Heat transfer across molecular junctions between nanoparticles

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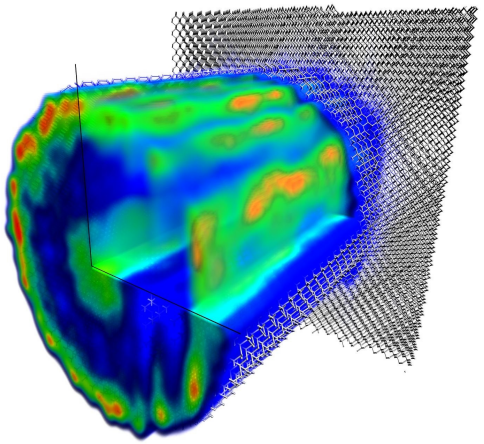
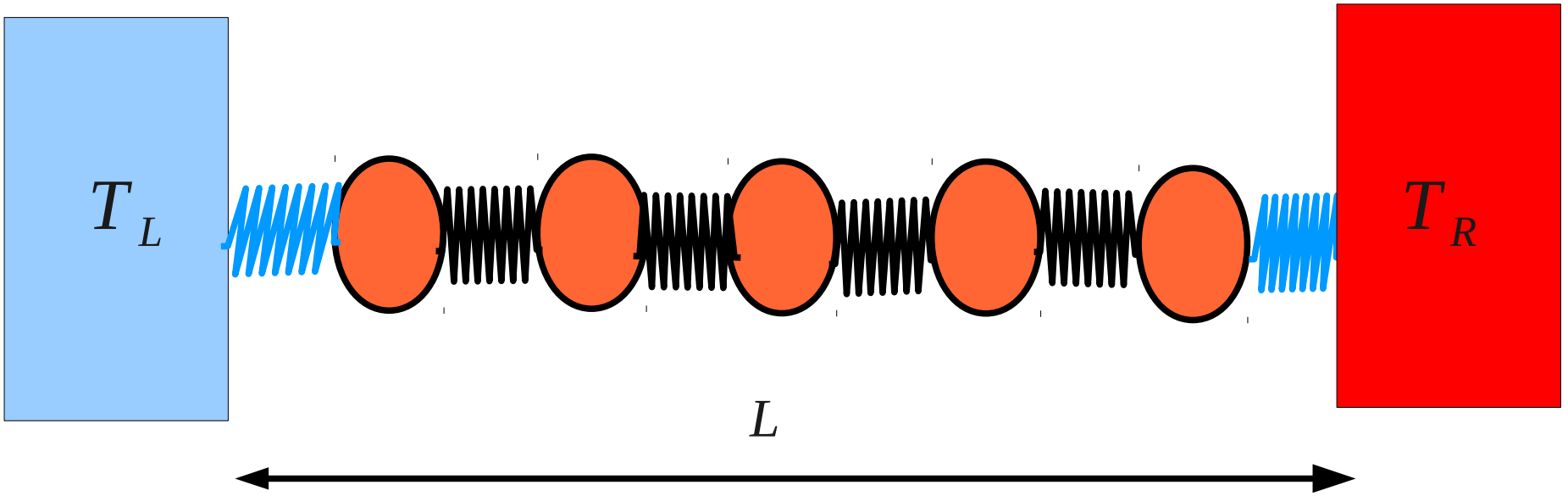


Workshop on « Charge and heat dynamics in nano-systems », Orsay October 2011

Heat Transfer in 1D objects



Thermal conductivity $\lambda(L)=J\,L/\Delta T$ Conductance : $\kappa(L)=\lambda(L)/L$

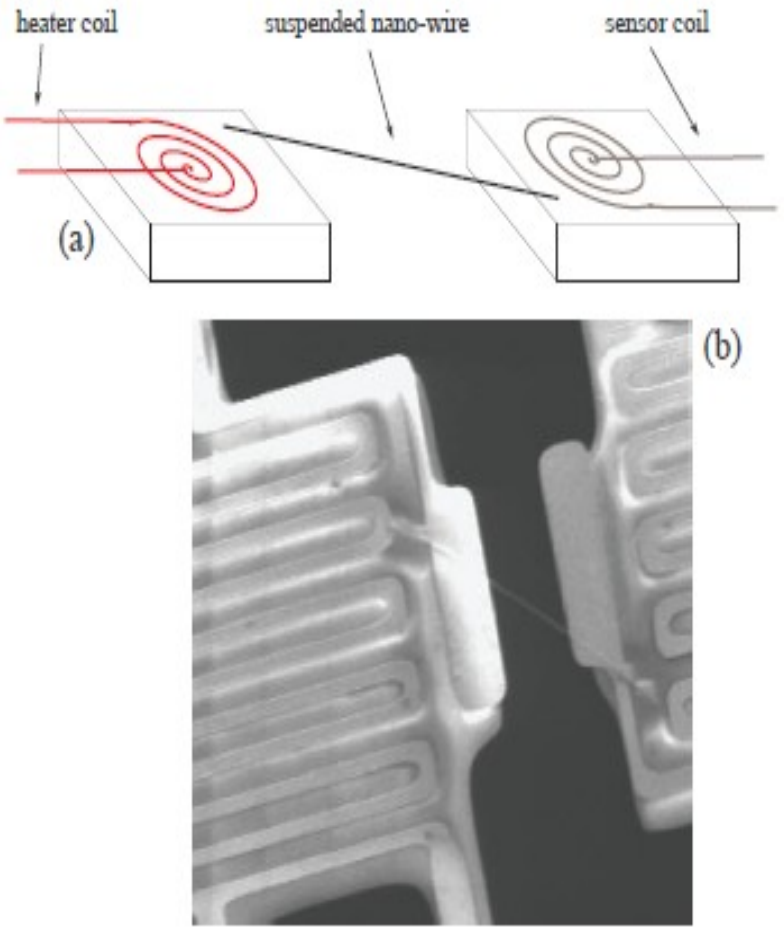
Fourier law : $\lambda\sim L^0$ Ballistic regime $\lambda\sim L$ $\kappa\sim L^0$

1D chain with anharmonic interactions : $\lambda(L)\sim L^{\alpha}$ $\alpha=1/3$
(momentum conserving systems)

Transition ballistic / diffusive transport: $\lambda(L)\sim L$ $L\ll\Lambda$

$\lambda(L)\sim L^{\alpha}$ $L\gg\Lambda$

Experiments : nanotubes



Dubi and Di Ventra, 2010
C.W. Chang Science 2006

$$\dot{Q} = \kappa \left(T_h - T_s \right)$$

Experiments on individual MWCNTs
(P. Kim et al., Phys. Rev. Lett. 2001)

Thermal conductivity

$$\lambda \simeq 3000\, W/mK$$

Phonon mean free path

$$\Lambda \simeq 500\, nm$$

Experiments on individual SWCNTs
(C. Yu et al., NanoLett. 2005,
E. Pop et al. NanoLett. 2006
C.W. Chang et al. Phys. Rev. Lett. 2008)

Large values of the thermal conductivity
Slow increase of the conductance with CNT length

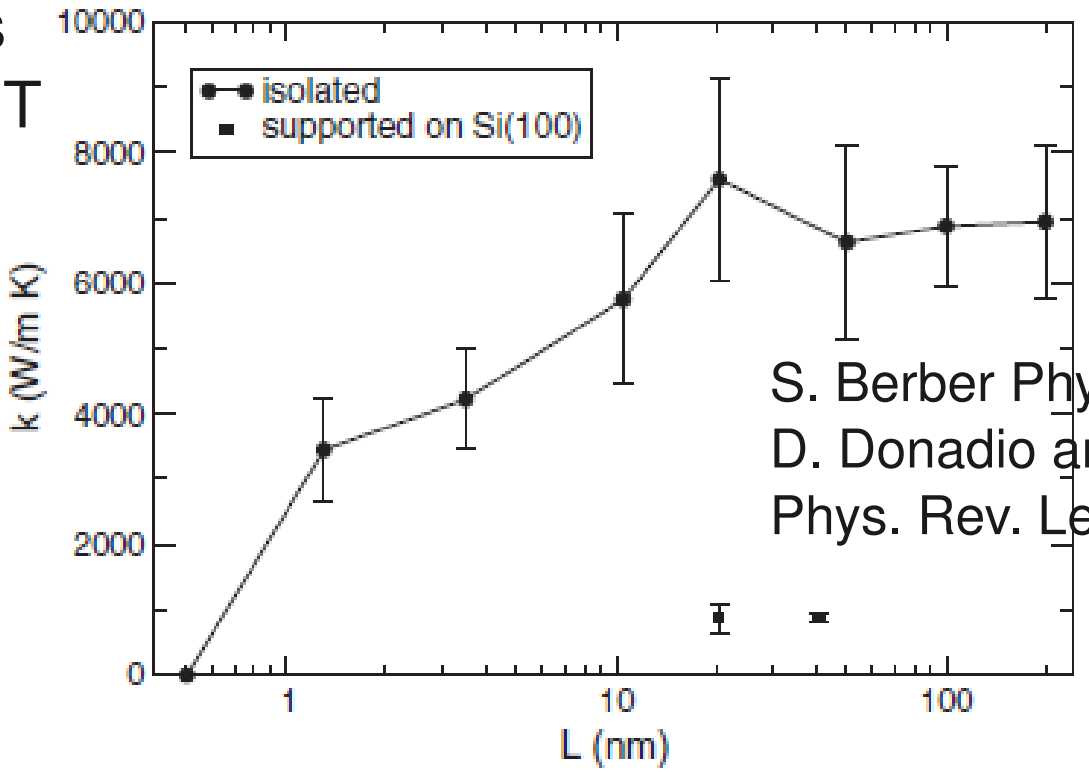
Molecular dynamics (MD) simulations

Equations of motion : $m\frac{d^2\vec{r}_i}{dt^2} = -\frac{\partial V}{\partial \vec{r}_i}$

Thermal conductivity : $\lambda = \frac{1}{V k_B T^2}\int_0^{+\infty} \langle \vec{J}(t)\cdot\vec{J}(0)\rangle dt$

$$\vec{J}(t) = \sum_i \frac{d(E_i \vec{r}_i)}{dt}$$

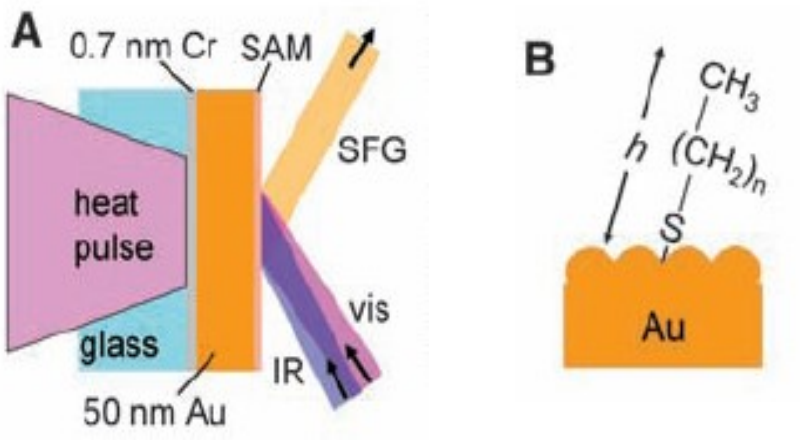
MD simulations
Isolated SWCNT



S. Berber Phys. Rev. Lett. 2000
D. Donadio and G. Galli,
Phys. Rev. Lett. 2007

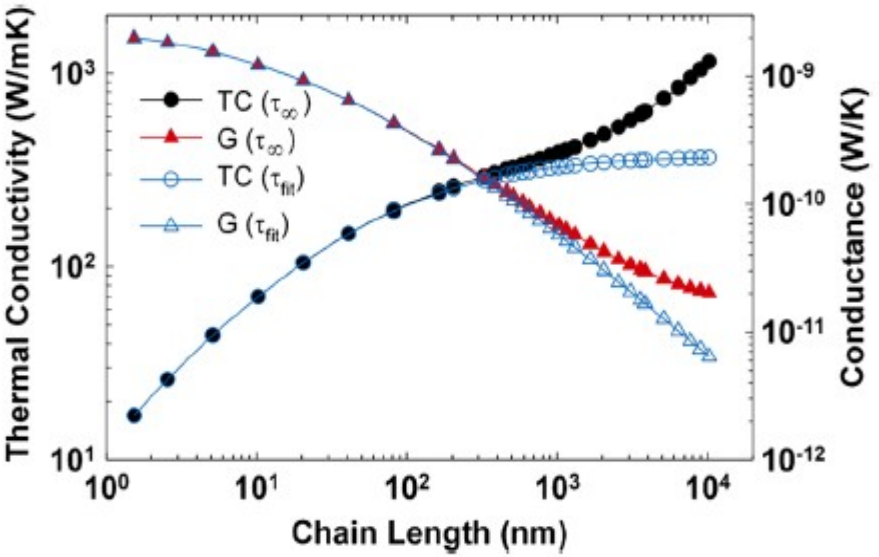
Heat transfer between nanoparticles: molecular junctions

Alkane-thiol chain



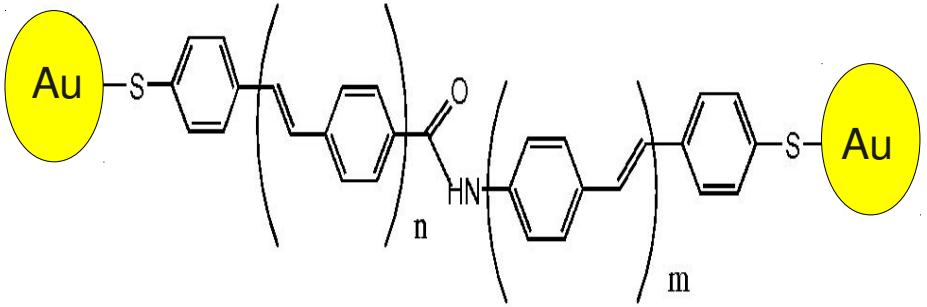
Z. Whang et *al.*, Science 2007

Polyethylene chain (simulations)

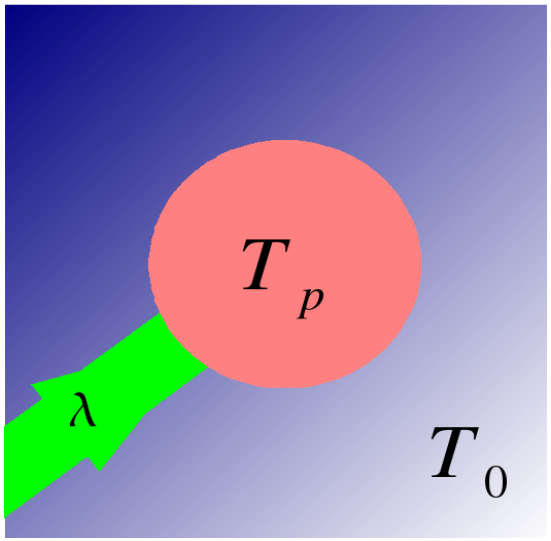


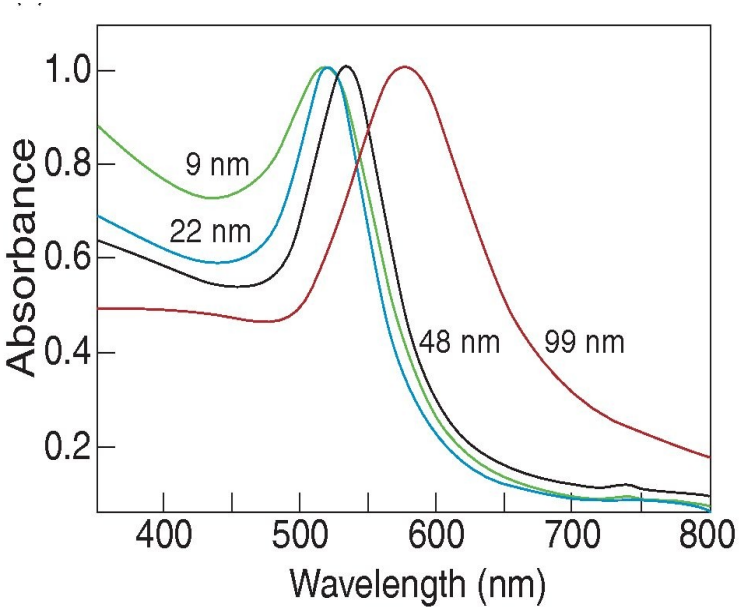
Henry & Chen, PRL 2008

Questions: thermal conductance, length dependance



Ultrafast spectroscopy



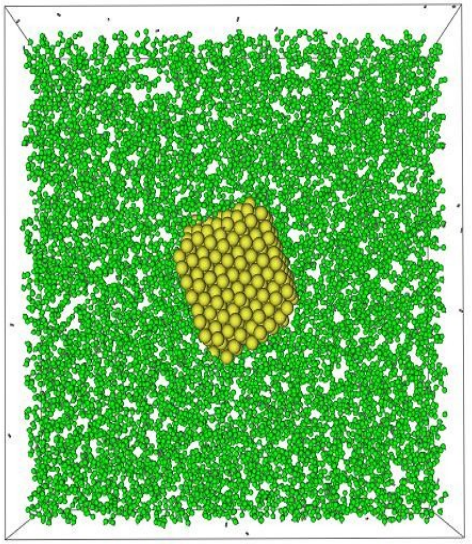


Selective strong heating of the nanoparticles (Hartmann, van Plessen, Kotaidis)

MD simulations heated gold nanoparticles in liquid octane

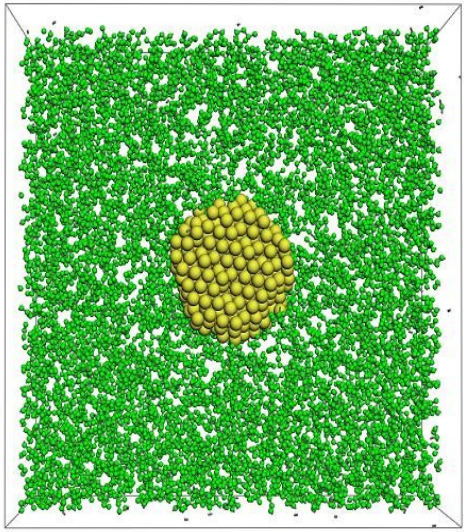
$300\,K$

$0\,nW$



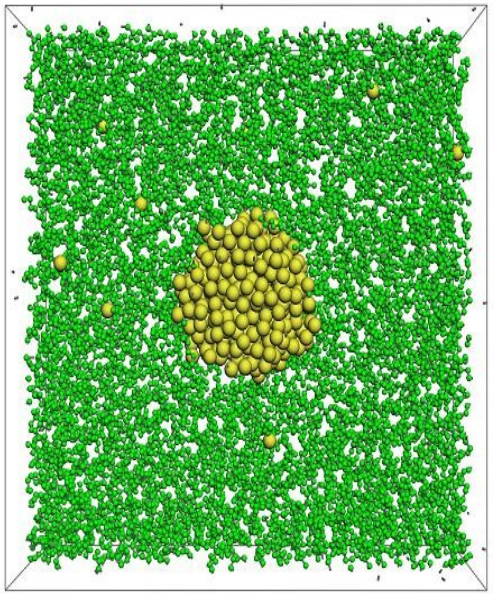
$800\,K$

$400\,nW$

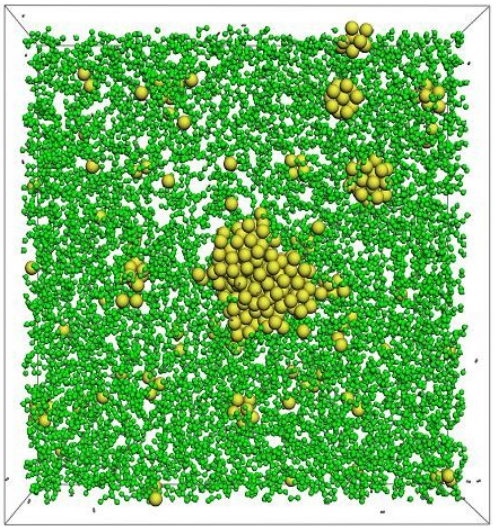


$1200\,K$

$600\,nW$

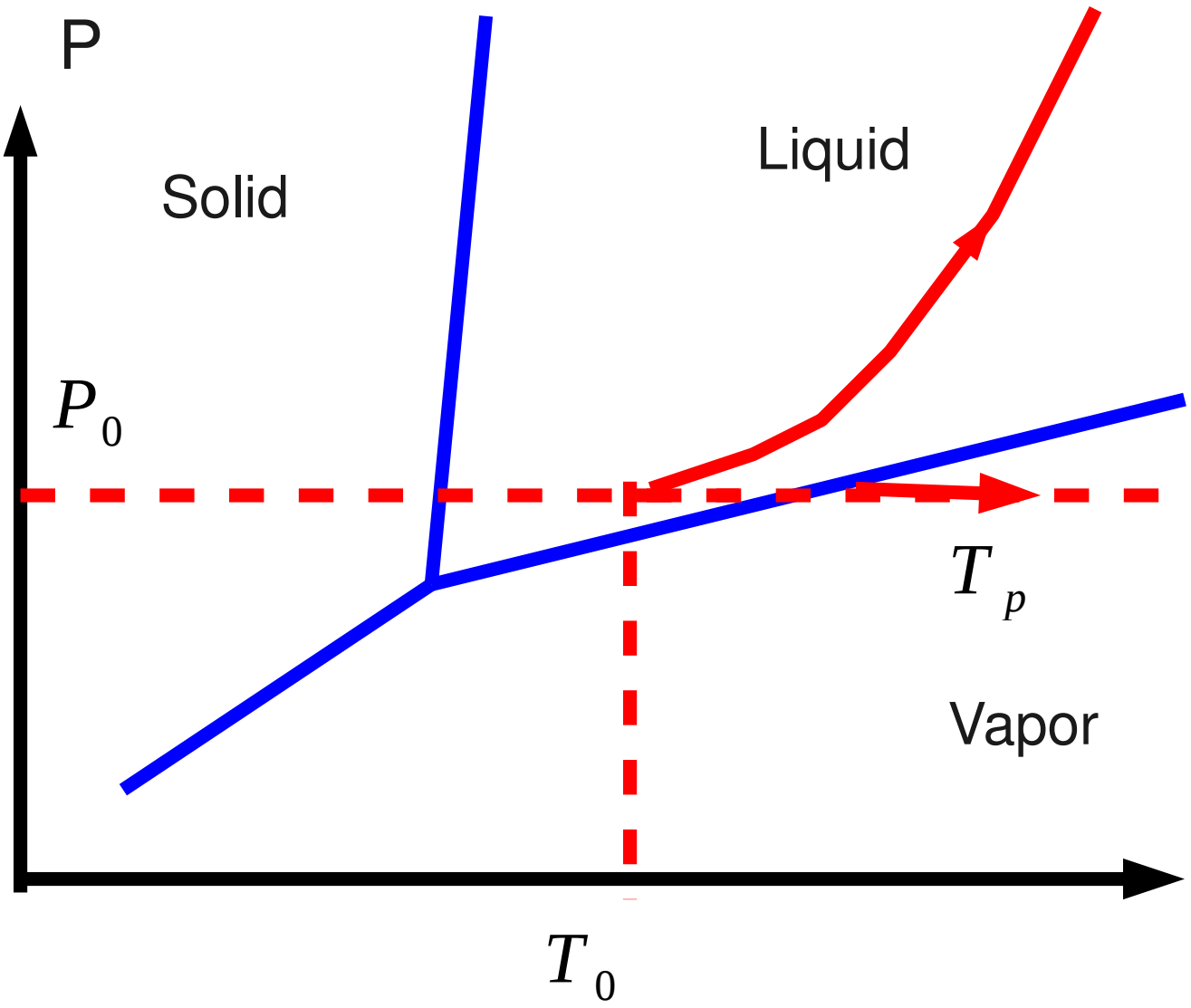


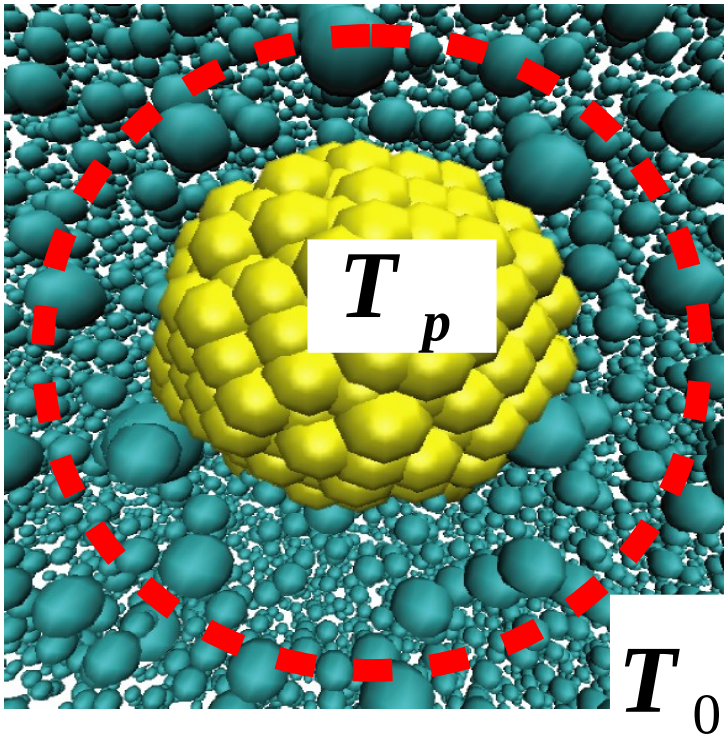
$1000\,nW$



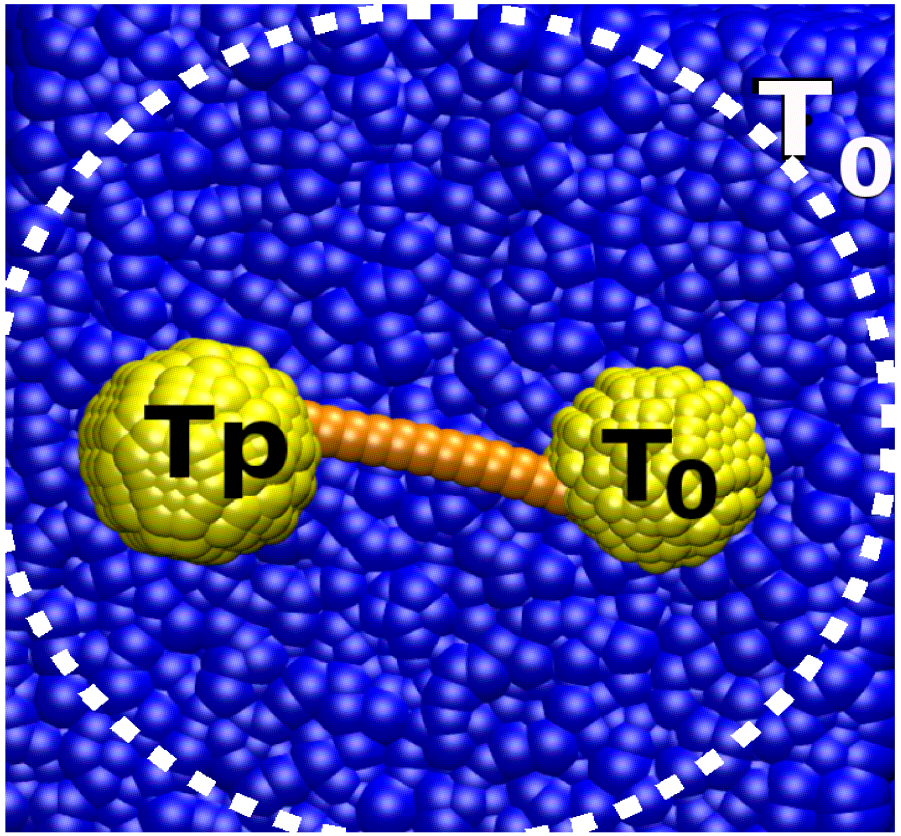
S. Merabia et al., PNAS 2009 Collaboration P. Keblinski N.Y.

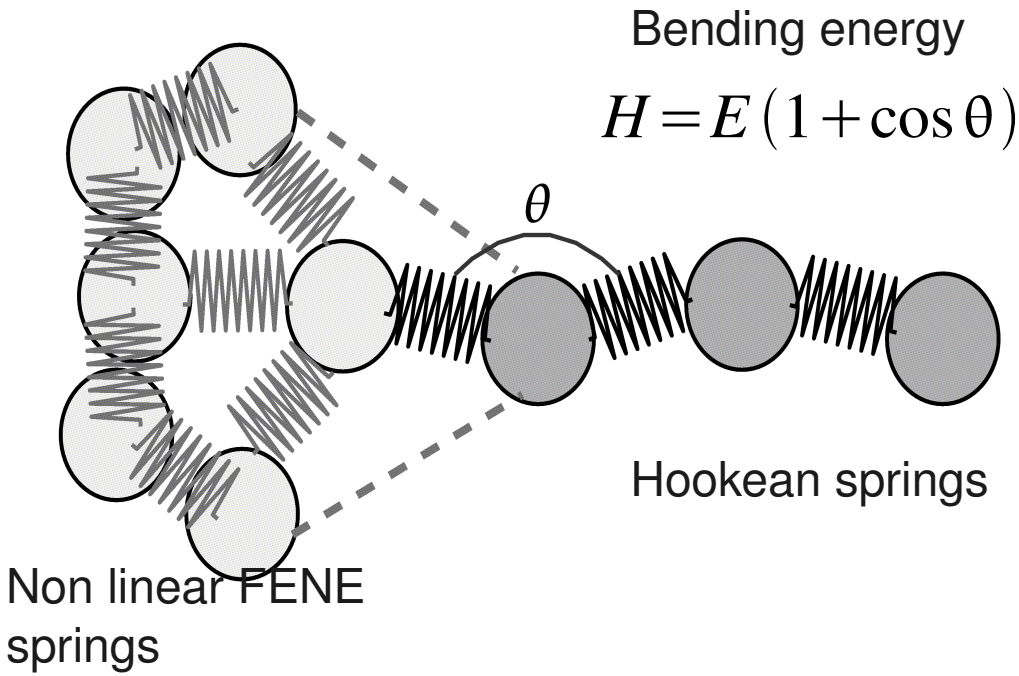
Heating up the nanoparticle





Heat transfer between nanoparticles: molecular junction model



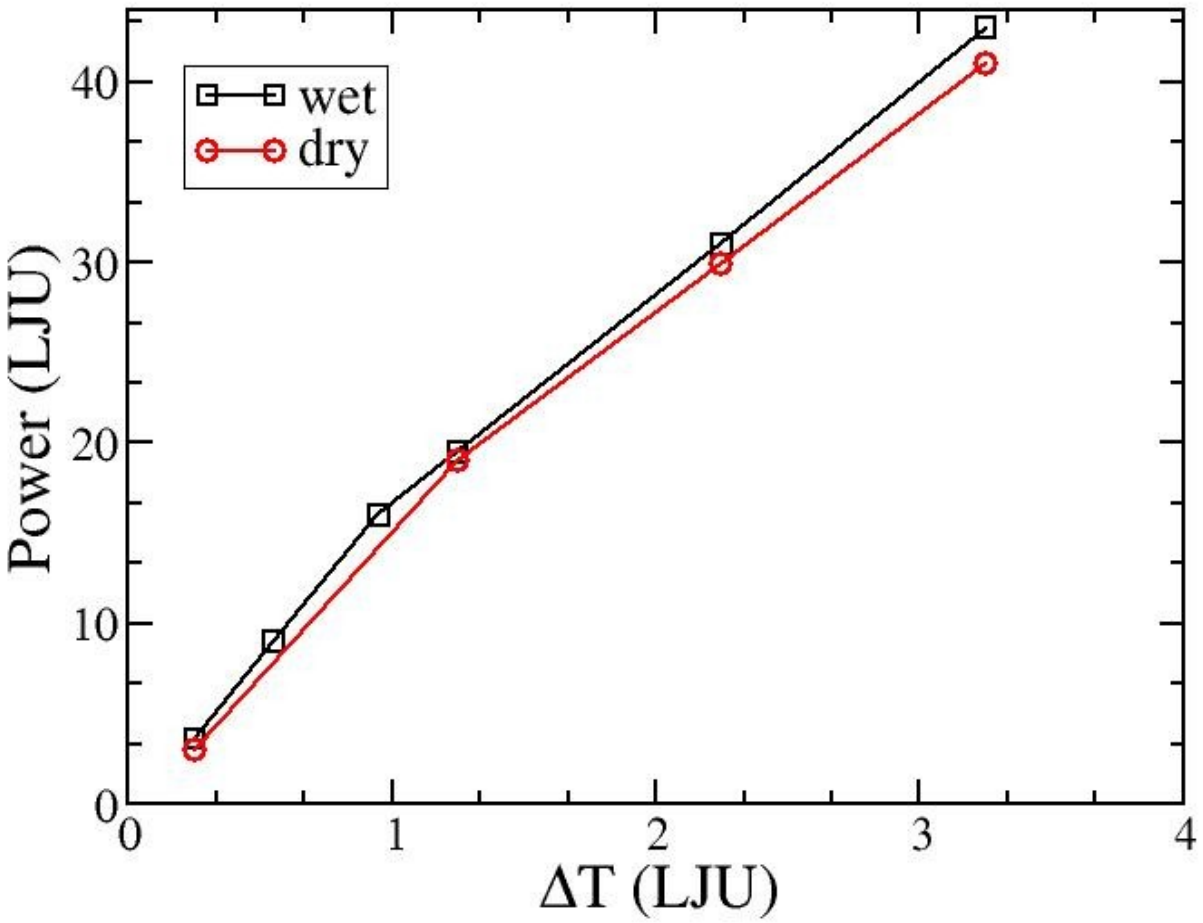


Lennard -Jones potential: $V_{\alpha\beta}(r) = 4\epsilon((\sigma/r)^{12} - (\sigma/r)^{6})$

Wetting interaction

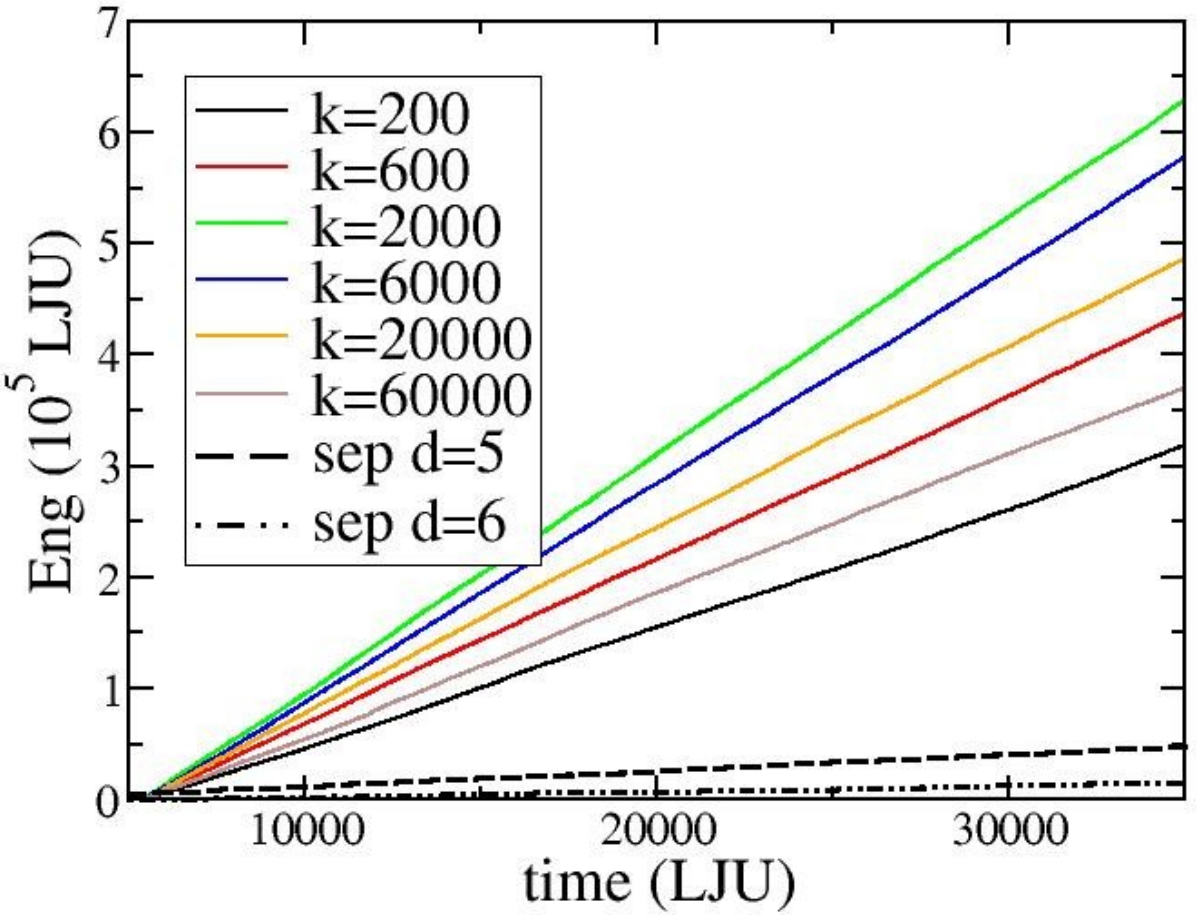
Steady state power vs temperature

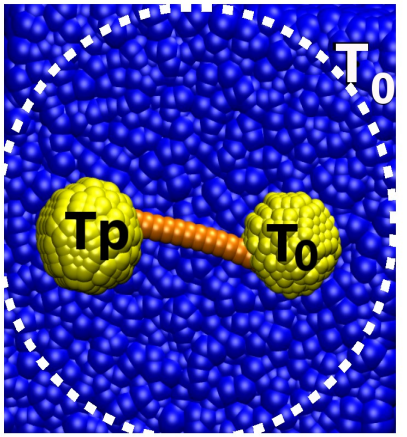
Short junction N=5 atoms



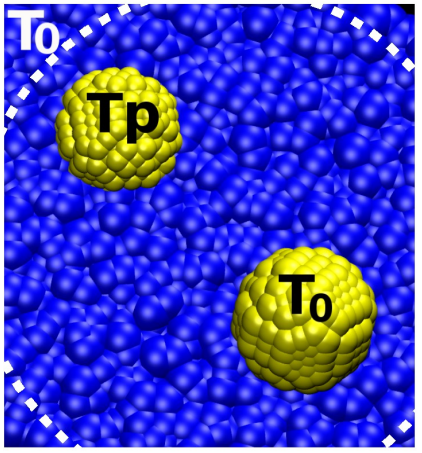
Energy transfer in the liquid

Short junction N=5 atoms



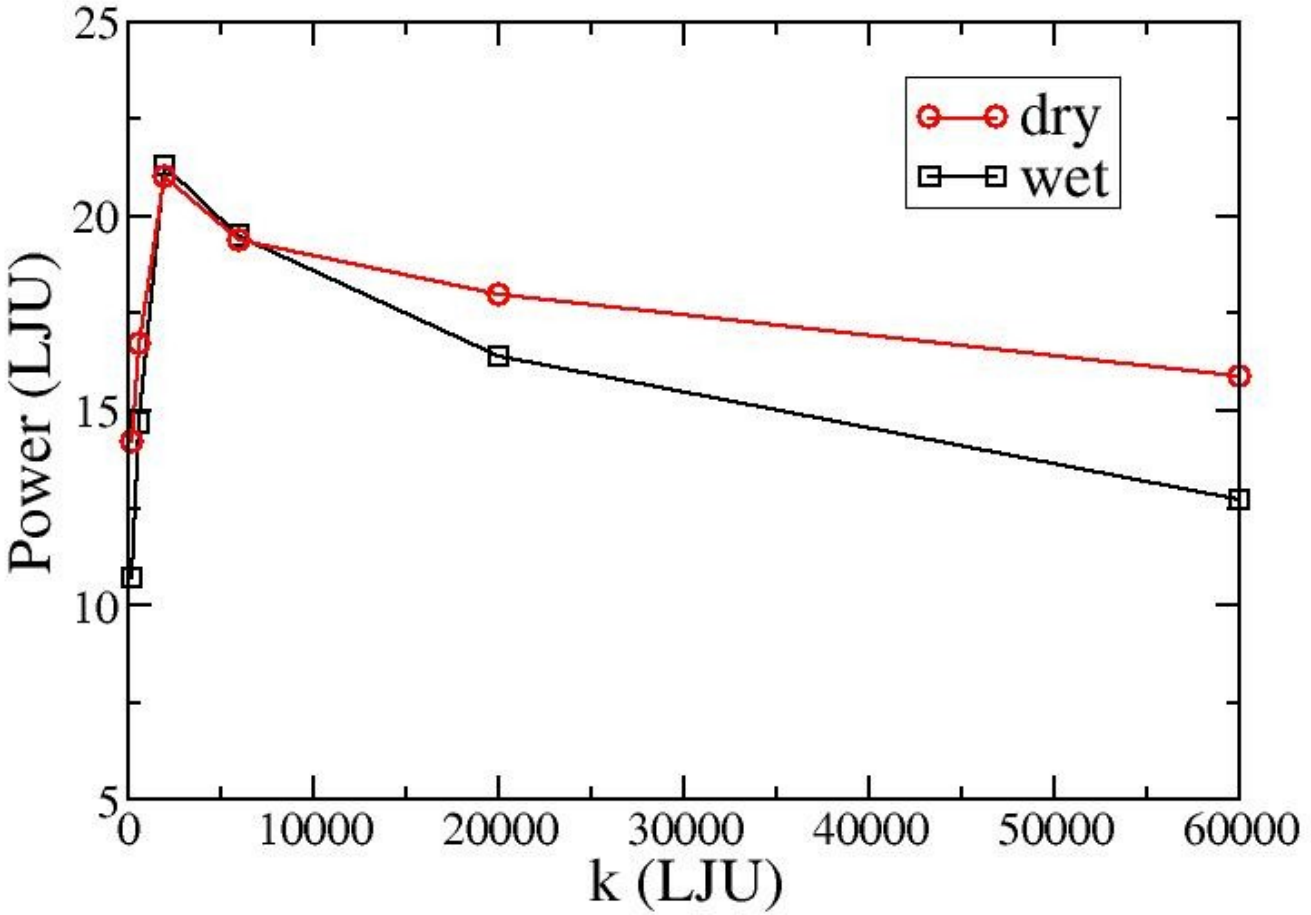


Liquid film

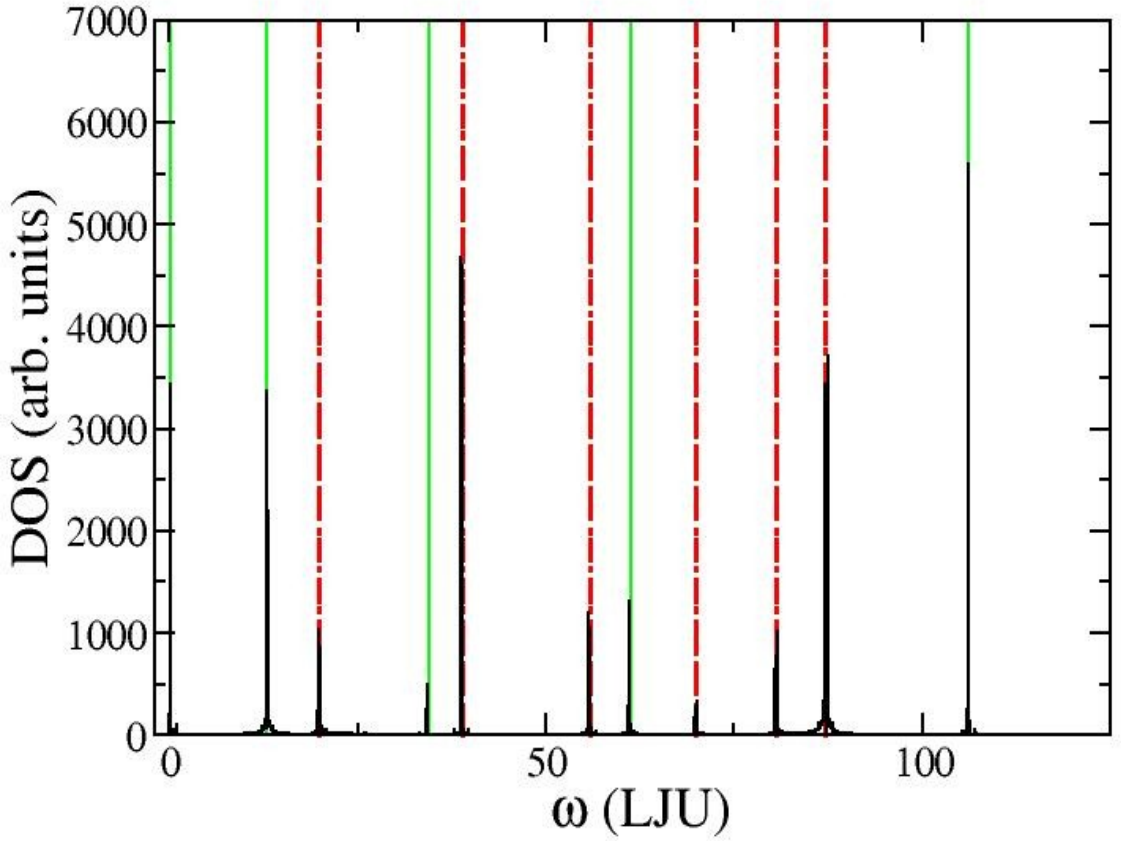


Steady state power vs junction stiffness

Short junction N=5 atoms



Spectrum of a dry junction



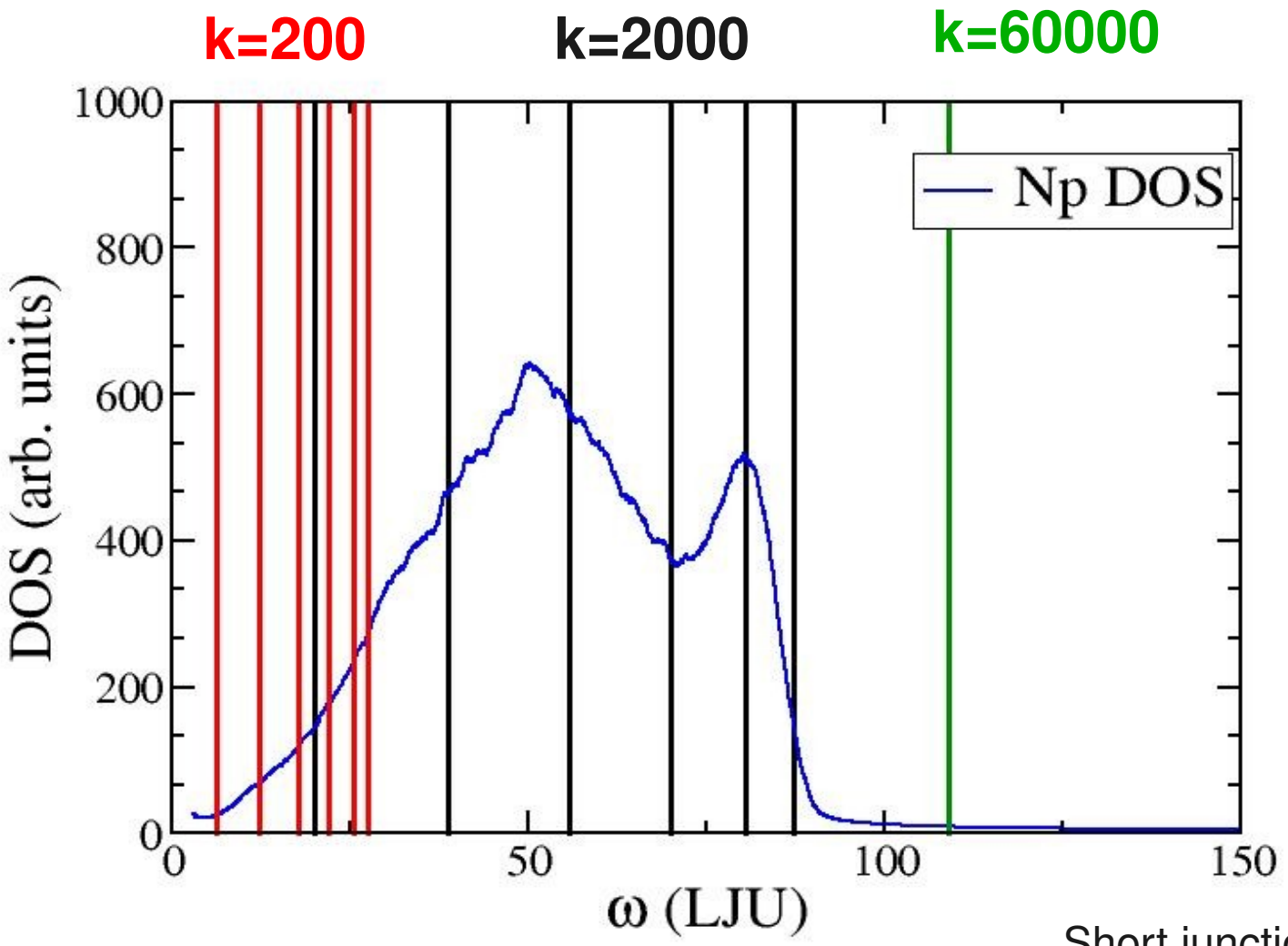
Junction N atoms

N+1 Stretching modes
N Bending modes

Stretching modes :

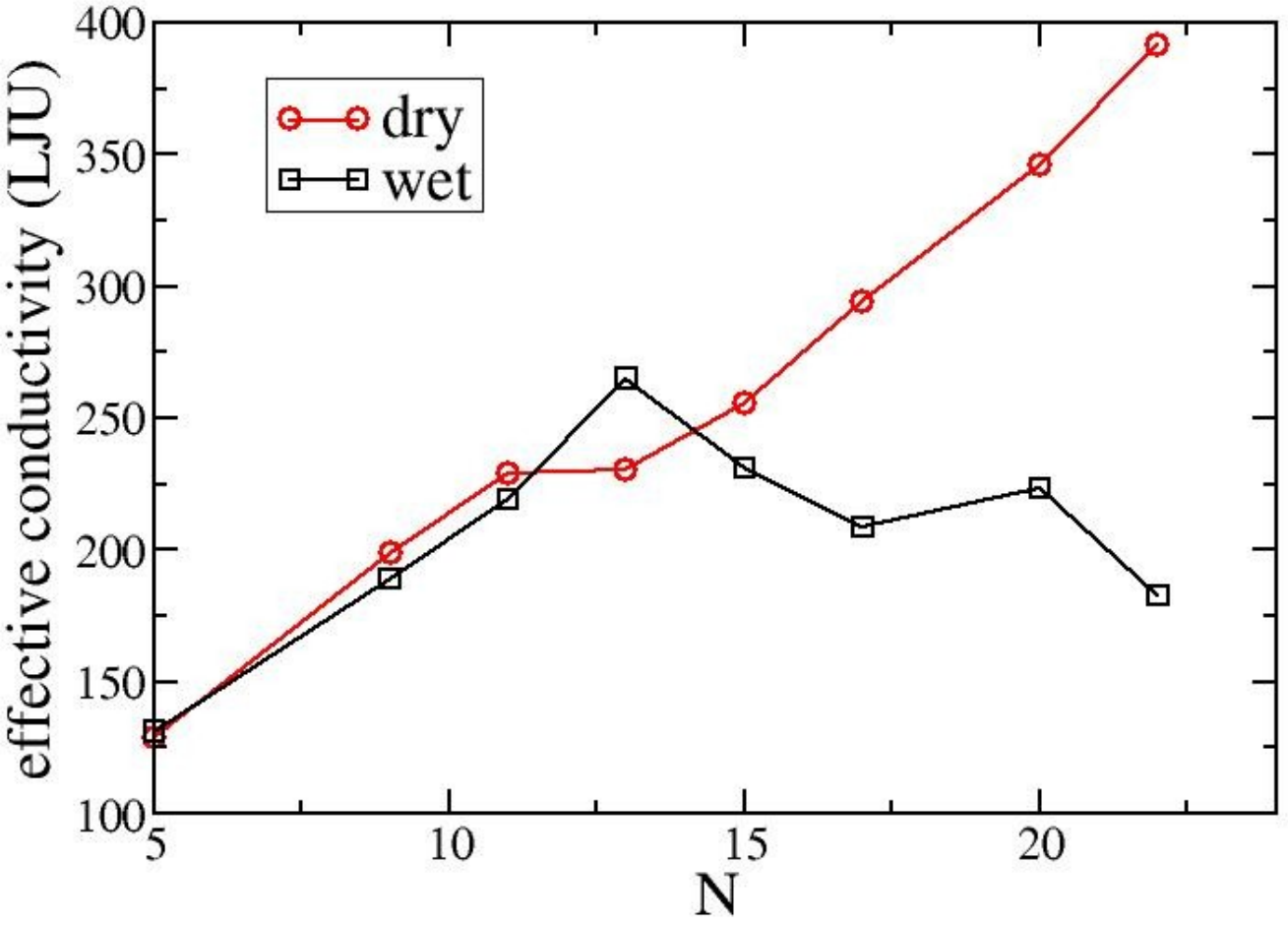
$$\omega_p = 2\sqrt{k/m}\sin(\pi\, p/(2(N+2))),\quad p\in[1,..,N+1]$$

Junction spectrum vs Nanoparticle DOS



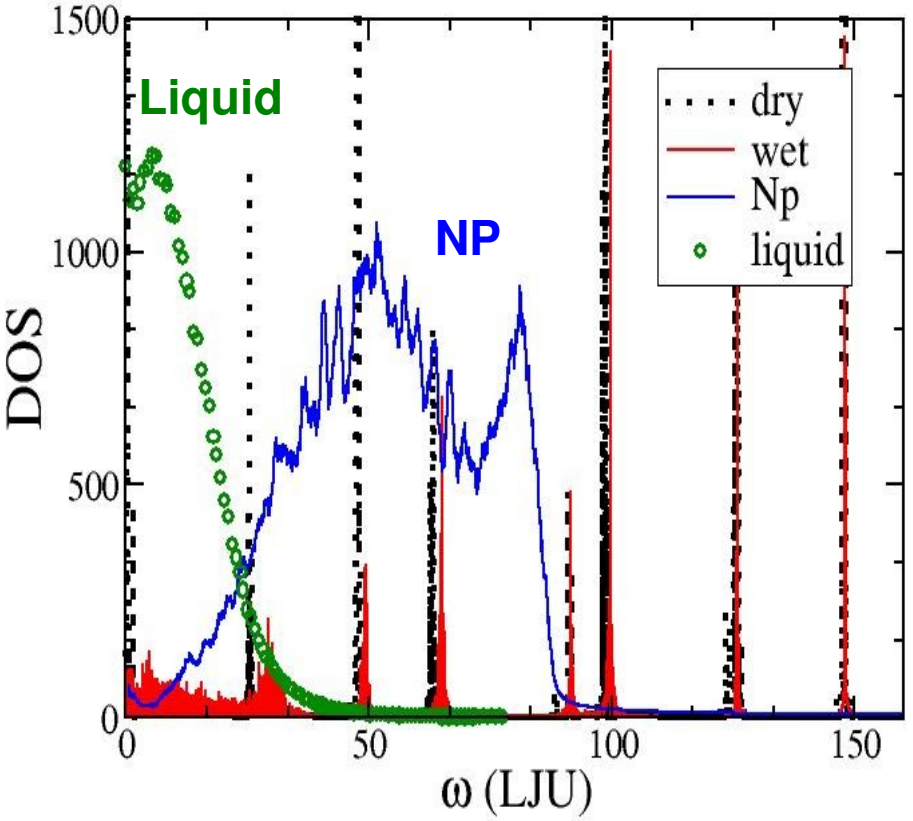
Short junction N=5 atoms

Conductivity as a function of length

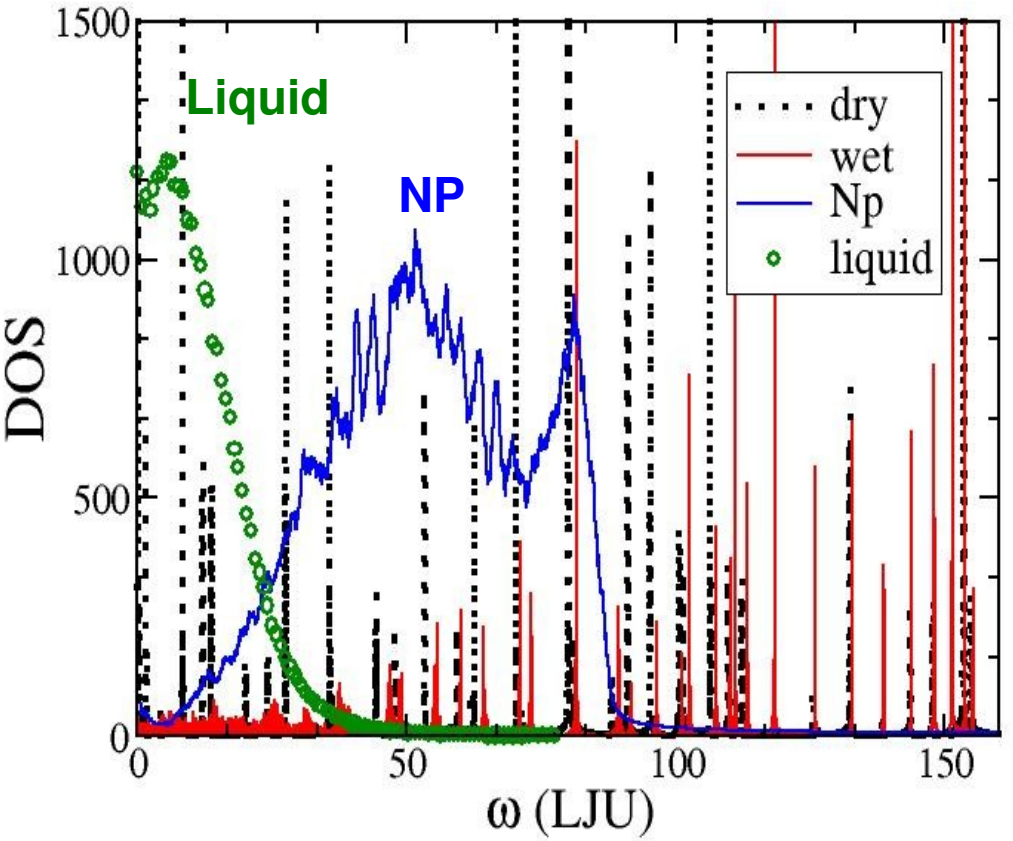


Spectrum of wet short and long junctions

Short junction N=5 atoms



Long junction N=20 atoms



Conclusions

New questions regarding heat transfer at the nanoscale

Characterization of the conductance of molecular junctions

Strong effect of the environment : dry vs wet

Perspectives : realistic modeling of the junction

Thermal rectification

L. Joly, S. Merabia, J-L. Barrat, *EPL* (2011)

S. Merabia, J.-L. Barrat L. Lewis, *J. Chem. Phys.* (2011)

Funding

Computer facilities

Pôle Scientifique de Modélisation Numérique-PSMN ENS Lyon

Thank you for your attention !