Generation of terahertz radiation in thin vanadium dioxide films undergoing metal-insulator phase transition

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***Abstract*— Generation of terahertz (THz) radiation was observed in epitaxial VO_2 films grown on R- and C-cut sapphire substrates above and below the metal-insulator phase transition temperature. Polarization analysis of the emitted THz radiation reveals strong in-plane anisotropy of the conductive phase of VO_2 which is not observed for insulating phase, generation efficiency increases up to 30 times after phase transition. Properties of generated THz radiation in VO_2 are defined by the displacement photocurrent at the film-air and film-substrate interfaces.**

I. INTRODUCTION

VANADIUM dioxide (VO_2) is a material that exhibits 1st order phase transition from insulating to conductive state at temperature near 68°C. It leads to up to 4 orders of magnitude increase in conductivity [1] and significantly changes it's optical and THz properties. For THz radiation the VO_2 film is transparent in insulating state, but becomes almost reflective in conductive state.

Since both conductive and insulating phases of VO_2 belong to centrosymmetric class of substances, second-order processes are not expected in the bulk material. Nevertheless, we find that thin (up to 300 nm) VO_2 films can emit broadband THz radiation as a result of interaction with femtosecond optical pulses from regenerative amplifier and THz emission efficiency shows step-like increase at phase transition temperature.

The aim of this work is to investigate the THz generation in thin VO_2 films in different phase states. We focus our attention on the polarization of the THz radiation since it is could give information on the generation mechanism and anisotropy of the free carrier dynamics in VO_2 films.

II. SAMPLES

We used a series of thin (30-200 nm) epitaxial VO_2 films grown by metalorganic chemical vapour deposition (MOCVD) or pulsed laser deposition (PLD) on R-cut and C-cut sapphire substrates. Both substrate types are transparent for terahertz and optical radiation. The best phase transition parameters of the films can be achieved on the R-cut sapphire substrates due to the best match of crystalline lattice dimensions of the VO_2 and substrate [2] which leads to epitaxial mode of growth for the VO_2 films. But these substrates are birefringent for THz and optical radiation and thereby complicates the analysis of emitted THz signal by time-domain techniques.

III. EXPERIMENTAL SETUP

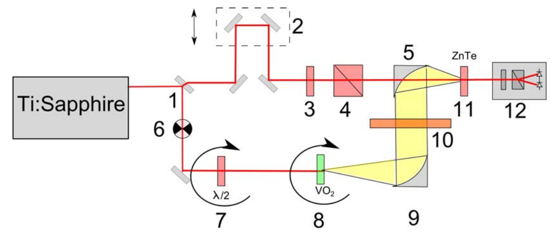

Fig. 1. The scheme of the experimental setup: 1 – beam splitter, 2 – delay stage, 3 – attenuator, 4- Glan prism, 5,9 – off-axis parabolic mirrors, 6 – optical chopper, 7 – half-wave plate, 8 – sample, 10 –THz polarizer, 11 – ZnTe crystal, 12 – balance detector

To investigate THz radiation from VO_2 films we implemented a THz-TDS spectrometer, based on 50-fs Ti:Sapphire regenerative amplifier with average power up to 3 W. In experiment we placed VO_2 film into the collimated beam from the amplifier and detected THz radiation using EO-crystal (ZnTe, 4mm) and balance detector.

To analyze THz polarization, we placed wire-grid polarizer into the collimated THz beam. Because our detector was polarization-sensitive, we decided to fix the angle of the polarizer and to rotate synchronously the sample and the polarization of optical pump radiation, so we could obtain the projections of THz radiation to the polarizer's direction.

For each studied sample, we carried out measurements at room temperature (20±2 °C) and above the phase transition temperature (80±2 ˚C).

IV. RESULTS

The irradiation of VO_2 films with the femtosecond pulses from Ti:Sapphire regenerative amplifier having energies up to 0.5 mJ lead to generation of THz pulses that have broadband

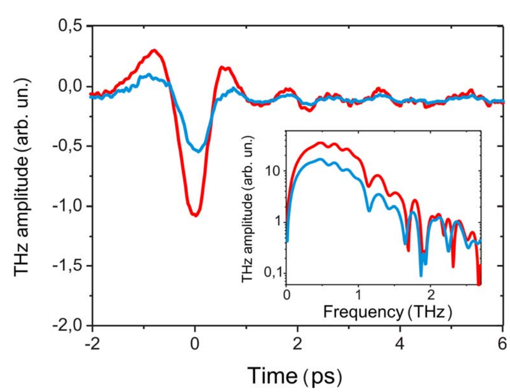

Fig. 2. Typical waveforms of THz radiation emitted by sample in conductive (red) and insulating (blue) phases.

spectrum and show no visible resonance features in the frequency range available for our detector (0.1-2.5 THz). After the phase transition, detected THz amplitude increases up to 30 times, but emitted spectrum remains identical [see Fig. 2].

Amplitude of emitted THz radiation was about 1% of that observed from the LT-GaAs surface in the same optical beam.

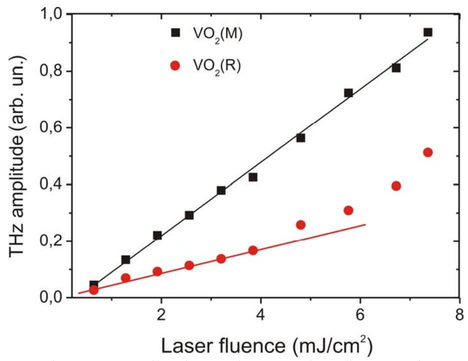

Fig. 3. Detected THz amplitude vs laser pulse fluence for semiconductor (red) and conductive (black) states.

THz amplitude shows linear dependence on the pump fluence above the phase transition temperature [see Fig. 3]. Below the phase transition temperature, difference from linear dependence can be explained as the light-induced phase transition which is expected to be at about 6.5 mJ/cm^2 [3]. Polarization of THz beam was linear in both states.

We observed different behavior of the polarization of THz radiation from VO_2 films in insulating and metallic states. In the former case, the THz intensity and polarization were dependent on the angle between the pump pulse polarization and the film orientation. In contrast, for rutile phase of the VO_2 film dependence of THz intensity and polarization on the pump beam polarization was very weak, and emitted THz pulses were polarized along the [100]VO_2(R) axis with almost constant intensity as the pump beam polarization was changed over 100°[see Fig. 4].

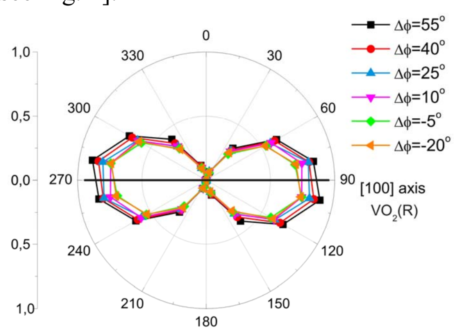

Fig. 4. Polarization of the THz radiation generated in the VO_2 film in conductive state for different angles between pump polarization and film orientation.

V. Discussuon

Conductive and insulating states of VO_2 belong to centrosymmetric space groups, so second-order nonlinear susceptibility tensor should vanish for the bulk material. But 2-nd order processes are not forbidden at interfaces film-air and film-substrate. It can be explained as excitation of displacement current at the surface of the conductive layer which accounts for emission of electromagnetic radiation at combinative frequencies - in our case, "zero" frequency which corresponds THz emission. This mechanism requires nonzero electric field component of incident optical radiation along the surface normal. Such component emerges for nonzero incidence angles or, as in our case, for limited dimensions of the optical beam.

R-cut sapphire substrate surface has symmetry m, which corresponds to the rectangular class, and its two orthogonal in-plane directions are not equivalent. That's why the nonlinear optical mixing process which occurs at VO_2-sapphire boundary has a preferred polarization axis linked to the substrate (and film) orientation, exactly as it happens in the experiment.

For the low-temperature phase of VO_2 the nonlinear response is influenced by optically-induced conductivity. Fundamental radiation leads to generation of the Frenkel excitons which decays rapidly into Wannier-Mott excitons. Conductivity appears due to tunneling of electrons from exciton states into an unoccupied excited states in the conductive phase [4]. It can explain different behavior of THz polarization in low-temperature phase state. In the three-wave mixing formalism, we can introduce from the experimental data for the low-temperature phase the effective tensor of nonlinear susceptibility $\chi^{(2)}_{eff}(\Omega,\omega,-\omega)$, which corresponds to the tetragonal symmetry group.

VI. Summary

In this work we studied the THz field emission in VO_2 films undergoing insulator to metal phase transition, emission occurs after irradiation with femtosecond laser pulses with high intensity. Amplitude of this emission was two orders of magnitude less than in LT-GaAs. Polarization studies of THz radiation revealed linear polarization for both monoclinic and rutile phase of VO_2 grown on R-cut sapphire substrates. In both the insulating and conductive phase states, the THz radiation is governed by the contribution of the surface displacement current induced by incident optical beam on VO_2-Al_2O_3 and VO_2-air boundaries. In the high-temperature phase, THz polarization is defined by anisotropic photocurrent influenced by intrinsic electrostatic field which is directed along the preferred [100] direction.

References

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