

## Coherent anti-Stokes Raman scattering microscopy (CARS) image contrast at interfaces: evidence of interference effects

David Gachet, Franck Billard, Nicolas Sandeau<sup>†</sup> and Hervé Rigneault  
Institut Fresnel, Mosaic group, UMR CNRS 6133, Domaine Universitaire Saint Jérôme,  
Université Paul Cézanne Aix-Marseille III, 13397 Marseille Cedex 20, France  
E-mail : [david.gachet@fresnel.fr](mailto:david.gachet@fresnel.fr), [herve.rigneault@fresnel.fr](mailto:herve.rigneault@fresnel.fr)

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Among the multiple nonlinear techniques (two-photon excited fluorescence, second and third harmonic generation) used in optical microscopy, coherent anti-Stokes Raman scattering (CARS) is unique as a coherent and resonant process. Its resonant and coherent nature is turned to account to obtain emission with molecular vibration selectivity and good directivity. A well-known feature of CARS is the presence of a non specific background (nonresonant contribution) in addition to the resonant contribution, both of them stemming respectively from the electronic and the vibrational response of the medium. The CARS field is dephased or not with the pump and Stokes exciting fields, whether it arises from the resonant or the nonresonant contribution. When spectrally scanning a CARS resonance, the dephasing is dependant on the vibrational resonance detuning [1]. The total CARS signal arises from the coherent summation of fields originating from the resonant and nonresonant contributions.

The interface between a “resonant” object and its “nonresonant” surrounding is expected to behave as a two-path interferometer when it is scanned in CARS microscopy. The dephasing between the two “paths” can be, in this case, modified by detuning the excitation from the probed Raman resonance. This effect was studied theoretically by the mean of a very simple one-dimensional model and a three-dimensional full-vectorial model. Both predict the alteration of the interface image. This was verified experimentally, for the first time at our knowledge, by imaging a polystyrene bead embedded in immersion liquid, around the polystyrene resonance at  $1003\text{cm}^{-1}$ . This experiment highlights the combined effects of spectral and spatial coherence associated with CARS microscopy. Such effects were demonstrated individually in CARS coherent control experiments [2] and giant unilamellar vesicles CARS imaging [3].

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<sup>†</sup> Current address - Laboratoire de Photonique Quantique et Moléculaire, UMR CNRS 8537, Institut d'Alembert - IFR 121  
École Normale Supérieure de Cachan, 61, avenue du président Wilson F-94235 Cachan, France.  
E-mail : [nicolas.sandeau@ida.ens-cachan.fr](mailto:nicolas.sandeau@ida.ens-cachan.fr)