The implication of using conductive nitrides as alternative plasmonic materials:
going beyond TiN and ZrN

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Birck Nanotechnology Center
Seminar, 04/30/2018

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Collaborators

Former and Current Students

G. Matenoglou (@Texas A&M)

L. Koutsokeras (@Cyprus Un. Tech.)

N. Pliatsikas (on going)

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Aristotle University

Spyros Kassavetis

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Christina Lekka

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University of Poitiers

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Grégory Abadias

Rensselaer Polytechnic Institute, NY

Daniel Gall and coworkers

University of Lorraine at Nancy

Institute Jean Lamour, France

Jean-François Pierson and coworkers

Nottingham Trent University, UK

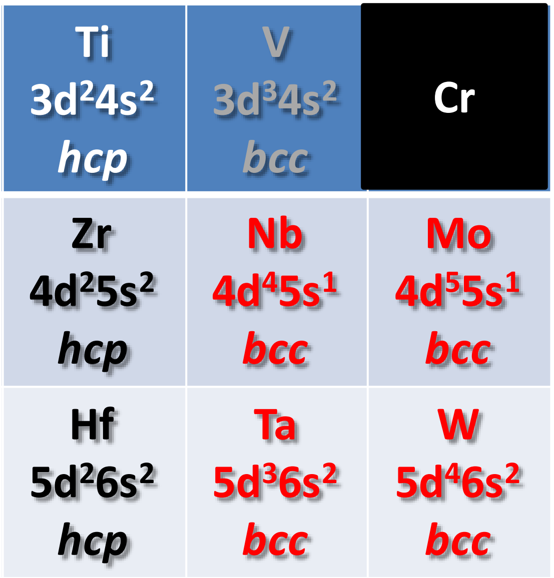
Nick Kalfagiannis

Layout

- A short long-story of conductive nitrides as optical conductors and their potential in plasmonics
  - Why the transition metal nitrides are good optical conductors?
  - Band structure and optical properties of continuous binary nitrides – identification of trends for plasmonics
  - Plasmonic performance of ternary nitrides: effects of blending, spectral tunability
- Implications:
  - Refractory character: the blessing turning into a curse
  - Intrinsic point defects in group Vb and VIb nitrides
  - Process-related defects: radiation damage and stress development during sputter-growth
  - Sputtering *vs.* alternative growth techniques: PLD
  - Extended *vs.* Local defects: grain/column boundaries vs. vacancies
- Polycrystalline *vs.* Epitaxial conductive nitrides

Cubic (B1) Mononitrides of the Group IVb-Vb-VIb Transition Metals

- One of the most widely studied and industrially implemented category of coating materials
- High hardness, chemical stability, refractory character
- **Electron conductors – electronic applications**
- Archetypical examples:
  - TiN and ZrN for mechanical applications (hard and wear-resistant coatings, among others)
  - TiN and TaN for electronic applications (Schottky contacts, diffusion barriers, ohmic contacts on GaN, etc)
- Emerging applications:
  - Plasmonics



Ti $3d^2 4s^2$ *hcp*	V $3d^3 4s^2$ *bcc*	Cr
Zr $4d^2 5s^2$ *hcp*	Nb $4d^4 5s^1$ *bcc*	Mo $4d^5 5s^1$ *bcc*
Hf $5d^2 6s^2$ *hcp*	Ta $5d^3 6s^2$ *bcc*	W $5d^4 6s^2$ *bcc*

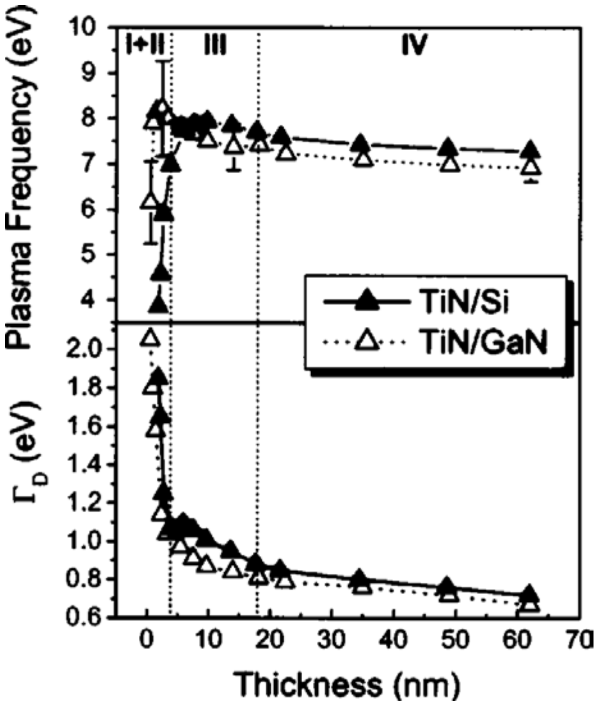
Nitrides for Plasmonics: TiN and ZrN

JOURNAL OF APPLIED PHYSICS VOLUME 93, NUMBER 2 15 JANUARY 2003

Interface properties and structural evolution of TiN/Si and TiN/GaN heterostructures

P. Patsalas[a)] and S. Logothetidis

Solid State Physics Section, Department of Physics, Aristotle University of Thessaloniki,
GR-54124 Thessaloniki, Greece

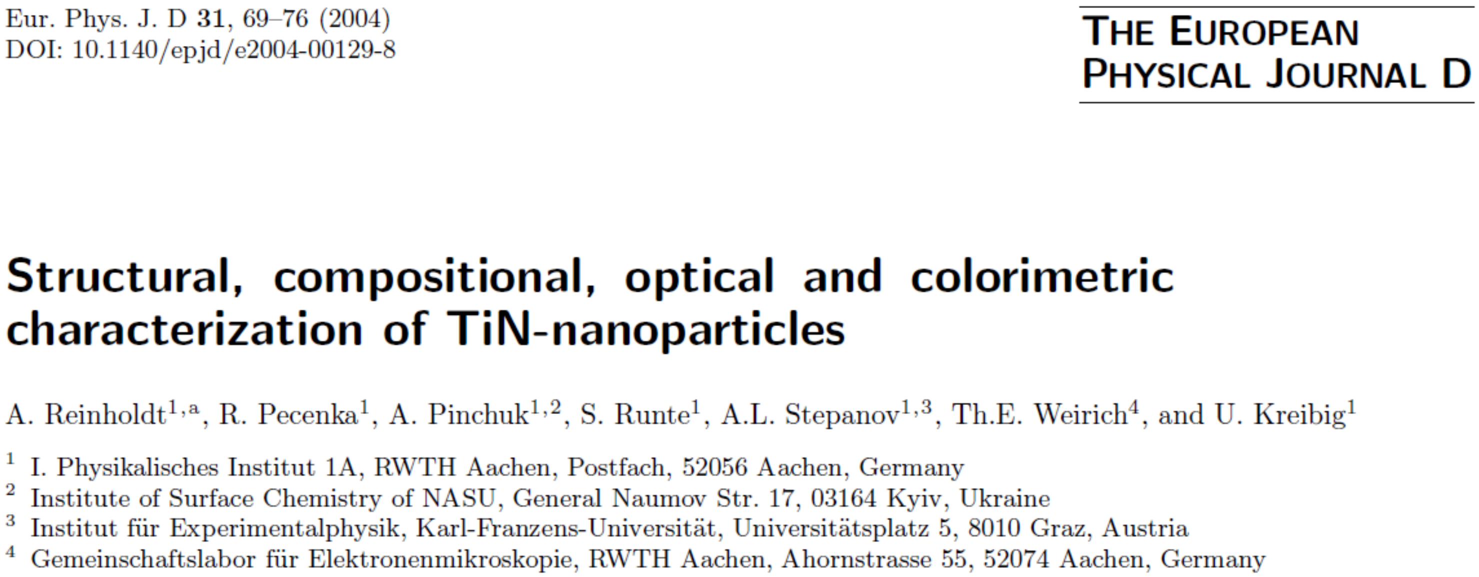


The first hint
Underestimated and undervalued

(by ourselves!)

*'At the nucleation stage of TiN/GaN, ϖ_{pu} exhibits much lower values than at the following stages of growth. This may be explained by the quasi-2D structure of the first deposited layer, which is equivalent with one TiN monolayer. The quasi-2D structure of the first layer may induce a **surface plasmon** vibration mode of the conduction electrons'*

Nitrides for Plasmonics: TiN and ZrN



Eur. Phys. J. D **31**, 69–76 (2004)
DOI: 10.1140/epjd/e2004-00129-8

THE EUROPEAN
PHYSICAL JOURNAL D

Structural, compositional, optical and colorimetric characterization of TiN-nanoparticles

A. Reinholdt[1,a], R. Pecenka[1], A. Pinchuk[1,2], S. Runte[1], A.L. Stepanov[1,3], Th.E. Weirich[4], and U. Kreibig[1]

[1] I. Physikalisches Institut 1A, RWTH Aachen, Postfach, 52056 Aachen, Germany
[2] Institute of Surface Chemistry of NASU, General Naumov Str. 17, 03164 Kyiv, Ukraine
[3] Institut für Experimentalphysik, Karl-Franzens-Universität, Universitätsplatz 5, 8010 Graz, Austria
[4] Gemeinschaftslabor für Elektronenmikroskopie, RWTH Aachen, Ahornstrasse 55, 52074 Aachen, Germany

The first report of plasmonic TiN

Nitrides for Plasmonics: TiN and ZrN

REVIEW

ADVANCED MATERIALS
www.advmat.de

Materials Views
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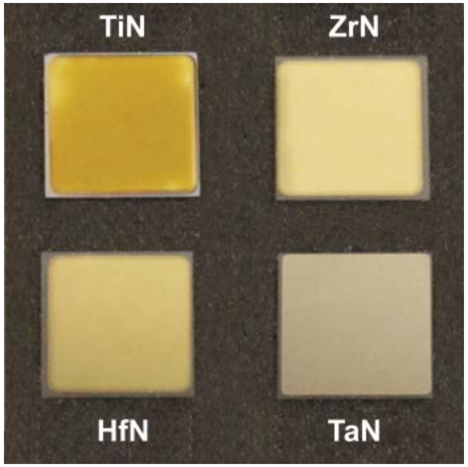
Alternative Plasmonic Materials: Beyond Gold and Silver

*Gururaj V. Naik, Vladimir M. Shalaev, and Alexandra Boltasseva**



The breakthrough!

Nitrides for Plasmonics: TiN and ZrN


Materials Views
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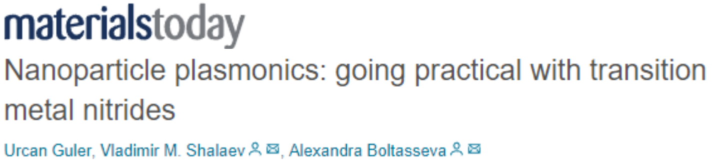
COMMUNICATION

Refractory Plasmonics with Titanium Nitride: Broadband Metamaterial Absorber

Wei Li, Urcan Guler, Nathaniel Kinsey, Gururaj V. Naik, Alexandra Boltasseva, Jianguo Guan, Vladimir M. Shalaev, and Alexander V. Kildishev**


materialstoday

Nanoparticle plasmonics: going practical with transition metal nitrides

Urcan Guler, Vladimir M. Shalaev, Alexandra Boltasseva


NANO LETTERS

Nonlinear Refractory Plasmonics with Titanium Nitride Nanoantennas

Lili Gui, Shahin Bagheri, Nikolai Strohfeldt, Mario Hentschel, Christine M. Zgrabik, Bernd Metzger, Heiko Linnenbank, Evelyn L Hu, and Harald Giessen


Materials Science and Engineering R 123 (2018) 1–55

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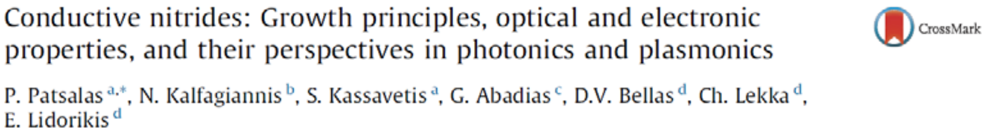
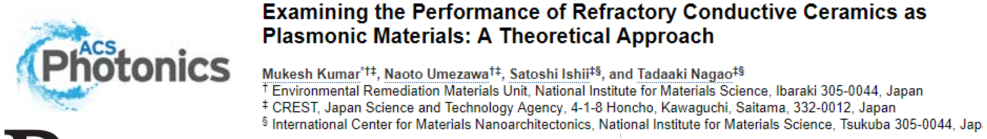

Conductive nitrides: Growth principles, optical and electronic properties, and their perspectives in photonics and plasmonics

P. Patsalas [a,*], N. Kalfagiannis [b], S. Kassavetis [a], G. Abadias [c], D.V. Bellas [d], Ch. Lekka [d], E. Lidorikis [d]


ACS Photonics

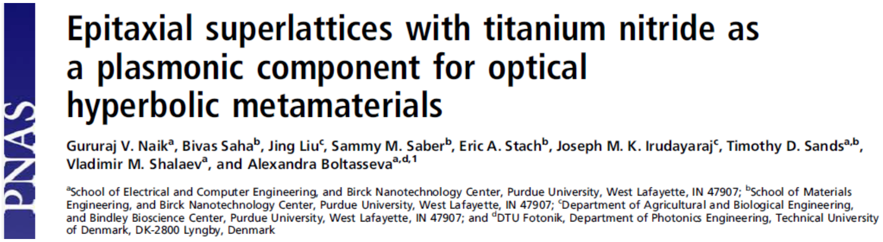
Examining the Performance of Refractory Conductive Ceramics as Plasmonic Materials: A Theoretical Approach

Mukesh Kumar[†‡], Naoto Umezawa[†‡], Satoshi Ishii[‡§], and Tadaaki Nagao[‡§]

[†] Environmental Remediation Materials Unit, National Institute for Materials Science, Ibaraki 305-0044, Japan

[‡] CREST, Japan Science and Technology Agency, 4-1-8 Honcho, Kawaguchi, Saitama, 332-0012, Japan

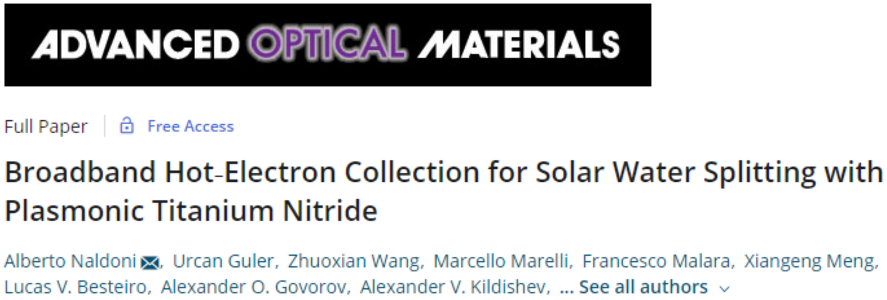
[§] International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba 305-0044, Jap


PNAS

Epitaxial superlattices with titanium nitride as a plasmonic component for optical hyperbolic metamaterials

Gururaj V. Naik[a], Bivas Saha[b], Jing Liu[c], Sammy M. Saber[b], Eric A. Stach[b], Joseph M. K. Irudayaraj[c], Timothy D. Sands[a,b], Vladimir M. Shalaev[a], and Alexandra Boltasseva[a,d,1]

[a]School of Electrical and Computer Engineering, and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907; [b]School of Materials Engineering, and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907; [c]Department of Agricultural and Biological Engineering, and Bindley Bioscience Center, Purdue University, West Lafayette, IN 47907; and [d]DTU Fotonik, Department of Photonics Engineering, Technical University of Denmark, DK-2800 Lyngby, Denmark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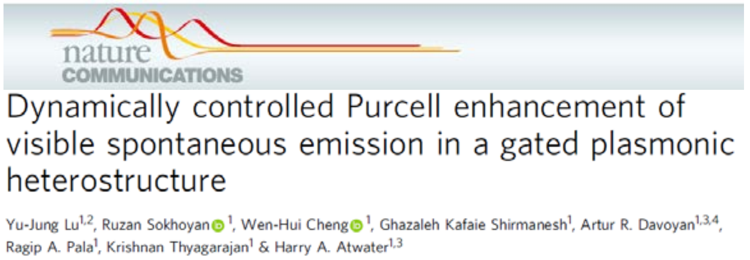
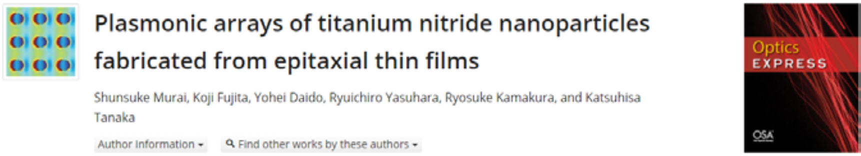
Broadband Hot-Electron Collection for Solar Water Splitting with Plasmonic Titanium Nitride

Alberto Naldoni, Urcan Guler, Zhuoxian Wang, Marcello Marelli, Francesco Malara, Xiangeng Meng, Lucas V. Besteiro, Alexander O. Govorov, Alexander V. Kildishev, ... See all authors


nature COMMUNICATIONS

Dynamically controlled Purcell enhancement of visible spontaneous emission in a gated plasmonic heterostructure

Yu-Jung Lu[1,2], Ruzan Sokhoyan[1], Wen-Hui Cheng[1], Ghazaleh Kafaie Shirmanesh[1], Artur R. Davoyan[1,3,4], Ragip A. Pala[1], Krishnan Thyagarajan[1] & Harry A. Atwater[1,3]


Plasmonic arrays of titanium nitride nanoparticles fabricated from epitaxial thin films

Shunsuke Murai, Koji Fujita, Yohei Daido, Ryuichiro Yasuhara, Ryosuke Kamakura, and Katsuhisa Tanaka

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ACS APPLIED MATERIALS & INTERFACES

Infrared Plasmonics with Conductive Ternary Nitrides

C. Metaxa,[†] S. Kassavetis,[†] J.F. Pierson,[‡] D. Gall,[§] and P. Patsalas[*,†]

PURDUE UNIVERSITY

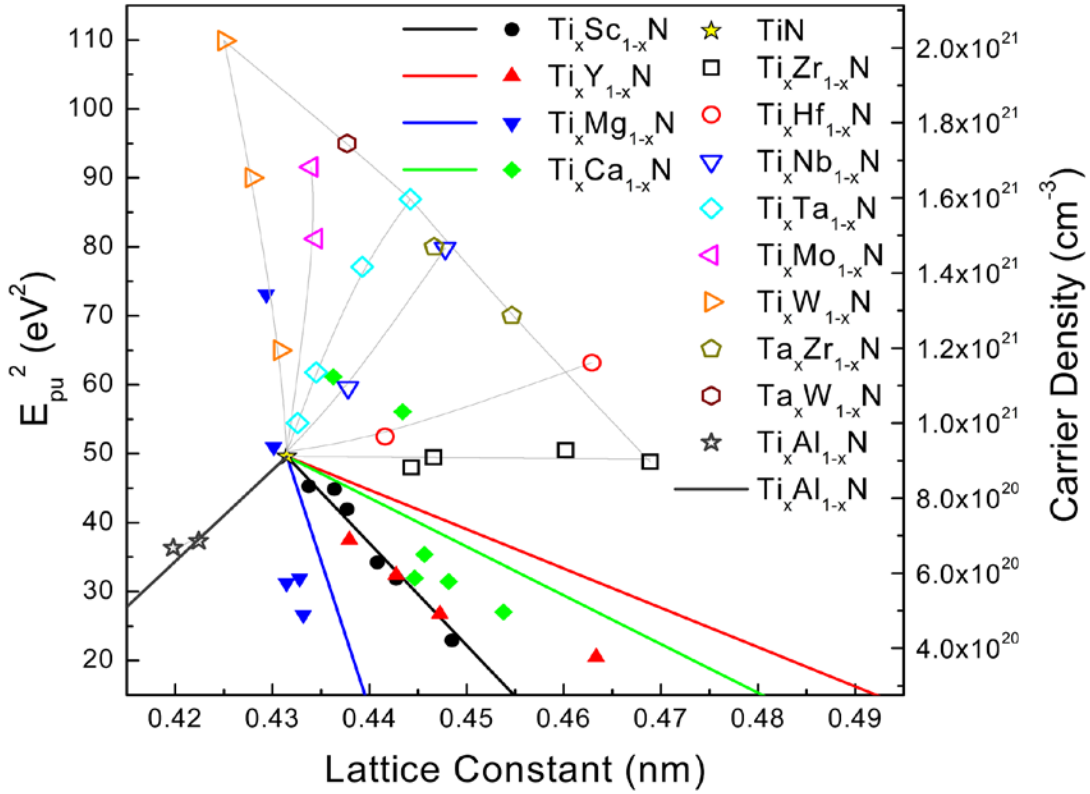
Birck Nanotechnology Center
Seminar, 04/30/2018

Aristotle University of Thessaloniki
Department of Physics
www.physics.auth.gr

Issues for electronic, photonic and *plasmonic* devices

Critical:

- Electrical conductivity
- Carrier density



Optical and electronic properties of conductive ternary nitrides with rare- or alkaline-earth elements

S. Kassavetis, A. Hodroj, C. Metaxa, S. Logothetidis, J. F. Pierson, and P. Patsalas

Citation: J. Appl. Phys. **120**, 225106 (2016); doi: 10.1063/1.4971407
View online: http://dx.doi.org/10.1063/1.4971407

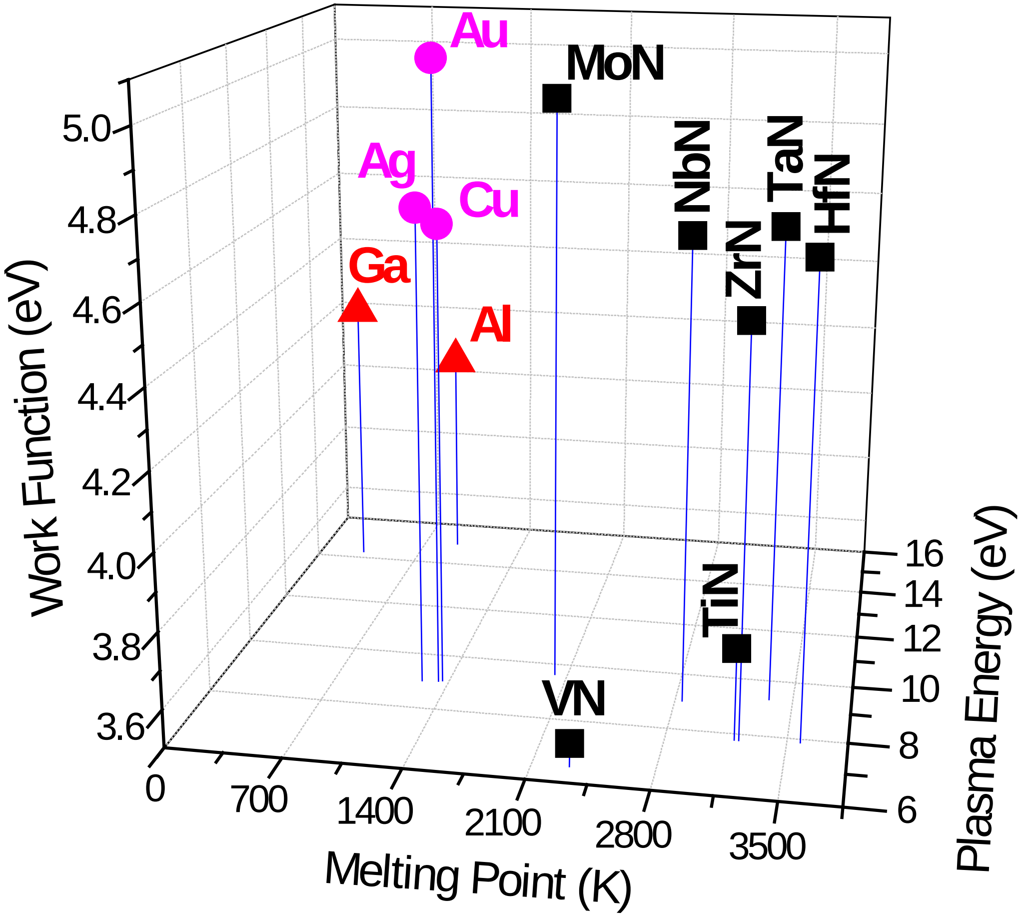
Issues for electronic, photonic and *plasmonic* devices

Critical:

- Electrical conductivity
- Carrier density
- *Zero real permittivity at optical frequencies*
- *Control of Electronic losses*
- *Control of Dielectric losses*

Desirable:

- CMOS compatibility
- Refractory character and *durability in strong fields*



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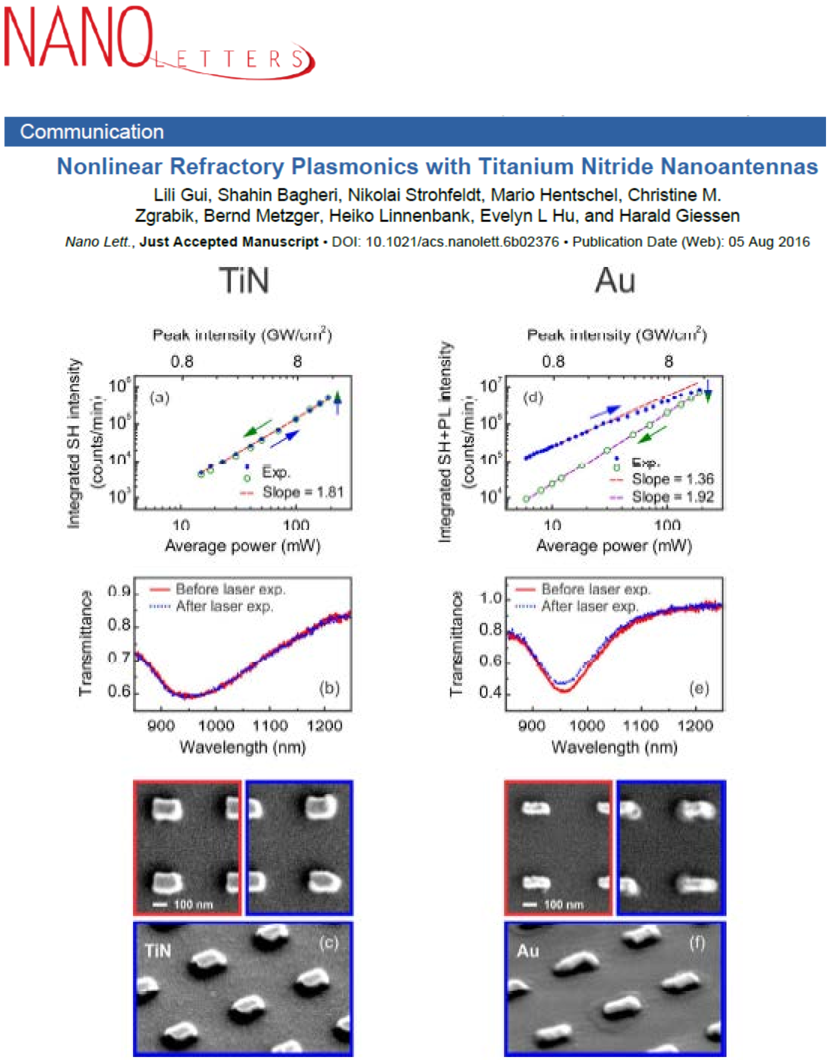

NANO LETTERS

Communication

Nonlinear Refractory Plasmonics with Titanium Nitride Nanoantennas

Lili Gui, Shahin Bagheri, Nikolai Strohfeldt, Mario Hentschel, Christine M. Zgrabik, Bernd Metzger, Heiko Linnenbank, Evelyn L Hu, and Harald Giessen

Nano Lett., **Just Accepted Manuscript** • DOI: 10.1021/acs.nanolett.6b02376 • Publication Date (Web): 05 Aug 2016

Issues for electronic, photonic and *plasmonic* devices

Critical:

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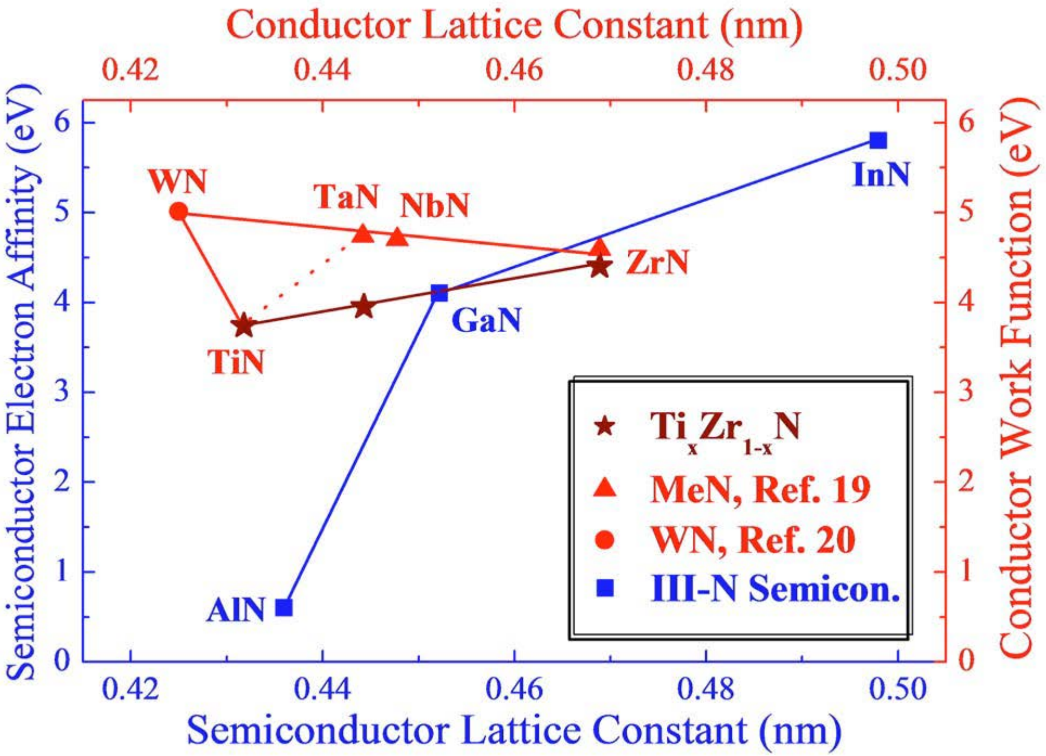
- *Zero real permittivity at optical frequencies*
- *Control of Electronic losses*
- *Control of Dielectric losses*

Desirable:

- CMOS compatibility
- Refractory character and *durability in strong fields*
- *Surface functionalization potential*
- Work function tunability (hot electrons)



APPLIED PHYSICS LETTERS **94**, 152108 (2009)

Plasma energy and work function of conducting transition metal nitrides for electronic applications

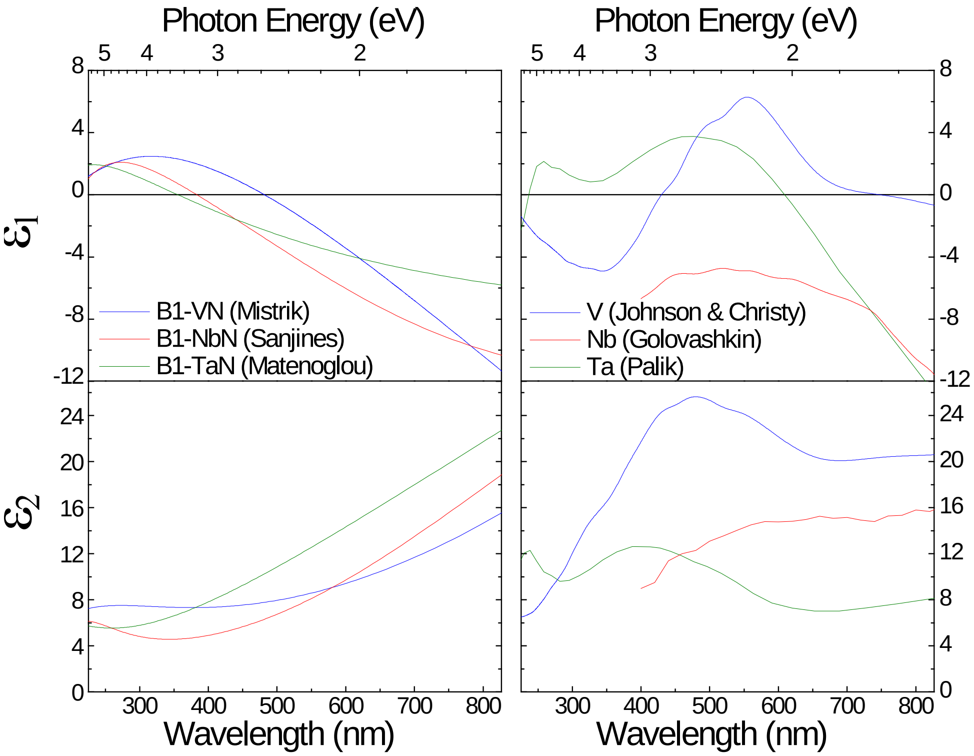
G. M. Matenoglou, L. E. Koutsokeras, and P. Patsalas[a]

Department of Materials Science and Engineering, University of Ioannina, Ioannina, 45110, Greece

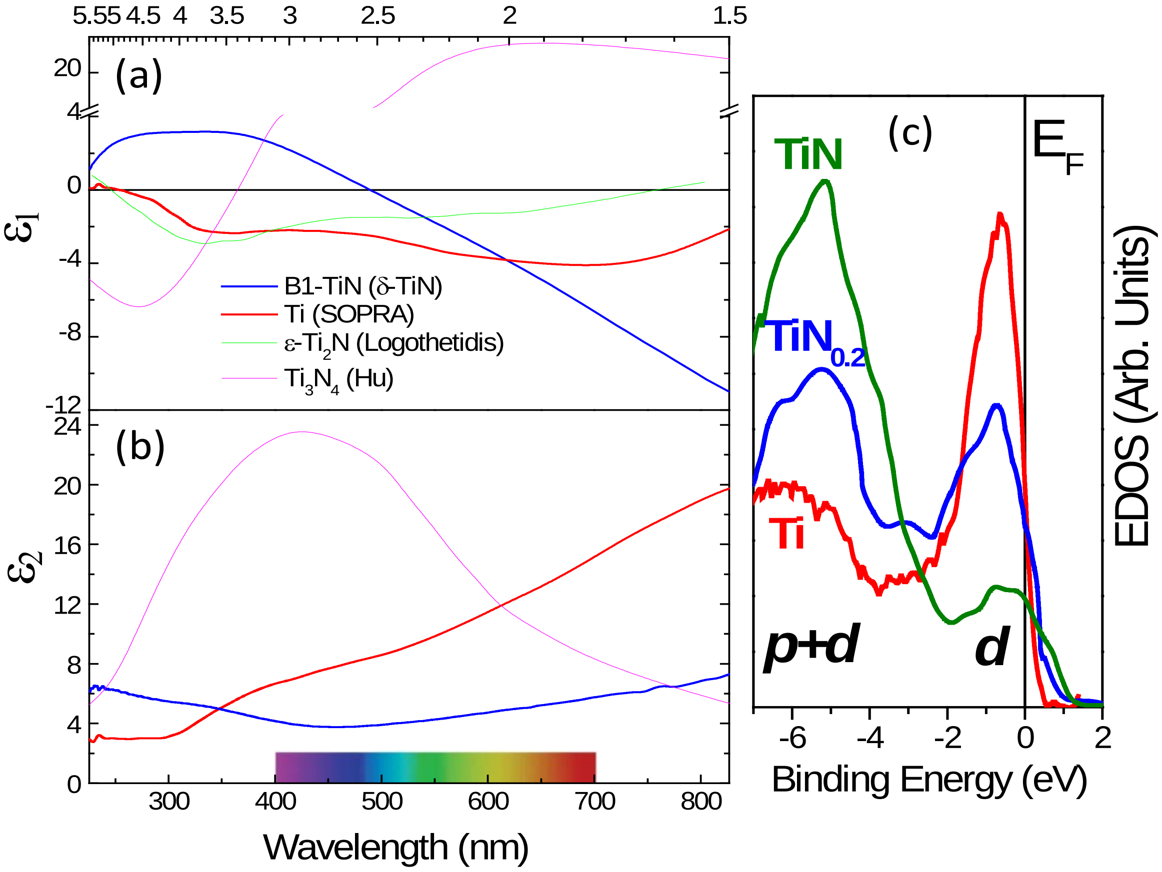
What does it make a good **optical** conductor?

When measuring DC or AC conductivity, we probe exclusively the conduction electrons; the DC/AC conductivity and mobility are affected only by the losses of the conduction electrons, either intrinsic (*i.e.* conduction electron density, electron relaxation time of the perfect single-crystal), or extrinsic (*e.g.* due to electron scattering at grain boundaries, point defects, *etc*)

In optical frequencies, we might probe bounded electrons, as well; consequently, the overall optical behavior would be screened by these bound electrons. Some authors call this screening 'dielectric losses'.



Why are the nitrides better optical conductors than the corresponding metals?



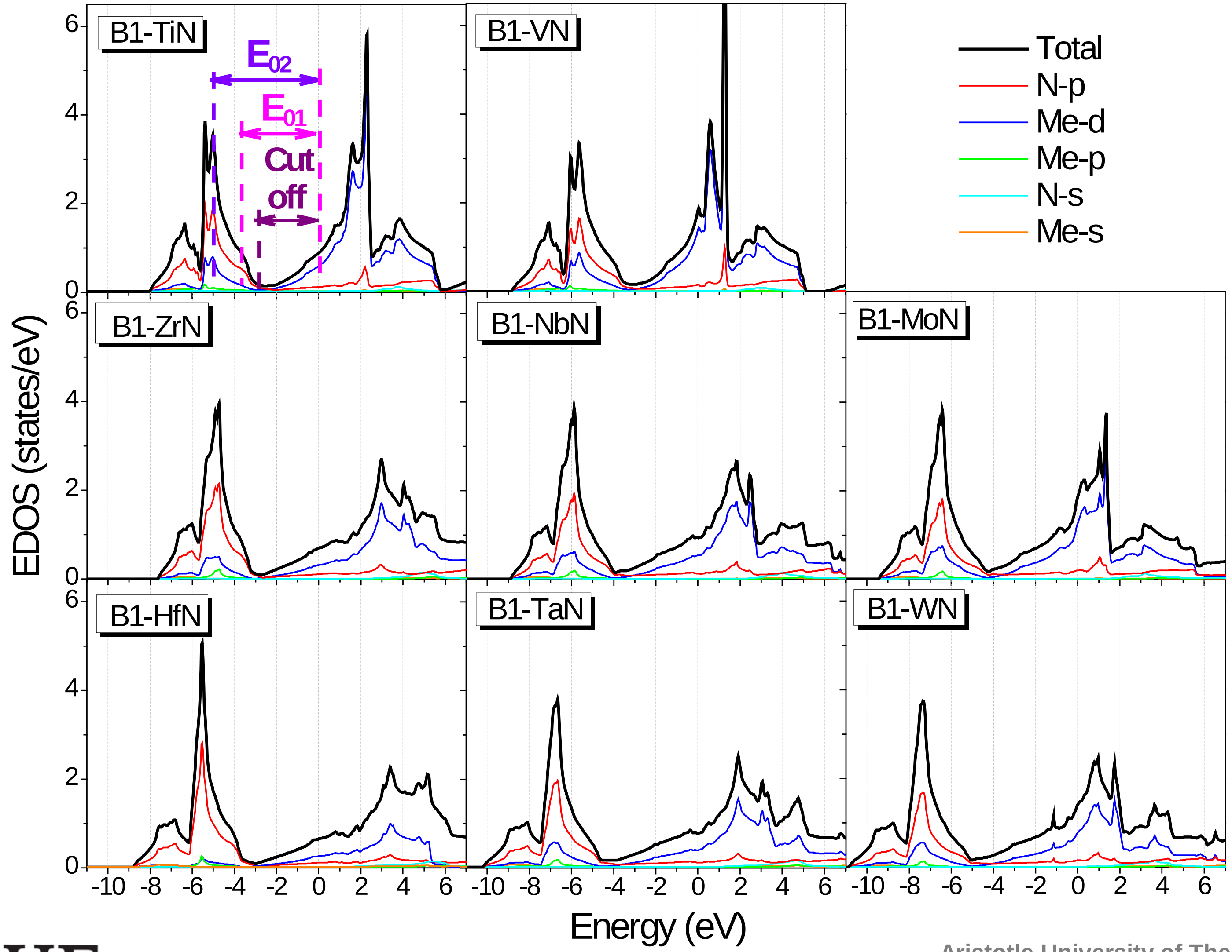
At the end of the day… the DC/AC conductivity of the metals might be still better than of the nitrides.

The nitrides are better conductors mostly in the optical (+NIR/MIR, UV) range.

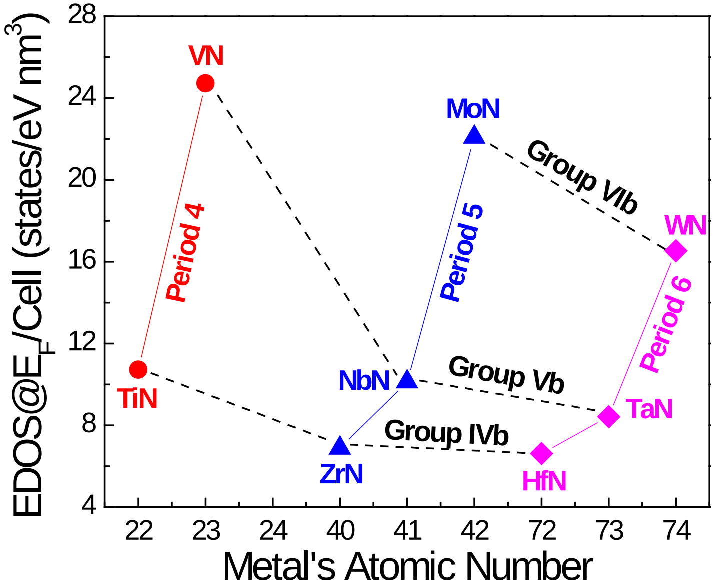
No need to revise your basic knowledge from the physics labs!

VB spectra from:
Vasile *et al*, JVSTA 8, 99 (1990) **TiN_x**
Fukuda *et al*, Surf. Sci. 91, 165 (1980) **Ti**

LAPW Calculations: The source of conductivity and dielectric losses



LAPW Calculations *vs.* Experiment



$$E_{pu} = \hbar\omega_{pu} \qquad \omega_{pu} = \sqrt{\frac{Ne^2}{\varepsilon_o m^*}}$$

APPLIED PHYSICS LETTERS **94**, 152108 (2009)

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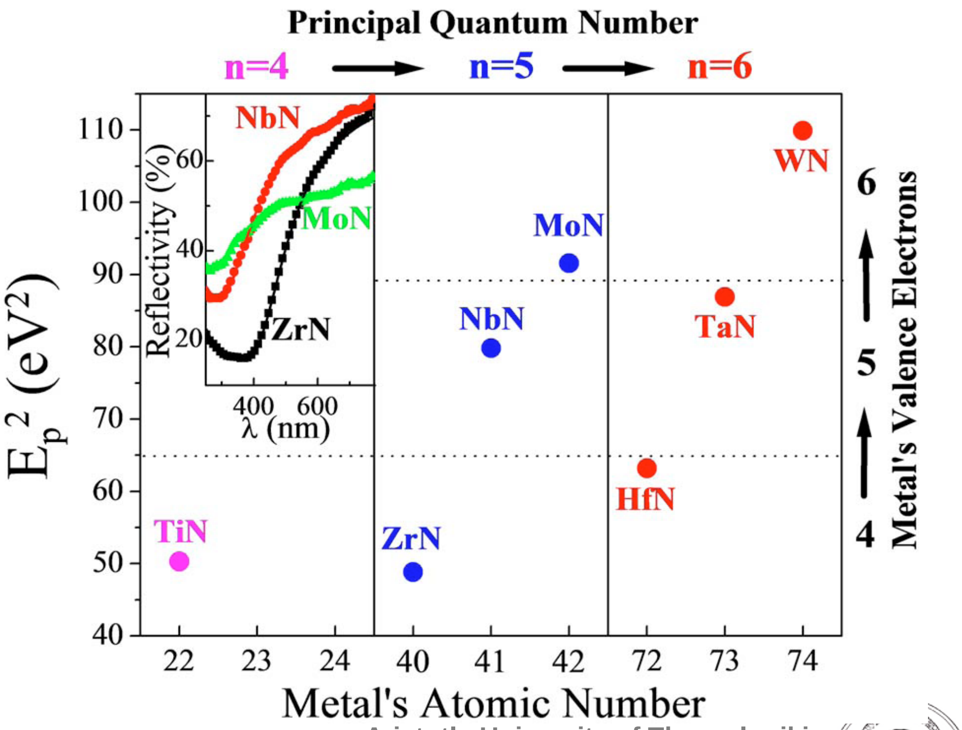
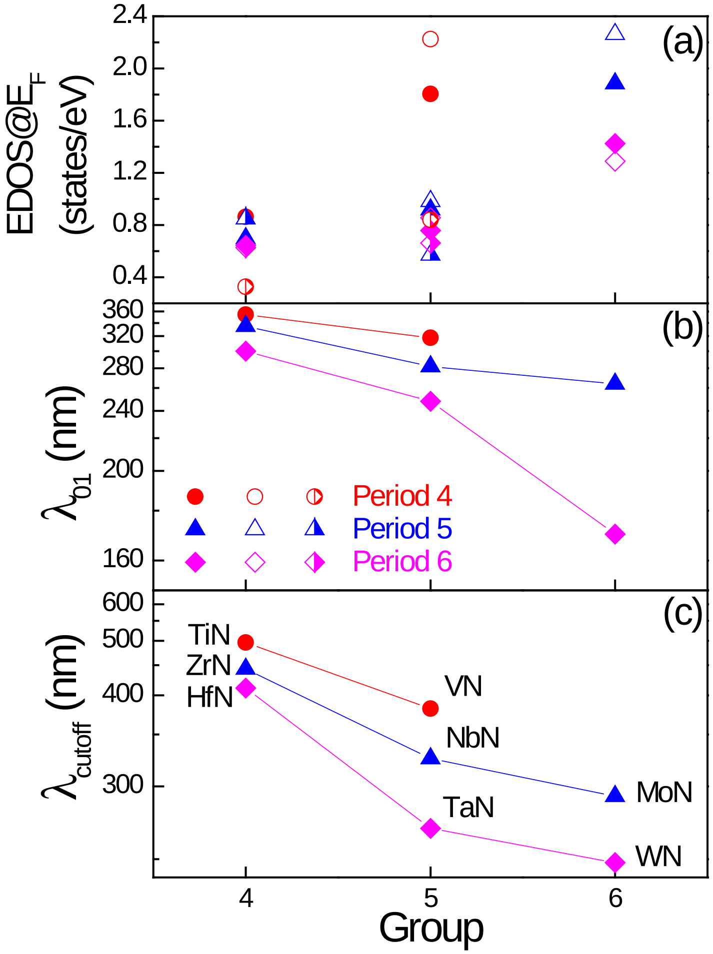
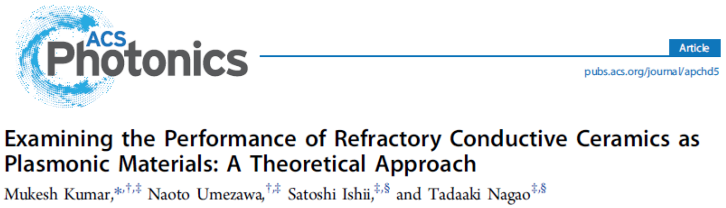


LAPW Calculations: Dielectric Losses



- •The dielectric losses are shifted to the UV with increasing Group number.
- •In B1 single-crystals, WN should be the best UV plasmonic material. Is it really?

Consensus among optical calculations


ACS Photonics

Article

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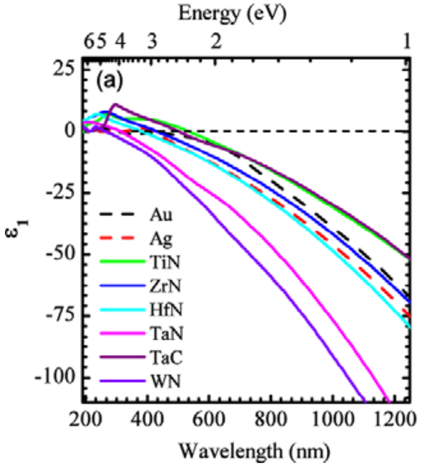
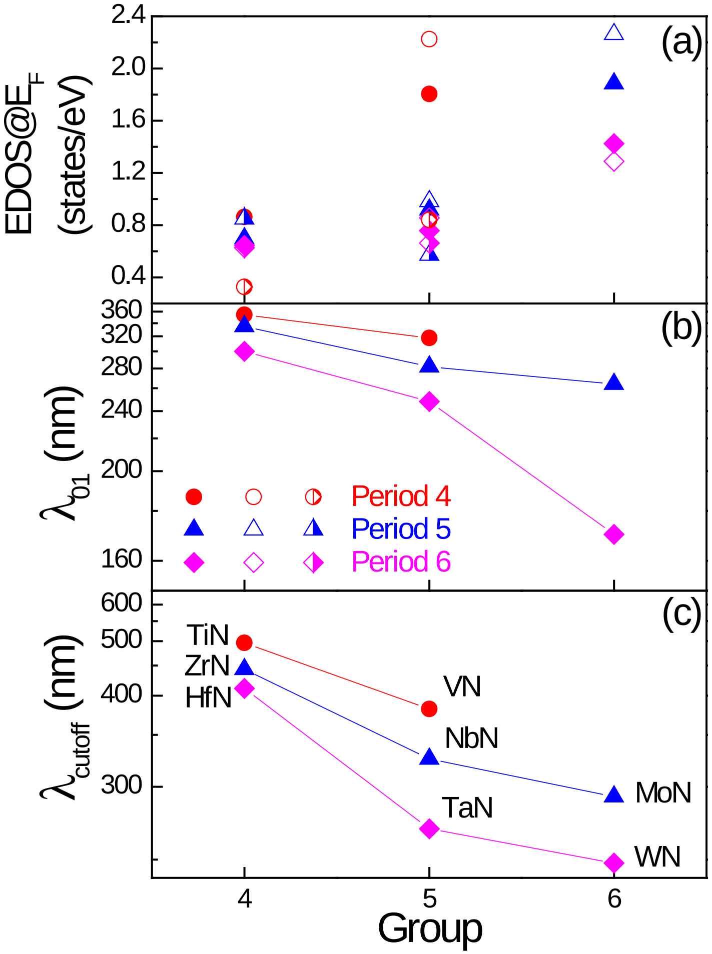
Examining the Performance of Refractory Conductive Ceramics as Plasmonic Materials: A Theoretical Approach

Mukesh Kumar,[*,†,‡] Naoto Umezawa,[†,‡] Satoshi Ishii,[‡,§] and Tadaaki Nagao[‡,§]

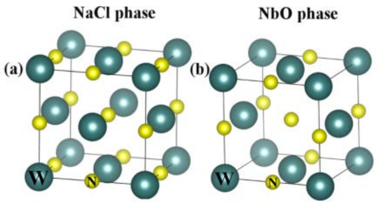


LAPW Calculations: Dielectric Losses



- The dielectric losses are shifted to the UV with increasing Group number.
- In B1 single-crystals, WN should be the best UV plasmonic material. Is it really?

Alas, the stability of the B1 structure and the growth reality tell another story; that of exceptionally lossy B1-WN




PHYSICAL REVIEW B **94**, 174111 (2016)

Vacancy-induced mechanical stabilization of cubic tungsten nitride

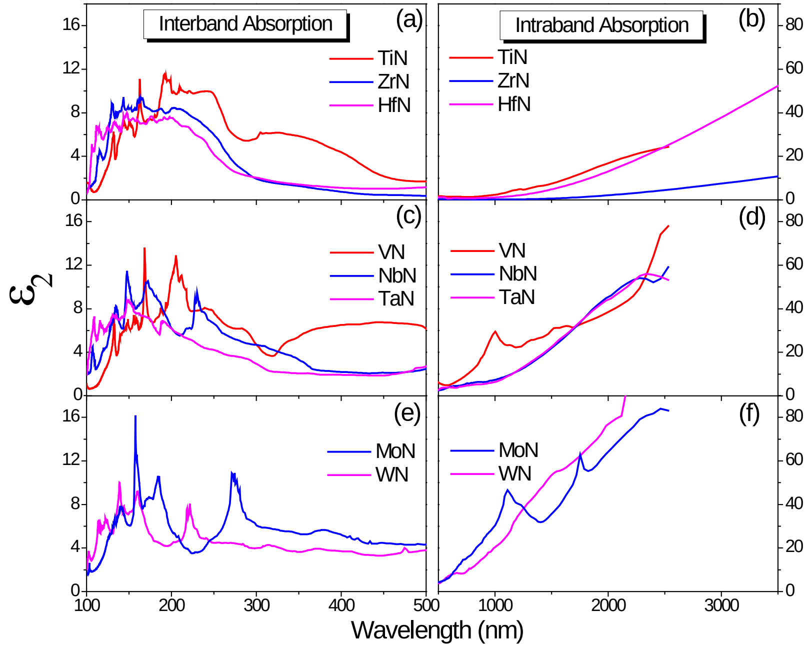
Karthik Balasubramanian,[1] Sanjay Khare,[2] and Daniel Gall[3]

[1]*Department of Mechanical, Nuclear and Aerospace Engineering, Rensselaer Polytechnic Institute, Troy, New York 12180, USA*

[2]*Department of Physics and Astronomy, The University of Toledo, 2801 West Bancroft Street, Toledo, Ohio 43606, USA*

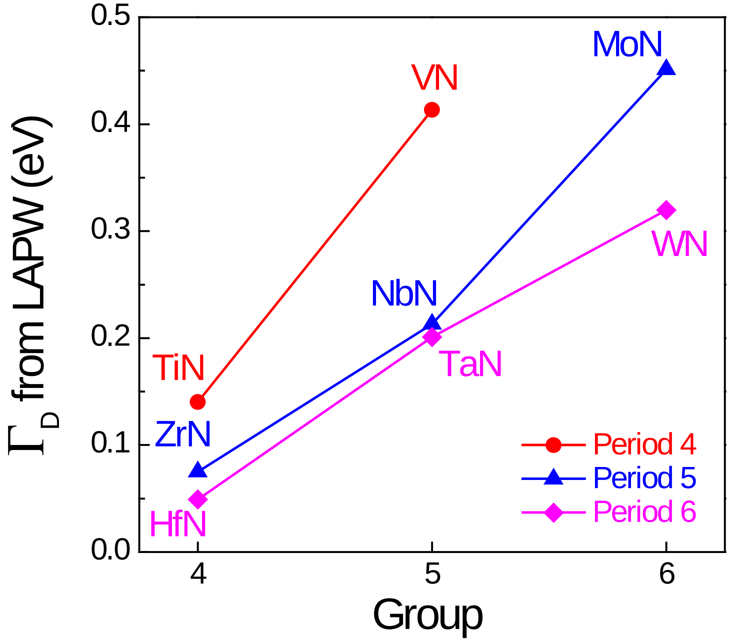
[3]*Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, New York 12180, USA*

LAPW Calculations: Electronic Losses



$$\tilde{\varepsilon}(\omega) = \varepsilon_1 + i\varepsilon_2 = 1 - \frac{\omega_{pu}^2}{\omega^2 - i\Gamma_D\omega},$$

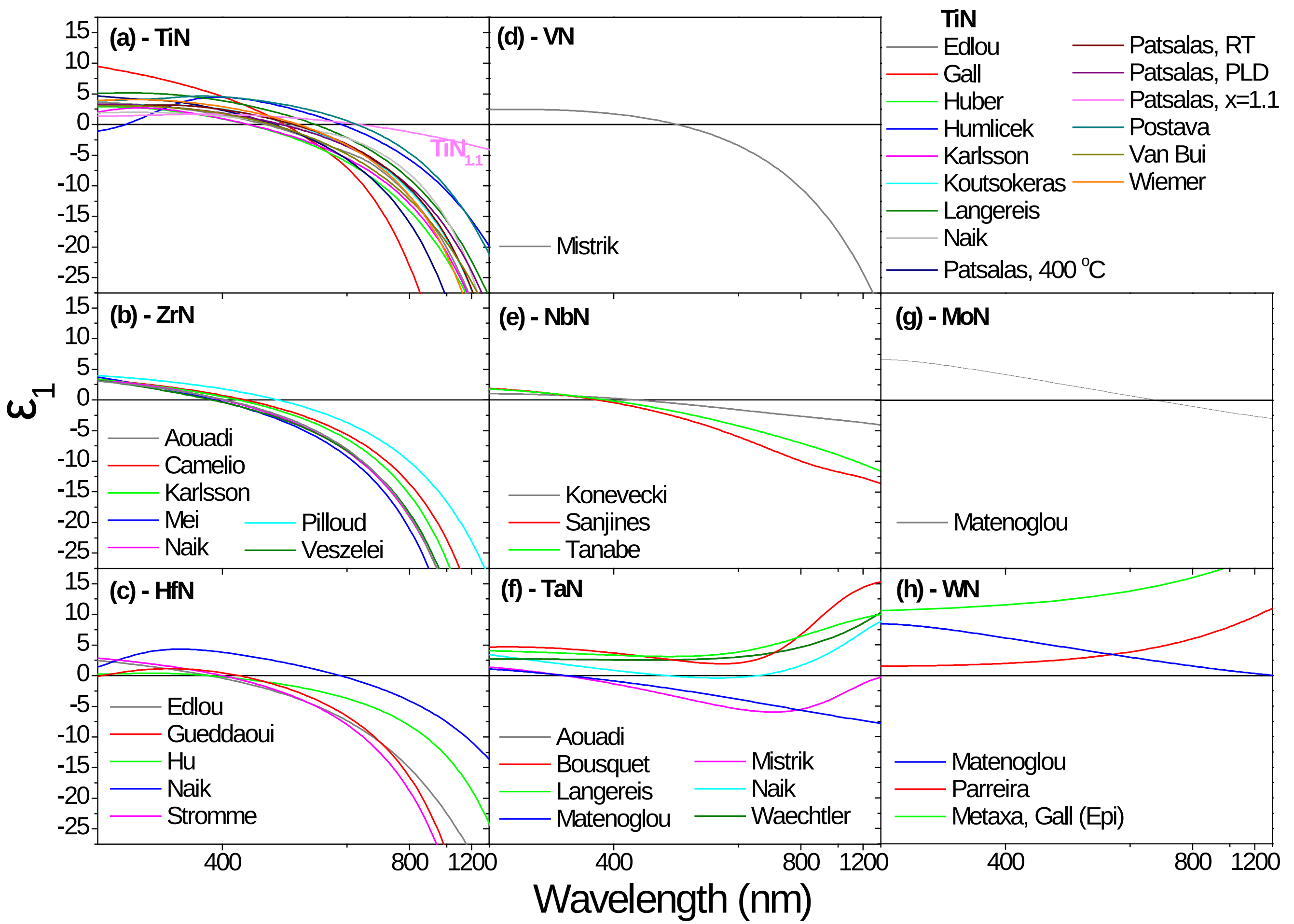
$$\Gamma_D(eV) = \frac{\hbar}{\tau_D(s)}$$



The electronic losses are increasing with increasing Group number

This competition between electronic and dielectric losses with increasing Group number calls for an optimal compromise!

Experimental Dielectric Function Spectra of Binary Nitrides

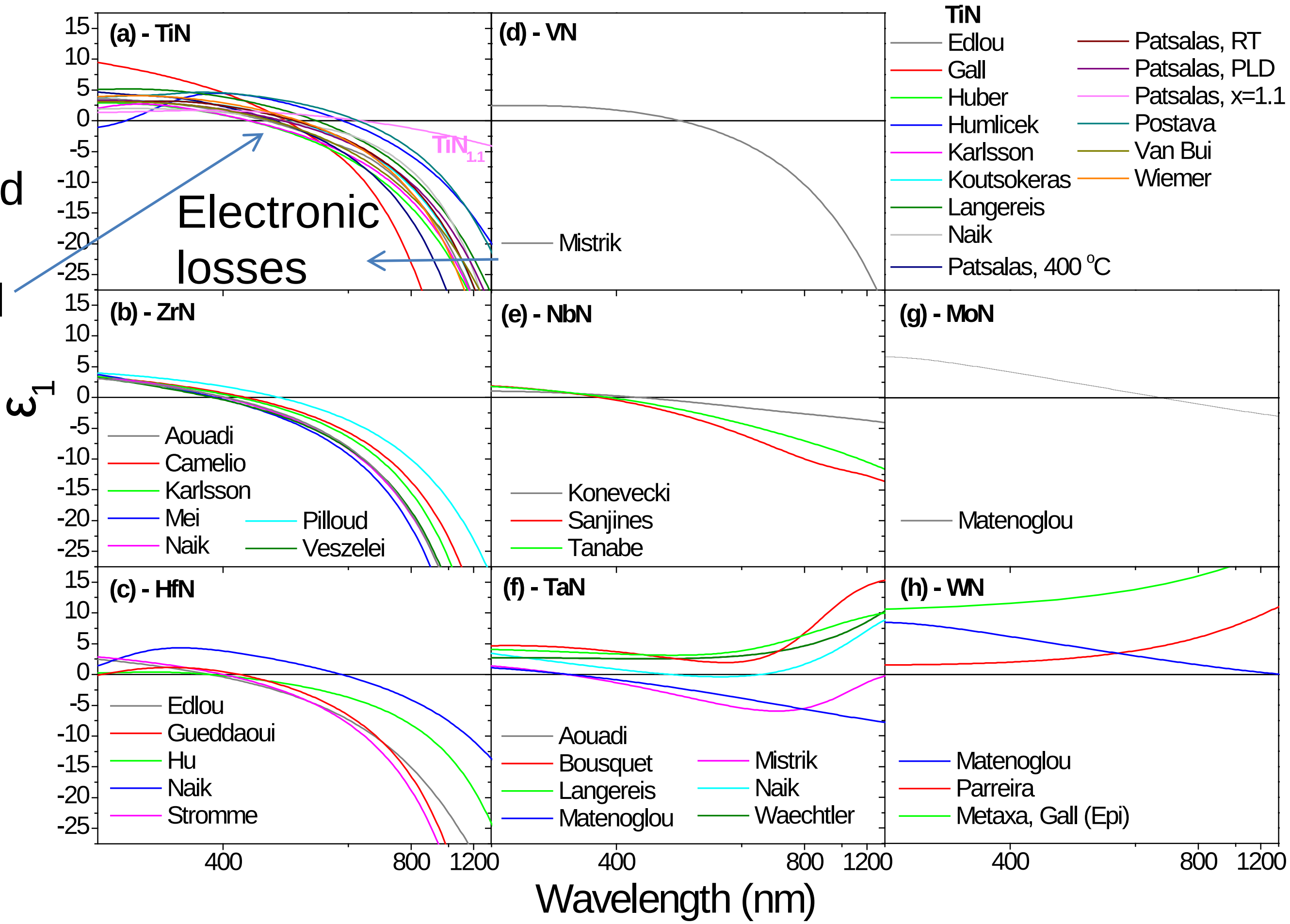


Data from groups worldwide

(Nancy, Poitiers, Strasbourg, Thessaloniki, Linköping, Uppsala, Augsburg, Aachen, Brno, Eindhoven, Lausanne, Barcelona, Coimbra, Urbana, Caltech, Purdue, RPI, Arizona, Texas, Ibaraki, Alberta, *etc*)

grown by Sputtering, CVD, ALD, CVA, IBD, *etc*

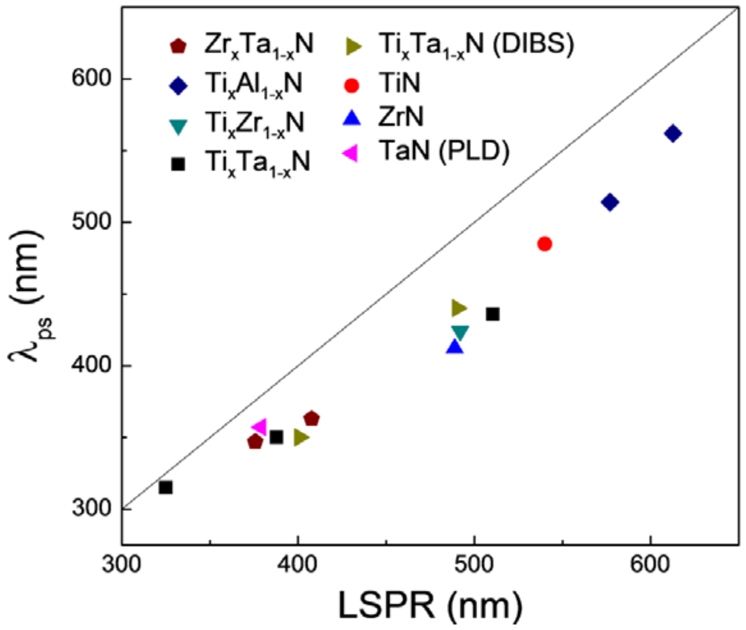
Experimental Dielectric Function Spectra of Binary Nitrides

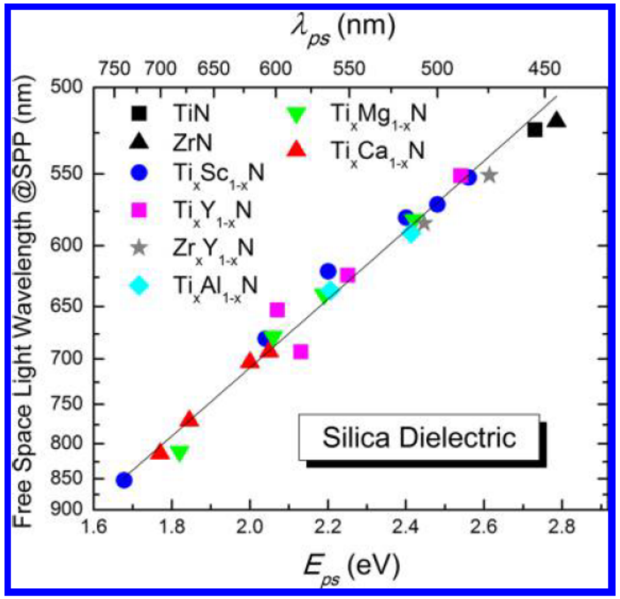


SPP and LSPR spectral position

Electronic losses

The importance of screened plasma energy E_{ps}





AIP Applied Physics Letters

Plasmonic spectral tunability of conductive ternary nitrides

S. Kassavetis, D. V. Bellas, G. Abadias, E. Lidorikis, and P. Patsalas

Citation: Applied Physics Letters **108**, 263110 (2016); doi: 10.1063/1.4955032
View online: http://dx.doi.org/10.1063/1.4955032

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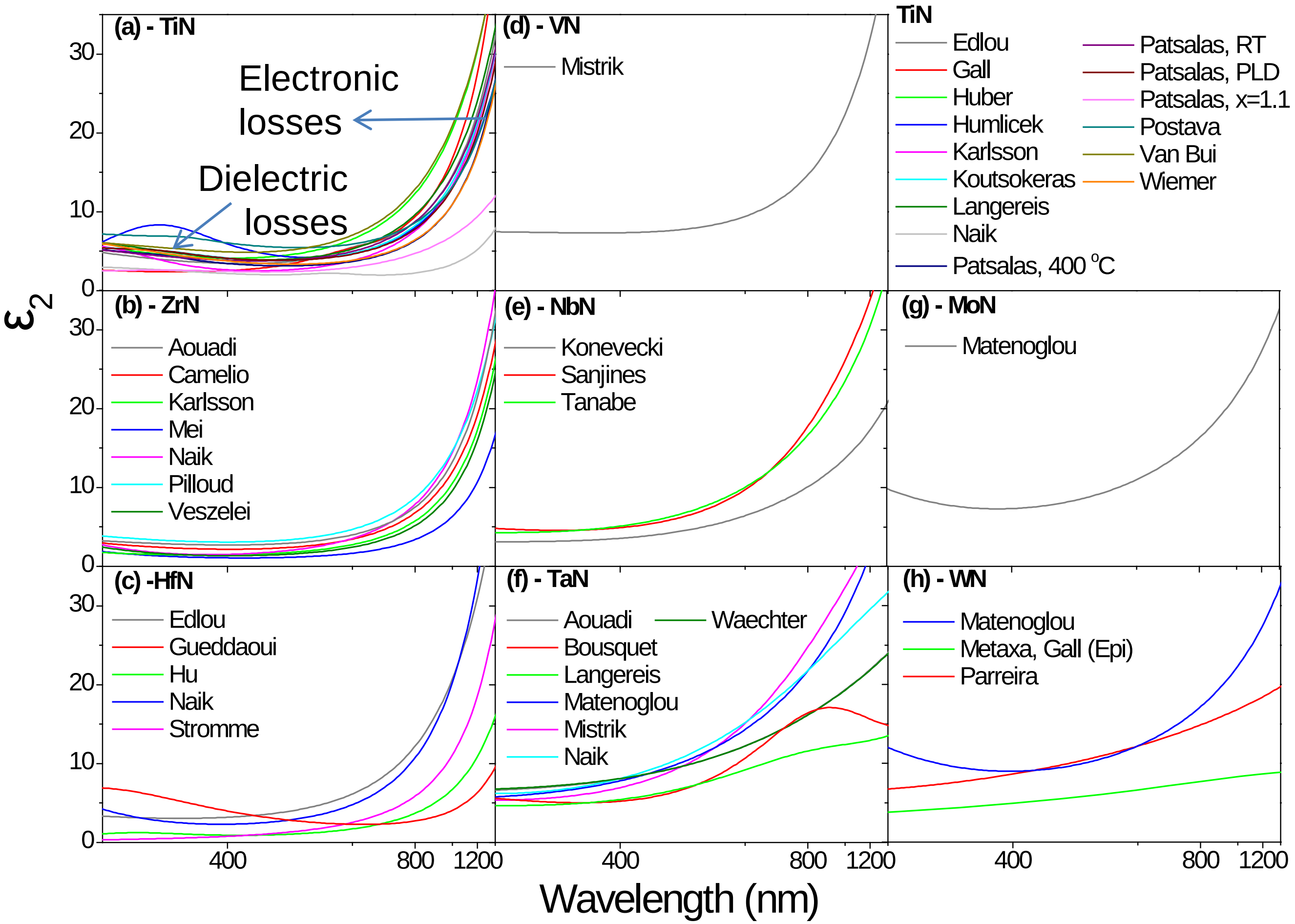
Infrared Plasmonics with Conductive Ternary Nitrides

C. Metaxa,† S. Kassavetis,† J.F. Pierson,‡ D. Gall,§ and P. Patsalas*,†

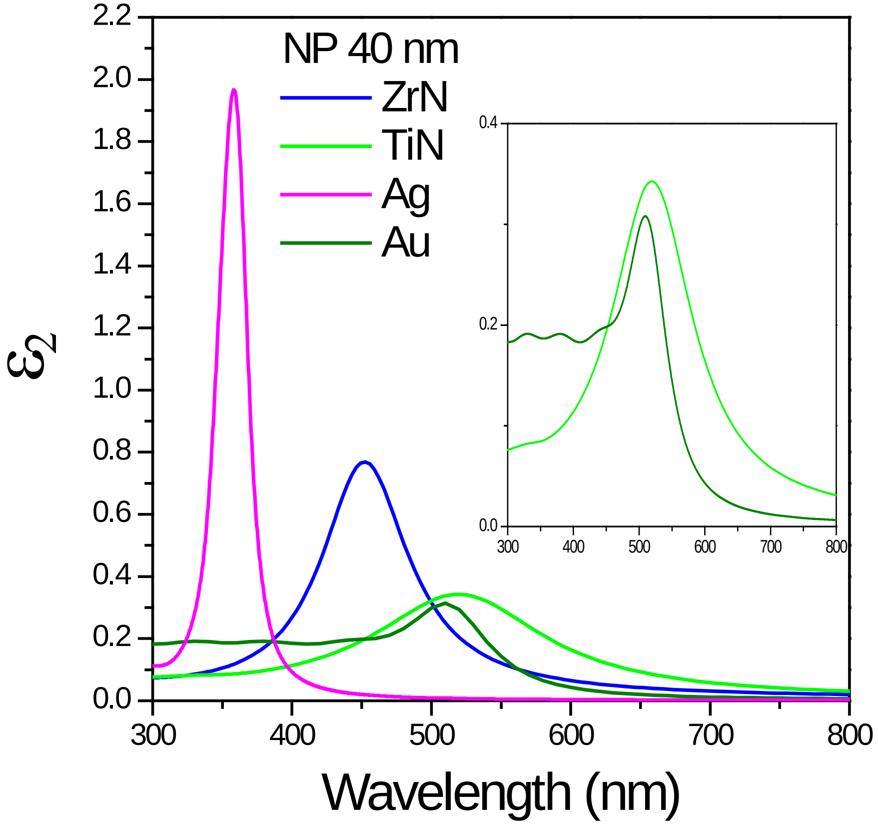
Experimental Dielectric Function Spectra of Binary Nitrides



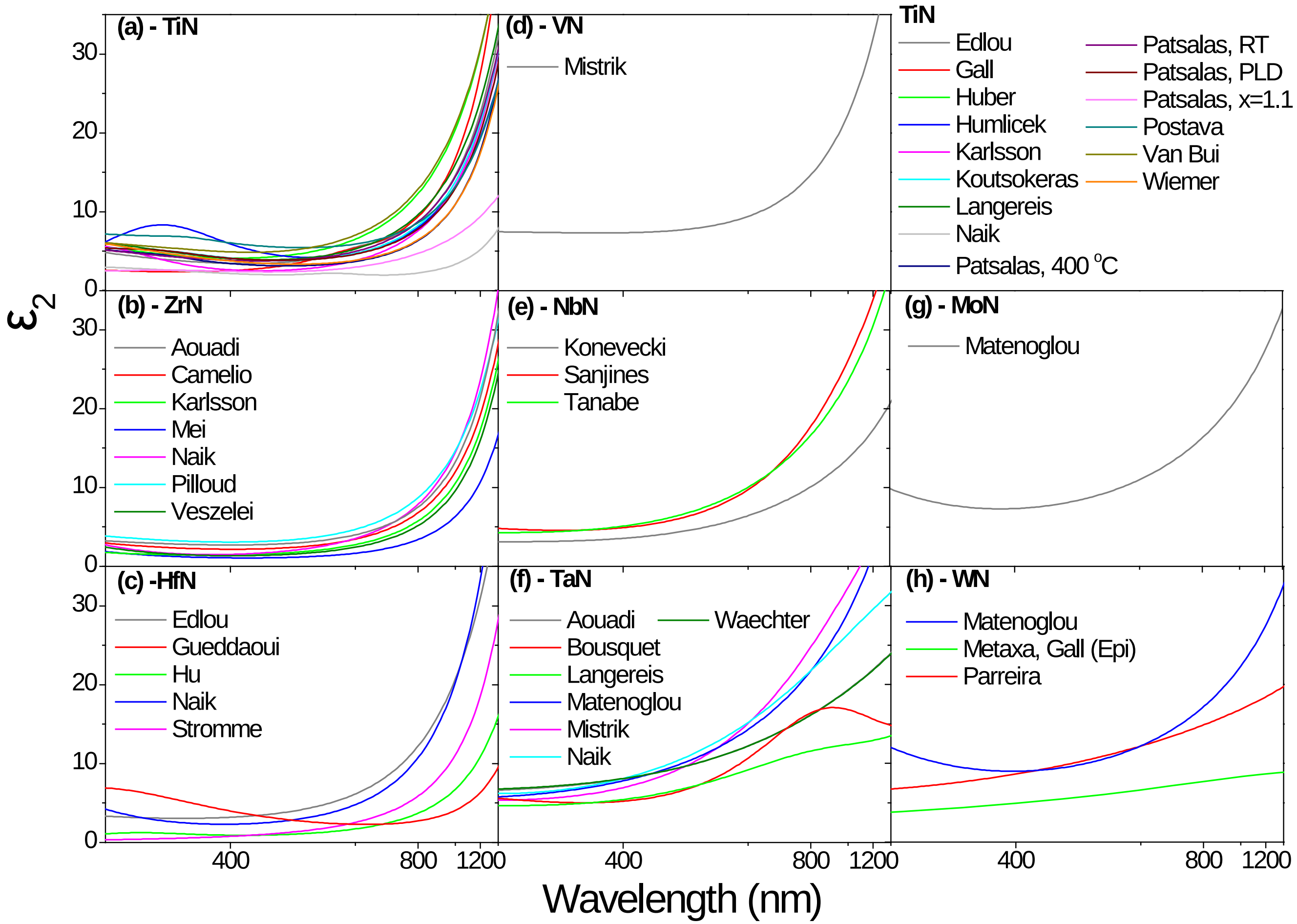
Predictions:

1) **ZrN** should be the best plasmonic nitride for the vis range and not the well studied TiN!
2) This is due to the **minimum electronic AND dielectric losses** observed for ZrN

Experimental Dielectric Function Spectra of Binary Nitrides



Experimental Dielectric Function Spectra of Binary Nitrides



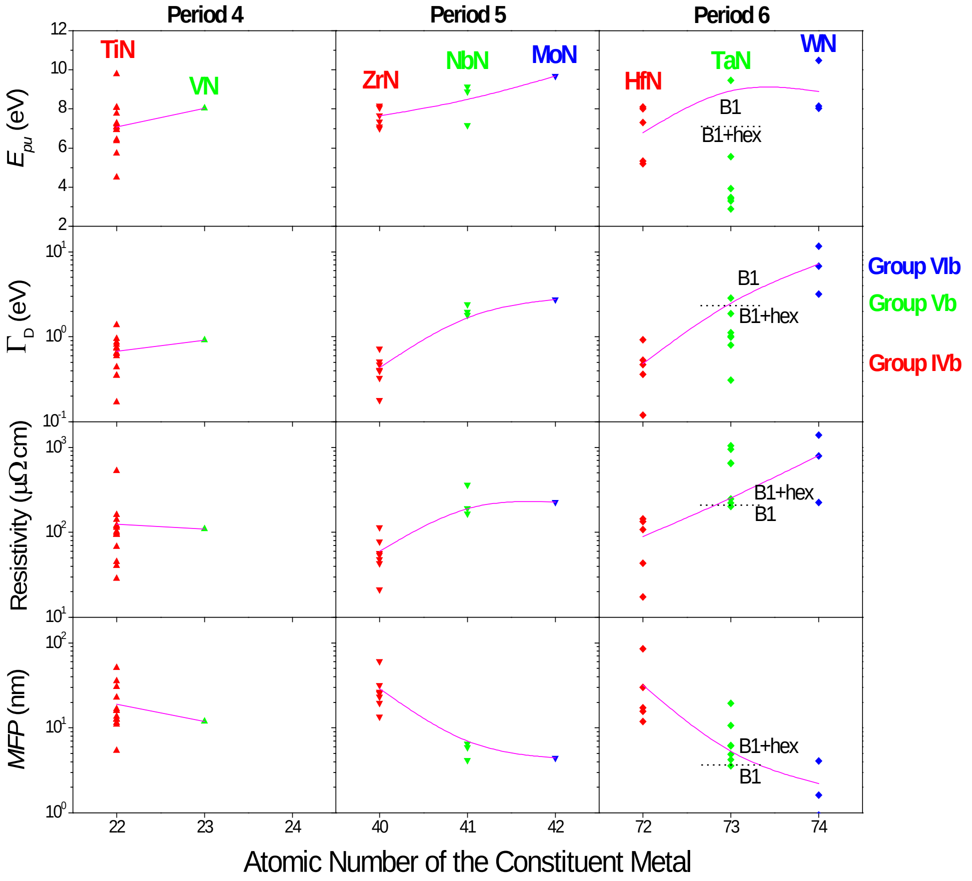
Predictions:

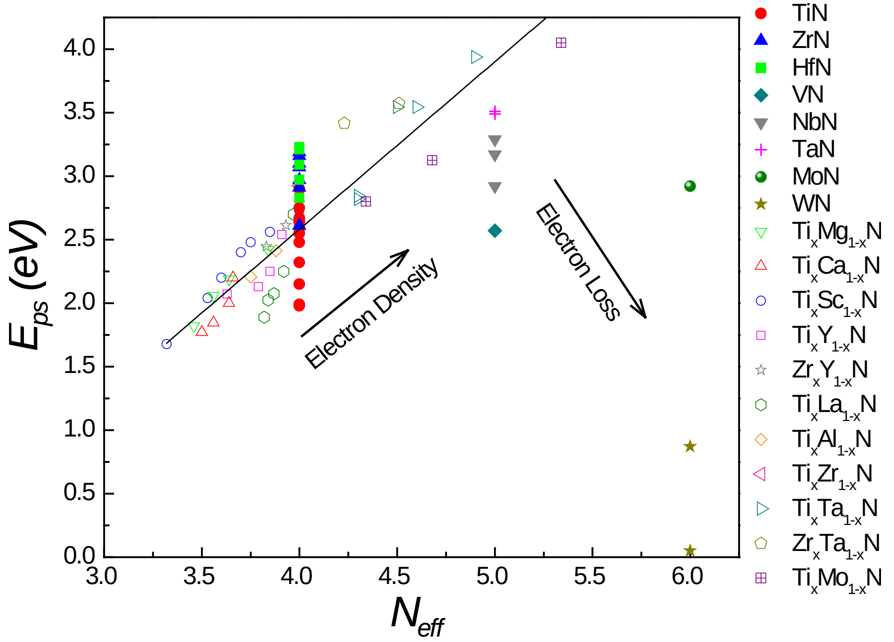
3) After eliminating B1-WN, **B1-TaN** should be the next **best UV plasmonic conductor.**
Is it?

UV Plasmonics and Photonics

$$\tilde{\varepsilon}(\omega) = \varepsilon_\infty - \frac{\omega_{pu}^2}{\omega^2 - i\Gamma_D\omega} + \sum_{j=1}^{n} \frac{f_j \cdot \omega_j^2}{\omega_j^2 - \omega^2 + i\gamma_j\omega}$$

$$\rho = \left(\frac{1}{\varepsilon_0}\right)\frac{\hbar^2 \cdot \Gamma_D}{E_{pu}^2}$$



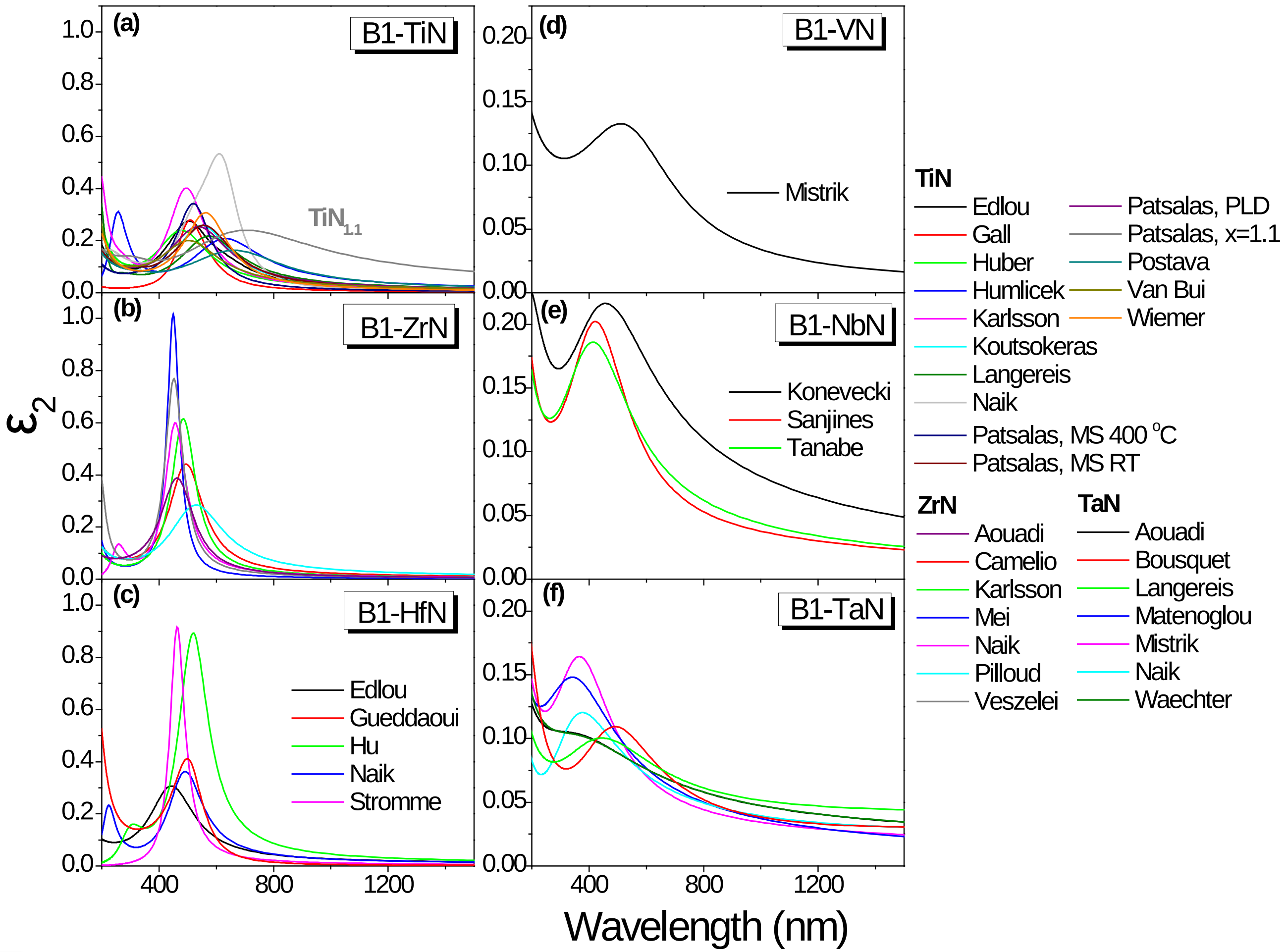


TaN, MoN, and WN are exceptionally hard to stabilize in the B1 structure; as a result, **B1-NbN** emerges as the best candidate among binary nitrides for UV applications.

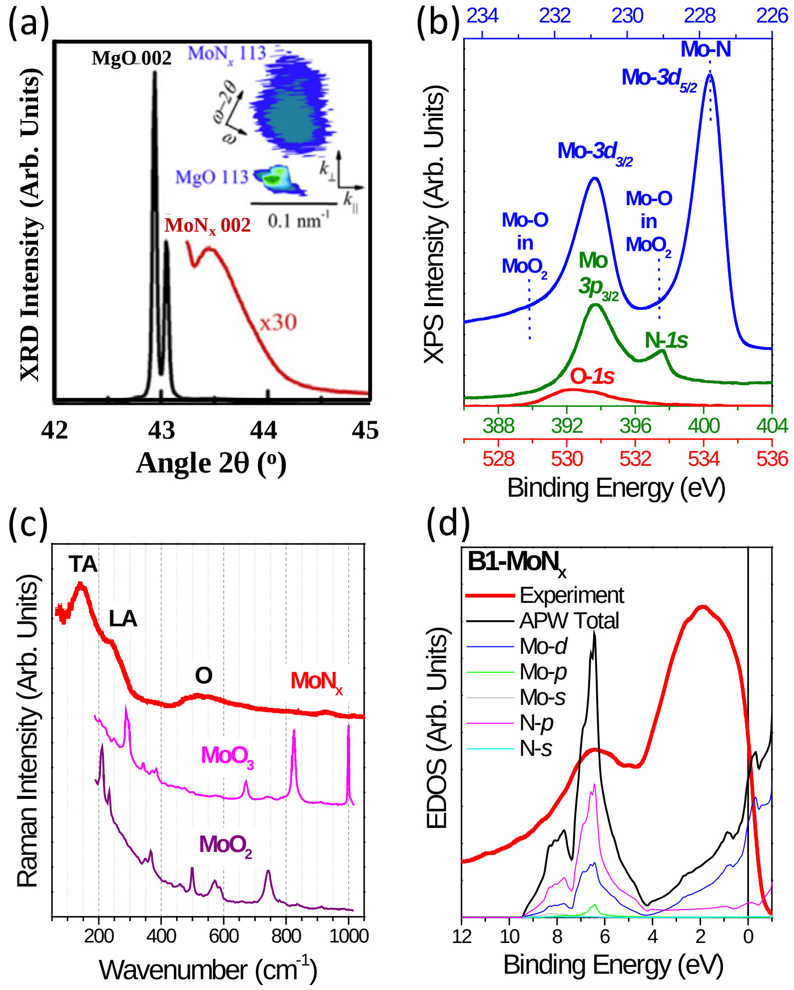
Me-rich $Ti_xMe_{1-x}N$ (Me=Ta, Mo) ternaries can also compete

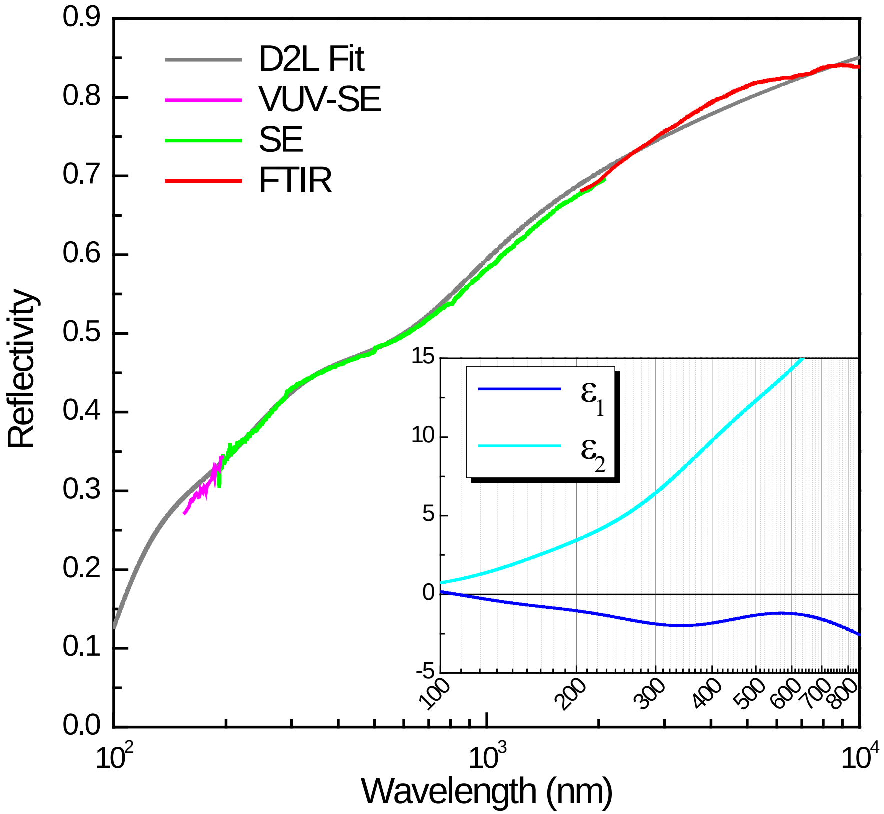
LSPR Spectra of Binary Nitrides: Going towards UV

MG-EMA: $$\varepsilon_{\text{eff}} = \varepsilon_m \frac{2\delta_i(\varepsilon_i-\varepsilon_m)+\varepsilon_i+2\varepsilon_m}{2\varepsilon_m+\varepsilon_i+\delta_i(\varepsilon_m-\varepsilon_i)}$$

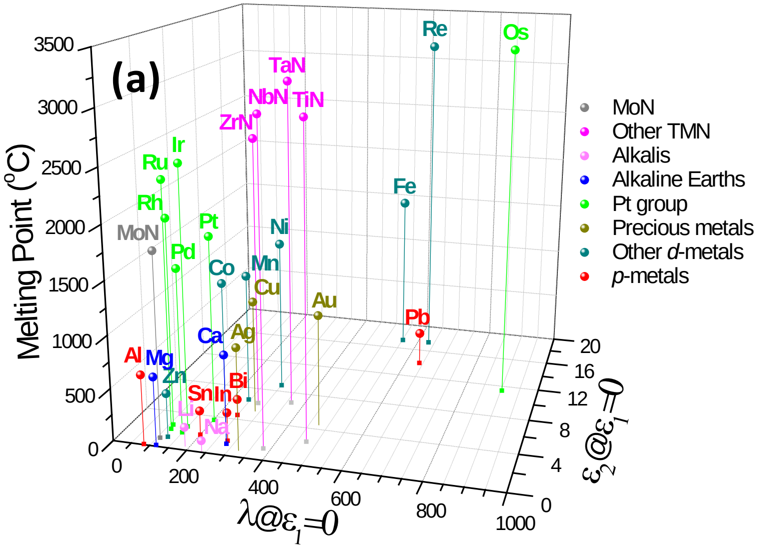


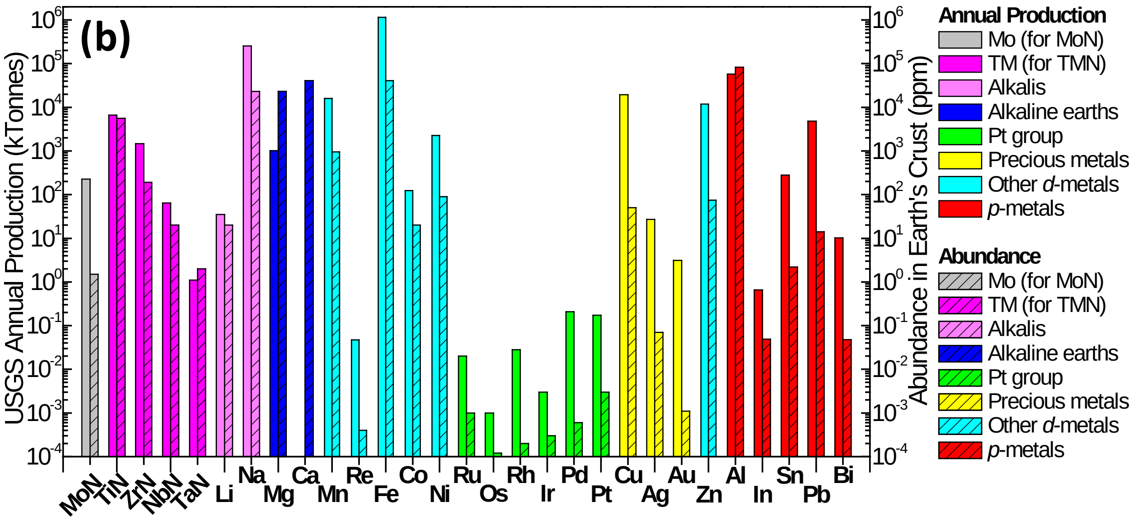
The stunning B1-MoN: defect-stabilized



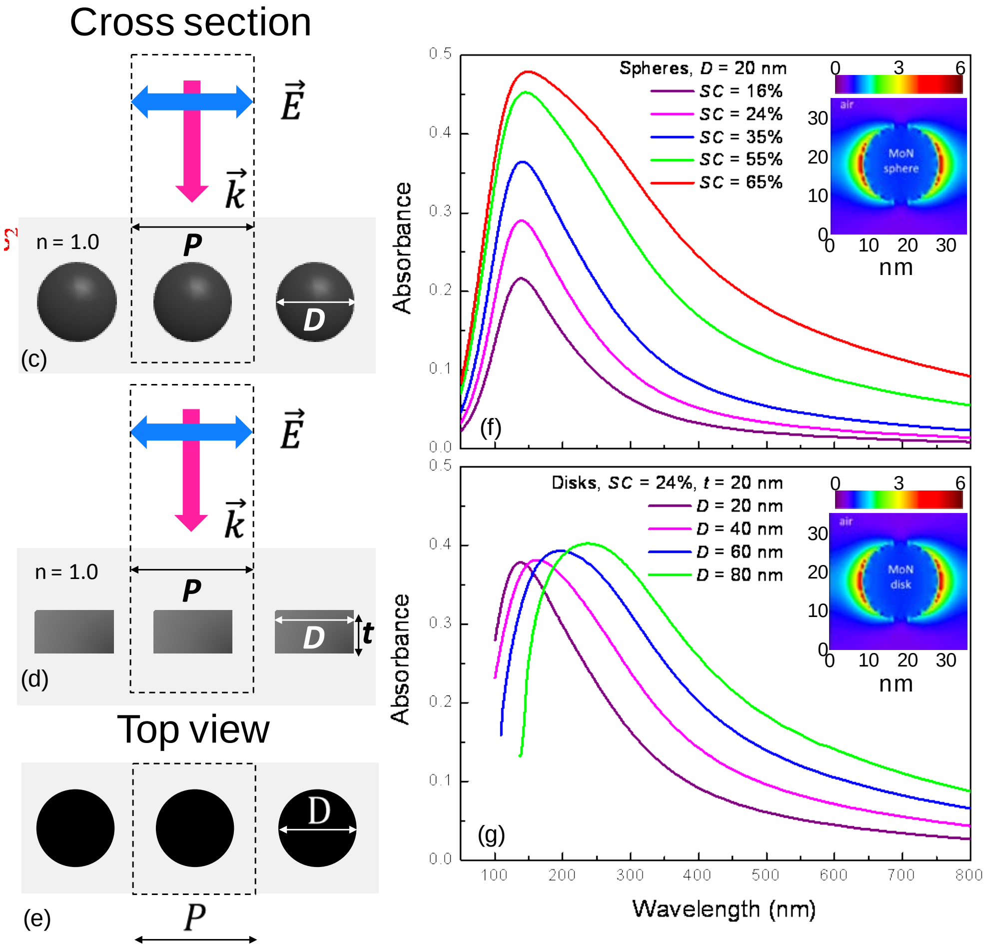


The stunning B1-MoN: defect-stabilized





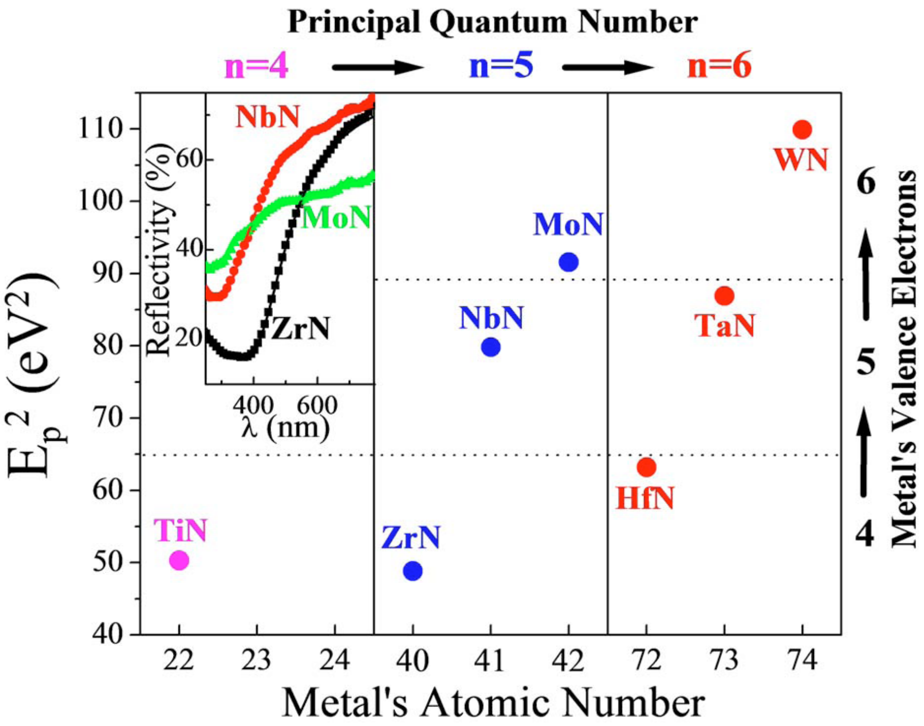
The stunning B1-MoN: defect-stabilized

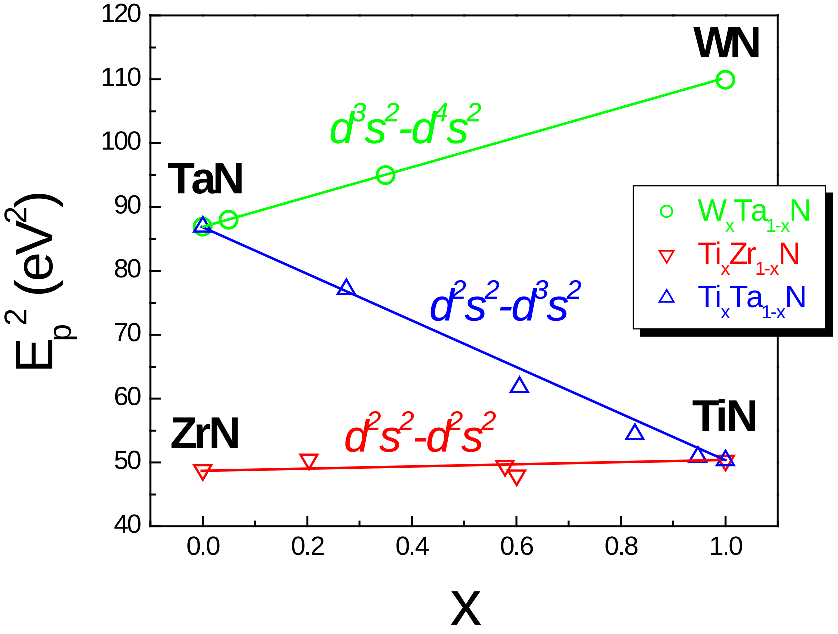


TMN-based compounds: Going towards IR and stabilizing UV

- Tune the electronic properties, such as carrier density and dielectric losses, to control the plasmonic response
- Tune the work function for hot electron applications
- Improvement of the microstructure and the structural stability of the B1 phase (*e.g.* by microstructure change from columnar to globular, stabilization of the B1 structure for TaN, and WN, *etc*)

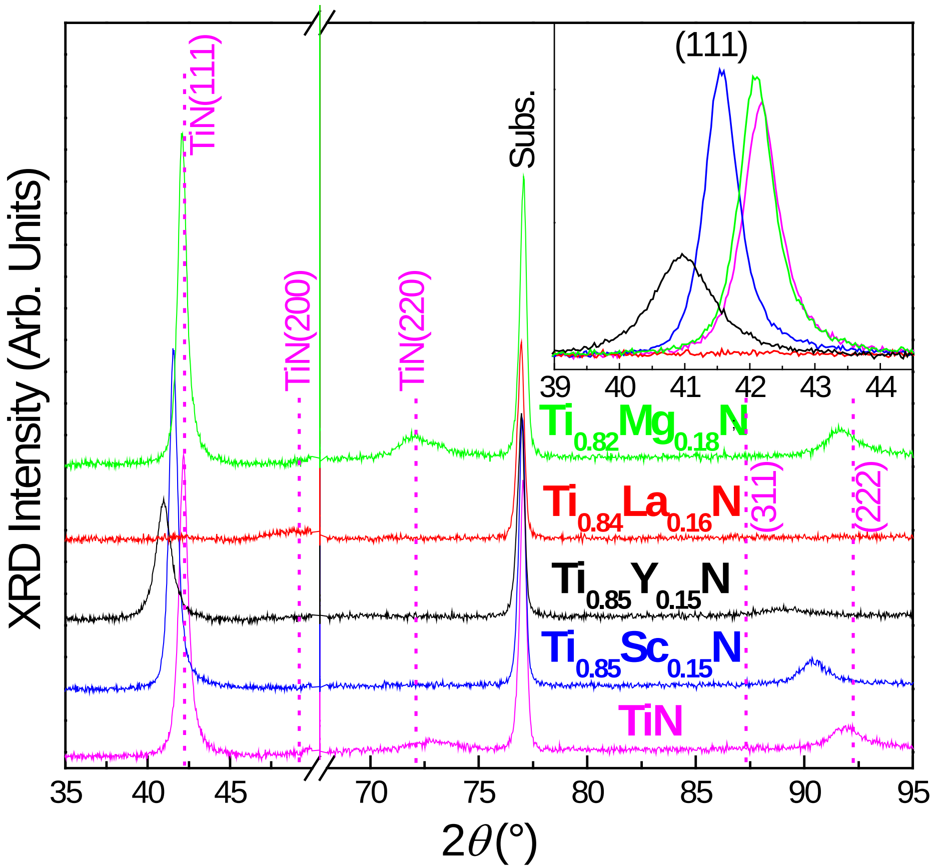
Tuning of the conduction electron density: towards UV response



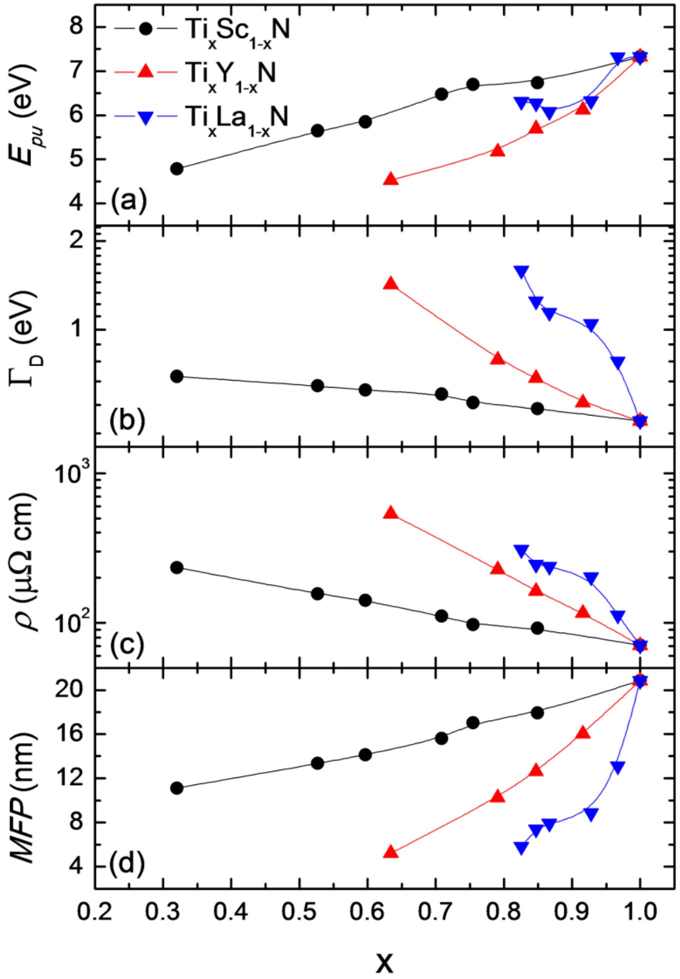


G.M. Matenoglou *et al*, Appl. Phys. Lett. 94, 152108 (2009)

B1 Ternary TMN – The lattice match effect: $Ti_xSc_{1-x}N$, $Ti_xY_{1-x}N$, $Ti_xLa_{1-x}N$, $Zr_xY_{1-x}N$

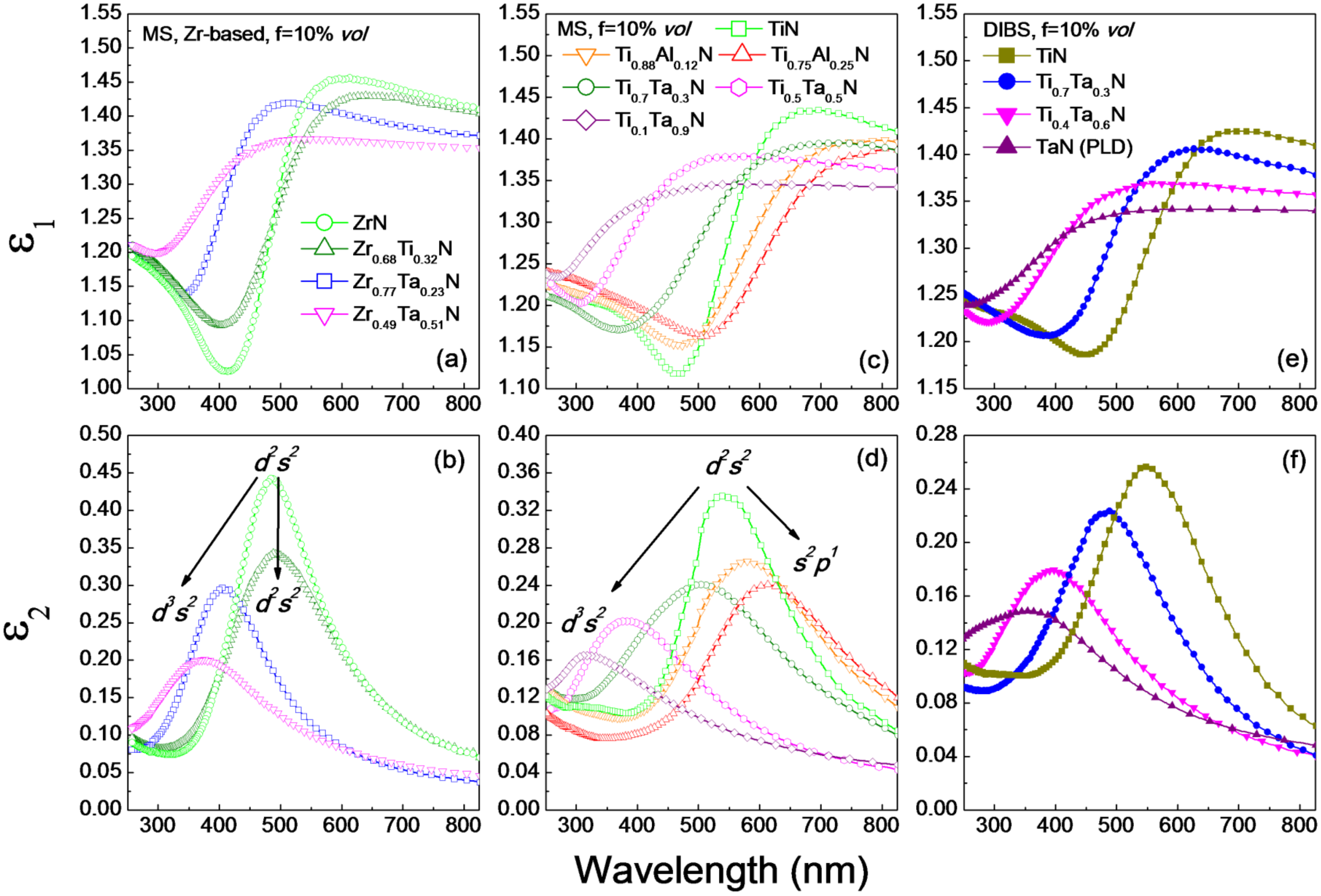


No XRD fine structure
Formation of Ternary Nitride (Solid Solution)



The best structural quality and the less optical loss is observed so far for $Ti_xSc_{1-x}N$

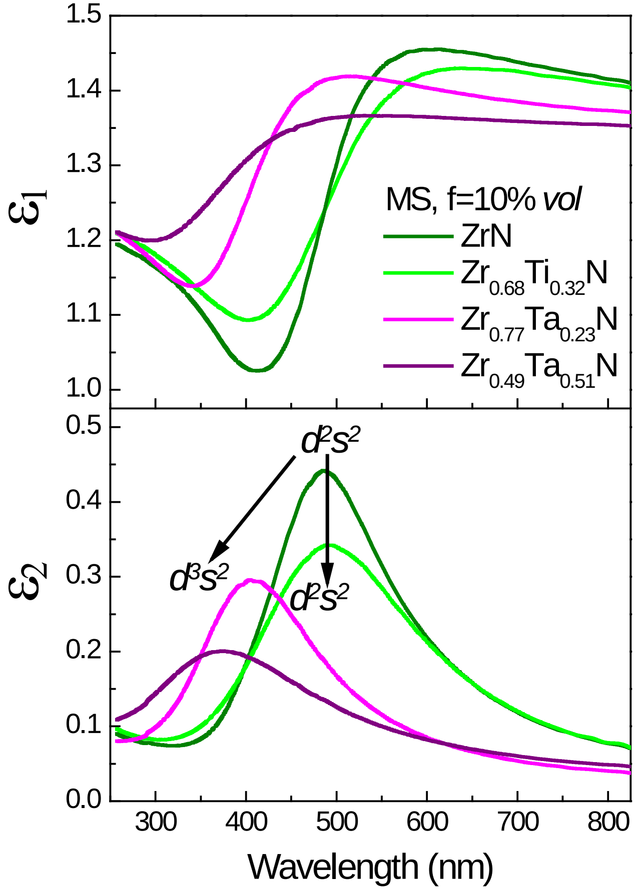
LSPR Spectra of Ti-based Ternary Nitrides: From Red to UV



LSPR Spectra of Zr-based Ternary Nitrides

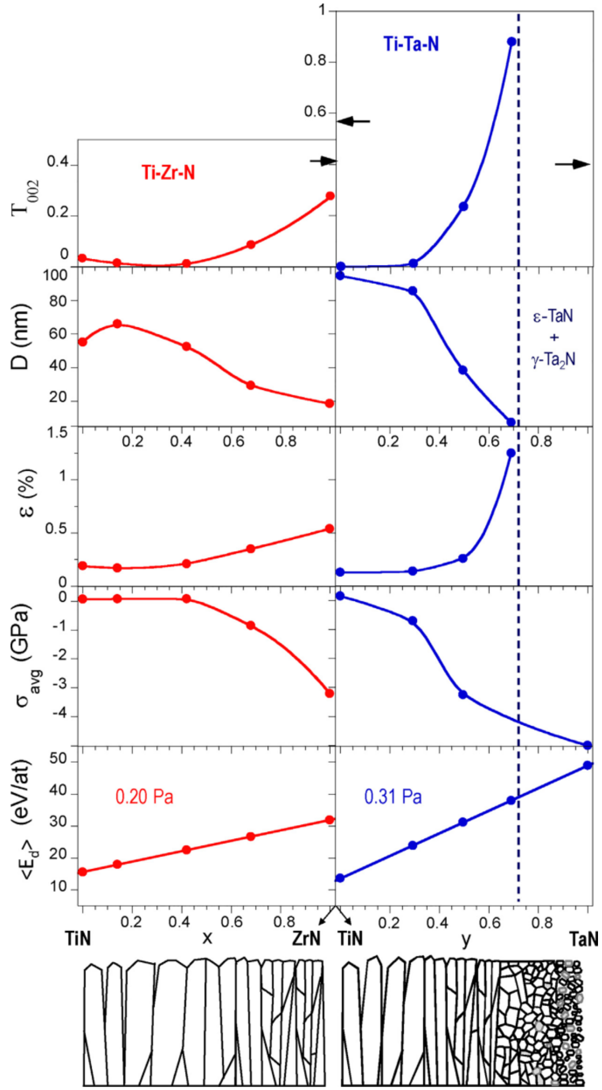
MG-EMA:

$$\varepsilon_{\mathrm{eff}} = \varepsilon_m \frac{2\delta_i(\varepsilon_i - \varepsilon_m) + \varepsilon_i + 2\varepsilon_m}{2\varepsilon_m + \varepsilon_i + \delta_i(\varepsilon_m - \varepsilon_i)}$$

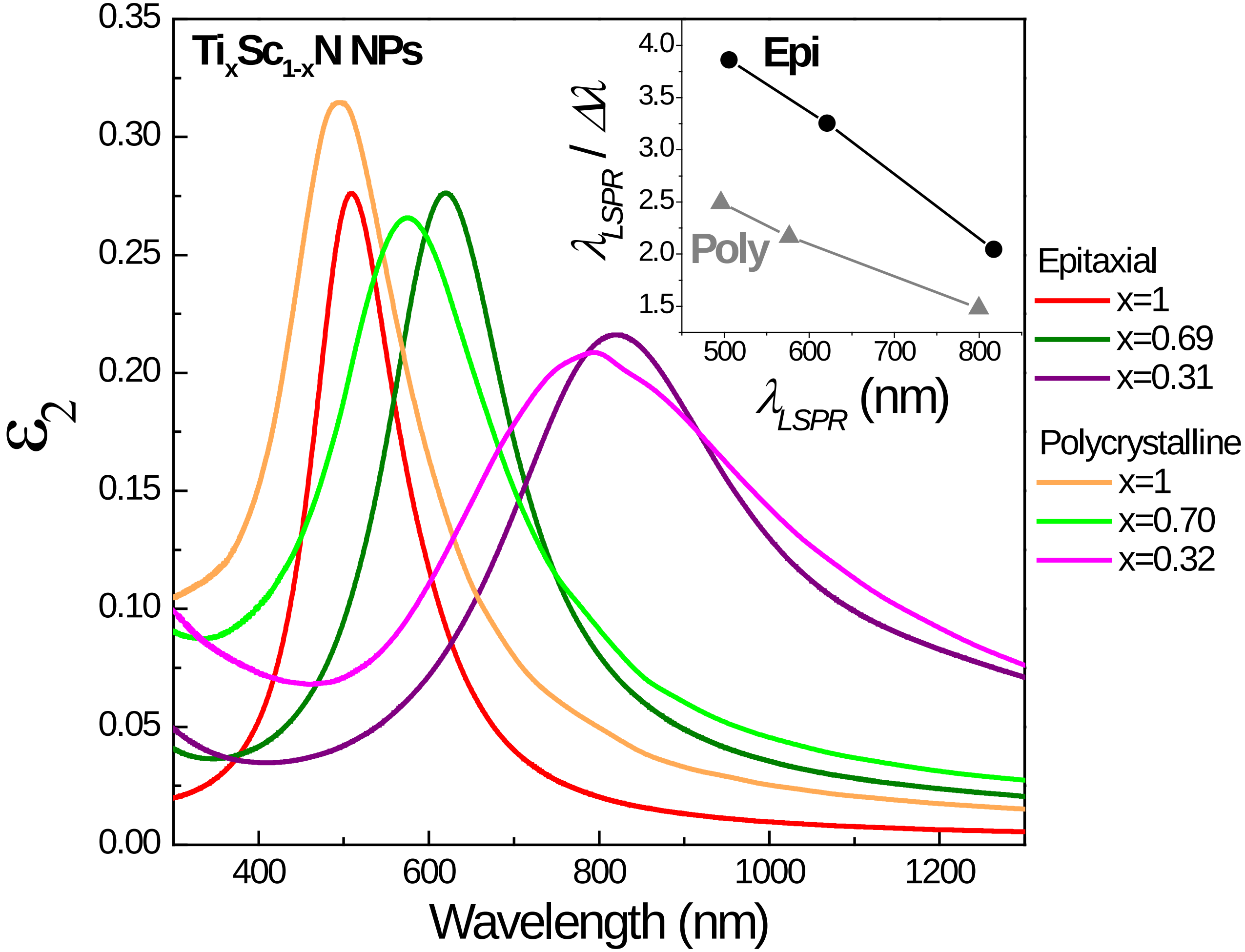


Although ZrN **IS** be the best plasmonic nitride for the vis range,

it is not the recommended blending element due to the large lattice size of ZrN

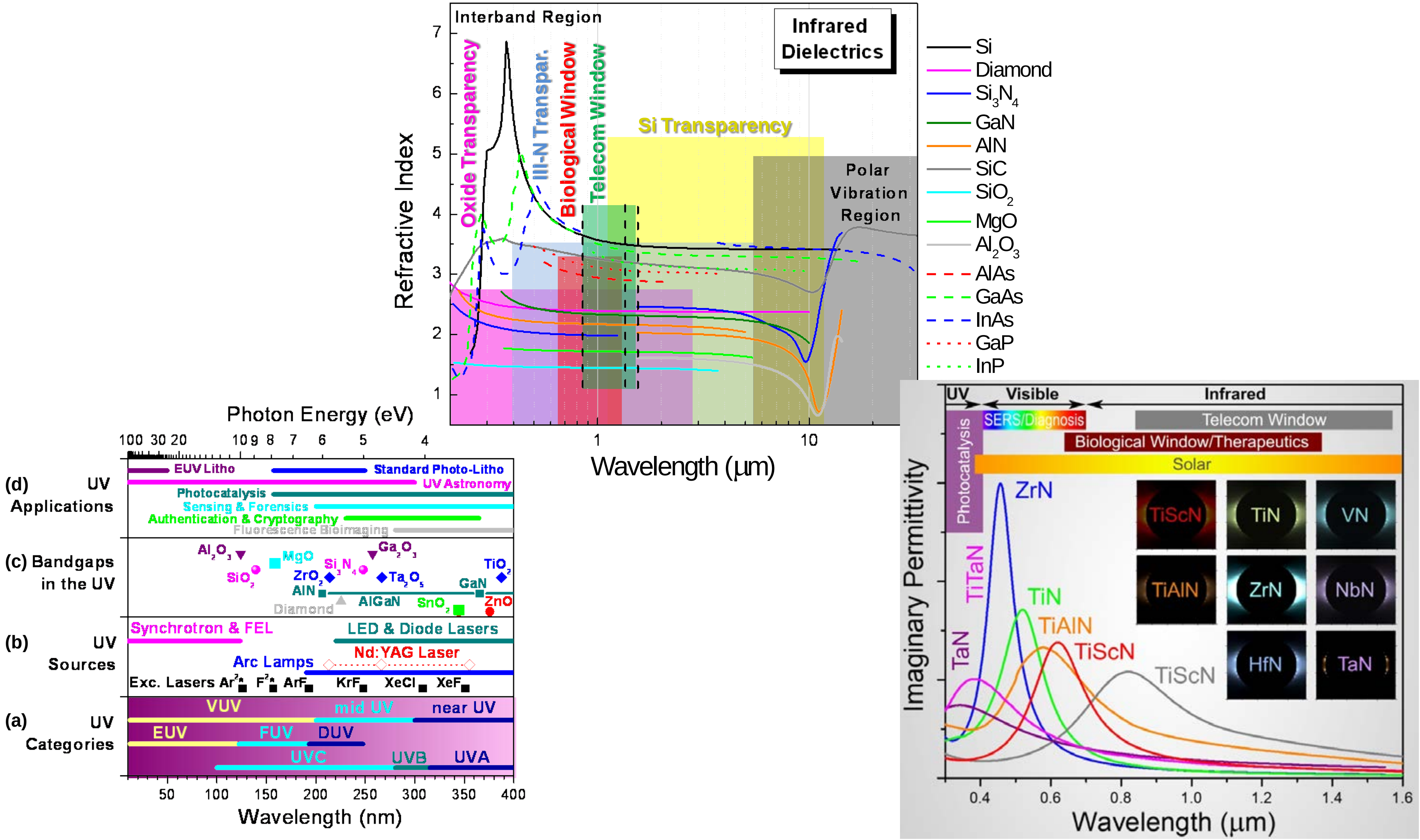


LSPR Spectra of Ti-based Ternary Nitrides: From Green to IR

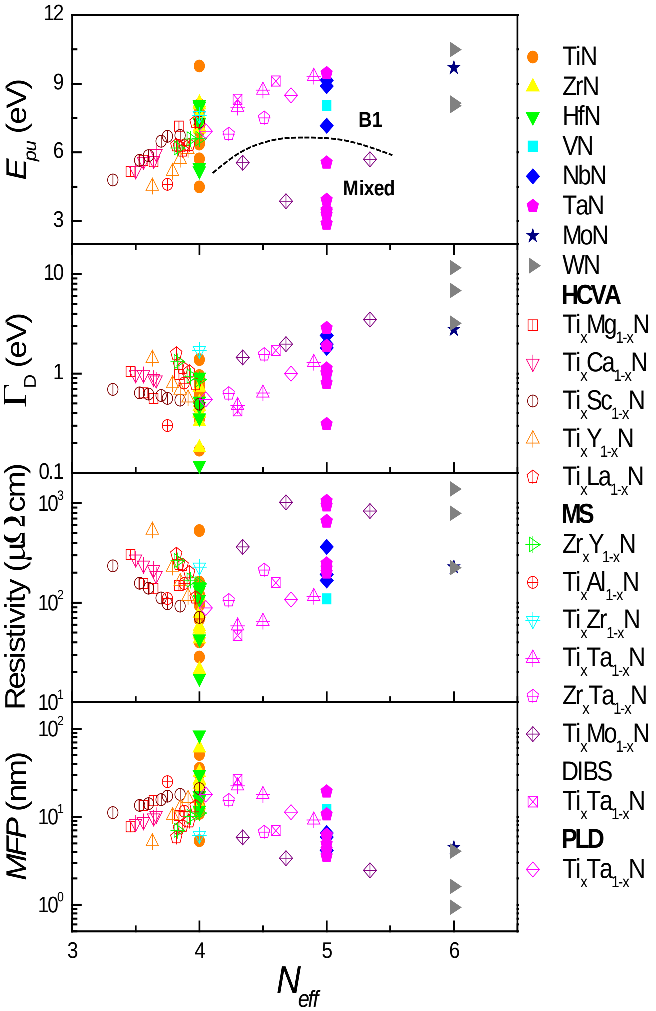


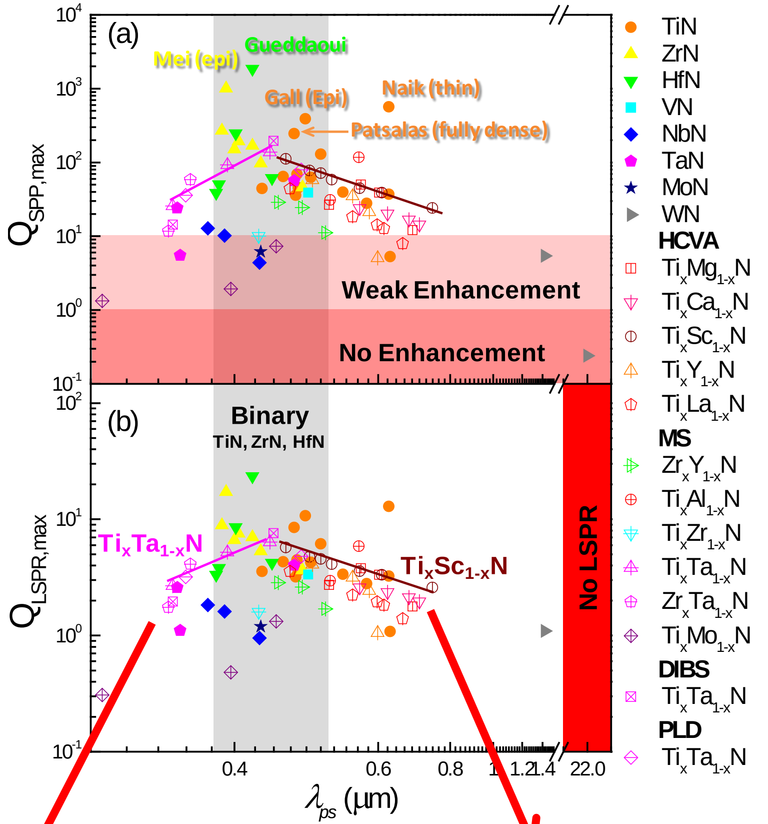
Institut Jean Lamour
PENSER LES MATÉRIAUX DE DEMAIN

Spectral ranges



An overall assessment of the current technology of plasmonic nitrides

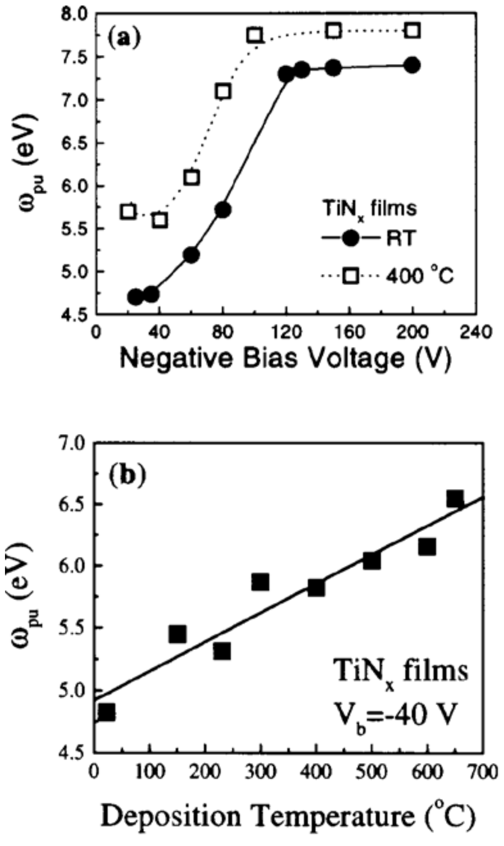




Mostly extrinsic (structure and growth related)

Combination of intrinsic (physically increased Γ_D) and extrinsic (structure and growth related)

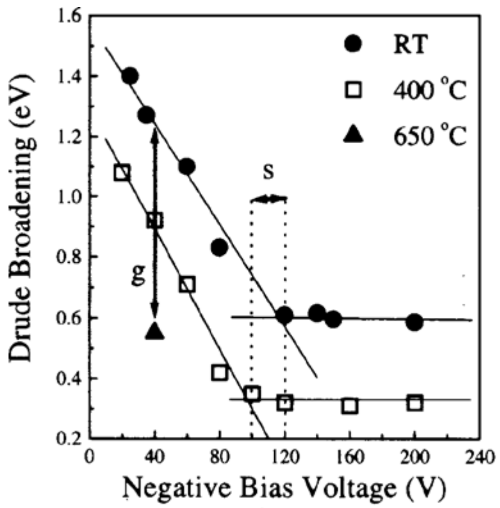
The refractory character: Implications to growth and optical properties

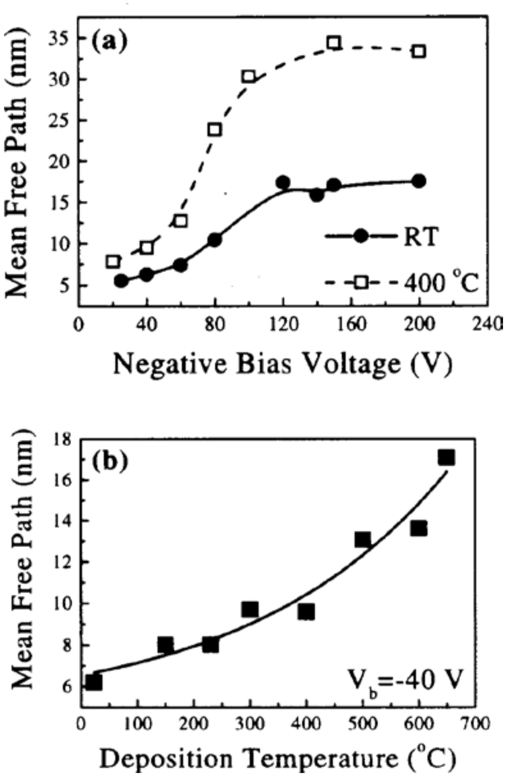


JOURNAL OF APPLIED PHYSICS VOLUME 90, NUMBER 9 1 NOVEMBER 2001

Optical, electronic, and transport properties of nanocrystalline titanium nitride thin films

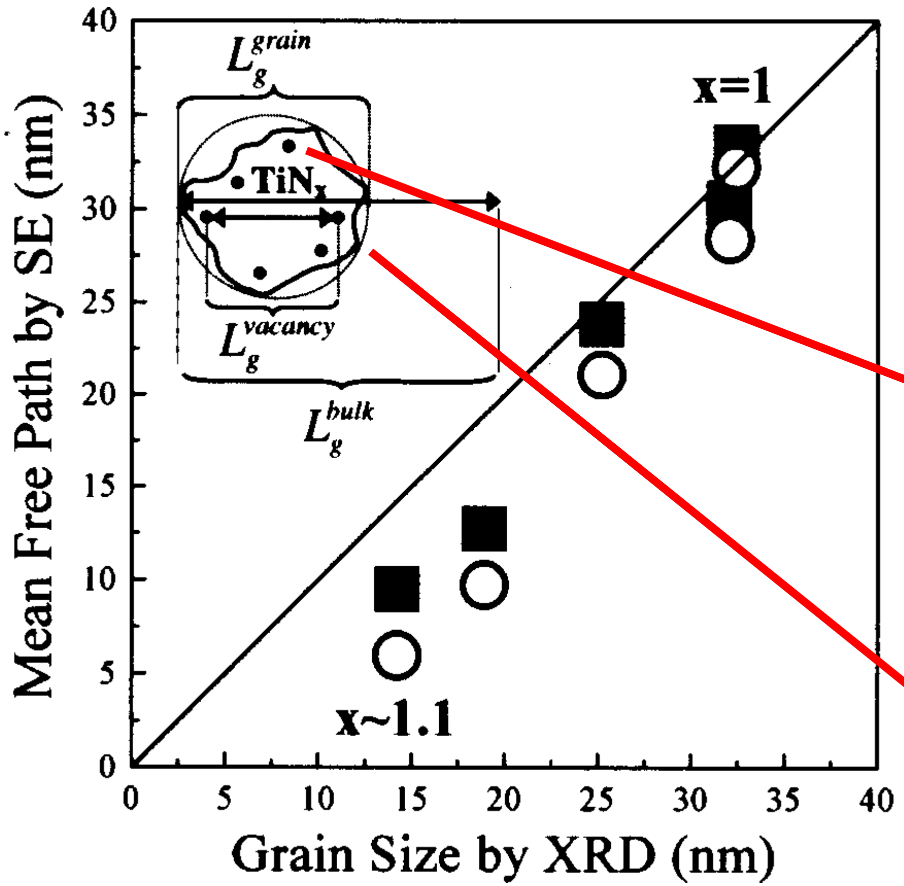
P. Patsalas[a] and S. Logothetidis





RT growth results in fine grains and porosity, and consequently to poor optical properties. Thus, sufficiently conductive nitrides are not compatible with self-assembly and mild lithography techniques, such as nanosphere lithography and makes RIE necessary.

What defects really do?




JOURNAL OF APPLIED PHYSICS VOLUME 90, NUMBER 9 1 NOVEMBER 2001

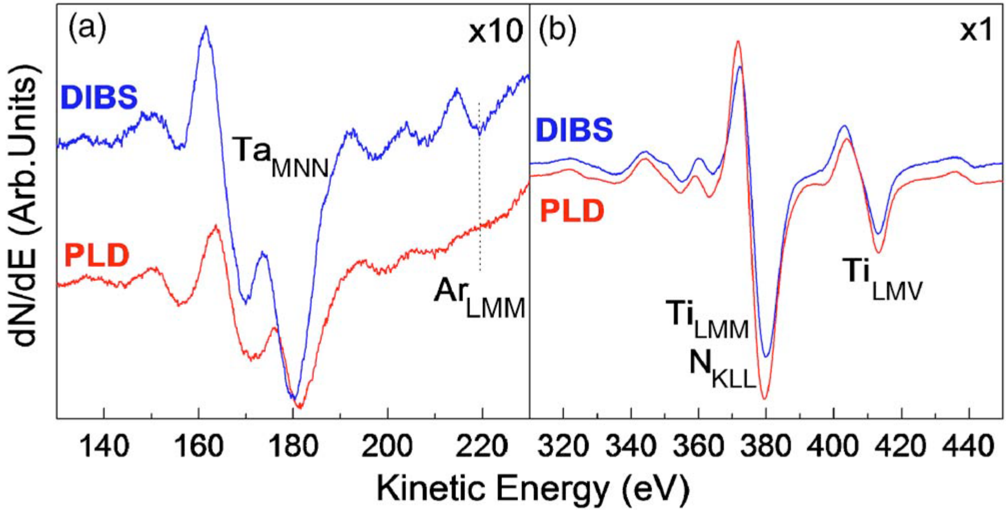
Optical, electronic, and transport properties of nanocrystalline titanium nitride thin films

P. Patsalas[a] and S. Logothetidis

Point defects: scattering of electrons AND trapping of them. *i.e.* varying carrier density. That’s why many groups reported TiN with plasmonic behavior in the IR

Extended defects: exclusively scattering of electrons

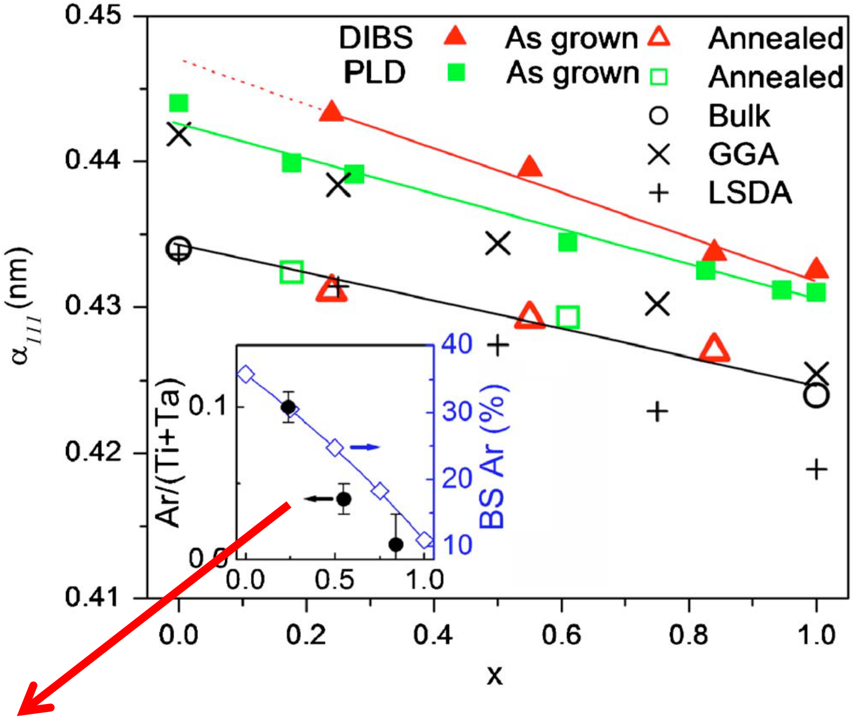
Sputter deposition: Backscattered Ar^+



APPLIED PHYSICS LETTERS 93, 011904 (2008)

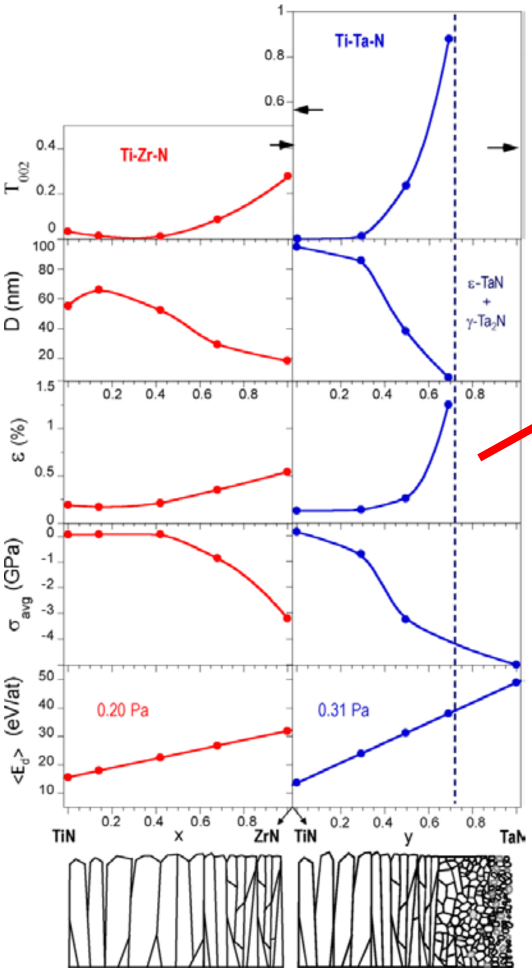
Conducting transition metal nitride thin films with tailored cell sizes: The case of δ-$Ti_xTa_{1-x}N$

L. E. Koutsokeras,[1,2] G. Abadias,[2] Ch. E. Lekka,[1] G. M. Matenoglou,[1] D. F. Anagnostopoulos,[1] G. A. Evangelakis,[3] and P. Patsalas[1,a)]

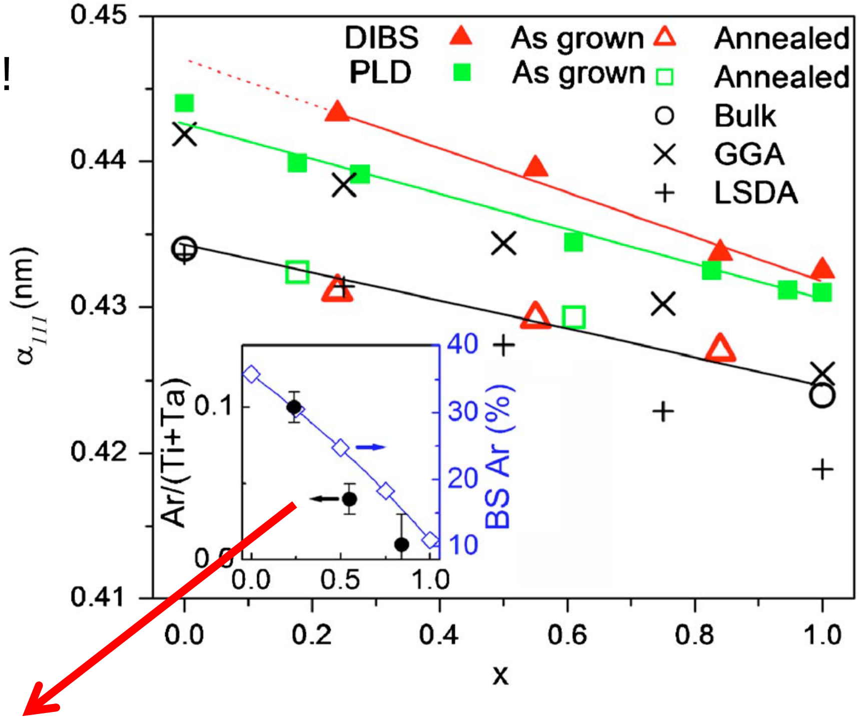


SRIM Calculations: The heavier the target atoms the more the backscattered Ar^+ trapped into the grown nitride

Sputter deposition: Backscattered Ar^+

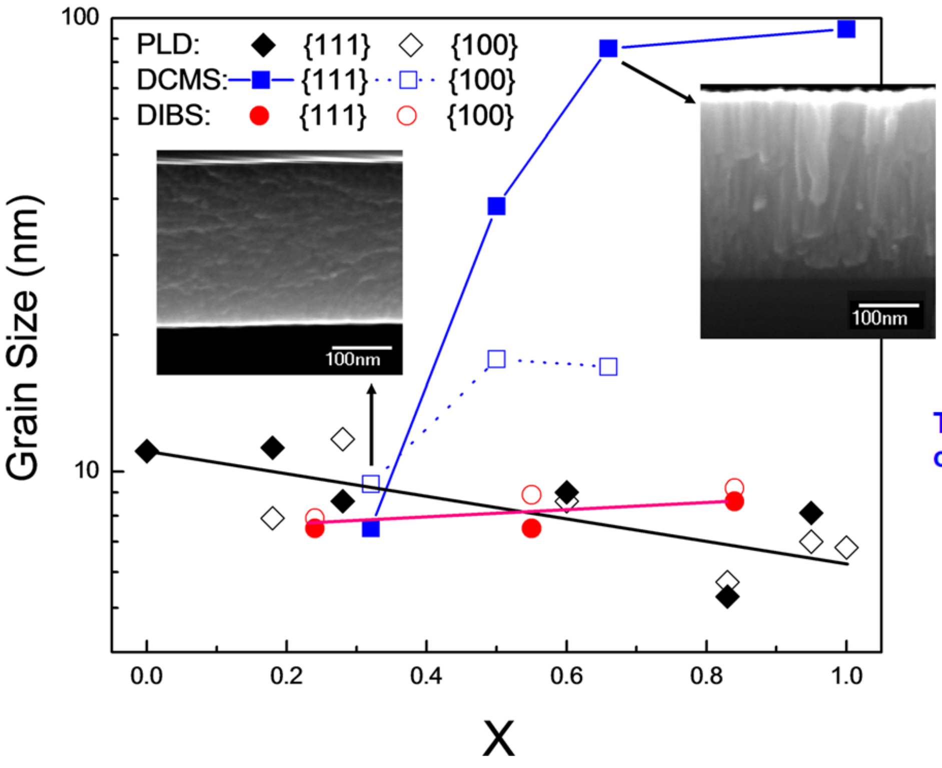


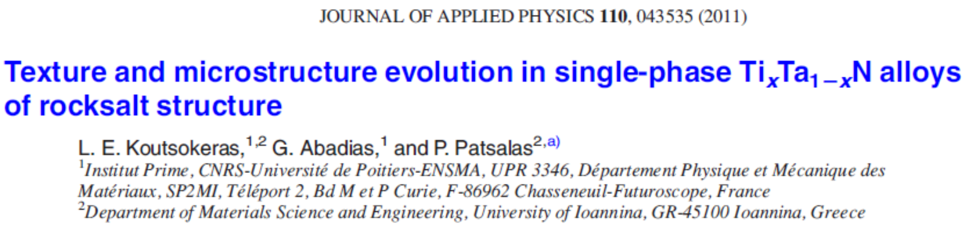
More point defects!



SRIM Calculations: The heavier the target atoms the more the backscattered Ar^+ trapped into the grown nitride

Sputter deposition *vs.* PLD (no Ar$^+$)




JOURNAL OF APPLIED PHYSICS **110**, 043535 (2011)

Texture and microstructure evolution in single-phase Ti$_x$Ta$_{1-x}$N alloys of rocksalt structure

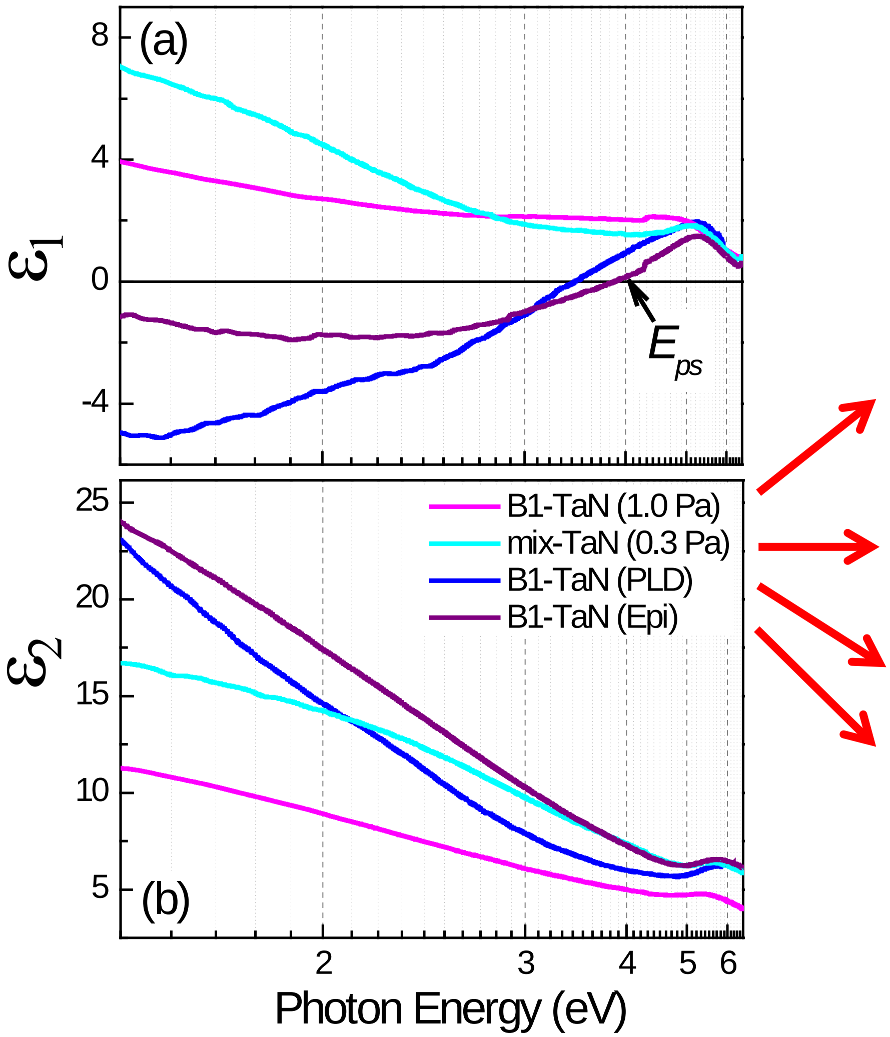
L. E. Koutsokeras,[1,2] G. Abadias,[1] and P. Patsalas[2,a)]

[1] *Institut Prime, CNRS-Université de Poitiers-ENSMA, UPR 3346, Département Physique et Mécanique des Matériaux, SP2MI, Téléport 2, Bd M et P Curie, F-86962 Chasseneuil-Futuroscope, France*

[2] *Department of Materials Science and Engineering, University of Ioannina, GR-45100 Ioannina, Greece*

For PLD the grain size increases with the Ta-content, *i.e.* the grain refinenement observed in both sputtering configurations is confirmed to be due to backscattering of Ar$^+$

Consequences to the optical properties: the case of TaN



The film with extended defects outperforms the sample with point defects!

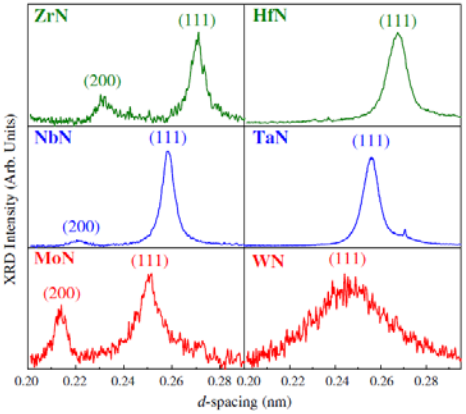
High pressure = reduced BS due to gas collisions, but low surface diffusion = small grains and O impurities

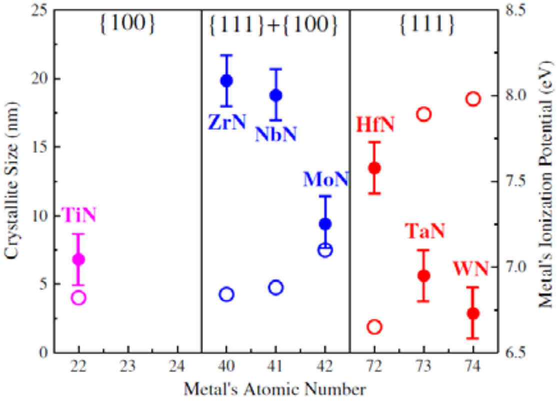
Low pressure on Si = high BS = high density of point defects and extended defects

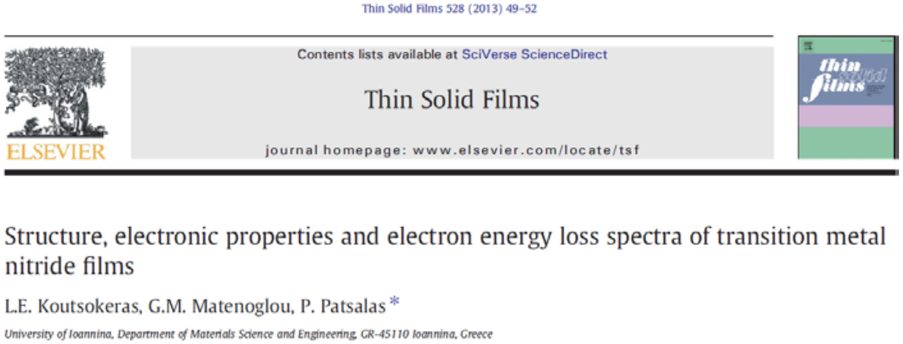
PLD = no BS = mostly extended defects

Low pressure on MgO = high BS but epitaxial films = high density of point defects and low density of extended defects

Defects beyond backscattering: PLD







Thin Solid Films 528 (2013) 49–52

Contents lists available at SciVerse ScienceDirect

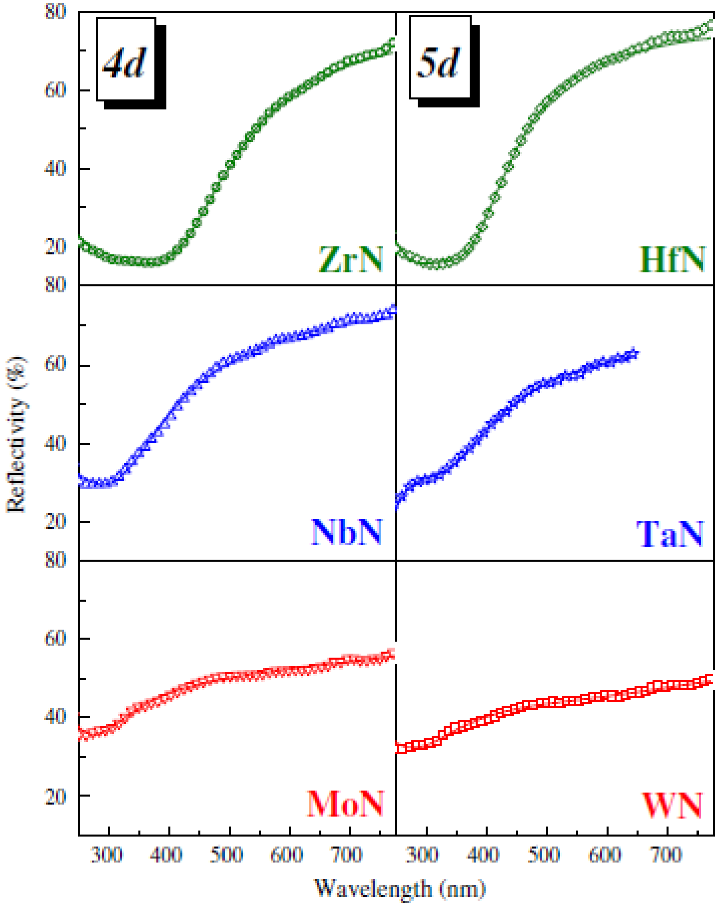
Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Structure, electronic properties and electron energy loss spectra of transition metal nitride films

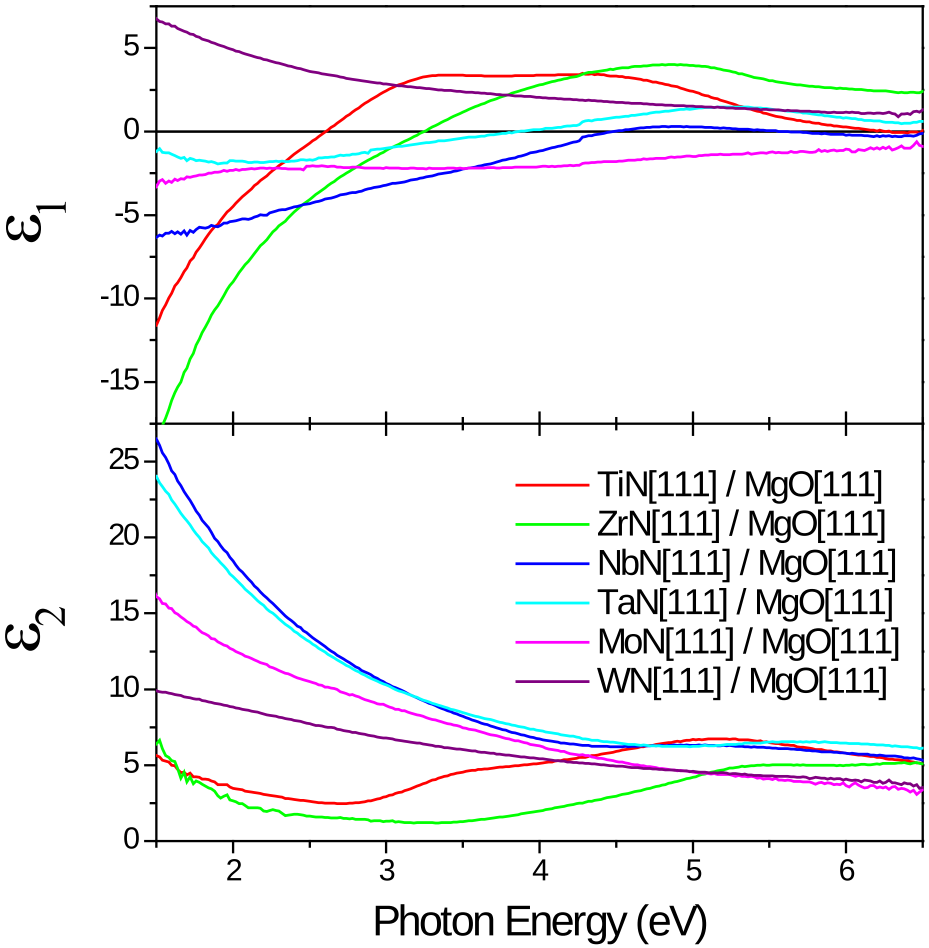
L.E. Koutsokeras, G.M. Matenoglou, P. Patsalas *

University of Ioannina, Department of Materials Science and Engineering, GR-45110 Ioannina, Greece

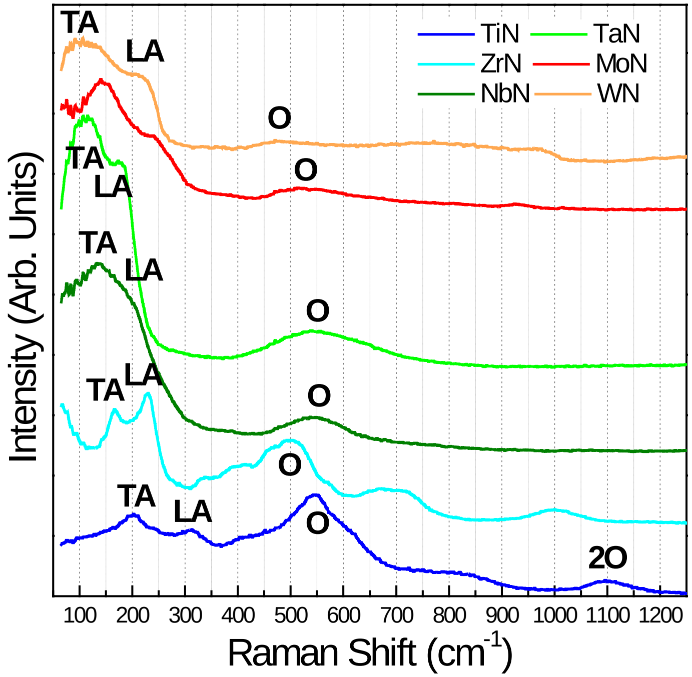


Even without BS increasing the period number (*i.e.* the mass of the metal) we do report variations of grain size even for films strictly stoichiometric ([N]/[Me]=1), mostly associated with surface diffusion length of deposited adatoms.

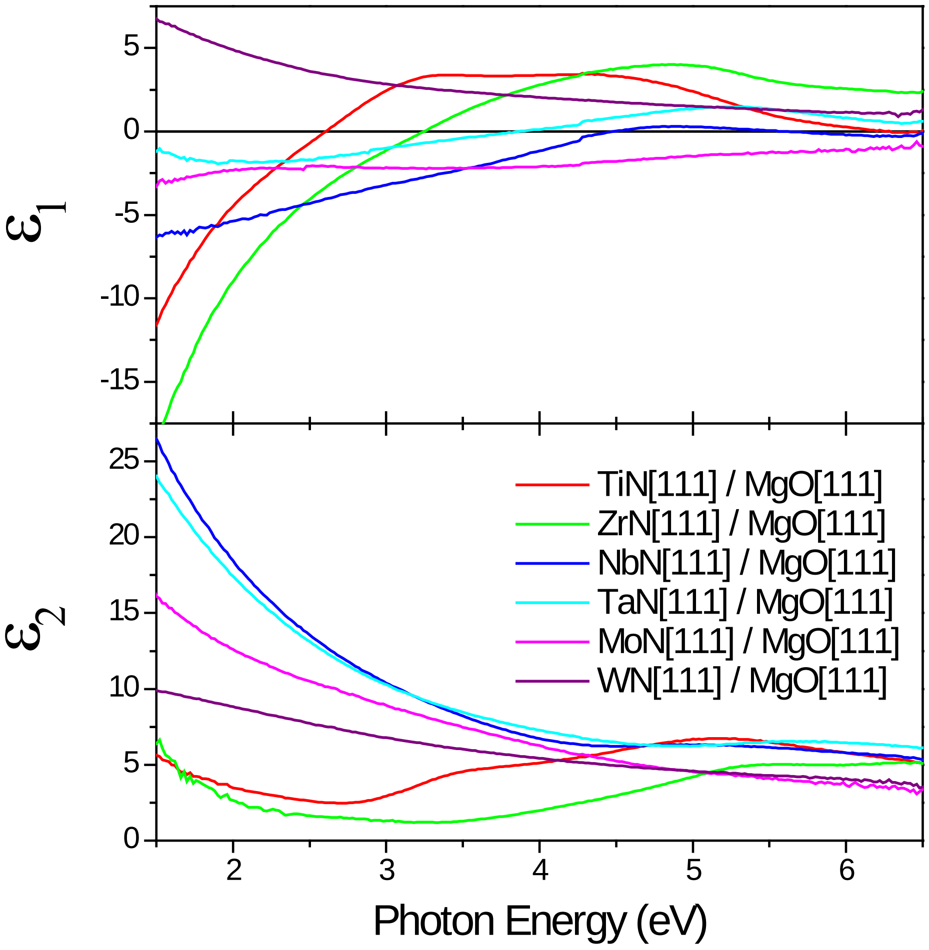
Optical performance of epitaxial sputtered nitrides



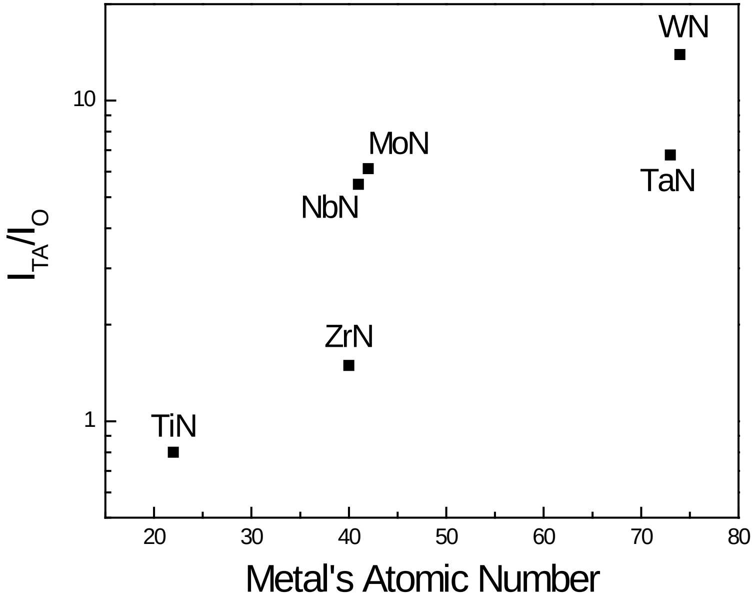
Period 5 nitrides (Zr, Nb, Mo) outperform period 6 nitrides (Ta, W)



Optical performance of epitaxial sputtered nitrides



Period 5 nitrides (Zr, Nb, Mo) outperform period 6 nitrides (Ta, W)

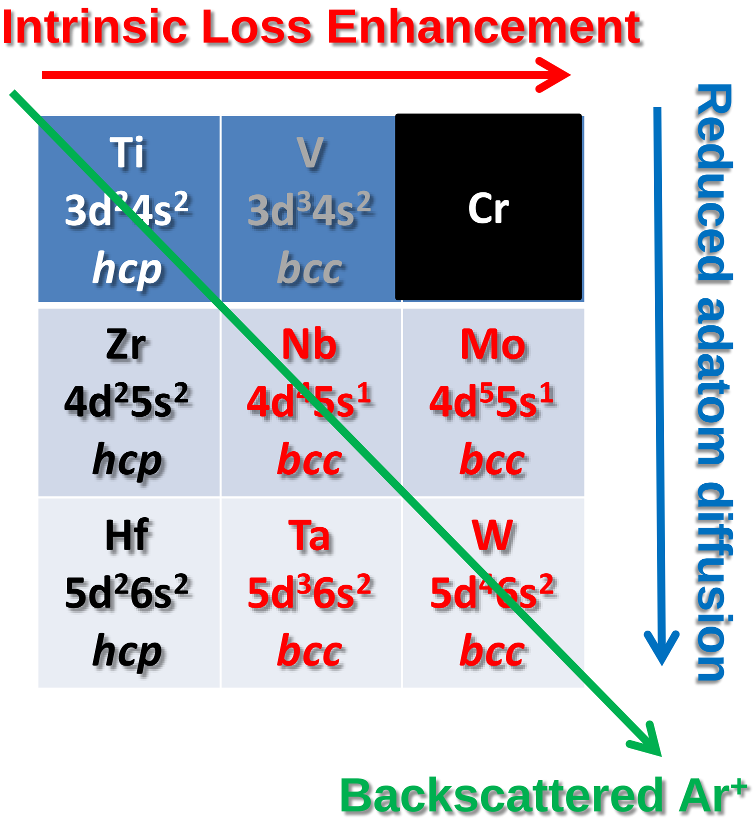


In most cases, the PLD polycrystalline films outperform the sputtered epitaxial films; further support of the more severe effect of point defects compared to extended defects.

Summary and outlook – Part II: Regarding the defects and optical loss

- We identified three electronic loss mechanisms that are manifested in TMN beyond TiN and ZrN:

  1. The intrinsic electronic loss increase with mostly group number but also secondarily with period number of the constituent metal, as revealed by the LAPW calculations
  2. The general tendency of reduced crystallinity, *i.e.* enhancement of extended defects, with the period number due to the limited surface diffusion of adatoms
  3. For sputtered growth the formation of point defects due to backscattered Ar^+



Intrinsic Loss Enhancement

Reduced adatom diffusion

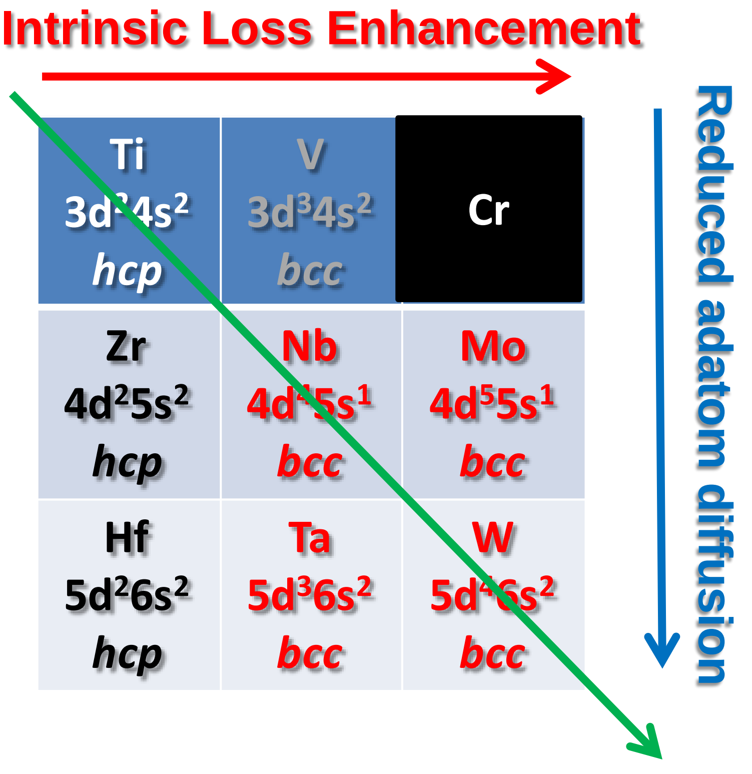
Backscattered Ar^+

Ti $3d^2 4s^2$ ***hcp***	**V** $3d^3 4s^2$ ***bcc***	**Cr**
Zr $4d^2 5s^2$ ***hcp***	**Nb** $4d^4 5s^1$ ***bcc***	**Mo** $4d^5 5s^1$ ***bcc***
Hf $5d^2 6s^2$ ***hcp***	**Ta** $5d^3 6s^2$ ***bcc***	**W** $5d^4 6s^2$ ***bcc***

Summary and outlook – Part II: Regarding the defects and optical loss

For low loss UV plasmonic nitrides (TaN, WN improved NbN, Mon), it is of utmost importance to minimize the point defects in the produced materials:

- Sputter deposition is usually accompanied by the Ar^+ BS: Use of other inert gases?
- Halide CVD: Halogen impurities which act as point defects and increase severely the resistivity
- MOCVD/ALD: Organic impurities which act as point defects and increase severely the resistivity
- PLD: Limited scaling up potential
- MBE: high melting point of the constituent metal and refractory products = extremely high temperatures



Ti $3d^2 4s^2$ *hcp*	**V** $3d^3 4s^2$ *bcc*	**Cr**
Zr $4d^2 5s^2$ *hcp*	**Nb** $4d^4 5s^1$ *bcc*	**Mo** $4d^5 5s^1$ *bcc*
Hf $5d^2 6s^2$ *hcp*	**Ta** $5d^3 6s^2$ *bcc*	**W** $5d^4 6s^2$ *bcc*

G.A. Olson, PhD, UIUC, 2016

We need to be creative!

Thank you for your attention!