

Thermal flux limited electron Kapitza conductance in copper-niobium multilayers

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This file contains details of the time-domain thermorefectance experiment, a list of the parameters used in the calculation of the EDMM, and the method for estimating the mean free path of electrons in the metallic multilayers.

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The thermal conductivities of the Cu-Nb multilayers were measured by time-domain thermoreflectance (TDTR) [1, 2, 3]. In our TDTR setup, an 80 MHz pulse train emanates from a Ti-Sapphire laser with pulse widths of about 100 fs and a central wavelength of 800 nm. The laser output is split by a polarizing beam splitter into separate pump and probe beams. The pump beam is modulated at 11.4 MHz frequency by an electro-optic modulator. The probe is delayed by a mechanical delay stage that gives a time delay of up to 5.5 ns. The pump and probe beams are coaxially focused on the sample down to $1/e^2$ radii of 25 μm and 6 μm , respectively. The probe beam is backreflected into a photodetector to monitor the change in the aluminum coating reflectance caused by the pump modulation. We use a lock-in amplifier to detect this change and relate it to the thermal properties of the Al film and materials underneath. We assume literature values for the heat capacity of the Al film and the heat capacity and thermal conductivity of the silicon substrate. We measure the electrical resistivity of the aluminum film by four-point probe and deduce the thermal conductivity from Wiedemann-Franz law. We treat the Cu-Nb multilayers as one layer and use a weighted average of the bulk heat capacity values of Cu and Nb. We assume that the interface resistance between the Cu-Nb multilayer sample and the silicon substrate is negligible in our analysis. Subsequently, the only unknowns in our thermal model are the Al/Cu-Nb Kapitza conductance and the thermal conductivity of the Cu-Nb film. These parameters are treated as free parameters and are varied to fit the data to a multilayer thermal model [1, 2, 3]. A total of five TDTR measurements were taken on each sample over the temperature range from 78 to 500 K in a cryostat with optical access that is kept under vacuum ($<10^{-6}$ Torr).

Table S1. Parameters used in the calculation of the EDMM

Metal	$D(\varepsilon_F) (\times 10^{47} \text{ m}^{-3})$	$\nu_F (\times 10^6 \text{ m s}^{-1})$
Cu	1.41 [4]	1.12 [5]
Nb	5.31 [6]	0.62 [7]
Ir	4.57 [8]	8.25 [8]
Pd	9.69 [9]	4.32 [10]
Al	1.26 [11]	1.33 [5]
Pt	7.05 [12]	0.33 [12]

The effective mean free path in the multilayers is given by Matthiessen's rule [13]:

$$\frac{1}{\lambda_{eff}} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2}. \quad (1)$$

where λ_1 and λ_2 are the electron mean free path in layers 1 and 2, respectively. λ inside each layer can be estimated by [14]:

$$\lambda = \frac{6\kappa}{\pi^2 k_B^2 D(\varepsilon_F) \nu_F T} \quad (2)$$

where κ is the metal thermal conductivity taken from literature [15].

References

- [1] D. G. Cahill, "Analysis of heat flow in layered structures for time-domain thermoreflectance," *Review of Scientific Instruments*, vol. 75, p. 5119, Nov. 2004.
- [2] A. J. Schmidt, X. Chen, and G. Chen, "Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance," *Review of scientific instruments*, vol. 79, p. 114902, Nov. 2008.

- [3] P. E. Hopkins, J. R. Serrano, L. M. Phinney, S. P. Kearney, T. W. Grasser, and C. T. Harris, "Criteria for cross-plane dominated thermal transport in multilayer thin film systems during modulated laser heating," *Journal of Heat Transfer*, vol. 132, no. 8, p. 081302, 2010.
- [4] F. L. Eckardt, H. and J. Noffke, "Self-consistent relativistic band structure of the noble metals," *J. Phys. F: Met. Phys.*, vol. 14, pp. 97–112, Jan 1984.
- [5] B. Gundrum, D. Cahill, and R. Averback, "Thermal conductance of metal-metal interfaces," *Physical Review B*, vol. 72, pp. 1–5, Dec. 2005.
- [6] A. R. Jani, N. E. Brener, and J. Callaway, "Band structure and related properties of bcc niobium," *Phys. Rev. B*, vol. 38, pp. 9425–9433, Nov 1988.
- [7] L. F. Mattheiss, "Electronic structure of niobium and tantalum," *Phys. Rev. B*, vol. 1, pp. 373–380, Jan 1970.
- [8] J. Noffke and L. Fritsche, "Band structure calculation and photoemission analysis of iridium," *J. Phys. F: Met. Phys.*, vol. 12, pp. 921–933, May 1982.
- [9] F. M. Mueller, A. J. Freeman, J. O. Dimmock, and A. M. Furdyna, "Electronic structure of palladium," *Phys. Rev. B*, vol. 1, pp. 4617–4635, Jun 1970.
- [10] R. Wilson and D. Cahill, "Experimental Validation of the Interfacial Form of the Wiedemann-Franz Law," *Physical Review Letters*, vol. 108, p. 255901, June 2012.
- [11] Z. Lin, L. V. Zhigilei, and V. Celli, "Electron-phonon coupling and electron heat capacity of metals under conditions of strong electron-phonon nonequilibrium," *Phys. Rev. B*, vol. 77, p. 075133, Feb 2008.
- [12] O. K. Andersen, "Electronic structure of the fcc transition metals ir, rh, pt, and pd," *Phys. Rev. B*, vol. 2, pp. 883–906, Aug 1970.
- [13] J. M. Ziman, *Electrons and Phonons: The Theory of Transport Phenomena in Solids*. Oxford University Press, 2001.
- [14] A. Belmiloudi, *Heat Transfer - Mathematical Modelling, Numerical Methods and Information Technology*. InTech, 2011.
- [15] C. Y. Ho, R. W. Powell, and P. E. Liley, "Thermal conductivity of the elements," *Journal of Physical and Chemical Reference Data*, vol. 1, no. 2, 1972.