A role of a picosecond strain in an ultrafast optically-driven phase transition in VO$_2$ nanostructures

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Abstract. We report on experimental picosecond acoustic studies of an ultrafast photo-induced insulator-to-metal and structural transition in VO$_2$ nanostructures epitaxially grown on Al$_2$O$_3$ substrates with different orientations. Applying a pump-probe technique with combined excitation of a sample with picosecond strain and femtosecond laser pulses we demonstrate that dynamical strain of moderate amplitude of 0.1% has a pronounced impact on ultrafast photo-induced phase transition in VO$_2$ nanohillocks. This enables novel path for controlling such transitions at picosecond and nanometer scales. Our experiments also allowed characterizing elastic and photo-elastic properties of the photo-induced metallic phase in VO$_2$ and to relate them to the properties of the equilibrium phase. Furthermore, we demonstrate the generation of picosecond strain pulses upon laser-induced excitation of thin epitaxial VO$_2$.

1. Introduction
Picosecond acoustics employs short strain pulses of high amplitude and various temporal and spatial profiles [1] to alter and control static and dynamic properties of nanostructures. Application of such pulses has been shown to enable generation of terahertz radiation [2] and electrical currents [3], control quantum-dot laser modes [4], excite magnetization dynamics [5], etc. The impact of picosecond strain pulses is considered as a promising purely-nonthermal alternative to the direct femtosecond laser excitation for driving ultrafast dynamics of electrons, lattice and spins. Recently, however, noticable interest emerged to dynamical strain generated upon various ultrafast laser-induced processes [6, 7], since this strain may provide valuable information about intricately coupled kinetics of lattice, electrons and spins. From this point of view, picosecond acoustic studies of ultrafast photo-induced phase transitions (PIPT) of various origins in strongly-correlated materials [9, 10, 11, 12] is of particular importance since it would provide novel information about kinetics of PIPT both far from thermal equilibrium and upon relaxation to the steady-state.

A problem of picosecond acoustic studies in the objects with ultrafast phase transitions can be separated into three main constituents, each capable of delivering novel information about the nonequilibrium and metastable photo-induced phases. First, what are the acoustic and
photo-elastic properties of a photo-induced phase, and how do they relate to the properties of a corresponding equilibrium phase? Secondly, how can picosecond strain pulse affect ultrafast PIPT, and can this be utilized for additional control of photo-induced phases? Third, what is the strain generated upon ultrafast PIPT, and what are the characteristics of such strain?

Here we report on results of experimental studies of the picosecond acoustics in VO\textsubscript{2} nanostructures. VO\textsubscript{2} is chosen as a model object possessing first order structural and insulator-to-metal transition above room temperature. This transition can be driven by femtosecond laser pulses on a subpicosecond time scale, [9, 13]. Ultrafast PIPT in VO\textsubscript{2} is also known to be sensitive to static strain [8]. In experiments we excite VO\textsubscript{2} by femtosecond laser pulses and picosecond strain pulses with variable timing between them. This technique allowed us to demonstrate impact of dynamical strain on PIPT, and characterize elastic and photo-elastic properties of the photo-induced metallic phase in this material. Further, we also considered reversed problem of picosecond strain pulse generation upon excitation of VO\textsubscript{2} by femtosecond laser pulse.

2. Experimental

The main sample was a layer of epitaxial VO\textsubscript{2} nanohillocks with a height of 70±20 nm and lateral size of 200±60 nm grown on a 350 \(\mu\)m-thick c-cut sapphire, Al\textsubscript{2}O\textsubscript{3} substrate by pulsed laser deposition [14, 15]. The nanohillocks are single crystalline with the [010]\(\text{M}_1\)-axis oriented perpendicular to the sample plane. The optical reflectivity \(R\) at a photon energy of 1.2 eV shows temperature dependence and a hysteresis typical for the insulator-to-metal transition in thin-film and nano-granular VO\textsubscript{2} with \(T_C = 340\) K and coercivity of 20 K [16]. A 140 nm thick Al film, serving as an opto-acoustic transducer [1], was sputtered on the back side of the sapphire. By means of conventional transient reflectivity measurements we [15] have get, in agreement with [13], that 170-fs laser pulses with the central photon energy of 1.2 eV induce the ultrafast PIPT in the VO\textsubscript{2} nanohillocks with the threshold \(W_T = 6\) mJ/cm\(^2\) and saturation \(W_S = 20\) mJ/cm\(^2\) fluencies. The increase of the laser fluence from \(W_T\) to \(W_S\) increases the fraction of VO\textsubscript{2} undergoing PIPT from 0 to 100%. Additionally, we have studied continuous epitaxial VO\textsubscript{2} films of 40 nm and 105 nm thicknesses grown on r- and c-cut Al\textsubscript{2}O\textsubscript{3} substrates. 30-nm Cr films were deposited on the back side of these samples.

We have developed a setup enabling measurements of the transient optical reflectivity following excitation by femtosecond laser and picosecond strain pulses (Fig. 1(a)). The laser source is a 170-fs Yb:KGd(WO\textsubscript{4})\textsubscript{2} regenerative amplifier with a central photon energy of 1.2 eV and a repetition rate of 5 kHz. The beam from the source is split into three beams. The optical pump beam is incident on the VO\textsubscript{2} nanohillocks. The second beam is used to generate the strain pulse and is incident along the normal onto an Al transducer. This beam is modulated with a mechanical chopper at a frequency of 625 Hz. The probe beam is incident to the VO\textsubscript{2} nanohillocks and after the reflection is steered onto one of the channels of the balanced photodetector (BD). To enable the balanced detection a part of the probe beam is used as a reference. A lock-in amplifier locked to the mechanical chopped frequency is used to measure the \textit{strain-related changes of the transient optical reflectivity}. The overall sensitivity of the setup reaches \(\Delta R/R \sim 10^{-6}\). The sample is placed on a copper electrical heater, which allows heating the sample up to 400 K. In experiments the transient reflectivity of the probe beam \(\Delta r\) is detected as a function of the time delay \(t\) counted from the moment when the front of the strain pulse enters VO\textsubscript{2}. The experiments are performed for the different timings \(t_0\) of the excitation of the VO\textsubscript{2} layer with optical pulses (Fig. 1(b)). This setup allows studying a variety of phenomena, e.g. optically-driven phase transition in the VO\textsubscript{2} layer, photo-elastic response of VO\textsubscript{2}, effect of the optical excitation on the photo-elastic properties of VO\textsubscript{2}, and the effect of the strain pulses on laser-induced transition. By turning the sample around, one can detect the strain pulses generated upon the laser-induced excitation of the VO\textsubscript{2} layer via photo-elastic effect in a metallic film.
3. Results and discussion

In Fig. 1(c) we compare the transient reflectivity $\Delta r$ measured in the equilibrium insulating and metallic phases when no optical pump is present. At insulating phase the transient reflectivity is governed by the photo-elastic response of VO$_2$. From the obtained curves we determine, in particular, the moments when the transient out-of-plane strain is maximal tensile ($t = 60$ ps, $\varepsilon_0 = 1.3 \times 10^{-3}$), or maximal compressive ($t = 95$ ps; $\varepsilon_0 = -2 \times 10^{-3}$). As one can see in Fig. 1(c), transition to the metallic phase results in the decrease of the measured signal, which is related to the change of the photo-elastic constants of VO$_2$. Next, we compare $\Delta r$ signals measured in the insulating and in the photo-induced metallic phases (Fig. 1(d), black and blue curves). The latter signal is obtained with the optical pump pulses of the fluence exceeding the saturation value ($W > W_S$) exciting the VO$_2$ layer several picosecond before the strain pulse enters it ($t_0 < 0$). As one can see, $\Delta r$ in the photo-induced metallic phase resembles that measured in the equilibrium metallic phase, i.e. only the decrease of the photo-elastic constants is clearly observed. If the VO$_2$ layer is excited by the laser pulse of the same fluence $W > W_S$ when the transient strain is already present, i.e. $t_0 > 0$ (Fig. 1(d), red curve), the quasi-instantaneous change of the photo-elastic response due to PIPT governs the measured signal.

The most important results were obtained in the similar series of experiments performed with the optical pump pulse of intermediate fluence ($W_T < W < W_S$), i.e. when only a fraction of VO$_2$ undergoes PIPT. As one can see in Fig. 1(e), $\Delta r$ under such conditions experiences additional changes (see the shaded area) when the VO$_2$ is excited by the optical pump pulse at $t_0 = 60$ ps corresponding to the maximal transient tensile out-of-plane strain. This is reliably interpreted as the strain-induced change of the fraction of VO$_2$ undergoing PIPT [15]. The sign of this strain-induced change of PIPT is dependent on the sign of the transient strain present...
at the moment of the photo-excitation. Furthermore, this addition to PIPT emerges at the moment of photo-excitation and slowly relaxes on a nanosecond time scale. Importantly, this slow relaxation appears to be insensitive to the dynamical strain. These features of the strain-induced contribution to PIPT are summarized in Fig. 1(f), showing the strain-induced changes of PIPT-related signal obtained when the optical pump pulse excites VO$_2$ at two different times $t_0$. The effect of the transient strain on the ultrafast PIPT is present only for the intermediate laser fluences $W_T < W < W_S$ (Fig.1(g)), and reaches $\sim$ 1% of the total reflectivity change upon PIPT induced by the optical pulses. Detailed analysis of the observed effect based on a phenomenological theory of the first order phase transitions and on the known effect of static strains on VO$_2$ [15], suggests that the ultrafast PIPT is controlled by the in-plane transient strain which changes the potential energy profile of the photo-excited state, and occur in the nanohillocks upon injection of the out-of-plane strain pulses.

Finally, we have performed experiments where VO$_2$ thin films are used as a photoelastic transducer, and the Cr film is acting as a detector of the strain pulses via photo-elastic effect. In these experiments we have detected the strain pulses of high amplitude and the observed features are related to the PIPT [17].

4. Conclusions
We have studied several aspects of picosecond acoustics of the ultrafast photo-induced phase transition in VO$_2$ nanohillocks. We have determined the photo-elastic response in equilibrium and photo-induced metallic phases of VO$_2$ and demonstrated their similarity. Further, we have shown that the impact of picosecond strain pulses with amplitude $\sim 10^{-3}$ decreases or increases by $\sim$ 1%, depending on the sign of strain, the fraction of VO$_2$ nanohillocks which undergo ultrafast PIPT from insulating to metallic phase. This impact occurs only at a sub-picosecond time range following optical pulse excitation, while the relaxation of the excess or deficient fraction of VO$_2$ in the metallic phase to the quasi-equilibrium goes on a nanosecond time scale and appears to be unaffected by the strain pulses.

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