

Prof. Ivan Ivanov - Linköping University

Customer Review – Raman Application

20th of February 2024

We purchased the 349NX laser from Skylark Lasers, a single-frequency ultraviolet (UV) laser with emission at 349 nm and 100 mW output power. Our motivation to purchase the laser was to replace an old argon laser (Innova 90), which was no longer producing adequate power. The laser was used to provide above-bandgap excitation for 4H-SiC (bandgap around 3.26 eV). **The 349 nm excitation (3.55 eV) is an excellent replacement for an Argon laser at 351 nm.**

The process of ordering the Skylark DPSS laser was very easy, the team were able to answer all my pre-sales questions and delivery was on time and the laser was well packaged.

While photoluminescence excitation does not necessarily require a single frequency laser, we also wish to use the laser for Raman spectroscopy. Raman does require single-frequency excitation, or at least laser line linewidth much smaller than the linewidth of any Raman peaks. In practice this requirement translates to linewidth of the exciting laser less than, say, 0.1 Å. The specified linewidth of the 349NX laser is 500 kHz (about 2 nanoelectronvolts, $2 \cdot 10^{-9}$ eV), which corresponds to $2 \cdot 10^{-6}$ Å at 349 nm. The long coherence length (> 100 m) resulting from this narrow linewidth can also be beneficial for other applications. Gas lasers can have linewidths up to 1 GHz (e.g., corresponding to 0.013 Å for He-Ne laser at 633 nm), which is still sufficiently narrow for Raman spectroscopy. However, **the Skylark 349NX has some significant advantages compared to a gas laser.**

Firstly, **the emission of the 349NX is spectrally pure** except, possibly, for some weak disturbances in the immediate vicinity of the laser line (see the report on the measured Raman spectra in Fig. 1). On the contrary, the emissions of gas lasers contain multiple plasma lines resulting from different non-lasing atomic (ion) transitions activated by the discharge in the active gas media which, if not filtered out, completely overwhelm the measured spectrum.

Secondly, **the efficiency of Skylark DPSS lasers is much better than that of gas lasers.** By efficiency we mean the ratio between the output optical power from the laser and the input electrical power; this ratio is much higher for the solid-state lasers (SSLs) in comparison with gas lasers. The reduced power consumed by SSLs leads to relaxed cooling requirements and much more compact design of the SSLs. In particular, our 349NX uses closed water-circuit chiller and the whole laser system (laser head + power supply + chiller) is easily portable between different labs (even more compact air-cooled option is also available). **Portability is an additional benefit** for our laboratories as our Raman and photoluminescence (PL) setups are in different labs.

Thirdly, the stability and RMS parameters of the Skylark 349NX and gas lasers are comparable but the **Skylark DPSS laser provides better longevity with cheaper maintenance.** If a gas laser fails one must change the expensive laser tube, the price of which is often half of the price of the entire laser. The repair of a failed DPSS laser can be cheaper if only the pumping diode(s) or a frequency-doubling crystal need to be changed.

Raman Tests

The first test we performed was to replace our 532 nm laser beam in a home-built micro-Raman system with the 349 nm from 349NX. We also replaced the dichroic mirror in front of the monochromator (Jobin Yvon, HR460) with a razor edge filter with cutoff wavelength at 355-nm from Semrock (LP02-355RE-25).

Optical elements such the objective, mirrors, and beam splitter, reduced the optical power to < 2 mW at the entrance pupil of the objective, which together with the Raman signal is further cut down by the objective (estimated transmittance ~30 % in the range 350 – 355 nm). Nevertheless, **using the Skylark 349NX we obtained clear Raman spectra from both 4H and 6H SiC**, as displayed in Fig. 1, including the second-order Raman bands.

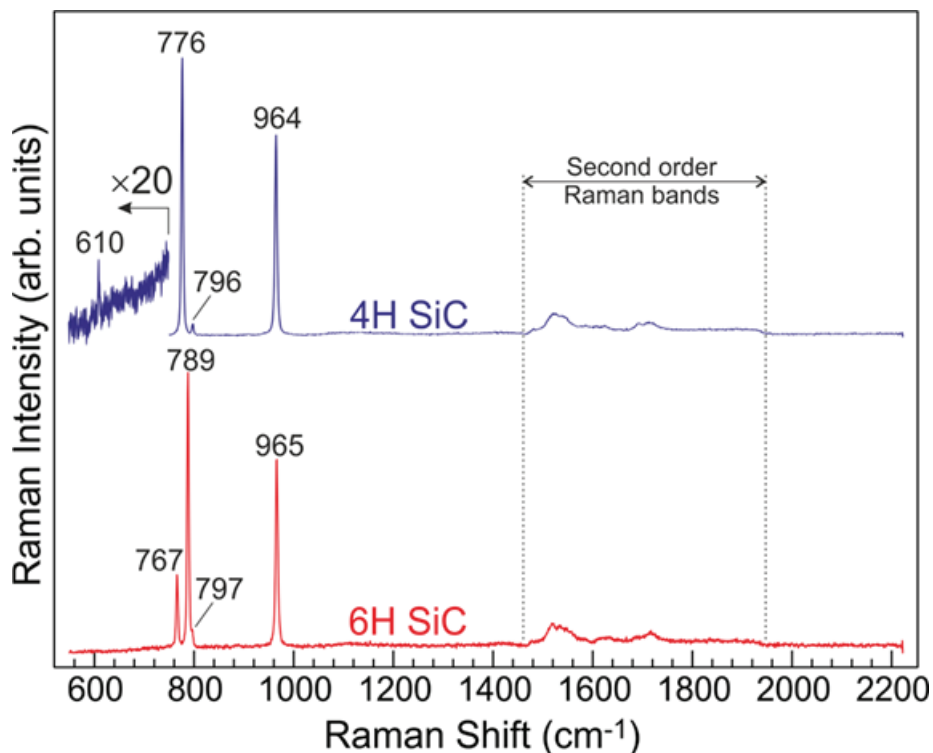


Fig. 1. Raman spectra of 4H and 6H SiC obtained in a micro-Raman setup with the Skylark 349NX laser. The dichroic mirror (long pass filter) at 355 nm intended to cut the laser also cuts the low-frequency part of the Raman spectrum (up to 520 – 550 cm⁻¹). Note the scale change in the 4H spectrum revealing the weak mode at 610 cm⁻¹.

We notice that the dichroic mirror used is not optimum for Raman measurement since the cutoff at 355 nm corresponds to a Raman shift of ~500 cm⁻¹ from the laser line at 348.85 nm (as estimated by the obtained Raman spectra). Hence, the spectrum below ~520 cm⁻¹ is cut down by the DM. However, micro-Raman measurements with the Skylark 349NX using a single monochromator coupled to a CCD camera would be perfectly possible with an adapted optical setup for 349 nm.

We repeated the measurements on 4H and 6H SiC using a double monochromator (SPEX 1404) equipped with a GaAs photomultiplier tube (PMT) and 1800 g/mm holographic gratings. Macroscopic UV lenses are used for collecting the signal and focusing the laser on the sample and the corresponding spectra are displayed in Fig. 2. In addition to the higher resolution offered by this system, the setup **using the Skylark DPSS also has the advantage that no filtering of the laser line is necessary**, hence the whole laser power is available for exciting the spectrum if needed, and the experimental setup is simpler and more flexible than if a filter-monochromator had to be used (e.g., to filter the plasma lines of a gas laser).

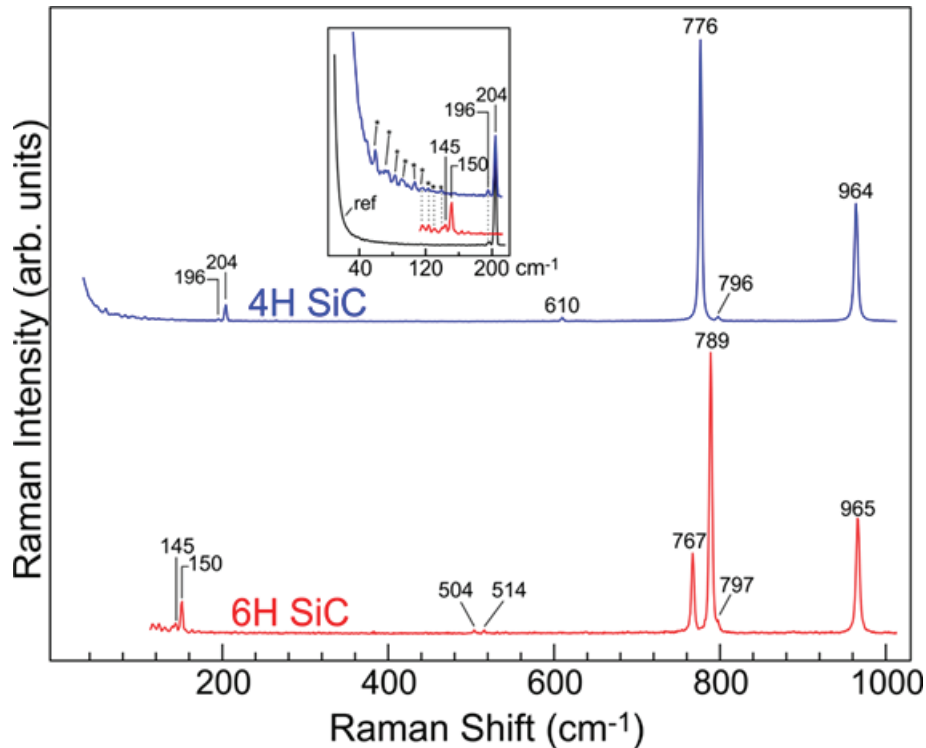


Fig. 2. Raman spectra of 4H and 6H SiC obtained with double monochromator without use of any filter in the excitation and receiving path. The inset is a close-up of the low-frequency region, where artefacts due to the excitation laser are seen (denoted with asterisks). The lowest (black) curve in the inset denoted 'ref' is obtained with a different laser (at 381.5 nm) and given as a reference of artefact-free Raileigh wing. The spectra illustrate also that there are no emission artefacts at Raman shifts $> \sim 160$ cm^{-1} .

The laser is directly guided to a lens focusing the beam on the sample, i.e., without any filtering. As expected, the spectrum in the region > 155 cm^{-1} is free from artefacts (no plasma lines). However, in the region of the Rayleigh wing (< 155 cm^{-1}) one can see some weak emissions (forming kind of broader lines) which do not originate from the sample and are due to the laser. These are demarked with asterisks on the inset image in Fig. 2 which shows that their intensity tends to be stronger the closer the features are to the laser line. However, it should be noted that such weak artefacts will not interfere with a Raman spectrum measured in a Raman setup with a razor-edge filter because the usual cutoff of such filters is about 150 – 180 cm^{-1} from the laser line they are designed for. Also, in the range below ~ 150 cm^{-1} these artefacts have **much lower intensity than plasma lines in the vicinity of a laser line from a gas laser**, which are difficult to filter away even with the use of a quality filter-monochromator.

Photoluminescence measurement

We have measured one spectrum from a 4H SiC sample in the region of the so-called AB lines, the spectrum for which is displayed in Fig. 3. The Skylark 349NX needs no filtering which, as already mentioned, greatly simplifies the experimental setup. The spectrum is free of laser-related artefacts and contains only the useful lines from the sample, a consequence of the spectral purity of the laser emission. For comparison, an unfiltered Ar laser at 351 nm would produce a forest of plasma lines, overwhelming the original spectrum.

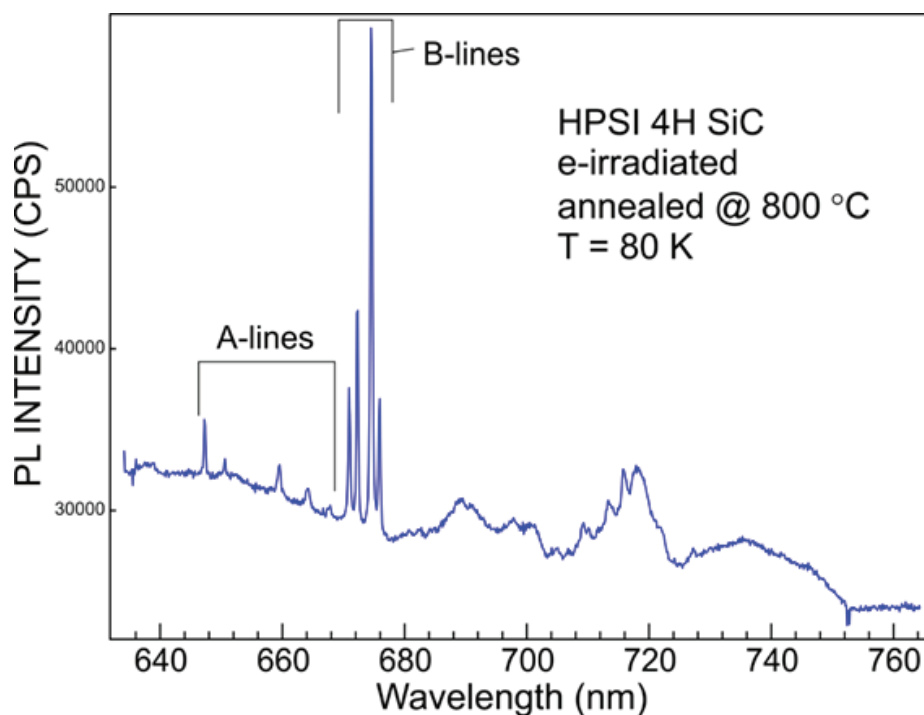


Fig. 3. PL spectrum of a 4H SiC in the region of the so-called AB lines (640 – 760) nm obtained with unfiltered laser at 349 nm. **All features in the spectrum are related to the samples, no artefacts from the Skylark laser emission are observable in the spectrum.** The spectrum is recorded using a single monochromator (Jobin Yvon, Triax) equipped with thermoelectrically cooled CCD camera.

Conclusion

The laser is easy to use, the startup time is comparable to that of an Ar ion laser (a couple of minutes to start, ~5 minutes to stabilize). **The wavelength of the Skylark 349 NX is perfect for my experiments on PL and Raman spectroscopy**, the shape of the beam is almost Gaussian and seems to be focused close to the diffraction limit through a microscope objective.

The Skylark laser is an excellent excitation source for Raman spectroscopy, comparable to an argon laser but with some advantages. The laser emission of the 349NX is spectrally clean unlike that from the Argon ion laser. Due to the presence of ubiquitous plasma lines the emission of the latter requires an additional filter-monochromator to filter out these parasitic lines.

The power of the Skylark DPSS laser is expected to remain constant over the lifetime which was the main driver for replacing our old argon laser. An additional unexpected benefit is that **the Skylark system is much smaller than our previous laser** making it portable between labs and experimental setups, hence, providing additional flexibility to our experiments.



If you would like further information regarding Skylark Lasers and how our lasers can support your application please contact:

Dr Lauren Fleming, Technical Specialist email: lauren.fleming@skylarklasers.com