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## A Tomographic Atom Probe laser assisted by a flexible optical system

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The use of ultra-fast laser pulses in Atom Probe Tomography (APT) opened the technique to the analysis of many non-conductive materials for micro- and opto-electronic, solar cells and even geologic materials. This technic, based on time of flight mass spectrometry (TOFMS) of field evaporated ions from a very thin needle, allows for 3d chemical mapping at nanometric scale. Nowadays, most of the APT systems use near ultra-violet (UV) light at a wavelength of  $\lambda \sim 350\text{nm}$ . It is the outcome of years of research on laser-tip interaction demonstrating that ultrashort near-UV light induces fast heating and cooling of the sample for almost all materials [1]. These fast thermal processes trigger the field evaporation on a short time scale ( $\sim 1\text{ns}$ ) resulting in high quality TOFMS.

Current APT systems use optical lenses to focus laser pulses on the tip apex, however, common lenses suffer from chromatic aberration (focal length change of about 25% from 200nm to 2000nm on fused silica) and reduce transmission range (350nm-2000nm for common BK7 glass). Thus, a modification in laser wavelength leads to a change in lenses positioning or to a complete change the optical system when the change in positioning is too large or when the lenses become opaque.

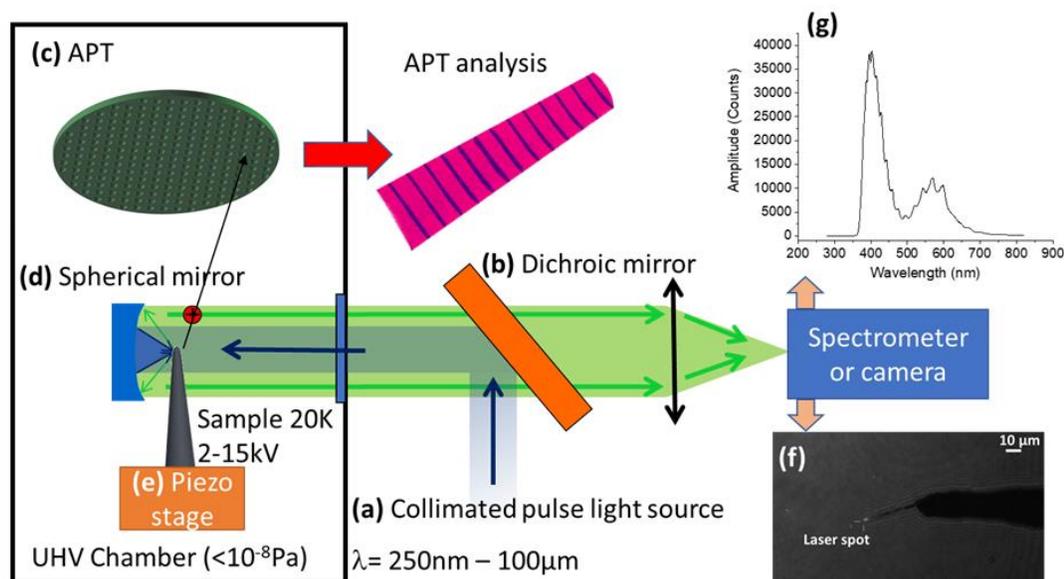
In this presentation, we propose to overcome this limitation using a reflective optical system that is fully achromatic. Many choices were available, but as a first approach, we use a spherical mirror inside the APT chamber. The proposed setup is “in axis”, which means that the collimated laser beam goes through the tip shaped sample (a part of it is stopped by the sample  $\sim 5\text{-}10\%$ ) and, as shown in **Figure 1**, it is focused on the tip apex by the spherical mirror. In our setup, this mirror is quite large (25mm) and its focal length is of 25 mm, resulting in the large numerical aperture, required to focus the laser beam. However, for near-UV light at 350 nm, only the centre of the mirror (4 mm of diameter) should be used to focus the laser beam on a spot size of  $2\ \mu\text{m}$  (limited by the numerical aperture), otherwise, spherical aberrations become too large and induce the formation of a caustic. On the contrary, the large aperture may become useful when the spherical mirror is used as a “condenser” to collect the light emitted by the sample.

This easy-to-use optical system is highly polyvalent, and in this presentation, we will report on two applications working with light pulses from the two ends of the optical spectrum, i.e. the deep UV at a wavelength of  $\lambda = 260\text{nm}$  and the extreme infrared at a wavelength of  $\lambda = 300\ \mu\text{m}$ , in the terahertz (THz) domain.

Thanks to the high focusing of the spherical mirror, the 3<sup>rd</sup> harmonic of a Titanium:Sapphire laser oscillator at 260nm, having an energy of  $\sim 2\text{nJ}$  and a tuneable repetition rate ranging from 10kHz to 4MHz, is used to trigger the evaporation of APT samples, such as large band-gap materials (GaN, ZnO) or optical structures (quantum dot, quantum well, colour centre). These samples are selected according to their ability to reemit light. Thanks to the high repetition rate, this photoluminescence (PL) signal is high enough to be collected by the spherical mirror (acting as a condenser) before being directed to an optical spectrometer in order to be analysed. We will present the results obtained on different samples using this new instrument, the Photonic Atom Probe (PAP) [2]. We will highlight that the high voltage applied to diamond nanoneedles can significantly modify the PL emission of the colour centres, allowing for the

measurement of the stress introduced by the high voltage. Furthermore, the modification of the optical signature of a multi-quantum well during APT analysis, allows for improving the optical in-depth resolution up to 20nm, which means that it becomes possible to discriminate the optical signatures of two neighbouring quantum wells.

We will also discuss our recent development coupling the APT equipped with a spherical mirror to a monocycle THz generator (0.1-10THz,  $\lambda = 3\text{mm}-30\mu\text{m}$ ). The spherical mirror is in this case very useful to focus such a large wavelength on a “small” spot size (200-400 $\mu\text{m}$ ). We show that this light can trigger field evaporation without heating the tip because the THz optical electric field adds to the static electric field in order to trigger the field evaporation of the sample [3].



**Figure 1.** Principle of the versatile APT using a spherical in axis mirror. An incoming pulse (a) (from deep UV to far infrared) is steered by a half transparent plate or a dichroic mirror (b) to spherical in axis mirror (d) through the sample inside of the APT chamber (c). The positioning of the sample is performed by the means of an accurate piezo stage (positioning resolution  $\sim 1\text{-}10\text{nm}$ ) (e). The same mirror performs the imaging of the shadow of the sample and the focusing of the spot at the tip apex. The collected light can be directed to a camera (f) or to a spectrometer (g)

## References

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- [3] A. Vella, *et al*, Sci. Adv. **7**, eabd7259 (2021).