A Coherent White Paper

Turnkey Femtosecond Amplifiers Enable Cutting-edge Applications

The superior beam quality and long-term stability of the Astrella regenerative amplifier, as well as the intrinsic flexibility of the Monaco ytterbium fiber amplifier, make them ideal turnkey engines for experiments in extreme operating regimes that formerly required complex sources and considerable laser expertise. This whitepaper explains how these industrial-grade ultrafast amplifiers can be used to reach pulse widths shorter than 5 fs, perform broadband two-dimensional spectroscopy scans lasting 48 hours, and even enable THz-driven nanoscopy with controlled carrier-to-envelope phase relationship.

Titanium:Sapphire Regenerative Amplifiers

While newer laser gain materials like ytterbium-doped fibers are increasingly used for applications in femtosecond science, the uniquely wide bandwidth and gain characteristics of titanium:sapphire (Ti:Sapphire) give this well-proven technology an edge for applications requiring very high pulse energies and/or extremely short pulse widths at the most popular repetition rates of 1-5 kHz. The best laser architecture for fulfilling these performances is based on Chirped Pulse Amplification (CPA) with a regenerative amplifier. Here the output of a ~80 MHz Ti:Sapphire femtosecond oscillator is stretched (chirped) to tens of picoseconds, then dropped to the kilohertz domain by a fast optical gate before being amplified in a single or multi-stage amplifier pumped by a Q-switched green laser. The amplified pulse – a replica of the input pulse – is then compressed to the initial pulse duration. Vertically integrated laser manufacturers like Coherent offer all these components – oscillator, amplifier, pump lasers – as separate devices that can be combined to achieve the desired performances. In order to make amplified femtosecond pulses available to the broadest possible user-base, we also provide “one-box” amplifiers where all these components are integrated inside a single, robust laser head.

Until recently, there was a marked trade-off in commercial Ti:Sapphire systems between complexity and performance. Open architecture systems enabled access to the shortest pulse widths and highest pulse energies, whereas one-box integrated amplifiers provided much simpler – often push-button – ease of use, at the expense of achieving cutting-edge performance. This has now changed with next-generation integrated amplifiers such as the Coherent Astrella series, where push-button performance up to 9 mJ per pulse at 800 nm with < 35 fs pulse width at a repetition-rate of 1 kHz greatly decreased the gap with more complex multi-box amplifiers. In conjunction with sophisticated accessories, Astrella now provides turnkey access to operating regimes that were formerly only available in a handful of specialist laser labs.

Ytterbium Fiber Amplifiers

While Ytterbium amplifiers cannot support the bandwidth or the energy per pulse delivered by Ti:Sapphire amplifiers, they provide much higher average power in a more compact package. With the selection of fibers as gain medium, Yb amplifiers achieve unprecedented flexibility in repetition rate by maintaining a constant average power over an extremely broad operating range. For example, Coherent Monaco produces up to 60 watts of average power between 750 kHz and 50 MHz repetition rate; below 750 kHz the energy per pulse is locked to 80 microjoule and the repetition rate can be arbitrarily down-selected to single shot. A motorized compressor enables pulse duration adjustment from <300 fs to 10 ps and all these parameters can be controlled with a laptop computer. Monaco's architecture includes several fiber amplification stages and two integrated acousto-optic modulators (AOM) that select the pulses to be amplified in the final fiber amplification stage and accurately control the energy per pulse (and average power). In addition, the fiber approach enables operation in "burst mode" where several pulses are produced at 50 MHz repetition rate, each one with the maximum pulse energy of 80 microjoule.

Industrial Simplicity and Reliability

Performance in a compact package is only part of the requirements needed to increase the adoption of ultrafast amplifiers; Astrella also combines ease of use with reliability and long-term stability. This rugged reliability/stability is the result of a program that Coherent calls the *Industrial Revolution in Ultrafast Science*. This encompasses design methodologies, materials qualification and sourcing, plus HALT/HASS testing protocols. In HALT (High Accelerated Life Testing), prototypes are iteratively tested to failure, re-designed and retested to eliminate any inherent weaknesses. HASS (Highly Accelerated Stress Screening) stresses production units beyond their specified operating environment before shipping to users. This process screens out any variances in manufacturing, packaging, etc. Figure 1 shows an Astrella loaded into our custom HALT/HASS testing chamber.

The reliability and stability resulting from these design and manufacturing protocols, together with a very compact size, enable advanced applications to spectroscopy, attosecond science, and terahertz nano-spectroscopy that we will describe in the following sections

Two-Dimensional Spectroscopy

Some of the most demanding applications for ultrafast amplifiers are the various embodiments of two-dimensional spectroscopy, which has become increasingly popular in the last decade. In conventional spectroscopy, the light signal (IR absorption, Raman scattering, etc.) is recorded as a function of the excitation wavelength. Here laser pulses with as large a bandwidth as possible are used to determine how strongly different molecular vibration or electronic levels are coupled, as well as the dephasing time of these couplings. The data is usually graphed as two-dimensional contours as shown in figure 2. The shape of the vibration contours also provides information to enable the homogenous and heterogeneous components of the excited state lifetimes to be independently determined.

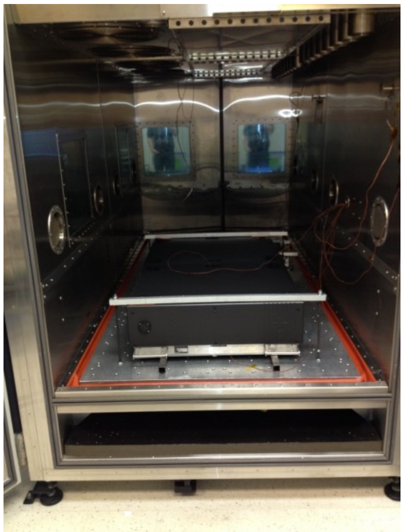

Figure 1: *HALT/HASS testing and screening are key factors enabling industrial reliability in Astrella amplifiers.*

Although the 2D spectroscopy concept is easiest to understand when plotted in the frequency domain, in most experiments the data is acquired as Fourier transforms in the time domain. This is accomplished using broadband pulses from femtosecond sources to simultaneously span the frequency region of interest. Here a single broadband source and a pulse shaper generate a pulse sequence. Timing between two of the closely synchronized pulses is transformed into the frequency domain and scanning the timing between the two pairs enables the dephasing lifetime to be determined, which is sometimes called 3D data.

For a more detailed explanation of how this works, and to see the full Xiong whitepaper, visit: https://edge.coherent.com/assets/pdf/SurfaceCatalystStructure_AstrellaProfXiong2D_Case_Study_FINAL.pdf

Professor Wei Xiong's group at the University of California, San Diego, is using 2D spectroscopy to examine how a heterogenous catalyst - Re(diCN-bpy)$(CO)_3Cl$ - is bound to a gold surface and how this binding affects its dynamics (see figure 2). This chemical is a CO_2 reduction catalyst and is thus a candidate for use in sustainable energy schemes.

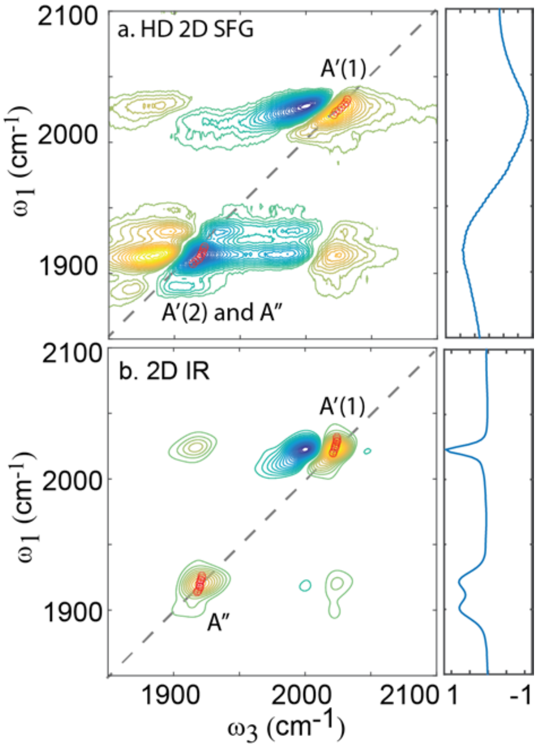

Figure 2: *Typical 2D data set contour plot. In this case the 2D SFG is compared with a standard 2DIR spectrum for the same sample*

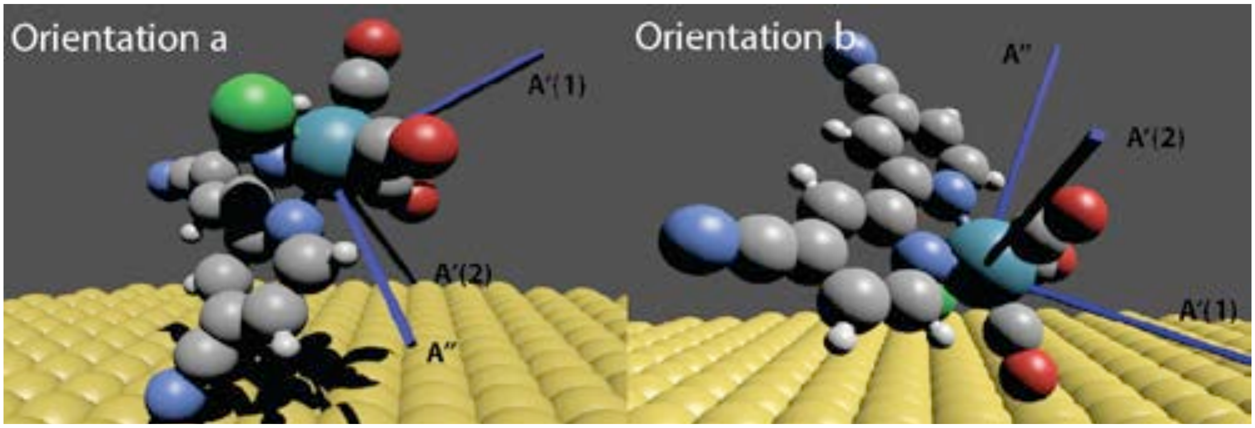

Figure 3: *Orientations of **the catalyst** on gold surface determined from HD 2D SFG data and data from reflection mode IR spectroscopy, simulated using Density Functional Theory (DFT). Orientation "**a**" is preferred. Blue sticks represent the vibrational modes direction. Inset shows 2D data plots from this study – 2D SFG data for the surface bound catalyst and 2D IR data showing the same vibrational information for the catalyst in solution.*

Xiong's teams are performing experiments based on sum frequency generation (SFG), a technique originally developed by Xiong as a graduate student in the lab of Martin Zanni. 2D SFG is ideal for studying surface-bound catalysts because SFG vibrational signals are only generated at surfaces and phase interfaces. This largely eliminates potentially huge background noise due to unbound (in solution) catalyst molecules. However, because the catalyst is bound as a monolayer, the SFG signal itself is extremely weak. And since the signal has non-linear dependence on laser intensity, high pulse energies and short pulse widths are absolutely essential; this is why Xiong selected Astrella for this work. The other reasons he cites for investing in Astrella as the lab's main ultrafast source are its ease of use and long-term stability. "To obtain a full set of spectra at different delay times – 3D data – we sometimes have to average data for 48 hours, which puts extreme demands on laser stability. During this period, it is critical that the amplifier output be stable and without drifts in beam pointing, beam quality, pulse energy, etc. The stability of Astrella means we can do these long data runs while remotely controlling the laser from an office near the laboratory." Xiong's group have used this setup to determine the specific orientation of the catalyst on the gold surface (see figure 3) and the effect of surface binding on the dynamic coupling between key vibrations.

Simple Access to High Energy sub-5-fs Pulses

Astrella provides turnkey access to pulse widths shorter than 35 fs with pulse energies in excess of 9 mJ. Yet, several important emerging applications in physics, photochemistry and materials science need even shorter pulses and/or higher peak powers – for example to generate attosecond and Extreme UV pulses, or to create bursts of relativistic electrons. Recently Coherent collaborated with Professor John Tisch and Dr. Daniel Walke from Imperial College of London, and scientists from Sphere Ultrafast Photonics to exploit the turnkey simplicity and stable beam quality from an Astrella amplifier to reach 5 fs pulse widths with pulse energies as high as 2 mJ. As shown in figure 4, a key element of this setup was a differentially pumped hollow fiber compressor (HFC) developed by Tisch's group that was used to generate the ultrashort pulses.

This approach takes advantage of the spectral broadening caused by self-phase modulation (SPM) in a hollow fiber containing a noble gas. The fiber acts as a dielectric waveguide, confining the beam and allowing for a long interaction length at a high intensity. This established approach is proven to allow the generation of high-power (up to 5 mJ), few-cycle laser pulses at kHz repetition rates.

A key innovation here is to differentially pump the HFC. As pioneered by Tisch and others, differential pumping reduces plasma formation at the fiber entrance where the laser intensity is highest. (In a statically gas-filled hollow fiber, plasma formation at the input side would otherwise cause a reduction in both coupling efficiency and shot to shot stability, by altering the size and position of the focus at the entrance from its optimum.) The Astrella pulses were focused by a 1 meter focal length lens to a beam waist of ~160 µm at the HFC input. The system ran repeatedly for many hours at a time, without any active feedback or re-alignment by the users, due to the high stability input beam from the Astrella amplifier.

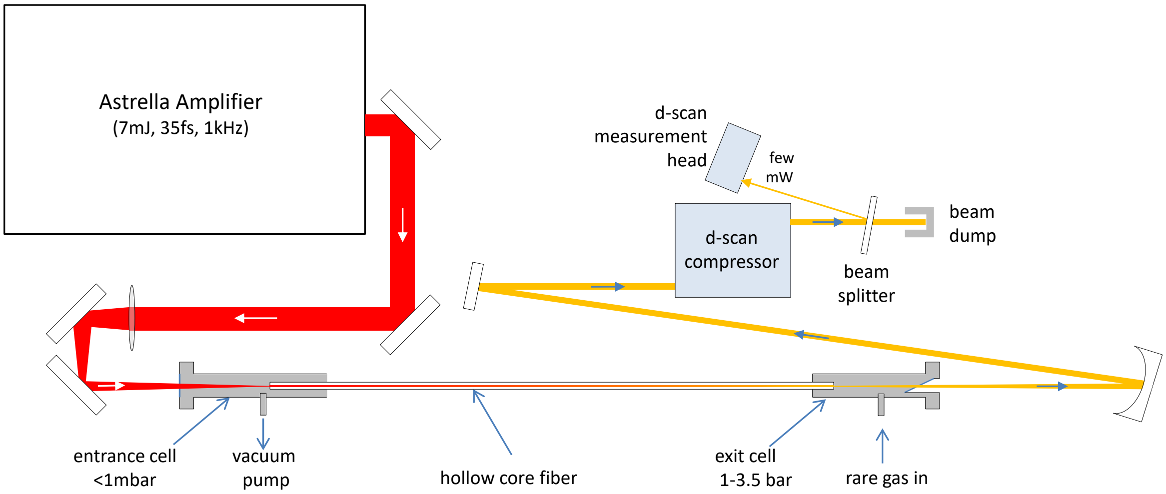

__Figure 4:__ Experimental set up for 5 fs pulse generation and measurement. The output of a Coherent Astrella amplifier is focused by a lens (f=1 m) into a 250 µm inner-diameter, differentially-pumped hollow core fiber, pressurized with either neon or helium gas. The pulse energy from the Astrella is controlled over the range 0-7 mJ with a waveplate-polarizer combination (not shown). The spectrally-broadened output from the hollow core fiber is re-collimated by a concave silver mirror (f=0.75 m) before it is both compressed and measured by the d-scan blue system. Only a few mW of average power is required for the d-scan measurement head, so beam splitters are used to sample the watt-level (~1 mJ @ 1 kHz) beam from the hollow fiber. The beam entering the beam dump would, in general, be available for experiments.

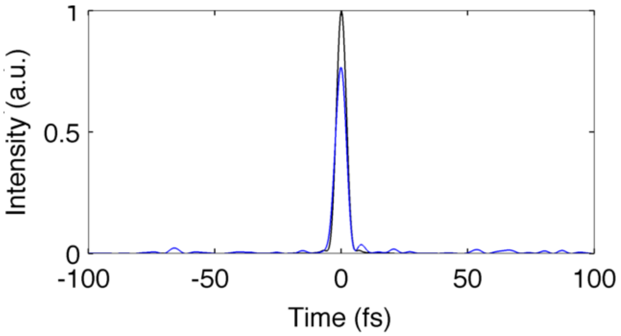

__Figure 5:__ Typical d-scan output data in the time domain. In this case, both the Fourier Transform limited pulse and actual retrieved pulse, revealing a duration of 5.1 fs FWHM.

There are several approaches to characterize various aspects of femtosecond pulses but the d-scan unit developed by Sphere and used in this demonstration offers a number of advantages, including its ability to measure and compress pulses in the few-cycle regime down to single-cycle pulses. The overall ease and speed of use makes d-scan the perfect tool for HCF measurement and optimization. First, it can perform both the compression/control and temporal measurement in a single unit. Second, it is a robust self-contained tool that is very tolerant of input beam misalignment (even ± a few degrees) and therefore quick to set up. Third, it is fast, providing a complete pulse characterization (phase and amplitude) in less than 1 minute for kilohertz pulse repetition rates.

As shown in the data plot in figure 5, this compact and relatively simple setup provides turnkey access to pulse widths of 5 fs with pulse energies at the millijoule level.

Extreme Stability of Monaco is Instrumental in Supporting Cutting-Edge Nanoscale Metrology

Prof. Hidemi Shigekawa of the Institute of Applied Physics at the University of Tsukuba, Japan, recently led a study on Sub-cycle Transient Scanning Tunneling Spectroscopy (STM) with Visualization of Enhanced Terahertz Near Field.

This technique replaces the DC field of conventional STM with the electric field of THz radiation. One key advantage is the capability to obtain Carrier-Envelope Phase (CEP) stable THz "light" consisting of nearly single-cycle THz pulses generated via optical rectification—that is the process of sending broadband optical pulses into a non-linear crystals such as $LiNbO_3$.

When combined with STM, controlling the CEP enables control over the direction of the transient electric field applied between the tip and sample. In addition, when a nanoscale metal structure is irradiated by electromagnetic waves, it becomes possible to obtain an intense terahertz near-field in a local region directly beneath the nanotip. Furthermore, by the manipulation of THz pulses with controlled CEP, it is possible to apply both a positive and negative electric voltage between the tip and sample. This results in a transient tunnel current between the two, making it possible to interrogate the localized region directly beneath the nanotip with the spatial resolution of an STM and enabling THz-driven nanoscopy. Because the applied voltage between the tip and sample is on the femtosecond timescale, dynamic effects that can be induced and/or interrogated by light can be studied on this same timescale.

Methodology

The pump and probe experiment set-up combines an Yb femtosecond fiber amplifier (Coherent Monaco) and an ultrahigh-vacuum STM system, which supports precise manipulation of the nanotip position. Electric field enhancement was achieved with a Platinum-Iridium-coated Tungsten STM nanotip. The nanotip was irradiated with two types of optical pulses: a THz pulse (generated by irradiating a $LiNbO_3$ crystal with an IR pulse) and an optical domain pulse (generated in a BBO crystal).

Experimental Setup

Figure 6 shows the system configuration with THz-STM system and the Monaco laser (40 watts at 1035 nm, Pulse width < 350 fs).

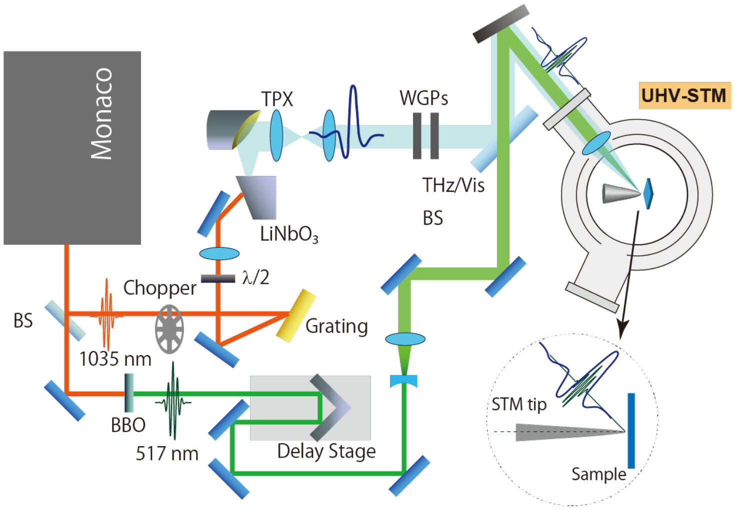

Figure 6: *System configuration*

Figure 7a shows time-resolved measurements performed with the THz-STM system. The specific sample material - 2H-$MoTe_2$ – has similar properties to a semiconductor; its STM image in Figure 7a shows the intrinsic defects (bright spots), here resolved at atomic level.

Figure 7b is a chart of THz-based measurement result. The delay time td < 0 indicates that the THz beam reaches the sample after the optical beam excites it and creates hot photoelectrons. The rectified current reflects the dynamics of the excited state due to photocarrier generation. For more details, refer to the published article [1].

Why the Coherent Monaco Laser

The most important laser requirements for these studies were high repetition rate and power. To efficiently generate THz radiation, high pulse energy is necessary while a high repetition rate enables accurate measurements in a short time. Monaco meets these needs perfectly.

The Monaco maintains its high stability over a wide range of operating conditions. After upgrading a 1 MHz model of Monaco to a 50 MHz repetition rate and adjusting several system components to be compatible with the higher power, the experimental set-up was operated at full power (40 W) and supported the higher repetition rate while maintaining the required stability.

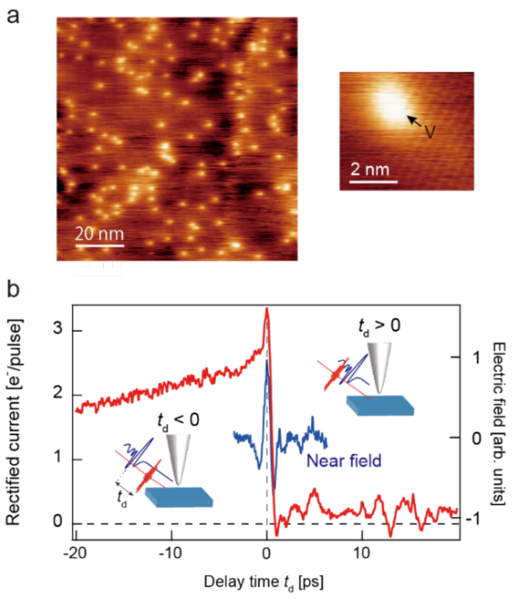

Figure 7: *Transient electronic dynamics obtained by THz-STM for 2H-MoTe$_2$.*
(a) STM image of the sample. (b) Delay-time dependence of 1 THz

Summary

Coherent pulses featuring extreme stability, broad bandwidth, and/or ultrashort (5 fs) pulse widths are required to support sophisticated experiments where the complexity of the available sources confined their use to laboratories with considerable laser hands-on expertise, precluding widespread applications. The availability of turnkey amplifiers like Astrella and Monaco, provides a simpler path to advanced femtosecond performance, widening the access to applications from attosecond physics to cutting-edge nanoscale metrology.

References

[1] ***Sub-cycle transient scanning tunneling spectroscopy with visualization of enhanced terahertz near-field***. Shoji Yoshida, Hideki Hirori, Takehiro Tachizaki, Katsumasa Yoshioka, Yusuke Arashida,Zi-Han Wang, Yasuyuki Sanari, Osamu Takeuchi, Yoshihiko Kanemitsu, and Hidemi Shigekawa, ACS Photonics, 6, 1356-1364 (2019).