Polymer physics in nanoscale cutting

Opportunities for improved control in nano-manufacturing?

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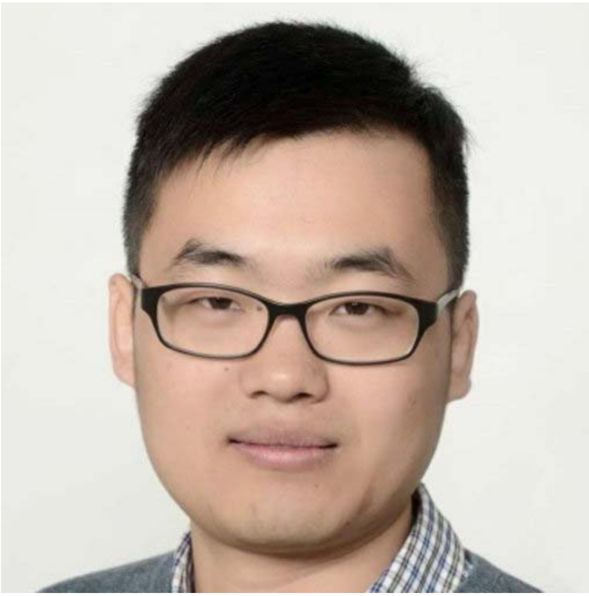
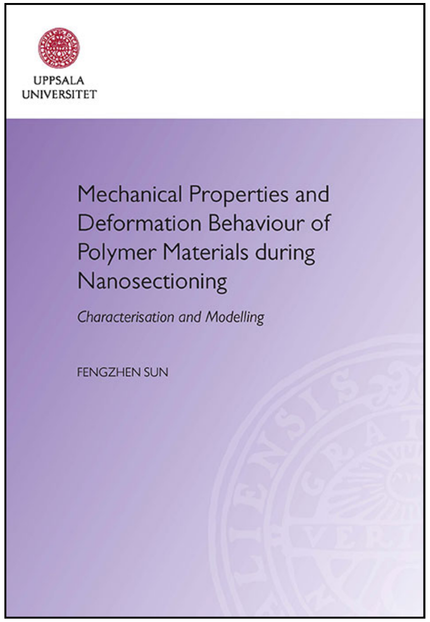
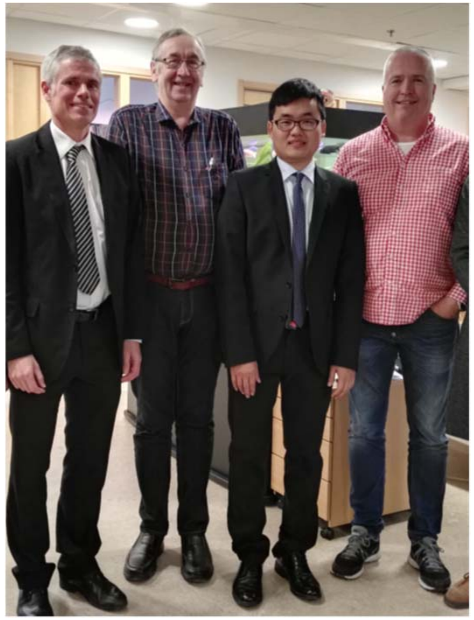
Work mainly carried out by Dr. Fengzhen Sun
PhD Uppsala University, 2017







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Design and potential of instrumented ultramicrotomy

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Ultramicrotomes are generally used for preparation of very thin sections for transmission electron microscopy. Recently it has been shown that when the sample holder of the ultramicrotome is instrumented with a force transducer, it is possible to measure the very small sectioning force during sectioning, and calculate the energy dissipated. In the present work, the instrumentation is further improved. The new sample holder, which uses two piezo-electric force transducers can measure two force components simultaneously. It is not only robust and stiff, but it also shows high sensitivity and reproducibility. It is possible to detect sectioning forces lower than 0.1 mN. The method is demonstrated on two amorphous polymers, poly(methyl methacrylate) and epoxy. Fracture energies in the same order of magnitude as theoretical predictions from chemical bond fracture only are recorded. It is therefore suggested that the method of instrumented ultramicrotomy is a useful tool when information on covalent bond density is needed. Potential future applications are identified including research on nano-scale fracture, characterization of molecular anisotropy and developments of the ultramicrotome. © 1997 Elsevier Science Ltd.

(Keywords: ultramicrotome; sectioning forces; fracture)

INTRODUCTION

An ultramicrotome is used for sample preparation for transmission electron microscopy (TEM). By use of a very sharp knife (diamond or glass), extremely thin sections or 'chips' of, for example, polymers or biological materials are prepared. The chip thickness is typically between 30 nm and 150 nm, and in order to get good sectioning results the sample needs to be in the solid, or preferably in the glassy state.

Since microtome sectioning creates two new surfaces one may look at the sectioning as a crack propagation process. Ericson and Lindberg[1] showed that if the ultramicrotome is instrumented in order to measure sectioning forces it is possible to calculate the sectioning energy during the crack propagation. Since the thickness of the sections are very small, this might give us some insight in very local fracture energy dissipation mechanisms.

Microtomes, which are used for section thicknesses from 0.5 to 10 μm, have been instrumented by several workers. Vincent *et al.*[2-5] used a rotary microtome fitted with a modified knife holder which was a load cell. They measured cutting forces, and when the thickness of the sections were decreased continuously from 1 mm to 1 μm the cutting energy also decreased for a number of biological materials. Atkins and Vincent[2] suggested that the sectioning energy extrapolated to zero thickness is equal to the critical fracture energy, G_C. No measurements on polymers have been published, but an interesting study on meat was published by Dobraszczyk *et al.*[6]. Wool and Rockhill[7] studied the molecular degradation during microtome sectioning by viscometry. Saubermann *et al.*[8] presented an alternative method to evaluate the sectioning forces on a microtome. However, as seen in the work of Ericson and Lindberg[1] for ultramicrotomes, it is impossible to extrapolate sectioning energies from experiments at the micrometre scale down to zero thickness without significant error. The reason is the dramatic nonlinearity in sectioning energy vs chip thickness in the thickness region below 200 nm. Therefore, in order to measure energy dissipation at a true microscopic scale, one needs to instrument an ultramicrotome.

Several attempts to instrument an ultramicrotome have also been presented in the literature[9-13], all with the purpose to determine sectioning forces. It is found that the techniques for instrumentation vary as much as the reasons for instrumentation. Hodson and Marshall[10] studied the thawing of frozen material during sectioning. They calculated the forces acting on the substrate by measuring the velocity of the specimen arm by a method of electromagnetic induction. When they knew the mass and the retardation of the arm it was possible to calculate the energy dissipated into the material during sectioning. Wikefeldt[11] placed two piezoelectric elements in the drive arm of the ultramicrotome. It was then possible to measure both forces acting perpendicular and parallel to the sectioning direction. The reason for his instrumentation was to record instability vibrations, so called 'chatter',

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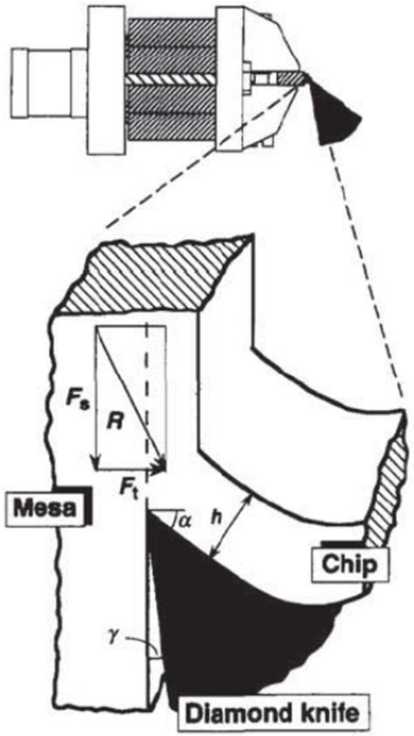
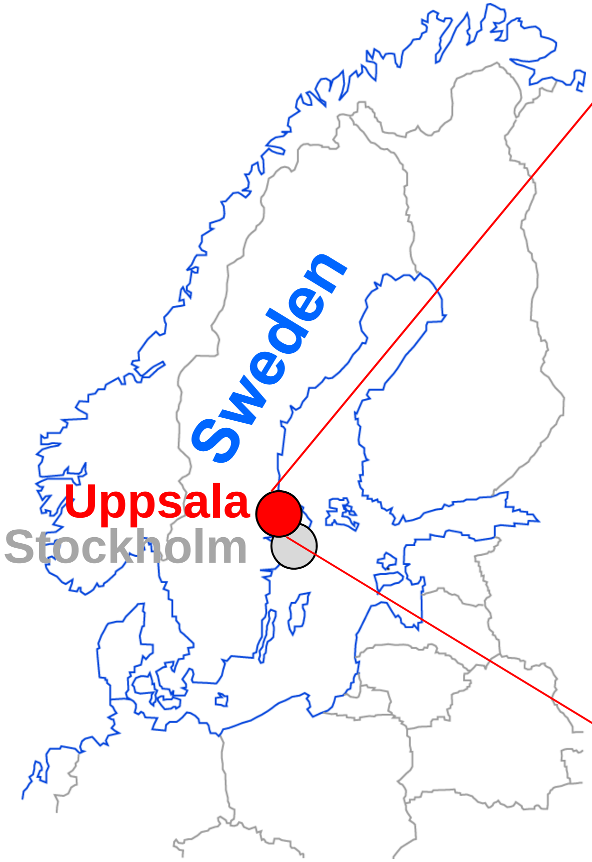


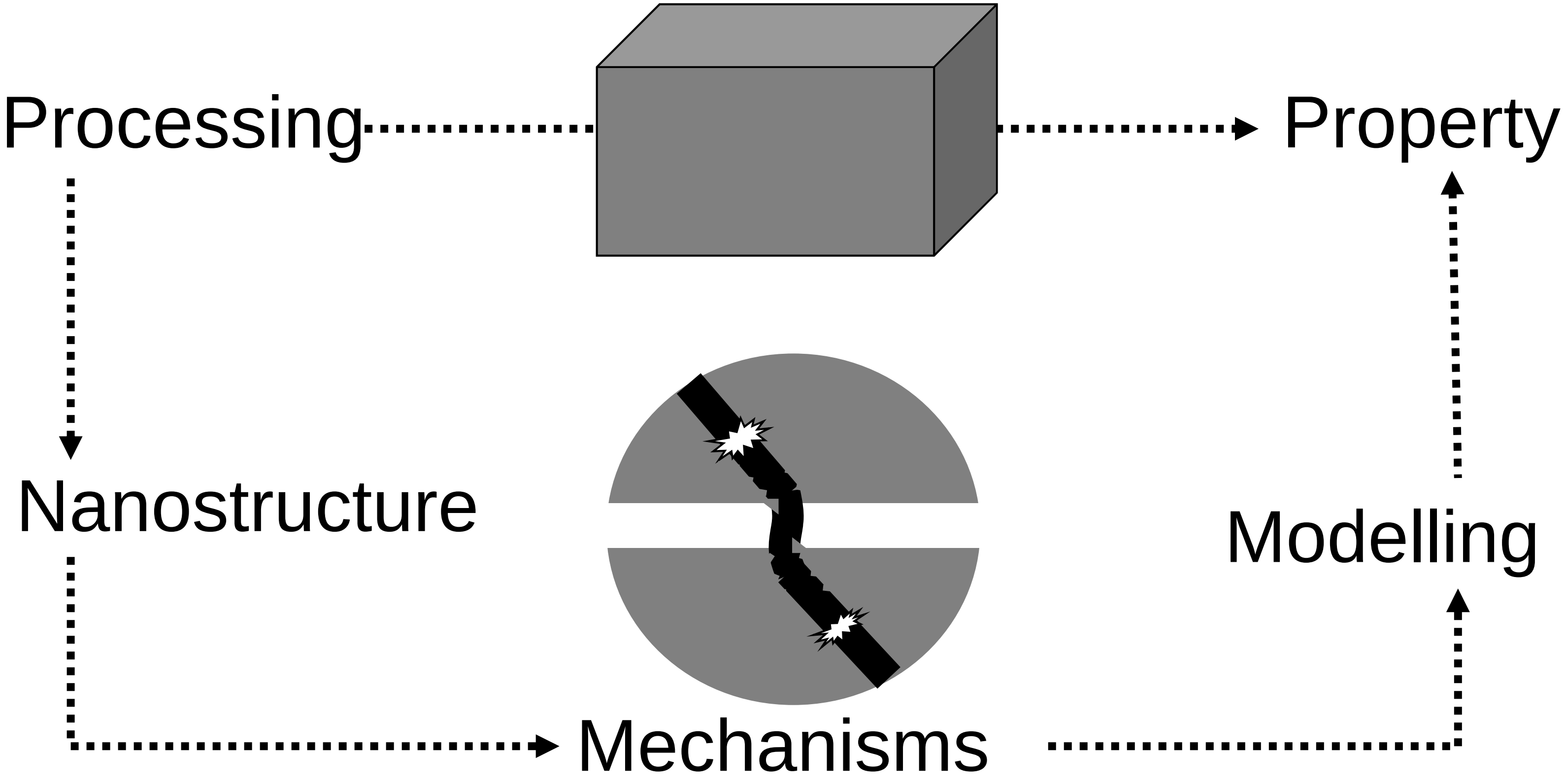
Ericson, M.L. and Lindberg, H., "Design and potential of instrumented ultramicrotomy", *Polymer*, **38** (1997), 4485

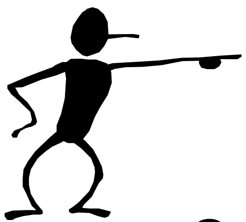
Uppsala University



Full university, founded in 1477
Physics, chemistry and engineering in the Ångström Laboratory

Background in material mechanics



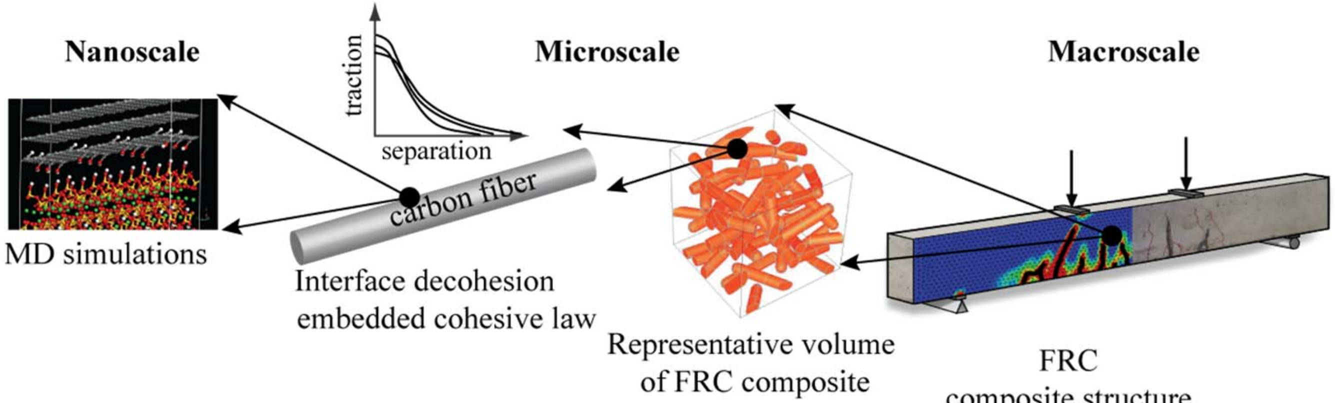


Vision: Experiments and modelling to develop predictive tools for optimized materials design

Composites, wood, paper, board, polymers, asphalt, implants…

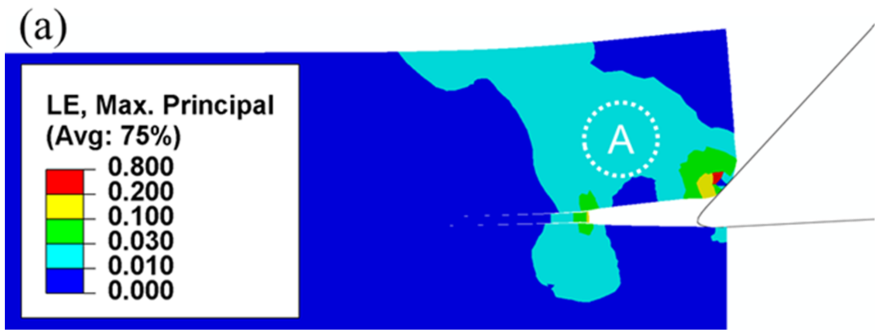
Experimentally characterized constitutive models on nanoscale …

① … to predict and design nanocomposites etc.



(Vanderbuilt University, Dept Mech Eng, 2017)

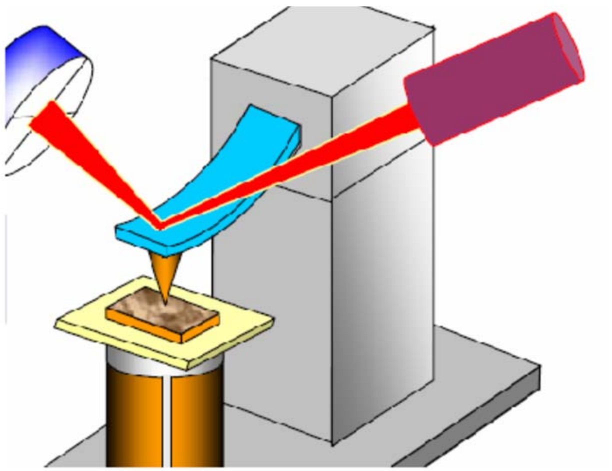
②… to predict and control in nanomanufacturing



Cross-disciplinary fertilization:

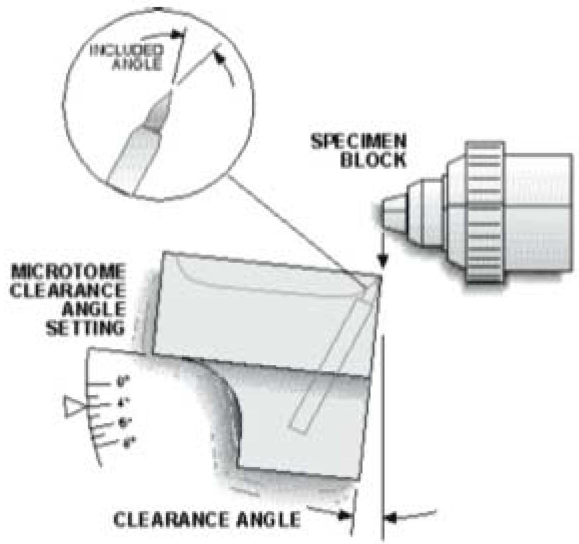
Methods developed for other applications also useful in nanomanufacturing

AFM



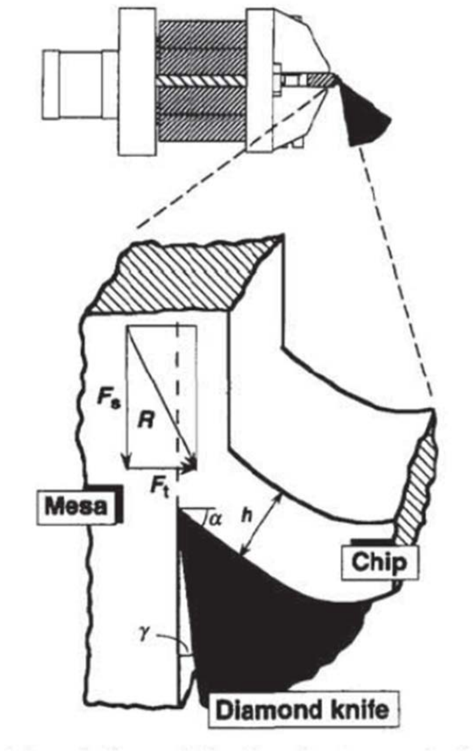
E.g. Yongda Yan (HIT), Emmanuel Brousseau (Cardiff) @ this conf.

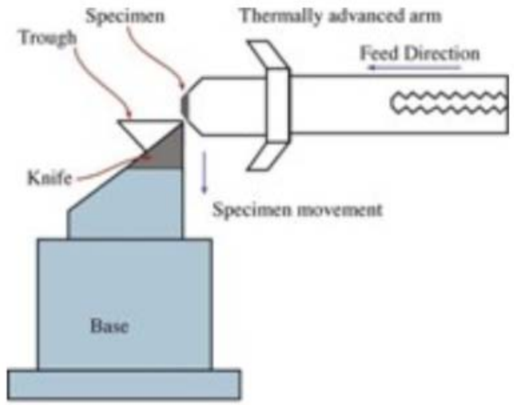
Ultramicrotomy

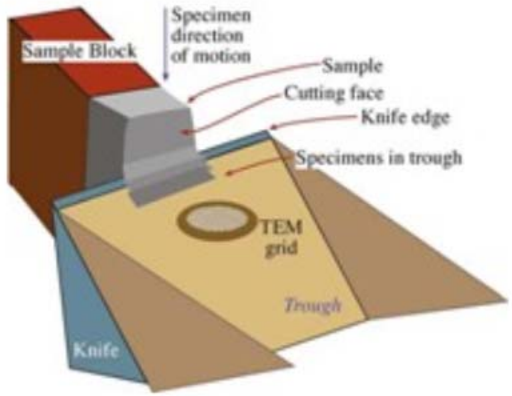


Ericson and Lindberg, 1997

Ultramicrotomy used for preparation of TEM samples

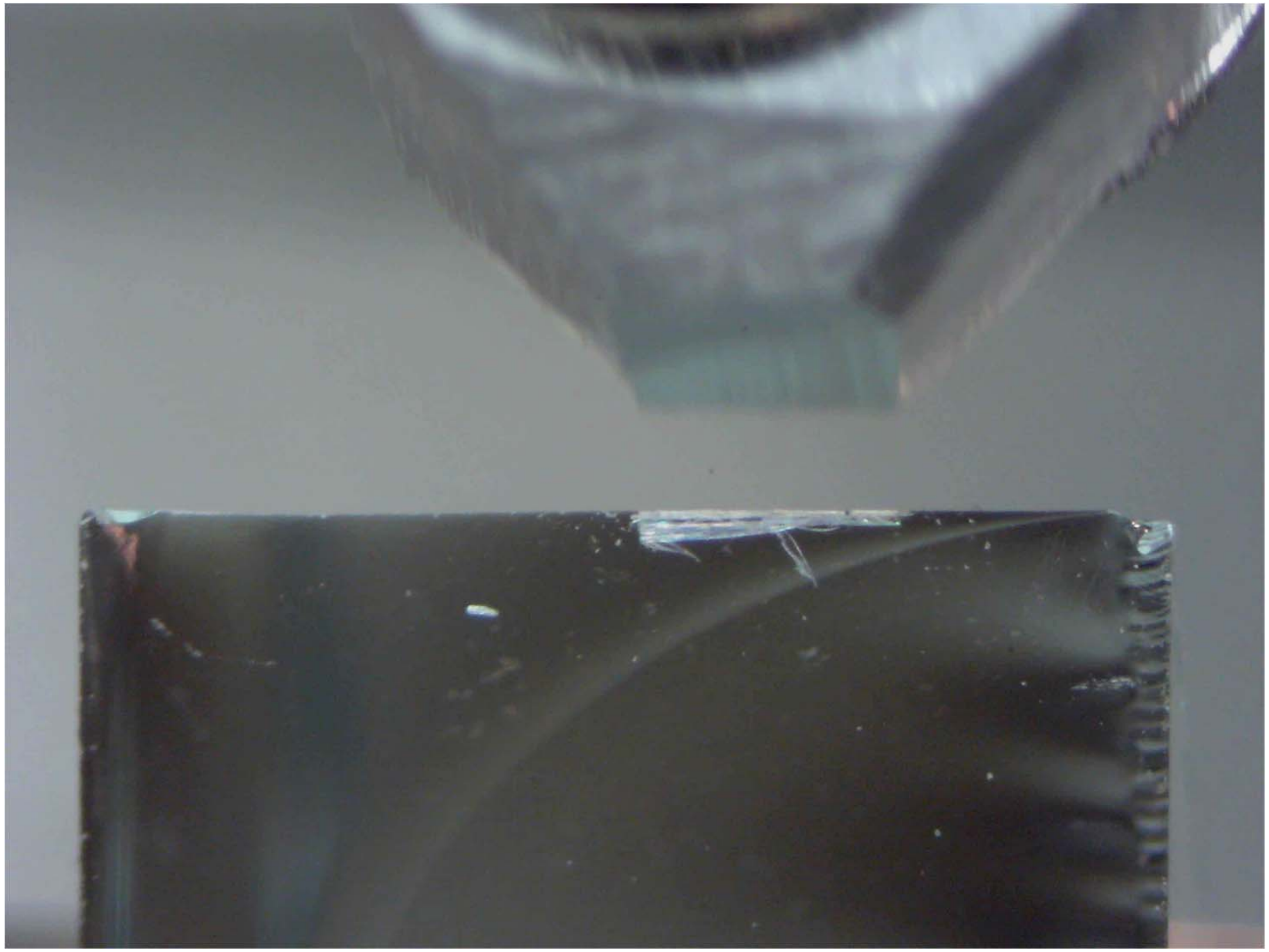




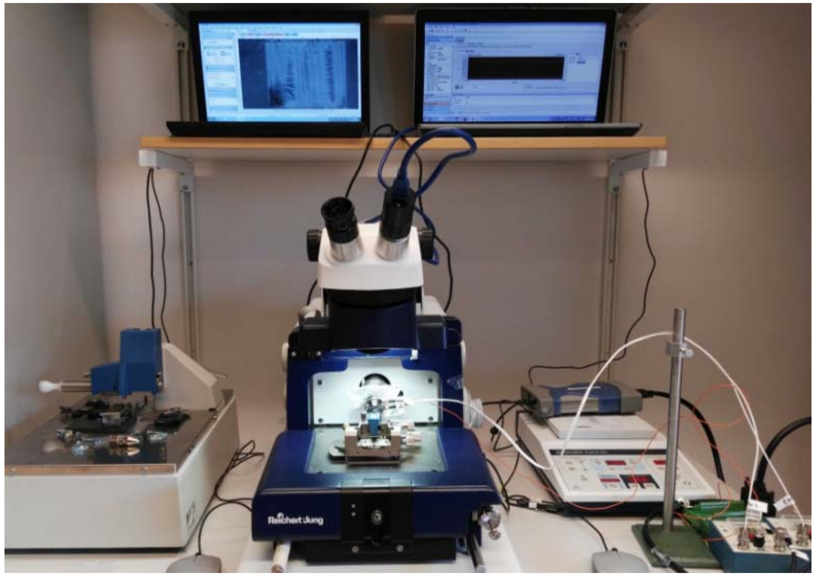


Chips with a thickness of down to ~30 nm

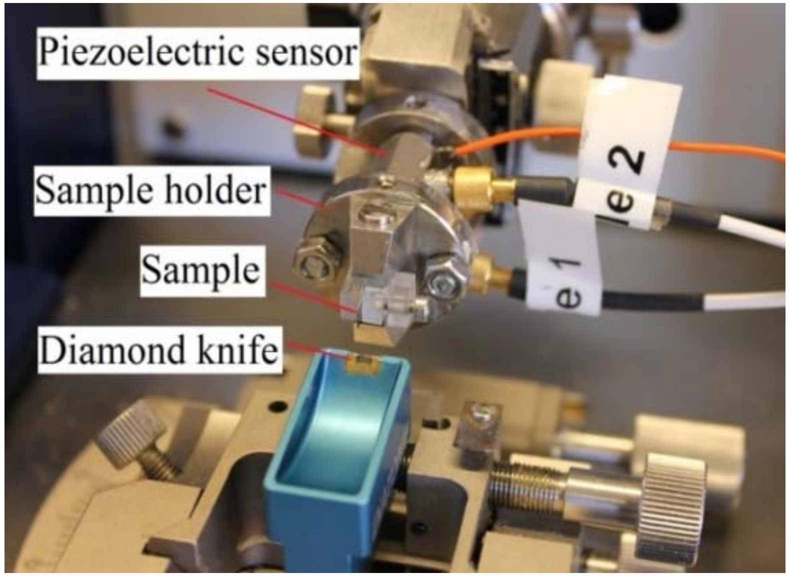
Sequential cutting of ultrathin chips in the ultramicrotome



Instrumented ultramicrotome

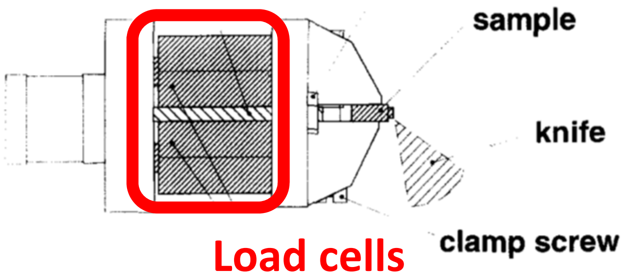


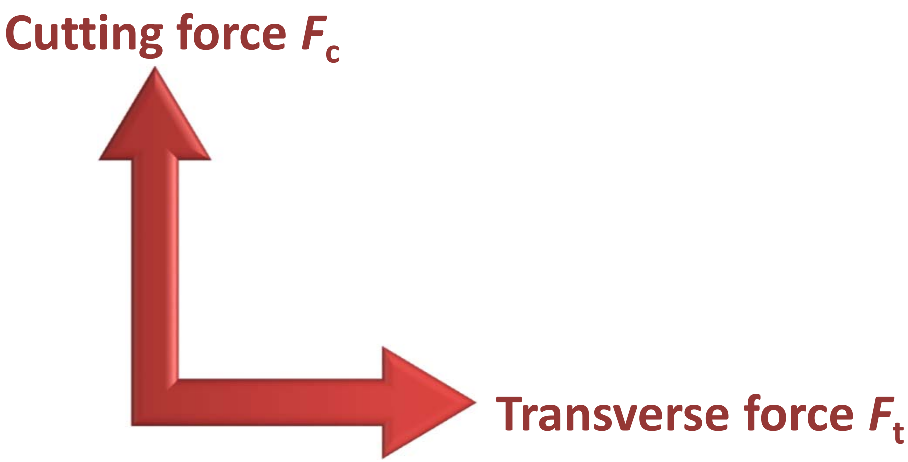
Sample holder



(*Ericsson & Lindberg, 1997*)

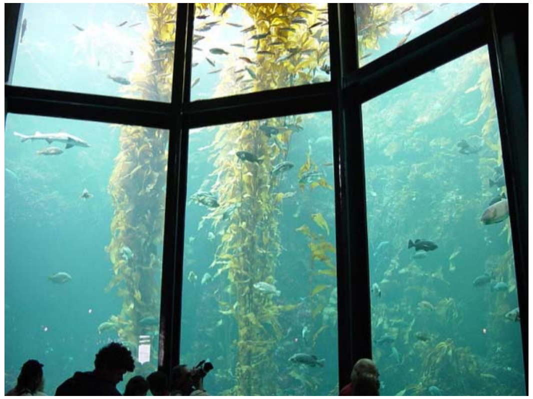
Sectioning thickness: 30-200 nm
Sectioning speed: 0.25-20 mm/s
Force sensor resolution: < 1 mN
Diamond knife: edge radius ~7 nm





Model material: Poly(methyl methacrylate), PMMA, acrylic glass or Plexiglas, a transparent amorphous glassy thermoplastic

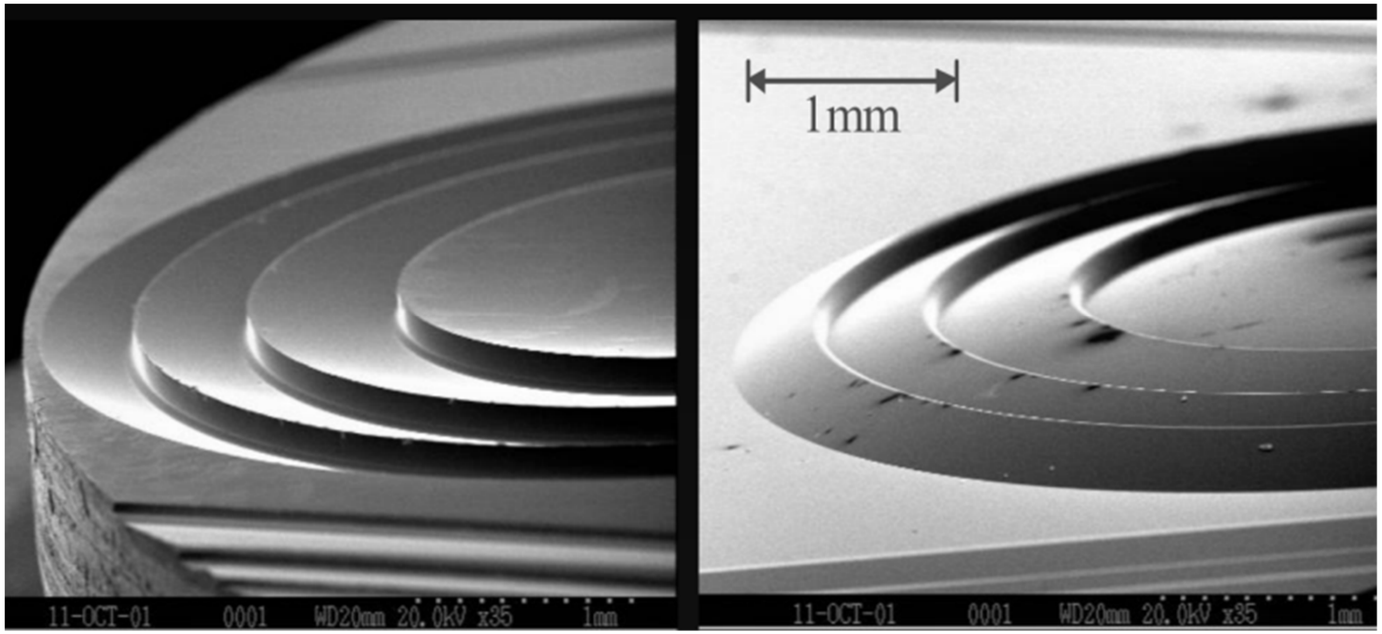





PMMA

Increasing demands on micro/nano manufacturing (miniaturisation in optics, electronics)

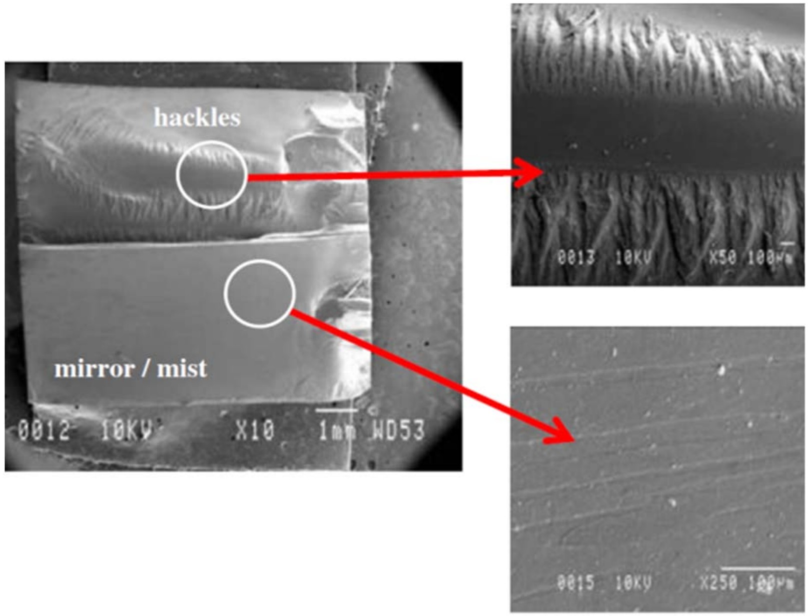
Diamond machined mould *Moulded PMMA optic*



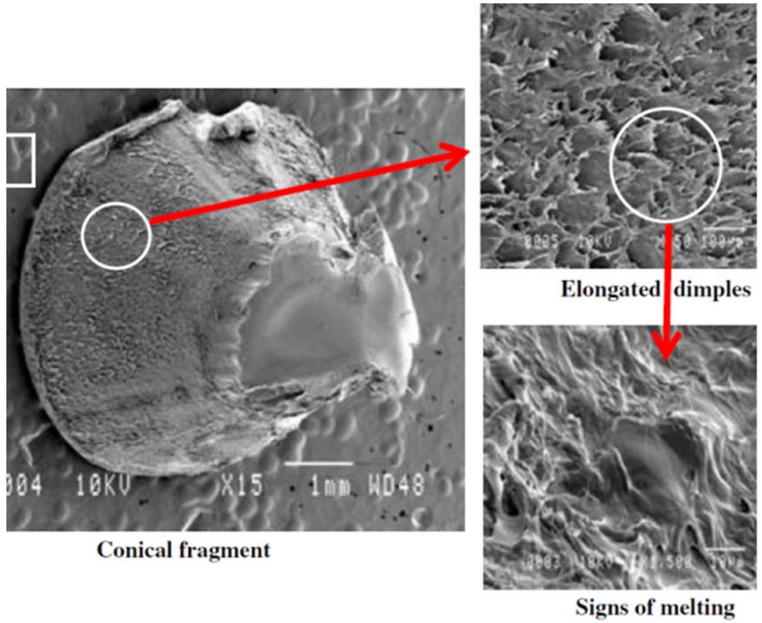
(*Gill & Dow*)

PMMA can show a complex deformation and fracture behaviour

Brittle failure of PMMA



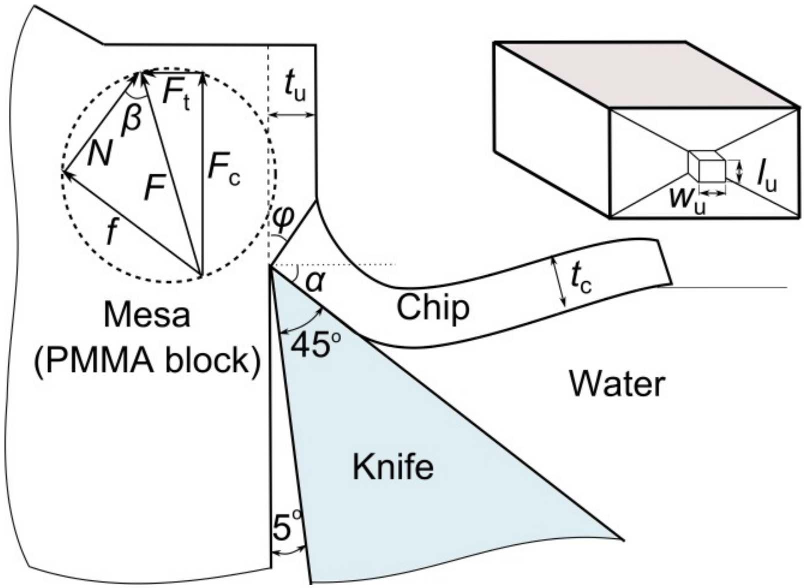
Ductile failure of PMMA



(Rittel & Brill, 2008)

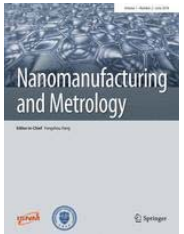
Instrumented ultramicrotomy:

Well-controlled cutting of PMMA on the nanoscale



Why?

- Measuring fracture toughness
- Investigate deformation and fracture mechanisms
- Propose predictive model
- Optimise processing conditions



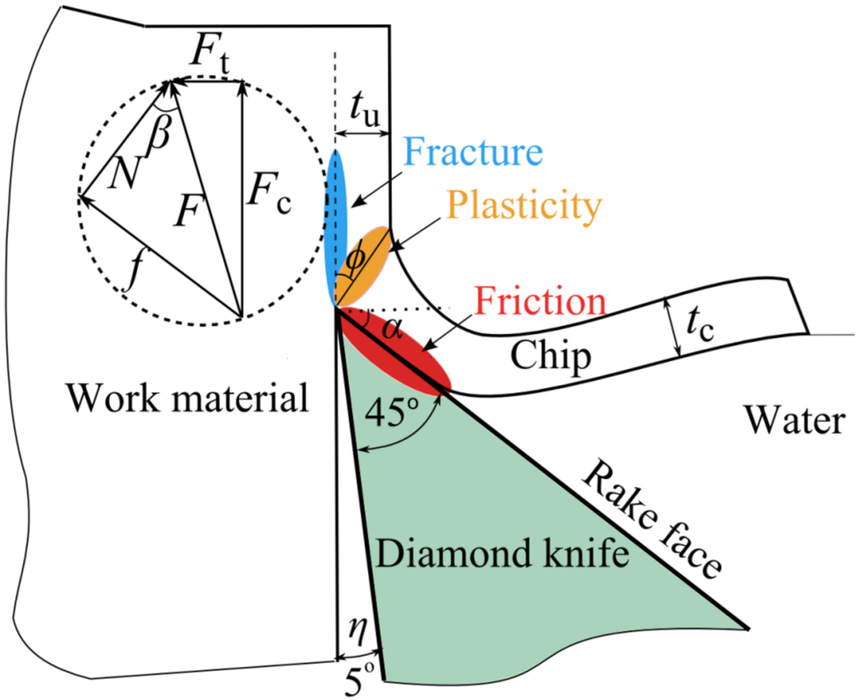
Review on nanocutting: Fang & Xu, *Nanomanufacturing and Metrology* **1** (2018), 4

Fracture toughness & Yield stress

Energy balance (*Atkins*, 2003)

external work =

plasticity+friction+fracture



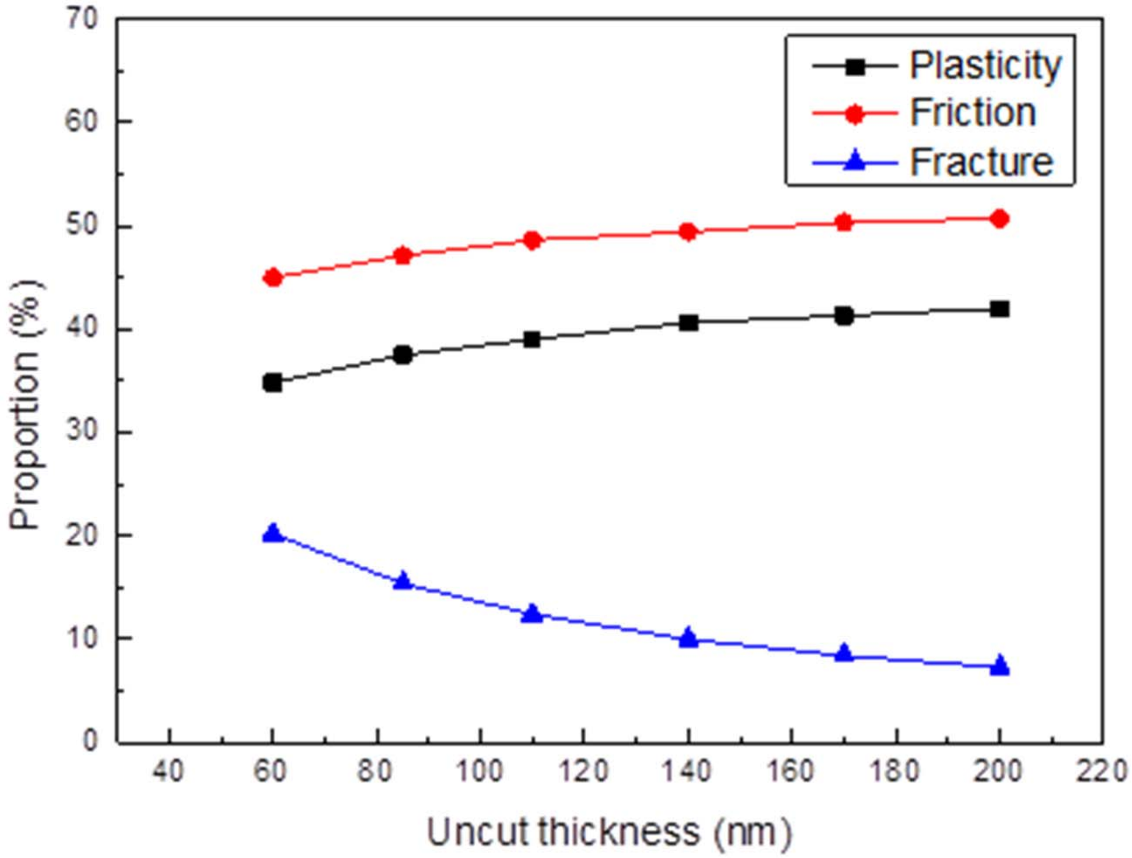
shear yield stress
MPa

$$F_c v_0 = (\tau_y \gamma)(t_u w v_0) + \left[F_c \sec(\beta - \alpha)\sin\beta\right]\frac{v_0 \sin\phi}{\cos(\phi - \alpha)} + R w v_0$$

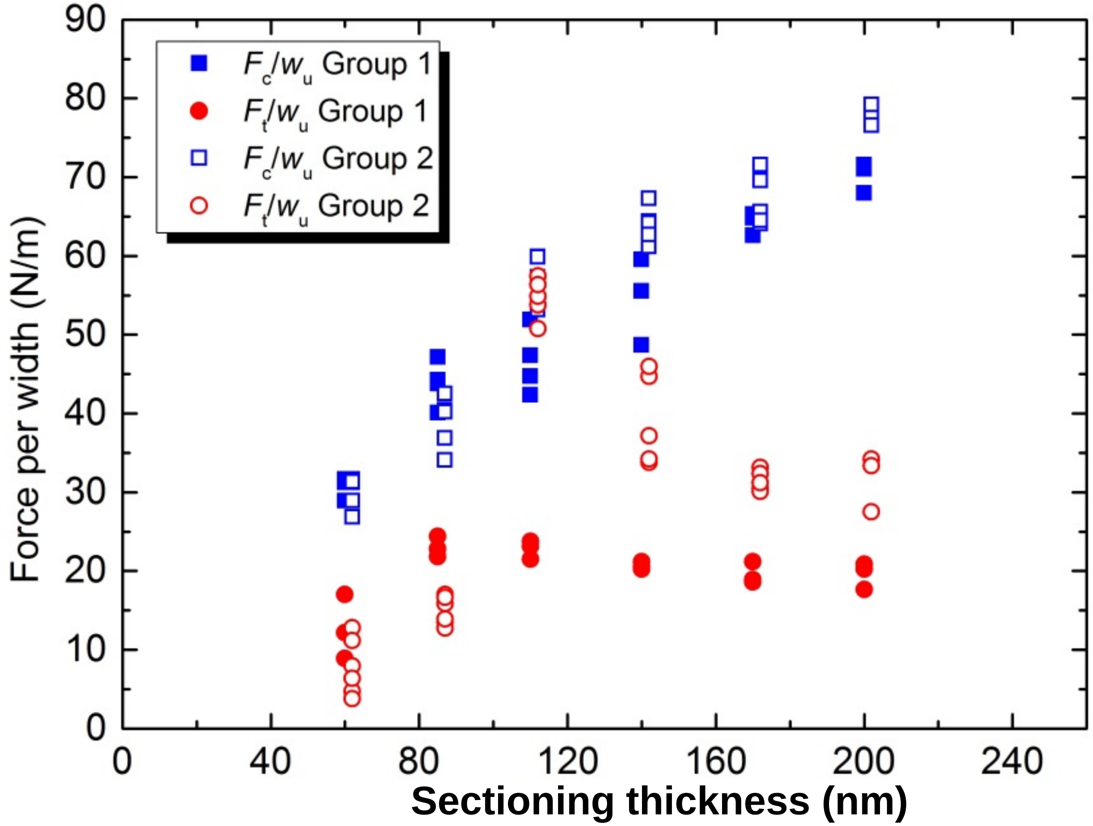
fracture energy
J/m^2

$$\frac{F_c}{w} = \left(\frac{\tau_y \gamma}{Q}\right) t_u + \frac{R}{Q}$$

Estimated division of dissipation mechanisms



Fracture toughness & Yield stress



Fracture energy *R*: ~10 J/m^2

Shear yield stress τ_y : ~110 MPa

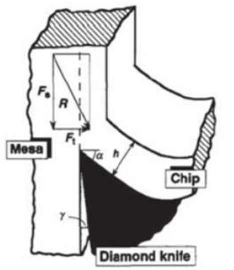
G_C=500-1000 J/m^2 at macroscopic

Close to *Patel et al*. (2009)

Scaling effect in polymer fracture

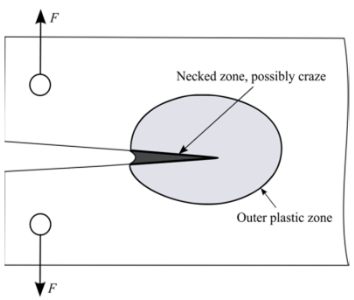
Intrinsic fracture energy
Breaking covalent bonds

1 J/m^2



Nanocutting
Fracture energy

10 J/m^2

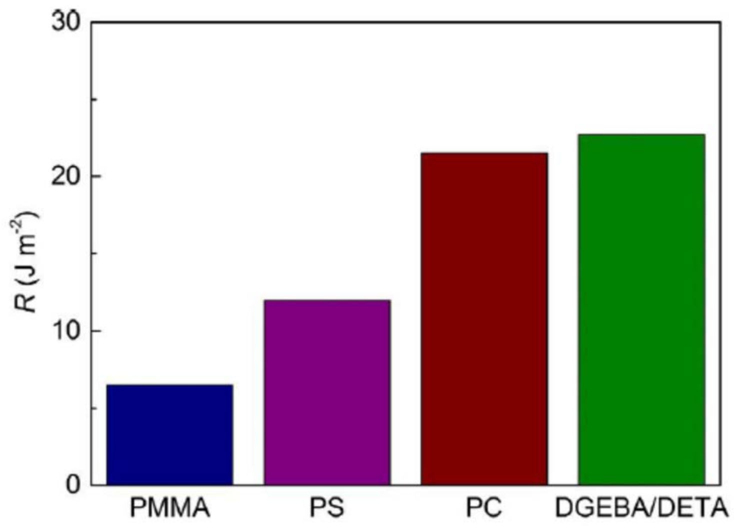


Macroscopic fracture toughness
Critical energy release rate

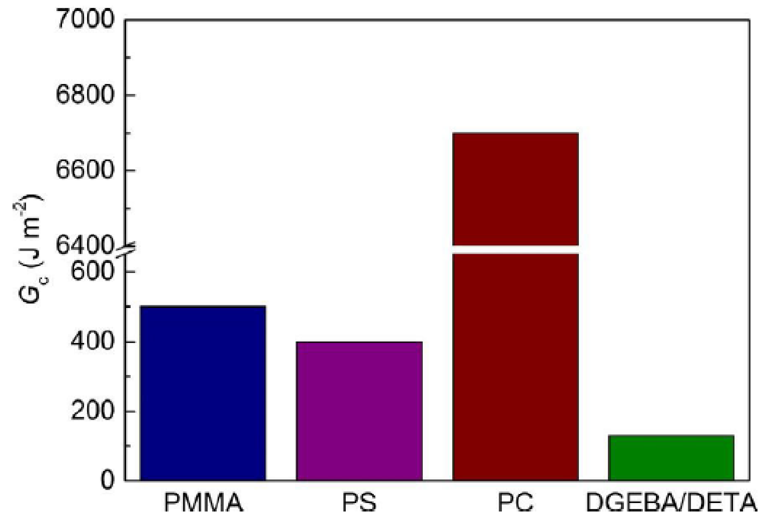
1000 J/m^2

Nano-macro scale difference

Nanoscale



Macroscale

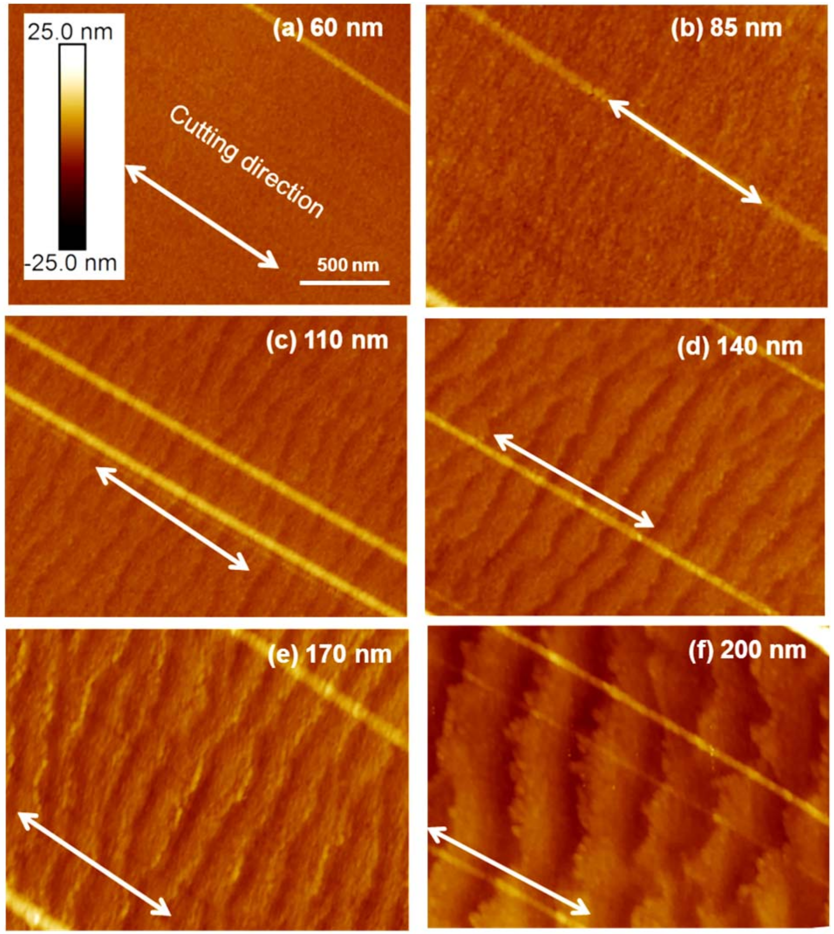


100× 100× 1000× 10×

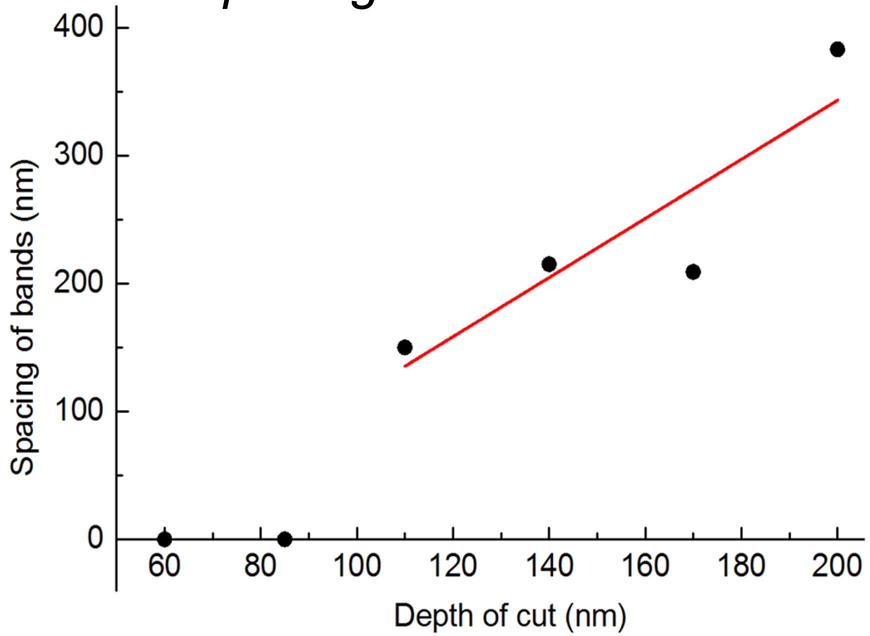
(Sun et al., 2017; Ericson & Lindberg, 1996, 1997; Kausch, 1987)

Characteristics of sectioned surfaces

AFM Morphologies, v_0= 1.0 mm/s



Spacing between bands

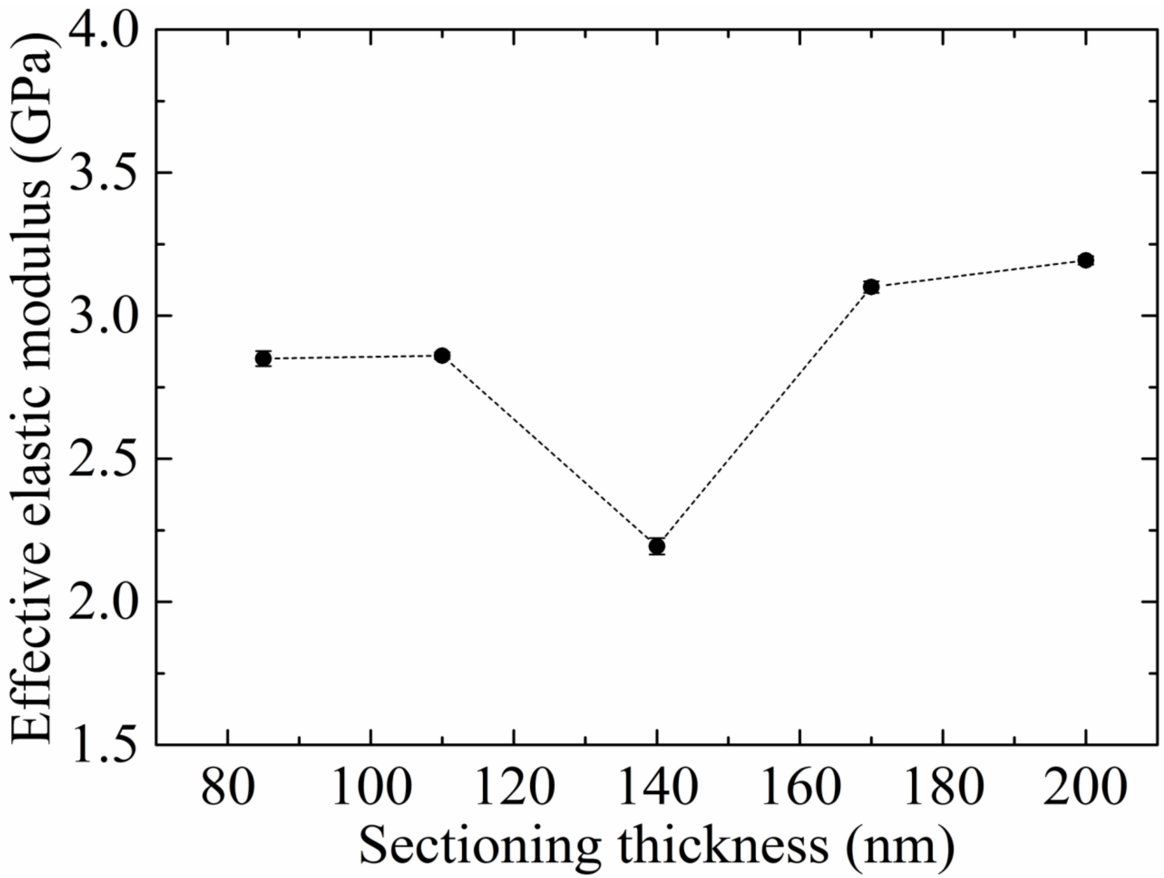


60 nm: flat and smooth

85 nm: feeble structures

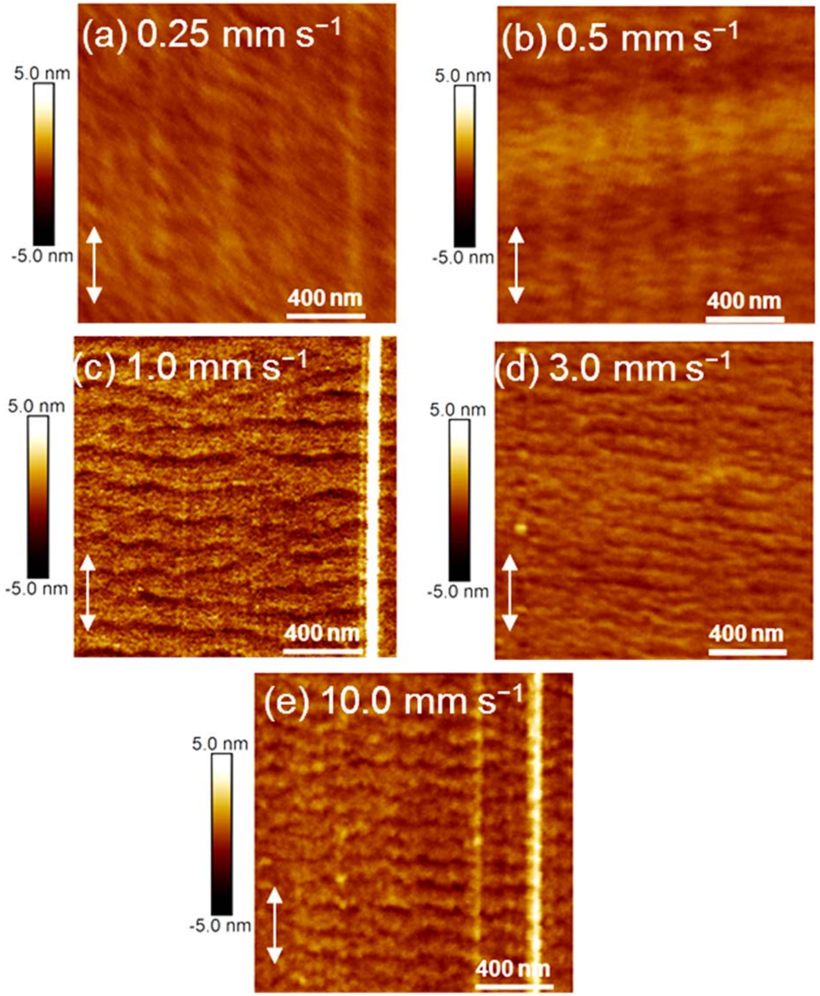
110-200 nm: periodicity

Surface stiffness

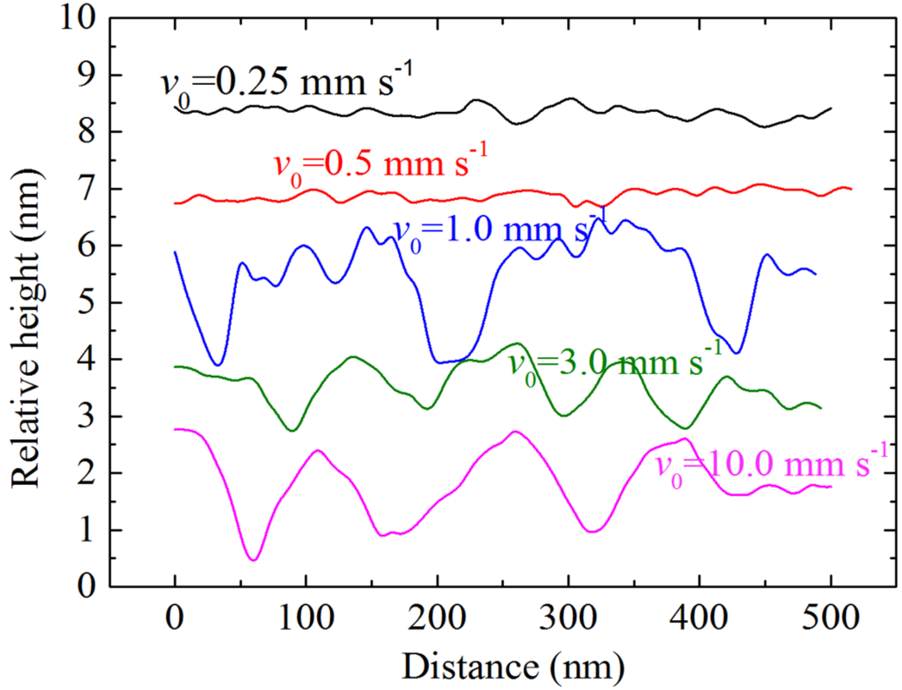


Characteristics of sectioned surfaces

AFM Morphologies, t_u=85 nm



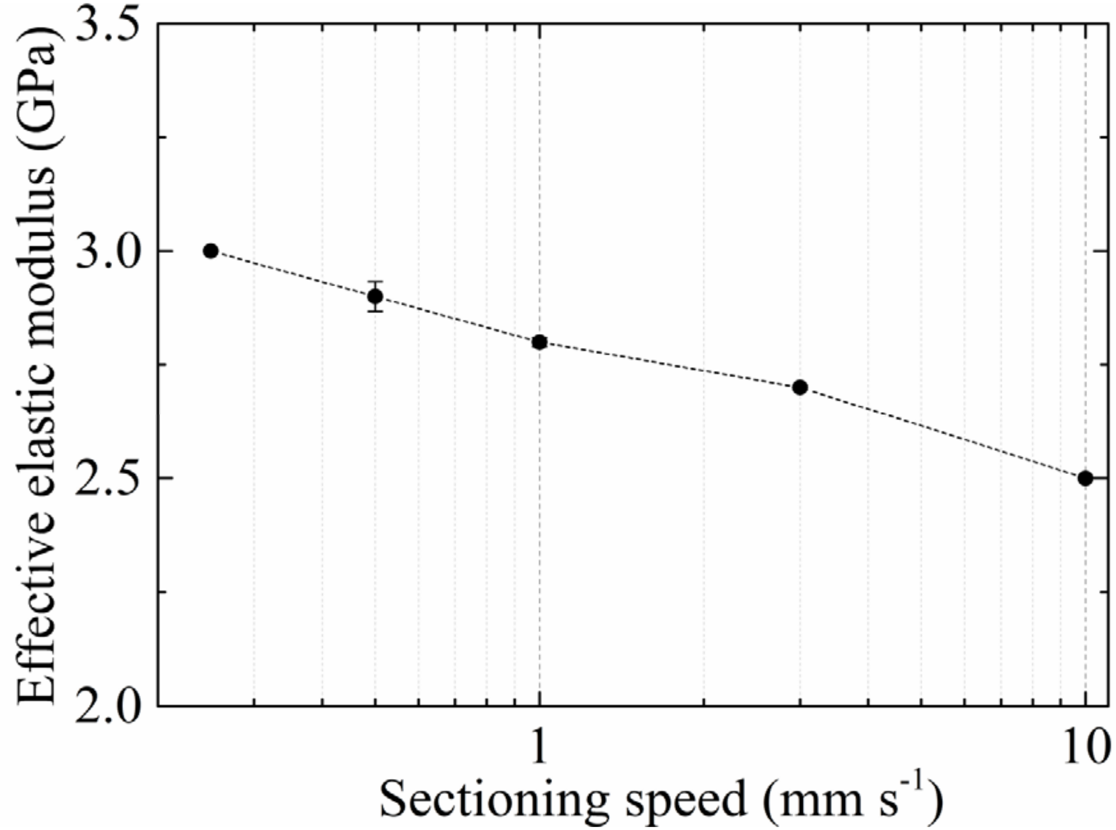
Height along sectioning



0.25, 0.5 mm/s: flat surfaces

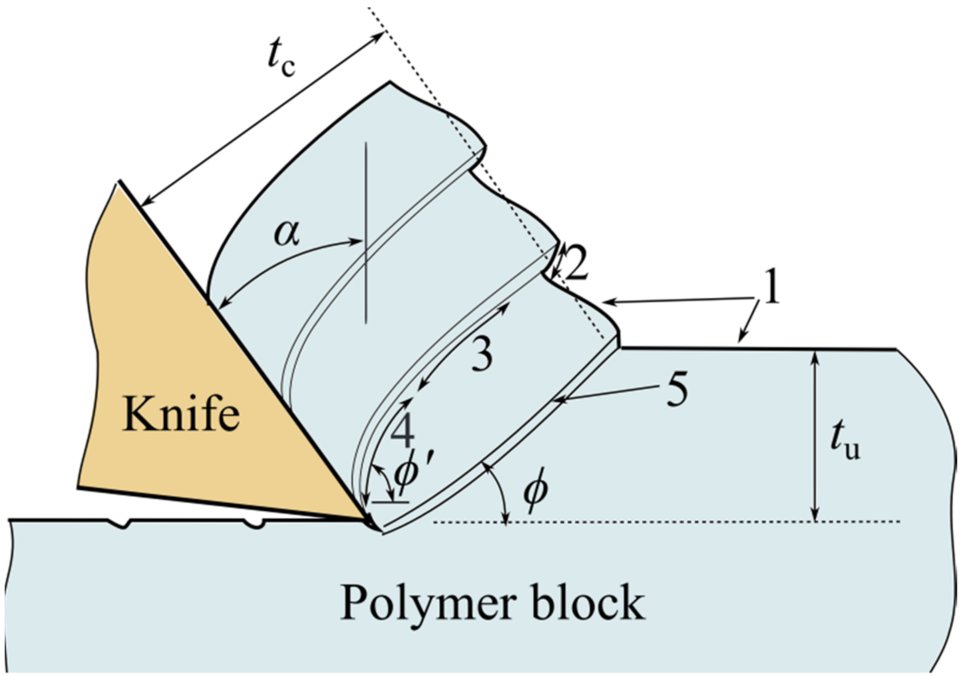
1.0-10.0 mm/s: periodicity

Surface stiffness



Hypothesis: shear banding

Critical sectioning conditions for the onset of periodic structures, a typical feature of shear banding in sectioning



(After *Komanduri & Hou*, 2002; *Turley et al*. 1982)

Heat generation

⇩

Heat localisation

⇩

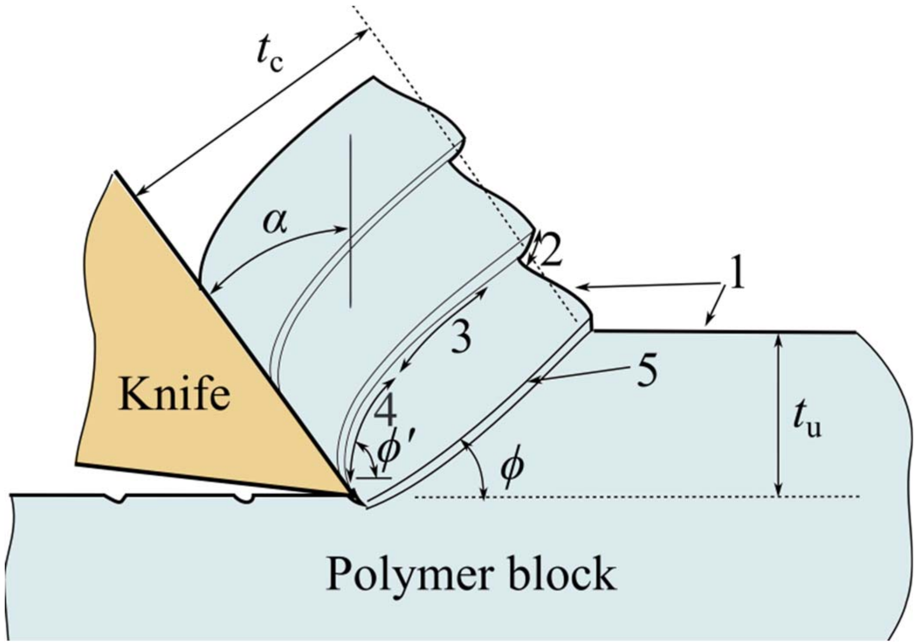
Thermal softening

⇩

Shear band

Several contributing heat sources affect shear yielding

Critical cutting conditions exist to form periodic structures on the surfaces, a typical feature of shear banding in cutting

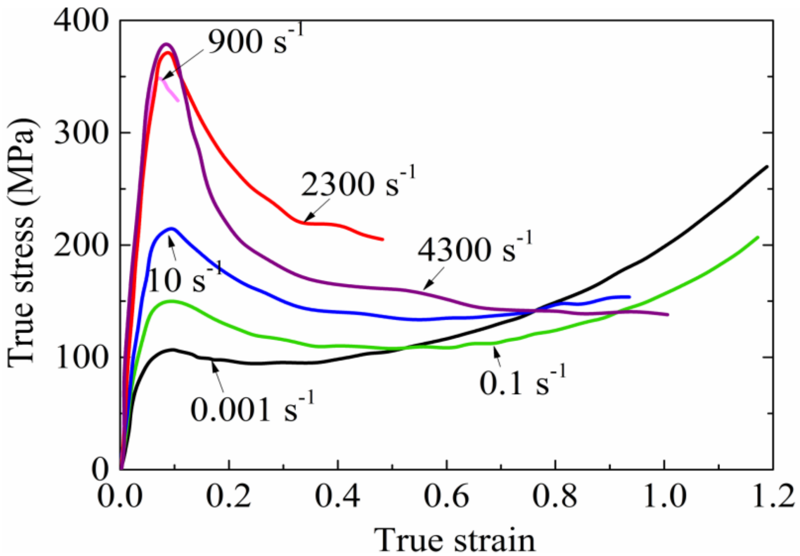


Schematic of the shear banding during cutting.
(after *Komanduri & Hou*, 2002; *Turley et al*. 1982)

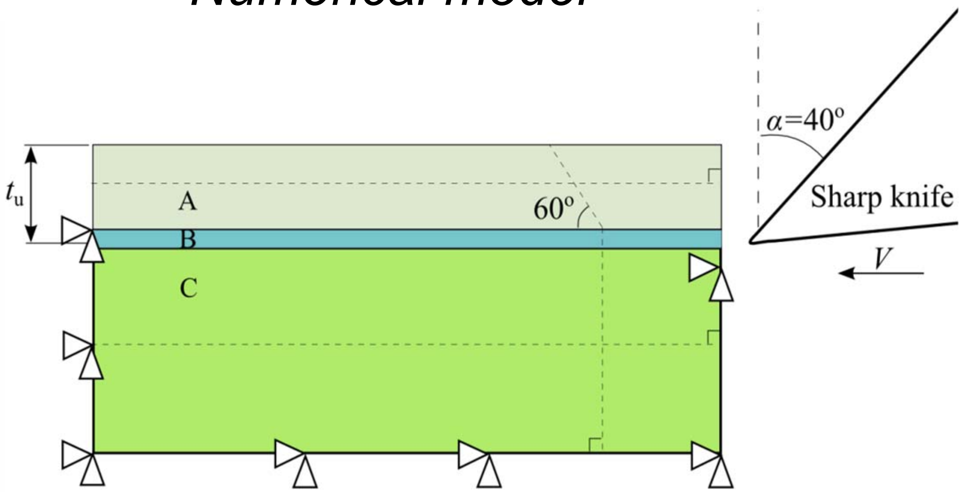
1. undeformed surfaces
2. yielded surface due to localised shear,
3. shear band
4. shear band segment near knife edge
5. localised deformation in the shear plane emanating from the knife tip.

FE modelling

σ-ε for PMMA, Richeton (2005)



Numerical model



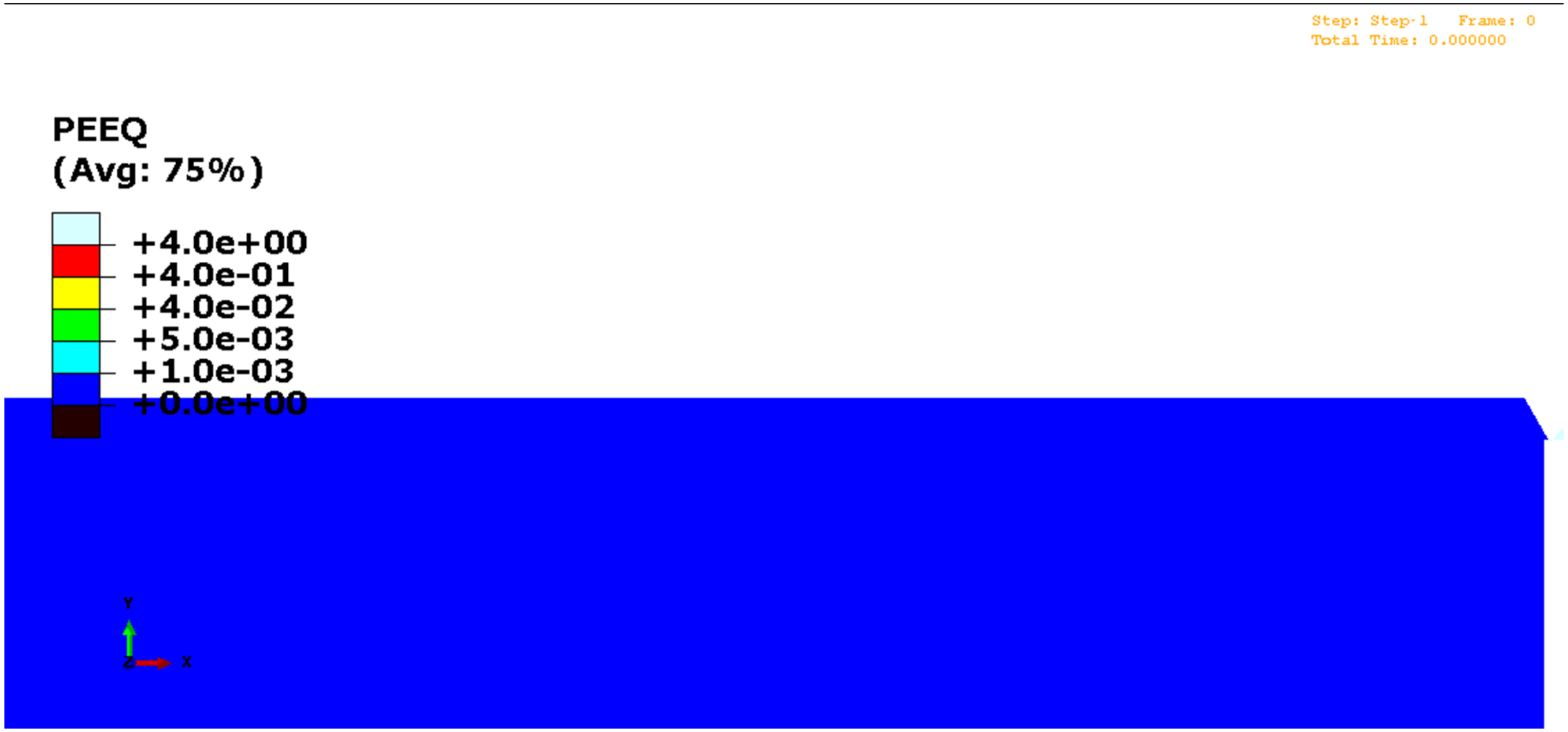
Abaqus/Explicit

Thickness: 85 nm, speed: 1.0 mm/s

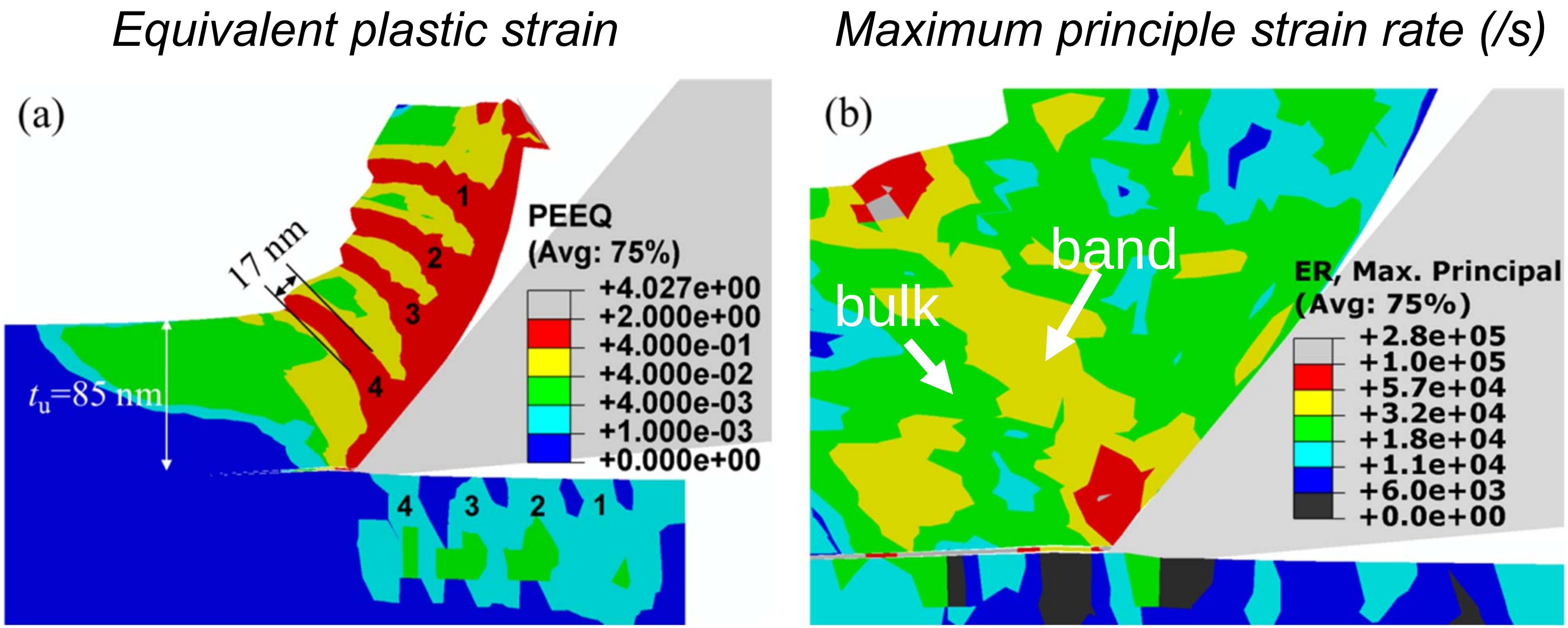
Yield & post-yield of PMMA: *Richeton* (2005) in A, B, C

Fracture model: strain based failure criterion in B

FE modelling



FE modelling

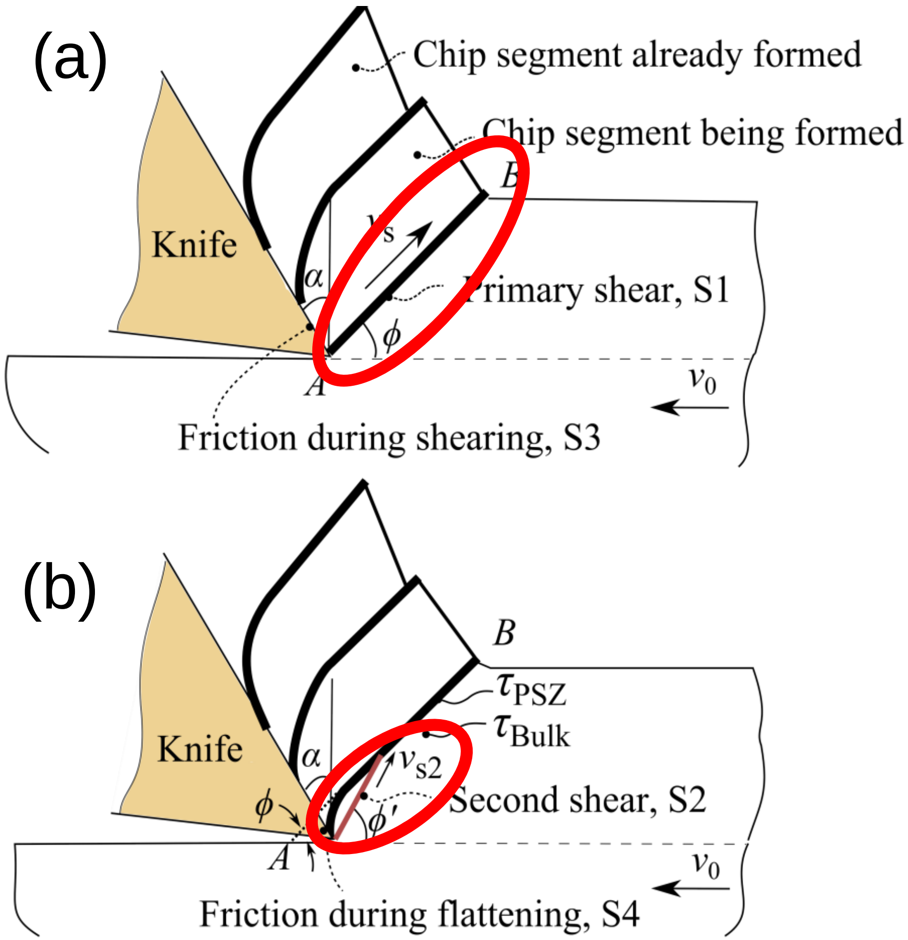


Band width = 1/5 of sectioning thickness

Strain rate in bulk = 1/3-1/2 of the rate in the band

(consistent with *Özel, 2006; Childs, 2013*)

Adiabatic shear modelling



Adiabatic shearing model by Komanduri & Hou (2002)

Onset criterion : $\tau_{PSZ} \leq \tau_{Bulk}$

Constitutive law for PMMA:

$$\tau_{PSZ} \ \& \ \tau_{Bulk} = f(T, \dot{\gamma})$$

$$\sigma_y = \sigma_i(0) - mT + \frac{2k_B T}{V_a} \sinh^{-1}\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0 \exp(-\Delta H_\beta / k_B T)}\right)^{1/n}$$

Richeton et al. (2005)

Strain rate:

$$\dot{\gamma}_{PSZ} = \frac{v_s}{\Delta h}$$

(from FEA)

$$\dot{\gamma}_{Bulk} = \dot{\gamma}_{PSZ} / 3$$

Adiabatic shear modelling

Temperature in primary shear zone:

Heat S1-S4 + Preheating due to S1-S4

e.g. Heat S1

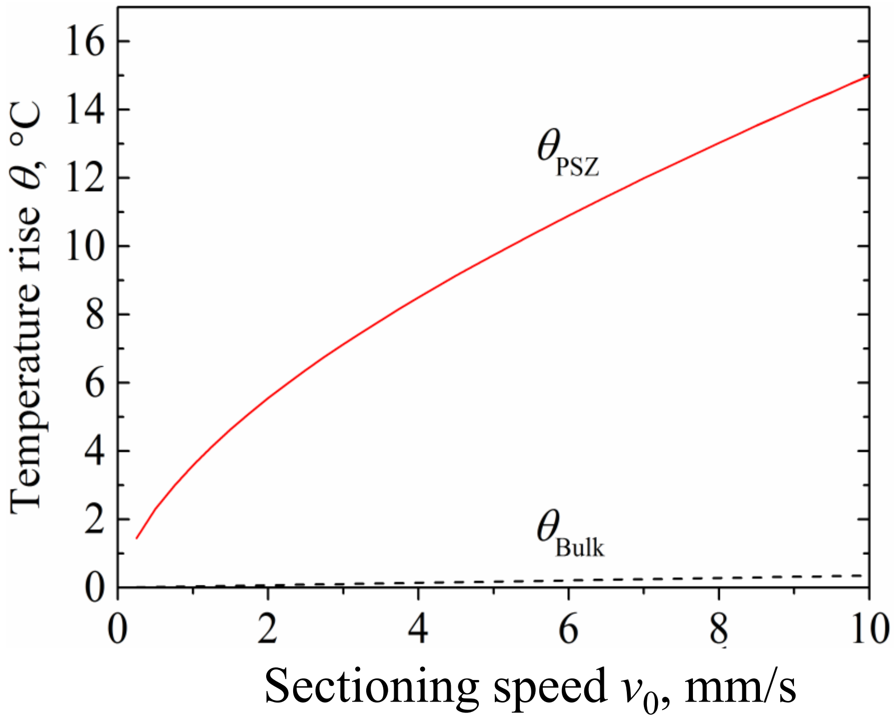
$$\theta_M = \frac{q_1}{2\pi\lambda}\left(1 - \frac{t}{2t_0}\right)\int_0^{l_s} \Omega(p)\mathrm{d}y_i + \frac{q_0}{16\pi\lambda\alpha_t t_0}\int_0^{l_s} r_i^2\,\chi(p)\mathrm{d}y_i$$

Temperature in bulk:

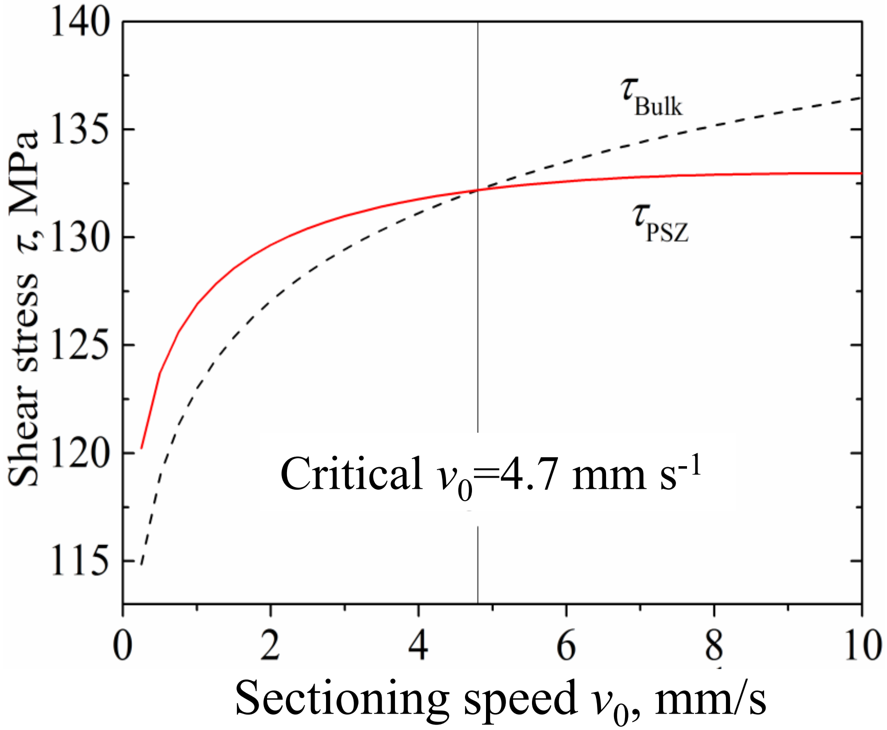
Preheating due to S1-S4

Adiabatic shear modelling

Temperature rise



Shear stress

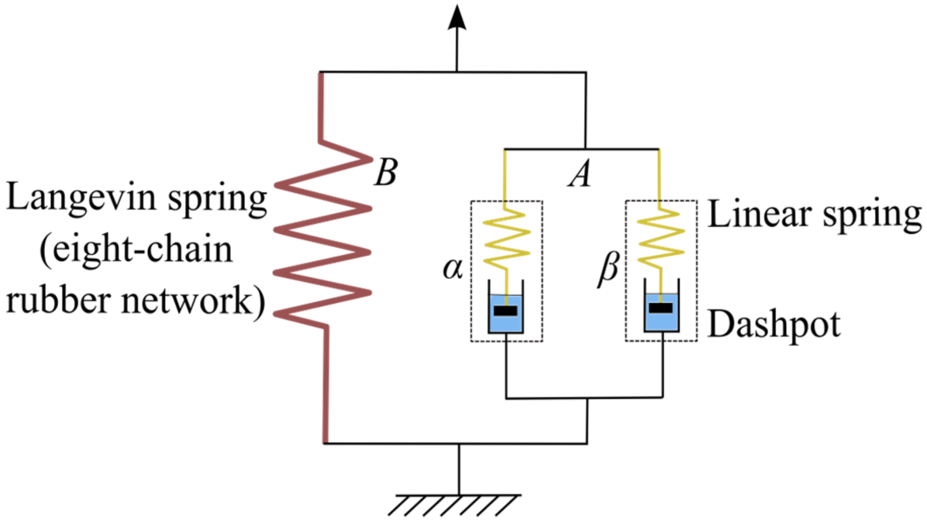


The estimated speed for the onset of shear bands agrees with the experiment

FE modelling of PMMA sectioning

Elastic viscoplastic model

Mulliken-Boyce model



Total stress :

$$\mathbf{T} = \mathbf{T}_\alpha + \mathbf{T}_\beta + \mathbf{T}_\mathrm{B}$$

Cauchy stresses in *α* and *β*

$$\mathbf{T}_\alpha = \frac{1}{J_\alpha} \mathsf{L}_\alpha^\mathrm{e} \left[\ln \mathbf{V}_\alpha^\mathrm{e} \right], \quad \mathbf{T}_\beta = \ldots$$

Back stress in B

$$\mathbf{T}_\mathrm{B} = \frac{C_\mathrm{R}}{3} \frac{\sqrt{N}}{\lambda_\mathrm{chain}^\mathrm{p}} \mathsf{L}^{-1}\left(\frac{\lambda_\mathrm{chain}^\mathrm{p}}{\sqrt{N}} \right) \overline{\mathbf{B}}'_\mathrm{B}$$

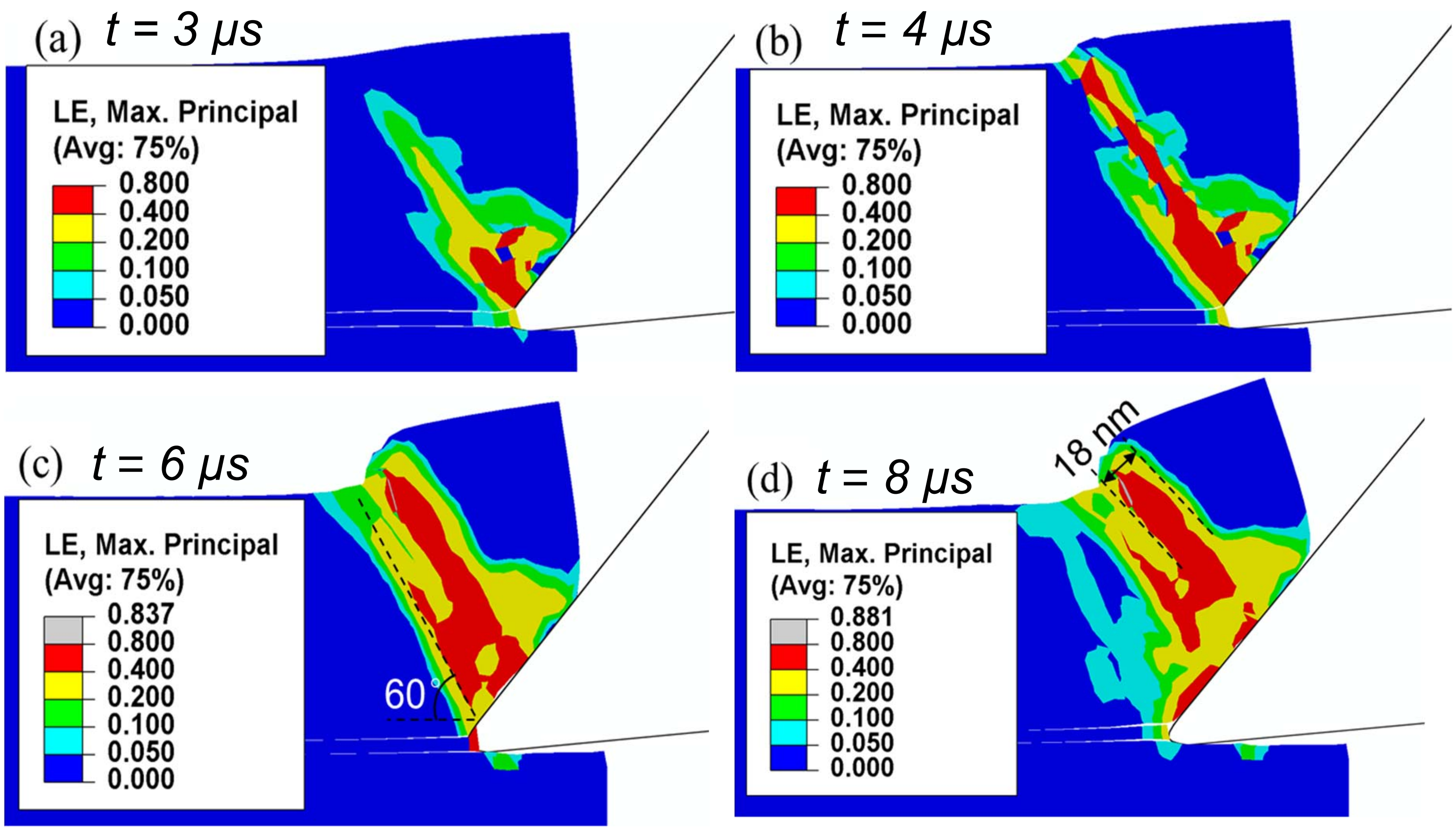
Plastic flow in *α* and *β*

$$\dot{\gamma}_\alpha^\mathrm{p} = \dot{\gamma}_{0,\alpha}^\mathrm{p} \exp\left[-\frac{\Delta G_\alpha}{k\theta} \left(1 - \frac{\tau_\alpha}{t_\alpha \hat{s}_\alpha + \alpha_{\mathrm{p},\alpha} p} \right) \right], \quad \dot{\gamma}_\beta^\mathrm{p} = \ldots$$

Adiabatic heating

$$\dot{\theta} = \frac{1}{\rho c} \left[\mathrm{tr}(\mathbf{T}_\alpha \widetilde{\mathbf{D}}_\alpha^\mathrm{p}) + \mathrm{tr}(\mathbf{T}_\beta \widetilde{\mathbf{D}}_\beta^\mathrm{p}) \right]$$

FEA results



Periodic structures are reproduced

Recapitulation so far

- The fracture energy of PMMA is as low as ~10 J/m^2 by nanosectioning test
- Critical sectioning conditions (sectioning thickness, speed) are identified for the onset of periodic structures
- The periodic structures are attributed to localised shear deformation
- The adiabatic shear model and elastic viscoplastic model could be used to predict shear banding during sectioning

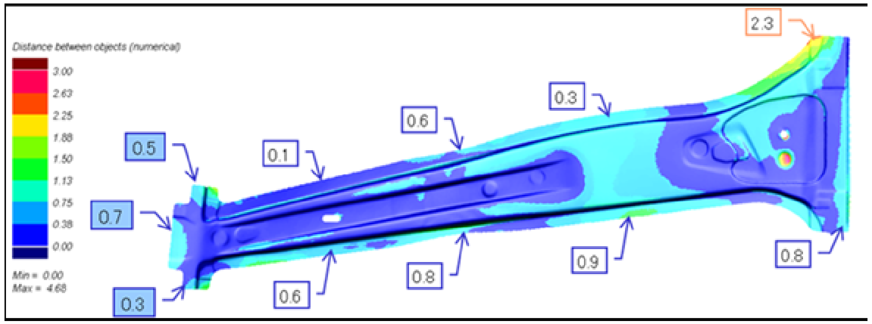
Macroscale:

Materials modelling and mechanical charactertization in metal forming

Sheet metals forming
Stamping

(Atlas Tool Inc.)

Finite element modelling
Including plasticity



(ESI Group)

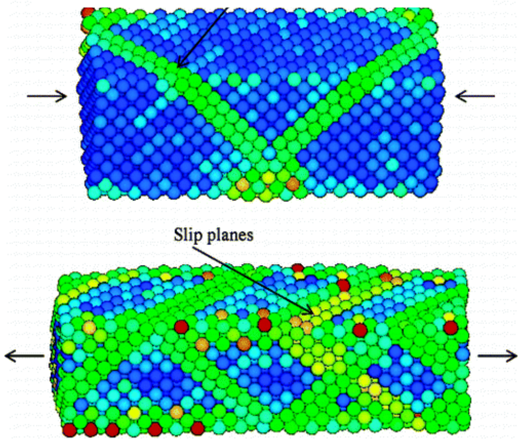
Modeling has unlocked breakthroughs in macroscale manufacturing

Nanoscale:

Materials modelling and mechanical charactertization at sub-micron scale

Metals

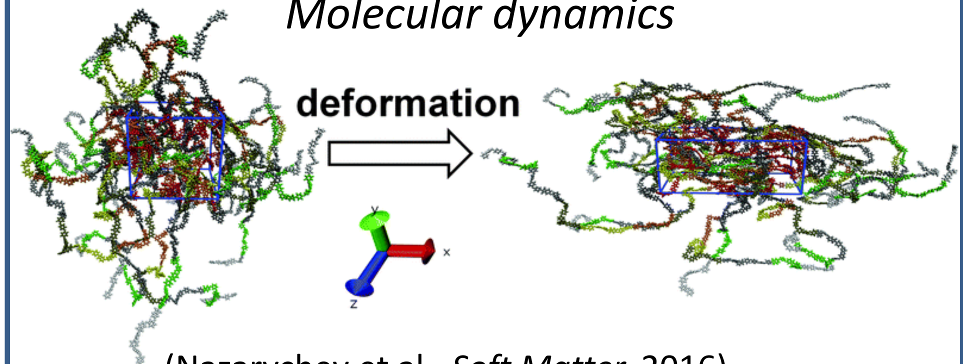
Atomistic simulations



(Diao et al., *Nano Letters,* 2004)

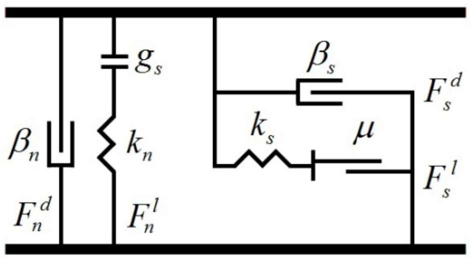
Polymers

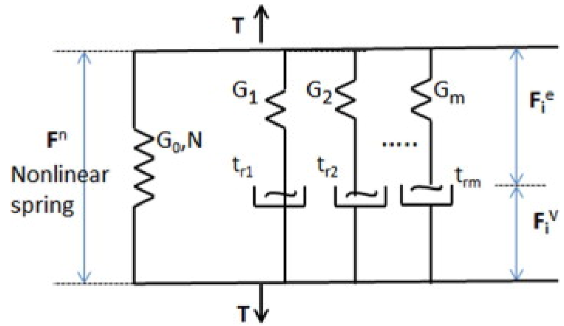
Molecular dynamics



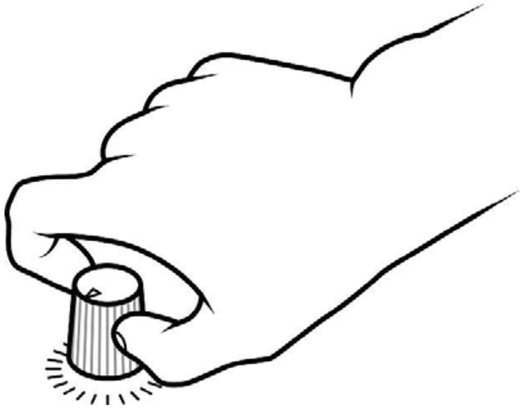
(Nazarychev et al., *Soft Matter,* 2016)

Continuum models





Model-based control of manufacturing parameters



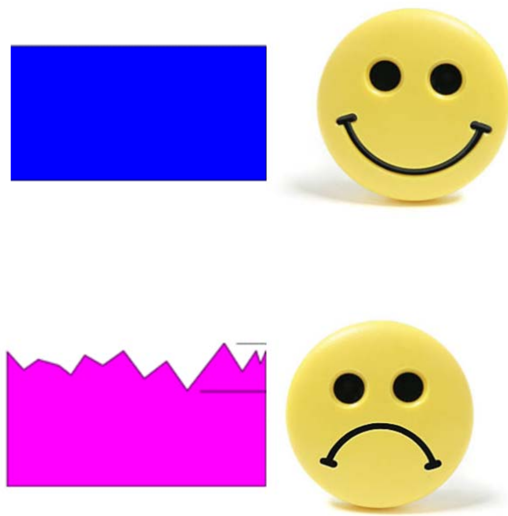
Turning the knobs sensibly…

- Feeding rate [µm/s]
- Rotational speed [rpm]
- Cutting depth [nm]
- Temperature [°C]

Smooth

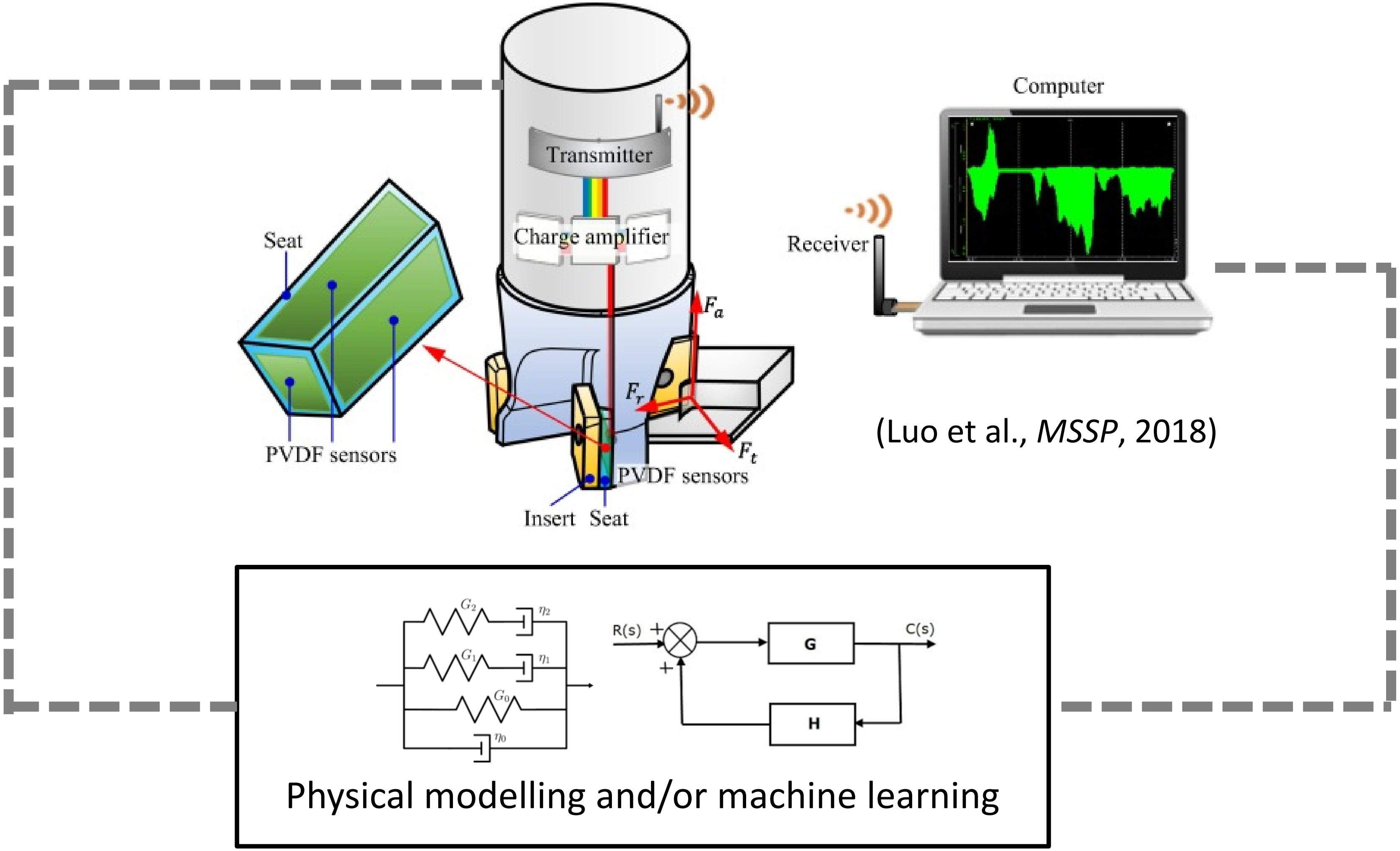
or

Rough



Future prospects:

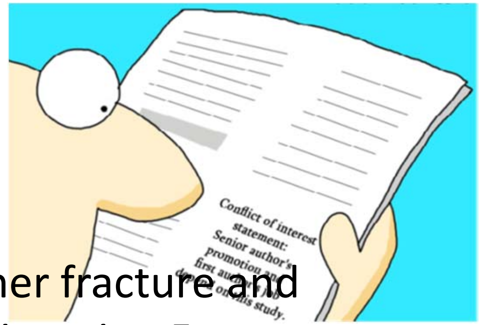
On-line monitoring of tool forces…



(Luo et al., *MSSP*, 2018)

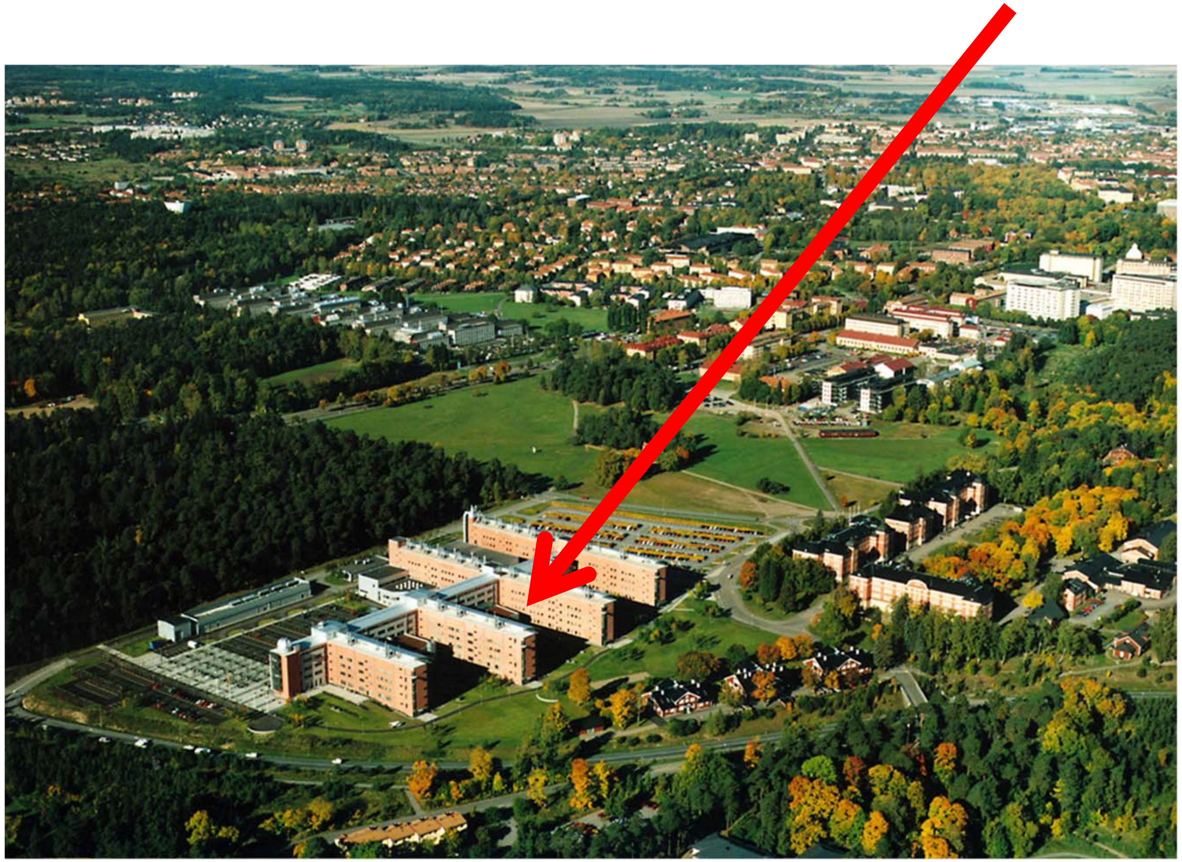
… with feedback for optimal control

Related publications:



- Sun, F., Li, H., Lindberg, H., Leifer, K. and Gamstedt, E.K., “Polymer fracture and deformation during nanosectioning in an ultramicrotome”, *Engineering Fracture Mechanics*, **182** (2017), 595-606. DOI: 10.1016/j.engfracmech.2017.05.044
- Sun, F., Li, H., Leifer, K. and Gamstedt, E.K., “Rate effects on localized shear instabilities in nanosectioning of an amorphous thermoplastic polymer”, *International Journal of Solids and Structures*, **129** (2017), 40-48. DOI: 10.1016/j.ijsolstr.2017.09.016
- Sun, F., Li, H., Leifer, K. and Gamstedt, E.K., “Effect of nanosectioning on surface features and stiffness of an amorphous glassy polymer”, *Polymer Engineering and Science*, available on-line, 2018. DOI: 10.1002/pen.24793
- Sun, F., Wiklund, U., Avilés, F. and Gamstedt, E.K., “Assessing local yield stress and fracture toughness of carbon nanotube poly(methyl methacrylate) composite by nanosectioning”, *Composites Science and Technology*, **153** (2017), 95-102. DOI: 10.1016/j.compscitech.2017.09.034
- Sun, F. and Gamstedt, E.K., “Experimental and numerical investigation on shear banding during nanomachining of an amorphous glassy polymer”, submitted to *International Journal of Mechanical Sciences*, 2018.

Welcome to the Ångström Laboratory at Uppsala University!



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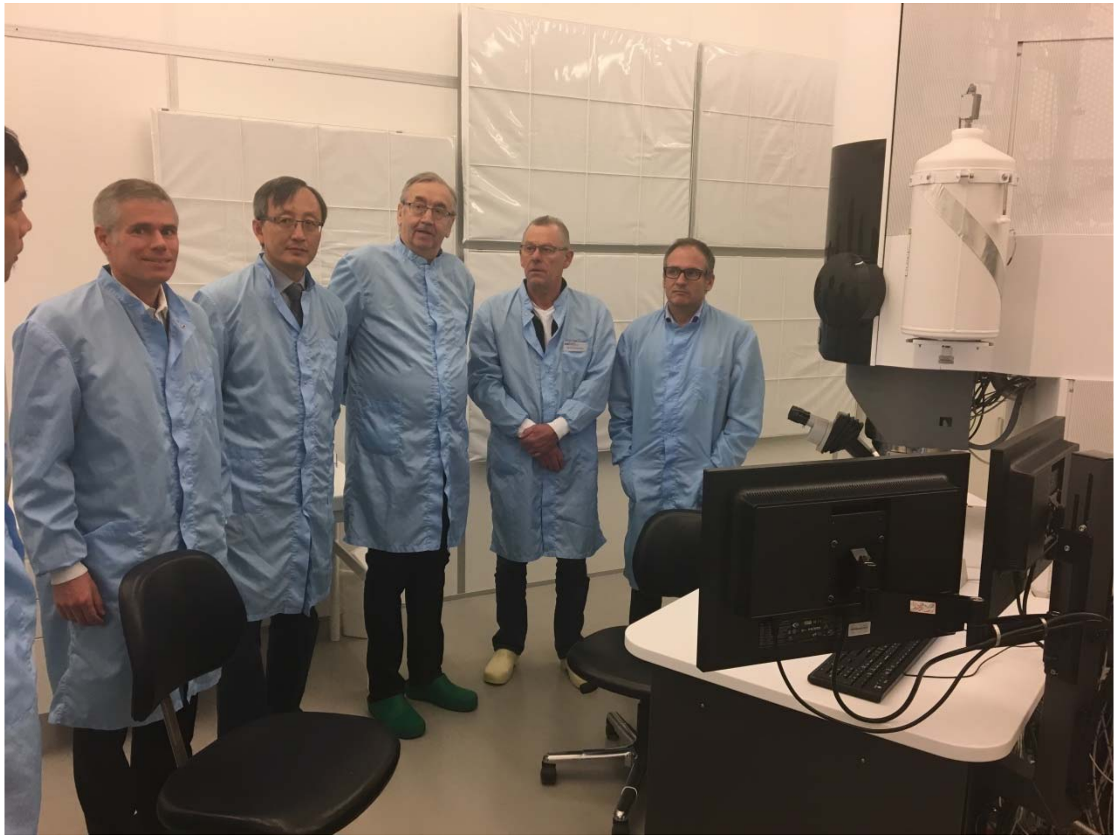
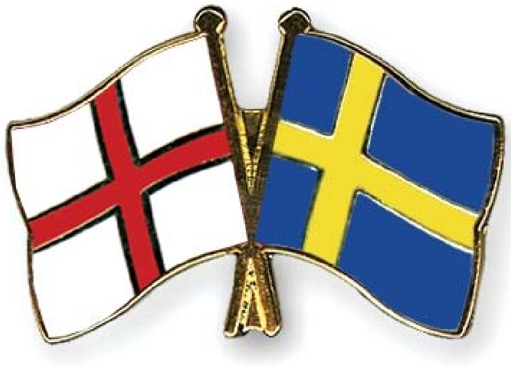
Particular thanks to Kai Cheng and his team!



Anders Ångström
Uppsala University

Visitng the Ångström Laboratory in Uppsala (2017)

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Questions or comments?