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Linear and nonlinear optical properties of large-area, sub-nanometer gap metasurfaces JAKE FONTANA, Naval Research Laboratory, MELISSA MALDONADO, Universidade Federal de Pernambuco, NICHOLAS CHARIPAR, SCOTT TRAMMELL, Naval Research Laboratory, RAFAELA NITA, NRC-NRL Postdoctoral Fellow, JAWAD NACIRI, ALBERTO PIQUE, BANAHALLI RATNA, Naval Research Laboratory, ANDERSON GOMES, Universidade Federal de Pernambuco — Emerging dispersion engineering technologies require accurate knowledge of the index of refraction. Furthermore, assembly approaches are needed with nanometer scale resolution while simultaneously providing high-throughput for device realization. Here, we created centimeter-scale area metasurfaces consisting of a quasi-hexagonally close packed monolayer of gold nanospheres capped with alkanethiol ligands on glass substrates. We show these metasurfaces, with interparticle gaps of 0.6 nm, are modeled well using a classical (without charge transfer) description, since ambiguities exist in the literature. We find a large dispersion of linear refractive index, ranging from values less than vacuum, 0.87 at 600 nm, to Germanium-like values of 4.1 at 880 nm, determined using spectroscopic ellipsometry. Nonlinear optical characterization was carried out using femtosecond Z-scan and we observe saturation behavior of the nonlinear absorption and refraction. We find a negative nonlinear refraction from these metasurfaces two orders of magnitude larger than previous reports on gold nanostructures at similar femtosecond time scales. We also find the magnitude of the nonlinear absorption comparable to the largest values reported.

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