On the reproducibility of ultra-intense terahertz experiments: Comment on "Sub-cycle insulator-to-metal transition in vanadium dioxide by terahertz-field-driven tunneling" by Giorgianni, Vicario,.. and Hauri (arXiv:1706.00616)

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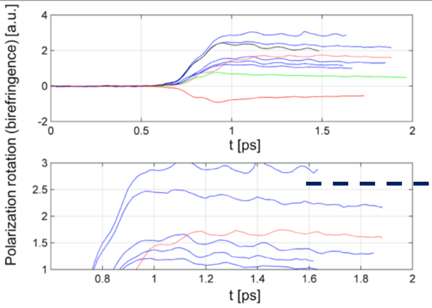
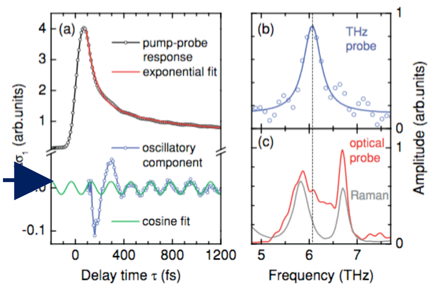
The manuscript by Giorgianni et al [1], and co-authored by myself, claims and concludes on a purely electronic insulator to metal transition (IMT) in VO_2 using ultra-intense THz fields without any lattice interaction. The underlying mechanism behind IMT in VO_2 is a highly debatable subject with the "electron-only" IMT concept being increasingly resisted in the field especially under intense excitations. If Giorgianni/Hauri's claim were sustained, it would be an important step towards fundamental understanding of complex systems such as the model VO_2 [2-4] and may have implications on low power ultrafast switching applications. The experiment was enabled by the uniquely-intense THz source I previously developed at the SwissFEL [5, 6] and is thus not possible to reproduce elsewhere under similar excitation conditions. However, some of the measurements under the reported experimental settings showed clear phonon oscillations on the short time scale, and sample damage using the maximum reported field intensities, which may be contradicting the purely electronic/non-phonon paper claims/conclusions. These measurements were known to all PSI authors (Feb./March 2017) before the paper was drafted. However, the principal authors (Giorgianni, Vicario, and Hauri) [1] decided to exclude these measurements from the paper, disapproved the call to further repeat and verify the paper measurements, and submitted it. They then publically published the text on the arXiv without the consent of all the authors.
The external co-authors (from U.C. Berkeley, USA, and Tsinghua U., China) requested the addition of the note "We (Kai Liu, Junqiao Wu and Kevin Wang) were unaware of the situation, of the hidden data, and of the PSI disagreements, and we have never been contacted or informed of any of these issues. We merely provided the samples."†
The experiment was performed at a SwissFEL laboratory, but not the experimental user laboratories. This comment should not lay responsibility on the SwissFEL facility. PSI attempted to withdraw the submission, but the arXiv is non-retractable.[2]

1. Phonon excitations

In transmission-type THz pump/optical probe spectroscopy, the basic experimental techniques are transmission modulation ΔT and birefringence/polarization rotation $\Delta\varphi$. Understanding of the structural dynamics in VO_2 strongly depends on the ultrafast electron and lattice dynamics which are coupled and difficult to disentangle using an optical probe. While ΔT reflects the conductivity change and IMT, $\Delta\varphi$ is the tool to trace phonon dynamics (excitations) [Table]. Generally speaking, phonons can also be measured through ΔT but the effect is negligible when compared to that of IMT [7]. The arXived text [1] shows only ΔT claiming purely electronic transition with no phonon excitations. As concluded, "*The pure electronic transformation is achieved by atomically strong THz field … … results in a prompt metallization without any lattice interaction* [1]." However, Fig. (a) shows measurements of $\Delta\varphi$ (excluded from the text [1]) with clear oscillation corresponding to 5.5-6 THz, phonon oscillations.

<table>
<tr><td>(a) Excluded measurements from Giorgianni, .. , Hauri, arXiv:1706.00616 [1]</td><td>(b) Pashkin, …., Leitenstorfer, Phys. Rev. B 83, 195120 (2015) [9]</td></tr>
<tr><td>
</td><td>
</td></tr>
<tr><td>Fig. (a) THz-induced $\Delta\varphi$ on a collinear 800 nm probe in 90 nm-VO_2. Different curves test on different THz excitation levels and 800 nm probe polarization. They all reveal traces of the 5.5-6 phonon. Exactly the same sample and experimental THz pump conditions as used for ΔT in [1].</td><td>Fig. (b) Phonon spectrum in VO_2 taken from ref. [9] and measured in a degenerate pump-probe experiment 1.55 eV. The spectrum corresponds to the THz oscillations in (a).</td></tr>
</table>

[1] In connection with [1] submission: Affiliations: C.V.: Staff scientist, SwissFEL, PSI; ext. funding: IZLRZ2_164051(Swiss SNF); C.P.H.: Head of the SwissFEL laser group, PSI/ EPFL (Lausanne); ext. funding: 200021_146769 (Swiss SNF).
[2] PSI: was informed on June 26th, 2017; requested from Nature Publishing Group to put the manuscript under review on hold on July 6th, 2017; attempted to remove the arXived version [1] on Sept. 1st, 2017.
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In the original text [1], these oscillations are not obvious because the paper shows only the transmission and focuses on long wavelength probe to magnify the change in the transmission [8]. Both ΔT and $\Delta\varphi$ were taken under the same conditions (experimental setup [6], sample, 2x 20 THz + 1x 6 THz QMC LPF excitation filters). Lower cut-off filters (4.2 THz) have also shown trace of phonon excitations. These oscillations match the previously reported phonons in VO_2 (Fig. b) [9,10].

2. Sample damage

The measurements in the paper [1] were done under very high field excitation of up to 18 MV/cm in the sub- 6 THz range. When the experiment was repeated (Fig. a), gradual sample damage was observed at such fields and exactly same experimental conditions in ref. [1] (such a damage at such a low repetition rate typically leads to irreproducible transition traces and reduction in the unmodulated transmission of the optical probe). Therefore, to take the scans in Fig. a, the maximum field was reduced (< 9 MV/cm) keeping the same spectral contents. This observation is consistent with previous report on damage around 4 MV/cm in [2]. However, the comparison between the two reports is complex. Ref. [2] uses higher (1 kHz) repetition rate (that has lower damage threshold) but the excitation frequency was low (~ 1 THz), this is stronger tunneling effect. In the present experiment, the repetition rate was 100 Hz but the excitation frequency was high and VO_2 absorption is higher. Nevertheless, sample damage changes the surface properties, transmission, and complicates the overall structural features being measured.

3. Conductivity calculation (minor)

The used formula "Tinkham" is an approximation that is valid only under specific conditions (when the film is so thin that the probe does' not experience phase change across the film). This is clearly not satisfied as the transmission change is > 30%. The fit on a logarithmic (tunneling) effect does not indicate much. Unless you have a very large dynamic range (not the case here), any high order function will probably give good matching.

Conclusion: *The published text on the arXiv by Giorgianni et al [1] does not reflect the opinion of all the listed authors. It does not fully represent the undertaken measurements. The excluded phonon measurements represent an integral part of the overall structural dynamics and may contradict the paper's conclusion. This case may raise concerns on the reproducibility of results in ultra-intense experiments in the THz community.*

Science is based on peer-review and reproducibility. Outside the SwissFEL laboratories, the most powerful existing THz sources (in the sub-6 THz range) are 100 times less intense (in W/m^2) than the one used for this experiment. Such intensities have shown to me nonlinear response from every sample I used in the recent years. However, extensive repetition and in-depth experimental analysis have proven that many of them were misrepresentations of results in an unprecedented regime of ultrastrong-low frequency fields. Most of these issues are not known in the field to the wide scientific (reviewers) community. Nor is there a way of getting such field intensities to reproduce such experiments elsewhere. In this regard, the underlying theory/simulations papers typically focus only on what they want to prove and do not deal with the associated unwanted other correlated dynamics. The author predicts this theory-experiment deviation to be a major issue in the coming years in the field of ultra-strong THz-induced structural dynamics, beyond the perturbative regime.

<table>
<tr><td colspan="2">Sub-system</td><td colspan="2">Phonon</td><td colspan="2">Electron</td><td colspan="2">Spin</td></tr>
<tr><td colspan="2">THz excitation</td><td>resonant</td><td>non-resonant</td><td>instantaneous</td><td>delayed</td><td>resonant (magnon)</td><td>non-resonant</td></tr>
<tr><td rowspan="2">POWER LAW</td><td></td><td>E (IR active phonons)</td><td>E^2 (ISRS), (Raman & IR active phonons)</td><td>E^2 (nonlinear optics Kerr), any material</td><td>E^2 (thermalization), mainly conductors and semiconductors</td><td rowspan="2">Same as resonant phonon excitation</td><td>$\mp H$ low field H^2 high field till T_c</td></tr>
<tr><td>Material damage occurs here</td><td colspan="2">$E^{2.5-3}$ (ISRS); coupling to other phonons, change phonon bandwidth/frequency</td><td colspan="2">Exponential response, easily seen in semiconductors</td><td>Coherent Magnetic switching (debatable)</td></tr>
<tr><td colspan="2">Optical probe</td><td colspan="6">Sensitivity could vary significantly depending on the probe photon energy, incidence angle, etc.</td></tr>
<tr><td rowspan="2">Signal / noise Rep. rate dep.</td><td>Transmission/ reflection</td><td colspan="2">Very weak (The reason why it is not clear in ref. [1])</td><td colspan="2">Tunneling is extremely strong (reported in ref. [1])</td><td></td><td></td></tr>
<tr><td>Birefringence/ polarization rotation.</td><td colspan="2">Very strong (excluded from [1])</td><td>strong</td><td>varies</td><td colspan="2">MOKE configuration + H direction.</td></tr>
</table>

Table: Crib sheet for THz-pump structural dynamics experiments: A rough and very generalized picture of how experimental nonlinear THz experiments could be designed and judged. (mainly for optical probe)

[1] Giorgianni, Vicario,..,and Hauri, arXiv:1706.00616v1 (2017).
[2] M. Liu, *et al*., *Nature* **487**, 345 (2012).
[3] A. X. Gray, *et al*. arXiv:1601.07490 (2016).
[4] E. Abreu, *et al.*, Phys. Rev. B 96, 094309 (2017).
[5] M. Shalaby & C.P. Hauri, *Nat. Commun*. **6**, 5976 (2015).
[6] M. Shalaby, C. Vicario, and C. P. Hauri, *APL Photonics* **2**, 036106 (2017).
[7] M. Forst, *et al.*, *Nature Physics* **7**, 854–856 (2011).
[8] M. Qazilbash, *et al.*, *Science* **318**, 1750 (2007).
[9] A Pashkin, *et al.*, *Phys. Rev. B* **83**, 195120 (2011).
[10] B. Mayer, *et al.*, *Phys. Rev. B* **91**, 235113 (2015).