ACS Photonics

Article

Cite This: ACS Photonics 2018, 5, 1058−1067

Comprehensive Study of Plasmonic Materials in the Visible and Near-Infrared: Linear, Refractory, and Nonlinear Optical Properties

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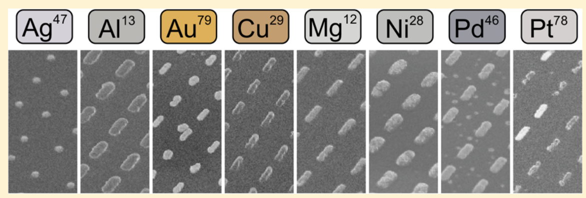
Supporting Information

ABSTRACT: Plasmonic nanostructures are used today for a variety of applications. Choosing the best suited plasmonic material for a specific application depends on several criteria, such as chemical and thermal stability, bulk plasma frequency, nonlinear response, and fabrication constraints. To provide a comprehensive summary, we compare these properties for eight different plasmonic materials, namely, Ag, Al, Au, Cu, Mg, Ni, Pd, and Pt. All these materials can be fabricated with electron beam lithography and subsequent evaporation of the desired material. First, we heated rod-antenna-type nanostructures made from these materials up to 1100 °C in air and investigated their linear optical response. Most structures lose their plasmonic properties at temperatures far below the melting point of the respective material. Gold, silver, and platinum structurally deform, whereas the other materials appear to chemically degrade. Second, to improve the thermal stability, structures with a 4 nm thin Al_2O_3 capping layer are fabricated. The thermal stability is significantly increased with the capping layer for all materials except for copper and magnesium. Lastly, the laser damage threshold is investigated for silver, aluminum, gold, and copper, which exhibit high nonlinear optical susceptibilities and are therefore particularly interesting for nonlinear optical applications.



KEYWORDS: *plasmonics, thermal stability, third-harmonic generation, linear and nonlinear properties, material comparison*

In recent years nanostructured plasmonic materials went from pure academic interest to applications. Localized surface plasmon resonances can be easily tailored to exhibit a specific optical response. Therefore, they are used in versatile applications such as plasmonically enhanced solar cells,[1] cancer therapy,[2] and optical sensing applications.[3] Most of these applications, in fact, use gold as plasmonic material, as it exhibits excellent optical plasmonic properties, is chemically inert, and can easily be nanostructured. However, gold is not the only material with plasmonic properties. Alongside the development of the applications, many new plasmonic materials were presented. For good optical plasmonic properties, a large free charge carrier concentration is necessary.[4] Therefore, most plasmonic materials are either metals or (doped) semiconductors. More exotic materials are Dirac systems such as topological insulators or graphene that can support edge state plasmons.[5,6] Metallic systems exhibiting plasmonic resonances include silver,[7−9] aluminum,[10,11] copper,[12] gallium,[13] magnesium,[14−16] molybdenum,[17] nickel,[18] palladium,[19] platinum,[20] and tungsten.[17,21] Also nitrides such as TiN and ZrN[22] and some hydrides such as YH_2[23] provide good plasmonic resonances. Additionally, metal oxides such as WO_3 and MoO_3 as well as transition metal oxides such as ReO_3 or VO_2 have been demonstrated to display plasmonic properties.[24] Furthermore, semiconductors such as transparent conducting oxides (ITO, AZO),[25] germanium,[26] and InSb[27] are used as plasmonic materials. From the group of topological insulators mainly bismuth telluride selenide compounds are investigated.[6]

For real world applications further aspects are relevant. First, the chemical stability is of major importance. Most materials are affected by oxidization or hydration and lose their plasmonic properties. Basically only gold and platinum are unaffected by these processes and are long-term stable under ambient conditions. Second, the thermal stability may play a major role. Many applications such as nonlinear optics or photovoltaics use the increased absorption at the plasmon resonance. Therefore, the plasmonic material can be heated locally to high temperatures. Some materials such as gold are known to reshape at temperatures as low as 100 °C,[28] which can easily be reached locally. Owing to the direct connection of geometrical shape to the properties of the plasmon resonance, this deformation leads to a low thermal stability. Besides the dimensional stability, the chemical stability also plays an important role at elevated temperatures, because higher temperatures lead to an increased chemical reaction speed. Third, for nonlinear applications the nonlinear optical susceptibility should be high. However, simultaneously the linear optical properties are also of significant importance owing to the local field enhancement. Last, the complexity to realize plasmonic nanostructures of high material quality varies strongly for different materials due to fabrication constraints.

It is not straightforward to achieve long-term stability of plasmonic structures in a harsh environment. All bare

Received: November 8, 2017
Published: January 3, 2018

DOI: 10.1021/acsphotonics.7b01346
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