American Institute of Aeronautics and Astronautics
SciTech Forum

Thermophysical Model for the Infrared Emissivity of Metals: Application to Nickel

Jeremy Orosco and Carlos F. M. Coimbra*
Center for Energy Research
University of California San Diego
January 9, 2019

*correspondence should be directed to this author: ccoimbra@ucsd.edu

CER Center for Energy Research

COIMBRA RESEARCH GROUP

UC San Diego
JACOBS SCHOOL OF ENGINEERING
Mechanical and Aerospace Engineering

AIAA
Shaping the Future of Aerospace

In this presentation

What?

- model derived for temperature-dependent infrared emissivity of metals
- applied to nickel

How?

- generalizing a well-known model for carrier transport
  1. derive anomalous carrier transport model
  2. determine functional form for the temperature-dependent parameters

Why?

- knowledge of a material's emissivity is important for a wide range of design applications: signal suppressing metamaterials [1], engine componentry [2], non-contact thermometry [3], etc.

[1] H. Kocer, *et al.*, "Reduced near-infrared absorption using ultra-thin lossy metals in Fabry-Perot cavities," *Sci. Rep.*, Vol. 5, 2015, p. 8157.

[2] G. Teodorescu *et al.*, "Normal emissivity of high-purity nickel at temperatures between 1440 and 1605 K," *J. Phys. Chem. Solids*, Vol. 69, No. 1, 2008, pp. 133–138.

[3] K.-H. Weng, C.-D. Wen, "Effect of oxidation on aluminum alloys temperature prediction using multispectral radiation thermometry," *Int. J. Heat Mass Tran.*, Vol. 54, No. 23, 2011, pp. 4834–4843.

Simple model for free-carrier transport

Drude(-Langevin) transport of a single charge carrier

- predicts response of "good" free-electron metals (*e.g.*, monovalent noble metals) [4]
- classically, a microscopic equation of motion in the carrier velocity [5]:

$$m^* \frac{dv(t)}{dt} + \zeta\, v(t) = q\,\mathcal{E}(t) + q\,\eta(t)$$

local field

random field fluctuations

- leads to the complex-valued (electric) susceptibility (*i.e.*, the Fourier transform of the polarization density):

$$\chi_d(\lambda) = \frac{i\,\lambda}{2\,\pi\,c_0\,\varepsilon_0}\sigma_d(\lambda) = -\frac{\lambda^2}{2\,\pi\,c_0\,\varepsilon_0}\left(\frac{\sigma_\gamma}{i\,\lambda + \lambda_\gamma}\right)$$

[4] F. Wooten, *Optical Properties of Solids*, Academic Press, New York, 1972.

[5] N. Pottier, *Nonequilibrium Statistical Physics: Linear Irreversible Processes*, Oxford graduate texts, Oxford University Press, Oxford, 2010.

Simple model for free-carrier transport

Drude(-Langevin) transport of a single charge carrier

- predicts response of "good" free-electron metals (*e.g.*, monovalent noble metals) [4]
- classically, a macroscopic equation of motion in the current density [5]:

$$\tau_c \frac{dj(t)}{dt} + j(t) = \left(\frac{n_d\, q^2\, \tau_c}{m^*} \right) E(t)$$

macroscopic (incident) field

DC photonic conductivity, σ_γ

- leads to the complex-valued (electric) susceptibility (*i.e.*, the Fourier transform of the polarization density):

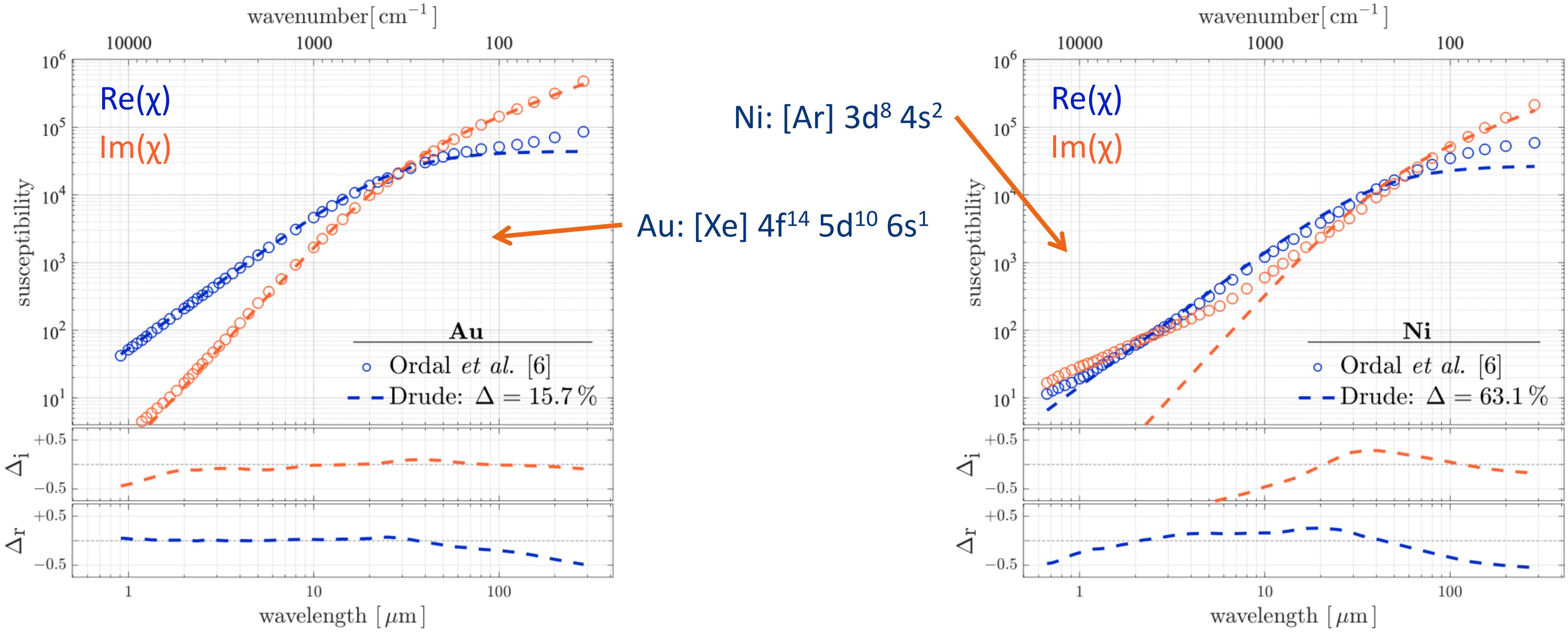
$$\chi_d(\lambda) = \frac{i\,\lambda}{2\,\pi\,c_0\,\varepsilon_0}\sigma_d(\lambda) = -\frac{\lambda^2}{2\,\pi\,c_0\,\varepsilon_0}\left(\frac{\sigma_\gamma}{i\,\lambda + \lambda_\gamma}\right)$$

[4] F. Wooten, *Optical Properties of Solids*, Academic Press, New York, 1972.

[5] N. Pottier, *Nonequilibrium Statistical Physics: Linear Irreversible Processes*, Oxford graduate texts, Oxford University Press, Oxford, 2010.

Applicability of the Drude model

Drude model is insufficient for explaining data produced by media that deviate substantially from the free electron model. Example: nickel.



[6] M. A. Ordal *et al.*, "Optical properties of Au, Ni, and Pb at submillimeter wavelengths," *Appl. Opt.*, Vol. 26, No. 4, 1987, pp.744–752.

Basis for the proposed model: field relaxation with memory

- Drude-Langevin transport for a single charge carrier:

$$m^* \frac{dv(t)}{dt} + \zeta\, v(t) = q\,\mathcal{E}(t) + q\,\eta(t)$$

- generalized with a (temporally) nonlocal relaxation in the (spatially) local field [7] :

$$\widetilde{\eta}(t) = \lim_{t_0 \downarrow -\infty} \overbrace{\int_{t_0}^{t} \mathcal{H}(t-s) \frac{d\mathcal{E}(s)}{ds}\, ds}^{\text{history integral}} + \eta(t)$$

history kernel

historical change rate

- we choose a history kernel the represents a generalization of the Markovian approximation, $H(t) = \delta(t)$:

characteristic time of field relaxation

$$\mathcal{H}(t) = \frac{(\tau_f/\,t)^{\mu}}{\Gamma(1-\mu)}$$

memory decay strength

generalized factorial function

[7] J. Orosco, C. F. M. Coimbra, “Anomalous carrier transport model for broadband infrared absorption in metals,” *Phys. Rev. B*, Vol. 98, No. 23, 2018, 235118.

Imparting temperature dependence on the model

- generalized fluctuating field leads to a fractional differential transport model:

$$\tau_c \frac{dj(t)}{dt} + j(t) = \sigma_\gamma \left(E(t) + \tau_f^\mu \frac{d^\mu E(t)}{dt^\mu} \right)$$

- temperature-dependence modeled in terms of its effect on the collisional rate:

$$\sigma_\gamma(T) = \frac{n_d\, q^2}{m^*} \tau_c(T)$$

- to account for the possibility of anomalous carrier dynamics in regimes distinct from those of the bulk Drude behavior, a (potentially temperature-dependent) time constant scaling law is defined:

$$\tau_a(T) = \alpha(T)\, \tau_c(T)$$

$$\Rightarrow \sigma_a(T) = \frac{n_d\, q^2}{m^*} \alpha(T)\, \tau_c(T)$$

Independent contributions to the current density

- resulting temperature-dependent macroscopic equations

$$\tau_c(T)\,\frac{dj_d(t)}{dt} + j_d(t) = \frac{n_d\,e^2}{m^*}\tau_c(T)\,E(t)$$

Drude component

$$\tau_a(T)\,\frac{dj_a(t)}{dt} + j_a(t) = \frac{n_d\,e^2}{m^*}\tau_a(T)\,\tau_f^{\mu}\,\frac{d^{\mu}E(t)}{dt^{\mu}}$$

anomalous component

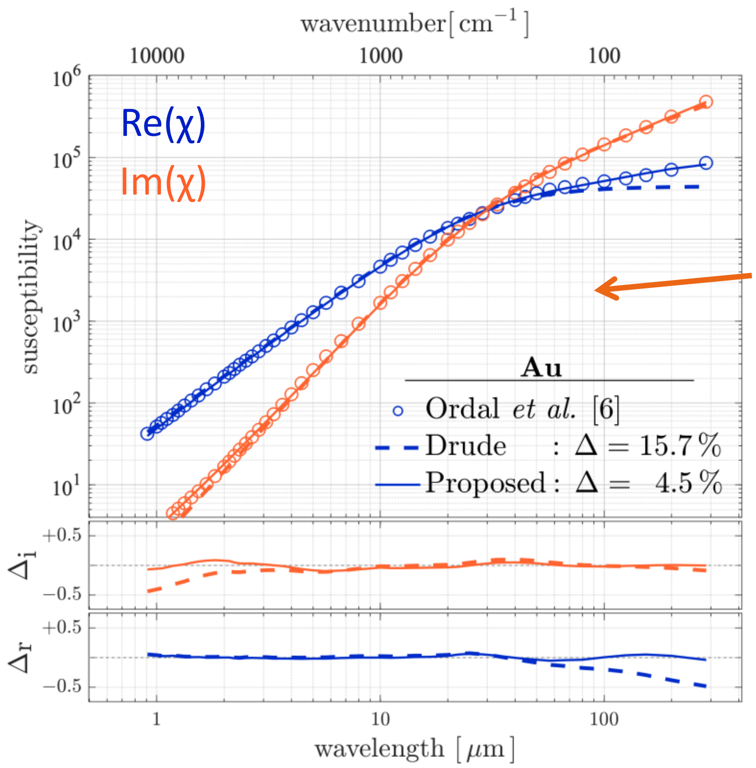
- total current density is the sum of the Drude component and the anomalous component, with the implied physical constraints:

$$\frac{\sigma_a(T)}{\lambda_a(T)} = \frac{\sigma_\gamma(T)}{\lambda_\gamma(T)} = \mathrm{constant}$$

- note: characteristic wavelengths are computed from time constants $\lambda(T) = 2\pi c_0/\tau(T)$

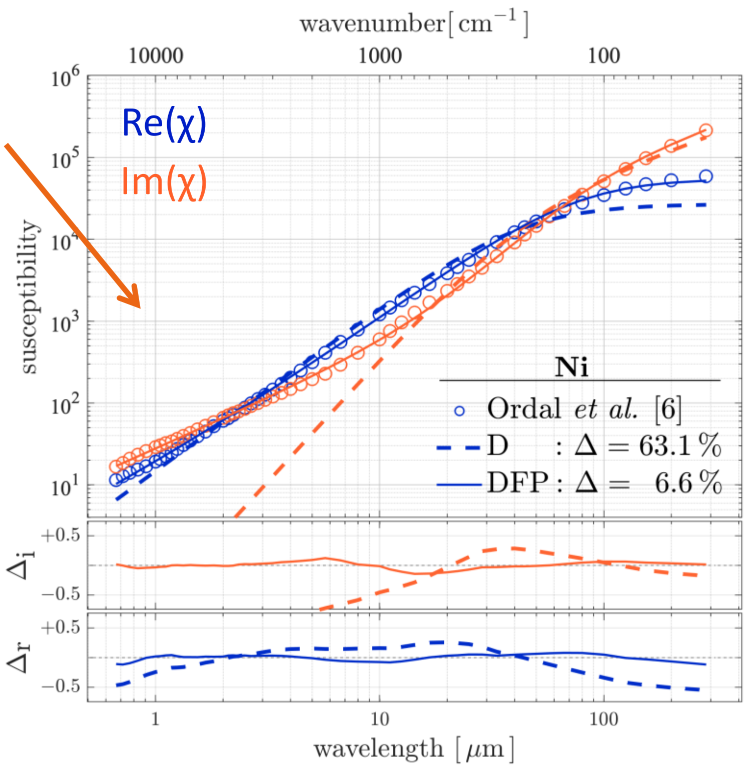
Fidelity of the proposed model

Applying the proposed model to a given set of data (*i.e.*, at a single temperature) leads to an improvement over the Drude model.



Au: [Xe] $4f^{14}$ $5d^{10}$ $6s^1$

Ni: [Ar] $3d^8$ $4s^2$



Temperature-dependent photonic conductivity

- temperature-dependent resistivity (inverse conductivity) is predicted, *e.g.*, by the solution to the Boltzmann equation in the relaxation time approximation [5]:

$$\rho_\gamma(T) = \rho_r + \rho_T(T) \propto \tau_c(T)^{-1}$$

- temperature-independent residual resistivity ρ_r is due to concomitant lattice imperfections (lattice errors, impurities, etc.)
- temperature-dependent resistivity can be captured over a limited temperature range with the power law

$$\rho_T(T) = \rho_{\mathrm{ref}} \left(T/\,T_{\mathrm{ref}}\right)^k$$

Procedural regression of parameters from experimental data

- 7 primary data sets were used: {4, 305, 583, 722, 1083, 1238, 1403} K [8-10]
- 1 auxiliary data set 294 K [6]
- parameter regression
  1. temperature-independent parameters fixed with low-temp data
  2. resulting structure regressed at each temperature
  3. functional relation for $\rho_\gamma(T)$ obtained from these values
- result: temperature- and wavelength-dependent complex-valued susceptibility model

[8] D. W. Lynch *et al.*, "Infrared and visible optical properties of single crystal Ni at 4K," *Solid State Commun.*, Vol. 9, No. 24, 1971, pp.2195–2199.

[9] D. K. Edwards, N. Bayard De Volo, "Useful Approximations for the Spectral and Total Emissivity of Smooth Bare Metals," *Advances in Thermophysical Properties at Extreme Temperatures and Pressures*, edited by S. Gratch, ASME, New York, 1965.

[10] G. W. Autio, E. Scala, "The normal spectral emissivity of isotropic and anisotropic materials," *Carbon*, Vol. 4, No. 1, 1966, pp.13–28.

Finalizing the model

- it was found that

$$\begin{aligned} \alpha(T)\,\tau_c(T) &\approx \text{constant} \\ \Rightarrow \lambda_a &\approx \text{constant} \\ \Rightarrow \sigma_a &\approx \text{constant} \end{aligned}$$

- leading to a free-carrier susceptibility model having the form

$$\chi_\phi(\lambda, T) = \chi_d(\lambda, T) + \chi_a(\lambda)$$

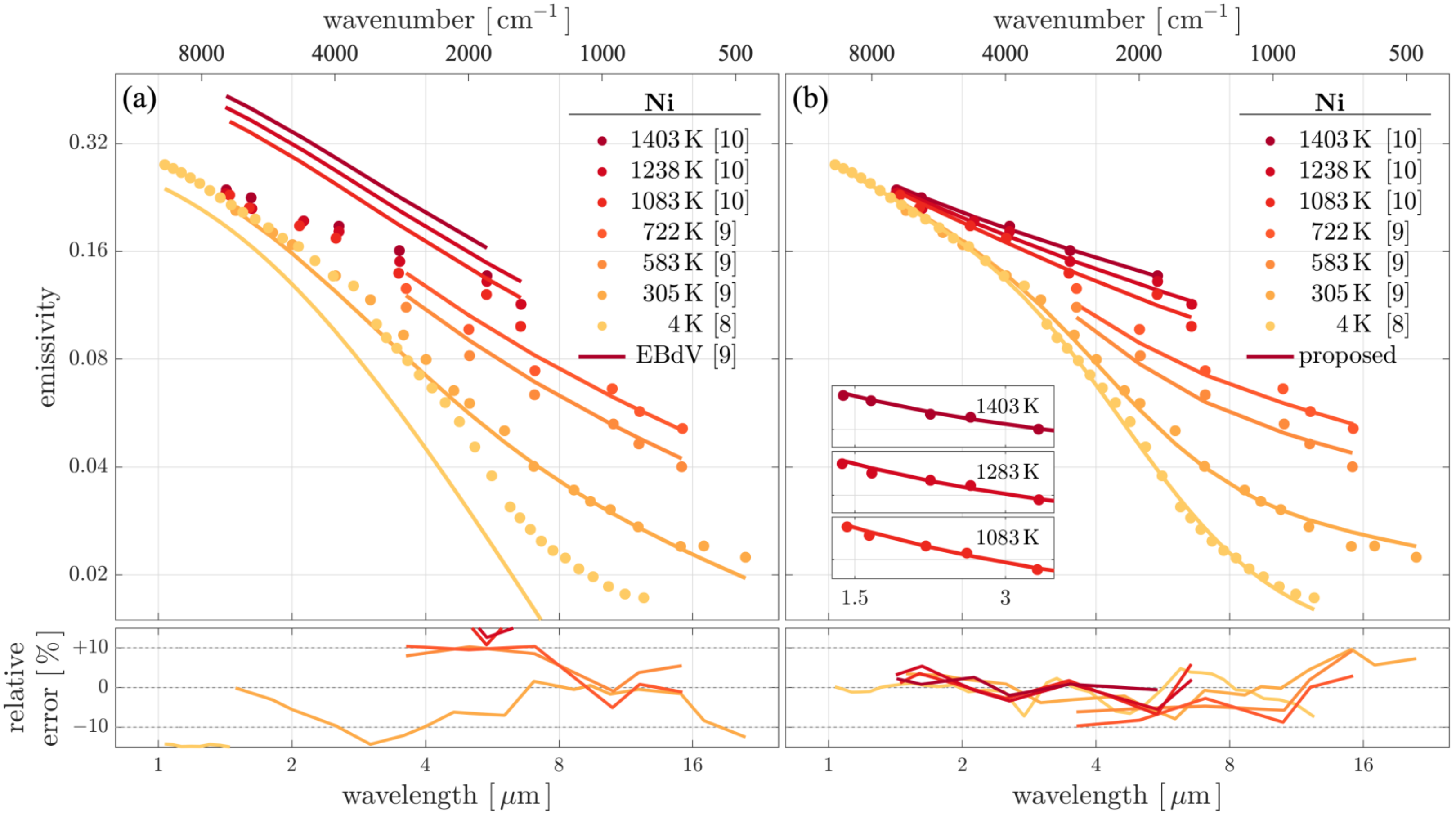
temperature-dependent
Drude component

temperature-independent
anomalous component

- the complex refractive index, complex Fresnel coefficients, and radiative parameters are then computed in the usual manner

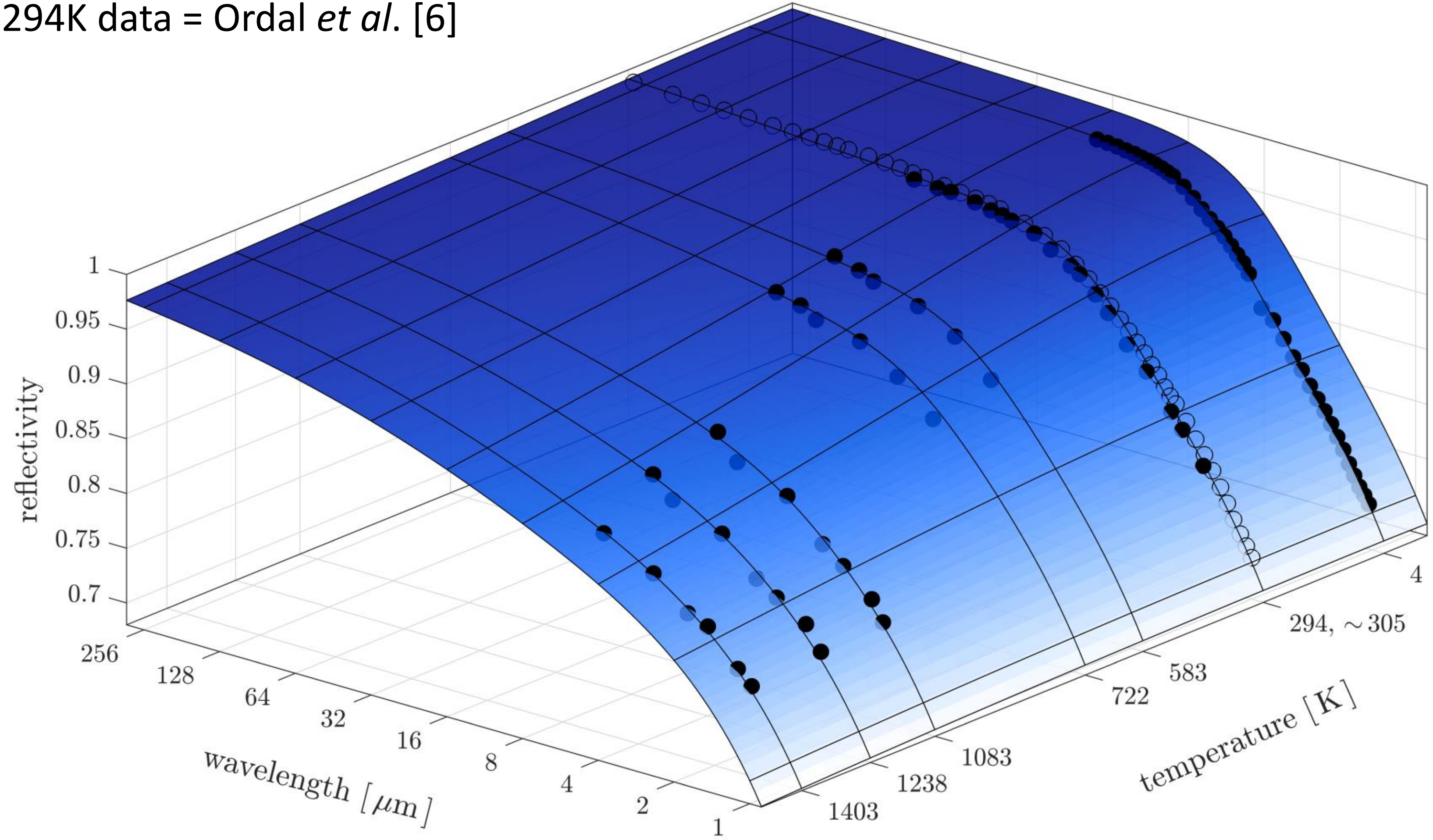
Comparative results

- EBdV = Edwards and Bayard De Volo model [9]



Prediction of auxiliary data

- 294K data = Ordal *et al*. [6]



Concluding remarks

- power law conductivity fit in relative agreement with the literature (ρ_{ref} = 92 nohm, k = 1.60)

Nickel, Ni	69	1.64

[11]

- model is realized in complex susceptibility, so we have access to
  1. temp.-dep. permittivity
  2. temp.-dep. refractive index
  3. temp.-dep. complex Fresnel coefficients
- can be used in more general optical and radiative frameworks (*e.g.,* non-smooth media [12])

[11] S. Kasap, P. Capper, editors, *Springer Handbook of Electronic and Photonic Materials*, Springer, New York, 2006.
[12] K. Tang, R. O. Buckius, "A statistical model of wave scattering from random rough surfaces," *Int. J. Heat Mass Tran.*, Vol. 44, No. 21, 2001, pp.4059–4073.

References

[1] H. Kocer, *et al.*, “Reduced near-infrared absorption using ultra-thin lossy metals in Fabry-Perot cavities,” *Sci. Rep.*, Vol. 5, 2015, p. 8157.

[2] G. Teodorescu *et al.*, “Normal emissivity of high-purity nickel at temperatures between 1440 and 1605 K,” *J. Phys. Chem. Solids*, Vol. 69, No. 1, 2008, pp. 133–138.

[3] K.-H. Weng, C.-D. Wen, “Effect of oxidation on aluminum alloys temperature prediction using multispectral radiation thermometry,” *Int. J. Heat Mass Tran.*, Vol. 54, No. 23, 2011, pp. 4834–4843.

[4] F. Wooten, *Optical Properties of Solids*, Academic Press, New York, 1972.

[5] N. Pottier, *Nonequilibrium Statistical Physics: Linear Irreversible Processes*, Oxford graduate texts, Oxford University Press, Oxford, 2010.

[6] M. A. Ordal *et al.*, “Optical properties of Au, Ni, and Pb at submillimeter wavelengths,” *Appl. Opt.*, Vol. 26, No. 4, 1987, pp.744–752.

[7] J. Orosco, C. F. M. Coimbra, “Anomalous carrier transport model for broadband infrared absorption in metals,” *Phys. Rev. B*, Vol. 98, No. 23, 2018, 235118.

[8] D. W. Lynch *et al.*, “Infrared and visible optical properties of single crystal Ni at 4K,” *Solid State Commun.*, Vol. 9, No. 24, 1971, pp.2195–2199.

[9] D. K. Edwards, N. Bayard De Volo, “Useful Approximations for the Spectral and Total Emissivity of Smooth Bare Metals,” *Advances in Thermophysical Properties at Extreme Temperatures and Pressures*, edited by S. Gratch, ASME, New York, 1965.

[10] G. W. Autio, E. Scala, “The normal spectral emissivity of isotropic and anisotropic materials,” *Carbon*, Vol. 4, No. 1, 1966, pp.13–28.

[11] S. Kasap, P. Capper, editors, *Springer Handbook of Electronic and Photonic Materials*, Springer, New York, 2006.

[12] K. Tang, R. O. Buckius, “A statistical model of wave scattering from random rough surfaces,” *Int. J. Heat Mass Tran.*, Vol. 44, No. 21, 2001, pp.4059–4073.

Questions?