

## Appendix 2

### Determination of the ionization efficiency

Total number of particles ablated per pulse:

$$N_{tot} = \frac{V \times \rho}{M} \quad [\text{particles/pulse}]$$

For 200 fs pulses at 4 J/cm<sup>2</sup> irradiating a sapphire target, we have:

$$N_{tot} = 0.8 \times 10^{12} \quad [\text{particles/pulse in the gentle phase}]$$

$$N_{tot} \geq 5 \times 10^{12} \quad [\text{particles/pulse in the strong phase}]$$

where:

$$M = \frac{3M_O + 2M_{Al}}{5} \quad \text{is the average mass for sapphire, } M=20.4 \text{ u, } V \text{ is the}$$

volume ablated per pulse and  $\rho$  is the material density [g/cm<sup>3</sup>].

The total ion yield is determined by the following procedure:

The measured yield for a fixed angle and extraction time is given by the integral of the ion peak in the mass resolved TOF spectra as in Fig. 2.2-2 middle and bottom part:  $I(t)$  [mV $\times\mu$ s]

The corrected yield in the velocity space is  $n(v)=I(t)t/D$  [mV $\times\mu$ s<sup>2</sup>/m] where  $t$  is the extraction time,  $v$  is the velocity,  $v=D/t$ , and  $D$  is the separation between the extraction zone and the sample (see Fig. 2.2-1).

Total ion yield at a fixed angle is:

$$Y(\theta) = \int_v n(v) dv \quad [\text{mV}\times\mu\text{s}^2/\text{s}]$$

The density of counts is determined by the active area of the extraction grids (covered by the solid angle determined by the MSP aperture).

$$\sigma(\theta) = Y(\theta) / A = \sigma_f(\theta)$$

Total number of counts is given by the integration of the angular distribution  $f_r(\theta)=\cos^p(\theta)$  over the hemisphere

$$N = 2\pi r^2 \sigma \int_0^1 f^p (\cos(\theta)) d \cos(\theta) = \frac{2\pi r^2 \sigma}{p+1} \quad [\text{V s}]$$

Since the MSP output has  $R=50 \Omega$  impedance, the measured charge is given by:

$$Q = \frac{N}{R} \quad [\text{C}]$$

The number of measured elementary charges is:

$$Q_{\text{measured}} = \frac{Q}{e}$$

The absolute number of ions is:

$$Q_{\text{absolute}} = \frac{Q_{\text{measured}}}{T\eta F}$$

where  $T$  is the grids transmission  $n_{\text{grids}} \times 90\%$ ,  $\eta$  is the detector efficiency, taking into account the active area ( $\sim 30\%$ ) and  $F$  is the amplification factor ( $\sim 10^3$ ).

The ionization degree is thus given by:

$$\alpha = \frac{Q_{\text{absolute}}}{N_{\text{tot}}}$$