Experimental Evaluation of the Interrelationships Between Laser Energy, Temperature, Applied Bias, and Measured Composition in Laser Pulsed Atom Probe Tomography

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Atom probe tomography (APT) relies on the use of controlled field evaporation of ionic species from the apex of a needle shaped specimen. From the first studies of field desorption it has been recognized, that this is a thermally activated process, such that increasing a specimen temperature decreases the field strength required to induce field evaporation [1, 2]. It has also been observed that laser pulsing may be used as a means of generating a transient temperature spike [3]. By calibrating the reduction in evaporation field due to increases in temperature with the reduction in field due to increases in laser power the average peak temperature reached by the apex of the specimen may be determined. Previous studies have illustrated these relationships between laser energy and temperature for elemental metals, such as W, Mo, and Rh [3, 4]. For the elemental semiconductors Si and Ge, the temperature rise caused during laser pulsing an atom probe specimen has been estimated based on the absorption by free carriers and the transfer of energy to the lattice [5].

This relationship between laser fluence and the resulting peak temperature has been virtually unexplored for non-metallic compounds. Likewise, there is little information regarding the role of the base temperature. A relationship between the laser absorption and the applied bias may also exist, thus further impacting the temperature rise [6]. Adding more complexity to the situation, previous analyses have shown that for non-metallic compounds, the measured composition can be affected, sometimes dramatically, by the relative contributions of the laser energy incident on the specimen and the applied bias [7-12]. Evaluation of the interrelationships between these variables can be used to better understand the field evaporation process, especially as it pertains to optimizing APT data collection.

In this work, systematic studies of the relationships between laser energy, base temperature, applied bias, and measured compositions for the compounds CdTe, GaN, Al₂O₃, and NiO were performed using a Cameca LEAP 4000X Si APT instrument. For CdTe, linear relationships were observed between the field and laser energy as well as between the field and base temperature (Figure 1). This is indicative of a thermally assisted process and allowed for the determination of the average peak temperature attained at the specimen apex [13]. Al₂O₃ and NiO also exhibited relationships between those variables, though they deviate from linearity as shown in Figure 2. In contrast, over the laser energies and base temperatures explored for GaN, increases in laser energy greatly decreased the necessary bias, but changes in base temperature exhibited relatively little effect as shown in Figure 1. In addition to those relationships, for all of these compounds, the measured compositions were affected to varying degrees by the other parameters. There does not appear to be a general relationship that applies to all of the materials; rather, the particulars of each material need to be considered when examining the effects of the variables on measured compositions.

References:

- [1] E.W. Müller, Physical Review 102 (1956) p. 618-624.
- [2] R. Gomer, The Journal of Chemical Physics 31 (1959) p. 341-345.
- [3] G.L. Kellogg, J. Appl. Phys. 52 (1981) 5320.
- [4] F. Vurpillot et al, Journal of Physics D: Applied Physics 42 (2009) p. 125502.
- [5] E.P. Silaeva et al, New Journal of Physics 14 (2012) p. 113026.
- [6] E.P. Silaeva et al, Nano Letters 14 (2014) p. 6066-6072.
- [7] T. Hashizume et al, Review of Scientific Instruments 57 (1986) p. 1378-1380.
- [8] R. Agrawal et al, Journal of Physical Chemistry C 115 (2011) p. 17688–17694.
- [9] R. Kirchhofer, M.C. Teague, and B.P. Gorman, Journal of Nuclear Materials 436 (2013) p. 23-28.
- [10] A. Devaraj et al, J. Phys. Chem. Lett. 4 (2013) p. 993-998.
- [11] D.R. Diercks et al, Journal of Applied Physics 114 (2013) p. 184903.
- [12] L. Mancini *et al*, The Journal of Physical Chemistry C 118 (2014) p. 24136-24151.
- [13] D.R. Diercks and B.P. Gorman, The Journal of Physical Chemistry C 119 (2015) p. 20623-20631.

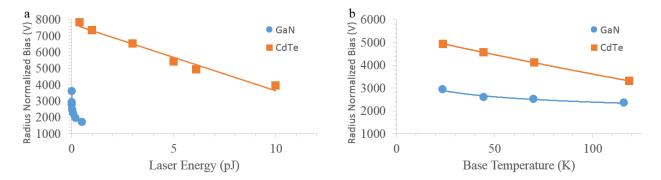


Figure 1. a) The relationship between applied bias and laser energy for GaN and CdTe at a fixed base temperature and detection rate. b) The relationship between applied bias and base temperature for GaN and CdTe at a fixed laser energy and detection rate.

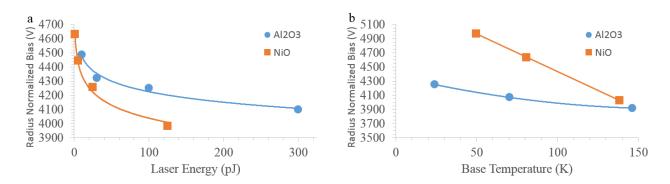


Figure 2. a) The relationship between applied bias and laser energy for Al_2O_3 and NiO at a fixed base temperature and detection rate. b) The relationship between applied bias and base temperature for Al_2O_3 and NiO at a fixed laser energy and detection rate.