Ultrafast Switching to a Stable Hidden Quantum State in an Electronic Crystal

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Transitions...in time

Stock market crashes

Elementary particle collisions

The Big Bang - hidden universes

D.M. What can physics tell us about stock market crashes, TEDx, Dec. 2013
The Big Data challenge

G. W. Burr, IBM, 2013

Problem (& opportunity): The access-time gap between memory & storage

Storage Class Memory

Access time...
(in ns)

Decreasing co$t

ON-chip memory

OFF-chip memory

ON-line storage

OFF-line storage

10^0

10^1

10^2

10^3

10^4

10^5

10^6

10^7

10^8

10^9

10^10

CPU operations (1ns)
Get data from L2 cache (<5ns)
Get data from DRAM/SCM (60ns)

Read a FLASH device (20 us)
Write to FLASH, random (1ms)
Read or write to DISK (5ms)

Get data from TAPE (40s)

1980

CPU

RAM

DISK

TAPE

Today

CPU

RAM

FLASH SSD

DISK

TAPE

Memory/storage gap
Our aim is to investigate trajectories of systems though symmetry-breaking transitions under nonequilibrium conditions, in real time.
Why CDWs?

CDW in 1T-TaS$_2$
Aharonov-Bohm effect in charge-density wave loops with inherent temporal current switching

M. Tsubota¹, K. Inagaki¹,², T. Matsuura¹ and S. Tanda¹,²

![Image]
Optical experiments: (at JSI)

- Ljupka Stojchevska
- Igor Vaskivskyi
- Tomaz Mertelj
- Jan Gospodaric
- Primoz Kusar
- Roman Yusupov
- Ian Mihailovic (IJS, FE-Uni-Lj)

Samples+

- I. Fisher (Stanford)
- P. Sutar (JSI)
- Lithography: D. Svetin (JSI)

Theory
- Serguei Brazovskii (Univ. Paris Sud Orsay)
- Patrick Kirchman
- ZX Shen (Stanford)

Time resolved ARPES
- Roman Yusupov
- P. Sutar (JSI)
- Lithography: D. Svetin (JSI)

Current switching experiments
- Ian Mihailovic (IJS, FE-Uni-Lj)
Destruction of Superconductivity by Laser Light

L. R. Testardi

Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 27 January 1971)

Superconductivity is destroyed by laser light in Pb films of thickness comparable to the optical penetration depth $\delta$ and less than the superconducting coherence length $\xi$. Thermal effects, which have been independently determined, cannot account for this. For films of thickness greater than $\delta$ and $\xi$, only the thermal effect is observed. In a proposed explanation it is shown that the electron gas may be heated from 3 to 18 °K above the lattice temperature by the light absorption in these experiments.
The electron relaxation process

Just heating ($T_e^* = T_L^* = \ldots$).
No doping.
The importance of e-h asymmetry for reaching photoinduced hidden states
The observable elementary excitations
with femtosecond spectroscopy

1. Quasiparticle (fermionic) excitations (detect the presence of a gap)

![Diagram showing quasiparticle excitations]

2. Collective mode (bosonic) excitations

The amplitude and phase modes

\[ \Psi = \Delta e^{i\varphi} \]
A theory should be more simple than the observations it is trying to describe.

(It should have fewer parameters)
The non-linear energy functional

The Landau non-linear energy functional originally written to describe a structural phase transition:

\[ F = \alpha \Psi^2 + \beta \Psi^4 + H \Psi \quad \text{where} \quad \alpha = \alpha_0(T - T_c) \]

The Ginzburg-Landau equation for a superconductor:

\[ F = F_0 + \alpha |\psi|^2 + \frac{\beta}{2} |\psi|^4 + \frac{1}{2m} (-i\hbar \nabla - 2eA)\psi|^2 + \frac{|B|^2}{2\mu_0} \]

Complex order parameter

\[ \Psi = \Delta e^{i\phi} \]

Lagrangian density, includes K.E.

\[ L(\phi) = \partial_\mu \phi^* \partial^\mu \phi - \alpha \phi^* \phi - \frac{\beta}{2} |\phi^* \phi|^2 \]
Time-dependent GL equation

Serguei Brazovskii, 2010

The energy of the system can be described in terms of a time-dependent Ginzburg-Landau functional†:

\[
F = \left(\alpha \Psi^2 + \beta \Psi^4 + H \Psi\right)
\]

where instead of the usual temperature dependence \((T - T_c)\), the first term is time-dependent:

\[
\alpha = \left[1 - \frac{T_e(t, r)}{T_c}\right]
\]

"Ergodicity"

The equation of motion is obtained via the Euler-Lagrange theorem:

\[
\frac{1}{\omega_0^2} \frac{\partial^2}{\partial t^2} A + \frac{\alpha}{\omega_0} \frac{\partial}{\partial t} A - (1 - \eta) A + A^3 - \xi^2 \frac{\partial^2}{\partial z^2} A = 0
\]

The order parameter, \( \psi(t) = A(t)e^{i\phi(t)} \)

†Phase fluctuations are assumed to be slow.
The predicted optical response of the collective mode

The reflectivity, $\Delta R(t) \propto \left( \frac{\partial R}{\partial \epsilon} \right)_\Delta \epsilon \propto \int [A^2_{DP}(t, r, \Delta t_{12}) - A^2_D(t, r)] e^{-z/\lambda} d^3 r.$
The response of the probe in all-optical experiments

1. Photoinduced absorption (PIA):

The polarisation selection rules are determined by the dielectric tensor

2. Coherent Raman-like (CRS) process:

The polarisation selection rules are governed by the Raman tensor $\chi_{kl}$

CRS and PIA probe processes can be distinguished by polarisation selection rules


The tritellurides are layered, quasi 2-dimensional metals with an orthorhombic (pseudo-tetragonal) crystal structure $\text{Cmca (D}_{2h})$

They exhibit a purely electronically driven 2nd order incommensurate CDW transition at $T_{c1} = 230\sim330K$

An AFM state exists at low $T_N$, some compounds exhibit another transition at low $T_{c2}$.

A Superconducting transition exists with $T_c = 3.5 K$ under a pressure of 75 kbar.

DiMasi ’94,’95, Fisher ’05,’08
The transient reflectivity $\Delta R/R$ after a quench at $\Delta t_{12}=0$ in TbTe$_3$
Quasi-particle (Fermion) evolution through $t_c$: gap recovery

$\Delta R/R (10^3)$

$\Delta t_{23}$ (ps)

$\Delta t_{12}$ (ps): 200, 30, 10, 8, 6, 4, 2.5, 1.6, 1.0, 0.75, 0.5, 0.35, 0.2

QP peak

Collective mode

Gap recovery

$\delta R = -\frac{2A\Delta^2}{\omega^2}n_p$.

$\tau = 650$ fs

Relaxation time recovery

$\tau \propto \frac{1}{\Delta}$
The collective mode (boson) spectrum as a function of time after quench

The most obvious feature: oscillations of intensity of the collective mode
Order parameter calculation for an inhomogeneous system

The eq. of motion:

\[
\frac{1}{\omega_0^2} \frac{\partial^2}{\partial t^2} A + \frac{\alpha}{\omega_0} \frac{\partial}{\partial t} A - (1 - \eta) A + A^3 - \xi^2 \frac{\partial^2}{\partial z^2} A = 0
\]

Calculated \( A(z,t) \) after quench:

Experimental parameters:

\( \tau_{QP} = 650 \text{ fs} \)
\( \omega_0/2\pi = 2.18 \text{ THz} \)
\( \eta = 2 \)
\( \alpha = 0.1 \)
Order parameter dynamics: theory vs. experiment

Theory predictions:
- Oscillations of $\Delta$ or $|\Psi|
- Critical slowing down
  (Collective mode softening)
- Domain annihilation
- $\Psi$ field (Higgs) waves

Experimental observations:
- Intensity oscillations
- Softening of $\omega$
- Distortions in $\omega$-t spectra
Critical dynamics near $t_c$.

Pre-transition behaviour: non-ergodic processes

“WIMP”s

Phonons: WIMPs from previous eons

“By analogy with the PM, dark matter excitations may be thought of as remnant excitations from before the SBT (the Big Bang).”

The trajectory to a hidden state in $1T$-$TaS_2$
What is a hidden state?

Hidden states of matter

A **hidden state** of matter is a state of matter which cannot be reached under ergodic conditions, and is therefore distinct from known thermodynamic phases of the material.[1] Examples exist in condensed matter systems, and are typically reached by the non-ergodic conditions created through laser photo excitation.[2][3] Short-lived hidden states of matter have also been reported in crystals using lasers. Recently a persistent hidden state was discovered in a crystal of Tantalum(IV) sulfide (TaS$_2$), where the state is stable for days at low temperatures.[4] A hidden state of matter is not to be confused with hidden order, which exists in equilibrium, but is not immediately

![Diagram showing a hidden system](image-url)
CDWs in 1T-TaS$_2$

A system with competing Coulomb, Fermi surface instability and lattice strain
Resistivity of $1T$-TaS$_2$ under equilibrium conditions

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{resistivity_graph.png}
\caption{Resistivity graph showing phases and temperature transitions.}
\end{figure}

- **Mott state**
  - (Commensurate CDW)

- **Incommensurate CDW**
  - Stretched honeycomb structure with domain walls in between
  - (Nearly Commensurate CDW phase)

- **Resistivity of $1T$-TaS$_2$**

  The resistivity measurements reveal the following phases and transitions:
  - At ambient pressure:
    - A metallic low-temperature phase, continuously evolving from the NCCDW state at ambient pressure.
    - Resistivity in the pressure range of 0–25 GPa and temperature range of 120–300 K for the whole pressure.
    - At pressures greater than 8 GPa, the resistivity is metallic-like.
    - Superconductivity first develops with pressure within the non-metallic phase.
  - At pressures greater than 8 GPa:
    - Fully metallic behaviour is present.
    - Transition from the incommensurate to the nearly commensurate CDW phase at 1.3 K to 300 K (Fig. 2).
    - NCCDW phase several tens of stars organize into roughly the kagome patchwork of the deformation is reduced, forming the planar structure that hexagonal CDW domains are separated by triangular regions where the amplitude hexagonal domains, locally reproducing the CCDW phase at temperatures above 550 K.
    - At temperatures above 550 K, an ICCDW phase appears.
    - At temperatures below 250 K, we observe a first-order transition.
    - The critical magnetic field would be of the order of 1.5 T.
    - Non-metallic over the entire temperature range for pressures of 0–4 GPa.
    - Superconductivity is observed at approximately 2.5 GPa, with a pressure of 0.8 GPa.
    - At low temperatures, the resistivity saturates fully suppressed at pressures of about 0.8 GPa.
    - The NCCDW phase is increased, and a metallic-like signature stabilizes in the incommensurate to the nearly commensurate CDW phase, which melts with a transition from the NCCDW to the CCDW phase, which melts at temperatures above 190 K.
  - At pressures of 0.8 GPa:
    - At temperatures above 550 K, an ICCDW phase appears.
    - At temperatures below 250 K, we observe a first-order transition.
  - At pressures of 2.5 GPa:
    - Metallic-like behaviour develops for low temperatures at pressures of 0.8 GPa.
    - Transition from the incommensurate to the nearly commensurate CDW phase.
  - At temperatures above 550 K, an ICCDW phase appears.
  - At pressures of 0.8 GPa:
    - Non-metallic over the entire temperature range for pressures of 0–4 GPa.
    - Superconductivity is observed at approximately 2.5 GPa, with a pressure of 0.8 GPa.
    - At low temperatures, the resistivity saturates fully suppressed at pressures of about 0.8 GPa.
    - The NCCDW phase is increased, and a metallic-like signature stabilizes in the incommensurate to the nearly commensurate CDW phase, which melts with a transition from the NCCDW to the CCDW phase, which melts at temperatures above 190 K.
Competing orders in 1T-TaS$_2$: Superconductivity under pressure, or Fe, or Se doping.

**Fe doping:**

**Pressure:**

Li et al. EPL 2012

Sipos et al (Nat.Mat. 2008)
$IT$-TaS$_2$: Collective mode switching

$W = 50$ fs “write”
$E = 50$ ps “erase”
$P =$ “pump” (50 fs)
$p =$ “probe” (50 fs)

Ljupka Stojchevska et al. Science 2014;344:177-180
Is the hidden state spectrum similar to any known equilibrium state?

No!
Switching to a hidden state in $1T$-TaS$_2$:
Resistance change after a (single) 35 fs pulse

$1T$-TaS$_2$ single crystal, ~100 nm thick.
Au contacts by laser lithography (LPKF LDI).

Dark resistance of 100nm film of $1T$-TaS$_2$

Resistance change after a (single) 35 fs pulse

$I$-TaS$_2$ single crystal, ~100 nm thick.
Au contacts by laser lithography (LPKF LDI).

Resistance after a laser pulse

L Stojchevska et al. Science 2014;344:177-180
Switching **only** occurs for short pulses $\tau_L < 4$ ps
Switching in ARPES

“Normal” ARPES:

Low Temperature ARPES of Switched 1T-TaS$_2$

Overview

C-state ERASE: multiple ~1μJ/cm$^2$ pulses or

Mott-gapped "VIRGIN" C-state

H-state WRITE: single >2mJ/cm$^2$ pulse changes
Landau theory of the CDWs in 1T-TaS$_2$

McMillan, PRB 1975

The Landau free energy: \[ F = F_1 + F_2 + F_3 \]

The order parameter (the charge density):
\[ \rho(\vec{r}) = \rho_0(\vec{r})[1 + \alpha(\vec{r})] \]
Where \[ \alpha(\vec{r}) = \text{Re}[\psi_1(\vec{r}) + \psi_2(\vec{r}) + \psi_3(\vec{r})] \]
\[ F_1 = \int d^2 r [a(\vec{r})\alpha^2 - b(\vec{r})\alpha^3 + c(\vec{r})\alpha^4 + d(\vec{r})(|\psi_1\psi_2|^2 + |\psi_2\psi_3|^2 + |\psi_3\psi_1|^2)] \]
\[ c(\vec{r}) = c_0 + c_1 \sum_i e^{i\vec{K}_i \cdot \vec{r}} \]

Impurity potential
\[ F_2 = \int d^2 r U(\vec{r})\rho_0(\vec{r})\alpha(\vec{r}) \]

Gradient terms:
\[ F_3 = \int d^2 r \left[ e(\vec{r}) \sum_i |(\vec{q}_i \cdot \vec{\nabla} - iq_i^0)\psi_i|^2 \right. \\
\left. + f(\vec{r}) \sum_i |\vec{q}_i \times \vec{\nabla} \psi_i|^2 \right], \]
where
\[ |\vec{q}_1| = |\vec{q}_2| = |\vec{q}_3| = 2\pi/\lambda, \]

The Fermi Surface of 1T-TaS$_2$ in the undistorted phase
The free energy of $1T$-TaS$_2$ with domain walls (the NC state)

Free energy:  
\[ F_c(n_c) = E_{DW}(C_0|n_c| + C_1|n_c|e^{-1/(\xi|n_c|)} - C_2\xi n_c^2 + C_4\xi^3 n_c^4) \]

\[ C - IC \text{ transition (MacMillan, 1975)} \]
\[ \text{Intersection of DW} \]
\[ \text{Repulsion between DW crossings} \]

Microscopic mechanism: Photo"doping" and subsequent ordering of voids

The photo-hole annihilates a polaron, creating a void.

\[ \text{Polaron annihilation by photoexcited hole} \]

\[ \Delta_{\text{Mott}} \]

\[ \Delta_{\text{CDW}} \]

\[ E \ (\text{eV}) \]

\[ h^+ \]

\[ h^- \]

\[ \text{Voids} \]

\[ \text{Domain walls} \]

L Stojchevska et al. Science 2014;344:177-180
Macroscopic quantum tunneling?

Condition for MQT:

The potential must change faster than the system can tunnel

Timescales:

The charge rearrangement in the H state occurs on a timescale:

\[ t_{\text{charge}} \sim \frac{\hbar}{E_{\text{Mott}}} \simeq 13\text{fs} \]

The polaron-forming ions reach new equilibrium positions on a timescale:

\[ t_{\text{ions}} \leq \frac{1}{4} T_{\text{AM}} \simeq 110\text{fs} \]

Tunneling time:

\[ t_{\text{tunneling}} \sim O(1) T_{\text{AM}} \simeq 500\text{fs} \]

Conclusion: the MQT condition is fulfilled
Kinetics cannot be described in a rigid band approximation.
**Electron and hole** relaxation kinetics beyond the rigid band approximation: **photodoping in terms of a time-dependent shift of the chemical potential**

Subject to conservation of charge

\[ n_e - n_h = n_v - n_i = n_d. \]

The kinetic equations for the electrons and holes:

\[
\begin{align*}
\frac{dn_h}{dt} &= -k_{eh}n_en_h(\mu_e + \mu_h) + k_{hd}n_h(\mu_h - \mu_d) + P(t) \\
\frac{dn_e}{dt} &= -k_{eh}n_e n_h(\mu_e + \mu_h) - k_{ed}n_e(\mu_e + \mu_d) + P(t)
\end{align*}
\]

\( \mu_i \) are time-dependent, and \( P(t) \) is the laser pulse.

Numerical solution:
**Chemical potential surfaces**

The chemical potentials:

\[ \mu_i = \frac{\partial F_i(n_i)}{\partial n_i} \]

for electrons, holes:

\[ \mu_{e,h}(n) = \Delta_{e,h} + k_B T \ln(e^{n_{e,h}/(k_B T N_{e,h})} - 1) \]

for the condensate:

\[ \mu_c(n_c) = E_{DW}(C_1(1 + \frac{1}{\xi|n_c|})e^{-1/(\xi|n_c|)} + C_0 - 2C_2\xi|n_c| + 4C_4(\xi|n_c|)^3)\text{sign}(n_c) \]

The system is stable in equilibrium when:

\[ \mu_e = \mu_h = \mu_c \]

where \( n_c = n_h - n_e \)
Calculated trajectory

Laser pulse energy above threshold ($U_W > U_T$):

The time evolution of $n_e$ and $n_h$:

System trajectory (parametric plot)
e-h Imbalance
C-Phase Near Threshold / H-State above Threshold

trARPES suggest that e-h imbalance drives switching
k-number conservation: A topological protection mechanism

The modulation wave vector

\[ \delta q = n/\pi \]

\( n \) is the number of void sites

The H and C states are topologically distinct
The relaxation mechanism (microscopic)

Relaxation of the H state

\[ R = R_0 \left[ 1 - \exp\left( -\left( t / \tau_H \right)^\beta \right) \right] \]

At low T (<10K) the lifetime is longer than the age of the Universe
Relaxation $\text{H} \rightarrow \text{C}$ (2).

Ostwald ripening

Incommensurate $\rightarrow$ commensurate

Relaxation (3)

Kontorova and Frenkel model (1938):

$$H = \int \left[ \frac{1}{2} \left( \frac{d\varphi}{dn} - \delta \right)^2 + V(1 - \cos p\varphi) \right] dn$$

$x_n$ is the position of the $n$th atom:

$$x_n = nb + \frac{b}{2\pi} \varphi_n$$

In the continuum limit: $\varphi_n - \varphi_{n-1} = d\varphi/dn$

A devil’s staircase

Only certain $q$-values are allowed
Energy-Efficient Superconducting Computing—Power Budgets and Requirements

D. Scott Holmes, Senior Member, IEEE, Andrew L. Ripple, and Marc A. Manheimer
Is it possible to rapidly switch to a H state by *charge injection*?

(It could be useful)
Switching to the H state using an ultrafast current source

Macroscopic Quantum Tunneling with current injection?

The state is spatially and temporally inhomogeneous.
**CDW memory performance**

**Switching speed**

![Diagram showing switching speed performance of different memory technologies.]

**Energy per bit**

![Graph showing energy per bit versus equivalent contact diameter for different memory technologies.]

**Equation:**

\[ E_B = \frac{\tau \omega^2 L w_t}{\rho_{xx}} \]

- \( E_B \): Energy per bit
- \( \tau \): Switching time
- \( \omega \): Angular frequency
- \( L \): Channel length
- \( w_t \): Transistor width
- \( \rho_{xx} \): Sheet resistance

**1T-TaS₂ device**

- Energy \( E_W \approx 0.25 \) pJ

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I.V. I.A.M and D.M., UK patent pend., 2014
The significance of hidden states of matter

New elementary particles under non-ergodic conditions
(different beam energies)

Hidden universes?