurements. (a) N/(Ti+Al) concentration ratio as a function of the charge state ratio of double and single ionized Al. (b) N/(Ti+Al) concentration ratio as a function of the electric field strength estimated from the charge state ratios in (a) using Kingham curves [5]. The colored region indicates the average N/(Ti+Al) ratio including the corresponding uncertainty quantified by combination of RBS/ERDA with the value 1.03 ± 0.05.](image-url)
The accuracy of laser-assisted APT for TiAlN. This can be rationalized by considering the above discussed enhanced ionization of neutral species in the presence of a high electric field.

The evolution of absolute Ti, Al and N concentrations obtained by laser-assisted APT at constant laser pulse energy is induced by the continuous increase of the DC voltage required to maintain the constant detection rate, causing a concomitant increase in electric field strength. Hence, the increase of e.g. the N concentration from 43.6 to 45.2 at% (figure 4(a)) is attributed to the enhancement of the electric field strength with increasing number of detected ions. However, also the specimen geometry is modified. The geometric change of the APT specimens which were utilized for the descending laser pulse energy variation and the constant 30 pJ series has been determined prior and after the composition measurement and is depicted in figure 7. Both specimens exhibited an initial tip radius of approximately 10 nm and an initial shank angle of approximately 15° as shown in figures 7(a) and (c). After APT measurements with 28 × 10⁶ detected ions, the radii increased to approximately 60 and 65 nm for the descending variation and constant laser pulse energy, respectively, while the shank angles were not changed significantly, figures 7(b) and (d). The mass spectra from the constant 30 pJ laser pulse energy series for ranges of 0–4 × 10⁶ and 24–28 × 10⁶ detected ions in figure 1 show that all peaks exhibit tails which are caused by delayed evaporation after the thermal pulse since the tip is not cooled instantaneously [24]. Further, it is visible that thermal tails are reduced for the detected ion range of 24–28 × 10⁶ since the peaks of the range 0–4 × 10⁶ are broader. It can be inferred that an increase in the tip radius enables more efficient cooling of the tip and less delayed evaporation resulting in improved accuracy.

4. Conclusions

We have systematically investigated the effect of laser pulse parameters on the accuracy of chemical composition measurements by laser-assisted APT for a TiAlN thin film. Significant compositional deviations with respect to ion beam analysis data were observed. N/(Ti+Al) ratios of all evaporation conditions were compared by the Al²⁺/Al⁺ charge state ratio which serves for estimation of the electric field strength. An increase of the electric field strength from approximately 25–28 V nm⁻¹ improves the accuracy of absolute concentrations measured by laser-assisted APT from 11.4 to 4.1 at% for N, from 8.8 to 3.0 at% for Al and from 2.8 to 0.9 at% for Ti. Thus, it can be learned that the measurement accuracy of laser-assisted APT for TiAlN is governed by the electric field strength. The smallest compositional discrepancies between ion beam analysis and APT are obtained for the maximum electric field strength of approximately 28 V nm⁻¹ at 10 pJ laser pulse energy. As only ionized N can be detected the above reported findings can be understood by considering that the ionization probability of neutral species is enhanced as the electric field strength is increased.

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