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Electrical Resistivity and Optical Reflectance Measurements of $ErH(D)_2$ Films on Silica Substrates

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Color change during loading relates to metallic to semiconductor (MS) transition. Both electrical resistivity and optical reflectance measurements seek to quantify this phenomena.

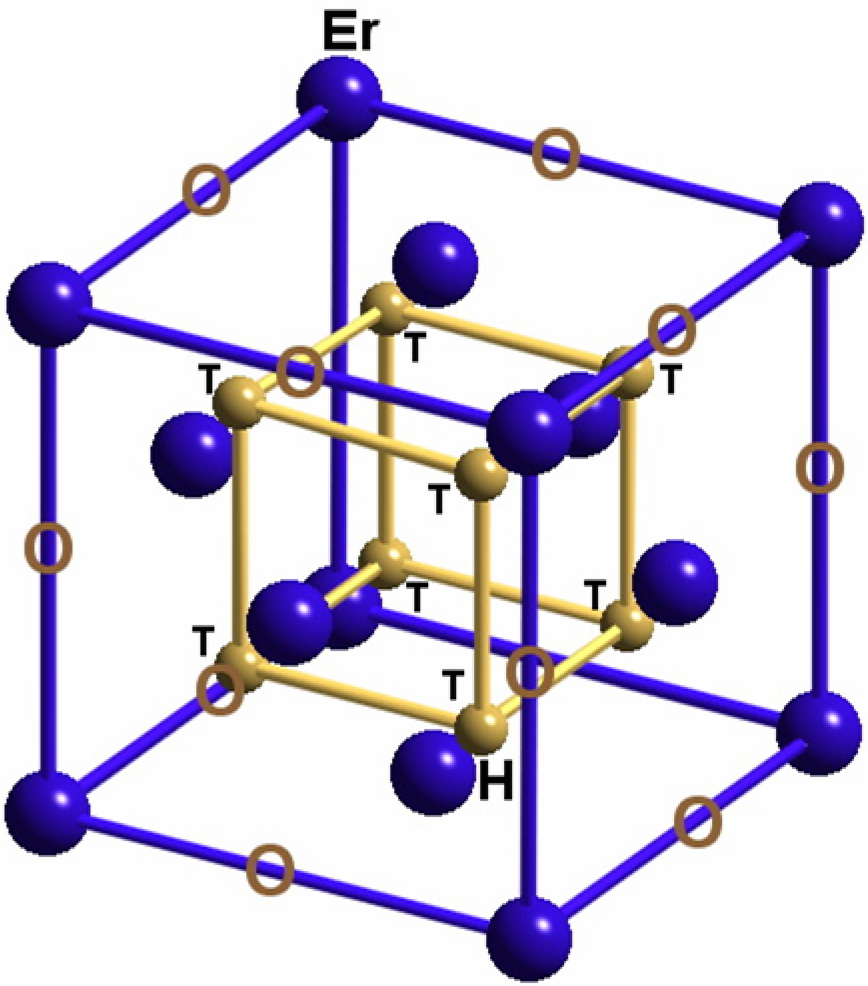
- Color changes (e.g. "robin's-egg blue,olive green") have empirically been used as an indicator of the loading process.

•Loading/unloading of Er films deposited on silica (transparent, insulating) substrates provides a useful "model" system to quantify the observed color changes in terms of optical and electrical properties measurements.

Silica substrates were chosen for the first attempt to correlate resistivity and reflectance on identical samples.

- SNL provides "mirror-like" Er coating on Silica
  - 25mm dia. x 3mm thick "standard blanks"
  - 400 nm Er coating with native oxide (10 nm)
- LANL D-loads films
- LANL measures temperature-dependent resistivity
- LANL measures absolute specular reflectance (ASR) and transmission

Temperature-dependent electrical resistivity is sensitive to hydrogen ordering in the crystal lattice.



β - phase (fluorite structure)

two tetrahedral sites ideally occupied.

Additional hydrogens *x* occupy octahedral

sites x_{max} ~ 0.1 $\rightarrow$ trihydride γ phase.

β-phase (metallic or semi-metallic).

Hexagonal γ-phase (semiconducting).

Temperature-dependent electrical resistivity is extremely sensitive to hydride stoichiometry, but not the oxidation layer thickness.

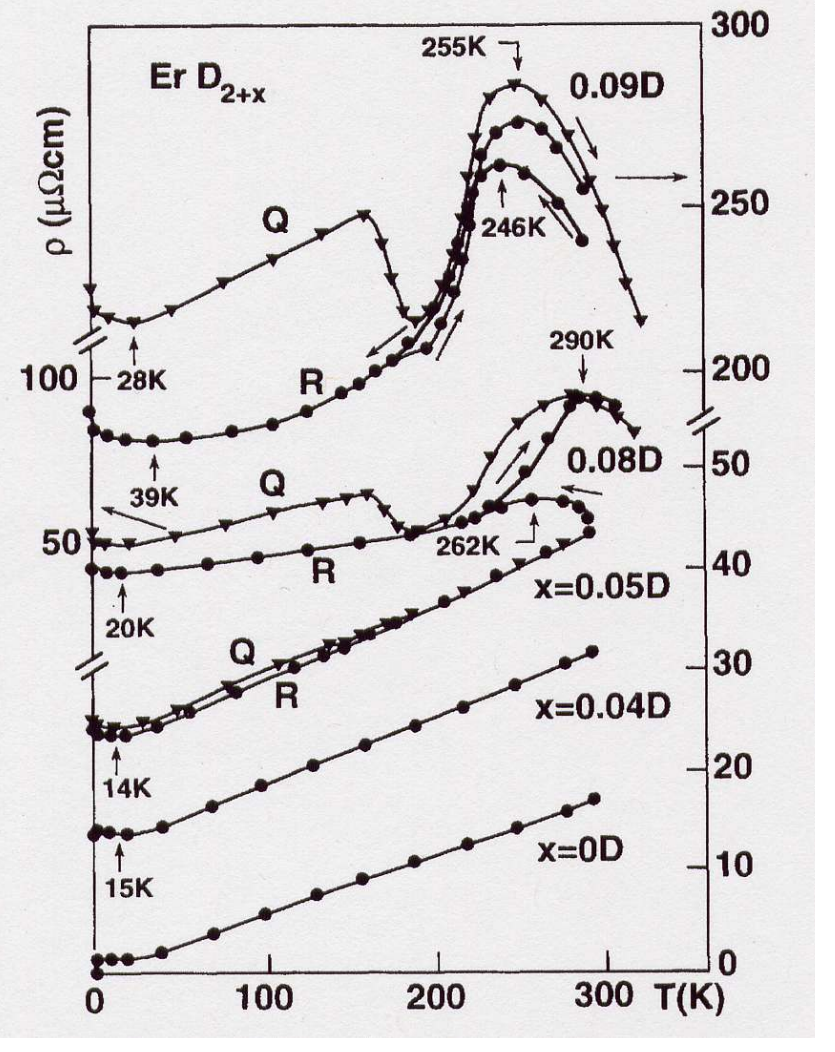
ErD_{2+x}

Complex system that exhibits:

(1) structural ordering in octahedral H sublattice,

(2) concentration dependent magnetic transitions, and

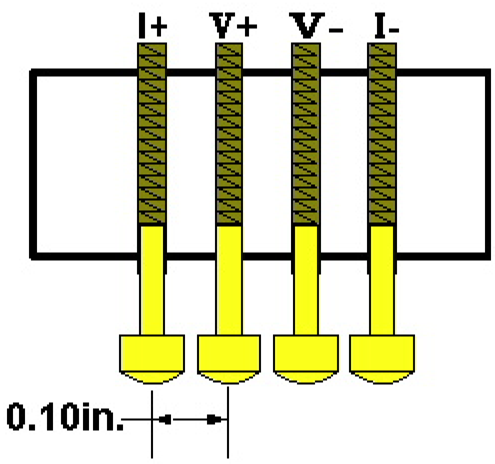
(3)metal-to-semiconductor transitions.



Vajda and Daou, *Phys. Rev. B* **49**, 3275 (1994).

The electrical resistivity measurement is performed using a cryostatically mounted four-point probe apparatus.

400-nm Er films deposited onto silica substrates (Sandia). D-loaded at LANL.



<u>Standard four-point probe</u>

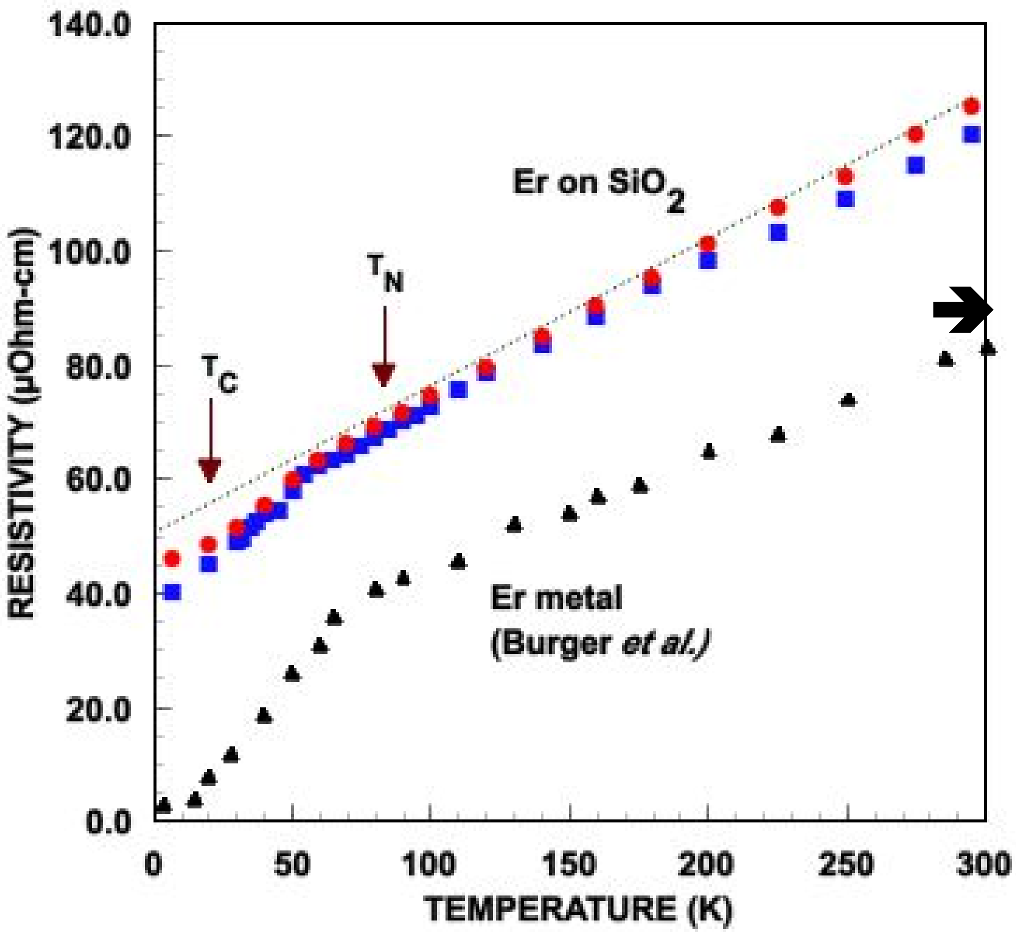
Resistivity for a thin film of thickness *t* (*t* << s, where *s* is probe spacing) is

$$\rho = \frac{\pi t}{\ln 2}\left(\frac{V}{I}\right)$$

Figure of merit:

$$\Delta\rho_{\min} \approx 0.05\mu\Omega - cm$$

Temperature-dependent electrical resistivity of Er/silica indicates presence of impurities in Er layer.



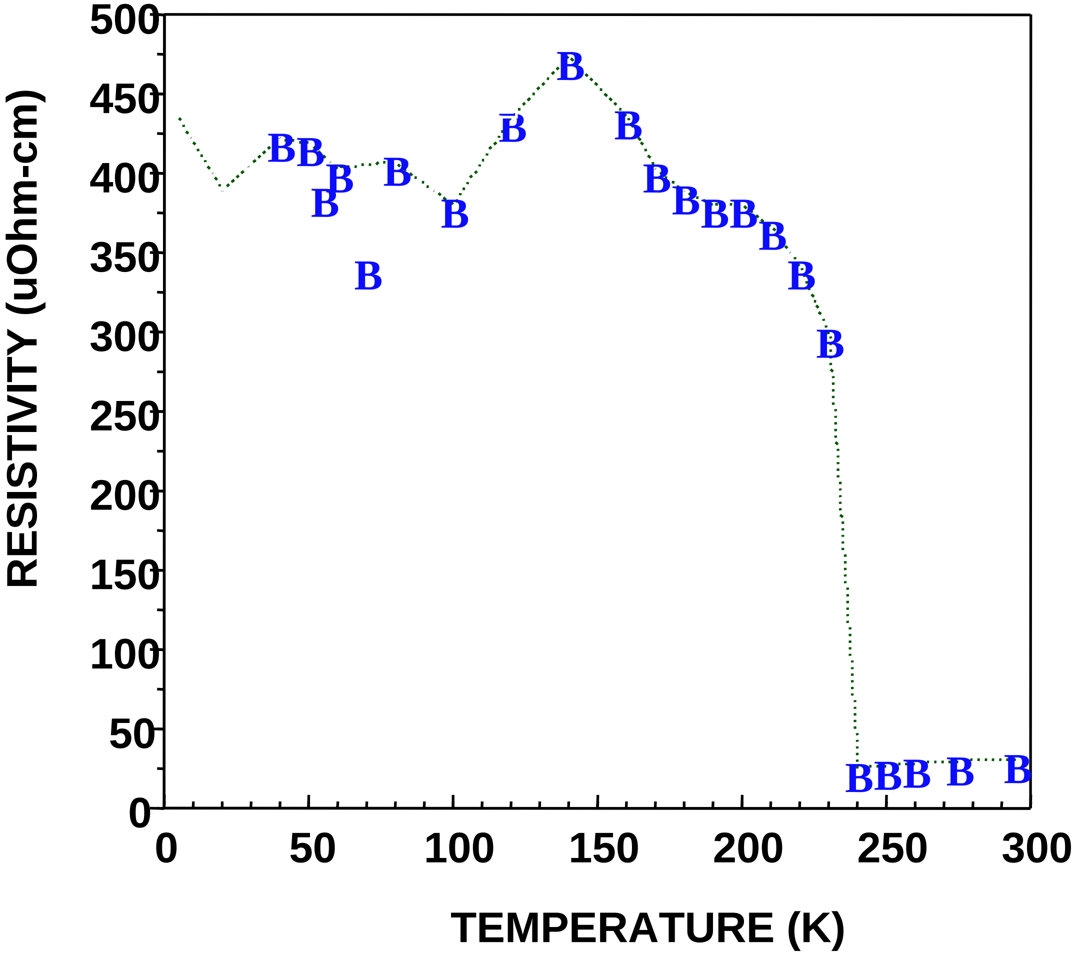
Residual resistivity ratio (RRR)

RRR =ρ(300 K)/ ρ(4 K)
RRR ≅ 125/42 ≅ 3
From Burger, *et al.*
RRR ≅ 83/4 ≅ 20

$$\rho(T) = \rho_r + \rho_{mag}(T) + \rho_{ph}(T)$$

residual | **magnetic spin scattering** | **phonon scattering**

Temperature-dependent electrical resistivity of "nominal" ErD_2/silica indicates lower room temperature resistivity.



$ErD_{2\pm x}$
As received

Complex electronic behavior.

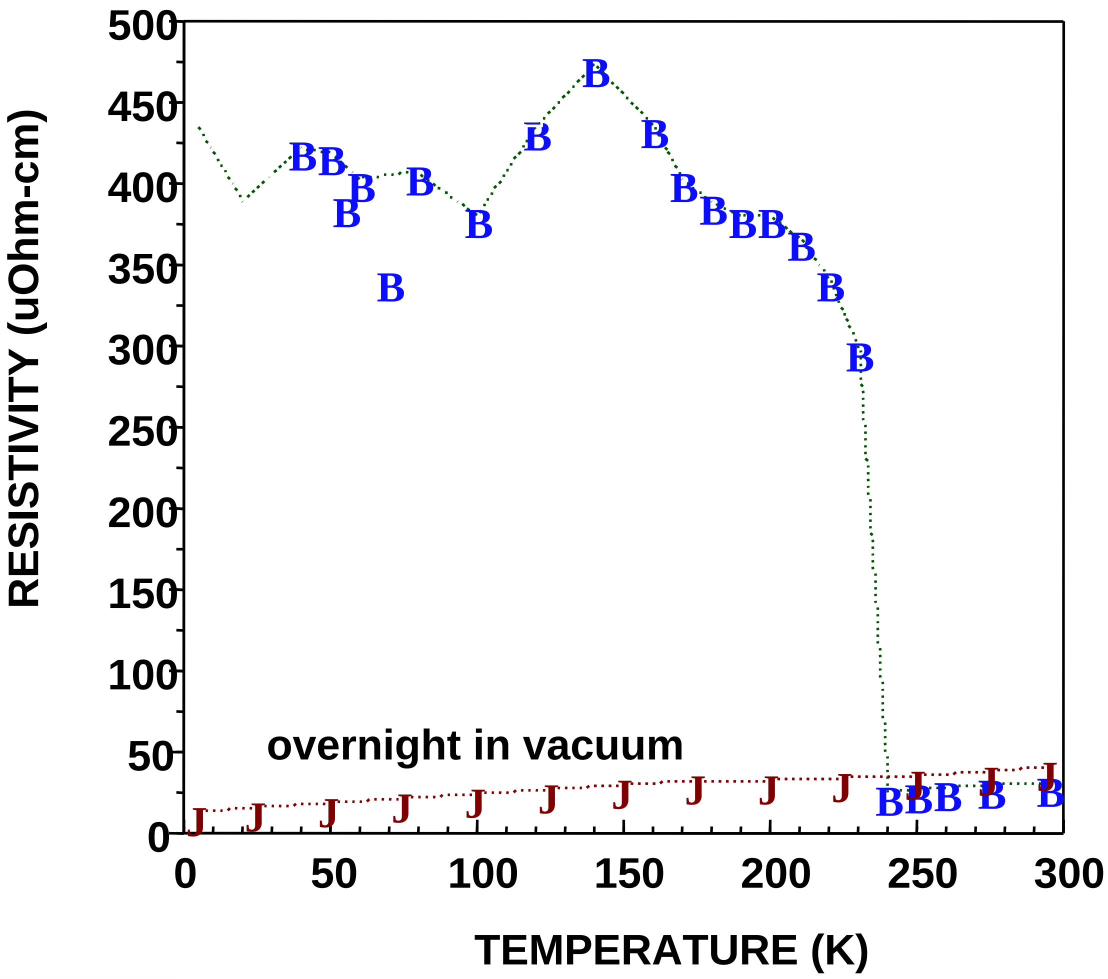
Very unstable--consistent with Vajda observations.

Stoichiometry may be close to β-γ transition.

ρ (295K) = 31 μΩ-cm

Similar value reported By Provo (29.1 μΩ-cm).

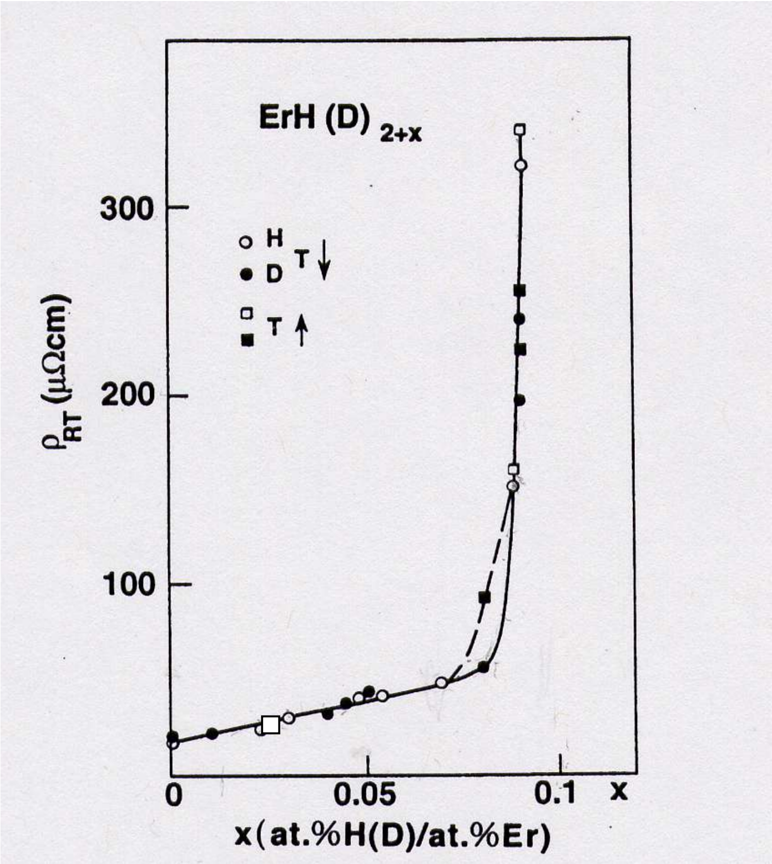
Temperature-dependent electrical resistivity of "nominal" ErD_2/silica shows metallic behavior after vacuum aging.



$ErD_{2\pm x}$

Sample was left in vacuum (~ 10^{-6} Torr) overnight with pump off. Res. vacuum (~ 10^{-1} Torr). Chamber evacuated, sample cooled to 4 K and data taken.

Room temperature electrical resistivity of ErD_2/silica suggests stoichiometry at $ErD_{2.025}$.



Vada and Daou, *Phys. Rev. B* **49**, 3275 (1994)

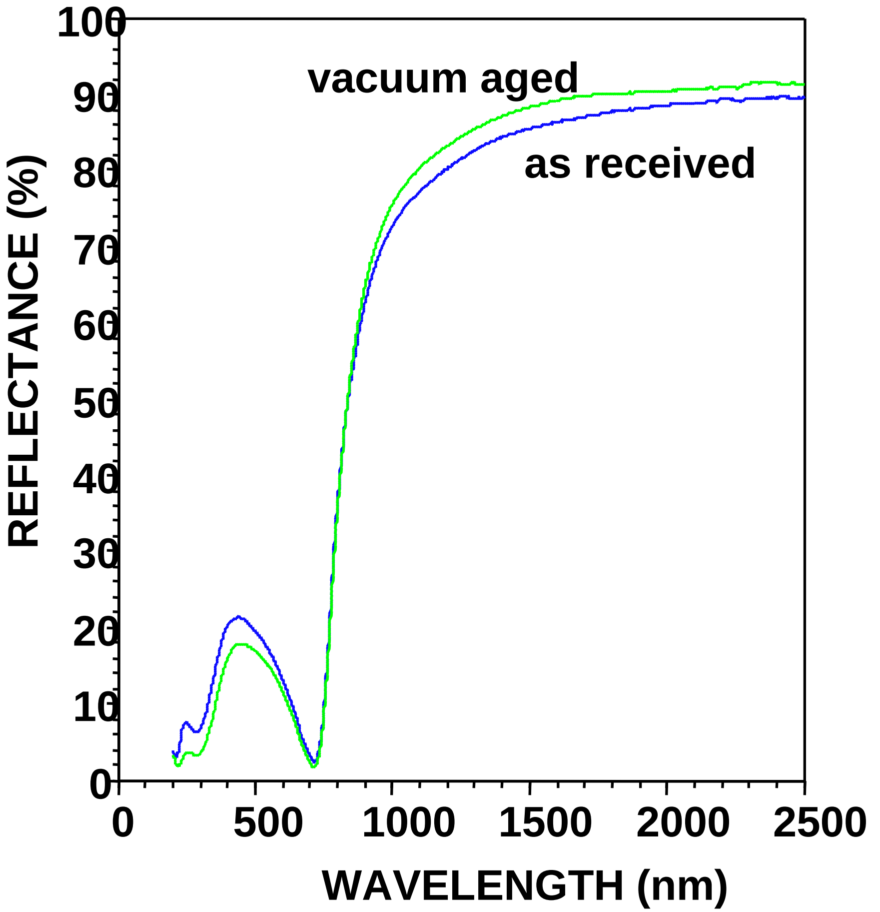
ErD_{2+x} resistivity vs. *x*

At the β limit (*x* ~ 0.09) there is strong divergence of room-temp. resistivity.

Our ErD_{2+x} data (either sample) show resistivity of *ca*. 40 μΩ-cm indicating stoichiometry near 2.025.

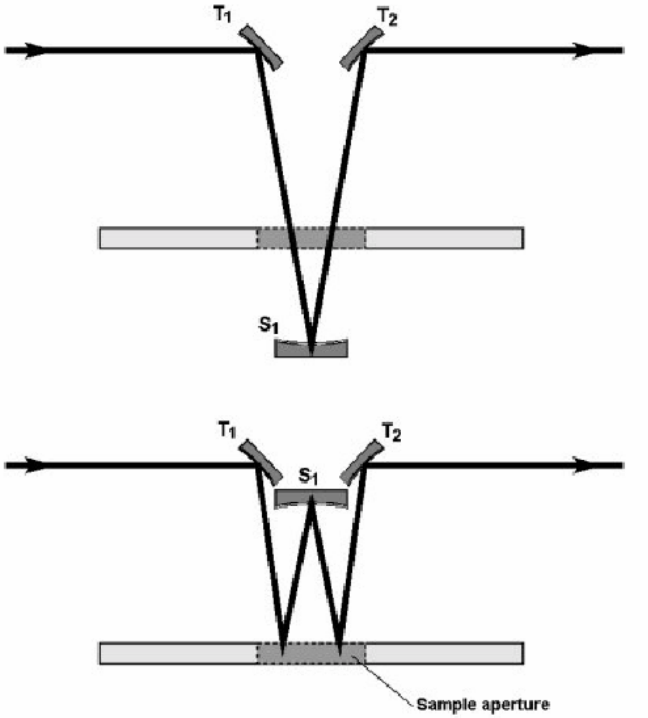
Extreme sensitivity of resistivity to stoichiometry near β limit !

$ErD_{2\pm x}$ Reflectance: observed changes in absolute specular reflectance (ASR) correlate with resistivity change.

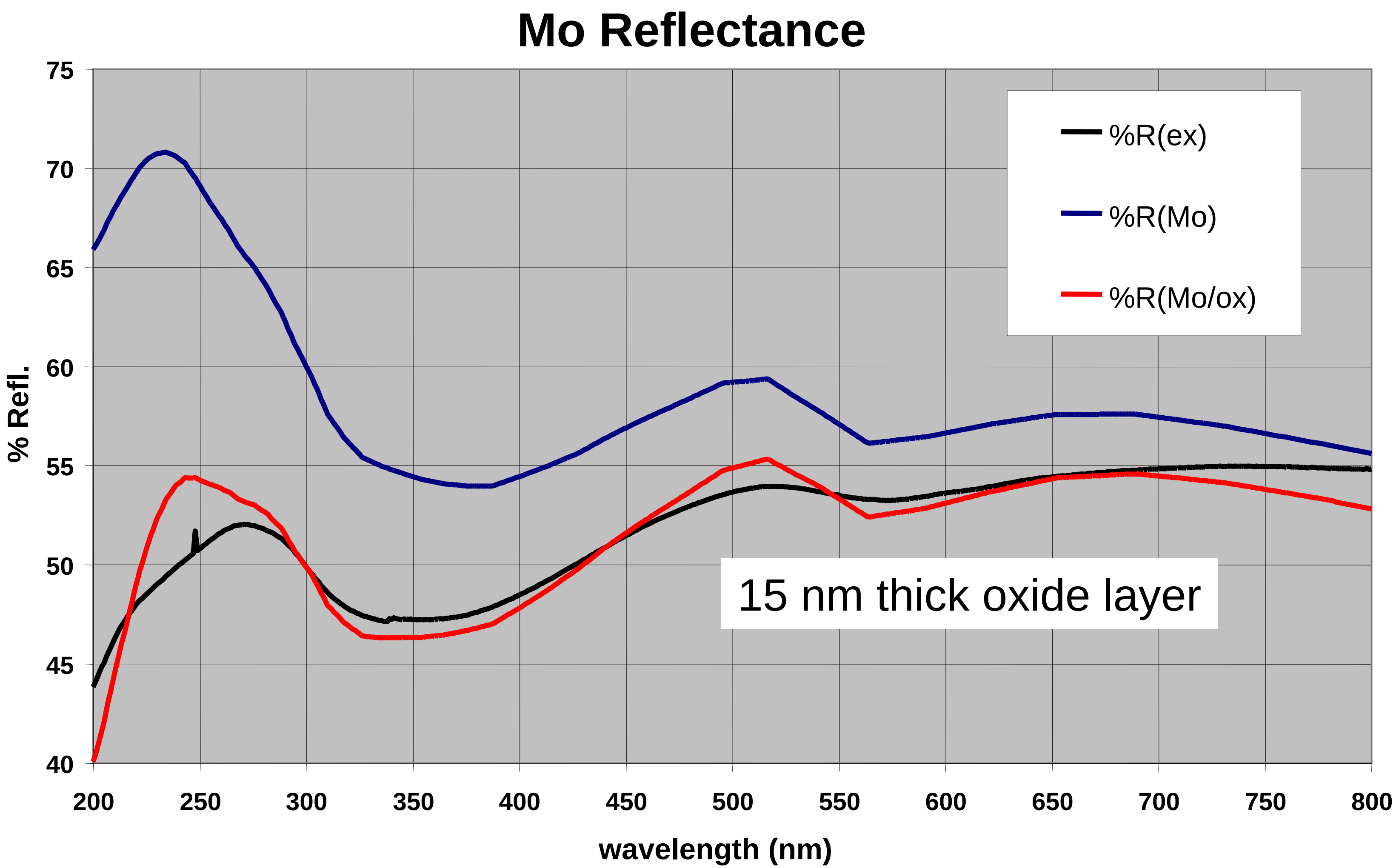


ASR

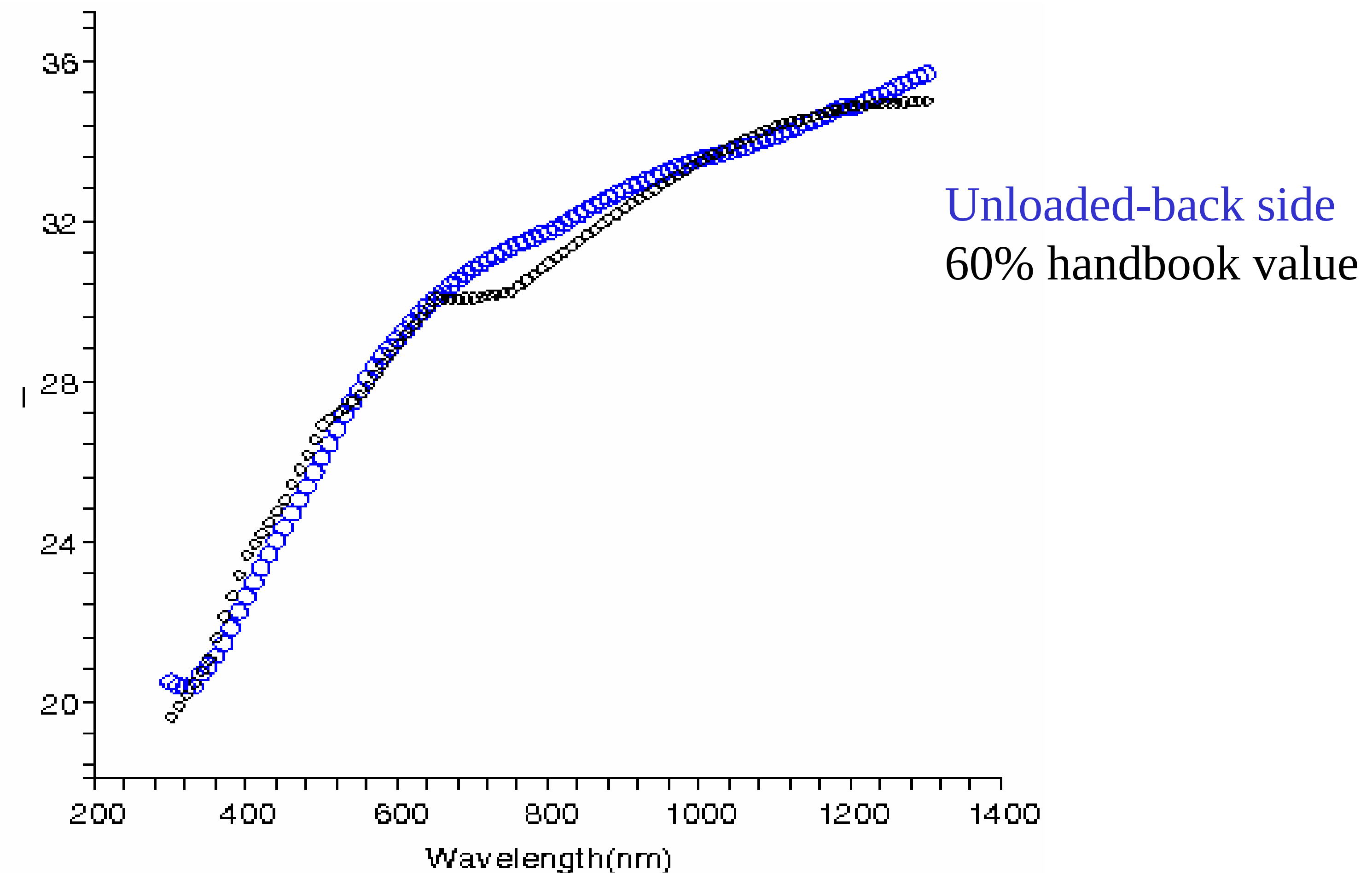
Absolute error ±2%
Relative error ±0.1%



Thin dielectric overlayers can dramatically reduce metal reflectance.



ASR:Unloaded Er film (substrate side)-The reduced reflectance implies a dielectric layer of higher refractive index than the incident media (n > 1.52).



The Extended Drude Model provides the simplest approach to calculate optical reflectance from metals *and* dielectrics.

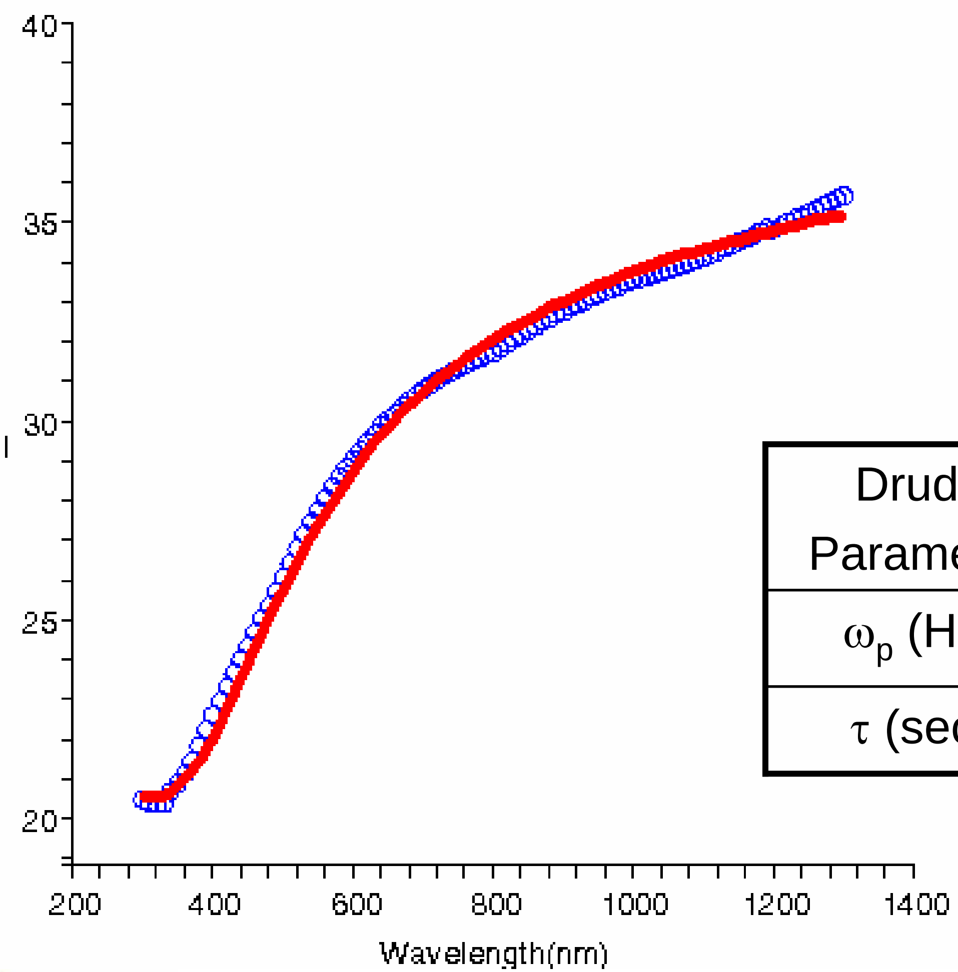
Fresnel Equation: $$R = \frac{(1-\sqrt{\varepsilon_r})^2}{(1+\sqrt{\varepsilon_r})^2} = \frac{(1-n)^2 + k^2}{(1+n)^2 + k^2}$$

Drude Equation: $$\varepsilon_r = \varepsilon_m \left(1 - \frac{\omega_{sp}^2}{\omega^2 + \frac{i\,\omega}{\tau}}\right) + \varepsilon_d \left(1 - \frac{\omega_{sp}^2}{\omega^2 - \omega_g^2 + \frac{i\,\omega}{\tau}}\right)$$

$$\omega_{sp}^2 = \frac{\omega_p^2}{\varepsilon_m} \qquad \omega_p = 4\frac{\pi\, n\, e}{m} \qquad E_g = h\,\omega_g$$

Optical conductance: $$\sigma(\omega) = \frac{\omega\varepsilon_2}{4\pi} \qquad \rho_{op} = 4\frac{\pi}{\tau\,\omega_p^{\;2}}$$

ASR:Unloaded Er film (substrate side)- The wavelength dependence is dominated by the “bulk” Er reflectance.

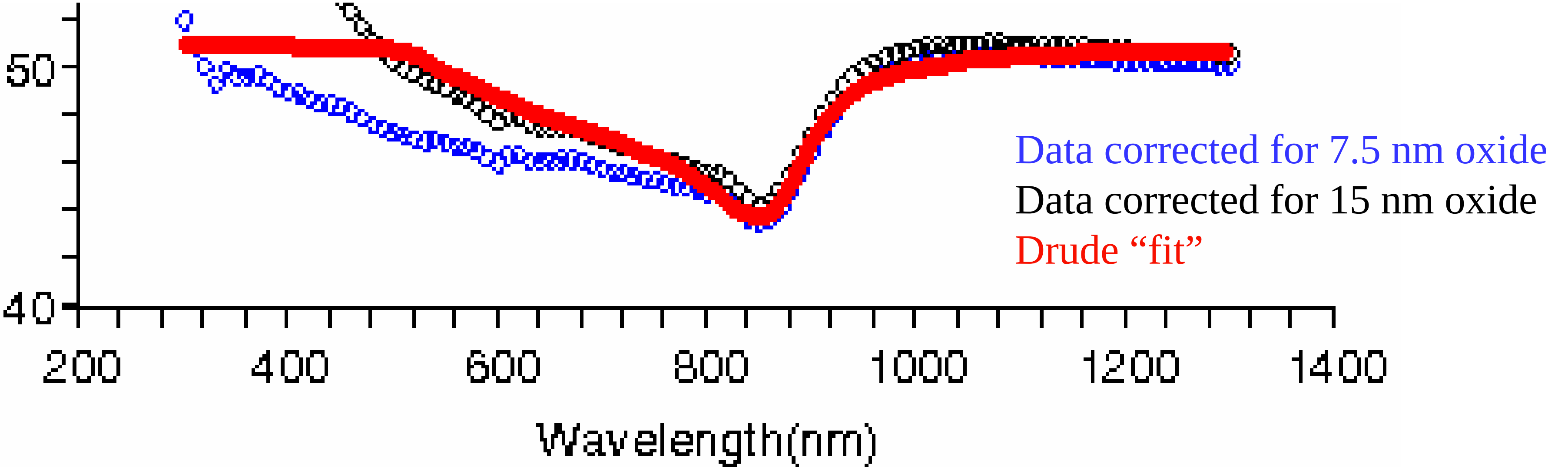


Unloaded-back side

Drude parameter “fit”

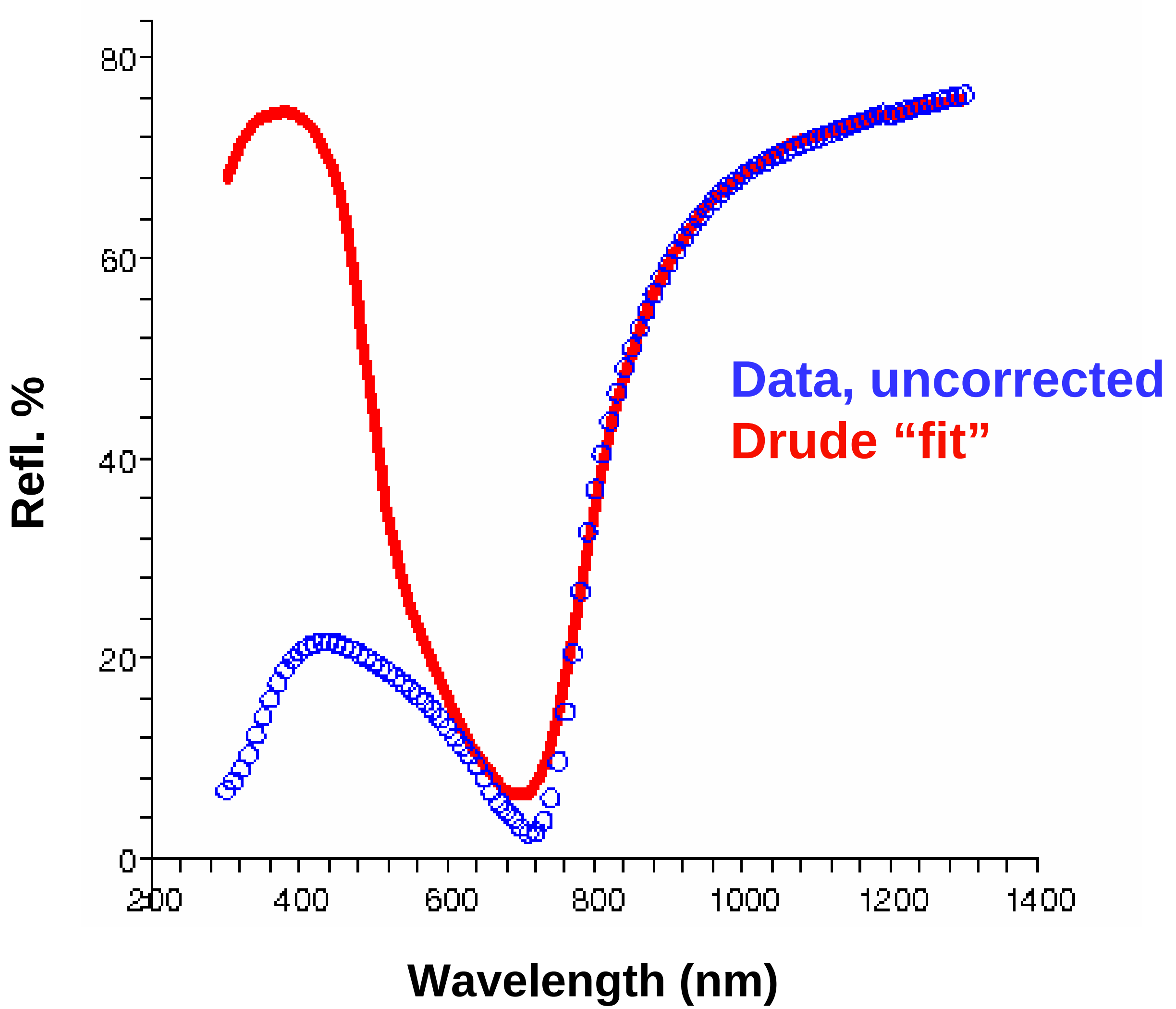
Drude Parameter	Calculated Value	Fit Value
ω_p (Hz)	2.84×10^{16}	2.52×10^{16}
τ (sec)	0.167×10^{-15}	0.25×10^{-15}

ASR:Unloaded Er film (front side)- To a first approximation, the reflectance is described by a thin oxide layer atop the Er metal.

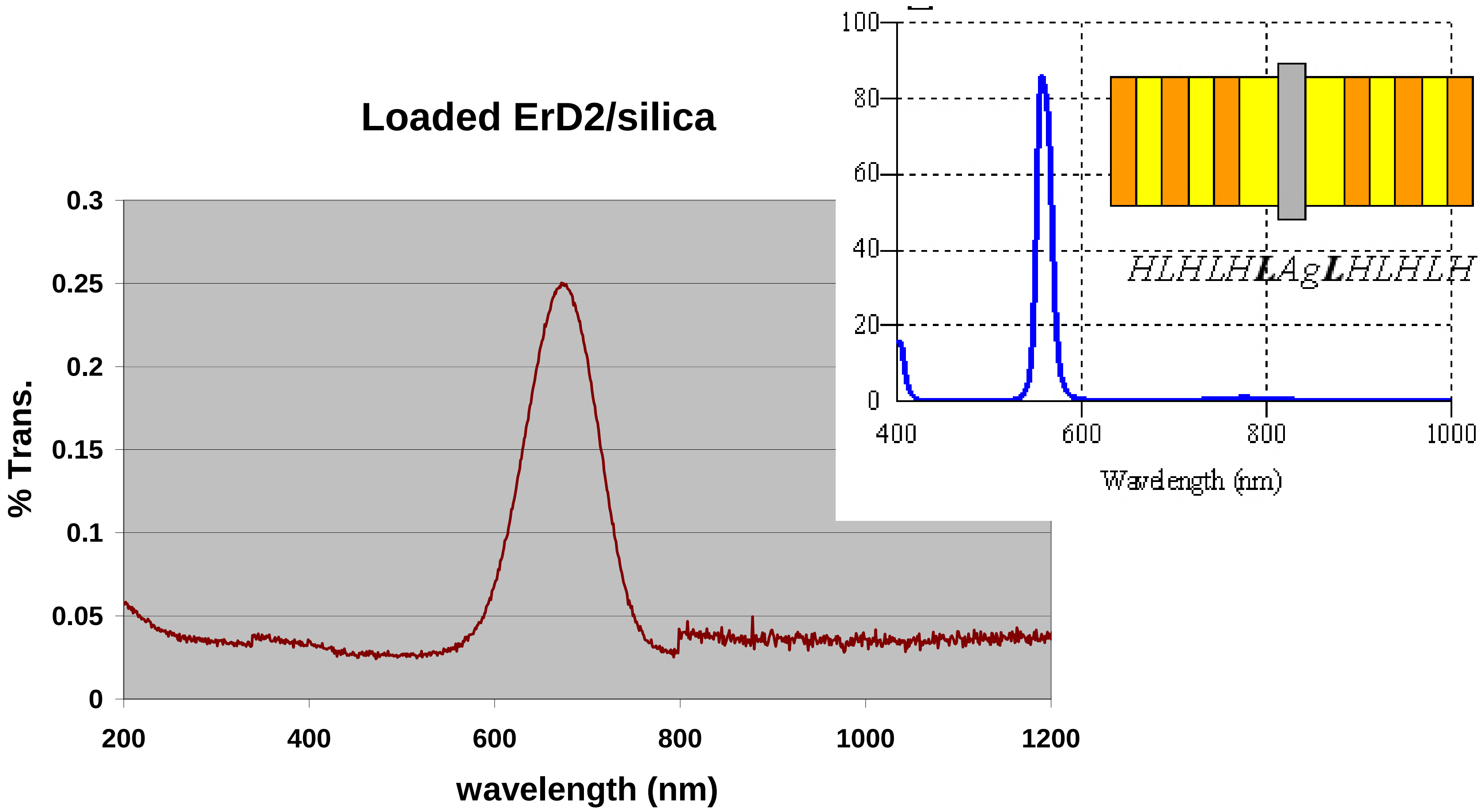


- **still not good fit in visible region**
- ω_g, ε_m, ε_d **variables highly correllated**

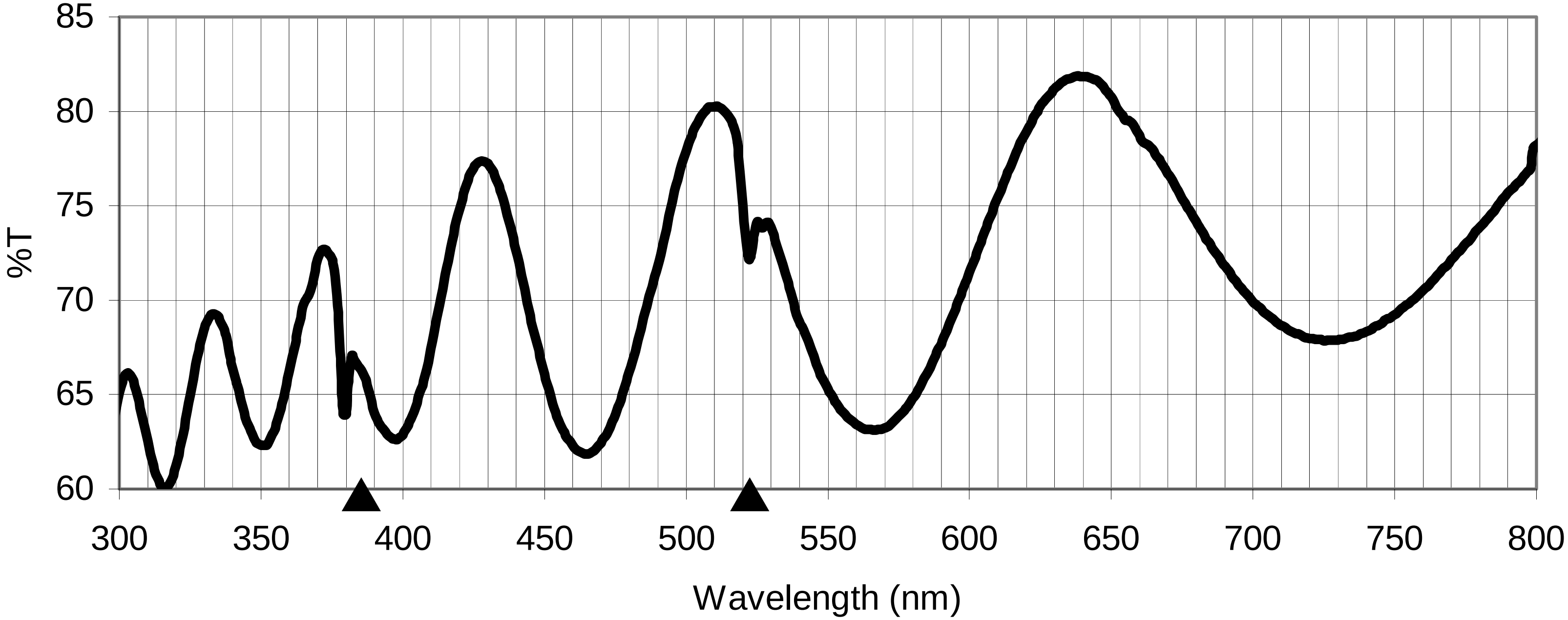
ASR:Loaded ErD_2 film (front side)- Measured reflectance in NIR due to Er metal, indicative of incomplete loading.



Transmission: Loaded ErD_2 – Similar to metal bandpass filter, indicates optically thick Er metal with symmetric dielectric layers.



Transmission: Oxidized Er_2O_3 film – Absorption peaks consistent with Er^{3+}-doped SiO_2



- **Annealed in oxygen at 800 C for 40 hours.**
- **$^4G_{11/2} \rightarrow {}^4I_{15/2}$ at 380 nm and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ at 520 nm Er^{3+} absorption**

Conclusions

- **Reaction between Er and silica substrate.**
  - **Increased ρ, decreased R**
  - **Er^{3+} doped glass observed after annealing.**
- **Loaded films exhibit complex behaviors**
  - **"Partial" loading due to low emissivity substrate**
  - **Resistive transition at 235 K; disappears after vacuum aging**
  - **Reflectance and transmission indicate Er metal still present.**
- **Repeat study using polished diamond substrates**
  - **Transparent and insulating**
  - **High thermal conductivity (thinner substrate)**
  - **Chemically unreactive**