RECOGNITION OF MULTIPOLAR SECOND-ORDER NONLINEARITIES IN THIN-FILM SAMPLES

K. Koskinen, R. Czaplicki, and M. Kauranen

Optics Laboratory, Department of Physics, Tampere University of Technology, Korkeakoulunkatu 3, FIN-33720 Tampere, Finland
email: kalle.o.koskinen@tut.fi

Second-order nonlinear optical processes provide the basis for many important optical phenomena such as frequency conversion and electro-optic modulation of light. A prime example of a second-order process is the second-harmonic generation (SHG), i.e., conversion of light at a fundamental frequency ω to light at the doubled frequency 2ω. The main limitation of SHG is that - within the electric-dipole approximation of the light-matter interaction - it can occur only in non-centrosymmetric materials. However, higher multipole (magnetic-dipole and electric-quadrupole) effects do not suffer from such restriction. Thus, multipole effects can provide an interesting avenue towards novel second-order materials.

Although multipole effects have been already utilized in nanostructured materials, the design guidelines for strong multipolar responses in bulk of materials are poorly understood, and such responses are difficult to address reliably in experiments. Multipole effects can be studied by second-harmonic generation with two non-collinear beams at the fundamental frequency [1,2]. However, this technique was originally developed for bulk samples and new materials are commonly characterized as thin films. Thus, the existing models used to interpret two-beam experiments fail in properly accounting for effects particular to thin films, including propagation effects and multiple reflections. This gives rise to significant difficulties in the recognition of multipolar nonlinearities.

We introduce a detailed model that fully accounts for all thin-film effects and show that apparent multipole nonlinearities can arise from such effects even when the nonlinearity has strictly dipolar origin. Our results show both theoretically and experimentally that reliable recognition of multipolar responses of new materials will require extremely careful experiments combined with detailed theoretical modelling.

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[2] S. Cattaneo, and M. Kauranen, Phys. Rev. B **72**, 033412 (2005).