15 years of Tsai lab

- From the eyes of a staff member -

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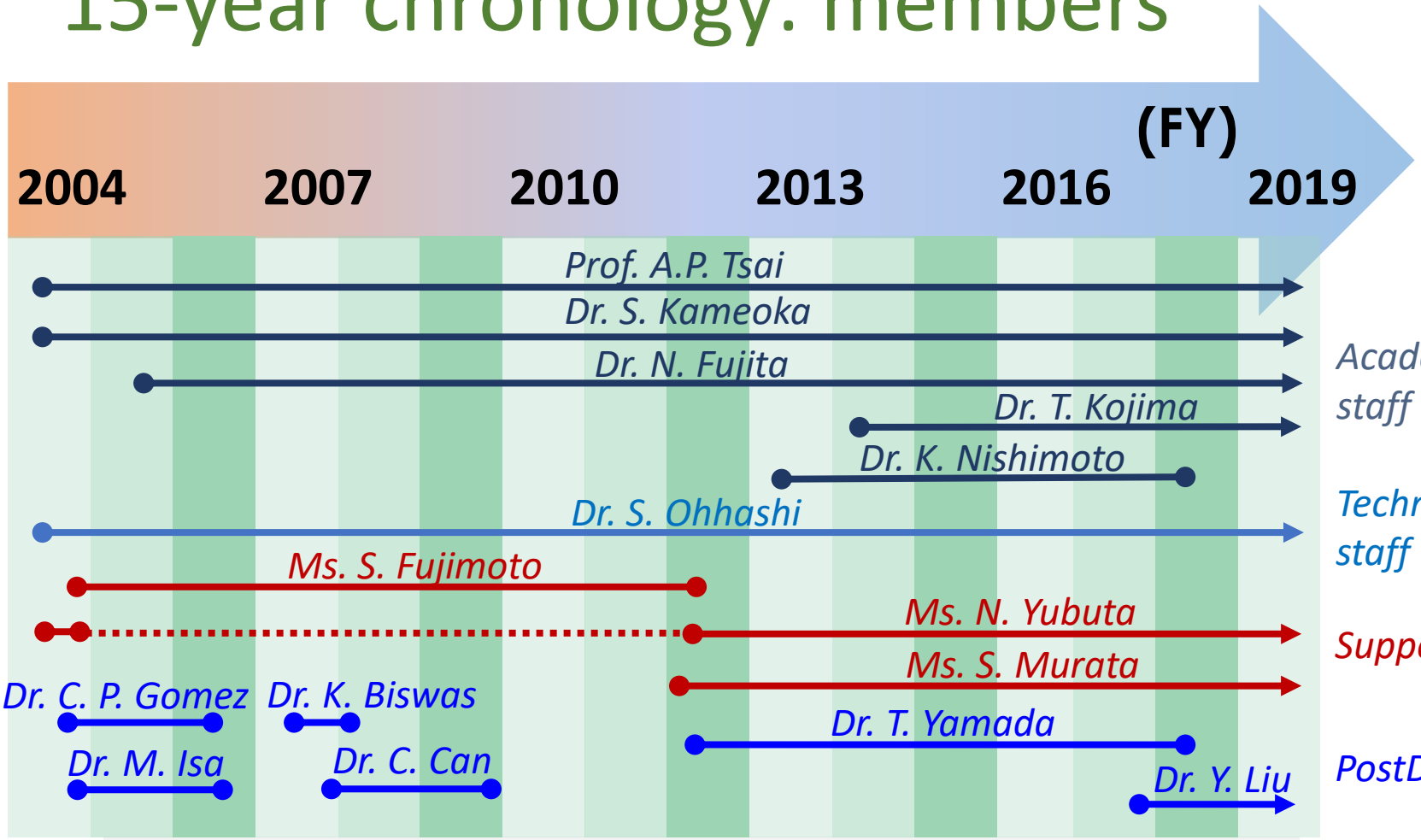
Prof. A.P. Tsai (my personal view)

- He was neither a physicist nor chemist, but metallurgist.
- He was an experimentalist by his nature, and one of the best experimentalists I ‘ve ever met.
- He was keen on something new & unexpected and on being a pioneer, but remained objective in thinking.
- He was humble and open minded in listening to his young colleagues & students.
- He allowed the lab members to have a lot of freedom and offered generous support for research.
- He was frank and honest when talking.

15-year chronology: members



Visiting scientists:
Prof. T. Janssen, Prof. P. Thiel, Prof. D. Shechtman, Prof. M. deBoissieu,
Prof. E. Belin-Ferre, Prof. J.M. Dubois, Prof. K. Chattopadhyay,
Prof. S.F. Wang, Prof. H.R. Sharma, Dr. C. Cui, Dr. G.H. Gebresenbut
Prof. S. Suzuki, Prof. H.R. Trebin, Dr. M. Mihalkovic, Prof. L.S. Hong

Staff members: combination of different disciplines (*experimental* !)

Prof. A.P. Tsai

Metallurgy

Dr. S. Kameoka

Catalyst chemistry

Dr. N. Fujita

Condensed matter physics (theory)

Dr. S. Ohhashi

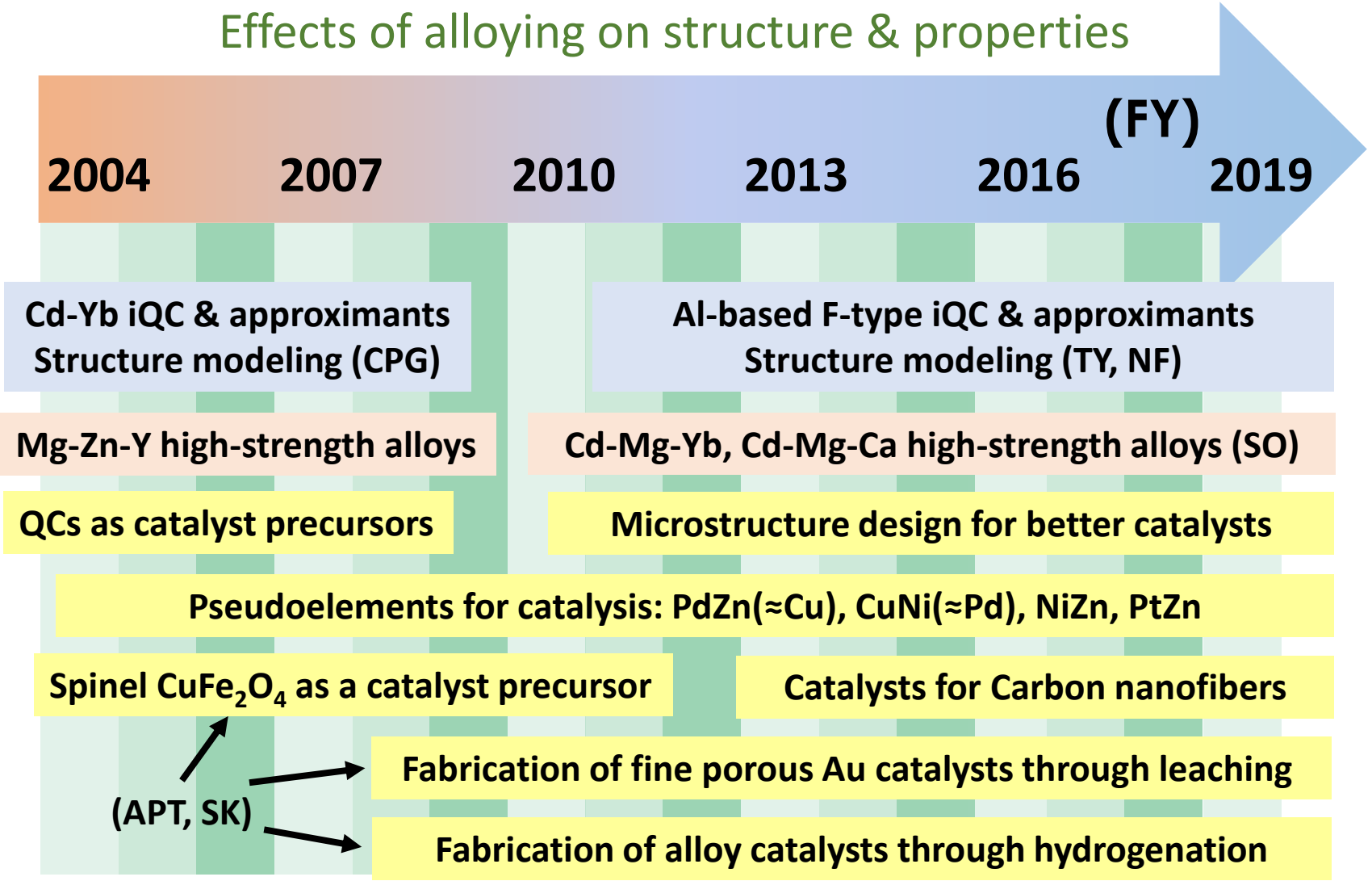
Technical staff: Synthesis of alloys, SEM, TEM

Dr. T. Kojima

Solid state catalysts, Magnetic materials, Metallic thin films

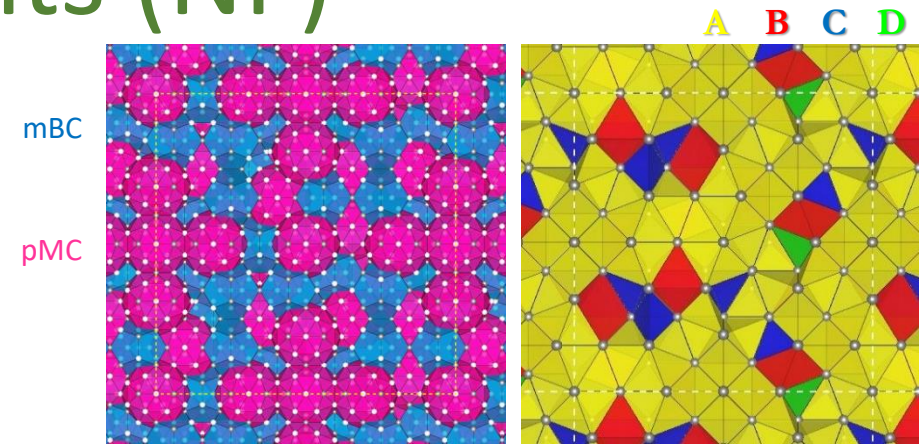
Research activities

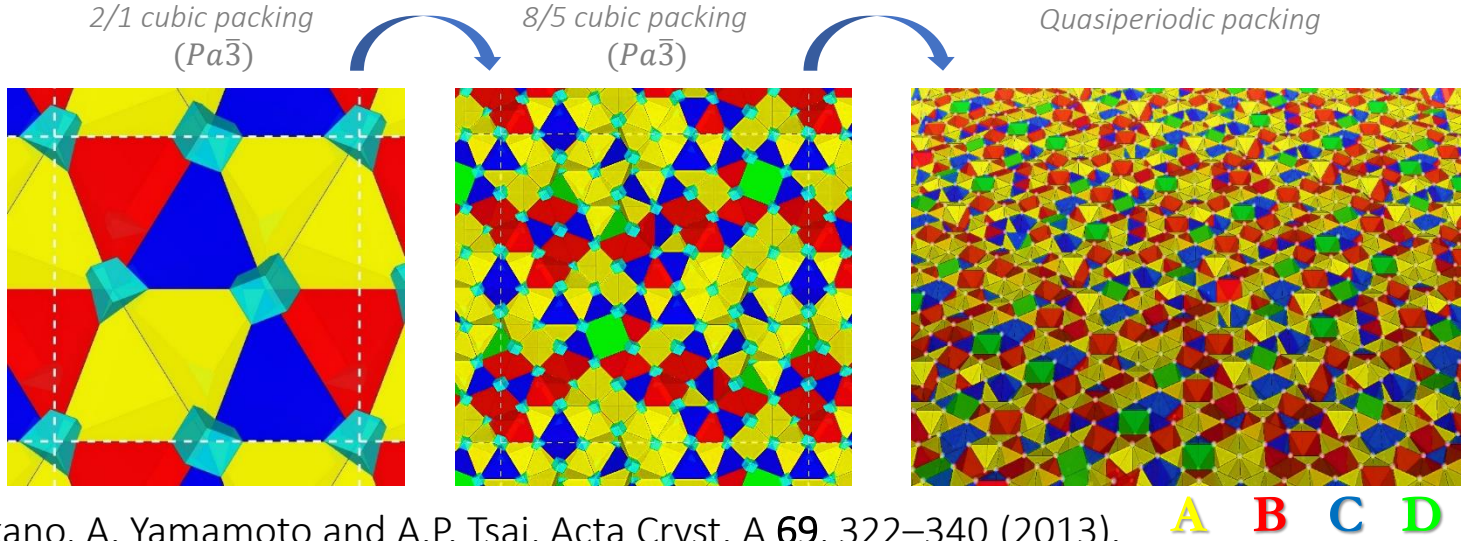
Effects of alloying on structure & properties



Clusters packing geometry in i-QC and approximants (NF)

We have renewed the way to understand the structure of Al-based F-type iQC & approximants as a packing of two kinds of small cluster centered at the nodes of a canonical-cell tiling.

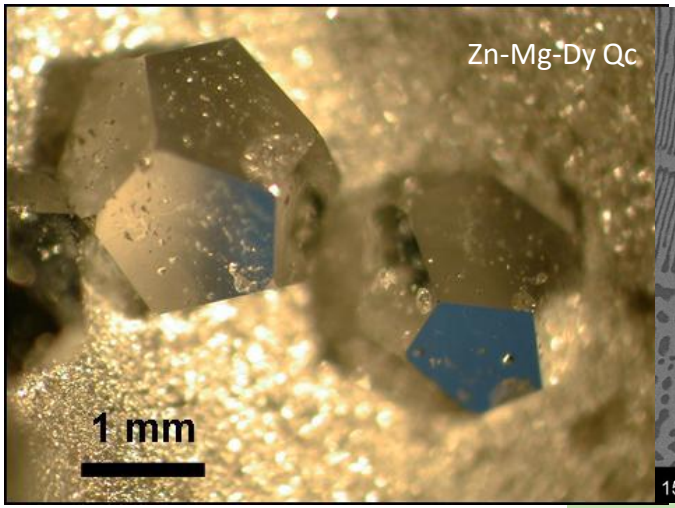




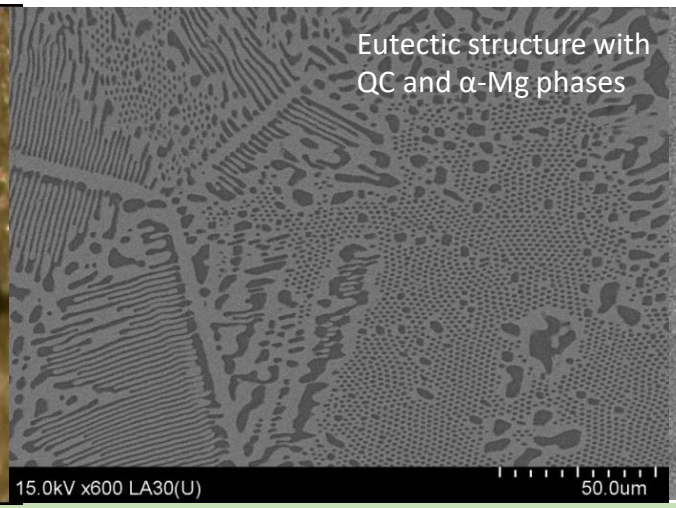
[1] N. Fujita, H. Takano, A. Yamamoto and A.P. Tsai, Acta Cryst. A **69**, 322–340 (2013).
[2] Y. Hatakeyama, N. Fujita and A.P. Tsai, Journal of Physics: Conf. Series **809**, 012007 (2017) .
[3] N. Fujita, Annals of Physics **385** (2017) 225.

Application of quasicrystals for reinforced Mg alloys (SO)

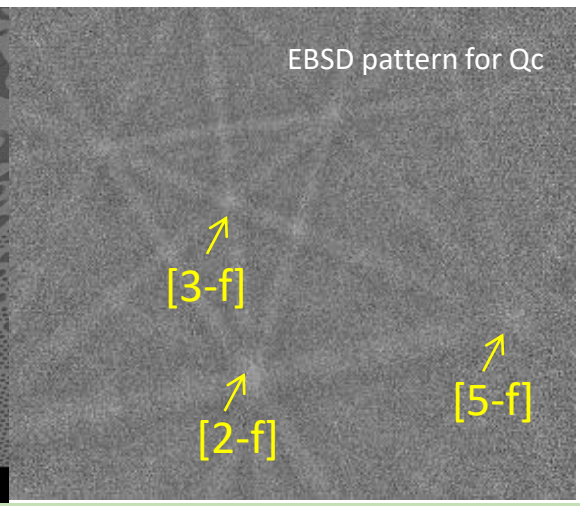
Single QC growth

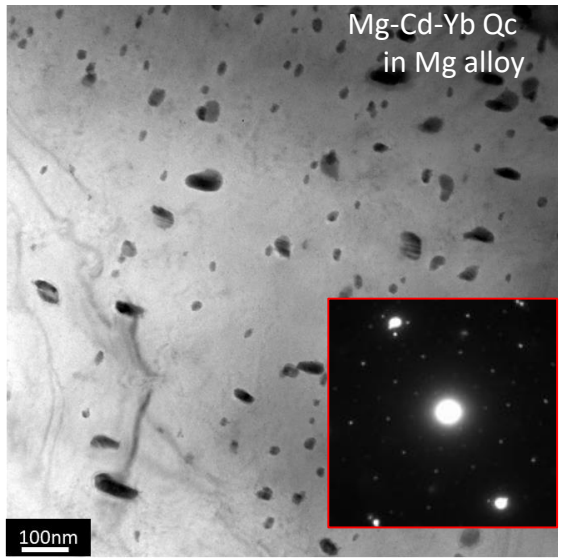


Microstructure containing QC



Orientation relationships





QC-reinforced Mg alloys: Zn-Mg-Zr Qc, Mg-Cd-Yb Qc in Mg

[1] S. Ohhashi, J. Hasegawa, S. Takeuchi, A. P. Tsai, *Philos. Mag.* **87** (2007) 3089.

[2] S. Ohhashi, E. Abe, M. Tanaka, A.P. Tsai, *Acta Mater.* **57** (2009) 4727.

[3] S. Ohhashi, A. Kato, M. Demura, A.P. Tsai, *Mater. Sci. Eng. A* **528** (2011) 5871.

[4] S. Ohhashi, K. Suzuki, A. Kato, A.P. Tsai, *Acta Mater.* **68** (2014) 116.

[5] R. Tanaka, S. Ohhashi, N. Fujita, M. Demura, A. Yamamoto, A. Kato, A.P. Tsai, *Acta Mater.* **119** (2016) 193.

[6] F. Labib, S. Ohhashi, A.P. Tsai, *Philos. Mag.* **99** (2019) 1528

Designing new catalysts through the fusion of metallurgy & catalyst chem.

Metallurgy (AP Tsai)

Knowledge about crystal structures, surfaces & microstructures

×

Catalyst chem (S Kameoka)

Knowledge about adsorption states & catalytic reactions

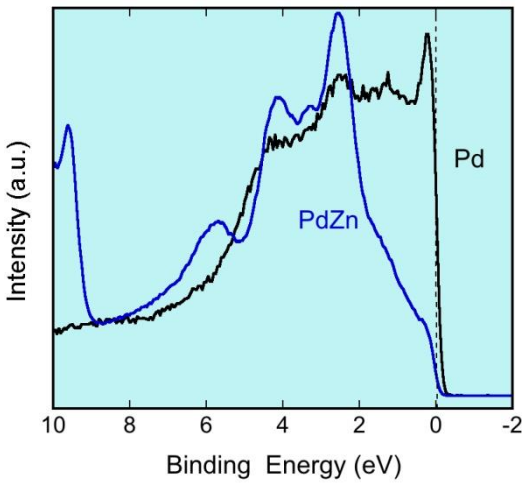
Aim: to find new (unexpected) routes to efficient catalysts.

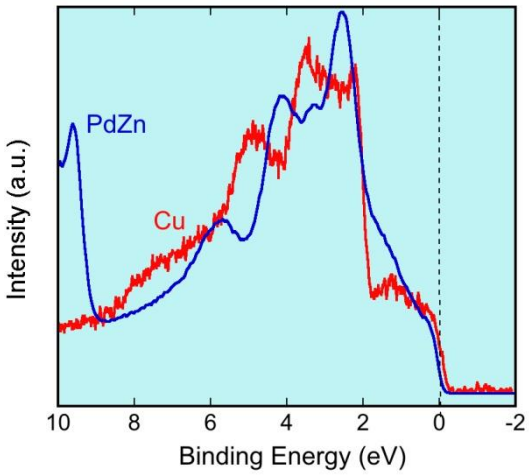
 # Tuning the electronic structure and/or microstructure

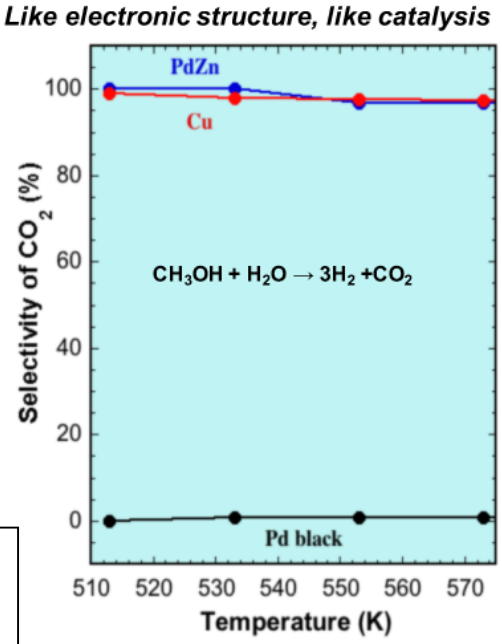
 # New understanding on the generation of active sites

Tuning the electronic structure for catalysis

(Pseudoelement: PdZn ≈ Cu for SRM)







CO_2 selectivity of steam reforming of methanol and valence electronic structures for PdZn, Cu and Pd

[1] A.P. Tsai, S. Kameoka and Y. Ishii, "PdZn=Cu: Can an intermetallic compound replace an element ?" *J .Physical Soc. Jpn.*, **73** (2004) 3270-3273.

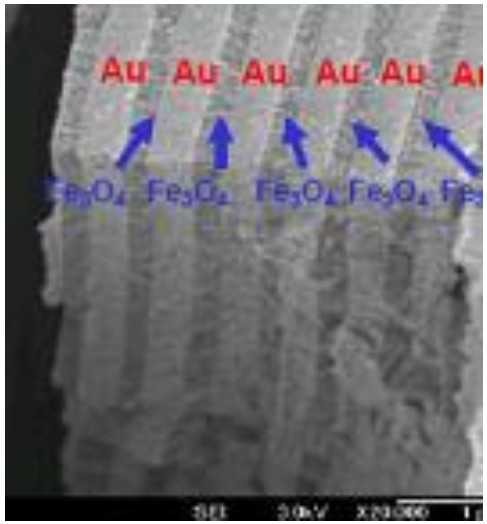
[2] K. Nozawa, N. Endo, S. Kameoka, A.P. Tsai and Y. Ishii, "Catalytic properties dominated by electronic structures in PdZn, NiZn and PtZn intermetallic compounds", *J. Physical Soc., Jpn.,* **80** (2011) 064801.

[3] A.P. Tsai, T. Kimura, Y. Suzuki, S. Kameoka, M. Shimoda and Y. Ishii, "Effect of electronic structures on catalytic properties of CuNi alloy and Pd in MeOH-related reactions", *J. Chem. Phys.,* **138** (2013) 144701.

[4] A.P. Tsai, S. Kameoka, K. Nozawa, M. Shimoda, Y. Ishii, "Intermetallic: A pseudoelement for catalysis" *Account for Chemical Research*, **50** (2017) 2879-2885.

Tuning microstructures through eutectic reaction + leaching

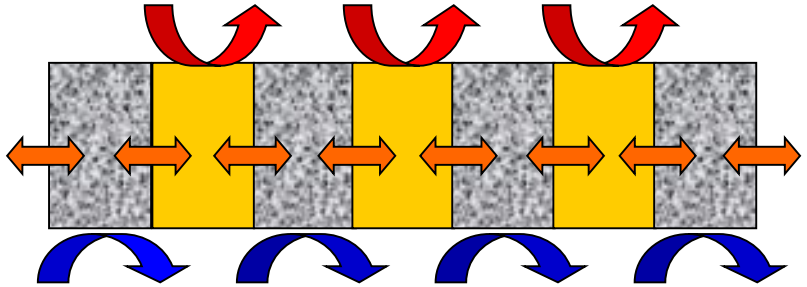
Eutectic microstructure (Lamellar) with porous Au (or Pt) layers



Fe_3O_4: $CO + H_2O \rightarrow CO_2 + H_2$ (exothermal)

Heat transfer

Mass transfer



Pt: $CH_3OH \rightarrow CO + 2H_2$ (endothermal)

Overall $CH_3OH + H_2O \rightarrow CO_2 + 3H_2$ (endothermal)

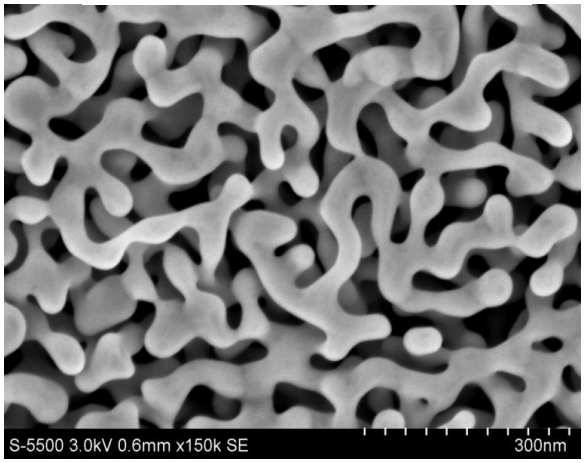
[1] S. Kameoka and A.P. Tsai, "Alternately layered Au/Fe_3O_4 with porous structure – a self-assembled nanoarchitecture for catalysis materials", *Journal of Materials Chemistry*, **20** (2010) 7348-7351.

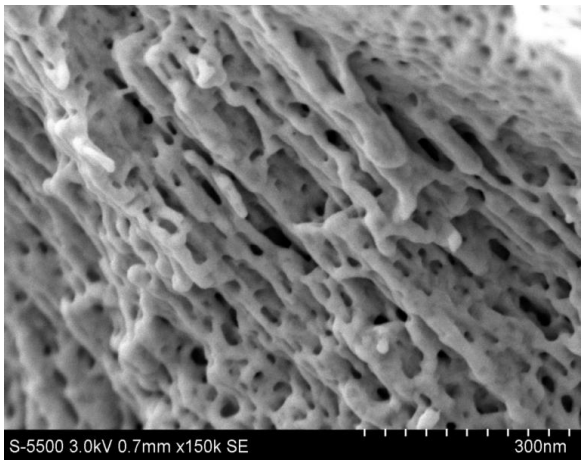
[2] S. Kameoka, S. Wakabayashi, K. Ohshima and A.P. Tsai, "Composite catalyst with lamellar Fe_3O_4/Pt/Fe_3O_4 structure and complementary dual catalytic functions", *Catalysis Letters*, **145** (2015) 1457-1463.

[3] S. Kameoka, S. Wakabayashi, E. Abe and A.P. Tsai, "One-step synthesis of high performance Pt-Fe_3O_4 catalyst: Intermetallic $Al_{13}Fe_4$ as a platform and precursor", *Catalysis Letters*, **146** (2016) 1309-1316.

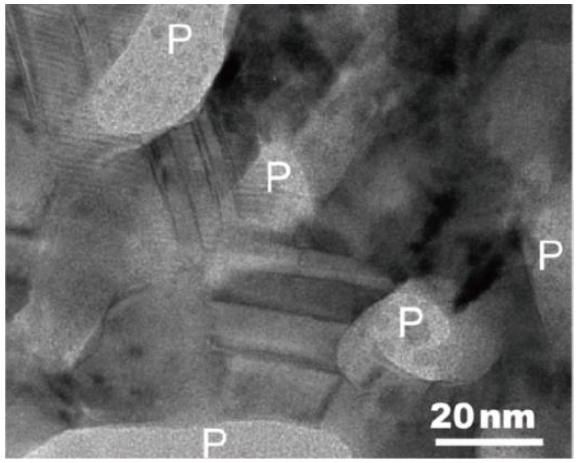
Creation of catalytic active sites at microscopic twin boundaries

PG(Ag_3Au) *with $cHNO_3$*





PG(Al_2Au) *with NaOHaq.*



Twin boundary (TB) defects in the fcc lattice of bulk gold can create close-packed rows of low-coordinated atoms (W-chains; CN= 5 or 6) as active sites on the stepped {211} surfaces of bulk Au.

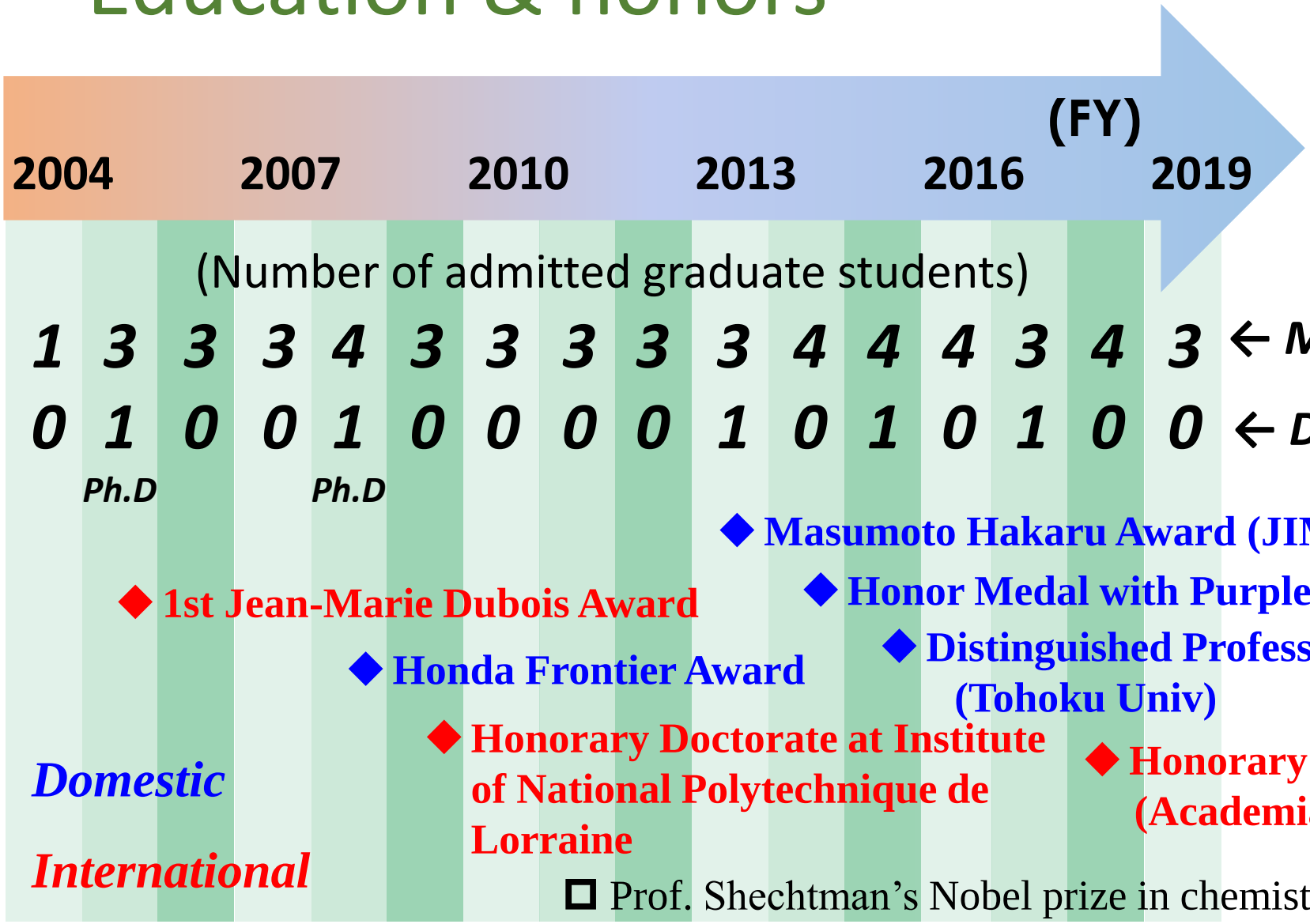
[1] S. Kameoka, T. Tanabe, K. Miyamoto and A.P. Tsai, "Insights into the dominant factors of porous gold for CO oxidation", *Journal of Chemical Physics,* **144** (2016) 034703.

[2] M. Krajci, S. Kameoka and A.P. Tsai, "Twinning in fcc lattice creates low-coordinated catalytically active sites in porous gold", *Journal of Chemical Physics*, **145** (2016) 084703.

[3] M. Krajci, S. Kameoka and A.P. Tsai, "Understanding the catalytic activity of nanoporous gold: role of twinning in fcc lattice", *Journal of Chemical Physics*, **147** (2017) 044713.

[4] S. Kameoka, M. Krajci and A.P. Tsai, "Highly selective semi-hydrogenation of acetylene over porous gold with twin boundary defects", *Applied Catalysis, A: General,* **569** (2019) 101-109.

Education & honors



Thank you very much!