Hot Electron Relaxation in Thin Titanium Nitride Films

H Ferguson[1], U Guler[2], N Kinsey[3], V. M. Shalaev[2,3], T Norris[1], A Boltasseva[2,3]

[1]*Electrical Engineering, University of Michigan, Ann Arbor, MI*
[2] *Nanometa Technologies, 1281 Win Hentschel Blvd, West Lafayette, IN, 47906, USA*
[3]*School of Electrical & Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette IN, 47907, USA*

Abstract: Pump-probe experiments using a femtosecond laser capture the dynamics of the electron thermalization and phonon relaxation. TiN is a plasmonic ceramic with many properties comparable to noble metals, but exhibits much weaker electron-phonon cooling.

OCIS codes: (240.0310) Thin films; (350.5340) Photothermal effects; (260.2160) Energy transfer

1. Introduction

Plasmonics has been an area of intense research for several decades driven by the promise of smaller faster all optical devices. Aside from the main obstacle in implementation, the loss in metals, another barrier is that the traditional plasmonic metals, Au and Ag are not CMOS compatible. TiN shows many of the same plasmonic properties as gold, but is a refractory material and CMOS compatible [1].

While many of the material parameters can be tuned to mimic the noble metals, electron-phonon interactions may be very different and are largely unexplored in these materials. This work investigates the phonon mediated hot-carrier cooling rate using pump-probe spectroscopy. Results show that TiN has a relaxation time that is more than one order of magnitude lower than gold. This could have implications in high-temperature refractory applications and nonlinear optical response, and for the development of fundamental theoretical models of TiN and related materials.

2. Experimental Results

The hot carrier cooling rates were measured by pump-probe differential transmission. A Ti:sapphire amplifier and OPA were used to generate a pump at 800nm and probe at 1.2um, with pulse widths of 100fs. The pump and probe powers were 20mW with a 200um diameter spot and 2mW with a 100um diameter spot, respectively. The samples were 10-nm TiN films grown epitaxially on double polished sapphire substrates at a low growth temperature and a high growth temperature, and their experimental results are shown in Fig 1. a) and b), respectively. Both samples were tested at 300K and 77K.

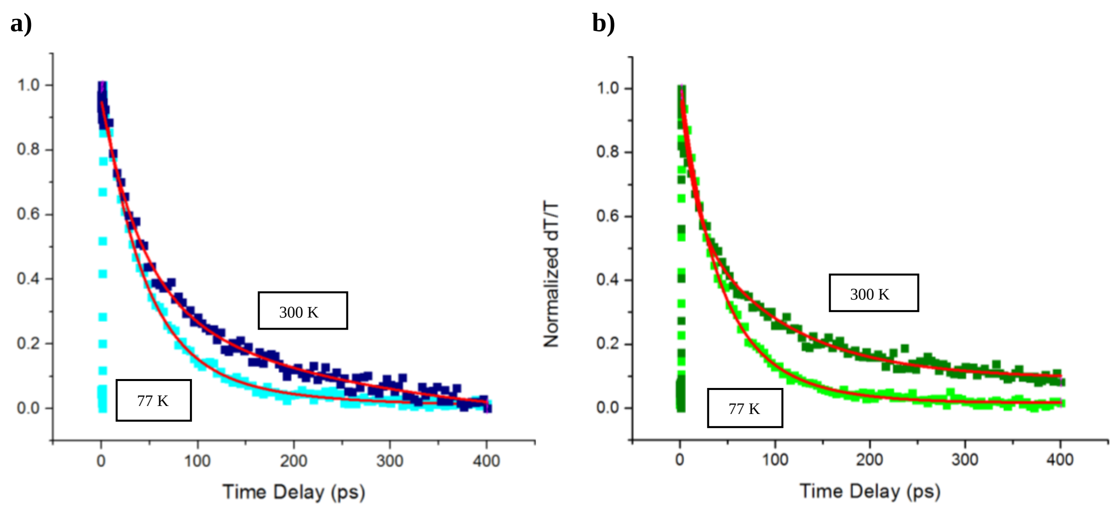

Fig. 1. Measured pump-probe differential transmission for two different samples **a)** low growth temperature and **b)** a high growth temperature. Each sample was measured at room temperature and 77K, with the solid lines showing the fitting for a biexponential decay.

Hot carrier thermalization in thin metals is usually modeled by the Two Temperature Model (TTM) [2], referring to the temperatures of the photoexcited hot electrons and the lattice. For an initial interpretation of the data, we have fit the dynamics using a double exponential; two time constants are extracted, a fast electron thermalization τ_{th} and a slower relaxation τ_p arising from the finite phonon lifetime. *The key observation is that the observed cooling* *dynamics are several orders of magnitude slower than for typical noble metals*. The electron-phonon interactions in traditional metals used in plasmonics have been studied extensively, and carrier cooling typically occurs on a single-picosecond time scale in those materials. By contrast, the electron cooling in TiN occurs with a time constant of approximately 10 ps, and the slow component arising from finite phonon lifetimes is nearly an order of magnitude longer. A direct comparison for TiN vs published results for gold is shown in Table 1. At present a detailed theoretical model accounting for this remarkable difference is not yet available. Further work in progress is motivated to sort out the role of electron-phonon coupling and the relevant densities of states. It should be noted that the development of refractory plasmonic materials is still in its early stage, and fundamental issues such as the band structure, deformation potentials, etc., remain to be understood; these results will provide experimental constraints on new theoretical models.

Table 1. Comparison of decay rates for Gold and TiN at 300K

	Reported for Gold Films [3]	TiN grown at a higher temperature	TiN grown at a lower temperature
τ_{th}	500fs	10ps	29ps
τ_p	1ps	67ps	162ps

3. Conclusion

The hot carrier cooling rate has been measured in TiN using transmission pump-probe and is much slower than that of gold. This is potentially due to a smaller electron- phonon coupling constant. More tests will help to solidify the reason for the slow cooling and provide a better understanding of TiN as a potential plasmonic material.

4. References

[1] Naik G. V., Shalaev V. M. & Boltasseva A. “Alternative plasmonic materials: Beyond gold and silver”, Adv. Mater. 25, 3264–3294 (2013).

[2] S.I. Anisimov, B.L. Kapeliovich, T.L. Perelman, “Electron emission from metal surfaces exposed to ultrashort laser pulses”, Sov. Phys. JETP 39(2), 375-377 (1974).

[3] Sun, C. K., Vallée, F., Acioli, L. H., Ippen, E. P., & Fujimoto, J. G., “Femtosecond-tunable measurement of electron thermalization in gold”, *Physical Review B*, *50*(20), 15337–15348. doi:10.1103/PhysRevB.50.15337 (1994).