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SHG on Transition Metal Oxides O. NEY, G. LEFKIDIS, W. HÜBNER, Department of Physics, Kaiserslautern University of Technology, Box 3049, 67653 Kaiserslautern, Germany — Optical second harmonic generation (SHG), within the electric-dipole approximation, is known to be sensitive to the surface electronic structure of inversion symmetric media. Besides, it allows to distinguish magneto-optically ferromagnetic from antiferromagnetic order of cubic (001) surfaces. Here, we compute the SHG from NiO(001) and CoO(001) surfaces from first principles. To address the notoriously difficult electronic correlations of transition metal monoxides we use a high-level quantum-chemical *ab initio* approach. We find that the gap states of CoO(001), due to level bunching, are equally well suited for ultrafast manipulation of the antiferromagnetic state as those of NiO(001), the latter having fewer resonances. Furthermore, we present calculations of the nonlinear optical susceptibility tensor $\chi^{(2\omega)}$ for the NiO(001) and CoO(001) surfaces, where we, besides the electric-dipole contributions, for the first time include both magnetic-dipole and electric-quadrupole contributions computed from our first principles theory. For a detailed disentanglement of bulk and surface contributions we additionally calculate the nonlinear optical susceptibility of bulk NiO and perform a detailed comparison of the computed nonlinear SHG spectra with recent experimental findings. In this way we offer an alternative explanation of the experimental results in terms of surface electric-dipole and bulk nonlocal electric-quadrupole contributions.

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