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Third-harmonic generation microscopy in highly scattering media: comment

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In their contribution "Third-harmonic generation microscopy in highly scattering media" [Appl. Opt. **39,** 5187 (2000)], C. M. Blanca and C. Saloma apply the Monte Carlo technique to the calculation of third-harmonic generation (THG) in strongly scattering samples. In that study the authors fail to appreciate the coherent nature of both the signal-generation and the image-formation process, inherent to THG microscopy. Here we wish to clarify several points. © 2002 Optical Society of America

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1. First Clarification

Even though third-harmonic generation (THG) is, under circumstances of normal dispersion [where $n(\omega_3) > n(\omega_1)$, with n the refractive index of the material and ω_1 and ω_3 the frequency of the fundamental and the third harmonic beam, respectively], highly efficient only when the fundamental beam is focused on an interface between two materials of different material properties, the third-harmonic beam cannot be considered as being generated at the interface itself as suggested by Blanca and Saloma.[1] Instead, THG is the result of the coherent buildup of third-harmonic radiation in the bulk. In contrast to what is subsequently suggested by Blanca and Saloma,[1] the phase mismatch is of prime importance for the efficiency of the process. The interface at focus is necessary for an efficient generation of third-harmonic photons, because the Gouy phase shift of the nonlinear material polarization differs from that of the third-harmonic beam. Without an interface, this difference in Gouy phase shift is such that the third-harmonic radiation generated before the focus is all converted back into radiation at the fundamental wavelength after the focus.

As a result of disregarding the coherent nature of THG, Blanca and Saloma[1] calculate an axial resolution of THG on a bulk–bulk interface that is significantly smaller than the confocal parameter of the focused excitation beam. Owing to the coherent nature of THG, the axial resolution of THG is, however, equal to the confocal parameter of the fundamental beam, as has been verified experimentally.[2]

2. Second Clarification

Since the Monte Carlo (MC) technique requires incoherence of the underlying physical process, Blanca and Saloma[1] consider an excitation pulse that is infinitely short in time. However, the (purely theoretical) limiting case of a delta-peaked light pulse requires a (purely theoretical) limit of zero group-velocity dispersion over an infinitely extended spectrum. Even if such a situation were experimentally realizable, the proposed MC results would not describe it correctly, because in this limiting case THG is again coherent. Moreover, a minimum pulse duration is required for spectral differentiation between the fundamental and the third-harmonic beams.

By way of conclusion, the MC calculations of Blanca and Saloma[1] may be, with due adaptations, applicable to incoherent generation of fluorescence light in, e.g., three-photon absorption, but cannot be applied to THG, which is an intrinsically coherent process.

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