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Effects of Refractive Index Mismatch on Stimulated Raman Scattering And Coherent Anti-Stokes Raman Scattering Microscopy

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Nonlinear optical microscopy techniques, such as stimulated Raman scattering (SRS) and coherent anti-Stokes Raman scattering (CARS), allow for label-free chemically-sensitive non-destructive video-rate imaging of biological processes[1]. SRS is of particular interest due to its improved image contrast, high spectral sensitivity and low acquisition times.

Correctly interpreting images produced by nonlinear optical processes is of vital importance. Earlier we showed AM-SRS signals depend upon the structure of $\chi^{(3)}$ in the background medium, and thus is not background free[2]. We now show that even for the modest linear refractive index mismatches typically found in biological tissues, near-field enhancements can cause significant signal distortions in both CARS[3] and SRS.

We employ finite-difference time-domain simulations to determine the near- and far-fields of wavelength-sized spherical Raman-active objects in a nonresonant Kerr medium illuminated by a tightly-focused laser source. We find that, depending upon the shape of the Raman scatterer, enhanced near-fields can create a signal an order of magnitude larger than what would be expected, and with a peak in the image that does not directly correspond to the object location. Additionally, the radiation pattern is heavily influenced and as a consequence we find that the numerical aperture of the collecting lens becomes important. Filtering techniques will not eliminate any of these effects as these distortions are caused by a microlensing effect within the scatterers. Understanding these distortions is key to correctly interpreting both CARS and SRS images.

Even without any Raman-active material present, the underlying $\chi^{(1)}$ structure can introduce background signals in AM-SRS and CARS. This highlights the need for frequency-based filtering methods such as FM-SRS and FM-CARS or hyperspectral analysis.

References

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2. K. Popov et al., **Opt. Lett.** 37, 473–475 (2012).
3. J. Lin et al., **Opt. Express** 17, 2423–2434 (2009).

Primary author: VAN DER KOLK, Jarno Nicolaas (University of Ottawa)

Co-authors: CALÀ LESINA, Antonino (University of Ottawa); RAMUNNO, Lora (University of Ottawa)

Presenter: VAN DER KOLK, Jarno Nicolaas (University of Ottawa)

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