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Research Workshop on Condensed Matter Physics
30 June - 22 August 1997
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QUANTUM MONTE CARLO SIMULATIONS OF LIQUIDS AND SOLIDS
30 JUNE - 11 JULY 1997
and
CONFERENCE ON
QUANTUM SOLIDS AND POLARIZED SYSTEMS
3 - 5 JULY 1997

"Equilibrium shape and spiral growth of c-facets in 4He crystals"

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These are preliminary lecture notes, intended only for distribution to participants.

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Equilibrium Shape and Spiral Growth of c-facets in ^{4}He-crystals

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OUTLINE:

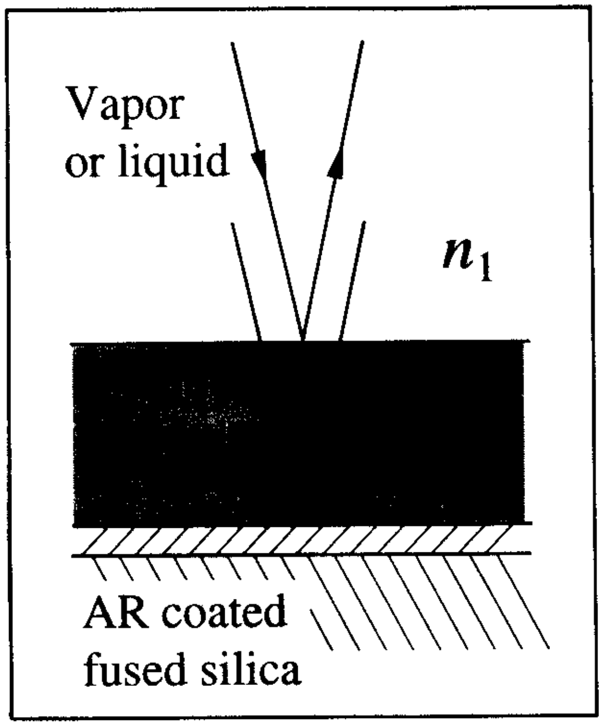
- **Introduction**
  Two beam interferometry
  How to do interferometry at mK-temperatures
- **Equilibrium crystal shape**
  Exponentially relaxing facet edges
  Reconstruction of the surface
- **Spiral growth of c-facets at low temperatures**
  Step velocities approach the speed of sound
  Modification of the classical spiral growth
- **Growth in crystals void of screw dislocations**
  How do such crystals grow?
- **Final remarks**
- **Future plans**

INTRODUCTION

- *Fizeau-type interferometry*
  - Equal thickness contours with:

$$\Delta L = \frac{\lambda}{2n} \approx 320 \text{ nm}$$

$\lambda = 632.8$ nm (He-Ne laser)
n = index of refraction



- *P.L. Marston and W.M. Fairbank (1977)*
  Superfluid meniscus in rotation: **100 nm**

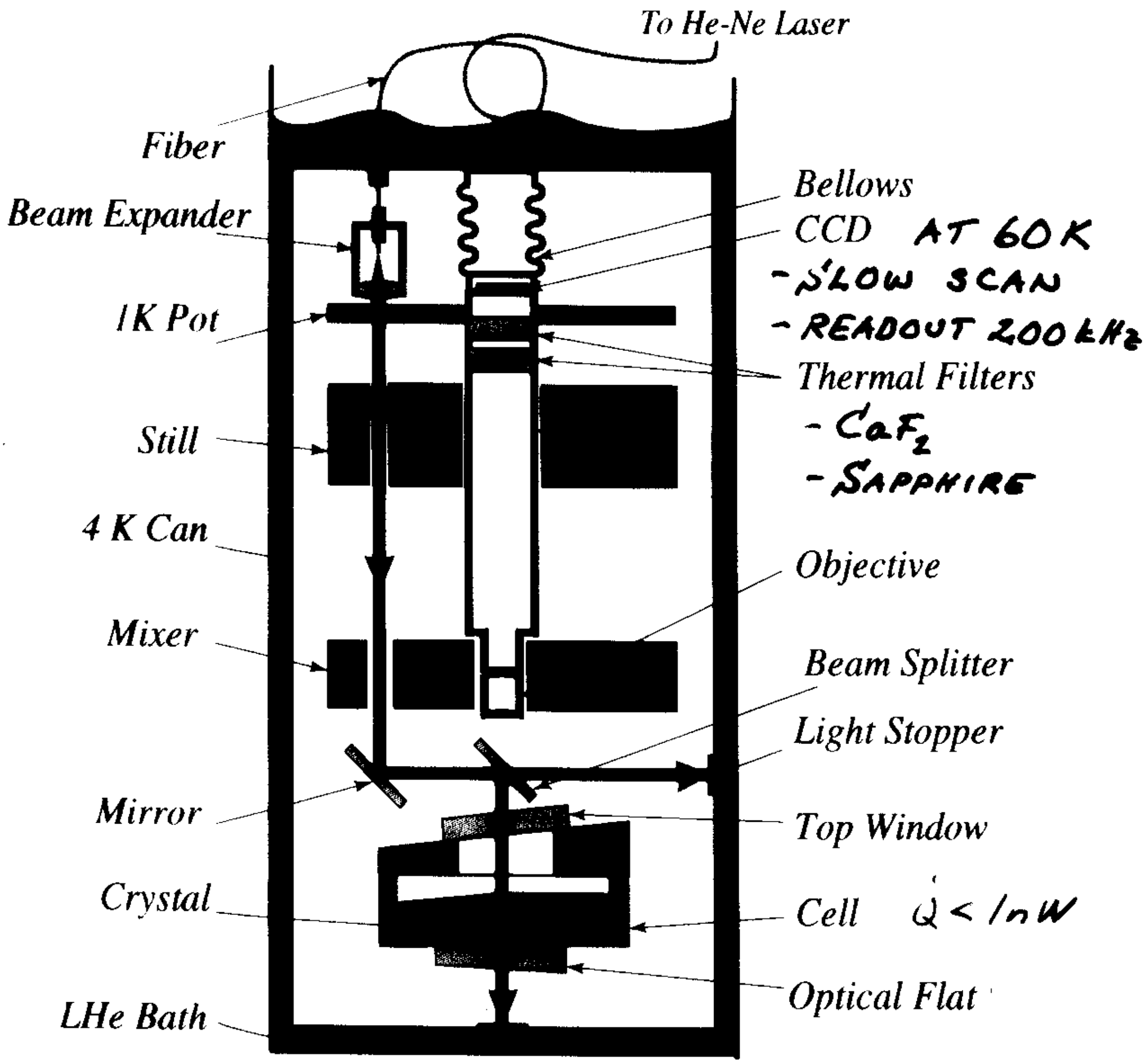
- Our work since 1992
  Resolution now better than **5 nm**

Problems:

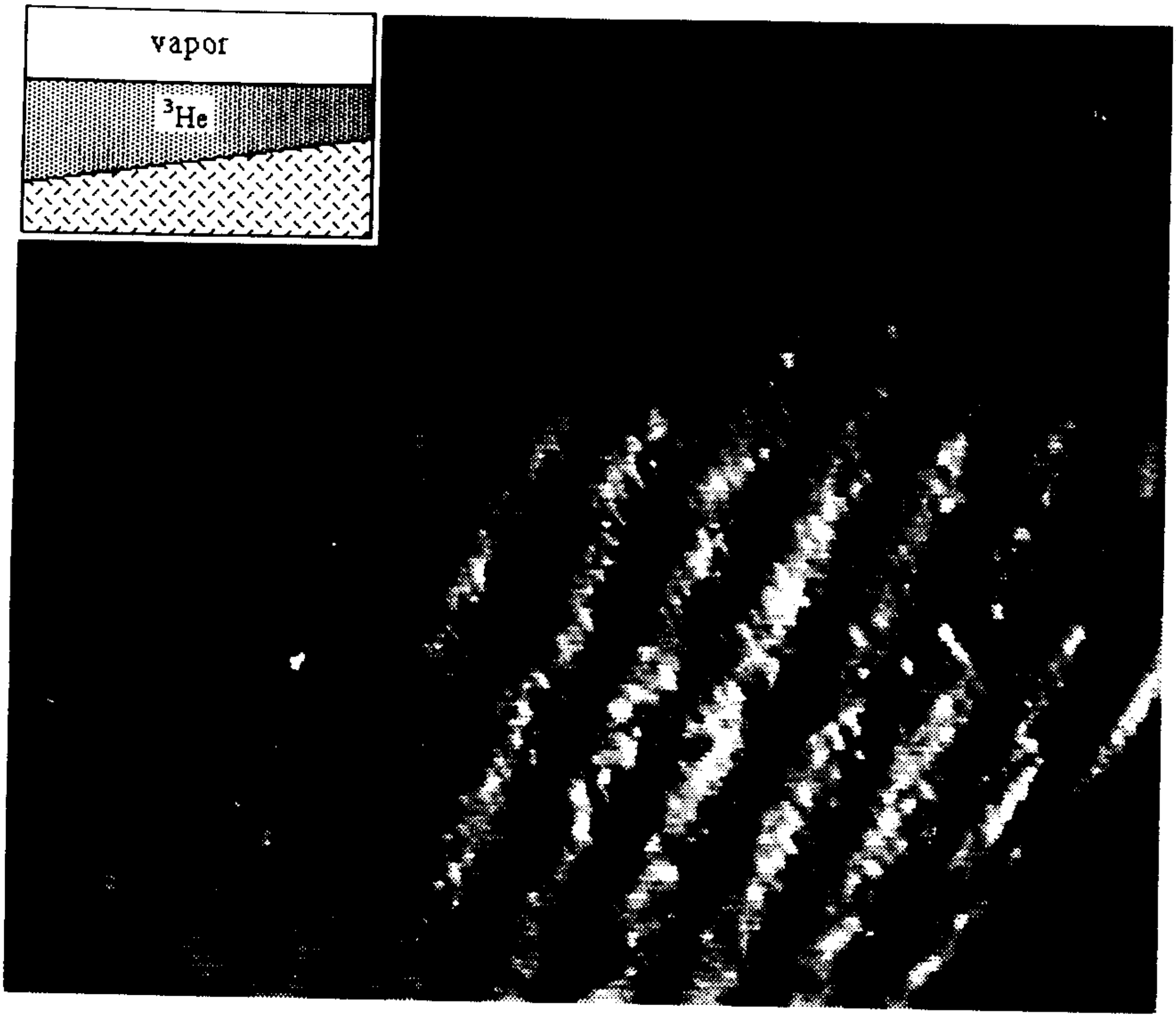
1) Small reflection
2) Cooling to milliKelvins

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2}\right)^2 \approx 10^{-6}$$

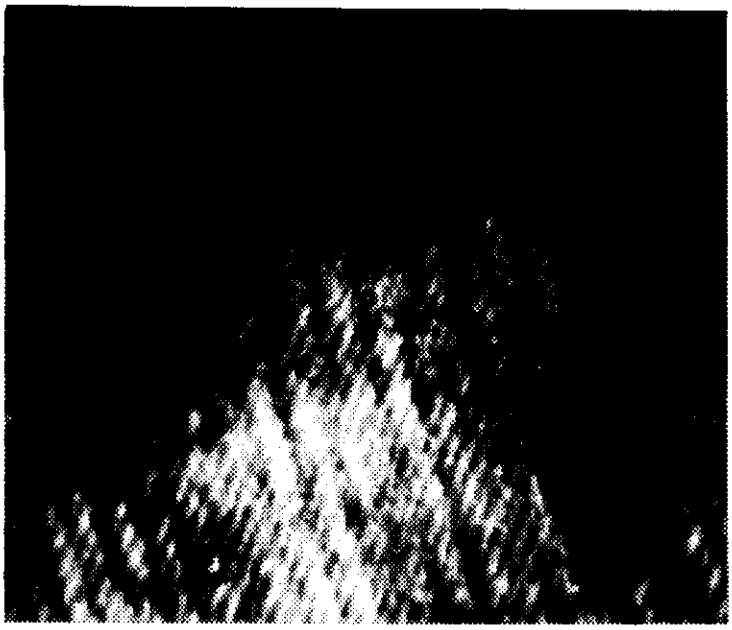
MANNINEN et al 1992,
WAGNER et al 1994



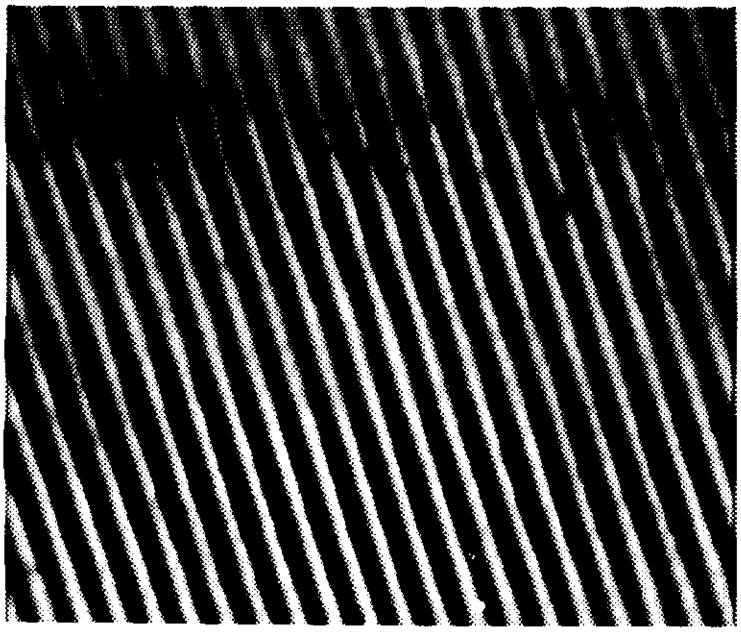
* GROWTH MEASUREMENTS:
  - BeCu PRESSURE GAUGE
  - $\Delta p \sim 0.3 \mu bar$



- IMAGED USING A VIDEO CAMERA
- WEDGE SHAPED LIQUID LAYER FROM ABOVE
- THICKNESS CHANGE:
  0.32 μm BETWEEN FRINGES
- REFLECTION $R = 1.2 \cdot 10^{-4}$



UNTREATED IMAGE



BACKGROUND SUBTRACTED IMAGE

- SUPERFLUID/SOLID INTERFACE
- REFLECTION $R = 3 \cdot 10^{-6}$
- IMAGED USING SLOW SCAN CAMERA

WHY TO STUDY ^{4}He CRYSTALS

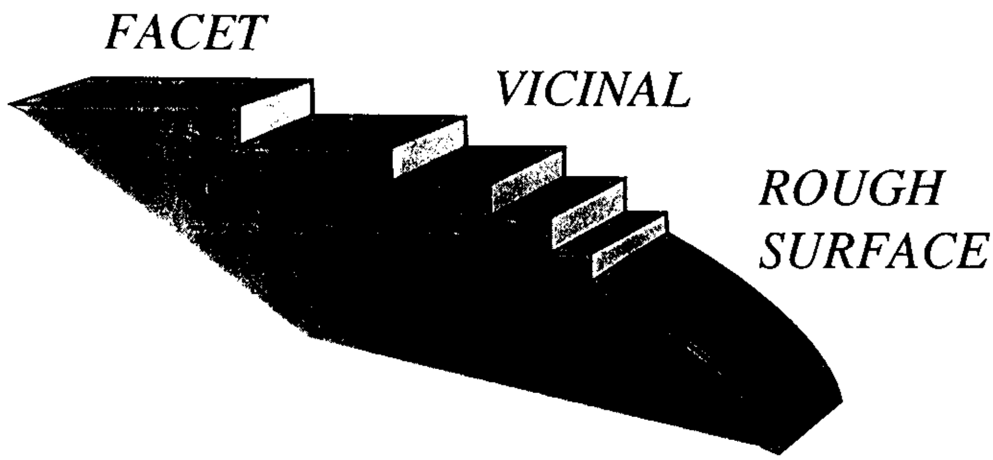
- ***High purity***
  - Impurities, except ^{3}He, frozen out
  - Natural purity 0.1 ppm; 10^{-14} achievable

- ***Equilibrium reached quickly, even in large crystals***
  - Latent heat very small
  - Thermal conductivity excellent
  - Studies of 10 mm crystals instead of 10 μm (Cu, Au)

- ***Surface phenomena not hampered by thermal effects***
  - Theoretically tractable situation
  - Elementary steps and their interactions
  - *Good model system for equilibrium crystal shapes*
    (Carmi et al. 1987, Andreeva et al. 1989, Rolley et al. 1994)

- ***Faceting in the limit $T \rightarrow 0$***
  - Infinite sequence of faceting transitions?
  - Only three transitions have been observed

CRYSTAL SHAPES



Surface free energy

$$\alpha(\phi) = \left(\alpha_0 + \beta\phi + \frac{\gamma}{6}\phi^3\right)\cos\phi$$

Terraces — Steps — Interactions

- *1/d* ^{2}
- *entropic*
- *elastic*

$\alpha_0 = 0.25 \frac{erg}{cm^2}$

$\beta = 0.014 \frac{erg}{cm^2}$

$\gamma = \gamma_{el} + \gamma_s(T)$
$\propto T^2$

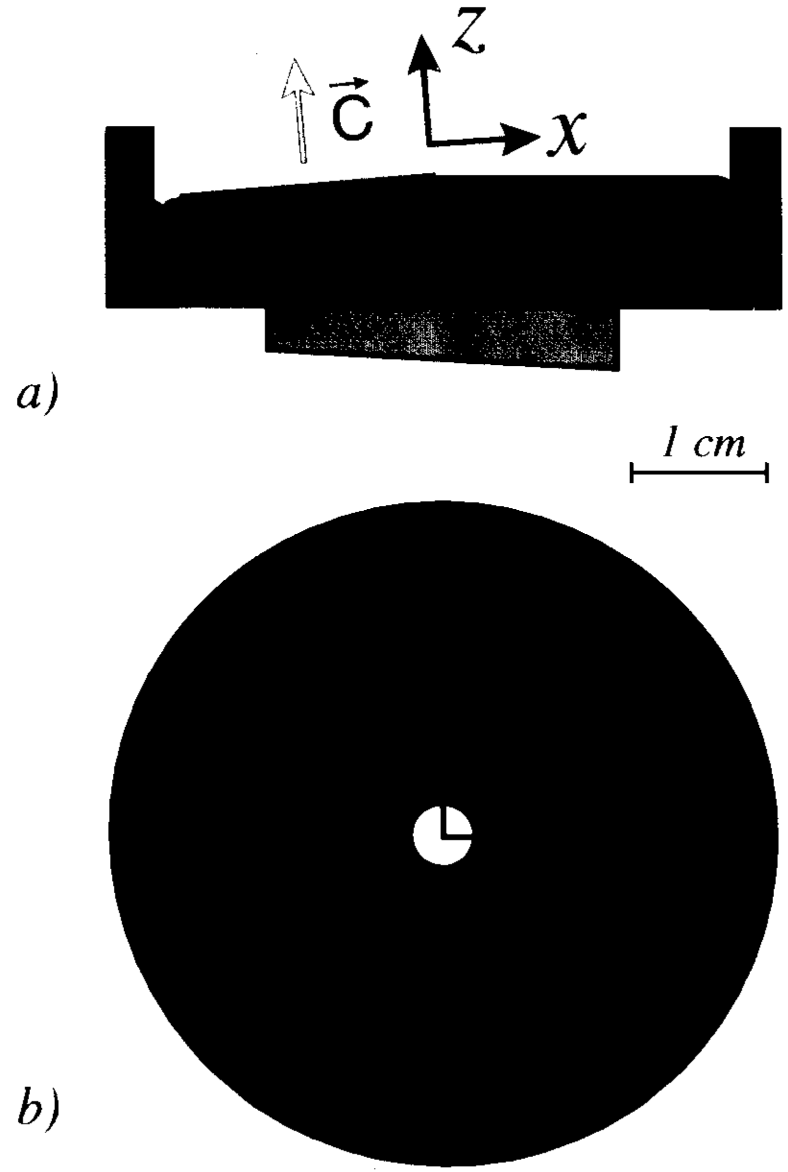
Surface stiffness: $\tilde{\alpha} = \alpha + \alpha''_{\phi\phi}$

$\tilde{\alpha}_{||} = \gamma\phi$ -Rolley *et al.* 1994 ($\phi > 10$ mrad)

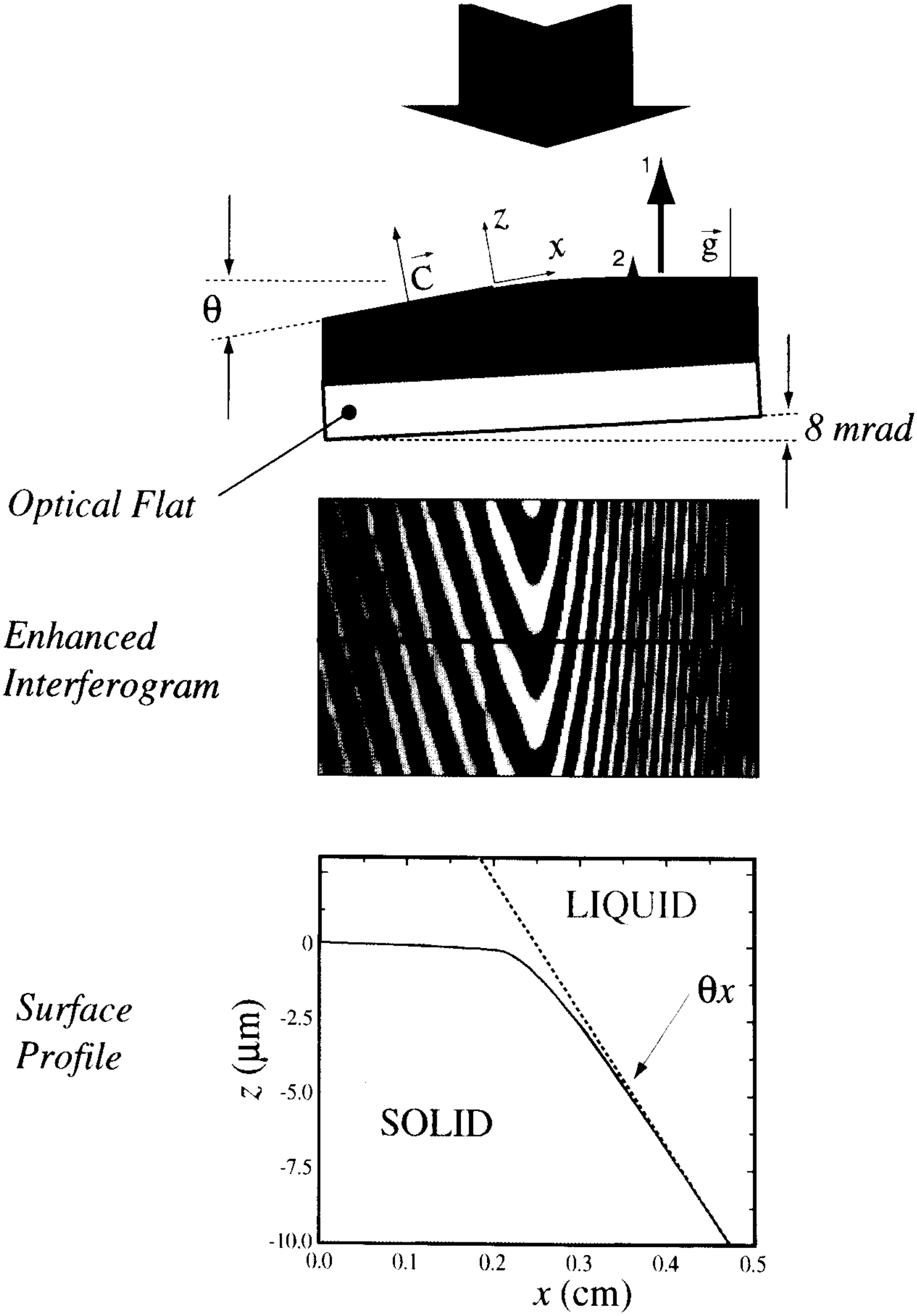
$$\tilde{\alpha}_{\perp} = \frac{\beta}{\phi}$$

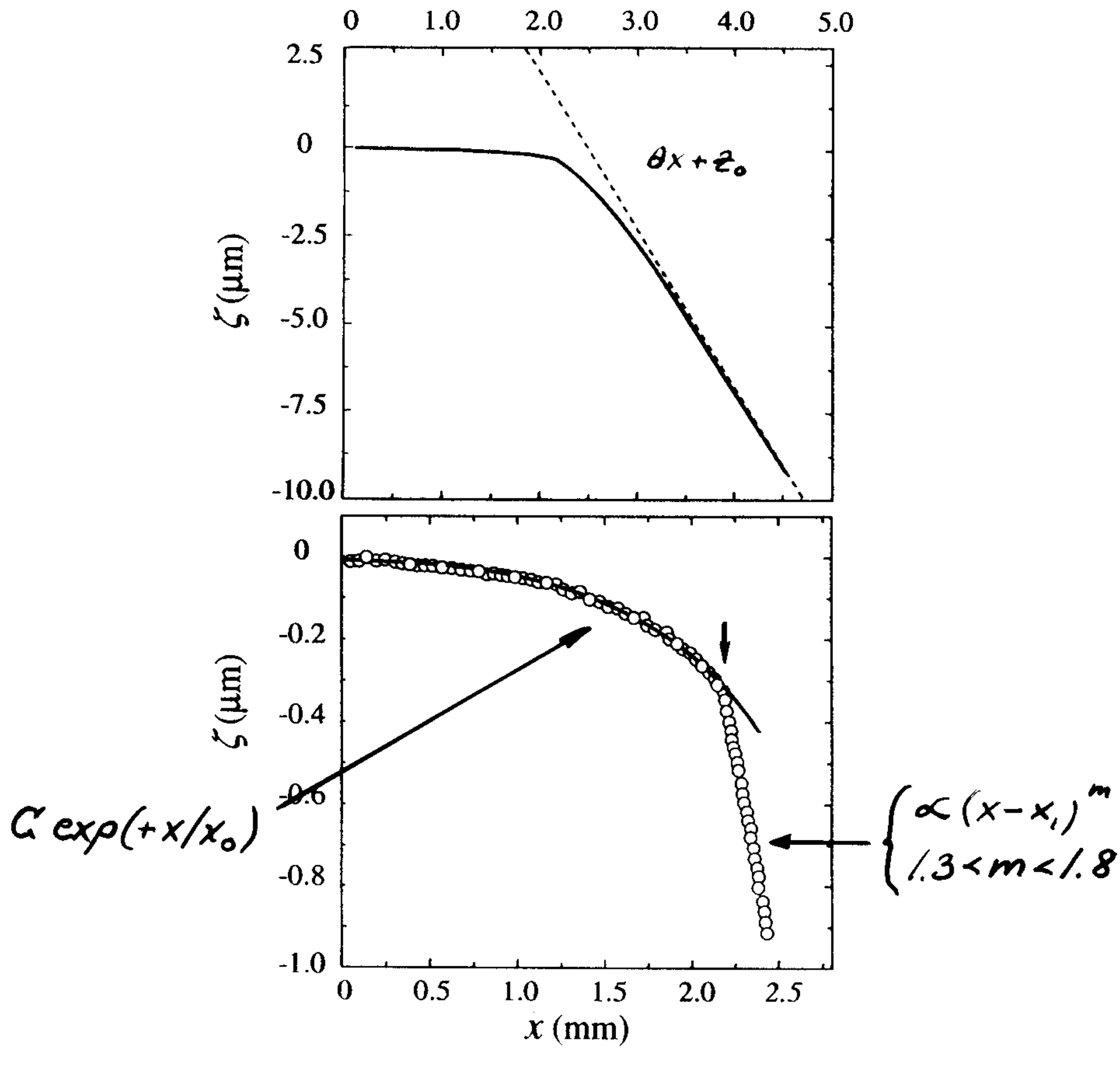
$\tilde{\alpha}_{||} = \gamma\phi \;\Rightarrow\; x^{3/2}$ profile

$\tilde{\alpha}_{||} = const. \;\Rightarrow\; x^2$ profile (1/*d* interactions)



$\vec{C}$
z
x
a)
1 cm
b)

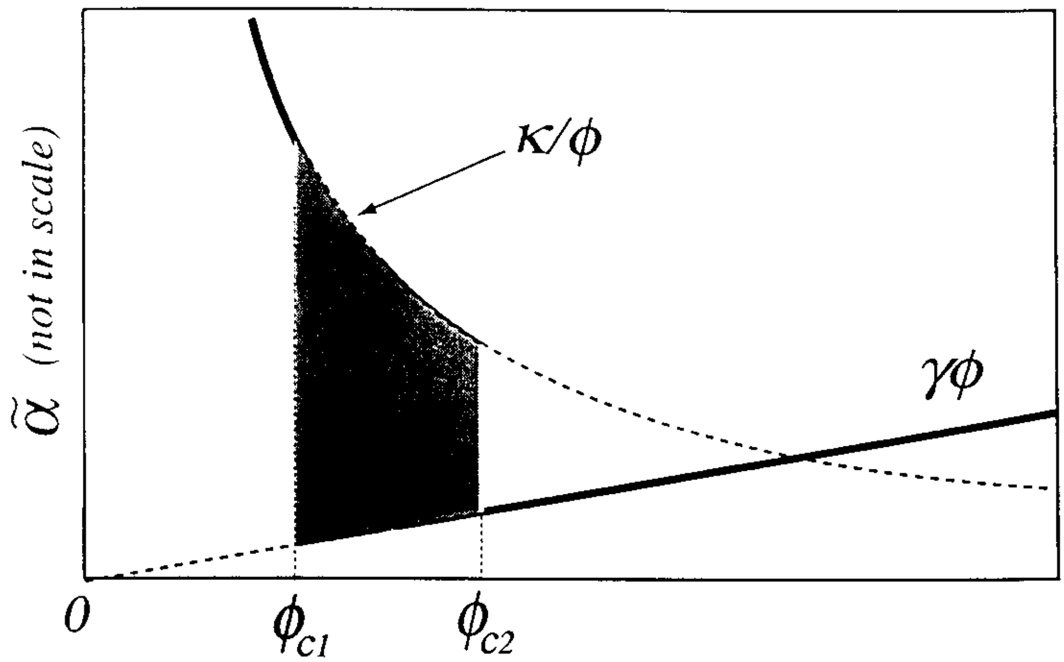

1
2
z
x
$\vec{C}$
$\vec{g}$
θ
8 mrad
Optical Flat
Enhanced
Interferogram
Surface
Profile
LIQUID
SOLID
θx
0
-2.5
-5.0
-7.5
-10.0
0.0
0.1
0.2
0.3
0.4
0.5
z (μm)
x (cm)



EQUILIBRIUM CRYSTAL SHAPE:

1-DIM. CASE $\quad \tilde{\alpha}\zeta''_{xx} = \zeta + \theta x + z_0$

$$\begin{cases} \tilde{\alpha} = \dfrac{\kappa}{\varphi} & \varphi < \varphi_{c_1} \simeq 400\,\mu rad \\ \tilde{\alpha} = \gamma\varphi & \varphi > \varphi_{c_2} \simeq 2\,mrad \end{cases}$$

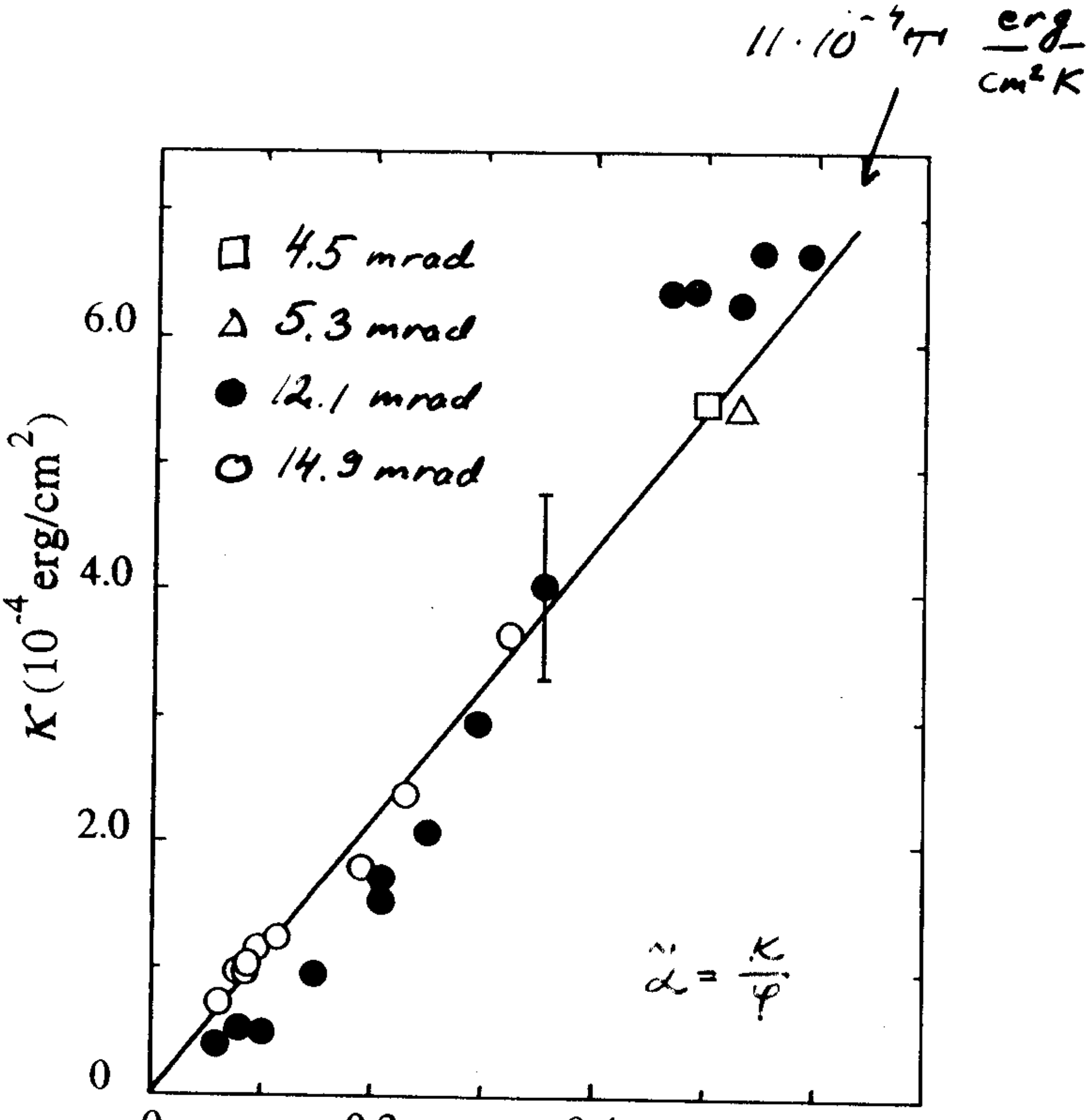


* FIRST ORDER TRANSITION BETWEEN TWO SURFACE STATES

* STRONG HYSTERESIS

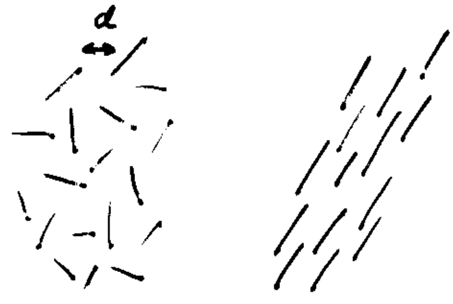
  $\varphi_{c_1} = 50 - 500\ \mu rad$

* METASTABILITY OF THE FACET SIZE!


11·10⁻⁴ T erg/cm²K
□ 4.5 mrad
Δ 5.3 mrad
● 12.1 mrad
○ 14.9 mrad
α̂ = κ/φ
6.0
4.0
2.0
0
0.2
0.4
0.6
κ (10⁻⁴ erg/cm²)
T (K)

POSSIBLE EXPLANATIONS

• ***Collective effects of dislocations***
(Uwaha and Nozieres 1987)
- Polarization of
Frank-Read sources
- Too few dislocations:
$d \approx 10\ \mu\text{m}$ needed
$d > 1$ mm in our crystals

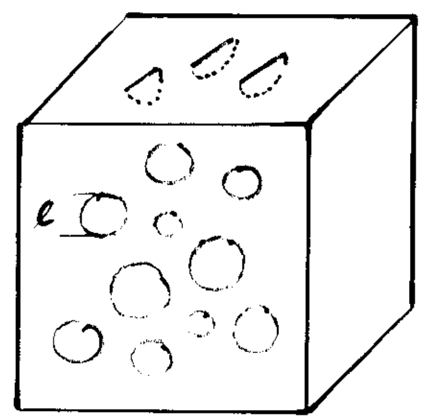


• ***Thermally excited dislocation loops***
(Andreev 1990, 95)

- Destroy long-range order
- Model calculation yields:

$$\tilde{\alpha}(\phi) = \frac{T}{\phi}(al)^{-1}$$

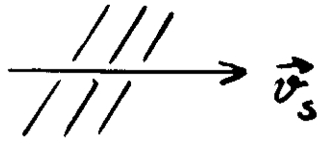
$l = 30$ nm, questionably big
- Excitation energy > 10 K



MORE POSSIBILITIES

- ***Instability due to superfluid flow*** (Andreev 1994)

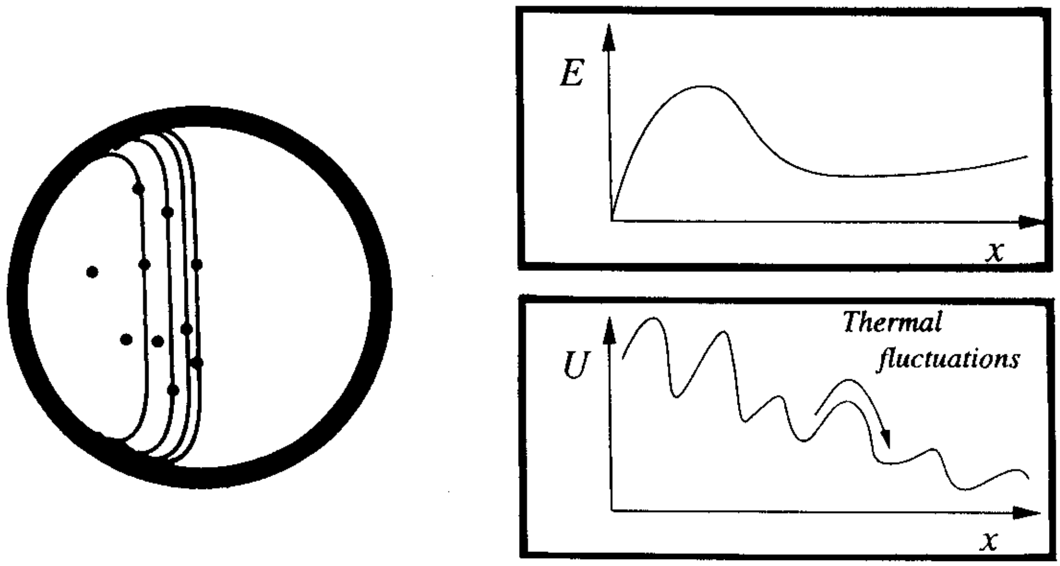
$$\tilde{\alpha} = \frac{\rho a v_s^2}{2\pi\phi}$$



  - 10^{10} times larger flow than can be present

- ***Surface stresses***
  - too small

- ***Step pinning by defects*** (Thuneberg 1996)
  - Driving force small close to equilibrium
    Even small pinning forces effective



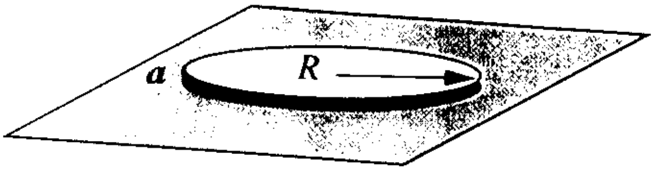
Exponential profile due to pinning probability:

$$\exp\left(\frac{\Delta U}{kT}\right) \propto \exp\left(\frac{x}{x_0}\right)$$

- many pinning sites?
- inclination angle dependence?

GROWTH OF c-FACETS

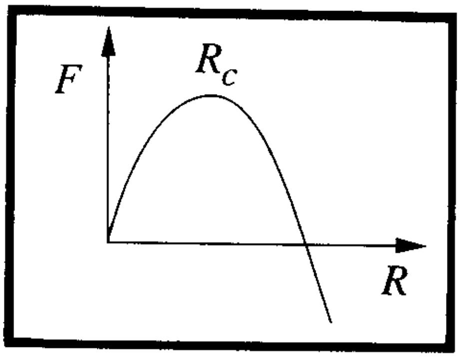
- *TERRACE GROWTH*



β STEP ENERGY
a STEP HEIGHT
$\Delta\mu = \frac{\Delta\rho}{\rho^2}\Delta p$

$$F = 2\pi R a\beta - \pi R^2 a\rho_s\Delta\mu$$

$$R_c = \frac{\rho}{\Delta\rho}\frac{\beta}{\Delta p}$$



$$\Delta p = 1\ \mu\text{bar} \Rightarrow R_c \approx 1\ \text{mm}$$
$$\Delta p = 100\ \mu\text{bar} \Rightarrow R_c \approx 10\ \mu\text{m}$$

- Nucleation unlikely at low T
- Observed close to T_R by Wolf *et al.* (1985).

- *SPIRAL GROWTH*
  - Step emerging from a screw dislocation

    ⇒ ***No nucleation barrier***
  - Spiral growth the only mechanism at low T

    Wolf *et al.* (1985)

    - Problems with irreproducibility

    Tsymbalenko (1995)

    - Intermediate behavior $T > 0.5$ K

SPIRAL GROWTH

J. WEEKS AND H. GILMER
ADV. IN CHEM. PHYS.
40, 157 (1979)

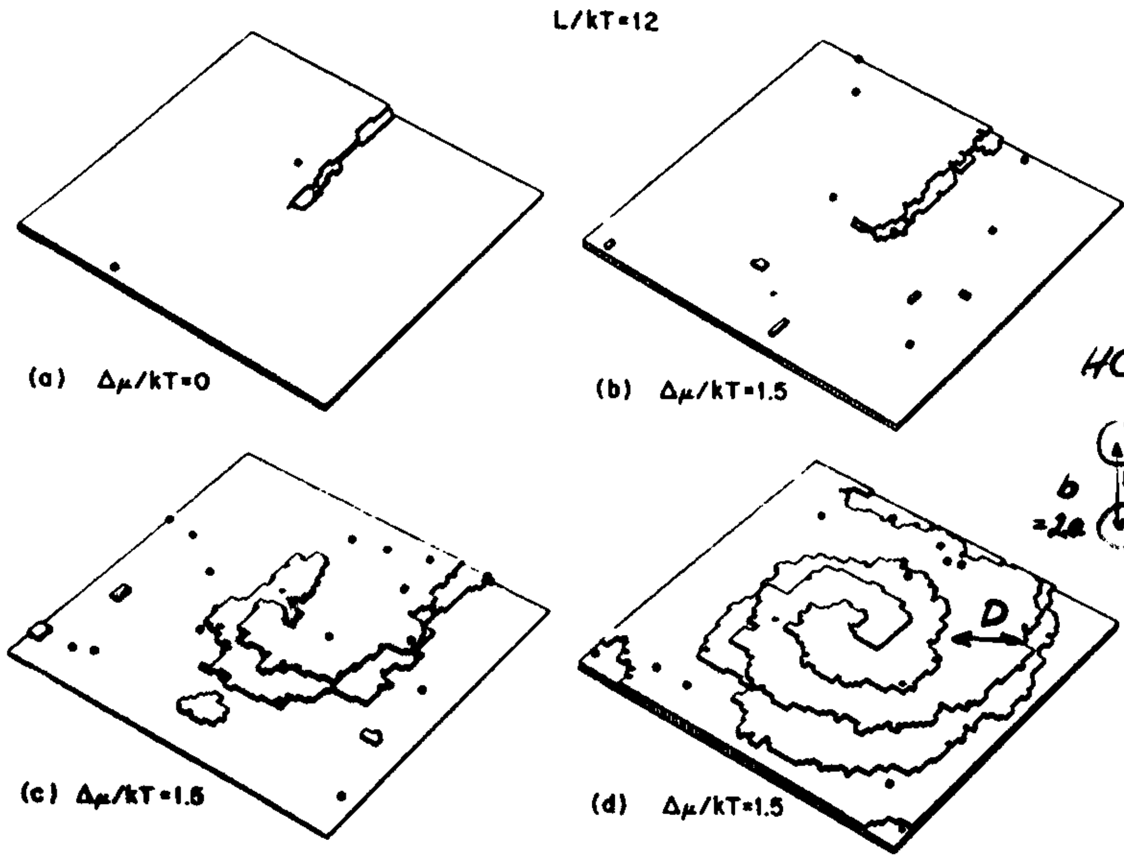
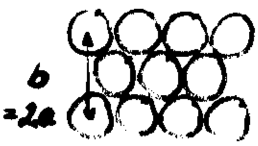


Fig. 12. An illustration of the formation of a double spiral. (*a*) is an equilibrium configuration, whereas (*b*) to (*d*) illustrate the step motion resulting from a driving force of $\Delta\mu = 1.5kT$.

HCP CRYSTAL



EQUATION OF STEP MOTION

$$f - \frac{\beta a}{R} - \frac{1}{\mu_s} v_s = 0$$

$f = a \frac{\Delta \rho}{\rho} \Delta p$ "SUPERCOOLING FORCE"

$v_s = \mu_s f$ STEP MOBILITY

R = RADIUS OF CURVATURE

BURTON, CABRERA, AND FRANK (1951)

- ASYMPTOTIC SOLUTION

$$D \sim 20 R_c$$

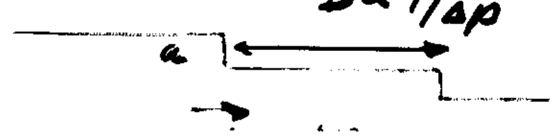
$$R_c = \frac{\rho}{\Delta\rho} \frac{\beta}{\Delta p}$$

100μm ≃ 10μbar

VERTICAL VELOCITY

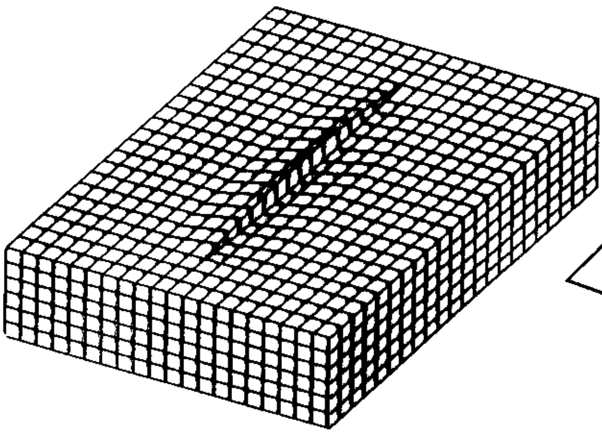
$$v = \frac{a}{D} v_s \qquad \propto (\Delta p)^2$$

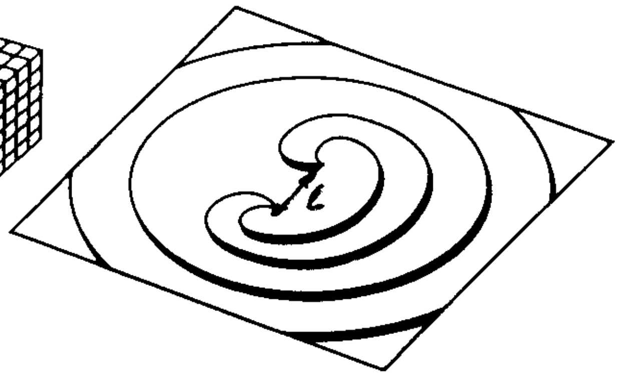
$D \propto 1/\Delta p$



GROWTH BY A FRANK-READ SOURCE

* TERRACE GENERATION WHEN ROTATING SPIRALS TOUCH



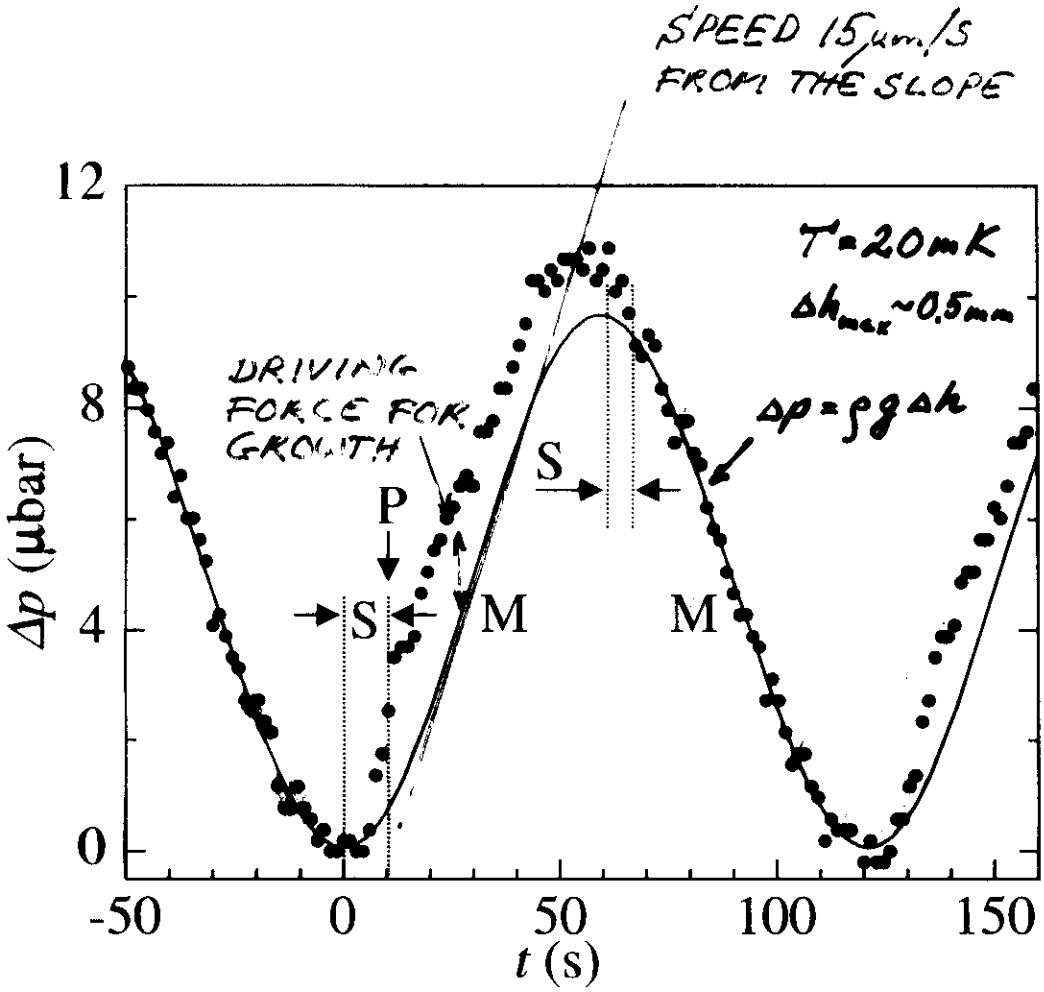


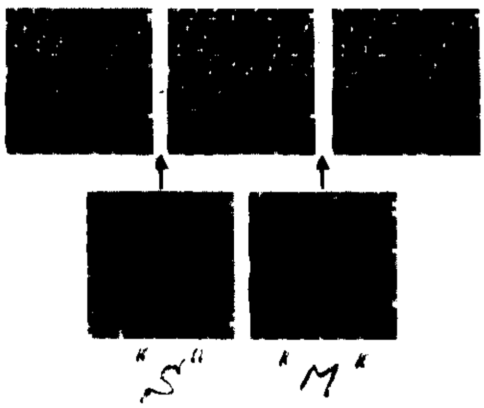
* GROWTH THRESHOLD: $\Delta p_c \approx \frac{\rho}{\Delta\rho} \frac{\beta}{\ell/2}$

$\Delta p_c \sim 1 \mu bar$

20 DISLOCATIONS/cm^2

* GROWTH RATE IS A PROPERTY OF A SINGLE DISLOCATION

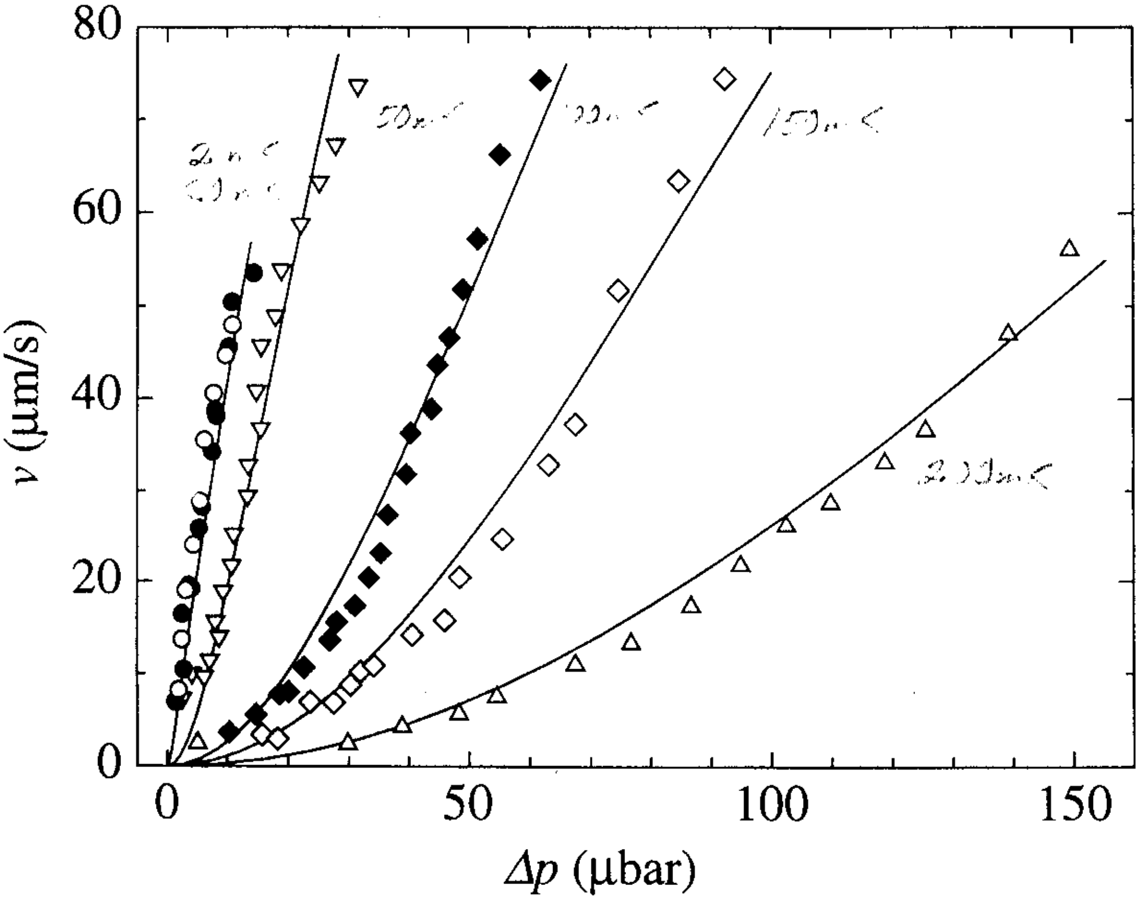




- $\Delta p_c \simeq 2 \mu bar$
- $100 \frac{1}{cm^2}$ OF SCREW DISLOCATIONS
- ASYMMETRY

  MELTING: HIGH MOBILITY ORIENTATIONS

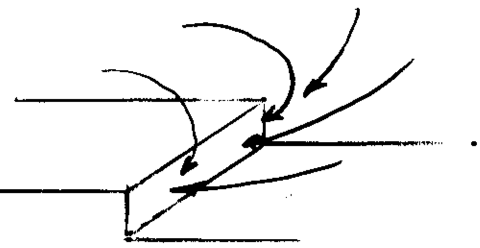
  GROWTH: MINIMUM MOBILITY


80
60
40
20
0
0
50
100
150
v (μm/s)
Δp (μbar)

MODIFIED THEORY OF SPIRAL GROWTH

Equation of step motion with ***inertial terms*** and ***kinetic energy*** added:

$$f - \frac{\beta^* a}{R} - \frac{1}{\mu} v_s = m\left(\frac{\partial v_s}{\partial t} + v_s \frac{\partial v_s}{\partial n}\right)$$



$\beta^* = \beta + (1/2) m v_s^2$

m = effective mass of the step

n = normal to the step

$f = a(\Delta\rho / \rho)\Delta p$

$m \approx \frac{\Delta\rho^2}{\pi\rho} a^2 \ln\frac{R}{a} \simeq 5\text{-}7 \cdot 10^{-18} \frac{g}{cm}$

Solution depends on the ratio $\gamma = \frac{Flow\ energy}{Step\ energy} = \frac{m v_s^2}{\beta}$

$v = \Lambda_0 \frac{\mu_0 a}{2\pi\beta} f^2 \quad \Lambda_0 = 0.33095$	at $\gamma << 1$	Classical picture
$v = \frac{a}{2\pi\sqrt{2m\beta}} f$	at $\gamma >> 1$	Inertia dominates

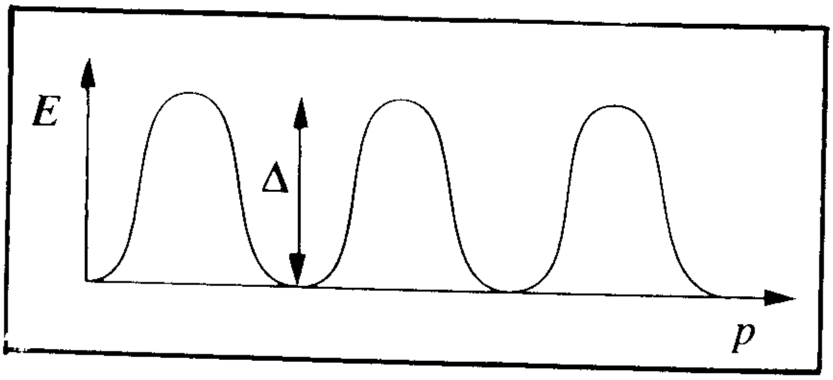
- does not depend on the mobility!
- steady state - *dissipation at the edge of the facet*
- flow of energy from the center to the edge

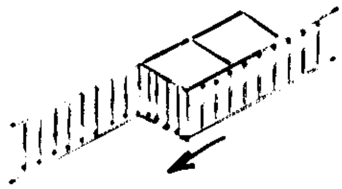
Spacing of the spiral arms $\quad D = 2\pi\mu\sqrt{2m\beta}$

Classical spacing $\quad D \cong \frac{2\pi}{\Lambda_0} \frac{\Delta\rho}{\rho} \frac{\beta}{\Delta p}$

LOCALIZATION OF STEPS

- Mobility of steps is made of motion of kinks along the step (Andreev and Parshin 1979).





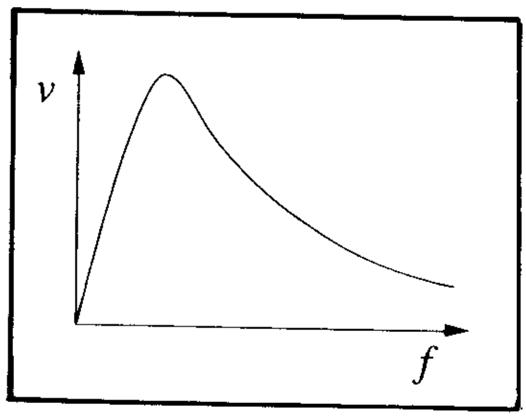
$$E = \varepsilon_0 - \frac{\Delta}{2}\cos\left(\frac{pa}{\hbar}\right)$$

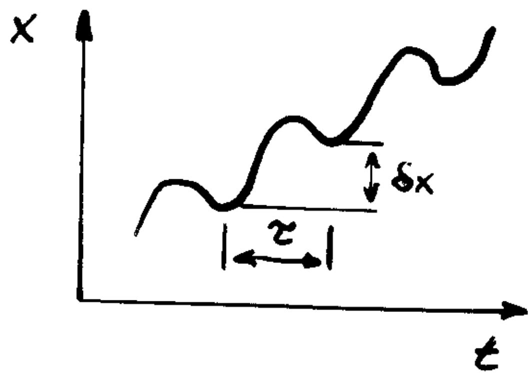
$$v = \frac{\partial E}{\partial p} = \frac{\Delta a}{2\hbar}\sin\left(\frac{pa}{\hbar}\right), \qquad v_{max} = \frac{\Delta a}{2\hbar}$$

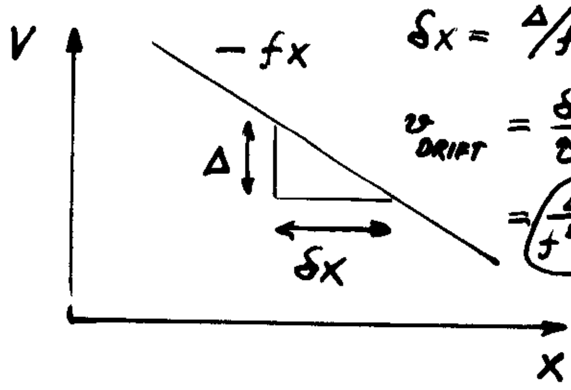
$$\dot{p} = f - \frac{1}{\mu} v$$

If $f > v_{max}/\mu$
the external force cannot be compensated by dissipative effects $\Rightarrow$ $v < v_{max}$

$$\frac{1}{\mu} = \frac{1}{\mu_0} + \eta f^2$$







$\delta x = \Delta / f$

$v_{DRIFT} = \frac{\delta x}{\tau} = \frac{\Delta}{f\tau} = \left(\frac{\Delta}{f^2\tau}\right) f$

$\eta = \frac{\tau}{\Delta}$

$\Delta \sim 10K$

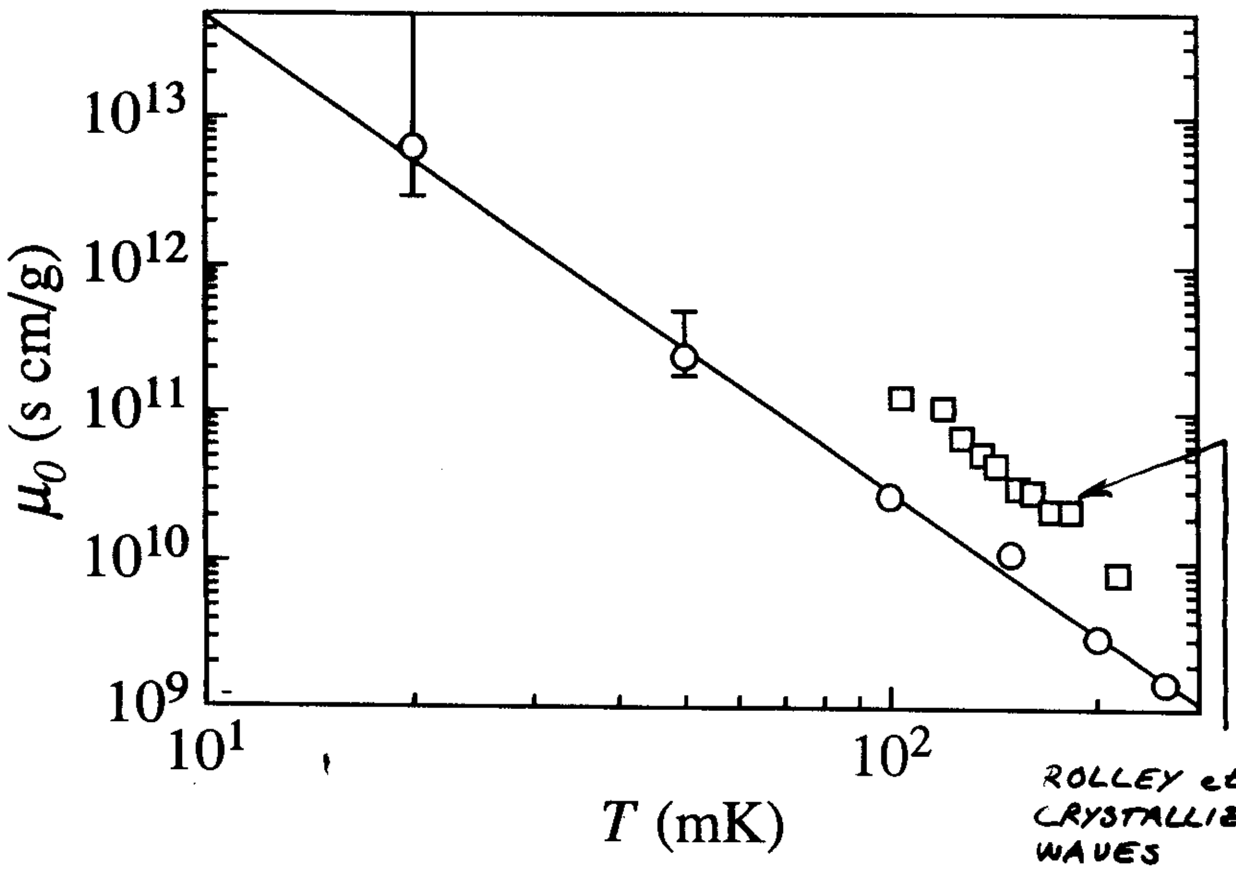
$$\begin{cases} f - \frac{\beta^* a}{R} - \frac{1}{\mu} v_s = m\left(\frac{\partial v_s}{\partial t} + v_s \frac{\partial v_s}{\partial n}\right) \\ \frac{1}{\mu} = \frac{1}{\mu_0} + \gamma f^2 \end{cases}$$

$m = 5 \cdot 10^{-18}$ g/cm   SLOPE AT LOW T

$\gamma = 300$ AVERAGE FROM FREE FITS

μ_0 FITTED

$\mu_0 \propto \frac{1}{T^n}$, $n = 3.3 \pm 0.3$ $\mu_0 \cong \frac{c a^2}{\theta_D}\left(\frac{\theta_D}{T}\right)^3$ NOZIERES AND UWAHA 87 PHONON DAMPING



ROLLEY et al 94 CRYSTALLIZATION WAVES

ANOTHER POSSIBILITY:

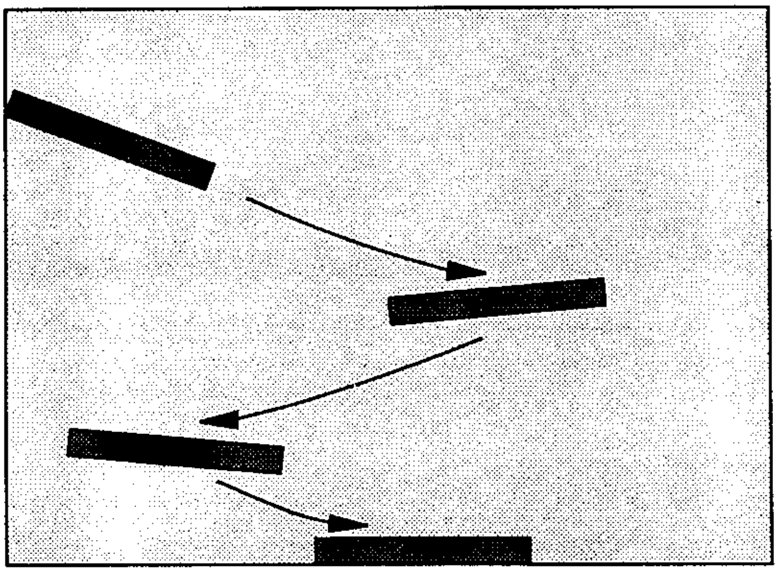
CERENKOV RADIATION AT $v > v_c$ "SUPERSONIC" PHONON/ROTON EMISSION

$$\frac{1}{\mu} = \frac{1}{\mu_0} + \gamma f^2 + \frac{1}{\nu(v)}$$

RESULTS WITH ^3He IMPURITIES DISFAVOR THIS MECHANISM

NUCLEATION OF CRYSTALS

- Proper orientation by trial and error using free fall
  "Throwing dice"



- Angle between the **c**-facet and the reference plate < 5 mrad
  - 2 mrad corresponds to 5 fringes/mm (200 tries needed)
  - Difficulties increase with requirements for orientation

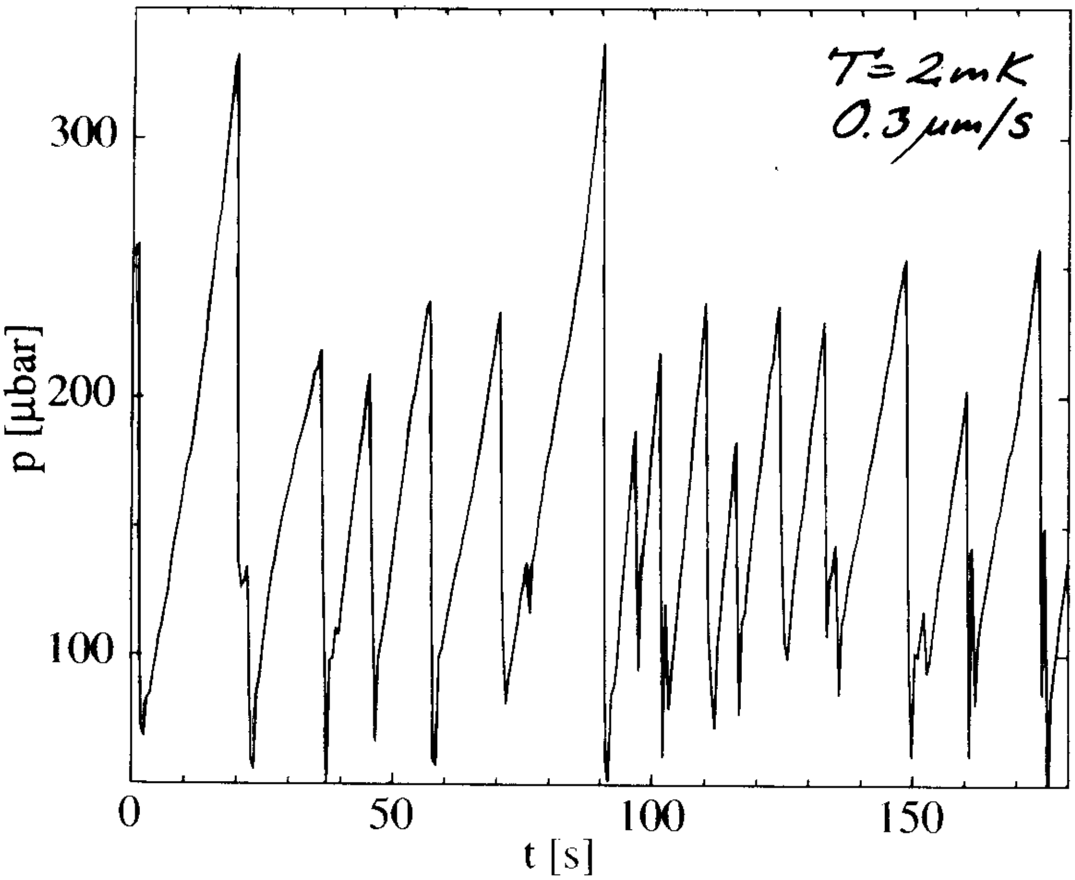
- Typically done at 0.9 K near the melting curve minimum
  - Only **c**-facets, i.e., pan-cake-like crystals
  - 100 screw dislocations/cm^2

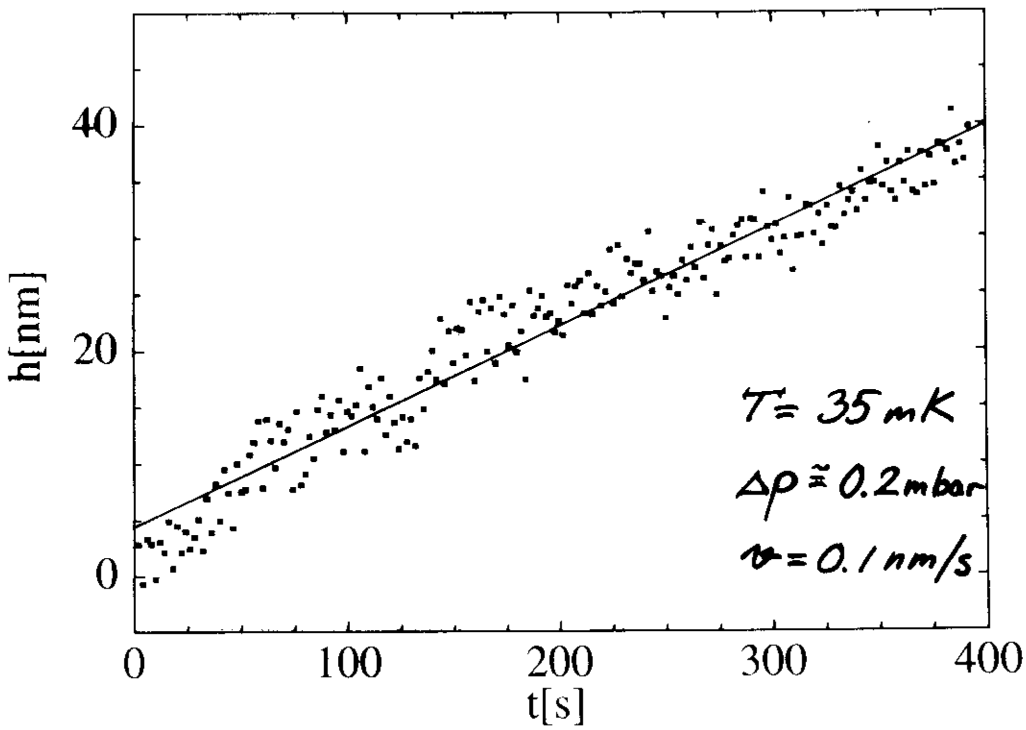
- We have created crystals also at 20 mK
  - More randomness in free fall
  - *Even crystals with no screw dislocations!*

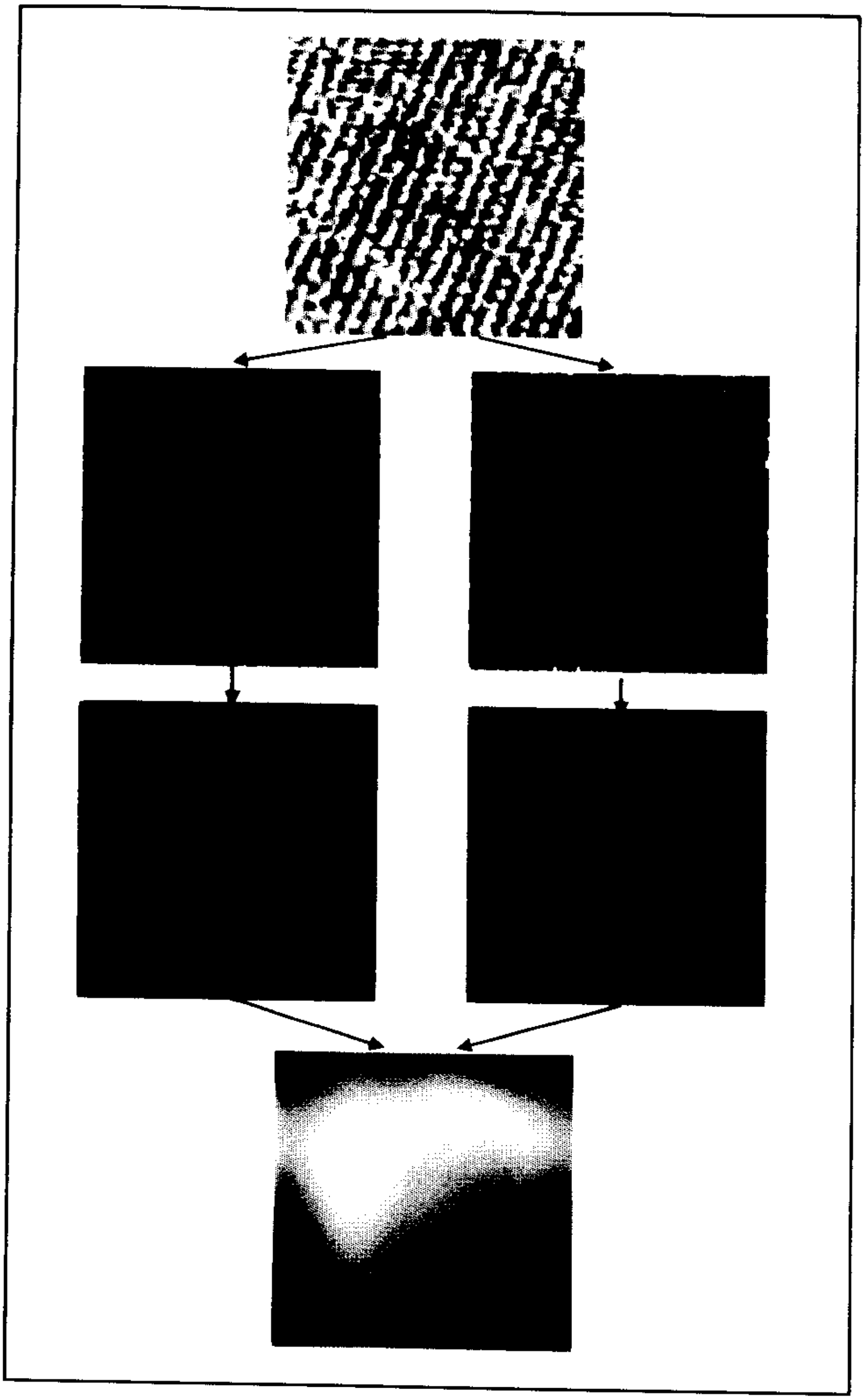
SLOW GROWTH OF c-FACETS

- NO SCREW DISLOCATIONS !?
  - NUCLEATION AT 20mK
  - NO ELECTRIC FIELD

- DISLOCATIONS CREATED AT 0.2-0.25K IN A FAST GROWTH




40
20
0
h[nm]
0
100
200
300
400
t[s]
T= 35 mK
Δp ≅ 0.2mbar
v = 0.1 nm/s



FOURIER TRANSFORM

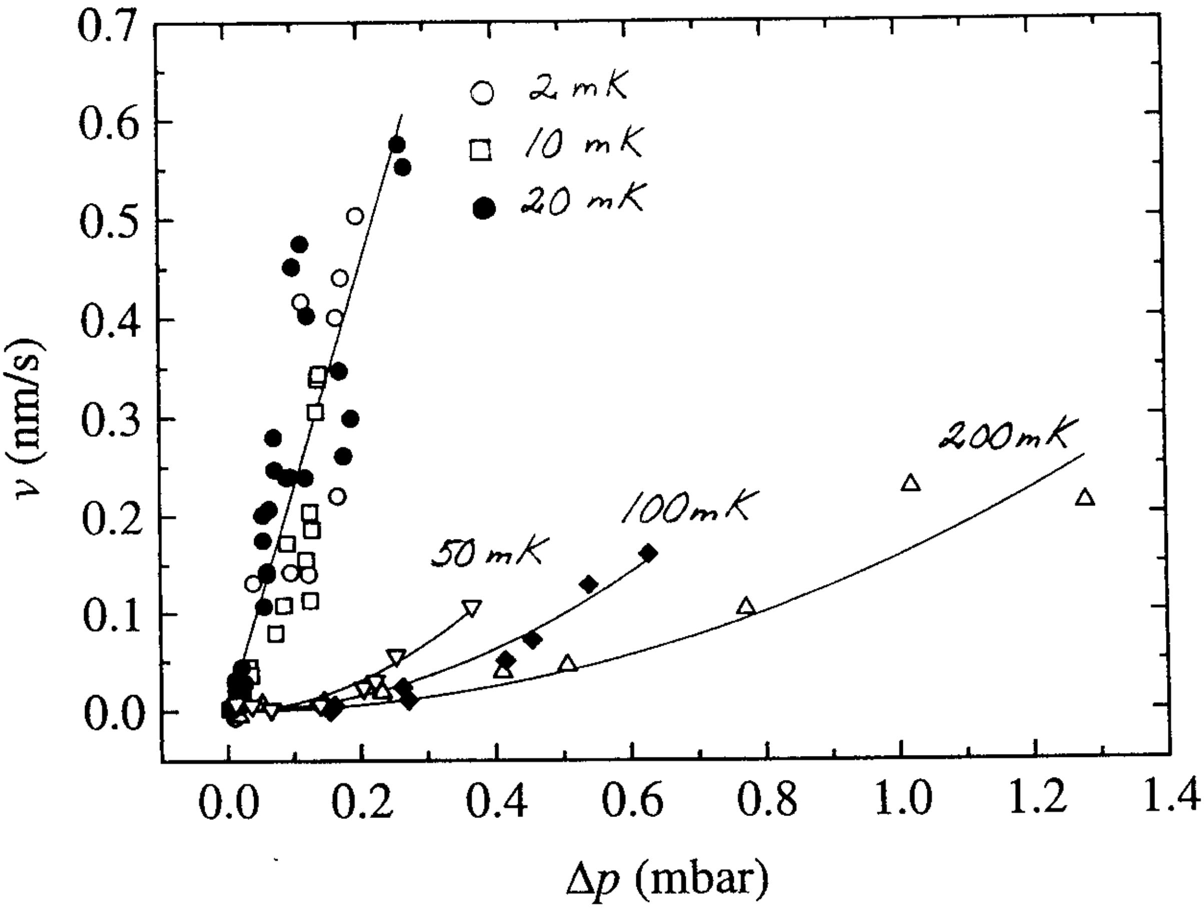
SYMMETRIC

GAUSSIAN FILTERING & SHIFT TO ORIGIN

INVERSE TRANSFORM & RATIO WITH REFERENCE IMAGE

⇒ $\Delta\varphi$ AS AN AVERAGE OVER THE IMAGE

REFERENCE:
KOSTIANOVSKI, LIPSON, RIBAK, APPL. OPT. 32, 4744 (1993)



- Similar to spiral growth:
  - Linear at low T, quadratic at high T
  - Step mass 10^7 times larger?
- Explanation?
  - Not due to ^{3}He impurities
  - Relation to facet curvature

FINAL REMARKS

Behavior of ^{4}He crystals more complicated than expected!

- EQUILIBRIUM CRYSTAL SHAPES

- ***First order transition between two surface states***

- Facet goes discontinuously over to a vicinal surface

- Exponentially relaxing facets
  Logarithmic interactions?
  No physical reason!

- SPIRAL GROWTH OF **c**-FACETS AT LOW *T*

- ***Linear pressure dependence due to step inertia***

• ***Inertia*** significant => $v \propto \Delta p$.
• ***Friction*** significant => $v \propto (\Delta p)^2$

- ***Step localization under large driving forces***

- Our model yields the step mass and the relaxation of kink

- Effect of ^{3}He impurities to be clarified

- SLOW GROWTH OF **c**-FACETS

- ***Growth even in the absence of screw dislocations***

- Relation to equilibrium shape?
  Large driving pressure: No facet curvature

- Nucleation at 20 mK without electric field
  Necessary for crystals without screw dislocations?

- *Spiral growth of massive steps?*

FUTURE PLANS

1) ^{4}He crystals

- *Effect of ^{3}He impurities*
    - step structure and mass
- *Waves on the curved part of the facet*
    - spatially varying surface stiffness
    - nonlinear behavior due to the growth threshold
    - observable below 0.1 K

2) ^{3}He crystals below 1 mK

- Magnetic properties of the interface
- Spin supercurrents
- *Crystallization waves* damped by magnons in solid
    - 0.2 mK needed for undamped waves

3) Defect-induced dimples at interfaces

- Single dislocations
- Improvement of resolution
    - three-beam interferometry?