Depinning of Driven Systems:
Time-resolved x-ray scattering measurements of the charge-density waves in $NbSe_3$

Joel D. Brock

Professor and Director
School of Applied & Engineering Physics

Director
G-line Division, Cornell High Energy Synchrotron Source (CHESS)
Cornell University, Ithaca, NY

Acknowledgements

Kristin Ringland[1], Robin Sampson, Jun-Dar Su, and Yanping Li[2]
School of Applied & Engineering Physics
Cornell University, Ithaca, NY 14853

S.G. Lemay[3], and R.E. Thorne
Department of Physics
Cornell University, Ithaca, NY 14853

Mark Sutton
Department of Physics
McGill University, Montreal, CANADA

1. McKinsey & Company, Stockholm, Sweden
2. Applied Materials Inc., Santa Clara, CA
3. Delft University of Technology, Delft, The Netherlands

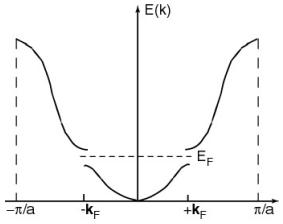
Outline

- Intro to CDW Physics/Phenomenology
- Intro to Modern X-ray Technology
- Properties of $NbSe_3$
- Experimental Details – Sample quality
- Kinetics of Relaxation from Sliding State
- Structure of the Sliding & Zero-field cooled States
- Low Temperature Phase Diagram
- Coherent X-ray Measurements: "Speckle"
- Summary

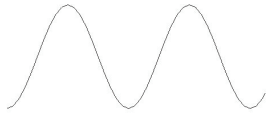
CDW Basics

In Quasi-One-Dimensional Metals...

- At *T=0,* Electronic energy is lowered by opening an energy gap at the Fermi surface.
- Periodic potential required to do this can be supplied by a longitudinal lattice-density wave (LDW).
- Elastic energy cost for LDW is less than electronic energy gain



$$\rho = \overline{\rho} + \Delta\cos\left(2k_F\hat{b}\cdot\vec{r} + \phi(\vec{r})\right)$$

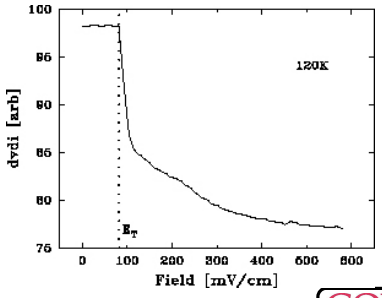


$$\vec{u}(\vec{r}) = \vec{u}_0\sin\left(2k_F\hat{b}\cdot\vec{r} + \phi(\vec{r})\right)$$

Sliding CDWs

Since the wavelength of a CDW is (usually) incommensurate with the periodicity of the lattice, there is no elastic restoring force. In an ideal crystal, the CDW can "slide" rigidly without friction.

Defects in the crystal lattice provide "friction," creating a threshold field.



General Predictions

- At zero field, periodicity of lattice → quasi long-range translational order
- Far below threshold: thermal activation → creep
- Topological defects → plastic flow
- Near elastic depinning threshold: similar to standard critical phenomena (velocity is order parameter).
- Far above threshold: recovery of perfect lattice

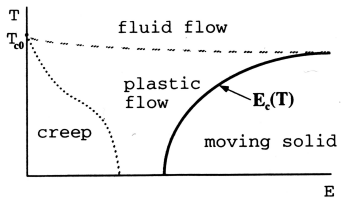


FIG. 1. Schematic phase diagram for the three-dimensional CDW. Hashed lines indicate (possible sharp) crossovers.

L. Balents and M.P.A. Fisher, PRL, **75**, 4270 (1995)

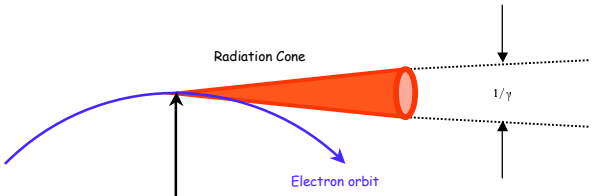
X-ray scattering is nearly ideal probe

- non-perturbative
- direct interpretation
- structural measurement

$$S(\vec{q},t)=\frac{1}{\Omega}\left|J_1(\vec{q}\cdot\vec{u}_0)\right|^2\int_{\Omega}e^{i\left[\left(\vec{G}\pm 2k_F\hat{b}\right)-\vec{q}\right]\vec{r}}\left\langle\overline{e^{i\phi(\vec{r},t)}e^{-i\phi(\vec{0},t)}}\right\rangle d\vec{r}$$

$$\left\langle\overline{e^{i\phi(x,t)}e^{-i\phi(0,t)}}\right\rangle=e^{-\frac{1}{2}g(x,t)}\approx e^{-\frac{1}{2}\left\langle\overline{[\phi(x,t)-\phi(0,t)]^2}\right\rangle}$$

Synchrotron Radiation

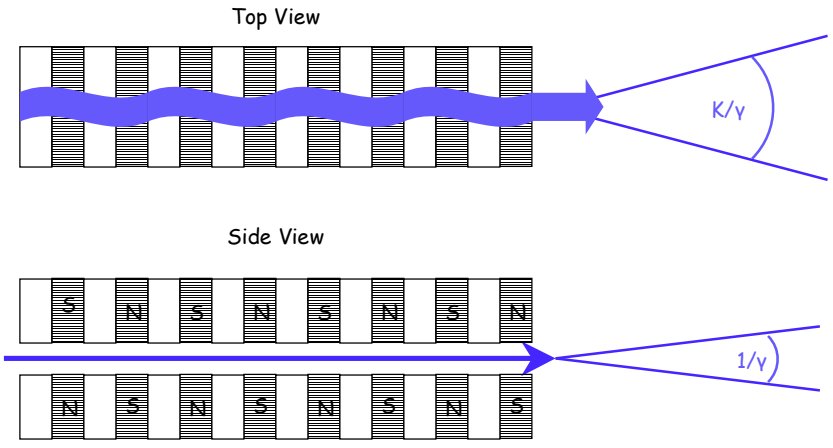


Dipole radiation pattern of accelerated charges is "folded" forward by special relativity, producing "search light" radiation pattern.

Characteristic opening angle is given by

$\gamma^{-1} = (E/mc^2)^{-1} \sim 10^{-4} \sim 0.1$ mrad

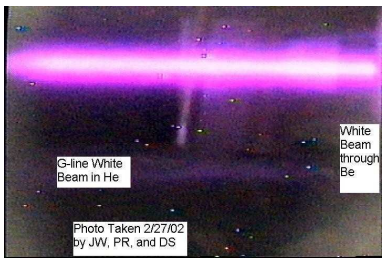
Increase Power with Wiggler Source



Why bother?

At comparable energy and angular resolution…

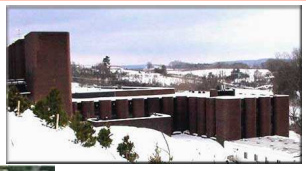
- **Rotating anode generator: ~10^7 x-rays/second**
- **Synchrotron source: 10^{12} – 10^{15} x-rays/second**



Not your father's x-ray source!

X-ray Synchrotron Sources

CHESS
Cornell High Energy Synchrotron Source



NSLS
National Synchrotron Light Source

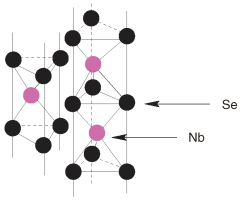
Advanced Photon Source
Argonne National Laboratory

$NbSe_3$ Properties

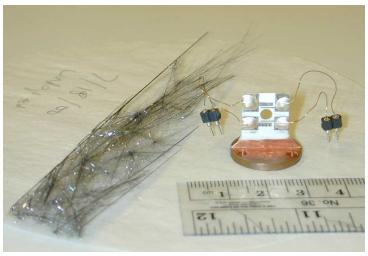
- monoclinic unit cell

  $a = 10.009$ Å
  $b = 3.4805$ Å $\beta = 109.47$
  $c = 15.629$ Å

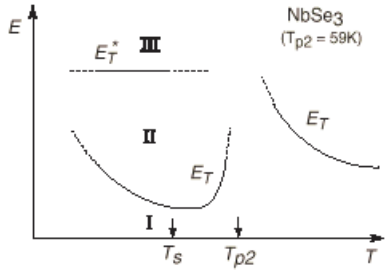
Hodeau *et al.*, J. Phys. C **11**, 4117 (1978)



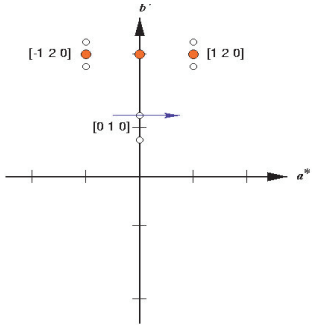
- whisker axis || to **b, b***
- width || **c**
- thickness || **a***
- $T_{P1} \approx 145$ K, $T_{P2} \approx 59$ K
- $Q_1 \approx (0\ 0.243\ 0)$
- $Q_2 \approx (0.5\ 0.26\ 0.5)$



$NbSe_3$ Properties



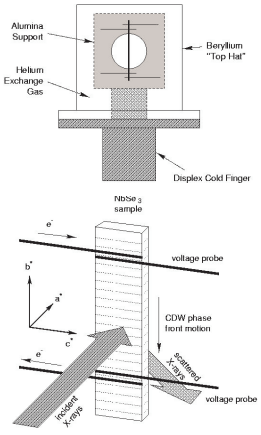
E-T Phase Diagram



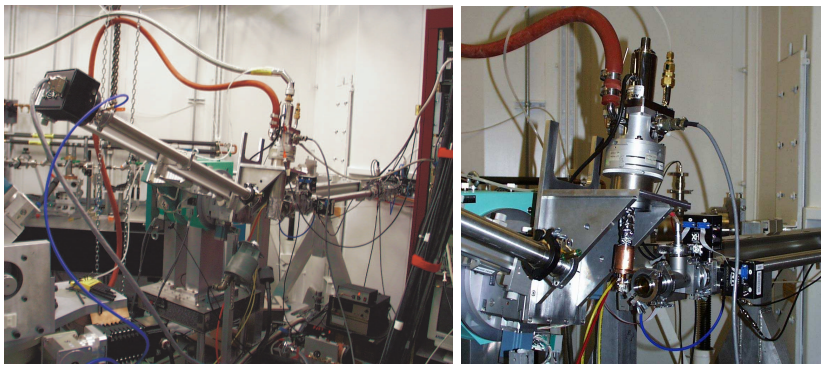
Reciprocal Space

Experimental Geometry

- **Beryllium Top Hat**
  - **x-ray transparent**
  - **contains He exchange gas**
- **Alumina Substrate**
  - **supports $NbSe_3$ whisker**
  - **four point connections**
- **Transmission Scattering Geometry**



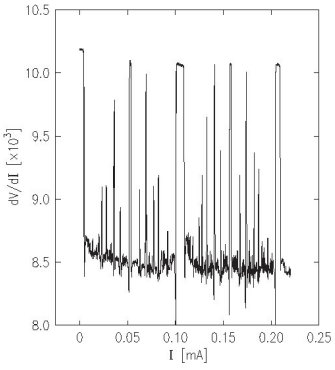
Experimental Geometry



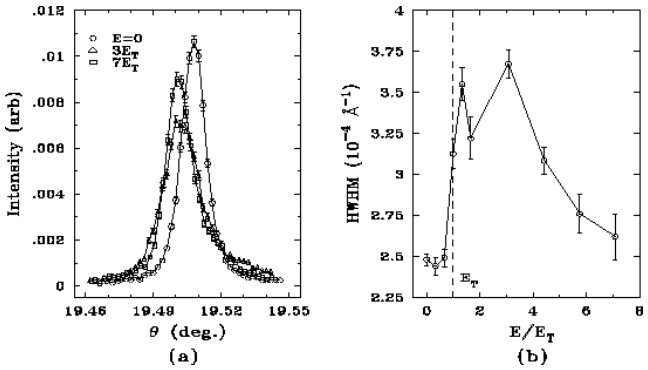
Sample Characterization

To study 'intrinsic' CDW properties, we need to make sure that there are no sample defects that might create extrinsic effects. Therefore, the criteria for choosing samples are:

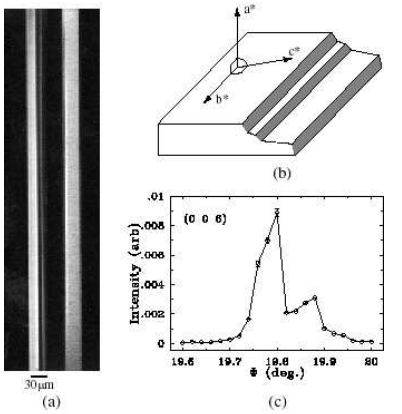
- Very regular cross sections
- Darwin limited (020) Bragg peaks
- Excellent mode-locking characteristics



Observe Predicted Recovery of QLRO?

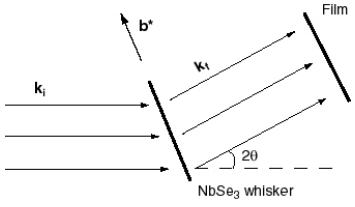


Diffraction Topography

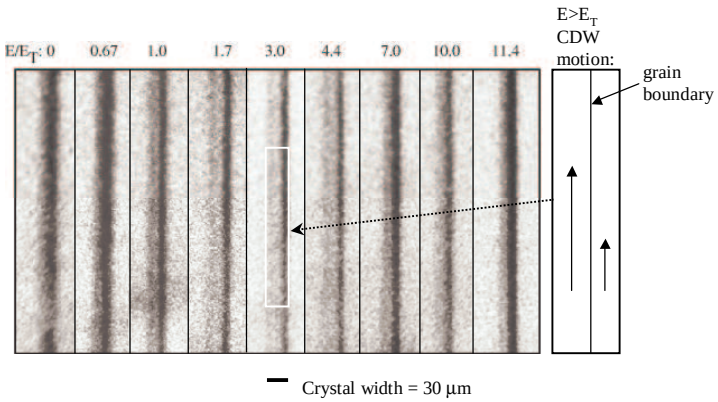


Y. Li, *et al.*, PRL **83**, 3514 (1999).

Use diffraction topography to image phase shifts across boundaries

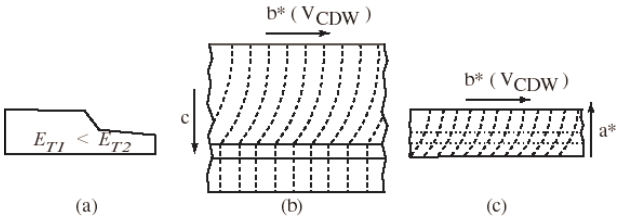


Due to grain structure of sample!



Y. Li, *et al.*, PRL **83**, 3514 (1999)

Shear in Sliding CDWs



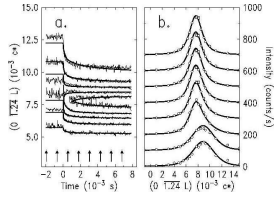
Y. Li, *et al.*, PRL **83**, 3514 (1999)

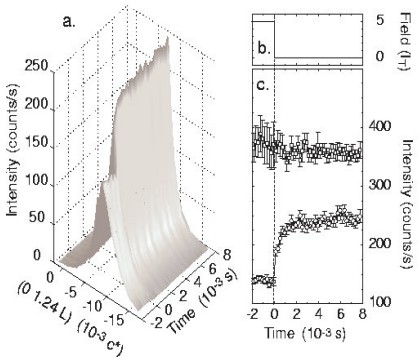
Time-resolved Experiments

Weak scattering signal requires signal averaging or time-periodic signal.

"Typical" Pinning Data

- Current is on for $t < 0$
- Current is off for $t > 0$
- CDW satellite responds by
  - **sharpening**
  - **(possibly) small rotation**
  - **integrated intensity remains constant**





K.L. Ringland *et al.*, PRL **82**(9), 1923-26 (1999).

Fukuyama-Lee-Rice Theory

Microscopic Model (based on Fröhlich Hamiltonian)

- P.A. Lee, T.M. Rice and P.W. Andersen. "Conductivity from charge or spin density waves." Solid State Comm., **14**, 703 (1974).

Phase only approximation (FLR)

- H. Fukuyama. "Pinning in Peierls-Fröhlich state and conductivity." J. Phys. Soc. Japan, **41**(2), 513 (1976).
- H. Fukuyama and P.A. Lee. "Dynamics of charge density wave. I. Impurity pinning in a single chain." Phys. Rev. B, **17**(2), 535 (1978).
- P.A. Lee and T.M. Rice. "Electric field depinning of charge density waves." Phys. Rev. B, **19**(8), 3970 (1979).

$$\Psi(x) = \overline{\psi}\, e^{i\phi(z)}$$

$$H_{FLR}[\phi(\vec{r},t)] = \int d\vec{r}\left\{\tfrac{K}{2}(\nabla\phi)^2 + \sum_i \rho_{CDW}\, V(\vec{r}-\vec{R}_i)\cos(\vec{Q}\cdot\vec{r}+\phi) - E\phi\right\}$$

Solution to Equation of Motion

- **Use FLR Hamiltonian**
- **Ignore thermal noise**
- **E=0**
- **Spatial Fourier transform**
- **Integrate EOM**
- **Assume δ-correlated, quenched random-field**
- **Integrate to obtain *g(r,t)***

$$\partial_t\phi = D\nabla^2\phi + V(\vec{r})$$

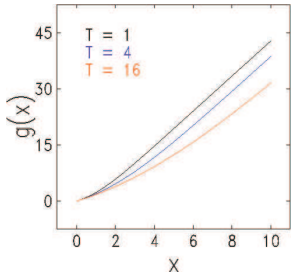
$$\left\langle\left|\tilde{\phi}(\vec{q},t)\right|^2\right\rangle = \left\langle\left|\tilde{\phi}(\vec{q},0)\right|^2\right\rangle e^{-2q^2Dt} + \frac{\left\langle\left|\tilde{V}(\vec{q})\right|^2\right\rangle}{D^2q^4}\left(1-e^{-q^2Dt}\right)^2$$

$$g(\vec{r},t) \cong \int_{1/r}^{\infty} d^3\vec{q}\left\langle\left|\tilde{\phi}(\vec{q},t)\right|^2\right\rangle$$

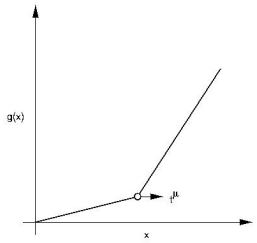
B. Jancovici, Phys. Rev. Lett., **19**, 20 (1967).

Approximate Form for g(x,t)

Numerical Integration of FLR Solution



Approximation to FLR Solution



$$g(x,t) \equiv \int_{1/x}^{\infty} d^3\vec{k} \left\langle \left|\phi(\vec{k},t)\right|^2 \right\rangle$$

$$g(x,t) \equiv \begin{cases} x/\xi_i & for\ x/t^{\mu} \geq 1 \\ x/\xi_f & for\ x/t^{\mu} \leq 1 \end{cases}$$

Approximate form for S(q,t)

Piece-wise integration using approximate form for *g(x,t)*

$$S(\vec{q},t) \approx \int_0^{t^{\mu}} d^3\vec{r}\ e^{i(\vec{G}\pm\vec{Q}-\vec{q})\vec{r}}\ e^{-|\vec{r}|/\xi_f} + \int_{t^{\mu}}^{\infty} d^3\vec{r}\ e^{i(\vec{G}\pm\vec{Q}-\vec{q})\vec{r}}\ e^{-|\vec{r}|/\xi_i}$$

$$S(\vec{q},t) \approx \frac{\xi_f}{1+\xi_f^2(q_\perp - G_\perp)^2}\left[1-e^{-(t/\tau)^{1/2}}\right] + \frac{\xi_i}{1+\xi_i^2(q_\perp - G_\perp)^2}e^{-(t/\tau)^{1/2}}$$

Approximate form for S(q,t)

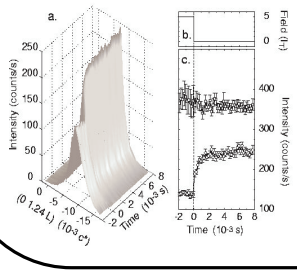
- $\mu \neq ½$, but keep $\alpha = ½$
- initial (sliding) state not Lorentzian
- integrate over broad directions of resolution function

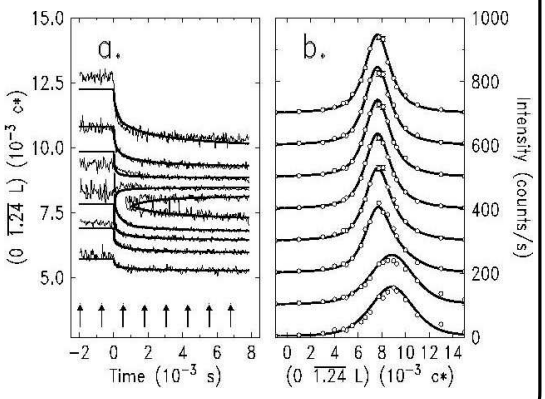
$$S(q_\perp, t) \approx \frac{\xi_f}{1+\xi_f^2 (q_\perp - G_\perp)^2} \left[1 - e^{-(t/\tau)^\mu}\right] + S(q_\perp, t=0)\, e^{-(t/\tau)^\mu}$$

Simple "stretched exponential" kinetics

Results of Analysis

- Solid lines are best fit results using only data from $t > \tau$
- 2 parameter fit (μ and τ)
- $\chi^2 \approx 1.5$
- $\sim 2 \times 10^4$ degrees of freedom.
- Arrows indicate times for the "slices" depicted.

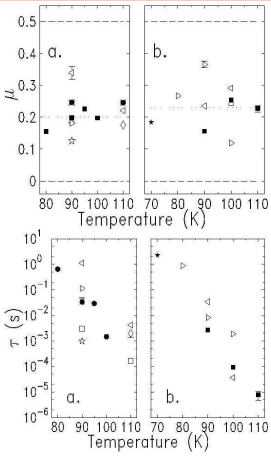




K.L. Ringland *et al.*, PRL **82**(9), 1923-26 (1999).

Results of Analysis

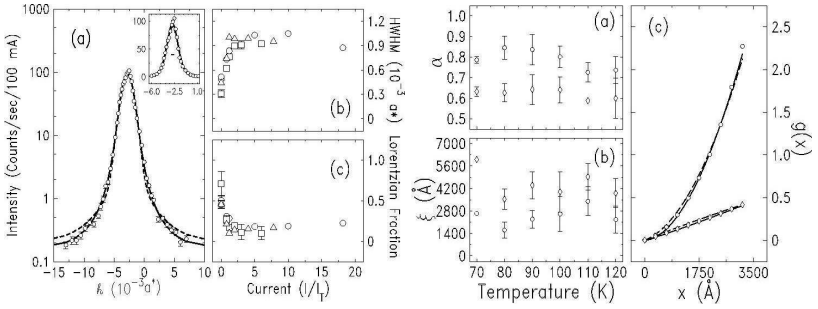
- μ is constant within fitting errors
- μ is not consistent with the value 1/2
- doping with tantalum does not have a statistically significant effect
- τ varies by nearly 6 orders of magnitude.
  This suggests an activated process with an activation energy barrier roughly equal to the CDW gap



K.L. Ringland *et al.*, PRL **82**(9), 1923-26 (1999).

Structure of Sliding State

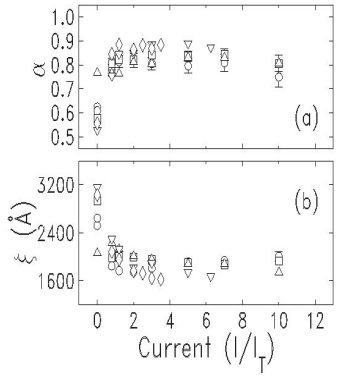
- **Expect** $g(x) \approx x^{2\alpha}$
- **Sliding state has α = 0.8±0.01**
- **pinned state has α = 0.6 ±0.01**
- **$\xi_{sliding} < \xi_{pinned} < \xi_{zero\text{-}field}$**

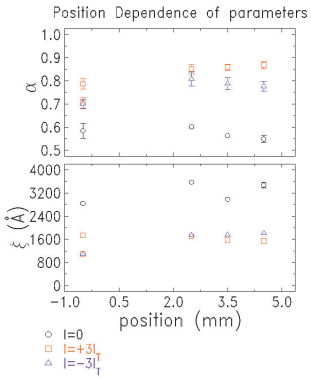


K.L. Ringland *et al.*, PRB **61**, 4405 (2000)

Structure of Sliding State (cont.)

α and ξ are insensitive to the value of the current, to the temperature or to the position on the sample.





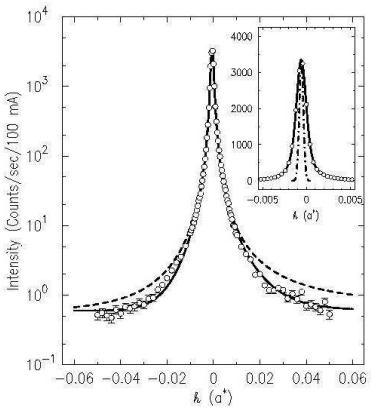
Structure of Sliding State (cont.)

- **Prediction is that system becomes more ordered at higher drives.**
- **Observation is that system disorders when it depins and does not reorder at accessible drives.**

Relationship to Zero-Field Cooled State

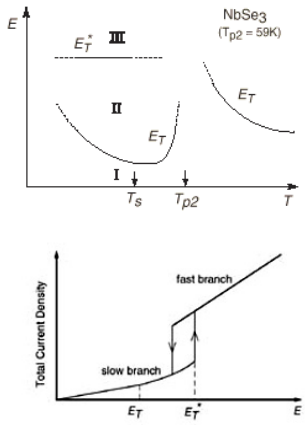
- **Zero-field Cooled**
- **New length scale required to describe short length scale behavior**
- **FLR Model is accurate**

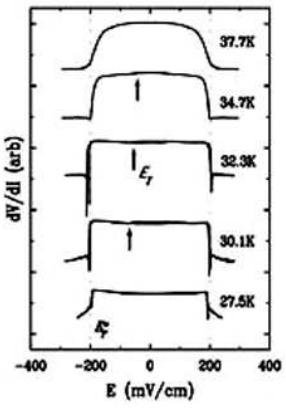
$$g(x) \cong \frac{\sqrt{a^2 + x^2} - a}{\xi}$$



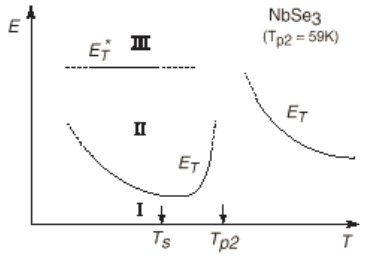
Brock *et al.*, PRL **73**, 3588 (1994)

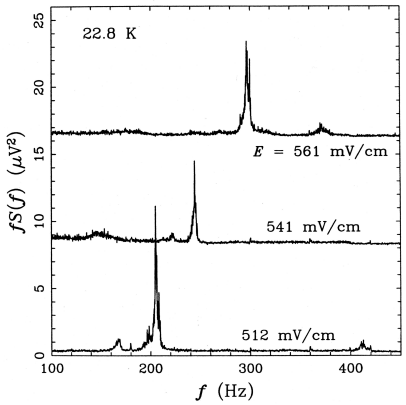
Low Temperature Behavior: Switching





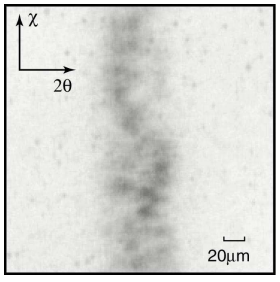
Coherent Flow in Region II!

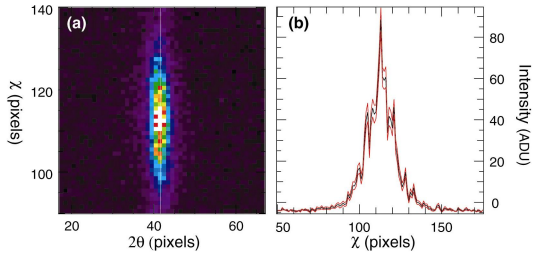




Lemay, *et al.* PRL, **83**, 2793 (1999)

Coherent X-rays and Speckle: Equilibrium Dynamics





Field Broadening

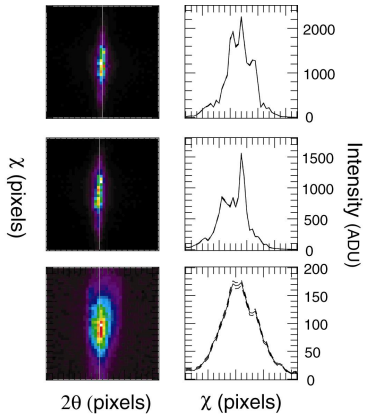
Q_1 CDW, T = 100K

Top: I = 0

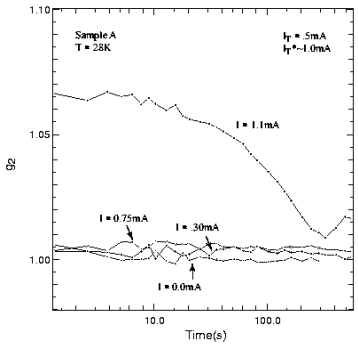
Center: $I = .8 \times I_t$

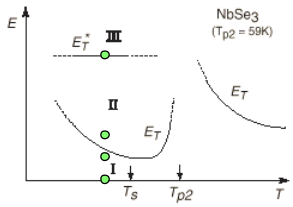
Bottom: $I = 2 \times I_t$

Averaged over 200 frames and 5 sec each.



Autocorrelation Function Results





E-T Phase Diagram

$E < E_T$: Static

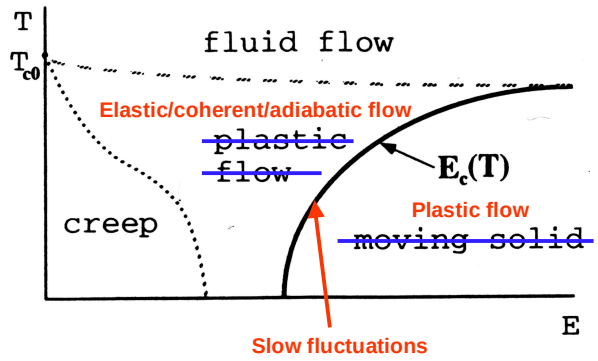
$E \geq E_T$: Static

$E \approx E_T^*$: Dynamic

$E >> E_T^*$: "Fast"

Suggests that we've got phase diagram "backwards."
Static (creep) → Adiabatic (elastic) → Plastic

Summary

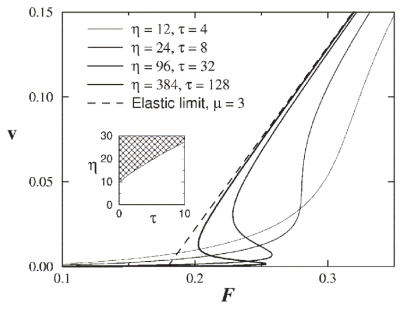


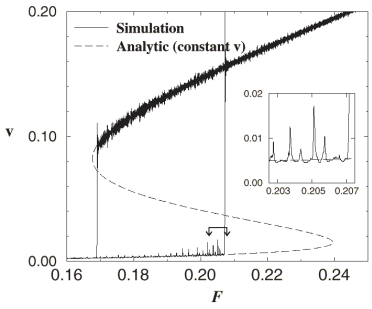
So what should we do?

Need to expand our description **of pinning/depinning**

- **New models**
  - **Amplitude fluctuations may be important**
  - **Elastic interactions may not be sufficient**
- **More data**
  - **Nail down XPCS data**
  - **X-rays measures lattice-distortion wave. Transport measures conduction electron density wave.**

Viscoelastic Coupling



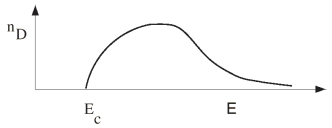


M.C. Marchetti, *et al.*, PRL **85**, 1104 (2000).

Replace elastic interaction of FLR model by couplings to the local velocity field that are non-local in time.

$$\gamma_0 \dot{u}_i = \sum_{(i,j)} \int_0^t ds\, C_{i,j}(t-s)\left[\dot{u}_j(s) - \dot{u}_i(s)\right] + F + F_i(u_i)$$

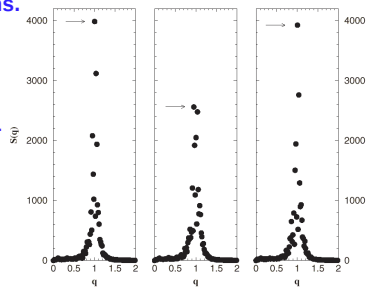
Modified Swift-Hohenberg Equation



Include both amplitude and phase fluctuations.

$$\frac{\partial \psi}{\partial t} + E\frac{\partial \psi}{\partial x} = \left[\varepsilon - \left(q_c^2 + \frac{\partial^2}{\partial x^2}\right)^2 + 4q_c^2\frac{\partial^2}{\partial y^2}\right]\psi - \psi^3 + \sum_i V_{imp}^i$$

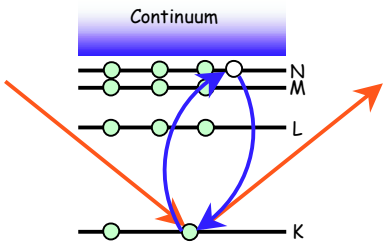
Dislocations proliferate near depinning transition. The dynamical generation of dislocations can make the system *more disordered* above threshold.



M. Karttunen, *et al.*, PRL **83**, 3518 (1999)

Resonant Enhancement

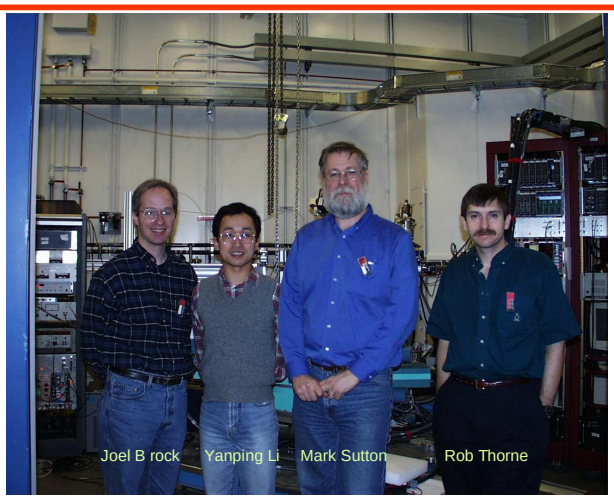
Resonant Scattering



- Use resonant x-ray scattering to sample (diverging) unoccupied density of states of conduction band at $2k_F$.
- Polarization analysis to suppress scattering from LDW

This September at CHESS

"The Crew"



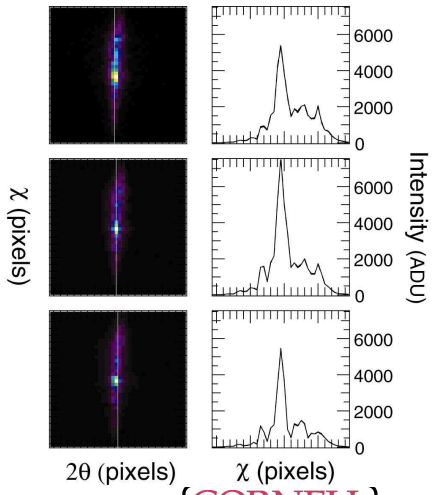
Effect of Slit Sizes

Speckle patterns taken on Q_1 CDW satellite with vertical slits sizes at

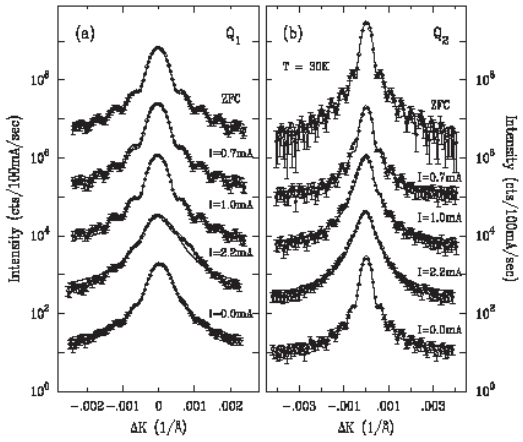
45 mm (top),

30 mm (center),

12.5 mm (bottom)

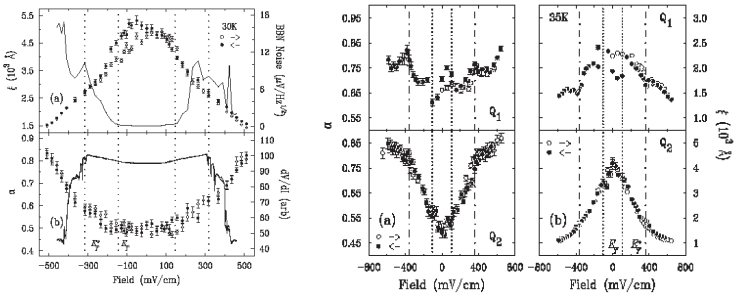


Finite Size Oscillations Demonstrate Coupling



Y. Li, *et al.*, PRB **63**, 041103(R) (2001)

Low Temperature - switching - regime



Y. Li, *et al.*, PRB **63**, 041103(R) (2001)

Observations

Non-Ohmic conductivity

- P. Monceau, N.P. Ong, A.M. Portis, A. Meerschaut, and J. Rouxel. "Electric field breakdown of charge-density-wave-induced anomalies in $NbSe_3$." *Phys. Rev. Lett.*, **37**(10), 602 (1976).
- R.M. Fleming and C.C. Grimes. "Sliding-mode conductivity in $NbSe_3$: Observation of a threshold electric field and conduction noise." *Phys. Rev. Lett.*, **42**, 1423 (1979).
- H. Matsukawa and H. Takayama. "Numerical study of statics and dynamics of charge-density-waves based on Fukuyama-Lee-Rice model." *Physica B*, **143**, 80 (1986).

Mode-Locking

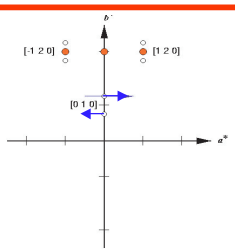
- J.C. Gill. "Transient non-linear phenomena and metastable states of charge-density waves in niobium triselenide." *Solid State Comm.*, **39**, 1203 (1981).
- M. Sherwin and A. Zettl. "Complete charge-density-wave mode locking and freeze-out of fluctuations in $NbSe_3$." *Phys. Rev. B.*, **32**(8) 5536 (1985).
- S.N. Coppersmith and P.B. Littlewood. "Interference phenomena and mode locking in the model of deformable sliding charge-density waves." *Phys. Rev. Lett.*, **57**(15), 1927 (1986).

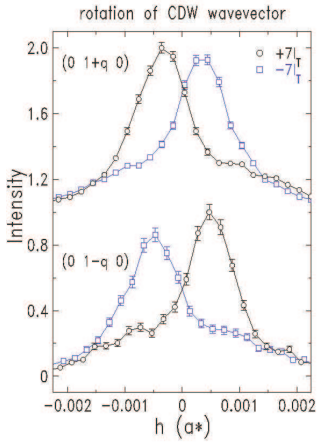
Narrow-Band Noise

- R.M. Fleming and C.C. Grimes. "Sliding-mode conductivity in $NbSe_3$: Observation of a threshold electric field and conduction noise." *Phys. Rev. Lett.*, **42**, 1423 (1979).
- S. Bhattacharya, M.J. Higgins and J.P. Stokes. "Harmonic generation and scaling behavior in sliding-charge-density-wave conductors." *Phys. Rev. Lett.*, **63**(14), 1503 (1989).
- P. Sibanl and P.B. Littlewood. "Critical dynamics of a pinned elastic medium in two and three dimensions: A model for charge-density waves." *Phys. Rev. Lett.*, **64**(11), 1305 (1990).

Rotations of Q

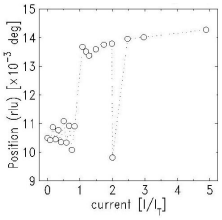
- Frequently, when CDW is sliding, the position of the satellite peak "rotates" away from the symmetric E=0 position
- The 1+Q satellite rotates in the opposite sense as the 1-Q satellite.
- The amount of rotation does not depend on position in the whisker.

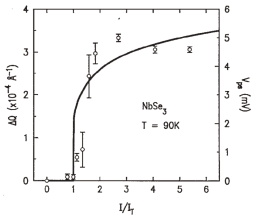




Rotation of Q (cont.)

- The rotation occurs rapidly as the E-field is turned on
- The form of this rotation is similar to that of the phase-slip induced strain and the phase-slip voltage, V_{PS}



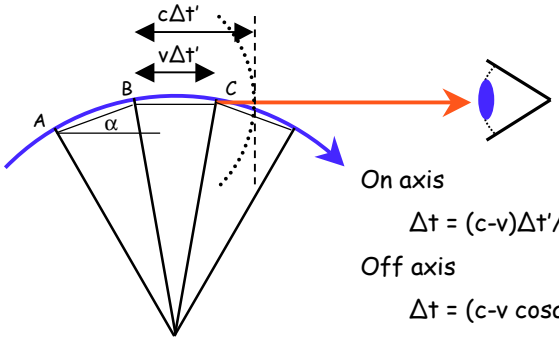


DiCarlo *et al.*, PRL **70**(6) 845-8 (1994)

Relationship to Surface Scattering/Growth

	CDWs	Surface Scattering/Growth
Structure Factor	$S(\vec{q},t) \propto \left\|J_1(\vec{q}\cdot\vec{u})\right\|^2 \int d^3\vec{r}\; e^{i(\vec{G}\pm\vec{Q}-\vec{q})\vec{r}} e^{-g(\vec{r},t)}$	$S(\vec{q},t) \propto \frac{1}{\left\|(\vec{G}-\vec{q})\cdot\hat{n}\right\|^2} \int d^3\vec{r}\; e^{i(\vec{G}-\vec{q})\vec{r}} e^{-q_z^2 g(\vec{r},t)}$
Correlation Function	$g(\vec{r},t) \equiv \frac{1}{2}\left\langle\left[\phi(\vec{r},t)-\phi(\vec{0},t)\right]^2\right\rangle$	$g(\vec{r},t) \equiv \frac{1}{2}\left\langle\left[h(\vec{r},t)-h(\vec{0},t)\right]^2\right\rangle$
Equation of Motion	$\partial_t\phi = -\Gamma\frac{\delta H_{FLR}}{\delta\phi} + \eta(\vec{r},t)$	$\partial_t\phi = -\Gamma\frac{\delta H}{\delta\phi} + \eta(\vec{r},t)$

Opening angle of Synchrotron Radiation



On axis

$\Delta t = (c-v)\Delta t'/c$

Off axis

$\Delta t = (c - v\cos\alpha)\Delta t'/c$

$$\Delta t \cong \left[1-\left(1-\frac{1}{2\gamma^2}\right)\left(1-\frac{\alpha^2}{2}\right)\right]\Delta t' \cong \left[\frac{1+(\alpha\gamma)^2}{2\gamma^2}\right]\Delta t'$$

Spectrum of Synchrotron Radiation

If T is period of revolution,

$\Delta t = [\gamma^{-1}/2\pi]\ T = 1/(\gamma\omega_0)$

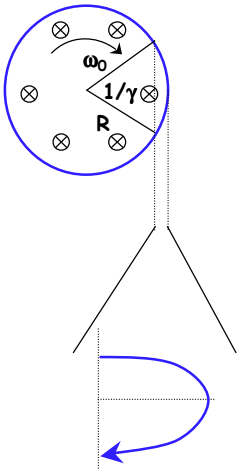
Observer "sees" an interval γ^2 shorter

$\Delta t = 1/(\gamma^3\omega_0)$

Inverse of pulse width gives frequency spread, so high cut-off frequency is

$\omega_c \equiv (3/2)\gamma^3\omega_0$

$E_c\ [KeV] = 0.665\ E_e^2[GeV]\ B[T]$



Sub-Threshold Quenches

- Clearly, phase-slip is occurring and FLR model is not able to describe the dynamics of pinning.
- Quenches from $I < I_T$ should avoid this difficulty.
- Preliminary data suggest that $\mu = ½$ for sub-threshold quenches -- consistent with FLR prediction.
- τ appears to be constant -- also consistent with FLR -- but data is not good enough yet.