Nonlinear optical properties of materials are important as tools in basic research and optical technology. Recently there has been a tremendous upsurge of interest in optical nonlinear effects, especially in crystals with curved bandstructure geometry. Such materials are candidates for applications based on the conversion of light to dc current. In this talk I describe our discovery that a family of Weyl semimetals has by far the largest second-order susceptibility of any previously known crystal. In puzzling over this result, we uncovered a surprising theorem relating the strength of optical nonlinearity to a quantum invariant property of the bandstructure that unites nonlinear optics with the celebrated “modern theory of polarization.” This quantum invariant provides a new strategy for algorithmic computational searches for nonlinear materials with optimal response functions.