

Quest for the Stoner instability in Pd quantum dots

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New Frontiers in the Physics of Quantum Dots

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Motivation

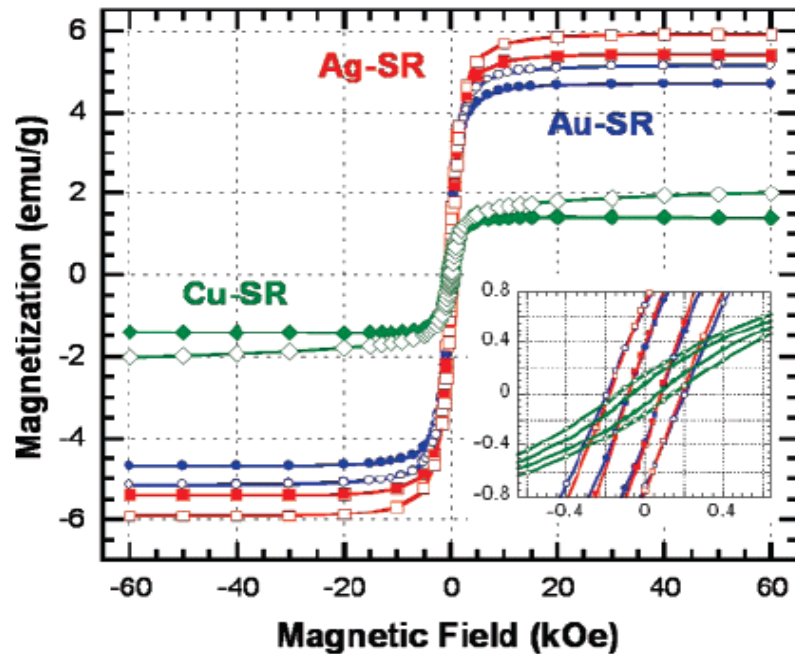


Figure 2. Magnetization curves of the Au, Ag, and Cu dodecanthiol-capped NPs. Open symbols represent measurements obtained at 5 K and filled symbols measurements obtained at 300 K. The similar coercive fields at 300 and 5 K and for the Au, Ag, and Cu NPs can be noticed in the inset.

Experiment:
a number of non-magnetic metals start to be «ferromagnetic» in nano-sized colloid form.

Explanation:

- surface interfacial effects (e.g., NanoLetters 8 (2008) 661)

OR

- volume effect: *space quantization would change balance between kinetic and magnetic energies and bulk of nanoparticle would make a crossover through the Stoner instability to the ferromagnetic phase.*

Nano Letters, 8 (2008) 661

Quest

Theoretical prediction (Burmistrov, Gefen, Kiselev 2010):

$$\frac{\chi_{dc}}{(g\mu_B)^2} = \frac{J_* - J}{2J^2} + \frac{1}{12T} \frac{J_*^2}{J^2} - \frac{1}{12T}$$

Curie-like with «spin»

$$S(S + 1) = (J_*^2 - J^2)/(4J^2)$$

Estimated effect: magnetization of $\sim 20 \mu_B$ per 5nm diameter Pd particle to develop at $T \sim 1 \dots 10K$.

- 1) To prepare the samples.
- 2) To measure magnetization.
- 3) To check whether the magnetic response comes from the bulk or from the surface of the sample

Samples

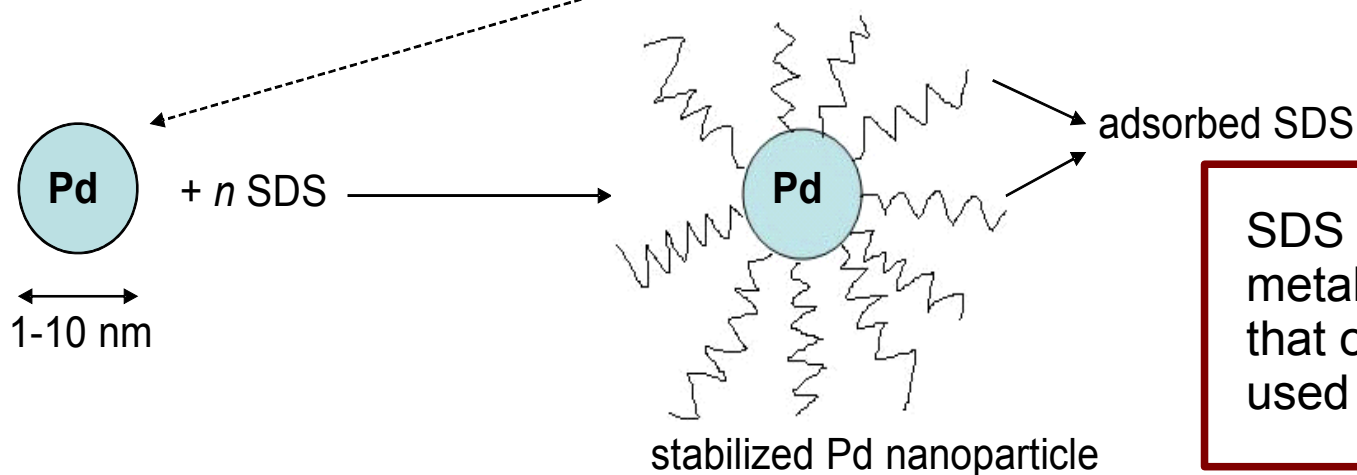
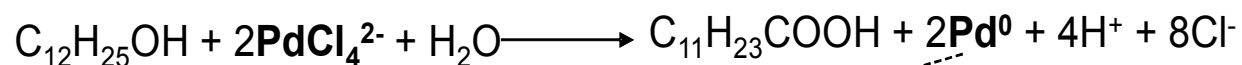
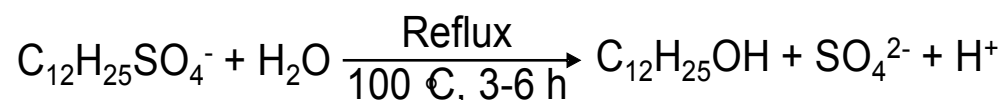
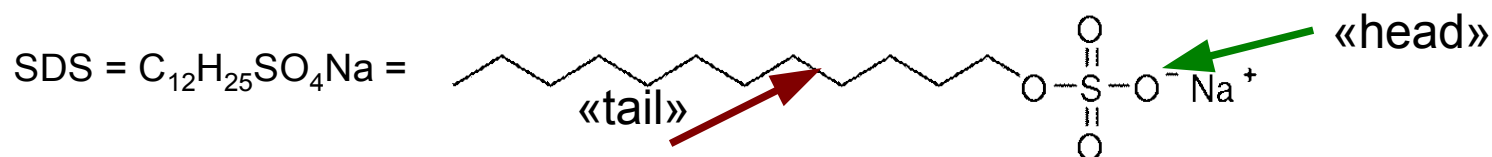
Pd nanoparticles stabilized by dodecyl sulphate (SDS) molecules were fabricated by wet technique

Sample ID No	Concentration of $[\text{PdCl}_4]^{2-}$	Synthesis duration, hours	Concentration ratio Pd:SDS	particle size (from TEM)
11	0.05 M	3	1:2	2.5nm
12	0.05 M	6	1:2	2.8nm
9	0.0025 M	3	1:40	3.1nm+12nm
10	0.0025 M	6	1:40	2.0nm+12nm

masses of all samples for magnetization measurements ~5mg
($\sim 3 \cdot 10^{15}$ particles)

Samples: preparation details

PdCl_4^{2-} chemical reduction and *in situ* stabilization by SDS (dodecyl sulphate).

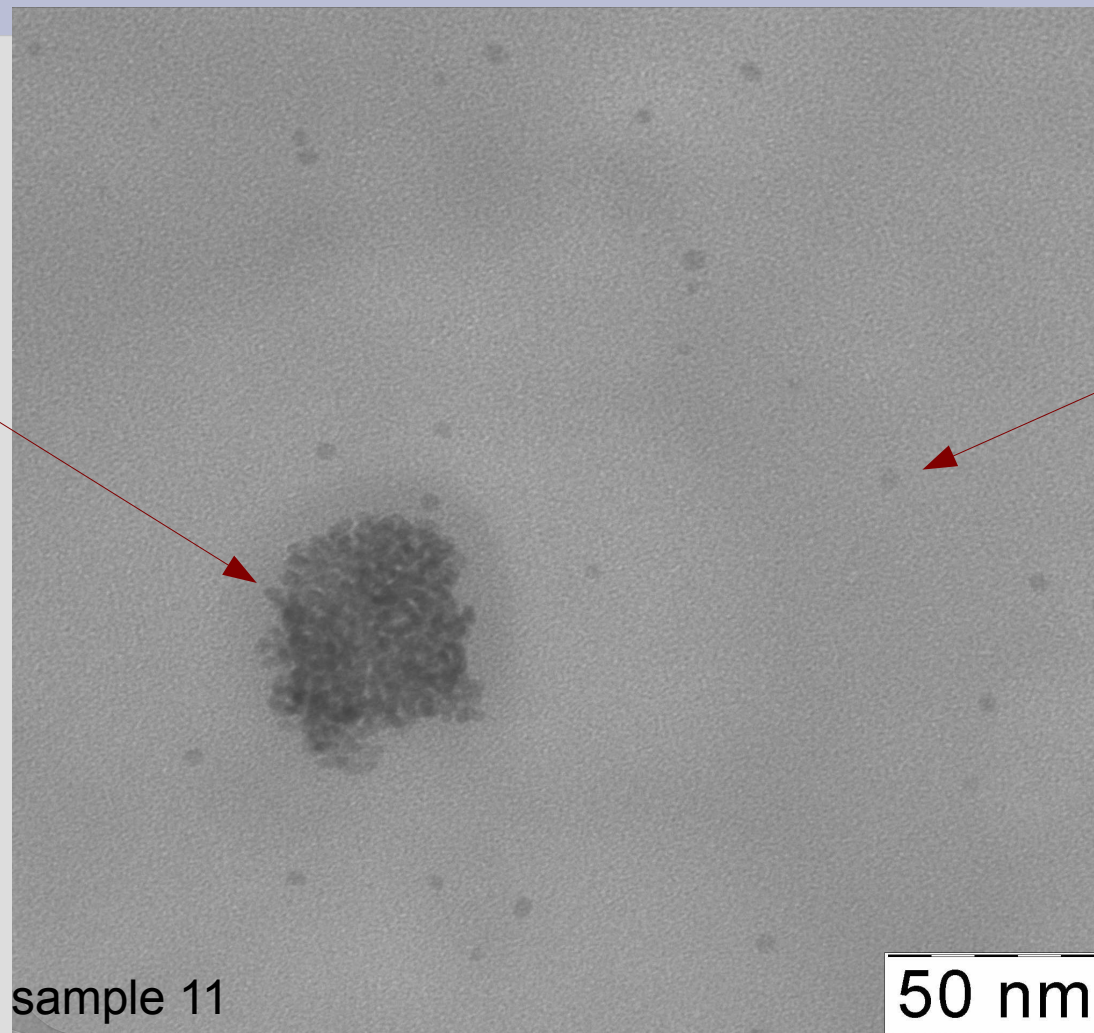


SDS interaction with metal is weaker than that of other spacers used in the art.

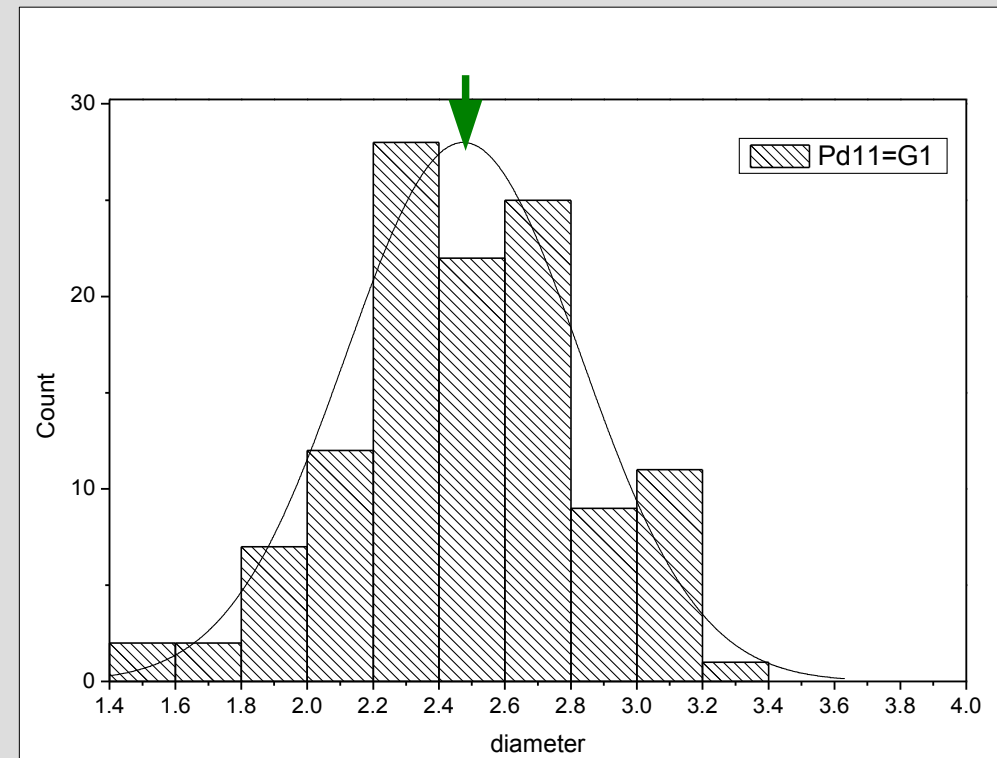
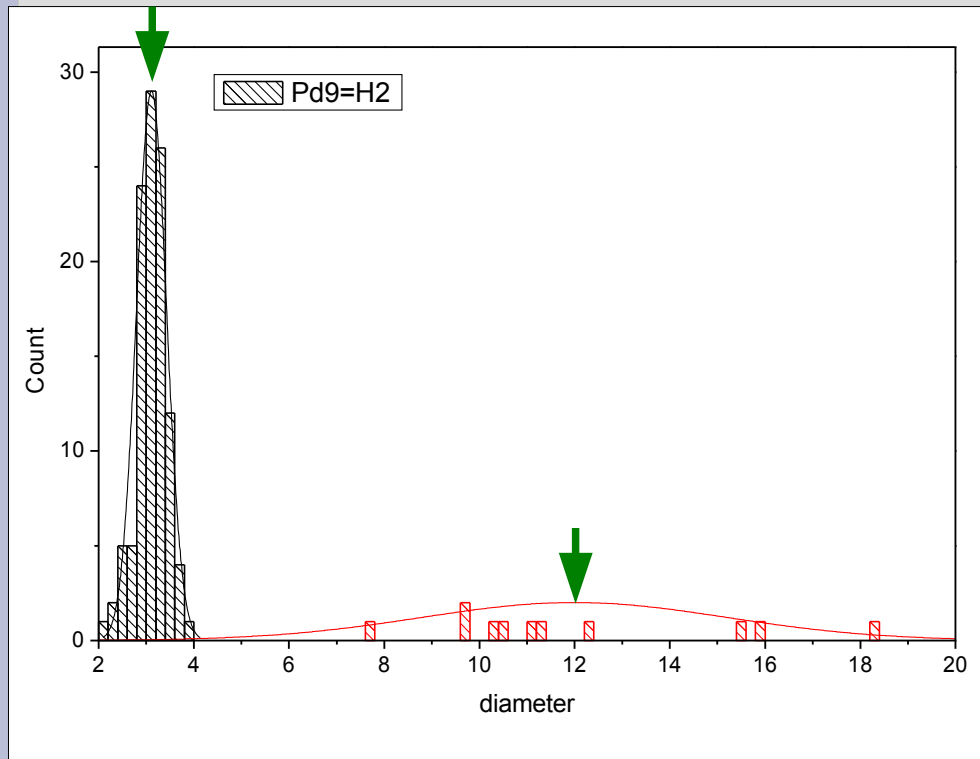
Samples: TEM image

agglomeration of
SDS-covered Pd
particles

single Pd
particle



Particle size distribution (TEM data)



Experimental details

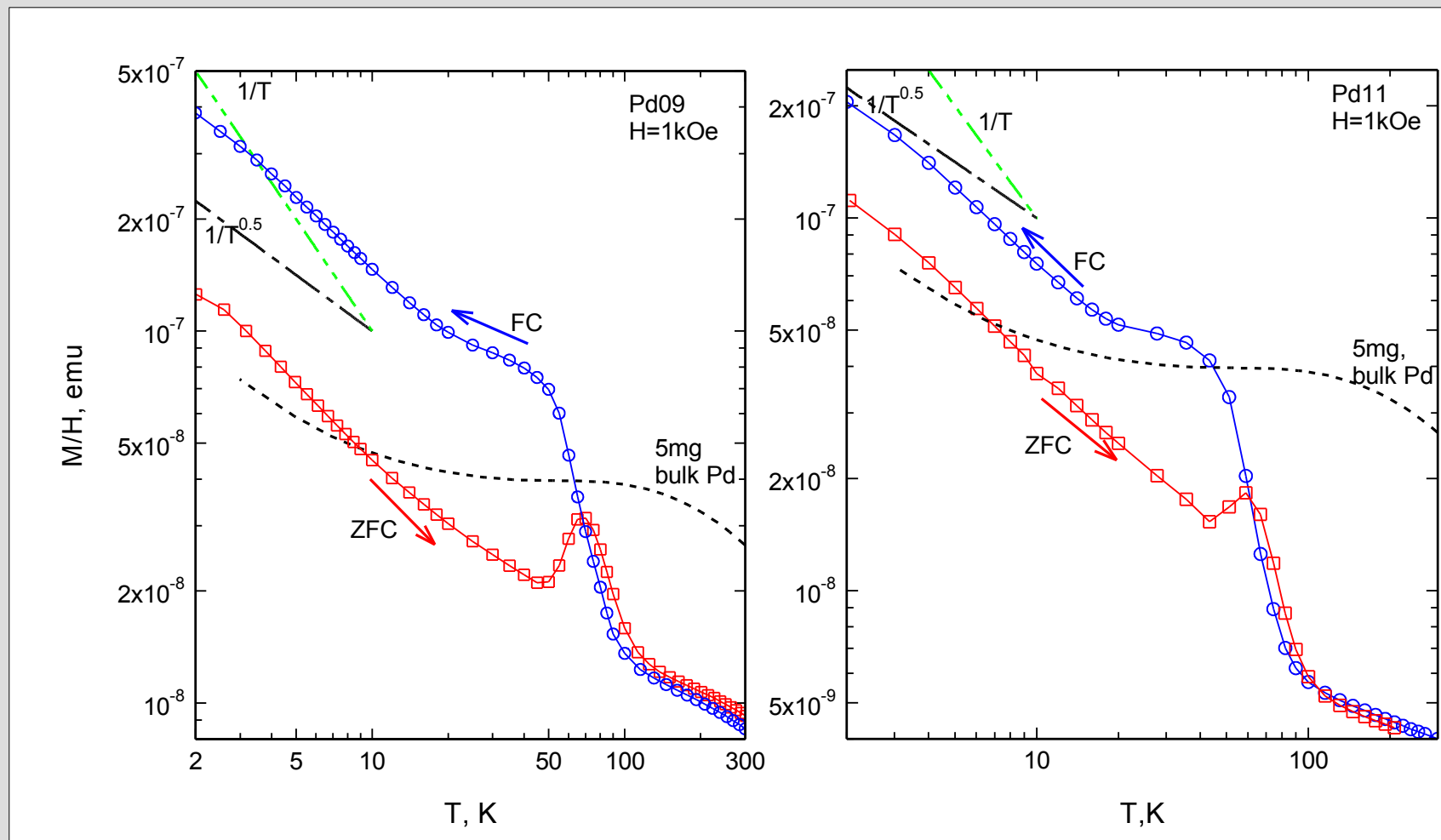
Magnetization measurements:

2K-300K: SQUID magnetometer (QD MPMS XL7)

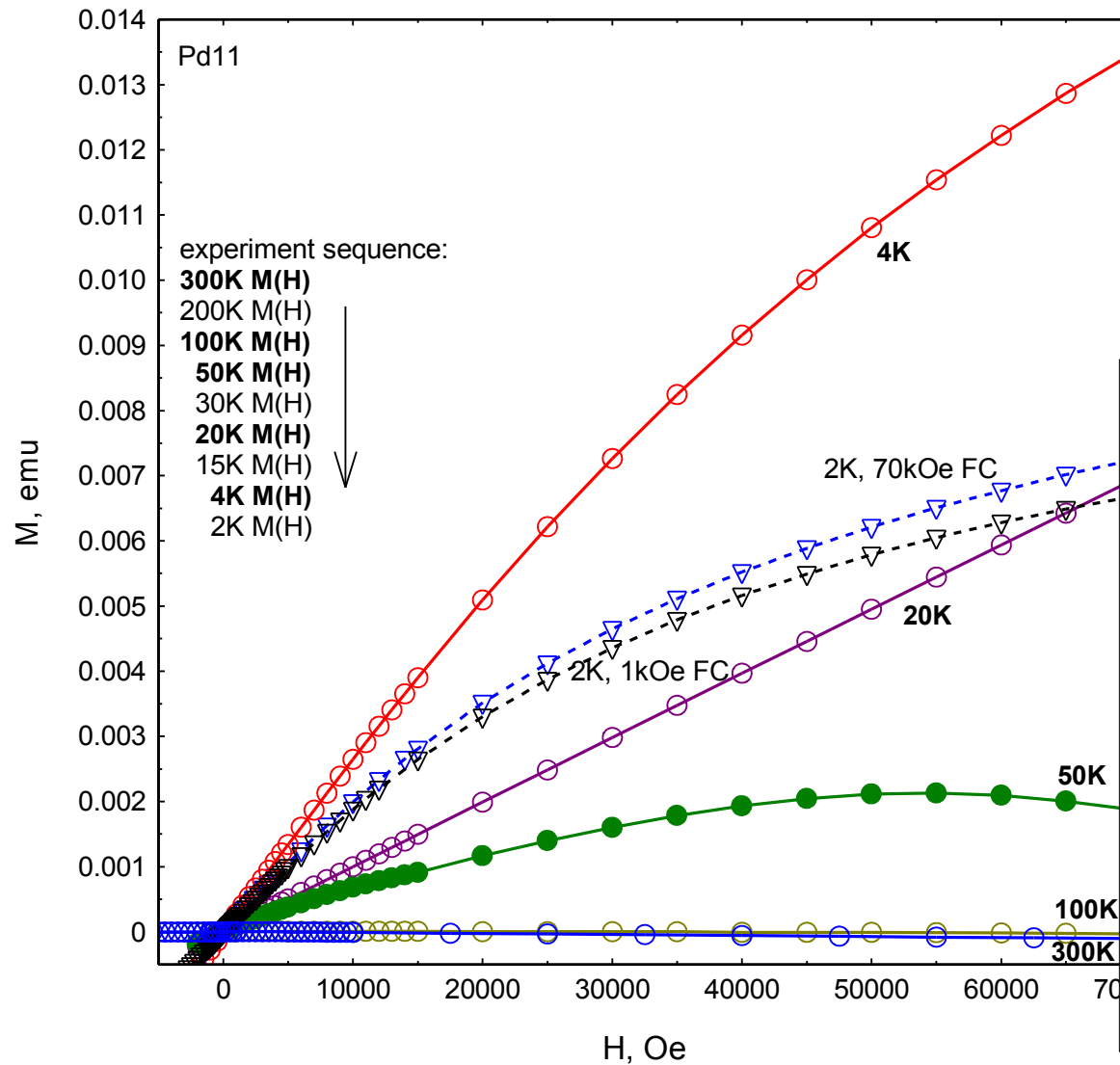
0.5K-2K : iQuantum He3 insert for MPMS

2K-300K, CO+He or H₂+He atmosphere: MPMS, home-made insert

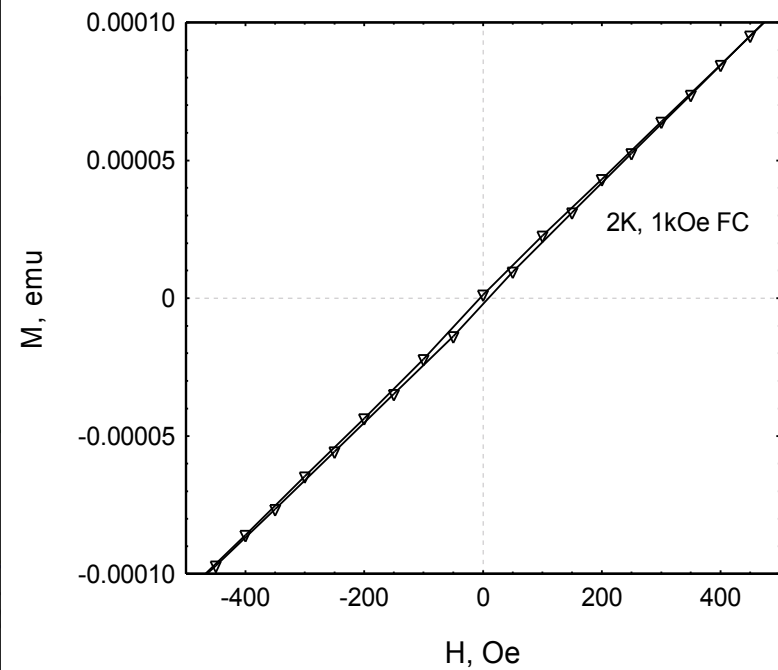
M(T) curves down to 2K



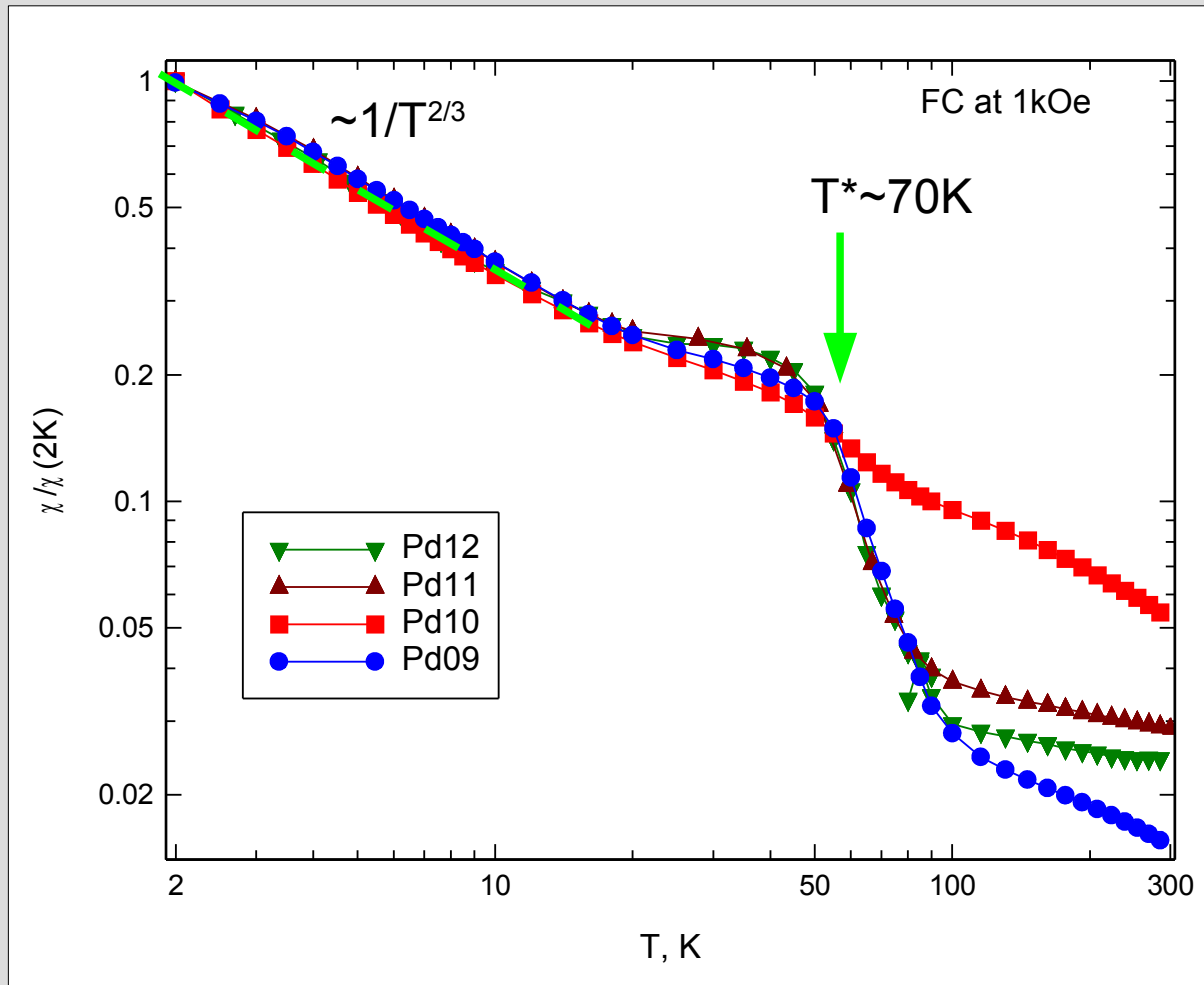
M(H) curves



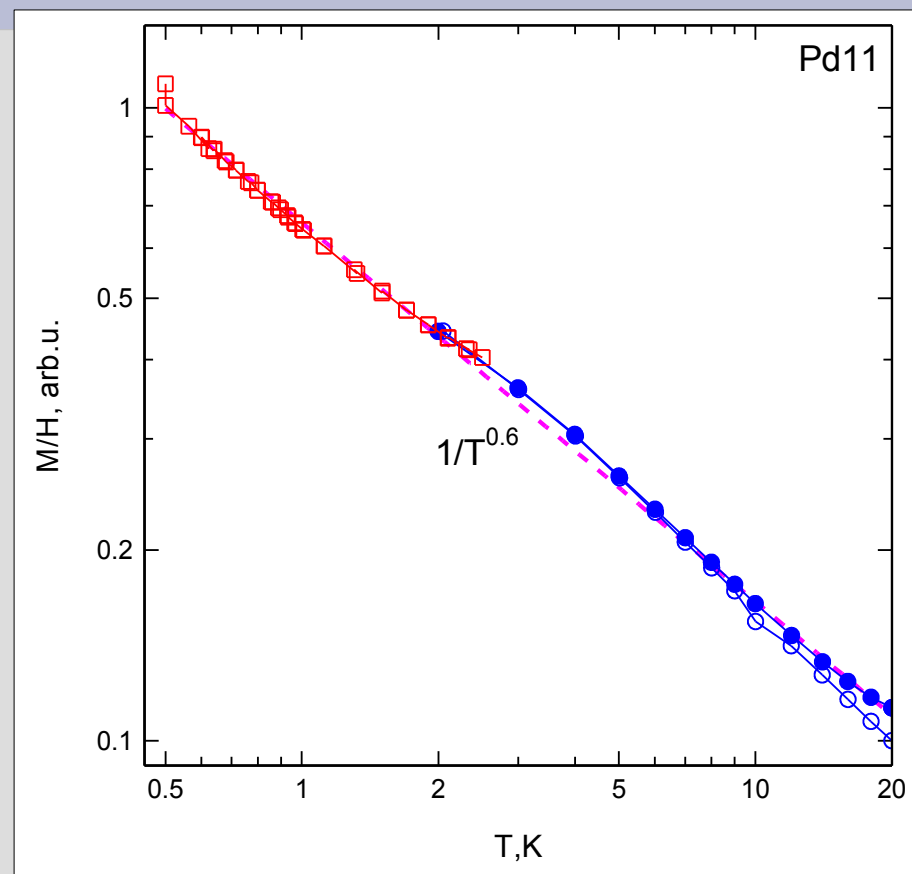
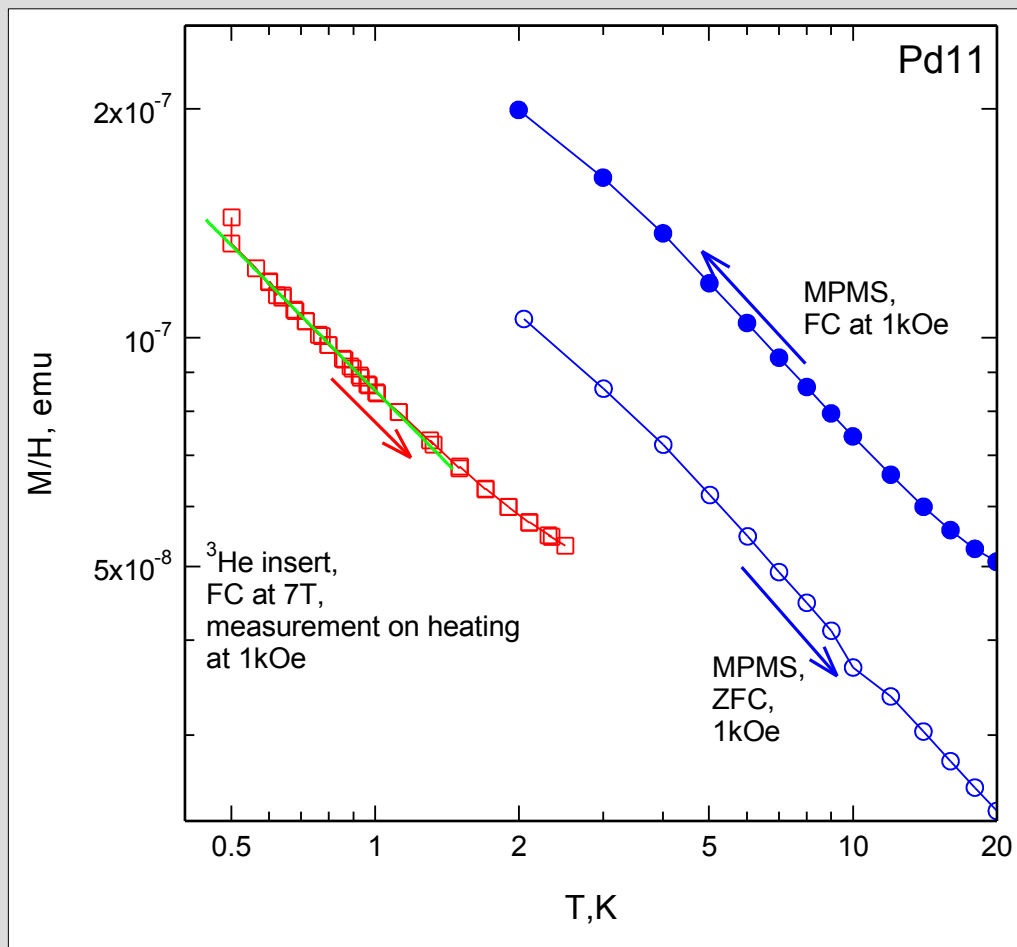
THEORY: *estimated
saturation value
 $M \sim 0.0006$ emu*



Scaling of all curves at low temperature



M(T) to 500mK



Experimental data summary

Low-temperature magnetization strongly depends on sample cooling history.

Low-temperature $M(T)$ curves do not follow $1/T$ law. Low-temperature behavior is the same for all samples irrelevantly to the particle size.

At 2K magnetization does not saturate up to 7T.

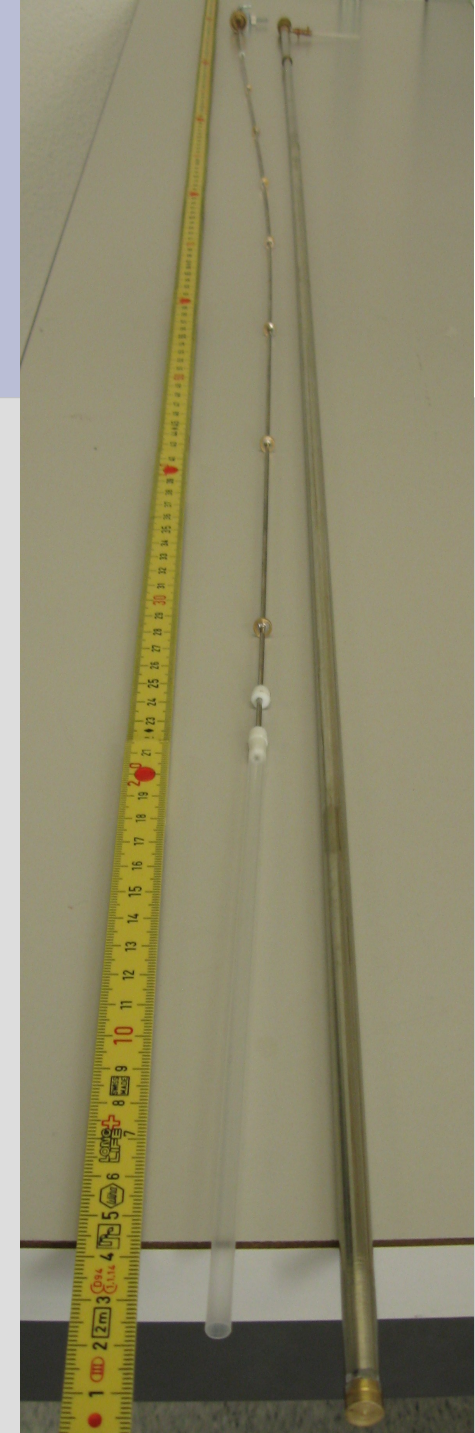
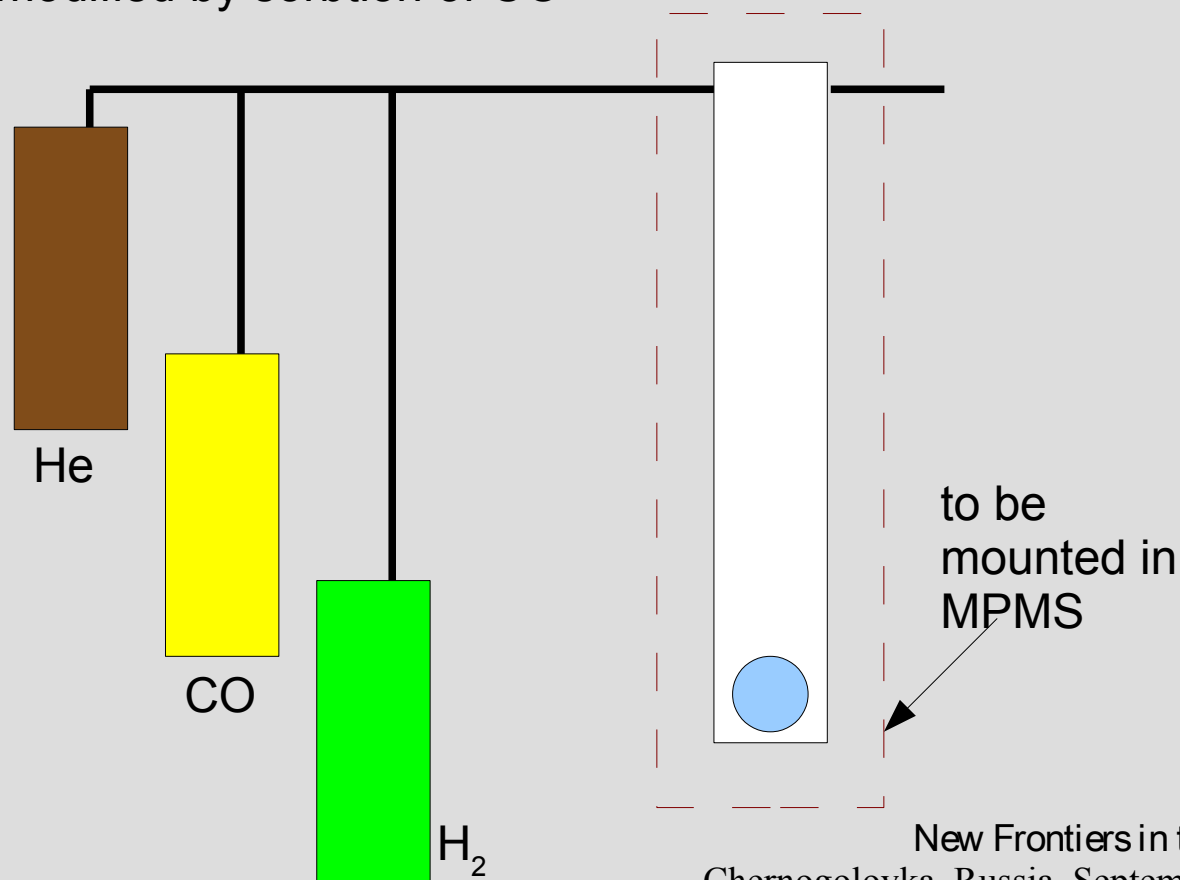
Observed magnetization value exceeds expected saturation value by an order of magnitude.

No hysteresis was observed on $M(H)$ loops.

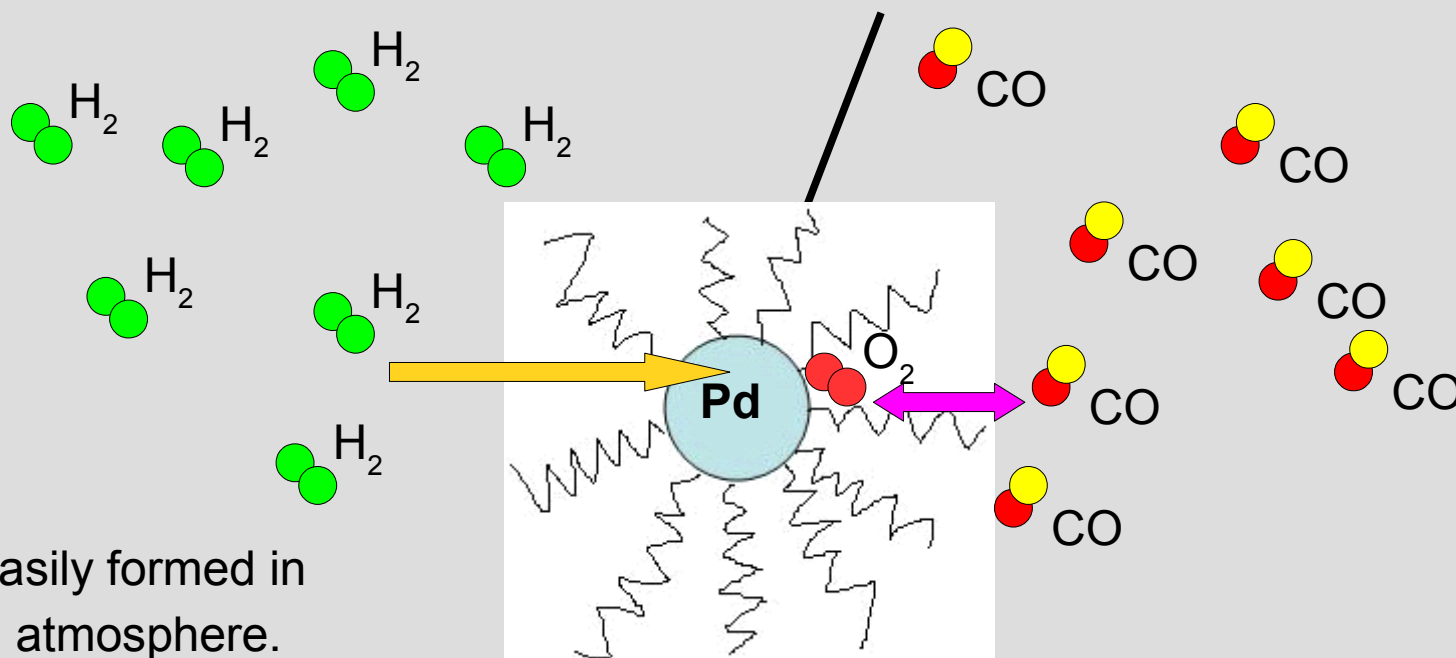
Surface or bulk magnetization?

Bulk of the Pd sample: can be reversibly modified by sorption of hydrogen.

Surface of the sample (e.g. oxygen defects): can be irreversibly modified by sorption of CO



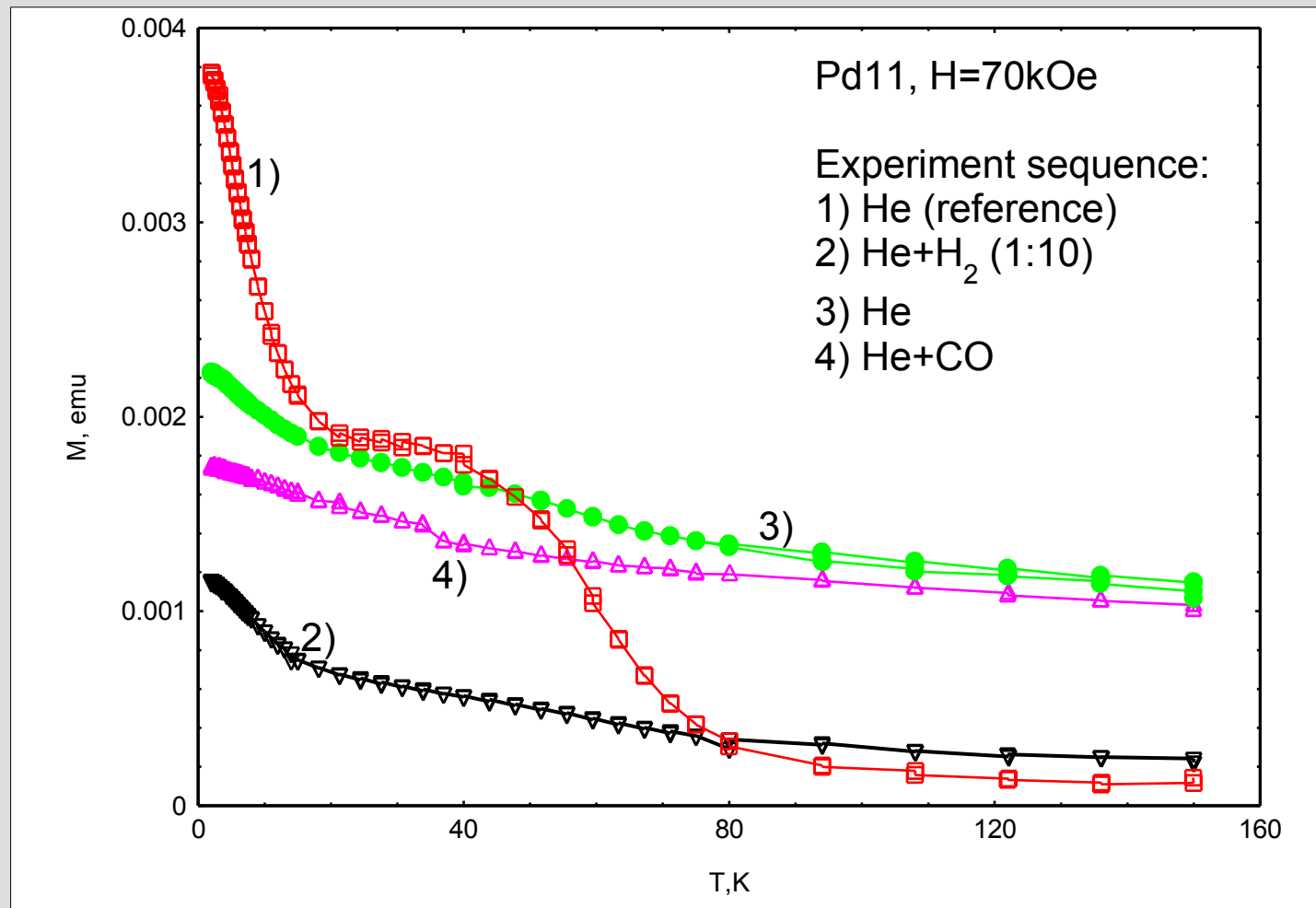
Expected CO and H₂ effect



PdH_x is easily formed in hydrogen atmosphere.
 At room temperature:
 at 1.0 atm of H₂ $x=0.65$
 at 0.1 atm of H₂ $x=0.60$
 below 0.01 atm of H₂ $x \rightarrow 0$
 Around $x=0.5$ PdH_x became a semiconductor

Samples were treated with CO or H₂ atmosphere for several hours. C.a. 10% of He were added for thermalization at low temperatures.

Effect of the atmosphere on the magnetization process



Surface or bulk?

Sorbption of hydrogen leads to reduction of magnetization — it is due to the volume.

Desorbption of hydrogen was (probably) accompanied by explosive destruction of the sample, which (probably) explains why magnetization curve after desorbption does not coincide with the initial one.

However, this bulk magnetization seems to be not dependent on particle size...

Polarization of Pd bulk by Fe impurities

(Larkin and Melnikov, JETP 61, 1231 (1971))

susceptibility per impurity

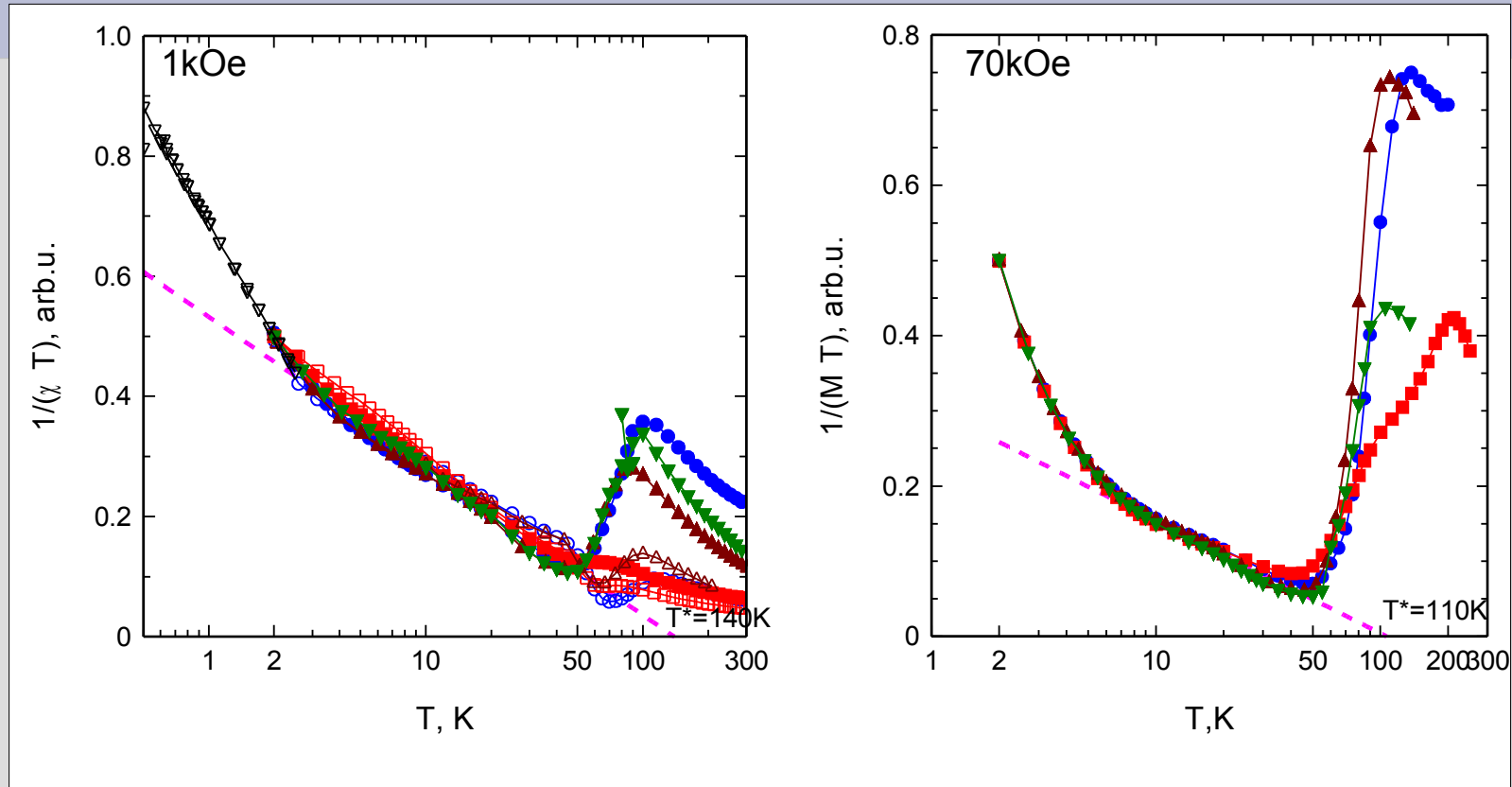
$$\chi = \frac{\mu^2 S(S+1)}{3T\lambda \ln(T_*/T)}$$

$$T_* = e^{1/\lambda} 4(1 + F_0^\sigma)^{3/2} E_F / (\pi a k_F)$$

$$\mu = g_{\text{imp}} \mu_B \left[1 + \frac{g_e}{g_{\text{imp}}} \frac{a k_F \sqrt{\lambda}}{\sqrt{1 + F_0^\sigma}} \right]$$

$$\frac{1}{\chi T} = A (\ln T^* - \ln T)$$

Comparison with the experiment



$$\frac{1}{\chi T} = A(\ln T^* - \ln T)$$

$T^* \sim 110 \dots 140$ K

+saturation effects at low temperatures.
Experimental magnetization corresponds
to impurities concentration $x \sim 10^{-3}$

Conclusions

Our set of samples does produce a magnetization response related to the sample bulk.

But, this response is not due to quantum-dot physics — it is due to the polarization of Pd by magnetic defects.

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