Influence of the Raman depolarization ratio on far-field radiation patterns in coherent anti-Stokes Raman scattering (CARS) microscopy

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Among optical nonlinear techniques, coherent anti-Stokes Raman scattering (CARS) has revealed to be a powerful non-invasive tool in microscopy as it maps particular intramolecular vibrational modes of imaged objects. The interaction between the two exciting beams and the investigated medium is described by the latter’s third-order tensor $\chi^{(3)}$. For isotropic media, it is defined by only two independent components, mutually dependent on the Raman depolarization ratio $\rho_R$. In the context of microscopy, $\rho_R$ lies between 0 and 0.75 [1]. Phase-matching relaxation and good spatial resolution are provided by high numerical aperture microscope objectives.

Basing on the Richards and Wolf’s framework [2], but neglecting the exciting field’s axial component, far-field CARS radiation patterns under tight focusing excitation have been previously investigated [3]. In this work, all the exciting field’s components are taken into account so that the induced third-order nonlinear polarization (and in particular its axial component), responsible for CARS generation, is dependent on the parameter $\rho_R$ of the probed medium.

We have found that thin objects experience higher alteration of their CARS far-field radiation pattern, with the $\rho_R$ parameter, than bulk objects. In the extreme case of an infinitely thin plane object, the ratio of epi- to forward-emitted power exhibits variations of nearly 30% on the range [0; 0.75]. To complete this study, we have introduced the non-resonant part of the tensor $\chi^{(3)}$, for which $\rho_R$ is identified to 1/3.

This work is the first full-vectorial treatment of CARS far-field radiation pattern at our knowledge [4]. In most cases, the radiation pattern shows no significant modification with the Raman depolarization ratio, thus validating the previous studies. In the particular case of thin objects, careful attention must be observed. It is particularly true when imaging biological objects where a few nanometres thick objects as cellular membranes are encountered.


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