WWU MÜNSTER

Aktuelle Fragen
der Nanophysik

**Freitag, 22.09.2023 um 13:30 Uhr**
**R87, Wilhelm-Klemm-Str. 10**

# Mapping resonantly enhanced nonlinear optical signals of 2D metals


## Prof. Kenneth L. Knappenberger, Jr.

**The Pennsylvania State University, USA**
**Department of Chemistry**



Nonlinear frequency mixing (e.g. harmonic generation) and polarization rotation of electromagnetic waves are the foundation of many important and emergent applications, which include laser technologies, optical switches, and frequency combs, among others. The current state-of-the-art for second-order harmonic generation is achieved using a sequence of multiple quantum wells that are designed to resonantly enhance transitions at both fundamental and harmonic frequencies. However, these systems are intrinsically limited to the mid infrared, precluding their operation at frequencies important for many optical imaging and telecommunications applications. Therefore, new materials that can achieve large nonlinear optical responses over a broader range of frequencies are desired. Here, near-infrared-to-visible (NIR-Vis) second harmonic generation mediated by two-dimensional polar metals formed from gallium, indium, and silver is described.
These systems exhibit exceptionally large NIR-Vis second-order susceptibilities ($\chi^{(2)}$) - approaching 10 nm$^2$/V. This extra-ordinary response results from the unique atomic-level interfacial structure and bonding properties of one-to-three-atom-thick crystalline metal films that were formed by a process called confinement heteroepitaxy (CHet). Resonance matching in CHet-formed 2D Ag leads to competitive nonlinear excitation and scattering pathways. Coherent multi-dimensional Fourier transform nonlinear optical microscopy is used to disentangle these competing structure-dependent channels. Depending on the excitation pathway, a range of NLO responses that include saturable and enhanced harmonic generation can be induced. The strong interplay between nanoscale structure and material nonlinear optical responses suggest that a range of macroscopic photonic properties can be optimized through microscopic structural control.