Self-detection of mechanical oscillations of charge-density wave conductors

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1. Introduction (long)
   a) CDW deformation. Interaction with the lattice
   b) Torsional strain – the main features
   c) Why self-detection?
2. The inverse effect: TS-induced current modulation
3. Self-detection of torsional oscillations
   a) 3-contact way
   b) Heterodyne mixing technique with frequency modulation
   c) Application to the CDW compounds
   d) Towards high frequencies
4. The progress in physical understanding (negative)
CDW crystals

Experimental arrangements

For AFM

Photo of a TaS$_3$ batch grown in IRE.

For torsional measurements
CDW basics. CDW-lattice interaction.

CDW – is deformable electronic crystal, – a spring inside the crystal.

In the simple 1D model the equilibrium CDW wavelength follows the lattice deformation:

\[ \frac{\Delta \lambda}{\lambda} = \frac{\Delta L}{L} \equiv \frac{\Delta c}{c}, \]

where L – sample length, c – lattice period.

However, \( \frac{\lambda}{c} \) strongly depends on c. Thus, the two springs interact somehow giving rise to an “analogue” of the piezoelectric effect.
The CDW basics. CDW-lattice interaction.

\[ \rho = \text{const} \]

\[ u = u_0 \sin(qx + \phi) \]
\[ \rho = \rho_0 \sin(qx + \phi) \]

\[ W = \frac{1}{2} \left[ Y_l \left( \frac{c - c_{eq}}{c_{eq}} \right)^2 + Y_c \left( \frac{\lambda - \lambda_{eq}(c)}{\lambda_{eq}} \right)^2 \right] \]
Different methods – AFM, TEM, interferometry for different kinds of non-uniform deformation: longitudinal, flexural, **BUT unexpectedly:**

**Torsional strain (TS) of TaS$_3$ under electric field.**

1) Whiskers arrangement allows free torsion.
2) Optical (mirror) methods for the strain registration are developed.

![Diagram of TaS$_3$ with laser and micromirror setup for strain measurement.](image-url)
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Torsional strain up to $10^\circ$ (shear $\sim 10^{-4}$ at $E \sim 300\text{mV/cm}$, “piezomodulus” $> 10^{-4} \text{ cm/V}$) is observed – a powerful intrinsic actuator.

Related to the CDW depinning and deformation: Hysteretic, threshold.
The fast linear (thresholdless) contribution

\[
\begin{align*}
&10^{-10} \\
&10^{-1} \\
&10^0 \\
&10^1 \\
&10^2 \\
&10^3 \\
&10^4 \\
&10^5
\end{align*}
\]

\[\Delta \phi, \text{ a.u.}\]

- **a)** \(f_0 = 6.6 \text{ kHz}\)

\[
\begin{align*}
&10^{-1} \\
&10^0 \\
&10^1 \\
&10^2
\end{align*}
\]

\[\Delta \phi, \text{ a.u.}\]

- **b)**

\[
\begin{align*}
&10^{-1} \\
&10^0 \\
&10^1 \\
&10^2 \\
&10^3 \\
&10^4
\end{align*}
\]

\[\text{frequency, Hz}\]

\[\Delta \phi, \text{ a.u.}\]
High-frequency mechanical oscillations?

\[ \omega_0 = \frac{\pi}{L} \sqrt{\frac{G}{\rho}}, \quad \text{[flexural \ } \sqrt{\frac{E}{\rho}} \frac{2\pi}{L} \frac{R}{L}] \]

\( L \) – sample length, \( G \) - shear modulus, \( \rho \) – density.

\( \text{TaS}_3: \ G=5 \ \text{GPa}, \ \rho=6.4 \cdot 10^3 \ \text{kg/m}^3, \ L=1 \ \mu\text{m} \Rightarrow \ \omega_0=2.8 \ \text{s}^{-1}. \)

Tortional modulation of the CDW conductivity

Ac voltage across a TaS$_3$ sample induced by modulation of torsional angle. $f=10$ Гц. [J. Nickols et al., PRB 79, 241110R (2009)]

$(\delta V/V)/G \sim 1-10 \Rightarrow$ a good sensor $\Rightarrow$ self-detection

(a) Ac voltage across a TaS$_3$ sample induced by modulation of torsional angle. $f=3$ Гц, amplitude $4^\circ$, $L=3$ mm. b) $R_d$ vs $I$ at the same condition [V.Ya. Pokrovskii et al., Physics Uspekhi].
An arrangement for observation of torsional oscillations. The electric scheme providing the feedback is shown. AC voltage is applied across 1-2, dc current is set between 3-2. In the resonance modes the lock-in detects torsional modulation of $V_{32}$.

Detection of mechanical resonance with heterodine mixing—allows 2-probe detection!

\[ I = V \times \sigma = V_0 \cos(\omega t) \sigma_0 (1 + \delta \phi_0 \Pi \cos(\omega t + \Phi)) = V_0 \sigma_0 \cos(\omega t) + \delta \phi_0 \Pi \cos(2\omega t + \Phi)/2 + \delta \phi_0 \Pi \cos(\Phi)/2 \]

\[ (\Pi \equiv (\delta \sigma/\sigma)/\delta z \quad \text{or} \quad \Pi \equiv (\delta \sigma/\sigma)/\delta \phi) \]


J. Chaste et al., APL 99, 213502 (2011); 4.2 GHz & 11 GHz
Detection of mechanical resonance with heterodine mixing - elastic anomaly in NbSe$_3$

Shamashis Sengupta et al., PRL 110, 166403 (2013)

Growth of Young modulus by 13%!! CDW plasmon mode?
Detection of torsional resonances in TaS$_3$ with heterodyne mixing

1) Torsional modes are excited.
2) The oscillations are excited not with a gate, but with intrinsic for CDW forces
3) Current is modulated not due to field effect, but due to intrinsic CDW current modulation
Detection of torsional resonances in TaS$_3$ with heterodine mixing

Signals from Lock-in 1 (translated in nA/kHz) (a) and lock-in 2 (in degrees), (b) as a function of $f$, detected simultaneously. The inset: integrated and normalized signal from Lock-in 1. The sample: $L=1.1$ mm, $w=6.4$ µm. $T=96$ K
Detection of torsional resonances in TaS$_3$ with heterodyne mixing – $f$ increase?

Signal at $V=\pm150,180,260$ mV. $L=161 \, \mu$m, $w=4 \, \mu$m. $T=105$ K.
Without mirrors.
Detection of torsional resonances in TaS$_3$ with heterodyne mixing – $f$ increase?

Frequency dependencies of the mixing current at several amplitudes of ac. voltage across the sample: $V=\pm 150$, 230, 300 and 490 mV (the bottom panel). $L=161$ µm, $w=4$ µm. $T=105$ K.
The physical ground: 
the electron transitions in sulphides

\[ \Delta \lambda(c)/\lambda \neq \Delta L/L \equiv \Delta c/c \]

The most probable reason: dependence of \( k_F \) on deformation. A slight change in S-S distance can result in a notable change of free carriers concentration. In [Meerschaut & Rouxel, in Crystal Chemistry and Properties of Materials with Quasi-One-Dimensional Structures, 205-279. J. Rouxel (ed.) © 1986 by D. Reidel Publishing Company] it is emphasized that in \( \text{MX}_3 \) “a lengthening of the X-X pair corresponds to a weakening of the X-X bond. The X-X pair behaves as an electron reservoir, which governs indirectly the electronic density available along the metallic chains.” The X-X pairs may be bonded with each other or only with the M atom. The general formula of \( \text{MX}_3 \): \( M_x^{4+}M_{(2-x)}^{5+}(X_2)^{2-}_{x/2+1}X^{2-}_{4-x} \), with the \( x \) value very uncertain. But \( x \) governs the electron concentration. Its variation changes \( k_F \) and, thus \( q=2k_F \).

Charge disproportionation?
The simplest case:
uniaxial expansion under uniform CDW strain.

ΔL/L ≈ 6 \times 10^{-5}

Thermally-induced deformation:

A.V. Golovnya, V.Ya. Pokrovskii, P.M. Shadrin,
Experimental ideas: how to estimate \( \frac{\Delta \lambda (c)}{\lambda} / \frac{\Delta c}{c} \)?


Connecting \( L(q) \) with \( q(\varepsilon) \) through the Le Chatelier principle.
How to estimate \((\Delta \lambda(c)/\lambda)/(\Delta c/c)\)? – \(\sigma(\varepsilon)\) dependence.

\[ \varepsilon \approx \frac{\delta y \cdot t}{L^2} \]
How to estimate \( \frac{\Delta \lambda(c)/\lambda}{\Delta c/c} \)? – \( \sigma(\varepsilon) \) dependence.

\[ (\frac{\Delta \lambda(c)/\lambda}{\Delta c/c}) \approx -0.1, \text{ but not 5} \]

\[ \sim 1\% \]
The conclusions

1) A number of CDW conductors in the Peierls state show unique electromechanical properties.
2) CDW strains result in the sample deformations. TS is the most studied.
3) Large “torsioresistance” observed for $E \gg E_t$ allows self-detection of torsional oscillations of TaS$_3$.
4) The mixing technique with FM allows 2-probe detection of torsional resonances.
5) The frequencies achieved (25 MHz) are already comparable with plasma ones and promising for search of elastic anomalies.
6) Shortening samples results in frequency growth $\propto \frac{1}{L}$. GHz range?
7) The anomalous electromechanical properties of the CDW conductors can be treated in terms of elastic lattice-CDW interaction, which can be strong due to electron transitions between bounded and conducting states governed by strain-induced S atoms rearrangements. However, the picture could be more complex.
8) Direct structural studies (at LT, under strain) are required to reveal the structural and charge changes.