Field Enhanced Nonlinear Optical Properties in Ultrathin TiN Films

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Recent advances in epsilon-near-zero (ENZ) materials, where the real part of the permittivity vanishes over specific spectral ranges, have opened new pathways for achieving subwavelength confinement and significant enhancement of field intensity and optical nonlinearity. In naturally occurring metals, ENZ characteristics can be observed in the visible range when the metal layers are ultrathin, with nanoscale thicknesses. Realizing a practical ENZ effect for photonic applications depends on the ability to fabricate high-quality, crystal-like metal films with precisely controlled thickness down to a few nanometers. However, the performance of these ultrathin metal layers is often limited by high optical losses that increase as the thickness decreases, thereby constraining their potential in integrated nonlinear photonics. Here, we report a substantial enhancement of nonlinear optical properties in ultrathin titanium nitride (TiN) films within their ENZ region, achieved through the growth of high-quality, epitaxial single-crystalline TiN layers using nitrogen plasma-assisted molecular beam epitaxy (MBE). These ultrathin TiN films, with thicknesses as low as 2 nm, exhibit considerably small optical losses across the entire visible range. By employing the z-scan technique in combination with transient reflection spectroscopy, we investigate the nonlinear optical responses of these TiN films. Under p-polarized excitation, ultrathin (less than 10 nm) TiN epilayers exhibit significantly enhanced optical nonlinearity as their thickness decreases to 2 nm, attributed to quantum confinement. Moreover, optical nonlinearity displays a pronounced increase near the ENZ wavelength and a distinctive incidence-angle dependence, observed exclusively in ultrathin films, indicating the presence of the ENZ effect.