Observation of Quantum Corrections to Conductivity of Disordered MoC and NbN Films up to Optical Frequencies

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Superconducting hybrids at Extreme, Štrbské pleso
28. 6. 2021

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  - Parametric amplifiers
  - Superconducting quantum bits
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  - Electron concentration n
- Need for a robust characterization method of the electric properties of disordered films in the metallic state

Optical conductivity of metals

- Drude model:

$$\sigma(\omega) = \sigma'(\omega) + i\sigma''(\omega) = \frac{\sigma_0}{1 - i\omega/\Gamma}$$

- $\sigma_0 = ne^2/m\Gamma$ is the DC conductivity, and $\Gamma = 1/\tau$

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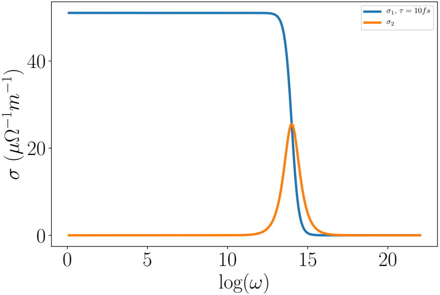

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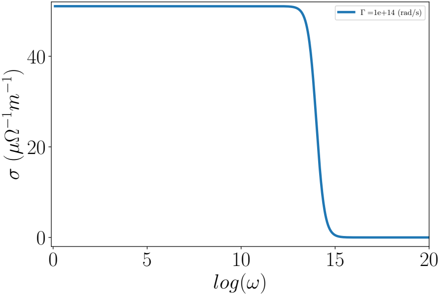

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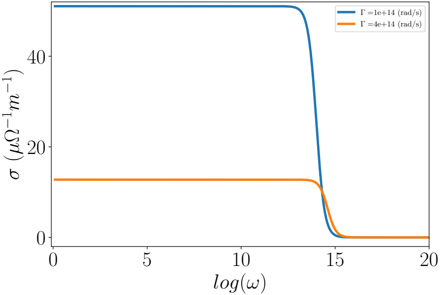

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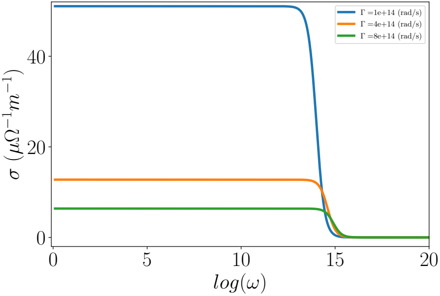

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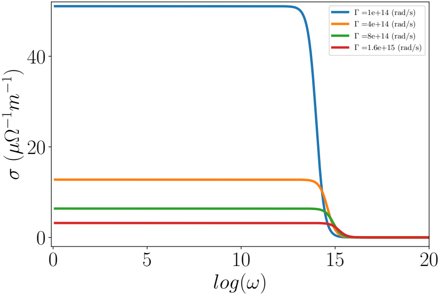

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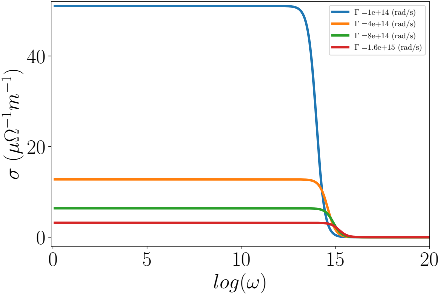

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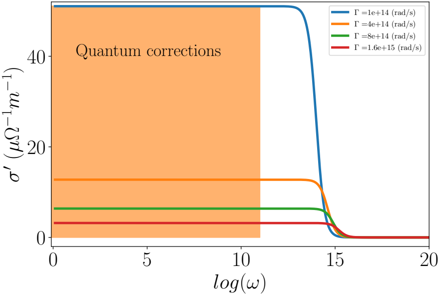

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- Normal conductivity:

$$\sigma_{\text{reg}}(\omega) \approx \sigma_0$$

- Quantum correction $\delta\sigma(\omega)$:
  - Weak-localization
  - Interaction effects
- The same functional form
  - 3D: $\delta\sigma(\omega) \sim \sqrt{\omega}$
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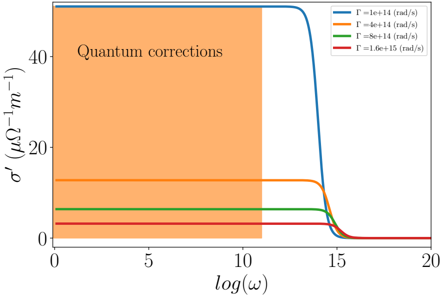

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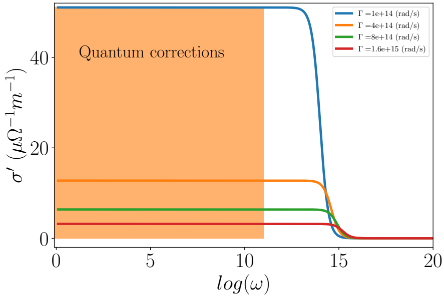

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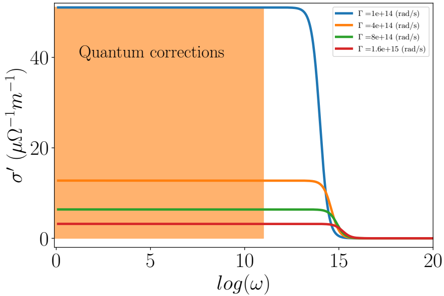

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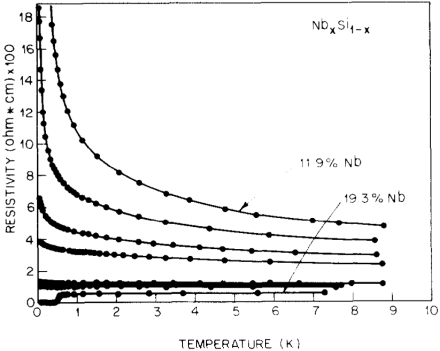
- Generally considered low-temperature phenomena
- Theoretically should be present up to $\omega^* \sim \Gamma$

Transport measurements at low temperatures

- Amorphous Nb_xSi_{1-x}, stoichiometry x - tuning of disorder
- Resistivity measuremets from 20mK to 9K:

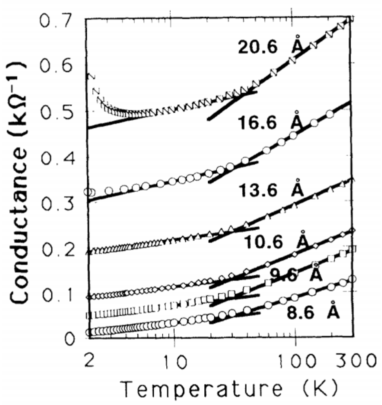
$$\sigma(T) = \sigma_0 + b\sqrt{T}$$



D.J. Bishop, E. G. Spencer, and R. C. Dynes, Solid State Electron. 38, 73 (1985)

Transport measurements of MoC

- MoC, thickness - tuned disorder
- 2D weak-localization and e-e interaction effects up to RT



S.

J. Lee et al., PRB **49**, 13882 (1994)

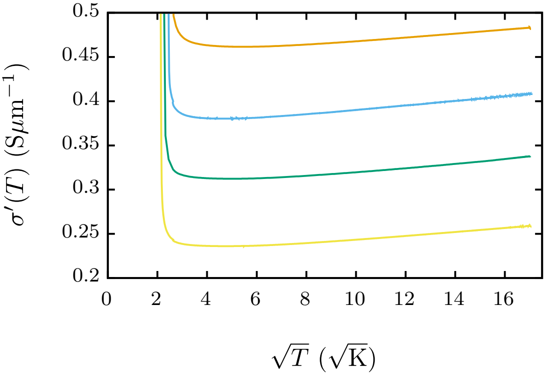
$$\sigma(T) = \sigma_0 + \{\alpha p + (1 - F)\} \ln(T)$$

MoC sample preparation

- Magnetron sputtered in argon-acetylene atmosphere
- Mo - target and sapphire substrates
- Mo_xC_{1-x}; stoichiometry (x) or thickness (t) tuned disorder
- Two sets of thin film samples:
  - 1. set: $R_\square$ from 420 to 720 Ω; fixed t=5 nm, varied x
  - 2. set: $R_\square$ from 100 to 220 Ω; varied t, fixed x

Transport measurements

- 1. set: $R_\square = 420, 500, 590$ a $720\ \Omega$



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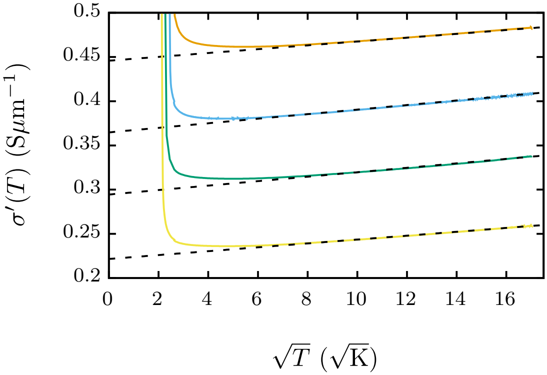

- $\sigma = \sigma_0 + b\sqrt{T}$; 3D quantum corrections up to T= 300K

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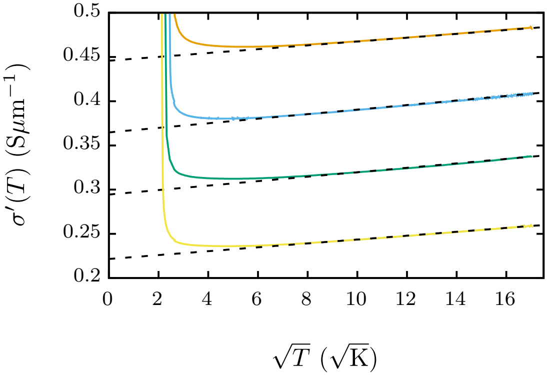

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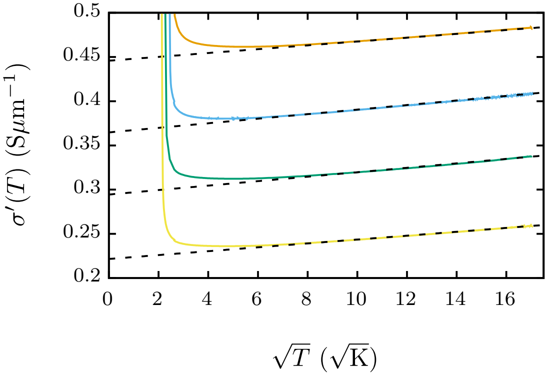

- $\sigma = \sigma_0 + b\sqrt{T}$; 3D quantum corrections up to T= 300K
- How far do they extend? The temperature range is limited.

Optical measurements

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$$\mathcal{T}(\omega) \approx \frac{\mathcal{T}_s(\omega)^2}{[1 + g'(\omega)/(n_s+1)]^2 + [g''(\omega)/(n_s+1)]^2}$$

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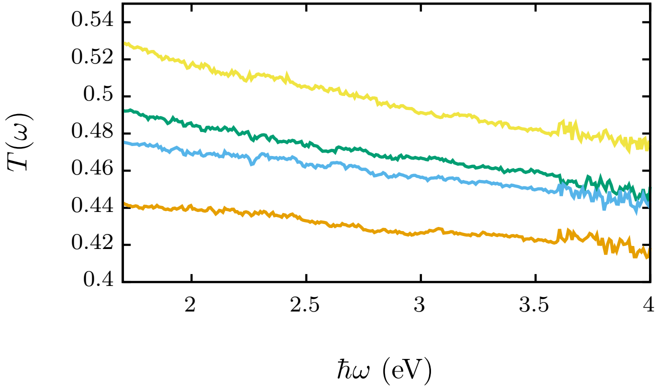
- **Spectroscopic ellipsometry**

- Reflection measurement, evaluation of σ_1 and σ_2
- Our experimental frequency range 300-810 THz

Optical transmission spectra

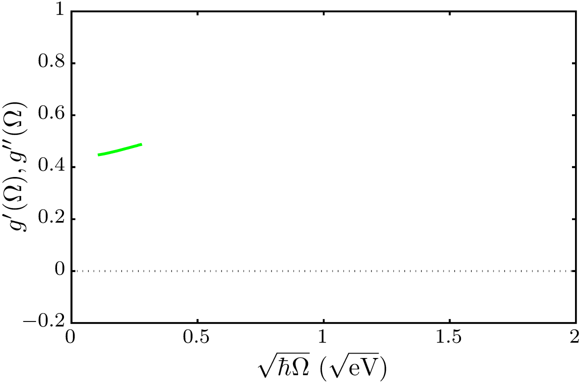
- 1. set of MoC: $R_\square$ = 420, 500, 590 a 720 Ω
- Smooth transmission spectra



- $T(\omega)$ decreases with frequency ω, the opposite is expected from the Drude model

Optical conductivity of MoC with $R_{\square}$=720Ω

- For interaction effects: $\Omega = \sqrt{\omega^2 + (\pi k_B T/\hbar)^2}$



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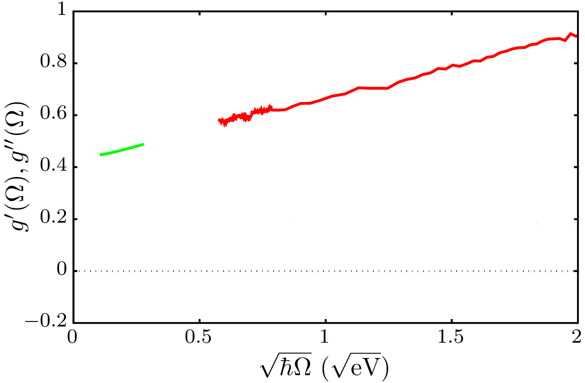

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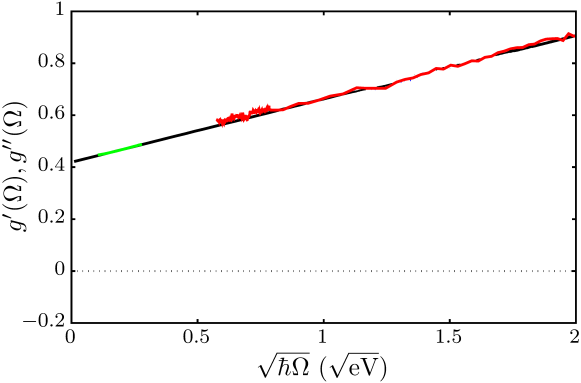

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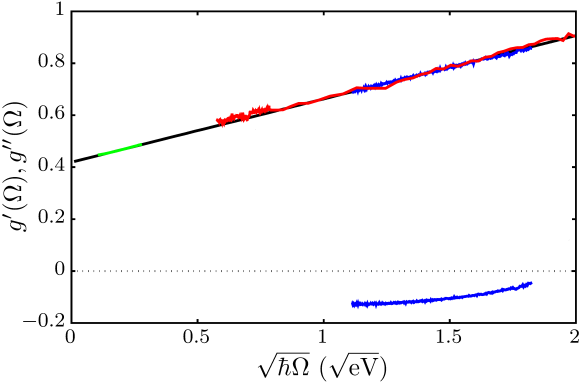


- Agreement between 3 data sets, $\hbar\Omega \approx 14$ meV up to 4 eV

Proposed model

- Postulate for the σ:

$$\sigma'(\omega, T) = \sigma_0 \left[1 - \mathcal{Q}^2 + \mathcal{Q}^2\sqrt{\Omega/\Gamma}\right] \quad \text{if } \Omega < \omega^*$$
$$\sigma'(\omega, T) = \frac{\sigma_0}{1 + (\Omega/\Gamma)^2} \quad \text{if } \Omega \geq \omega^*$$

- 3 parameters:
  1. $\mathcal{Q}$ - "Quantumness"
  2. Γ - Scattering rate
  3. $\sigma_0 = ne^2/m\Gamma$, whereas $\sigma(0,0) = \sigma_0(1 - \mathcal{Q}^2)$
- Fermi liquid theory, for interaction effects:

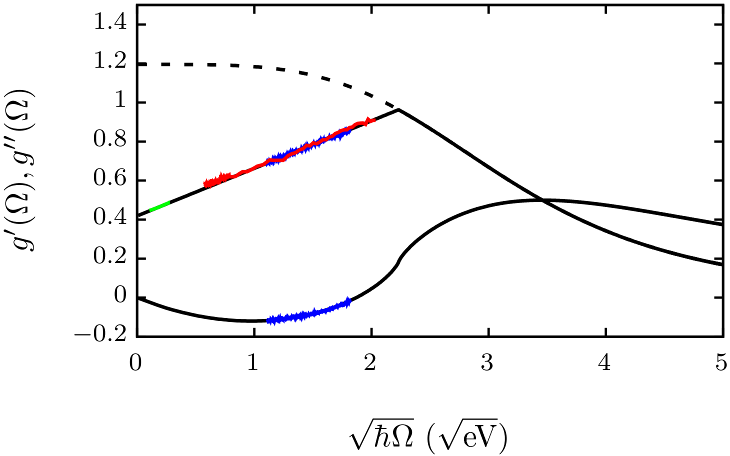
$$\Omega = \sqrt{\omega^2 + (\pi k_B T/\hbar)^2}$$

- Imaginary part σ'' given by Kramers-Kronig relations

- Contribution of bound electrons to permittivity

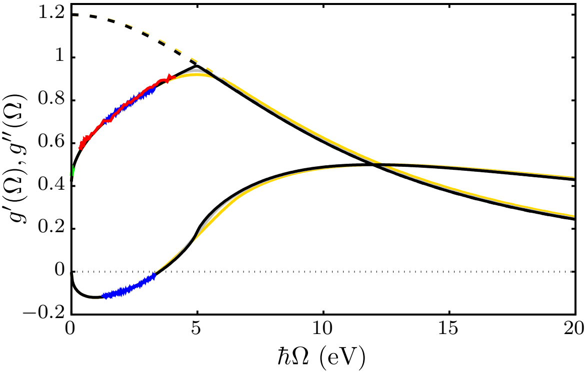
$$\sigma''_{\text{bound}}(\omega) = -\epsilon_0(\epsilon_\infty - 1)\omega, \; \epsilon_\infty = 1.4$$

Conductivity $\sigma(\omega)$ fit



- Observation of quantum corrections to conductivity up to optical frequencies, Phys. Rev. B 100, 241106(R) (2019)

- Smoothing cups at $\omega = \omega^*$- "tailoring"with 3^{rd}-order polynomial $f(\omega)$ for $\omega_- < \omega < \omega_+$, for different ω_-,ω_+



Extracted parameters of MoC with $R_\square = 720\Omega$

- Lorentz vs. Gaussian

	Drude		Gaussian	
ϵ_∞	1.0	1.4	1.0	1.4
$g_0 = Z_0 \sigma_0 d$	1.25	1.20	1.32	1.26
$\mathcal{Q}^2$	0.66	0.65	0.68	0.67
$\hbar\Gamma$ (eV)	11.5	10.1	13.6	12.0
n (10^{23} cm^{-3})	4.1	3.5	5.1	4.3

- Smoothened Lorentzian

$\hbar\omega_-$ (eV)	3.3	4.1	5.0
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- Scatter in σ_0, $\mathcal{Q}$, and $\Gamma \leq 15$ %

  $\rightarrow$ Prolongation procedure is robust

Characteristic parameters of MoC with $R_\square$=720Ω

- $\mathcal{Q} \approx 0.82 \pm 0.01$ strong quantum corrections
  $\rightarrow \sigma'(0) = (1 - \mathcal{Q}^2)\sigma_0 \approx \sigma_0/3$
  $\rightarrow$ 3x reduced classical value

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- Normalized conductivity $g_0 \approx 1.26 \pm 0.06$
  $\implies n = (4.3 \pm 0.8) \times 10^{23}$ cm^{-3}
  $\rightarrow$ surprisingly large electron concentration

Characteristic parameters of MoC with $R_\square$=720Ω

- Large $\Gamma \implies$ Electronic bands separated by $\leq \hbar\Gamma$ merge
  - Concentration of Mo $n_{\mathrm{at}} = 5.1 \times 10^{22}$ cm^{-3}
  - Valence electron configuration within $\pm\hbar\Gamma$ from ε_F
    Mo: 4d^5 5s^1 a C: 2s^2 2p^2
  - Corresponding electron density
    $n =$**10**$\times n_{\mathrm{at}} = 5.1 \times 10^{23}$ cm^{-3}, in the range of the estimated $n = (4.3 \pm 0.8) \times 10^{23}$ cm^{-3}

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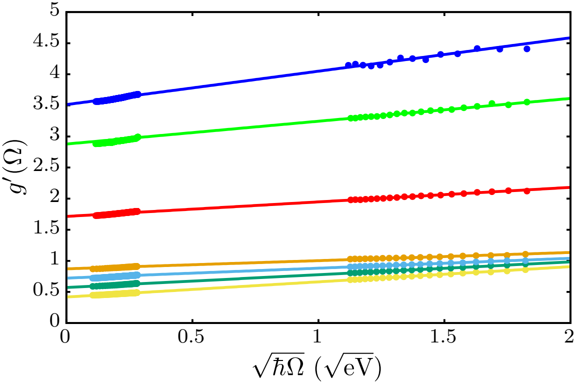
- Absence of interband transitions → Broad electronic band
- High Fermi energy $\epsilon_F = 20.65$, $k_F l \approx 3.5$
- Similarly, highly disordered NbN $n \approx 4.2 \times 10^{23}$ cm^{-3}
- Large n should be a generic property of dirty metals

Optical conductivity with the 2. set of MoC

- Broad range of $R_\square$ =100, 120, 220, 420, 500, 590 a 720 Ω



- Details of microstructure are not important
- The control parameter is the degree of disorder

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  - Randomly generated conductivity curves that obey the Kramers-Kronig relations and fit the experimental data to a chosen degree
  - Prolongation is given by the average of the selected curves

Prolongation of optical conductivity

- Experimental bandwidth is limited
- Fit depends on the chosen model - not trivial problem
- Model independent prolongation of optical conductivity
  - Reasonably smooth conductivity without rapid changes and oscillations in strongly disordered films
  - Randomly generated conductivity curves that obey the Kramers-Kronig relations and fit the experimental data to a chosen degree
  - Prolongation is given by the average of the selected curves
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- Numerical extrapolation method for complex conductivity of disordered metals, S. Kern et al., PRB **103**, 134205 (2021)

Prolongation of optical conductivity

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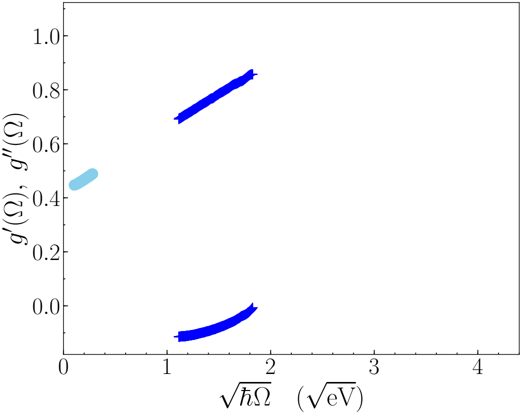

- S. Kern et al., Phys. Rev. B **103**, 134205 (2021)

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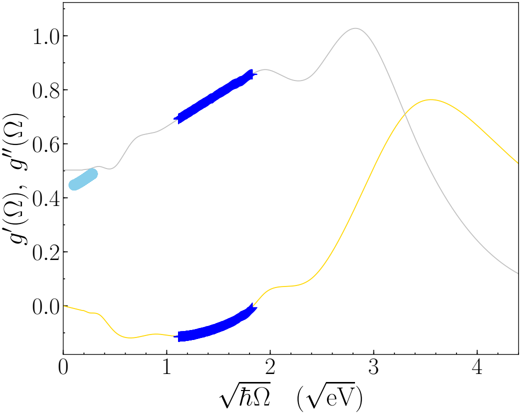

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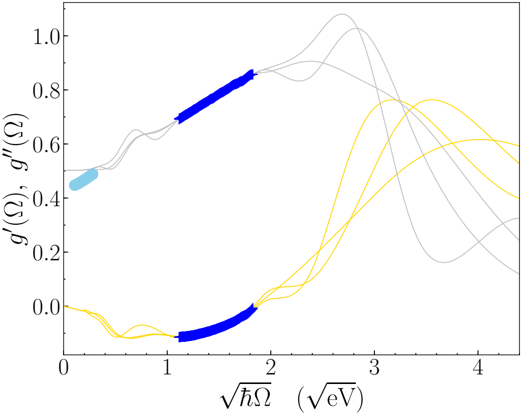

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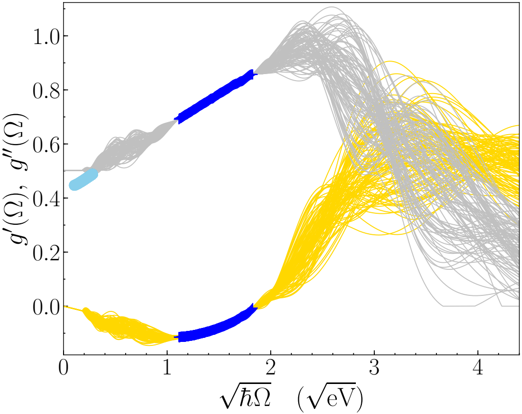

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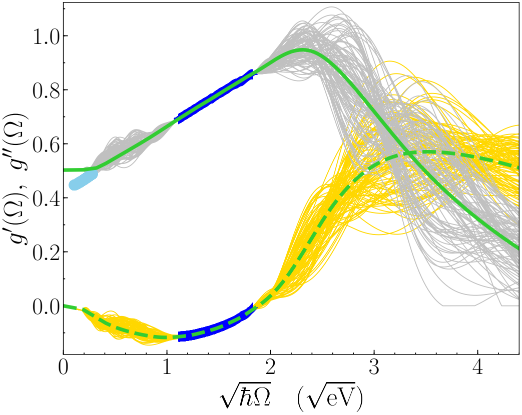

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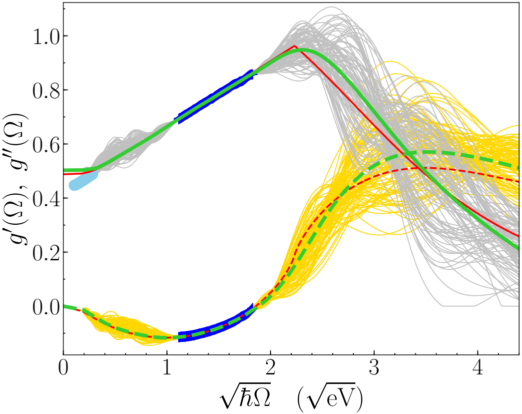

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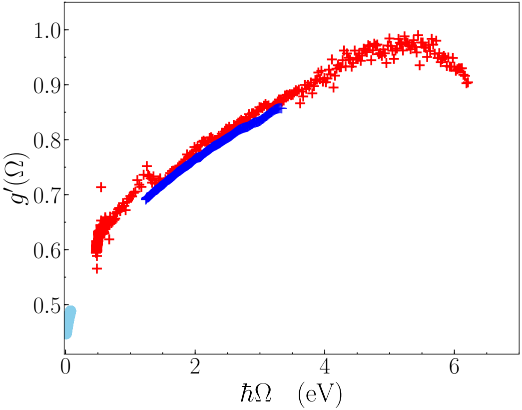
Optical transmission

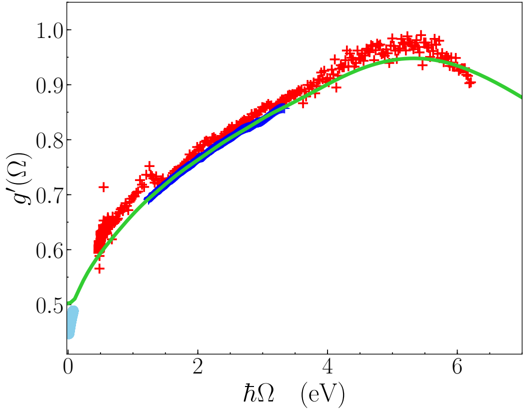
- 5 nm MoC with $R_\square$=720Ω



- Transmission spectra from 0.5 to 6 eV

Optical transmission

- 5 nm MoC with $R_\square = 720\Omega$



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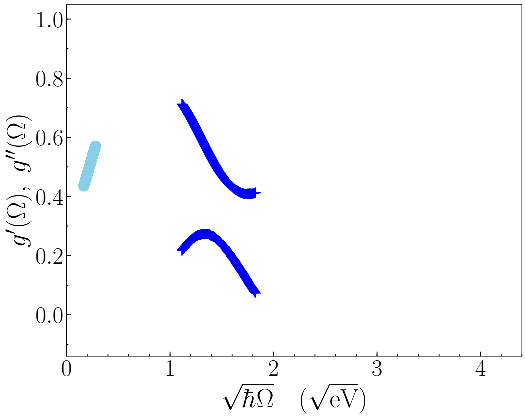
Optical conductivity of NbN

- Pulsed laser ablation; Nb-target, N_2 atmosphere
- Film thickness t=3.5 nm and $R_\square = 655\Omega$



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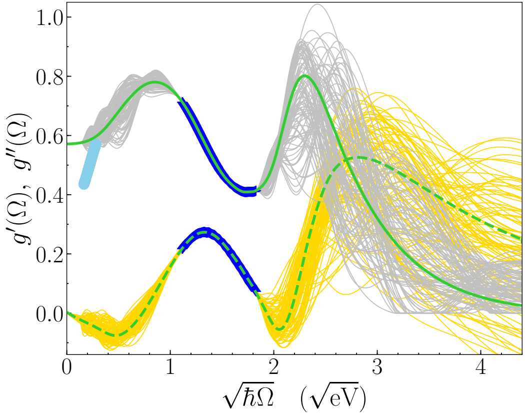

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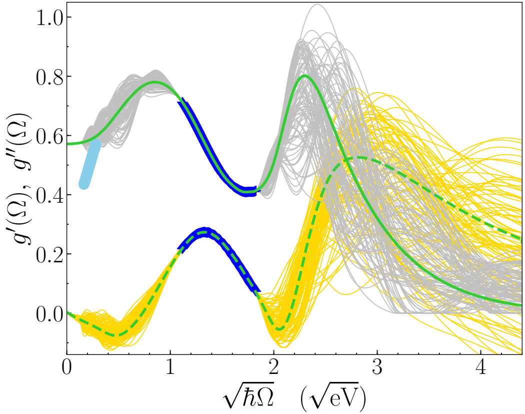

- Strong deviation from Drude-Lorentz model

Conclusion

- Observation of quantum corrections to conductivity up to optical frequencies in highly disordered films
- Method to extract σ_0, $\mathcal{Q}$ a Γ from the combined knowledge $\sigma'(\omega)$ and $\sigma''(\omega)$
- Estimation of the magnitude of the quantum correction $\delta\sigma' = -\mathcal{Q}^2\sigma_0$
- Numerical extrapolation method for complex conductivity of disordered metals

Thank you for your attention!

Neilinger et al., Phys. Rev. B **100**, 241106 (R) (2019)
S. Kern et al., Phys. Rev. B **103**, 134205 (2021)