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Plasmons under extreme dimensional confinement

HANNO WEITERING, Department of Physics and Astronomy, The University of Tennessee

In our studies, we explore how surface and bulk plasmons emerge under extreme dimensional confinement, i.e., dimensions that are orders of magnitude smaller than those employed in 'nanoplasmonics'. Atomically-smooth ultrathin Mg films were epitaxially grown on Si(111), allowing for atomically-precise tuning of the plasmon response.[1] While the single-particle states in these 3-12 monolayer (ML) thick films consist of a series of two-dimensional subbands, the bulk-plasmon response is like that of a thin slice carved from bulk Mg subject to quantum-mechanical boundary conditions. Remarkably, this bulk-like behavior persists all the way down to 3 ML. In the 3-12 ML thickness range, bulk loss spectra are dominated by the n=1 and n=2 normal modes, consistent with the excitation of plasmons involving quantized electronic subbands. The collective response of the thinnest films is furthermore characterized by a thickness-dependent spectral weight transfer from the high-energy collective modes to the low-energy single-particle excitations, until the bulk plasmon ceases to exist below 3 ML. Surface- and multipole plasmon modes even persist down to 2 ML. These results are striking manifestations of the role of quantum confinement on plasmon resonances in precisely controlled nanostructures. They furthermore suggest the intriguing possibility of tuning resonant plasmon frequencies via precise dimensional control.

[1]M.M. Özer, E.J. Moon, A.G. Eguiluz, and H.H. Weitering, Phys. Rev. Lett. **106**, 197601 (2011).