Experimental characterization techniques

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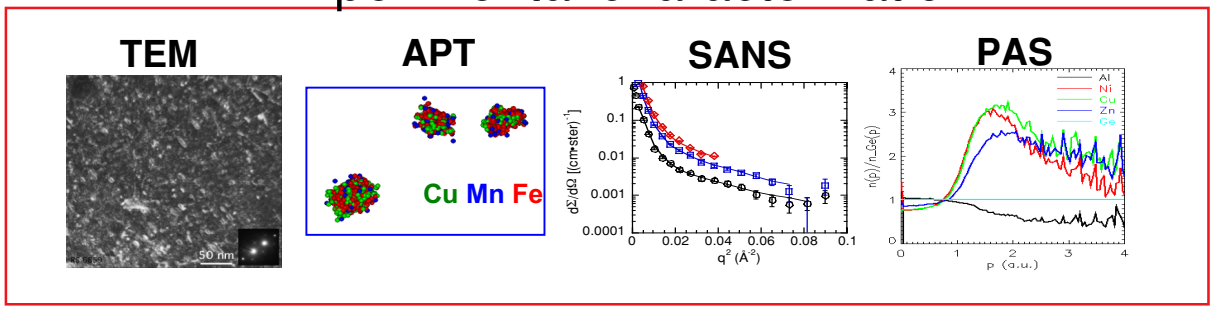
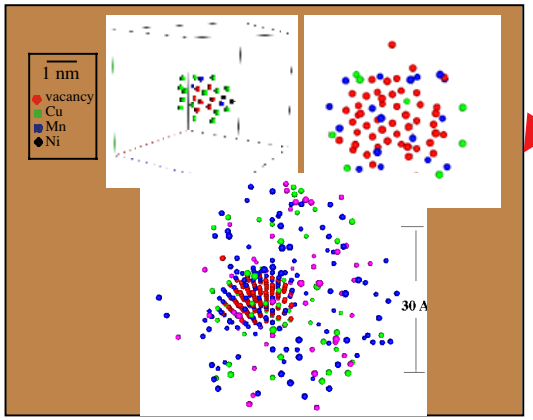
Computational microscope

Experimental characterization

feature 'signals'



Multiscale modeling

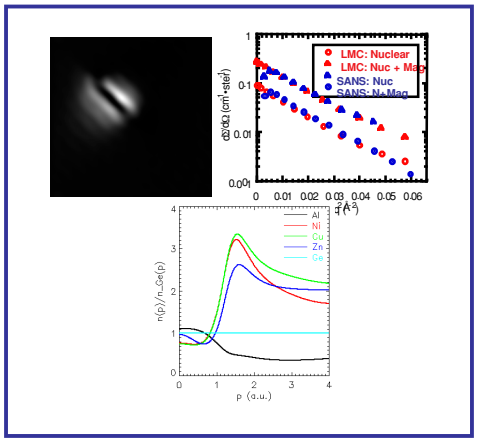


self-consistent 'understanding'

predict features

simulate observables

TEM, SANS, Positron theory

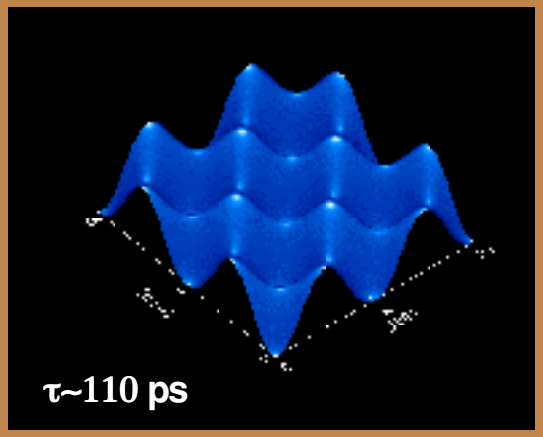


Apply complementary experimental measurements, closely coupled to modeling and positron theory

Positron annihilation spectroscopy

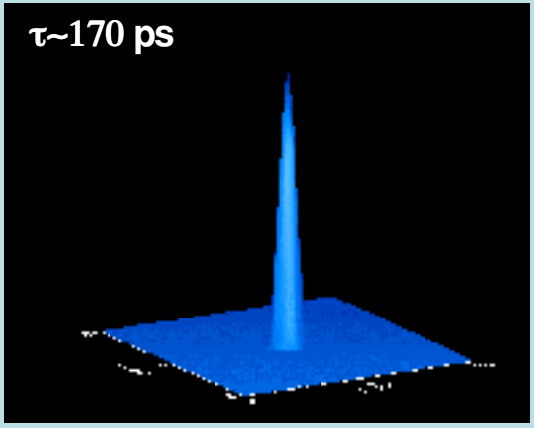
Positrons diffuse and localize ('trap') at defects or regions of strong positron affinity before annihilation

Bulk metal without defects

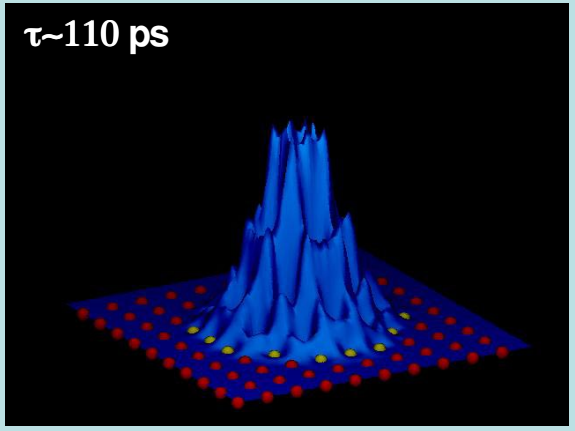


Delocalized positron density

Vacancy defects



Embedded particles of stronger positron affinity

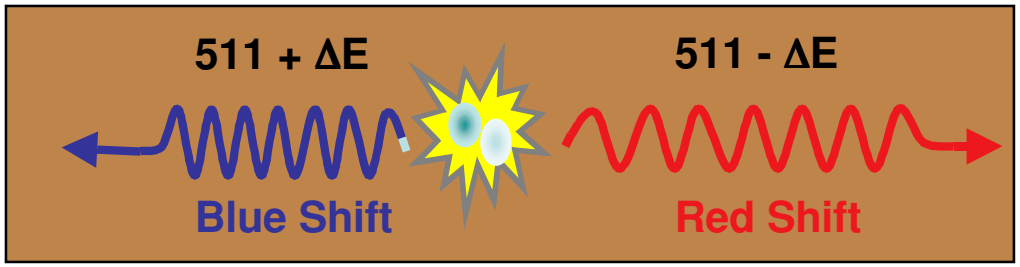


Strongly localized positron density

Each defect has a characteristic positron lifetime, τ, determined by the overlap of electron and positron charge densities

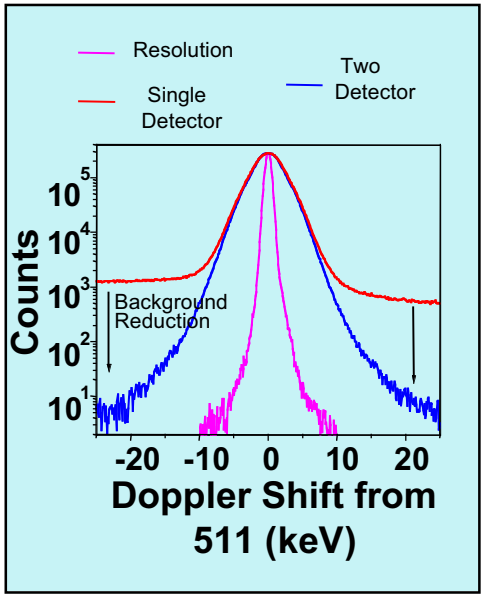
Doppler broadening of annihilation photons

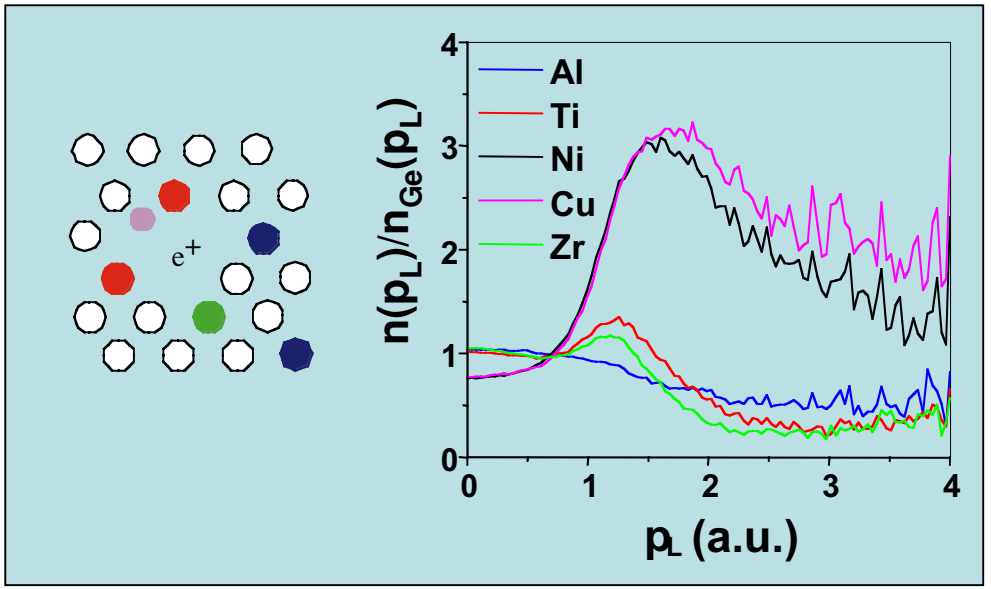
Doppler shift probes the local electron momentum



Doppler shift, ΔE Is proportional to electron momentum, p_L

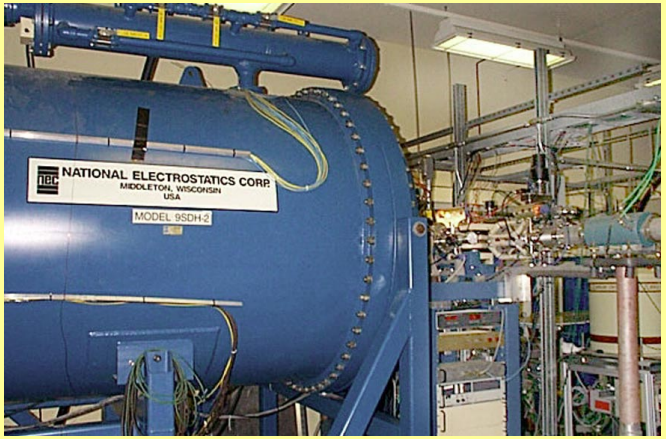
Coincident, two - detector spectroscopy improves signal-to-noise ratio





Orbital Electron Momentum Spectrum (OEMS) provides elemental identification at positron 'trapping' (localization) sites

PAS systems at LLNL

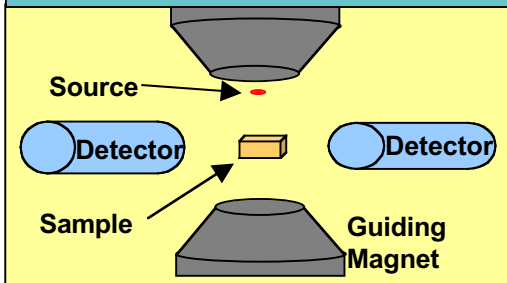


Pelletron

- 2-5 mm
- 2-3 MeV

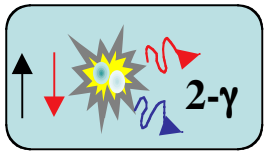
- Measurements of:
  - positron lifetime
  - OEMS using coincidence Doppler broadening

Low energy source

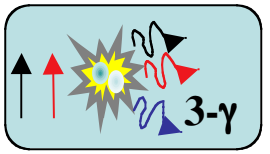


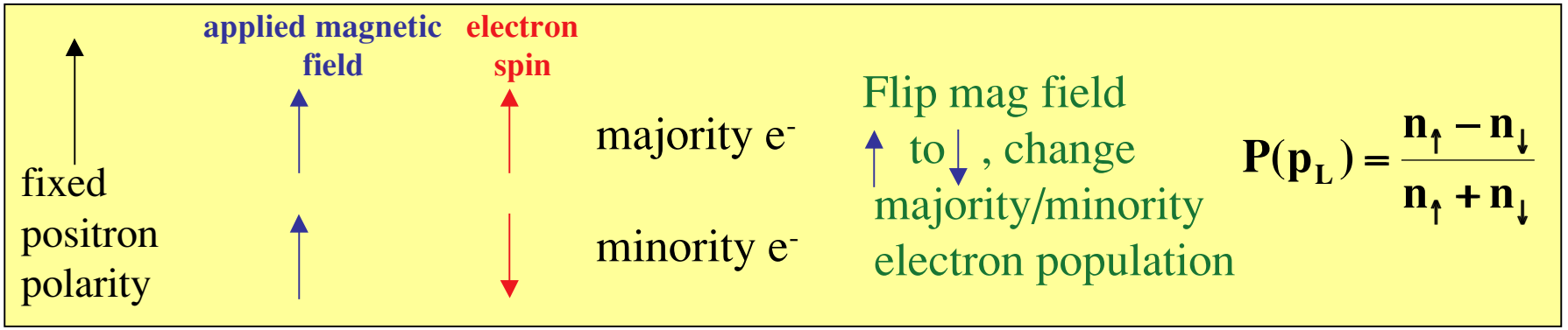
- 30-100 μm

- Measurements of:
  - OEMS using CDB
  - Low temperature OEMS
  - Magnetic polarization OEMS



Magnetic polarization OEMS





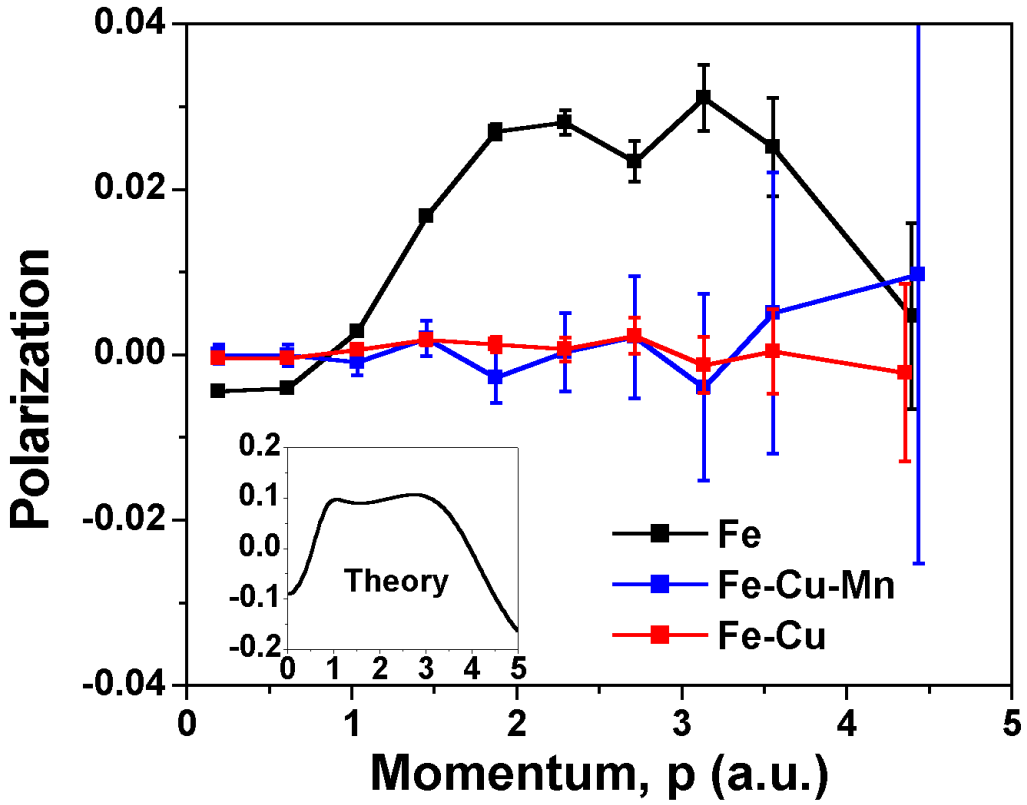
Spin dependent measurements confirm Cu precipitates are non-magnetic

OEMS measured with magnetic field reversal

Contrast obtained when polarized positrons annihilate with electrons of parallel and anti-parallel spins.

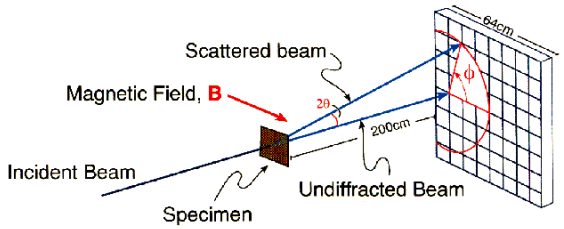
$$P = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}}$$

In aged alloys, analysis sets an upper limit of 2% for Fe contribution.



Measurements indicate the nanometer Cu-Mn precipitates are non-magnetic & confirm SANS analysis

Small angle neutron scattering (SANS)

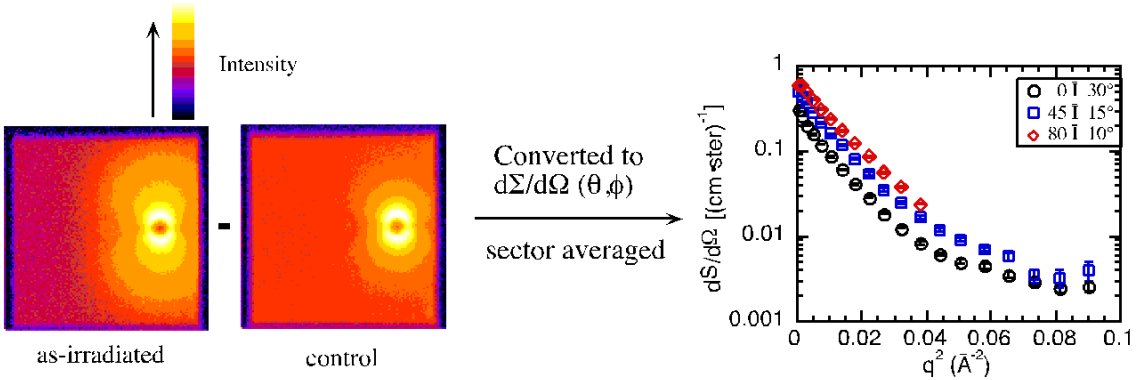


- SANS measurements on 8-m instrument at NIST in a strong, > 1.7T magnetic field and scattering measured on a 64x64 cm (128x128 pixel) position sensitive detector, 2-m from sample and rotated 5° off axis. Effective q-range:

  0.04 - 0.33 Å^{-1} parallel

  0.04 - 0.23 Å^{-1} perpendicular

- A defect signal is obtained by subtracting the corrected signal of an unirradiated control specimen and converting to an absolute cross section using a water standard



- Magnetic/Nuclear scattering ratio determined over the q-range ($0.09 < q < 0.19$ Å^{-1})

Single and two-feature non-linear least squares fits assuming a log-normal size distribution of spherical, magnetic holes to obtain: mean radii, volume fractions and number densities

Key assumption - precipitates are non-magnetic

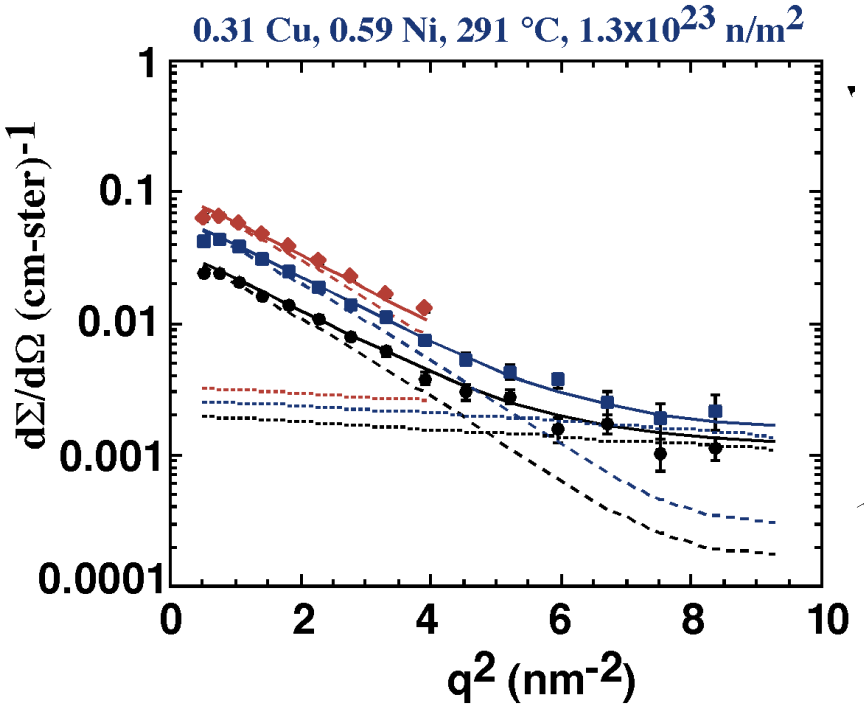
SANS data analysis

$$\frac{d\Sigma(q,\phi)}{d\Omega} = N_d V^2 \Delta\rho^2 S(qr)$$

$$d\Sigma/d\Omega(q,\phi) = d\Sigma/d\Omega_N(q) + d\Sigma/d\Omega_M(q)\sin^2(\phi)$$

$$\Delta\rho^2 = \Delta\rho_n^2 + \Delta\rho_m^2 \sin^2(\phi)$$

$$S(qr) = 3\{[\sin(qr) - qr\cos(qr)]/(qr)^3\}^2$$



- Assume log-normal size distribution of spherical, magnetic holes, fit parameters (r_m, β, M/N and $d\Sigma/d\Omega_M(0)$) with fixed $\Delta\rho_m^2$

$$\langle r \rangle = r_m \exp(0.75\beta^2)$$

$$N_d = (3/4\pi)^2\{\exp(-9\beta^2)/[r_m^6\Delta\rho^2]\}d\Sigma/d\Omega_M(0)$$

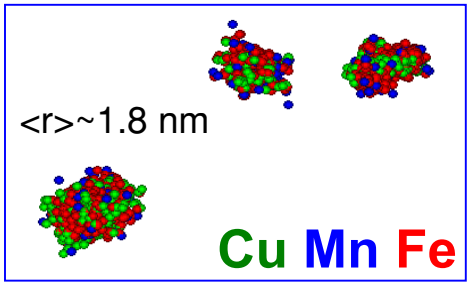
$$f_v = (3/4\pi)\{\exp(-6.75\beta^2)/[r_m^3\Delta\rho^2]\}d\Sigma/d\Omega_M(0)$$

- Composition estimated from Magnetic to Nuclear scattering ratio

$$\sqrt{N/M} = 1.58 - 1.58X_{Fe} - 1.22X_{Cu} + 0.59X_{Mn} - 1.63X_{Ni}$$

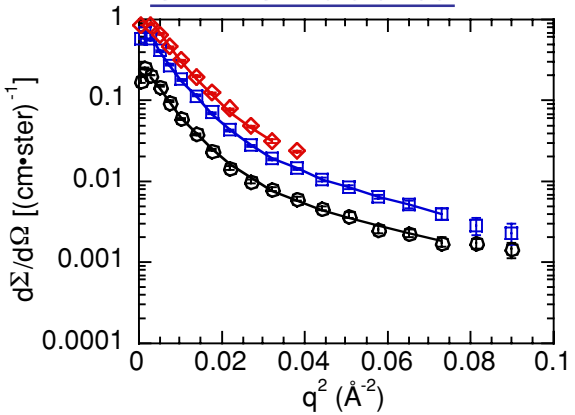
Fe-0.9%Cu-1.0%Mn
288°C irradiated, 1.0×10^{23} n/m^2

Atom Probe results



52% Fe, 44% Cu, 4% Mn

SANS results



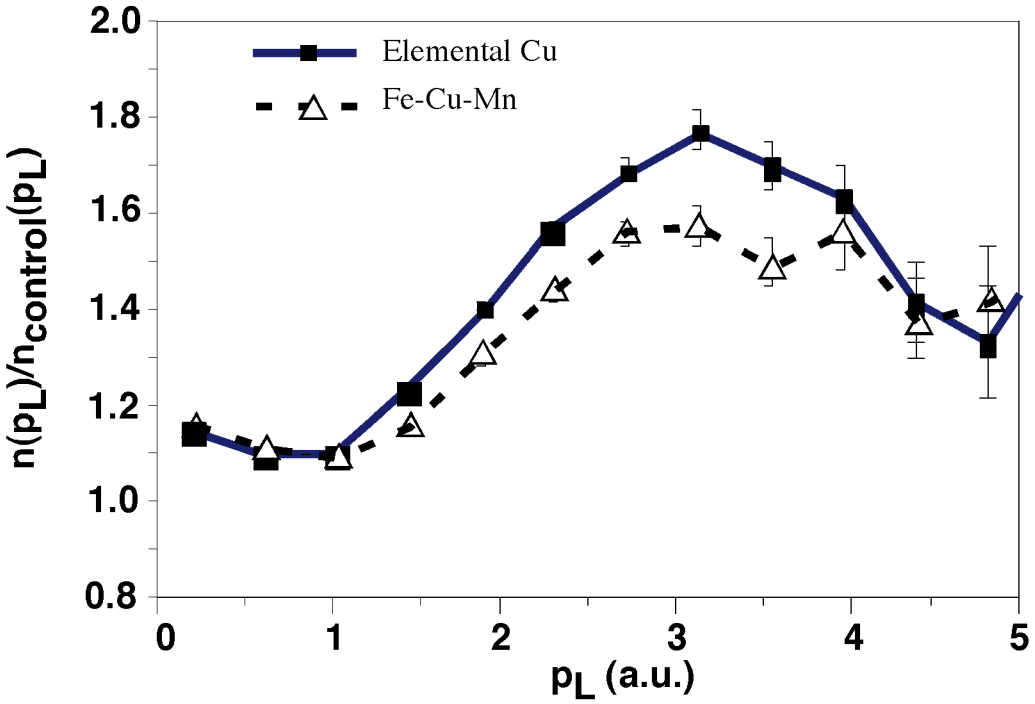
<r> ~ 2 nm

94% Cu/6% Mn

<τ> = 120 ps

87 ps (31%)

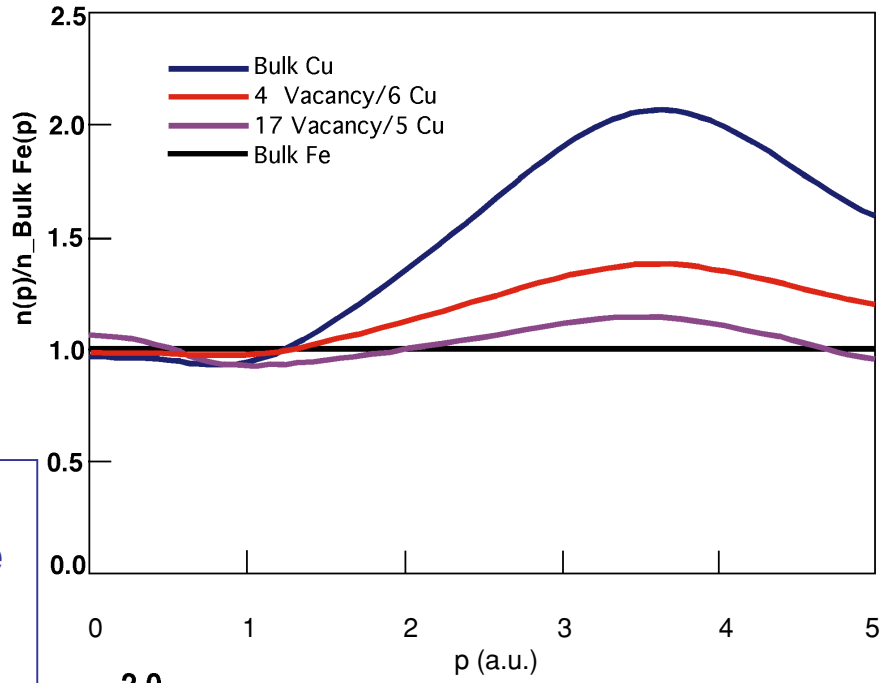
134 ps (69%)



Positron lifetime & OEMS reveal that positrons localize in Cu precipitates - defect free & nearly pure Cu (~90%)

Modeling predictions & positron theory

Predicted positron OEMS

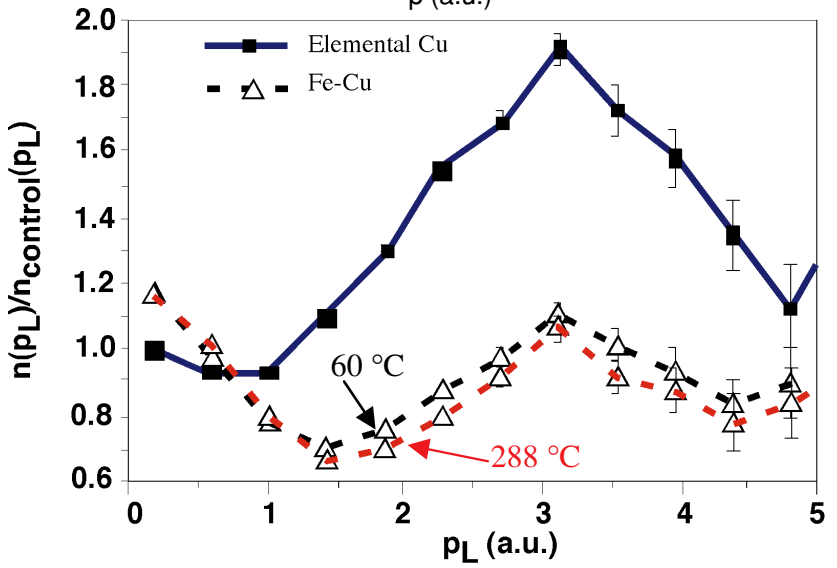


Predicted positron lifetimes

Vac/Cu	τ (ps)
3/6	173
4/6	216
17/5	350
>50/x	520

Qualitative OEMS, Quantitative lifetime agreement confirm Vac-Cu clusters

Measured positron OEMS Fe-0.9%Cu



Measured positron lifetimes

τ (ps) - 60°C
178
355
τ (ps) - 288°C
222
520

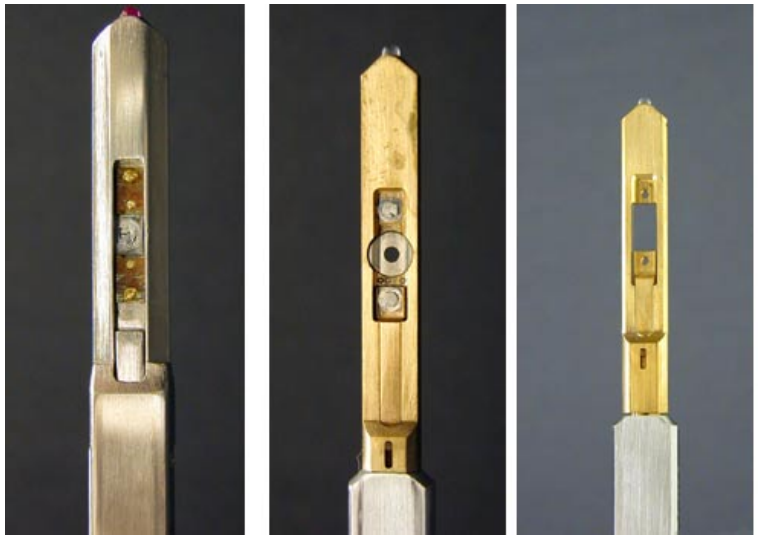
In-situ TEM Deformation

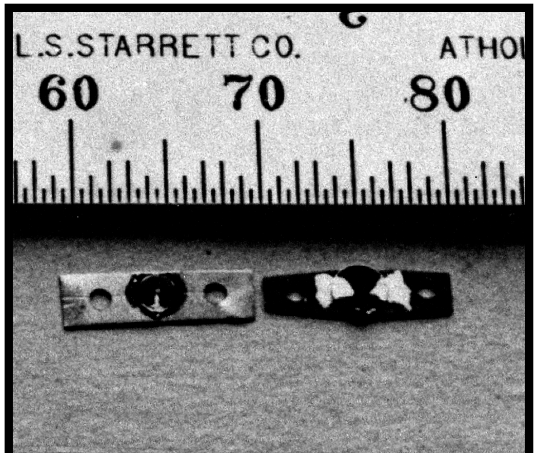
- allows observation of dynamic events (video)
- high spatial and temporal resolution

Specialized TEM stages allow

- heating and straining
- cooling and straining
- double tilt and straining

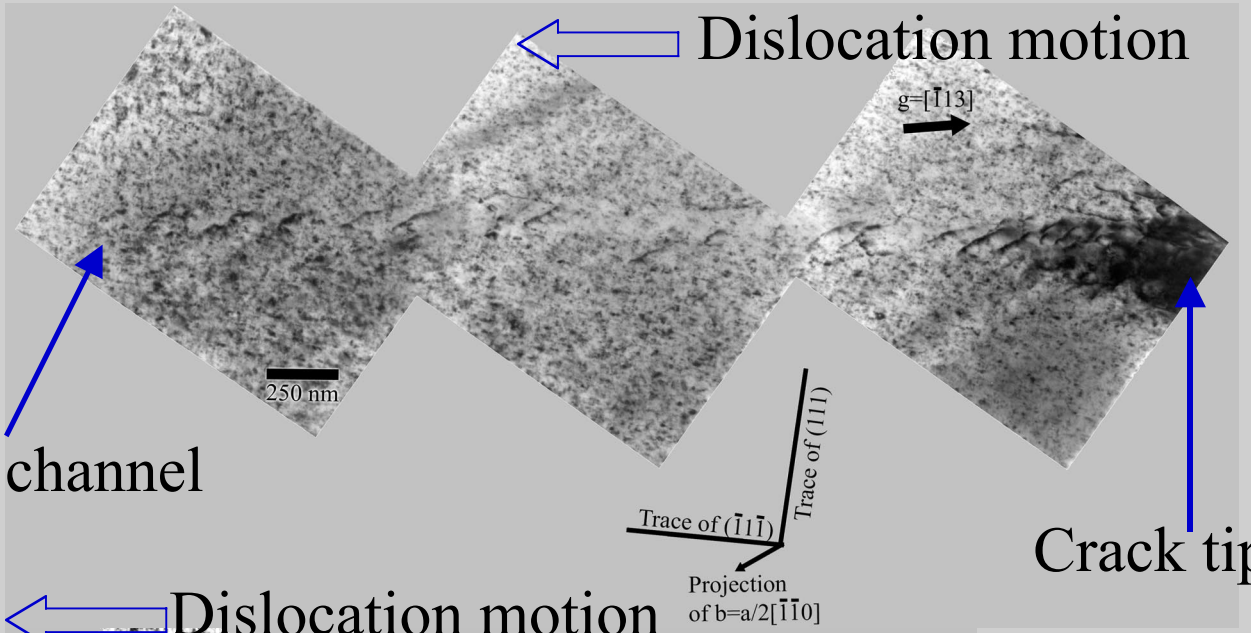
Ambient Heating Cooling





Observed Dislocation Sources

Channel initiated by crack tip



Channel initiated by grain boundary