Interband Effects on Hot Carrier Relaxation in Titanium Nitride Films

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Abstract: Hot carrier cooling rates determined for 30nm TiN film using transmission pump-probe. Experiments were conducted using a 400nm pump/800nm probe, as well as a 650nm pump/800nm probe. The bandstructure gives insight into the long cooling times observed.

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1. Introduction

Inherent to the speed and use of a material used in electronic or optical applications is the carrier relaxation dynamics. While the hot carrier relaxation processes of the noble metals have been well characterized over the past several decades, many materials of great interest are still being studied. Titanium Nitride is a plasmonic material that has on chip applications due to its plasmonic properties and CMOS compatibility [1]. As a refractory metal, in conjunction with its plasmonic properties, it has also been experimental demonstrated as an absorber for thermophotovoltaic applications.

Under the right growth conditions, the dielectric functions of TiN mimic those of Au, which is a conventional plasmonic material; however the carrier dynamics in TiN are more than an order of magnitude slower than those of Au when pumped at 800nm and probed at 1.2um [3]. Gold has a single band which crosses the Fermi level, so carrier thermalization and relaxation is fast and efficient [4], while carrier relaxation in the transition metal nitrides is more complex, involving intraband and interband relaxation processes.

2. Experimental Results

Hot carrier cooling times were measured by ultrafast pump-probe differential transmission with an 800nm probe and two pump wavelengths: 400nm and 650nm, the experimental results are shown in Fig 1. a) and b), respectively. A Ti:sapphire amplifier, BBO crystal and visible OPA were used generate the pump, 400nm probe, and 650nm probe, respectively. The pump and probe powers were 2 mW and 200uW with pulse widths of ~100 fs. The sample is a 30-nm TiN film grown epitaxially on a double polished sapphire substrate 800 °C.

Fig. 1. Measured pump-probe differential transmission at two pump wavelengths a) 400nm pump with 800nm probe b) 650nm pump with 800nm probe.
The band structure of TiN has three bands close enough to the Fermi level to play a contributing role. The valence band sits ~2eV below the Fermi level ($E_F$). The lower conduction band ($C_1$) dips below the $E_F$ by ~0.7eV and the upper conduction band ($C_2$) is ~0.6eV above the $E_F$. The 400nm (3.1 eV) probe is able to excite from the valence band to $C_2$, while the 650nm probe cannot. However, interband processes will contribute in both cases between the conduction bands. For each of these experiments, the cooling processes will take place within both the $C_1$ and $C_2$ bands, as well as between the conduction bands; each of these is expected to be fairly slow.

For an initial interpretation of the data, we have fit the dynamics using a double exponential; two cooling time constants are extracted, one on the order of tens of ps and the other on the order of hundreds of picoseconds. Following the pump pulse there is a rapid thermalization in the bands that remains for a separate study. The key observation is that the observed cooling dynamics are orders of magnitude slower than metals, but not as slow as materials with a bandgap. For the band structure this makes intuitive sense since it has bands that cross the $E_F$, like a traditional metal, but it also has other bands that are accessible with the pump energies chosen, so interband processes play a role. Details of the specific bandstructure will be presented. Work to theoretically model these processes is in progress.

3. Conclusion

The hot carrier cooling rates in TiN have been studied using transmission pump probe. Pump energies were chosen to correspond with different energy gaps in the bandstructure. Further studies that use only a near IR pump and probe, which would be only capable of perturbing the bands near the Fermi level, would solidify the role that interband transitions are playing in the slow cooling rates, since that area near the $E_F$ is very similar to the single band in the noble metals which leads to rapid thermalization.

4. References


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