March, 2018

SAND2018-1820C

Optimizing the soft magnetic behavior of iron nitride

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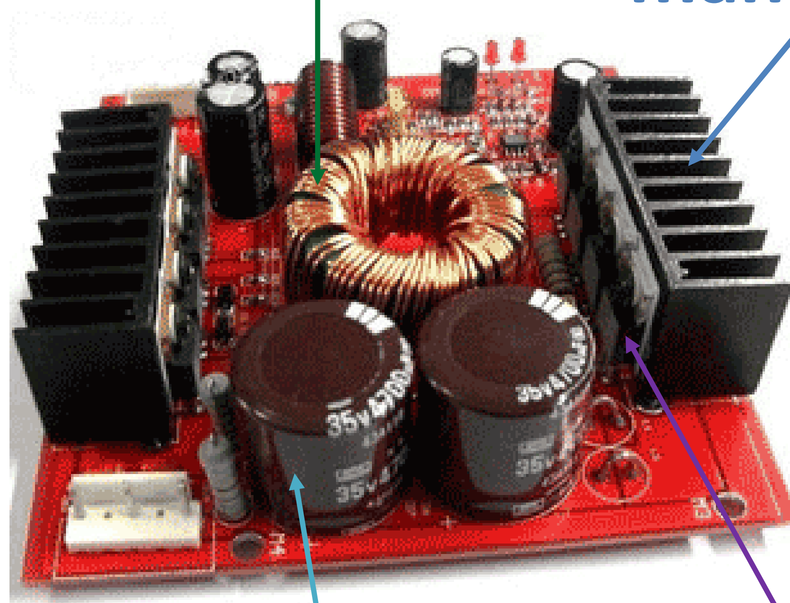
The authors acknowledge support for this work from Dr. Imre Gyuk and the Energy Storage Program in the Office of Electricity Delivery and Energy Reliability at the US Department of Energy

Magnetic Devices Impact Power System Volume and Weight

Passive elements and thermal management comprise the bulk of the volume and mass of a power converter

WBG/UWBG materials enable higher switching frequency and better thermal management



Magnetics

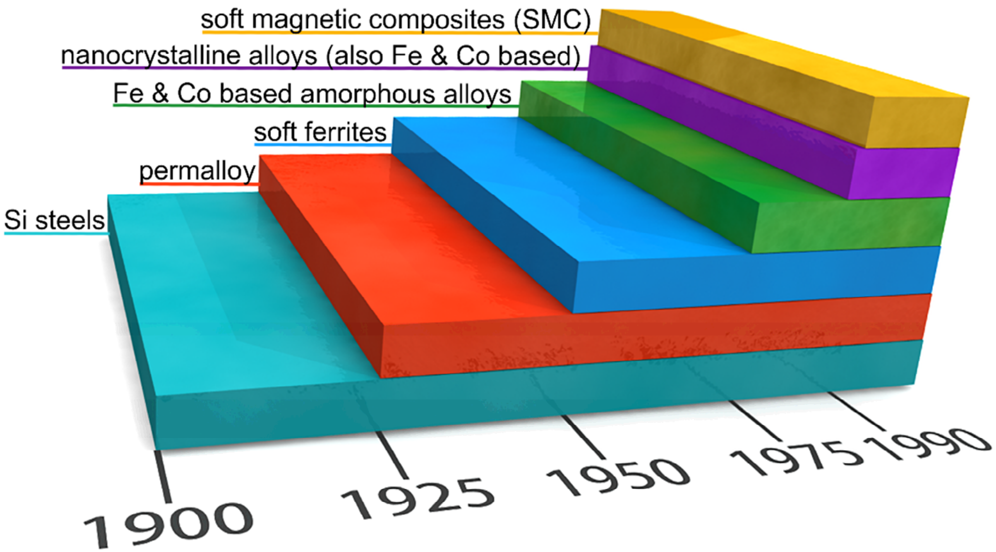
Thermal management

Capacitors

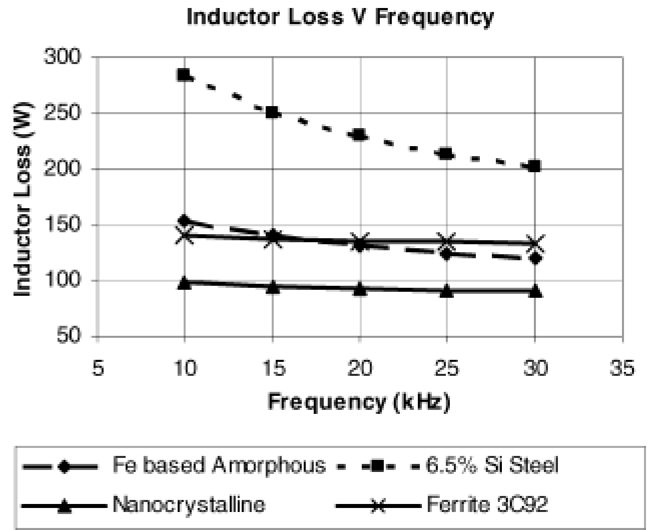
Semiconductor switches

J. Neely, J. Flicker, B. Kaplar

Soft Magnetic Material Development



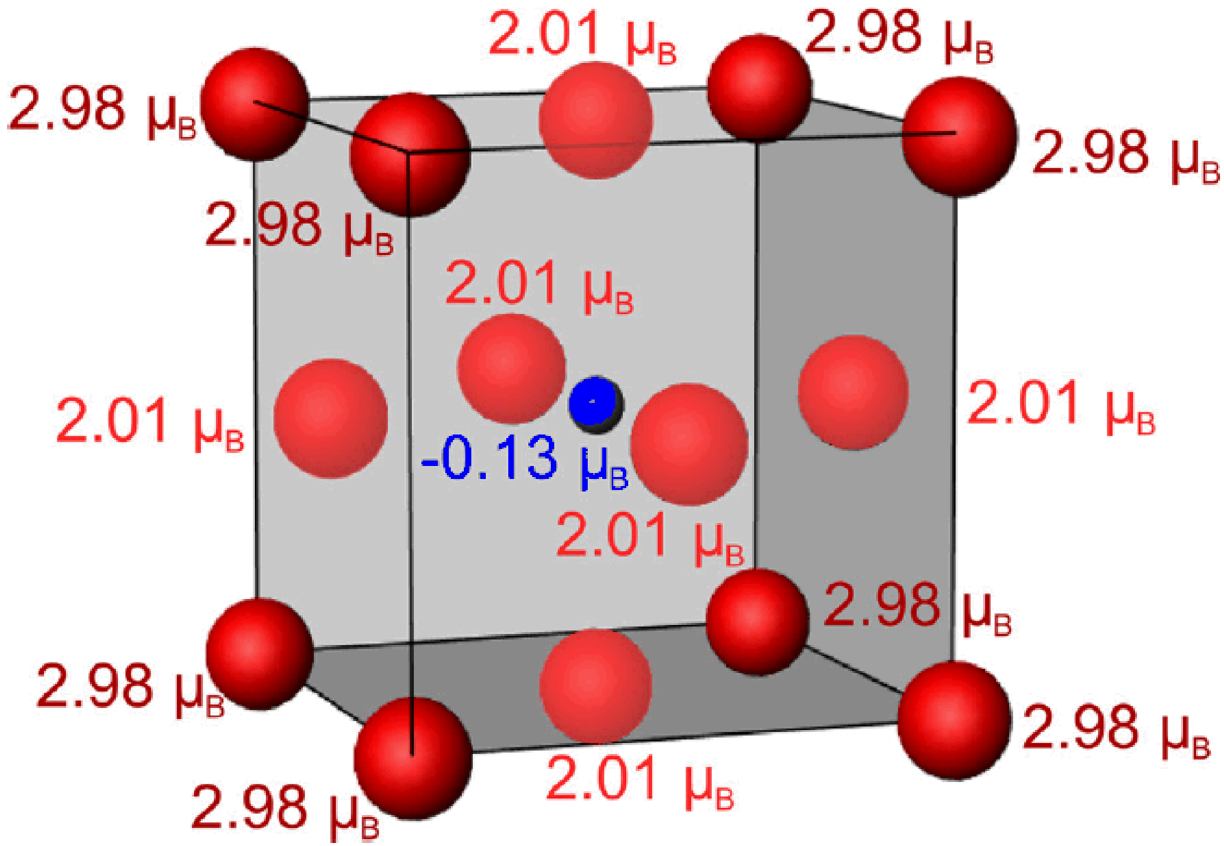
Adapted from: L.A. Dobrzański, M. Drak, B. Ziębowicz, Materials with specific magnetic properties, Journal of Achievements in Materials and Manufacturing Eng., 17, 37 (2006).



B.J. Lyons, J.G. Hayes, M.G. Egan, Magnetic Material Comparisons for High-Current Inductors in Low-Medium Frequency DC-DC Converters, IEEE, 71 (2007).

Magnetic Material	J_s (T)	$\rho(\mu\Omega\cdot m)$	Cost
VITROPERM (Vacuumschmelze)	1.20	1.15	High
Metglas 2605SC	1.60	1.37	High
Ferrite (Ferroxcube)	0.52	$5x10^6$	Low
Si steel	1.87	0.05	Low
γ'-Fe_4N	1.89	> 200	Low

γ'-Fe_4N Unit Cell



fcc γFe structure stabilized by interstitial nitrogen in the body center

G. Scheunert, et al., A review of high magnetic moment thin films for microscale and nanotechnology Applications, Appl. Phys. Rev., 3, 011301 (2016).
J.M.D. Coey, *Magnetism and Magnetic Materials* (Cambridge University Press, Cambridge, UK, 2012).

Previous Syntheses of γ'-Fe_4N

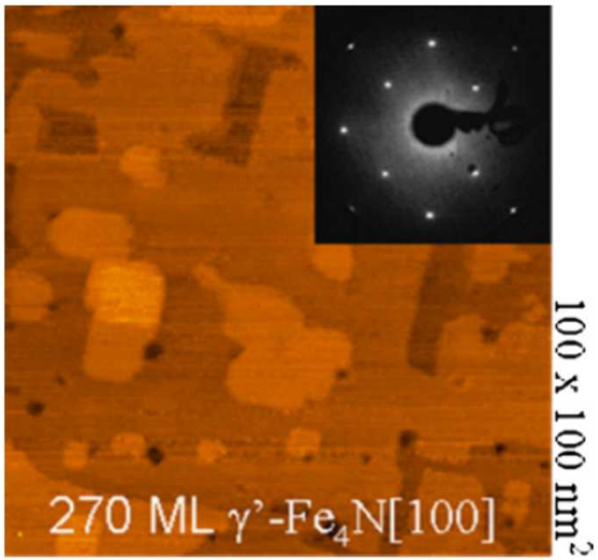
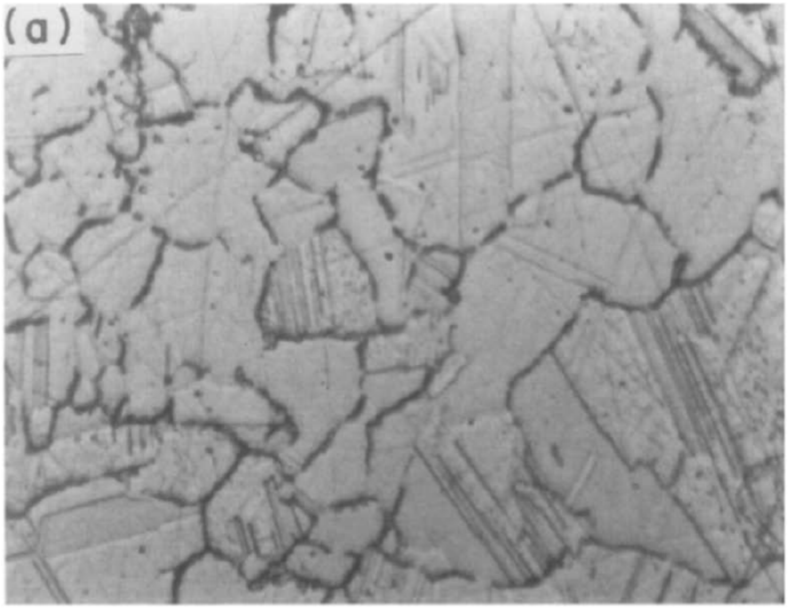


Fig. 1. STM image of a 270 monolayers (ML) thick γ'-Fe_4N film grown on Cu(1 0 0). The inset shows the corresponding LEED pattern (110 eV).

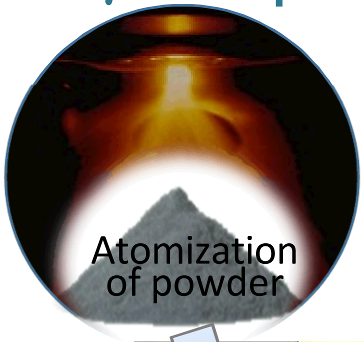


D. Ecija, et. al., "Magnetization reversal of epitaxial films of γ'-Fe_4N on Cu(100)", J. Magn. Mag. Mat., 316, 321 (2007).

S.K. Chen, et. al., "Synthesis and magnetic properties of Fe_4N and $(Fe, Ni)_4N$ sheets", J. Magn. Mag. Mat., 110, 65 (1991).

γ'-Fe_4N Synthesis and Processing

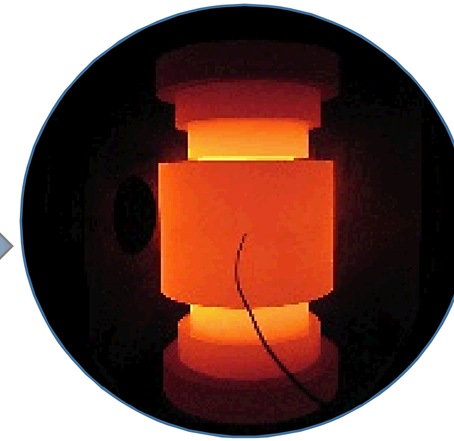
U.S. Patent Filed January 2016 (#15/002,220)



Cryomilling
Severe Plastic deformation

Fluidized Bed Furnace

SPS Consolidation



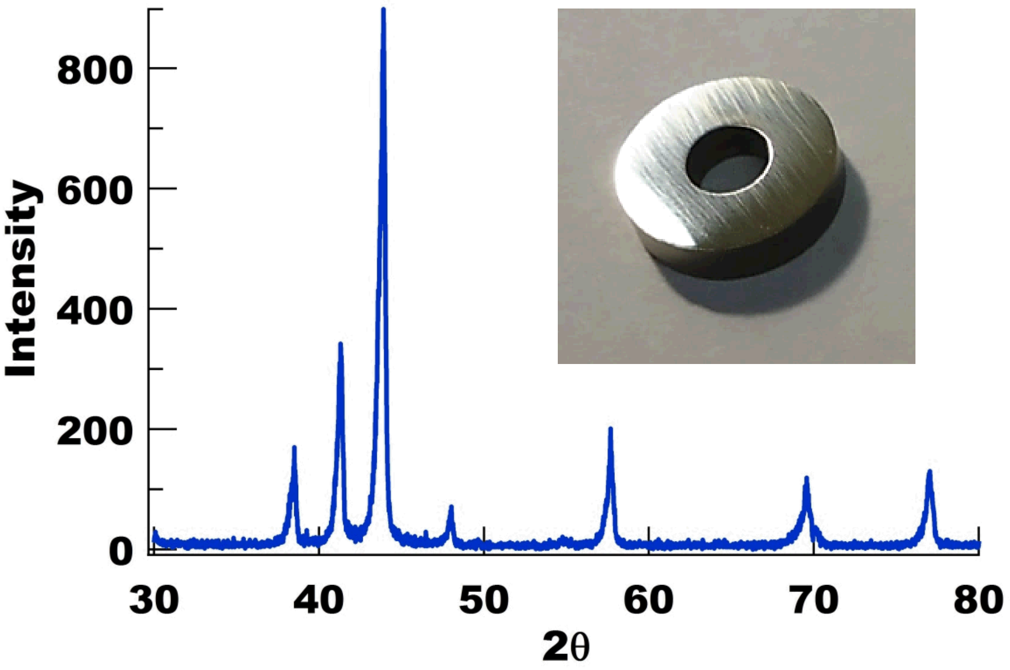
Spark Plasma Sintering
Fast sintering

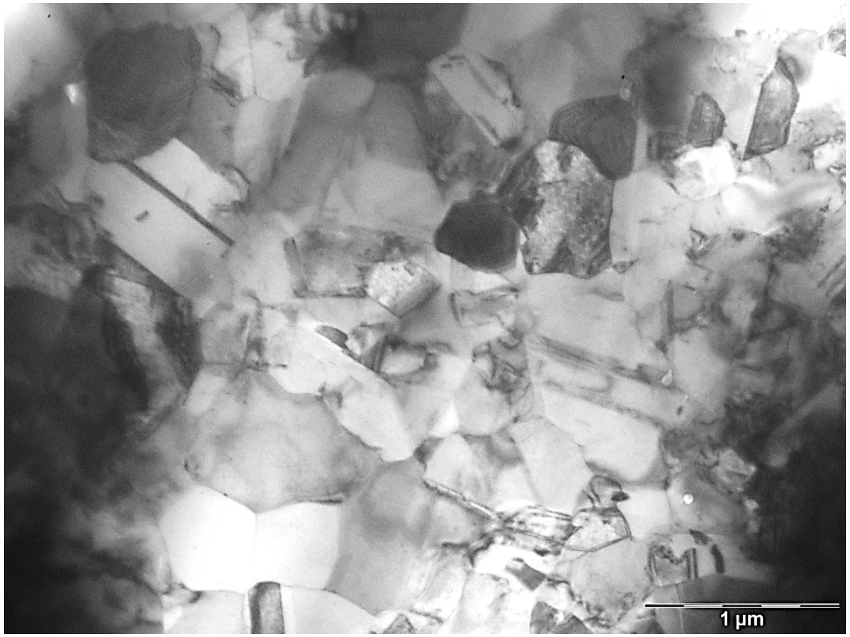
- **Pressure and pulsed current assisted sintering process**
- **Precision control over heat and pressure**
- **Restrain grain growth**
- **Full densification**

Sandia National Laboratories

SPS consolidated Iron Nitride

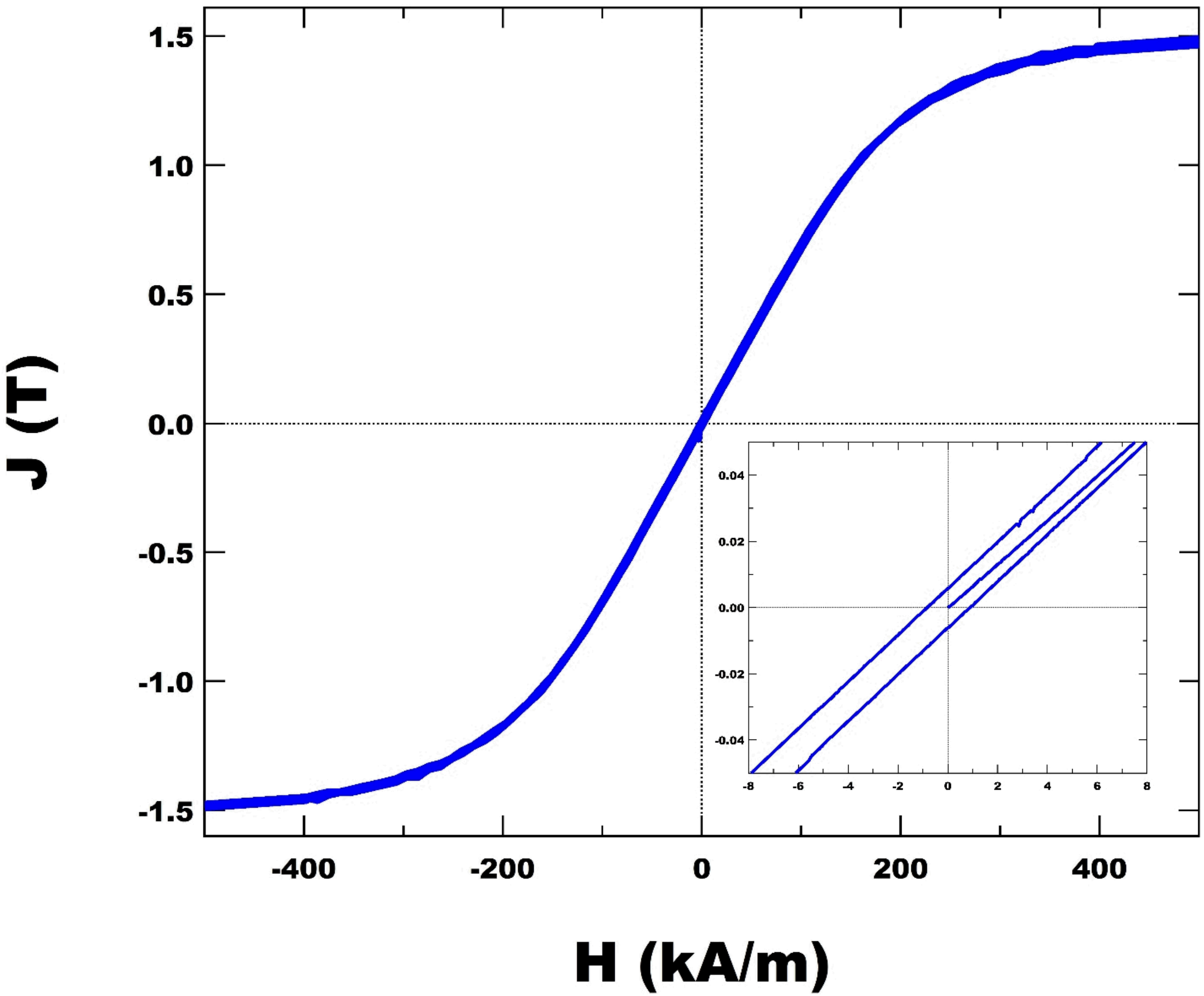
First ever bulk γ'-Fe_4N!





- **Fe nitride powders well consolidated with little porosity**
- **Grain sizes 200 nm – 1 µm → fine grain size = low loss**
- **γ'-Fe_4N primary phase**
- **Fe_3N secondary phase from mixed phase starting material**

Magnetic Characterization



- SPSed at 550°C and 100 MPa
- J_s = 1.62
- Theoretical J_s = 1.89 T (SiFe is 1.87 T)
- H_c < 1000 A/m

Coercivity as a Function of Particle Size

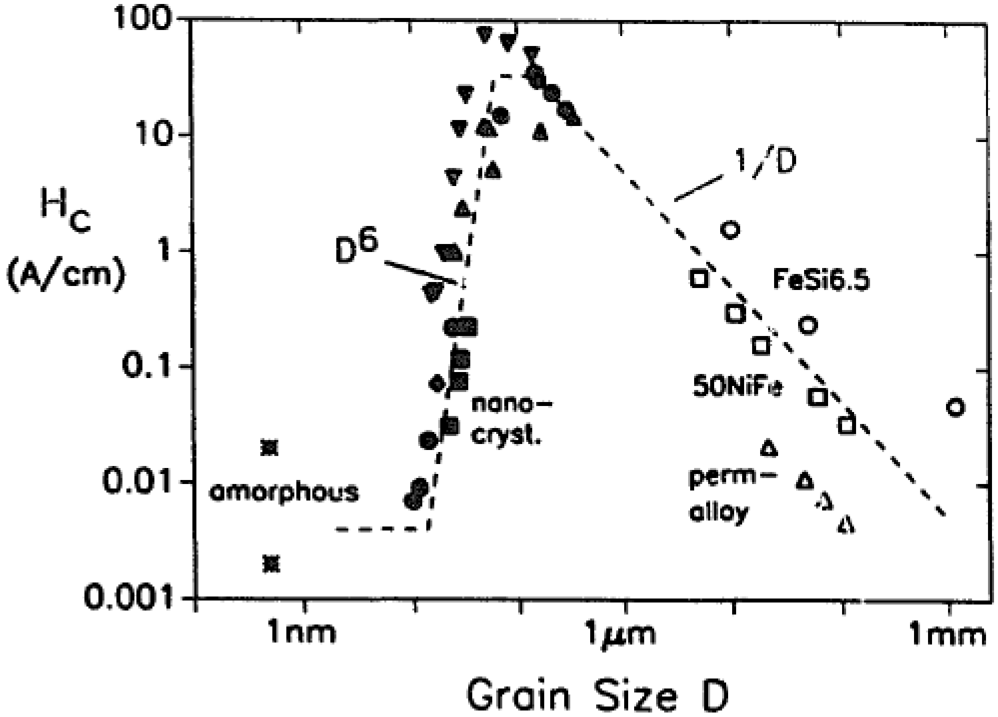
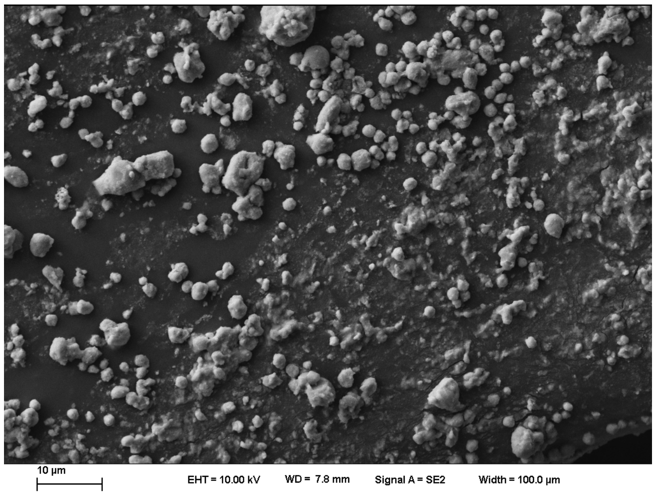


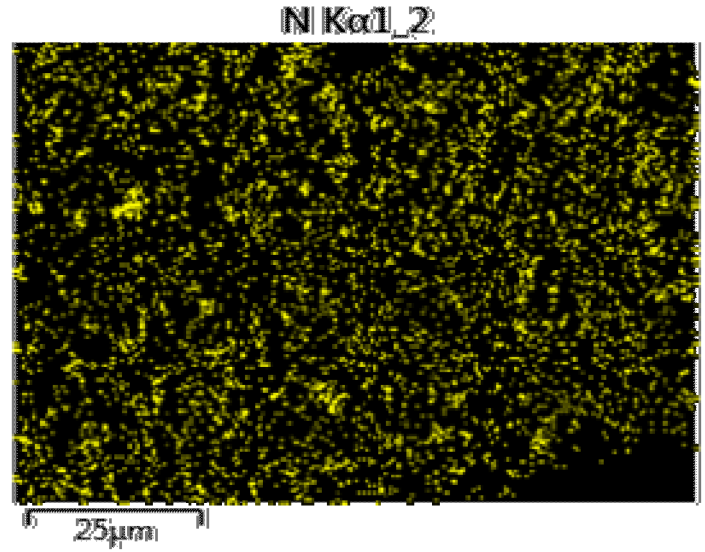
Fig. 2. Coercivity H_c vs. grain size for various soft magnetic metallic alloys. The data of the nanocrystalline material refer to (▲) FeNbSiB and (●) FeCuNbSiB [14], (◆) FeCuVSiB [15], (■) FeZrB [4] and (▼) FeCoZr [16].

G. Herzer, Nanocrystalline Soft Magnetic Materials, J. Magn. Mag. Mat., 112, 258 (1992).

Synthesis of fine grained γ'-Fe_4N

Fe nanoparticles $\xrightarrow{NH_3\ (l)}$ Fe_xN

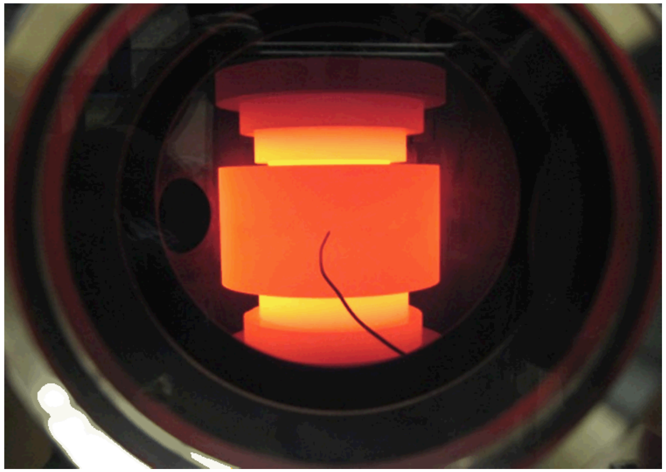




- **SEM-EDS confirms nitrogen content**
- **Optimizing conditions to yield phase pure material**

Conclusions

- γ'-Fe_4N has the potential to serve as a new low cost, high performance transformer core material
  - M_{sat} > Si steel
  - Increased current and field (and therefore power) carrying capability
  - Resistivity 200X greater than nanocrystalline and amorphous alloys
  - Only comprised of low cost and abundant elements (Fe & N)
- The fabrication of bulk γ'-Fe_4N using SPS has been demonstrated
  - SPS can consolidate iron nitrides without material decomposition
  - Parts can be fabricated directly using net-shaping



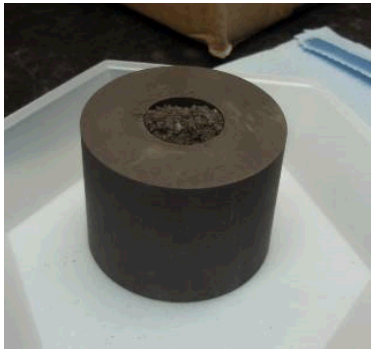
Acknowledgements

- γ'-Fe_4N R&D supported by Dr. Imre Gyuk and the Energy Storage Program in the Office of Electricity Delivery and Energy Reliability at the US Department of Energy
- We thank Robert Delaney (Eldorado High School) for his assistance with magnetic data fitting and analysis

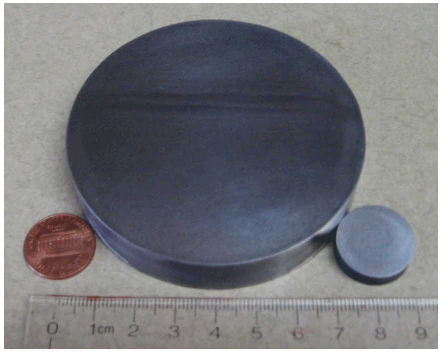
Extra Slides

Spark Plasma Sintering (SPS)

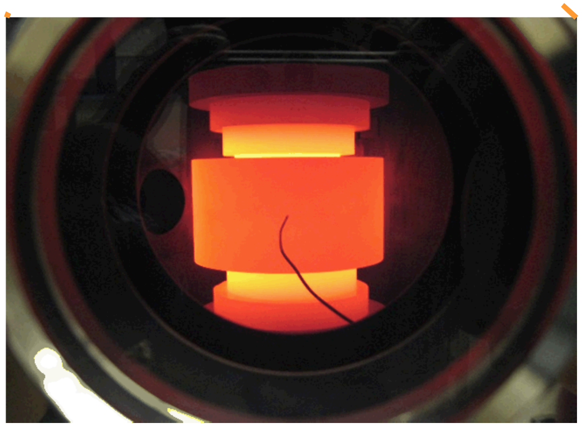
SPS Model: SPS-825S Dr. Sinter® at UC Irvine



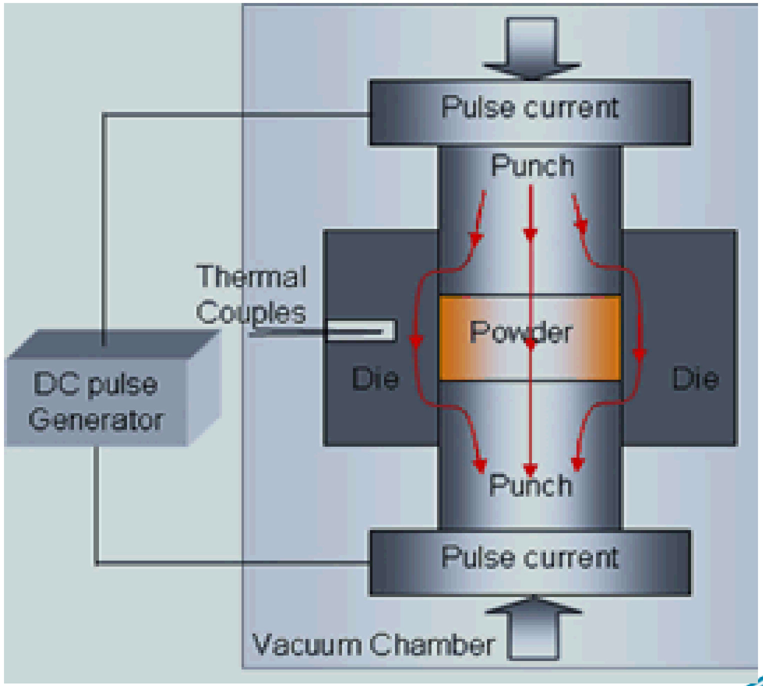
Starting Powder in Die



End Product

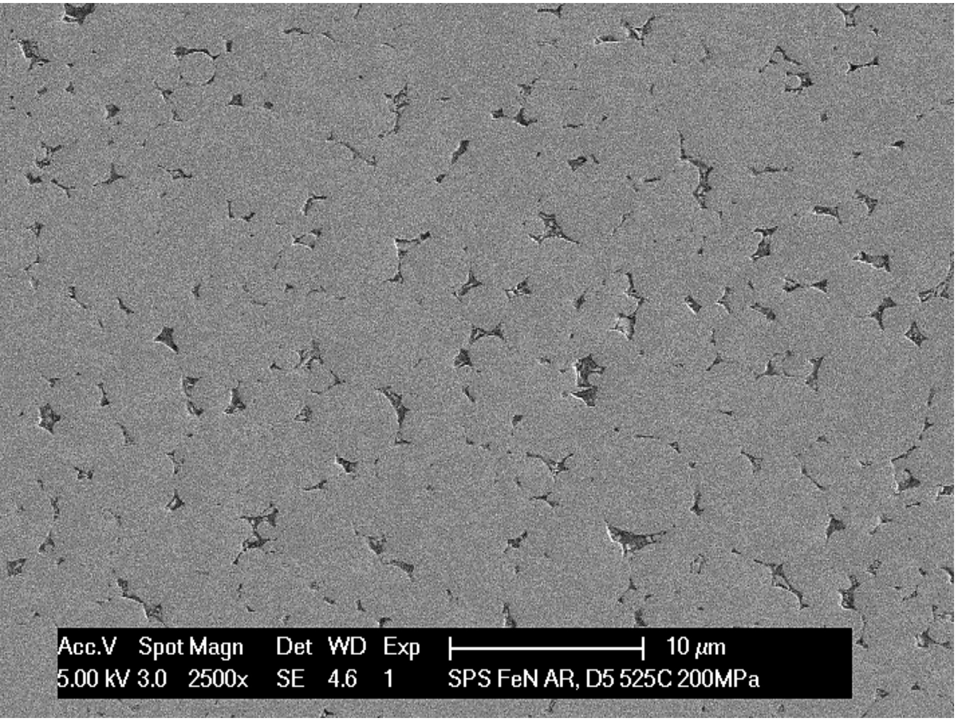


SPS Chamber

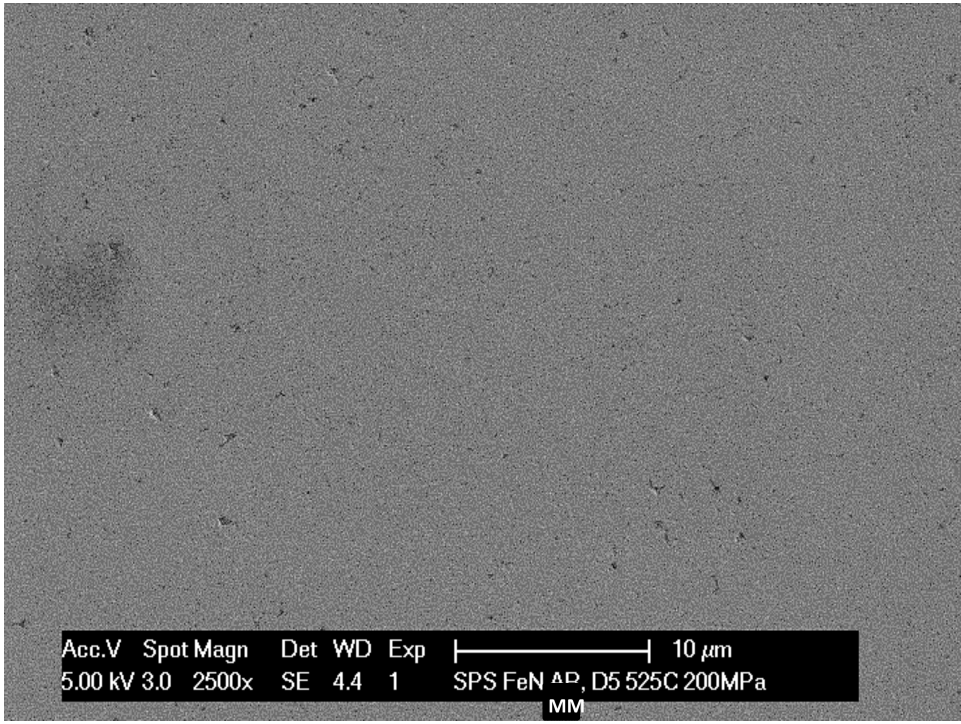


SEM of SPSed FeN samples, (Ø5mm, 525^{o}C, 200MPa)

W/ as-received FeN powder



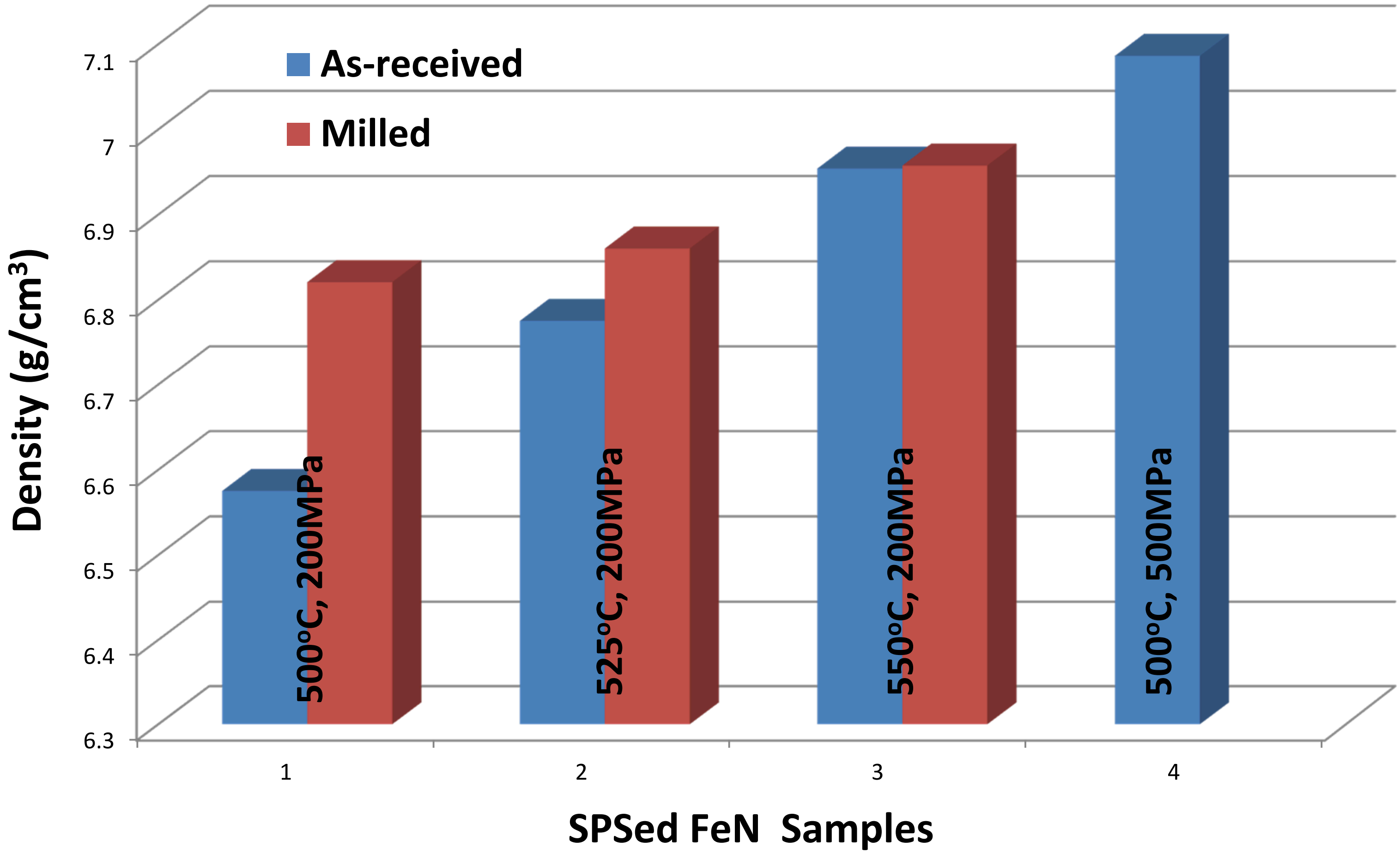
W/ milled FeN powder



- **Milled FeN powder produces more uniform and dense SPSed billets**
  - **Higher packing density with smaller particle size**
  - **Enhanced diffusion with smaller grain size of milled powder**

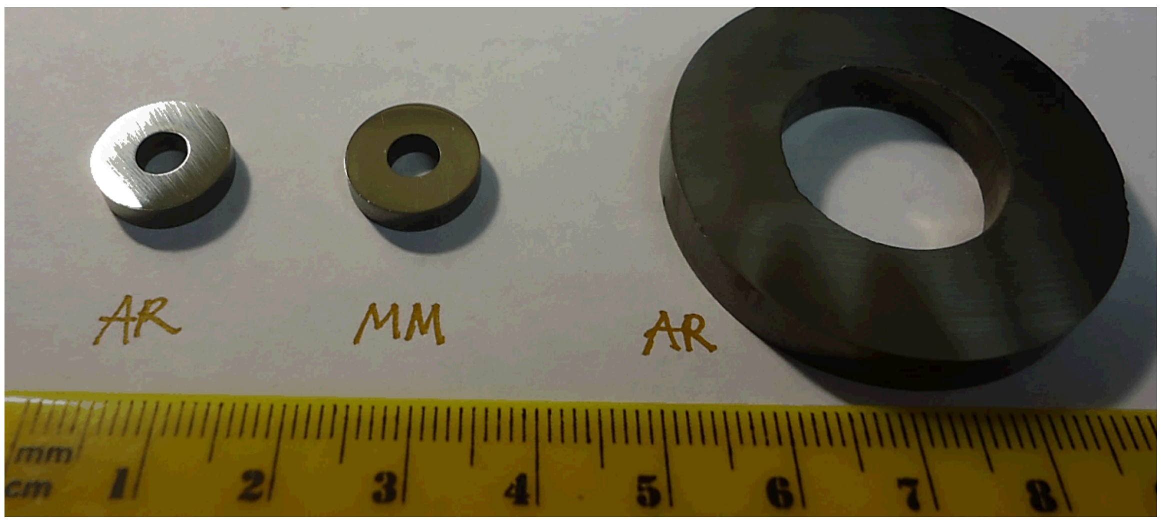
Density of SPSed Fe_4N samples

ρ_{theory} = 7.212 g/cm^3



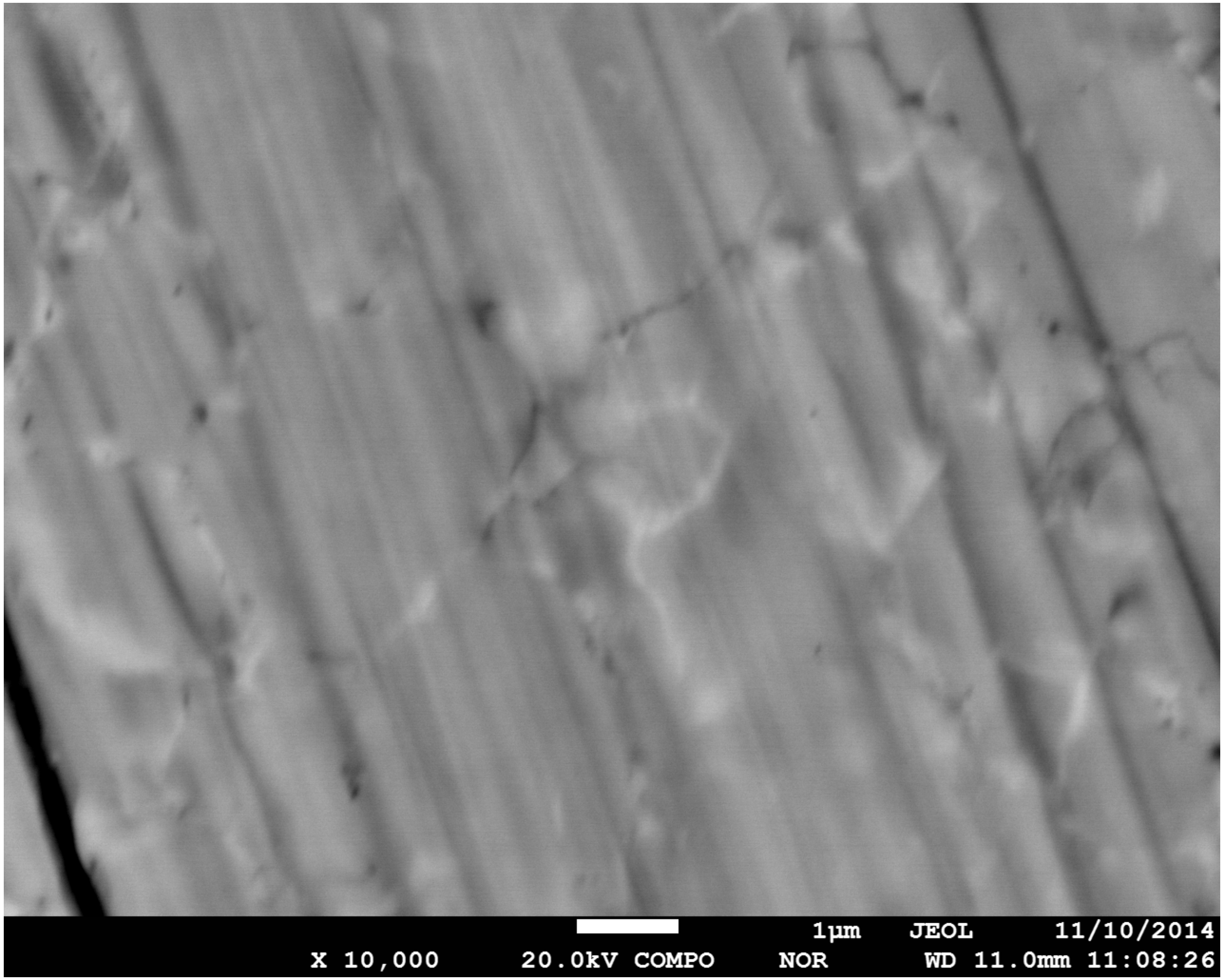
- Density increases with increasing SPS temperature and pressure
- Higher degree of variation in SPSed samples using as-received powder
- Milling improves density and uniformity

Net-Shaping of Transformer Cores



- **Can sinter toroids and other shapes directly (net-shaping)**
- **Eliminates the need for machining**

Toroid Surface SEM and EDS

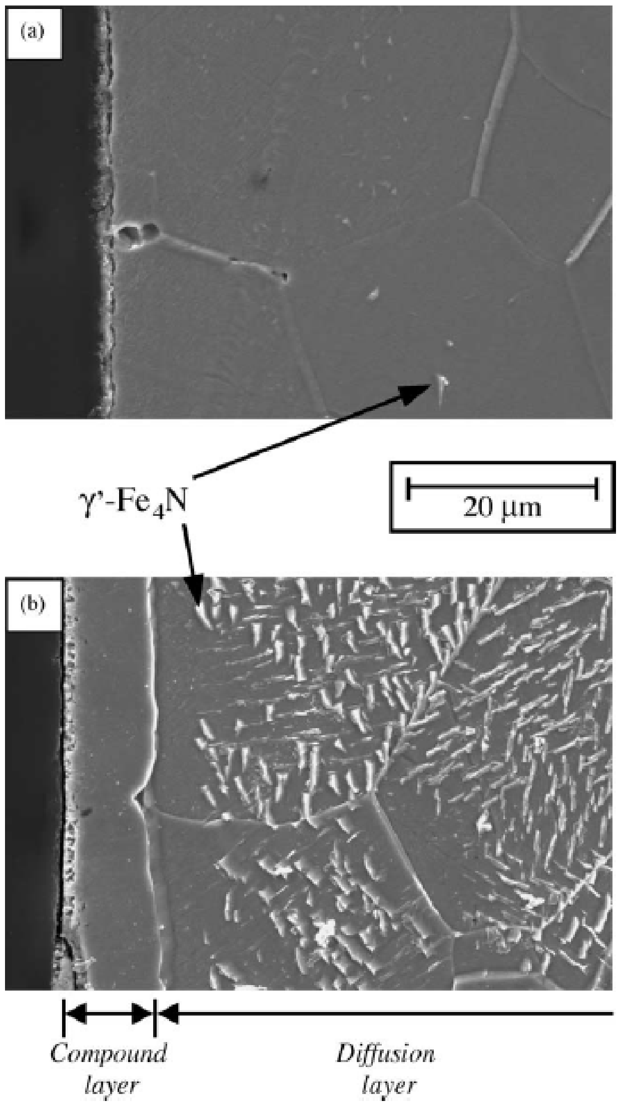


- Small variation in composition between grain boundary and center
- Grain center stoichiometry ≈ Fe_4N
- Grain boundary is ≈ 3 Atomic% richer in iron

*SEM/EDS analysis completed by Dick Grant (SNL)

Location	Fe (Atomic %)	N (Atomic %)
Grain center	81.3	18.7
Grain boundary	84.2	15.8

In House Synthesis of Raw Materials: Electrochemical Nitriding of Iron…plus more



H. Tsujimura, T. Goto, Y. Ito, Journal of Alloys and Compounds, 376, 246 (2004)

- **Growth of γ'-Fe_4N demonstrated by Japanese electrochemists**
- **Formed γ'-Fe_4N at the surface of Fe(0) electrode using Li_3N as nitride source**
- **Demonstrates electrochemical synthesis of iron nitride possible**
- **Our goal is to demonstrate autonucleation of iron nitride with flowing N_2**

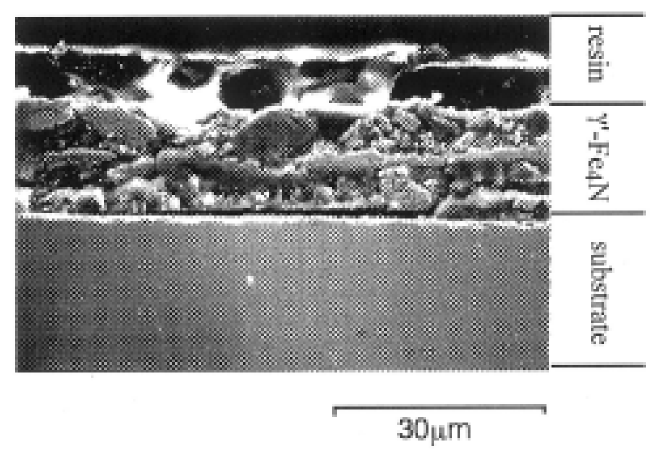
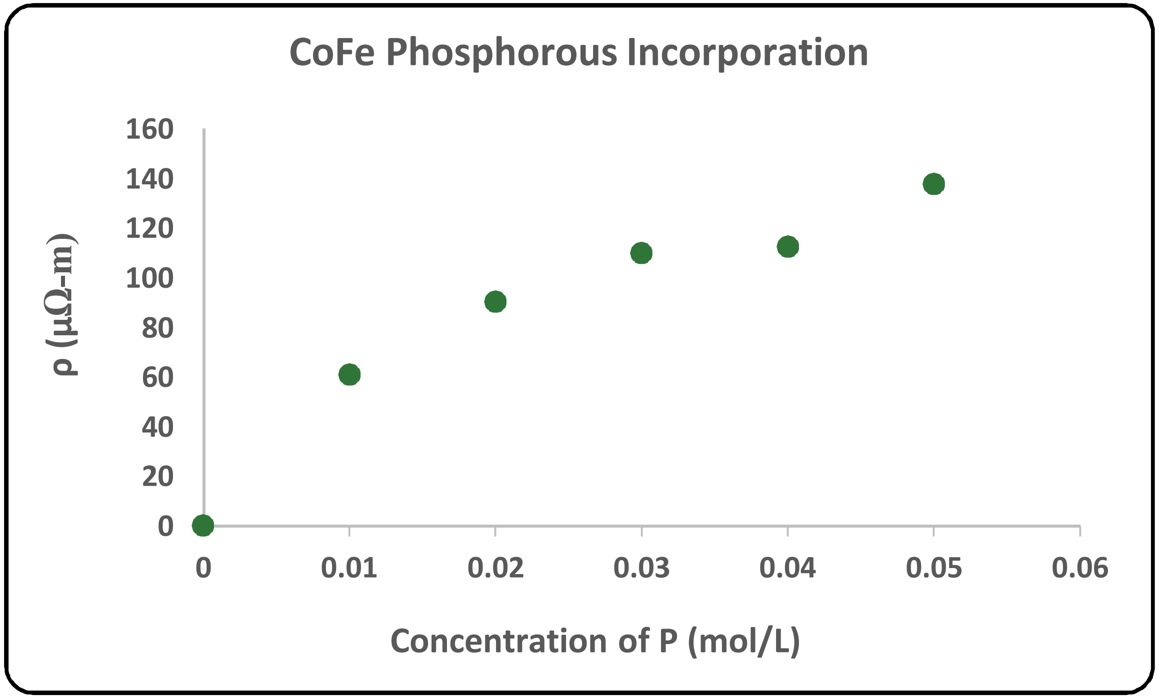


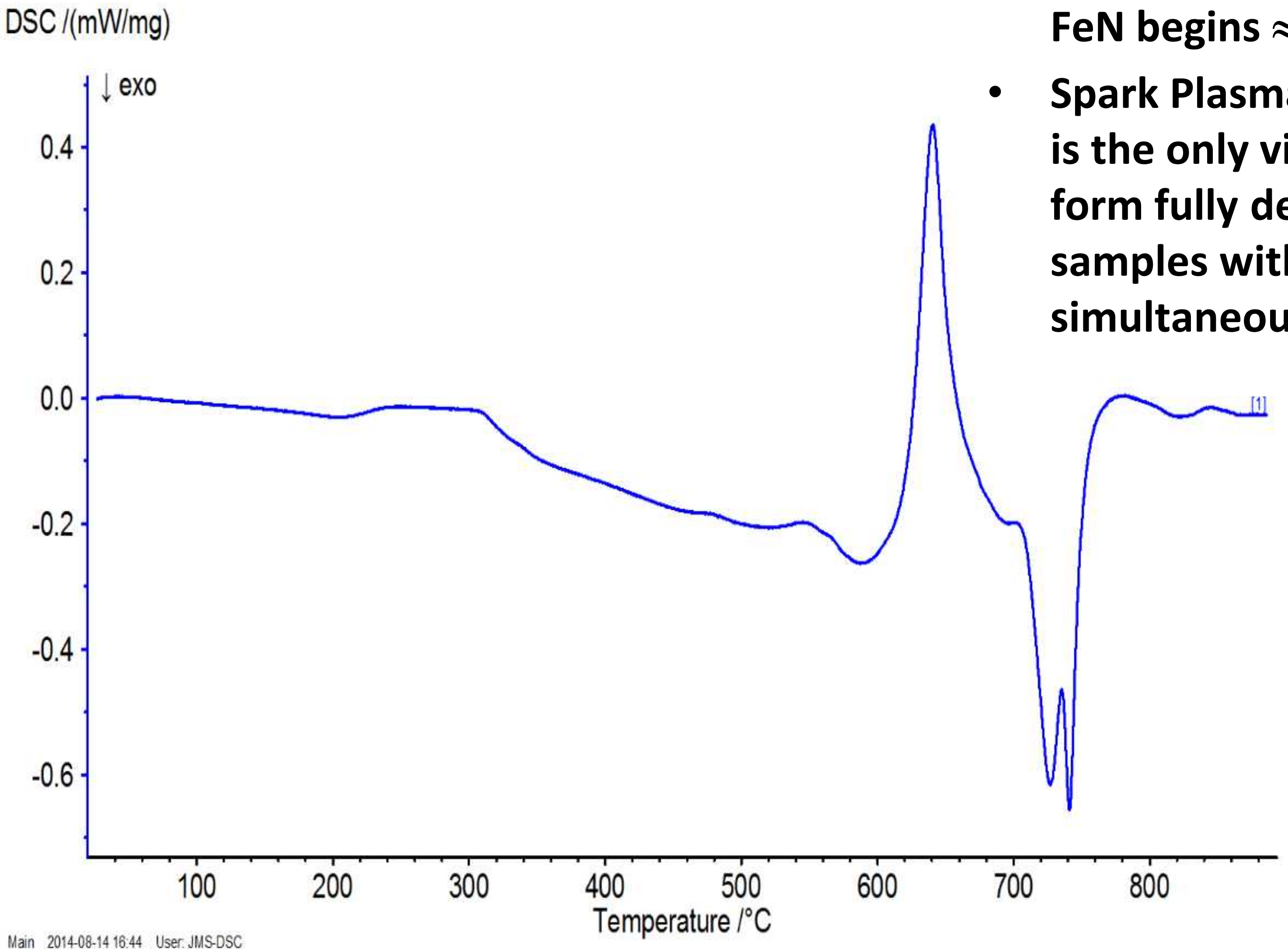
Fig. 10. Cross-sectional SEM image of iron electrode after potential pulse electrolysis for 1 h.

T. Goto, R. Obata, Y. Ito, Electrochimica Acta, 45, 3367 (2000)

Increasing resistance with phosphorus



Differential Scanning Calorimetry (DSC) of Fe_4N



- **Decomposition of sintered FeN begins ≈ 600°C**
- **Spark Plasma Sintering (SPS) is the only viable route to form fully dense bulk Fe_4N samples without simultaneous decomposition**

Iron nitrides crystalline structure

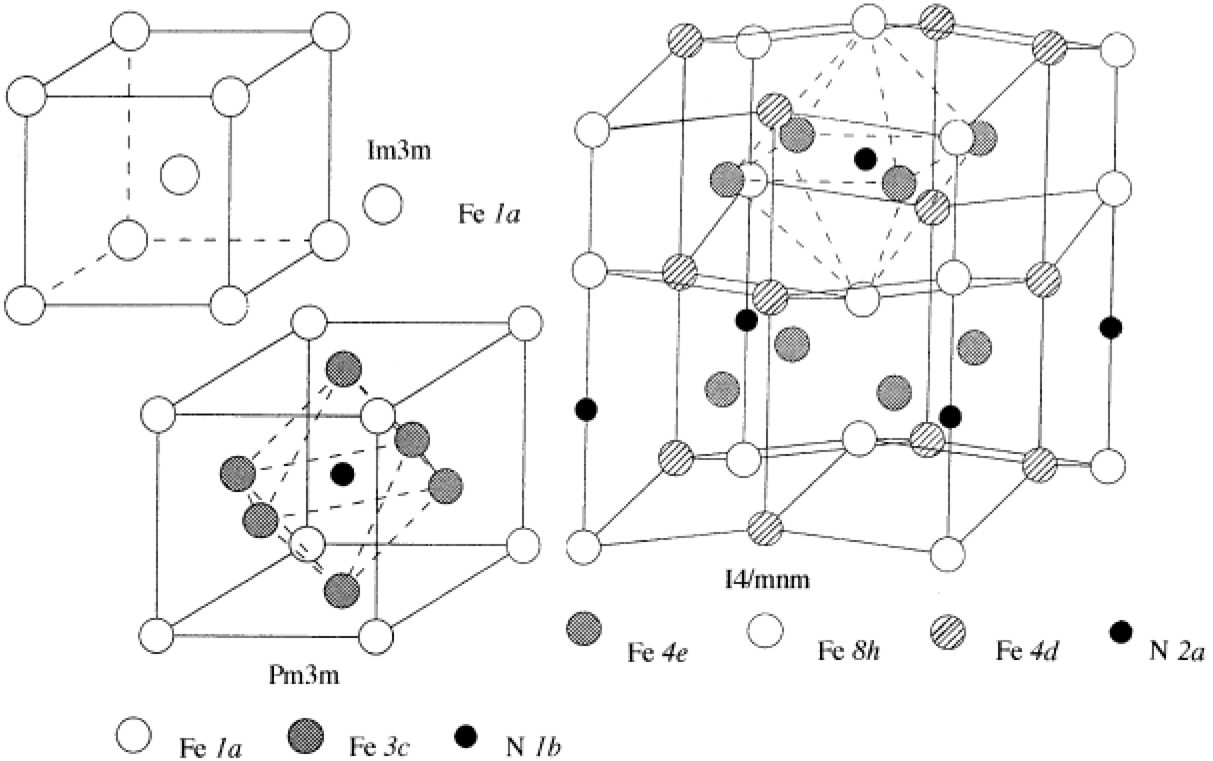


Fig. 1. Crystal structures of αFe, $\gamma'Fe_4N$ and $\alpha''Fe_{16}N_2$, drawn to scale.

J.M.D. Coey, P.A.I. Smith, Magnetic Nitrides, J. Magn. Mag. Mat., 200, 405 (1999).

Magnetic Moment Variation with Nitrogen Concentration

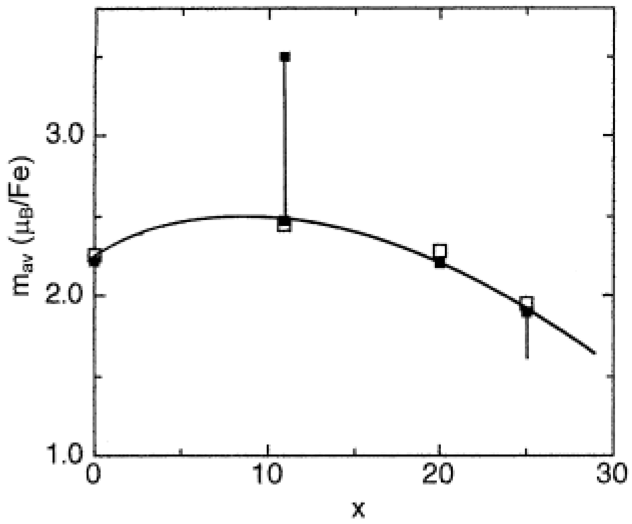


Fig. 3. Calculated (open symbol) and measured (solid symbol) average iron moments in Fe–N compounds.

J.M.D. Coey, P.A.I. Smith, Magnetic Nitrides, J. Magn. Mag. Mat., 200, 405 (1999).

Other Magnetic Nitrides of Interest

Material	Phase	σ_s (Am^2/kg)	J_s (T), if available	T_c (K)	H_c (A/m)
FeN	rocksalt (fcc or fct)	209			
γ'-Fe_4N	antiperovskite-like	209	1.89	769	460
α''-$Fe_{16}N_2$	tetragonal	230 - 286	2.3	810	
α''-$Fe_{90}N_{10}$		230			
g-C_4N_3	graphitic	62			
MnN	rocksalt	194-308			4000
α-Fe	bcc	217	2.15	1044	70

- **Nitrides will have higher resistivities than current transformer core materials and will not require laminations of inactive material to mitigate eddy current losses**

SPS for Manufacturing Ceramics

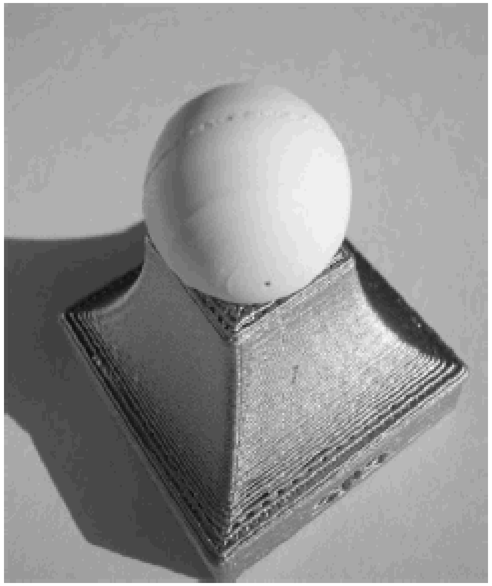


Fig. 9. Al_2O_3 sphere obtained in one single step by SPS.[63]

J. Galy, Private Communication, 2007.

Hungría et. al., Adv. Eng. Mater. Vol. 11 (2009) 616
DOI: 10.1002/adem.200900052

SPS System Manufacturers:

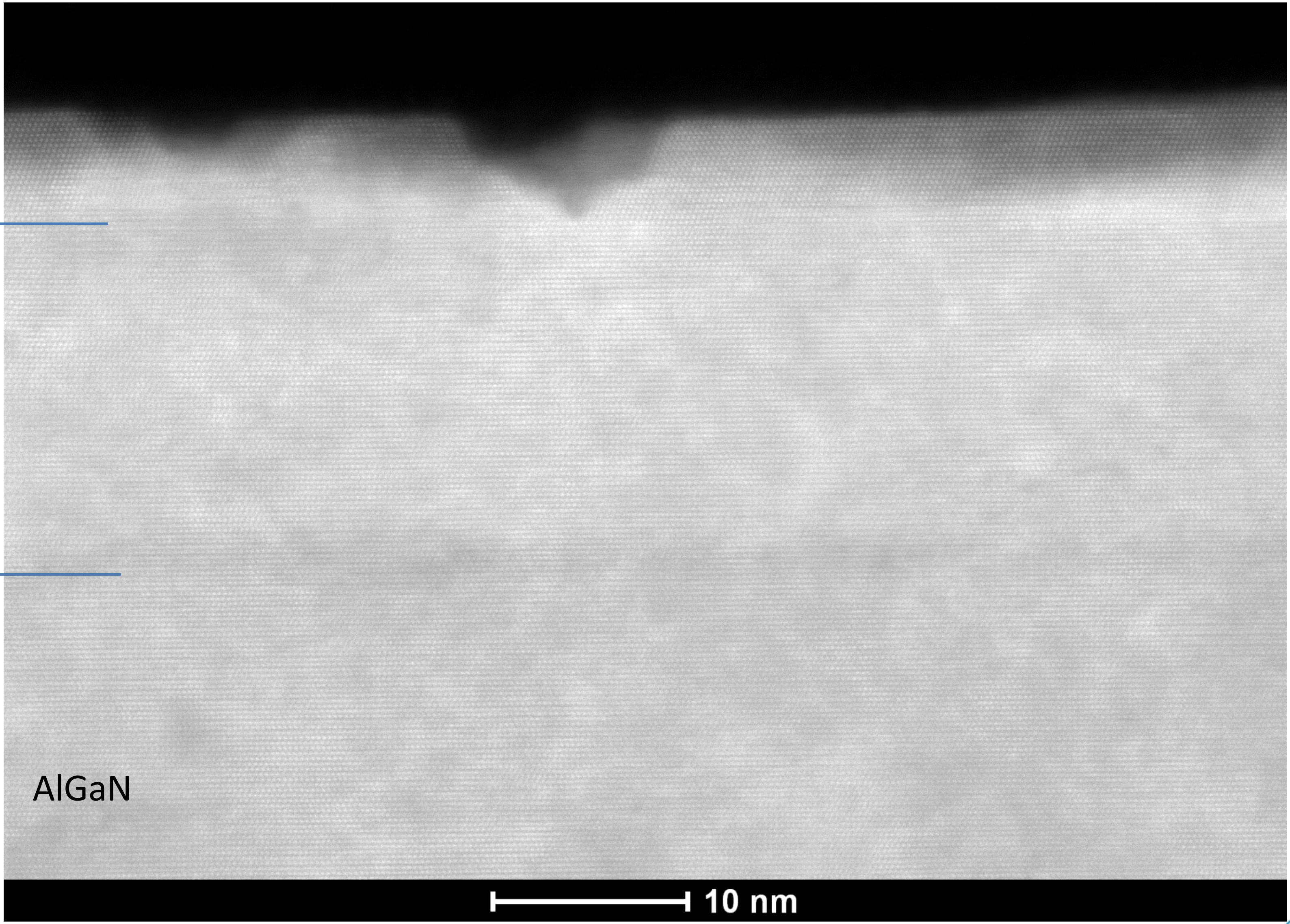
- **Fuji Electronic Industrial Co. (Japan)**
- **FCT Systeme GmbH (Germany)**
  - **Can make components up to 500 mm (~20”) in diameter**
- **Thermal Technology LLC (Santa Rosa, CA)**

- **Size of equipment increasing to accommodate commercial needs**
- **Technology for continuous SPS under development**
- **A large number of companies have acquired SPS but often request this info to not be made public to maintain a competitive advantage**

Electrochemical Solution Growth of GaN

c-GaN

h-GaN



Moving Forward

- SPS processing parameters are being modified to improve phase purity, grain structure, and magnetic performance
- Parallel development of improved synthesis routes to phase pure nanocrystalline γ'-Fe_4N raw powders
- Evaluation and comparison of transformers at high frequency and elevated temperatures

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