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# Interaction of electromagnetic waves with VO<sub>2</sub> nanoparticles and films in optical and millimetre wave ranges: Prospective for nano-photonics, nano-antennas, and sensors

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Abstract. Recently great interest in the field of nano-optic is attracted to nano-antennas in both basic and applied research. Nano-antennas are represented by metallic nanoparticles and holes in thin films with arrays of the holes. The usual nano-antennas have fixed functionality, but it will be very interesting to get tunability of nano-antennas controlled by external field. Materials with phase transitions are candidates for the development of tuneable nanoantennas. In this report metal-dielectric phase transition in VO2 micro- and submicron particles, films and holes are studied experimentally and theoretically. The complex dielectric permittivity of VO<sub>2</sub> is varied drastically with temperature due to structural phase transition, accompanying metal-insulator transition (MIT) at critical temperature (Tc=340 K). In this work, VO<sub>2</sub> film on glass substrate were prepared and investigated in MM wave range (27-37 GHz). Then submicron holes arrays were formed on VO<sub>2</sub> films by FIB milling and their optical Raman spectra were studied. The MM wave response of nanosized VO2 films reveals strong anomalies near MIT. The holes and arrays show strong change of the Raman spectra at the wavelength 530 nm under heating by laser radiation. The optical and MM wave electromagnetic properties of homogeneous VO<sub>2</sub> nanospheres embedded in the air are studied theoretically. Size effects on the optical properties of the VO<sub>2</sub> nanosphere are investigated. In VO<sub>2</sub> nanosphere, converting into metallic phase by heating leads to formation of a localized surface plasmon resonance (LSPR) which red shifts by increasing dimension.

#### 1. Introduction

In the last decades a real breakthrough has been made in the field of nano-photonics, nano-plasmonics, NAs, and metasurfaces (optical nanomaterials). One of the main problems facing the wide application of these devices is that the properties of such devices are fixed at time, and cannot be tuned. Controllable, or as they are called in these areas "active" (not to be confused with energy-generating) devices would not only be much more in demand, but could create many new scientific and technical opportunities. Recently developed devices like nano-optic, nano-plasmonic, nano-antennas, optical metamaterials/metasurfaces for many applications need the operations either at different frequency bands or different radiation characteristics (such as pattern or polarization) it will be optimal that the devices of nano-photonic, will have multiple characteristic. Thus one of the challenges in this area is to find mechanisms that can provide the control of the properties of functional materials and the devices based on these phenomena. There are many types of reconfigurable antenna that according to its function are divided into frequency reconfigurable antenna, polarization reconfigurable antenna, pattern reconfigurable antenna and hybrid reconfigurable antennas [1, 2]. The different tuning mechanisms have been studied in the literature can be classified into three general categories: (1) Physical tuning

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(shift, deformation, microfluidic); (2) material tuning (liquid crystal, ferromagnetic/ferroelectric, semiconductors, phase change materials); (3) circuit tuning (open/short, semiconductor devices, MEMS, Non Foster, varactor). One of the most promising mechanisms of tuning is using of phase transition materials, such as VO<sub>2</sub>. In this report the phenomena that gives the possibility to create nano-photonic devices, such as nano-antennas in wide frequency range from optics to millimeter range in VO<sub>2</sub> thin films and NAs are studied.

#### 2. VO<sub>2</sub>- phase change material

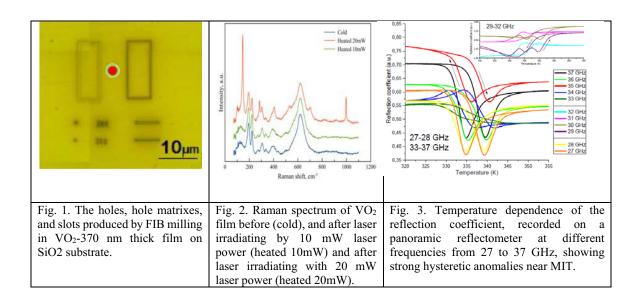
One of the most promising phase change material is  $VO_2$  [3-5]. Single crystals of  $VO_2$  have high transparency and low absorption due to a low concentration of free carriers ( $\sim 10^{18}$  cm<sup>-3</sup> at f < 6.7 THz). At a temperature of  $T_c = 340$  K [3,5]  $VO_2$  undergoes metal-isolator phase transition (MIT) at which a monoclinic crystallographic structure is transformed into a tetragonal crystalline structure [3-5]. Under the illumination with a laser pulse with a dose of 0.15 mJ/cm<sup>2</sup>, switching occurs for 8 ps [8]. Under the dose of 2.0 mJ/cm<sup>2</sup> for pumping, the switching time lies in the range of several hundred femtoseconds [4-5]. The transition is accompanied by a sharp change in the optical properties, IR, THz and MM wavelenths [6]. The goal of this report is study of  $VO_2$  films properties in the MM wave range (8 mm) ranges, to make prototypes of nanoantennas (NAs), micro-holes, and slots (see Fig.1), as well as to study their interaction with electromagnetic wave in the visible range using Raman scattering methods, and to study theoretically the optical characteristics of  $VO_2$  nanospheres.

#### 2.1. Raman spectra of NAs based on VO<sub>2</sub>

The spectra obtained from the structure of two adjacent holes in the VO2 film and the surface of the film after subtraction of the background and normalization show the follows: the relative intensity of the 620 cm-1 peak obtained in the region of the bridge between the holes is much smaller than that of the analogous peak in the spectrum, obtained on the film. See Figs.1 and 2. There is also a shift to smaller wave numbers at the peak obtained at 614 and 622 cm<sup>-1</sup>. On the spectrum obtained after irradiation of the VO<sub>2</sub> film usig laser of 20 mW, there are additional peaks 146, 284, 531, 703 and 997 cm<sup>-1</sup>, compared with the unirradiated one. The origin of these peaks may be due to photoinduced phase instability of the material [7].

#### 2.2. MM wave range properties of the $VO_2$ films

Fig. 3 shows the reflection coefficient of VO<sub>2</sub> in the range 27-37 GHz at different temperatures in the vicinity of MIT in thin VO<sub>2</sub> film: below MIT at 331 K, near MIT at 340 K and above MIT at 345 K. Also with this method the hysteretic behavior of the reflection coefficient of the VO<sub>2</sub> film is shown for direct and inverse MIT, while the shape of the curve depends strongly on the frequency (Fig. 11). To explain the frequency dependence of the reflection coefficient, a theoretical model has been proposed within the framework of the Drude theory [9].

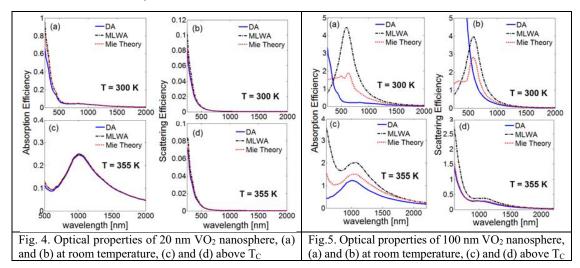


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### 2.3. Properties of VO2 nanosphere in optical spectrum

The optical properties of  $VO_2$  nanosphere and also the temperature, size and surrounding medium effects on these properties are explored theoretically by exploiting dipole approximation (DA) [10], modified long wavelength approximation (MLWA) [11] and Mie theory [10]. For this purpose, the optical constant of  $VO_2$  are extracted from [12] in two different temperature: i) 300  $^{\rm o}K$ , in which the  $VO_2$  nanosphere acts as an insulator nanosphere, ii)355  $^{\rm o}K$ , where the  $VO_2$  nanosphere manifests metallic properties. These two temperatures are far from the critical temperature ( $T_C$ ) in which insulator-metal transition occurs in  $VO_2$ . In the temperatures near the  $T_C$  the insulating and metallic domains coexist, therefore the effective medium theory elucidates the optical constants of  $VO_2$  [13]. In the metallic phase of  $VO_2$ , the nanosphere experiences the LSPR. So there is an enhancement in optical properties of  $VO_2$  nanosphere which is disappeared in insulator case. See Fig. 5 and 6. By increasing the radius of nanosphere, the LSPR wavelength slightly redshifts. There is a peak in scattering and absorption spectra of  $VO_2$  nanosphere in insulator state, in visible range corresponding combined modes. According to the obtained results, it is obvious that by increasing the radius of nanosphere, the DA method lose its accuracy.



The optical constant of VO<sub>2</sub> is shown in Fig.6. According to this data, the real part of VO<sub>2</sub> optical constant is near zero at LSPR wavelengths of VO<sub>2</sub> nanosphere with different radiuses. This is in contrary to LSPR condition (Fröhlich condition) [14]. It seems that the imaginary part of VO<sub>2</sub> has tremendous effect on the LSPR wavelength. This effect is examined in Fig.6. The extinction spectrum of 20 nm nanosphere approaches to Fröhlich condition as the value of imaginary part for VO<sub>2</sub> is lowered by 0.5, 0.25 and 0.1 factors. It can be concluded that the principle of resonance around 1000 nm is the LSP which is influenced severely by the imaginary part of the dielectric constant.

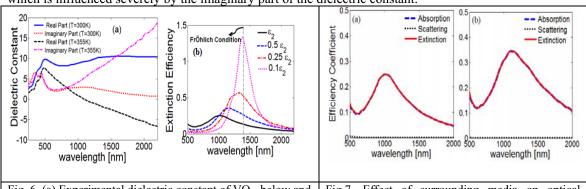


Fig. 6. (a) Experimental dielectric constant of VO<sub>2</sub>, below and above the T<sub>C</sub>, extracted from [12], (b) Effects of imaginary part of VO2 dielectric constant on spot of LSPR wavelength.

Fig.7. Effect of surrounding media on optical properties of  $VO_2$  nanosphere in metallic phase, (a) in air, (b) in a medium with  $\mathcal{E} = 2.25$ .

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The effects of surrounding media (  $\varepsilon = 2.25$  ) on the LSPR wavelength of 20 nm VO<sub>2</sub> nanosphere are investigated in Fig.7. The LSPR wavelength red shifts by increasing the permittivity of surrounding medium as expected from theory [10].

#### 3. Conclusion

The results of the work are as follows:

- 1)  $VO_2$  film on glass substrate were prepared and investigated in MM wave range (27–37 GHz). The MM wave response of nanosized  $VO_2$  films reveals strong anomalies near MIT.
- 2) The submicron holes arrays were formed on  $VO_2$  films by FIB milling and their optical Raman spectra were studied. The holes and arrays show strong change of the Raman spectra at the wavelength 530 nm caused by laser radiation heating.
- 3) The optical and MM wave electromagnetic properties of homogeneous VO2 nanospheres embedded in the air are studied theoretically. Size effects on the optical properties of the VO2 nanosphere are investigated. In VO<sub>2</sub> nanosphere, converting into metallic phase by heating leads to formation of a localized surface plasmon resonance (LSPR) which red shifts by increasing dimension.

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