Summary of the PhD thesis of Nathalie Vermeulen
Department of Applied Physics and Photonics (IR-TONA)
Vrije Universiteit Brussel

Modeling continuous-wave Raman lasers:
towards an intrinsic heat mitigation mechanism

The invention of the laser in 1960 has been a very important milestone for our present-day society. This remarkable new light source not only triggered a revolution in science and technology but also gave rise to a significant improvement of the quality of our daily life. An important type of laser is the so-called Raman laser, the lasing mechanism of which is based on Stimulated Stokes Raman Scattering (SSRS). Some remarkable Raman laser examples are the hydrogen-based Raman laser and the revolutionary silicon-based Raman laser, both of which have a myriad of application potentialities e.g. in atmospheric spectroscopy, remote sensing, and laser medicine.

In our thesis, we present our original research contributions in three different 'previously unexplored' domains of Raman laser research. First, considering a working point at exact Raman resonance, we reveal **the physical nature and behavior of Coherent Anti-Stokes Raman Scattering (CARS)**, a four-wave mixing process which often accompanies the SSRS lasing process in continuous-wave Raman lasers and which is also applied in a variety of optical analysis techniques. Secondly, we present **a novel powerful modeling method for continuous-wave Raman lasers** which outperforms the previously used modeling methods. Thirdly, we introduce **a novel CARS-based approach to intrinsically mitigate the heat dissipated in Raman lasers** caused by the pump-Stokes quantum defect.

Our first original contribution is the derivation of an adequate and full interpretation of CARS at exact Raman resonance in which we explain how the involved pump, Stokes and anti-Stokes photons interact with each other for a given level of pump depletion and for a given phase mismatch. We start from the basic classical Raman formalism and apply it to a single-pass Raman converter. Using this formalism, we find that CARS at exact Raman resonance behaves in a completely different way than most researchers have assumed so far. Indeed, the most commonly used model for CARS is the model introduced by Terhune in 1963, where CARS is thought of as a process that simultaneously generates Stokes and anti-Stokes photons without exchanging energy with the Raman medium. We, however, find that depending on the phase mismatch value and on the level of pump depletion CARS either converts Stokes photons to anti-Stokes photons while annihilating phonons or converts anti-Stokes photons to Stokes photons while creating phonons. We also discover that, whatever the value of the phase mismatch, the CARS process always generates anti-Stokes photons and therefore annihilates phonons in the Raman medium in the initial part of the propagation path. Furthermore, our interpretation of CARS supports different experimental findings, which were seemingly contradictory in the framework of the CARS theory introduced by Terhune.

We can thus conclude that our new CARS model not only deepens the fundamental understanding of this important Raman mechanism, but will also be a useful tool for further optimizing existing CARS-based (analysis) techniques. In addition, it might even inspire researchers to develop new CARS applications that have not been considered before. Specifically in the context of our thesis, we have gained with this new CARS model more physical insight in the way anti-Stokes radiation is generated in Raman lasers, thereby realizing that CARS could be used for intrinsically mitigating the heat dissipation in Raman lasers.

Our second original contribution is the development of a widely applicable and easily implementable modeling method for describing the Stokes and anti-Stokes emission of continuous-wave Raman lasers. To construct the equations of this so-called "Stokes–anti-Stokes iterative resonator method" (IRM), we again rely on the basic Raman formalism. When comparing our Stokes–anti-Stokes IRM with the rate equation method previously used for modeling conventional hydrogen-based Raman lasers, we find that our IRM is superior with respect to the implementation of the longitudinal field distributions inside the cavity and the incorporation of the differences in scattering direction of the different Raman processes. When making the comparison with the power distribution method previously used for modeling near-infrared silicon-based lasers, we conclude that our IRM is the only modeling method that: correctly incorporates the interference effects between the incident and intra-cavity pump waves; is able to provide us with data on the anti-Stokes emission; and has the capability of describing the lasers' transient characteristics. What is more, we find that our IRM is also applicable to the newly developed hydrogen Raman lasers based on a hydrogen-filled hollow-core photonic crystal fiber and to the forthcoming mid-infrared silicon Raman lasers. We remark that for the latter our modeling method predicts a high lasing efficiency. Thus, our Stokes–anti-Stokes IRM is a very powerful tool not only for studying and optimizing existing continuous-wave Raman lasers but also for designing future Raman laser types and for predicting their laser performance. In the context of our thesis, our modeling method has also proven to be very useful for investigating the feasibility of our CARS-based heat mitigation technique for Raman lasers.

Our third original contribution is the introduction of a novel technique that intrinsically mitigates the quantum-defect heating in Raman lasers by the use of CARS. This so-called "CARS-based heat mitigation" relies on the principle that increasing the ratio of the number of out-coupled anti-Stokes photons to the number of out-coupled Stokes photons causes a decrease of the average quantum-defect heating per out-coupled Stokes lasing photon. To obtain effective heat mitigation, it is first of all important to suppress as efficiently as possible heat generating mechanisms other than the quantum defect. Secondly, we find that the CARS-based heat mitigation efficiency can be enhanced by achieving (quasi-)perfect phase matching for the four-wave mixing process of CARS. Thirdly, one can reduce the "absorption" of anti-Stokes photons through Stimulated Anti-Stokes Raman Scattering or SARS (another process that often occurs in continuous-wave Raman lasers) by lowering the gain of backward Raman scattering. After detailed investigation, we find that all three effectiveness-enhancing methods can be more easily applied to a mid-infrared silicon Raman laser and to a Raman laser based on a hydrogen-

filled hollow-core photonic crystal fiber than to a near-infrared silicon Raman laser. As a result, using our Stokes–anti-Stokes IRM we obtain approximately the same CARS-based heat mitigation efficiencies for the two former laser categories (namely 35% and 30%, respectively) and a much smaller CARS-based heat mitigation efficiency for the latter laser type (namely 15%). The fact that heat mitigation efficiencies of as much as 30% and even higher are predicted for two very promising types of Raman lasers demonstrates the viability of our CARS-based heat mitigation technique. Taking also into account that as a by-product of our heat mitigation mechanism the Stokes-lasing Raman lasers also generate significant anti-Stokes emission, we find that with our CARS-based heat mitigation technique important progress could be made in the development of dual-wavelength, compact lasers with undoubtedly perspectives for numerous novel applications.

With our PhD work we believe to have contributed to the further development and exploitation of Raman lasers in general and of hydrogen-based and silicon-based Raman lasers in particular. We hope that our research contributions will open new pathways for original research and technology developments, and that they will as such lead to the realization of advanced Raman-based devices which can open up new horizons, for example in the domain of silicon photonics.