JOURNAL OF NANO- AND ELECTRONIC PHYSICS
Vol. 16 No 5, 05001(4pp) (2024)

ЖУРНАЛ НАНО- ТА ЕЛЕКТРОННОЇ ФІЗИКИ
Том 16 № 5, 05001(4cc) (2024)

REGULAR ARTICLE

Surface Plasmon's Dispersion Properties of Vanadium Oxide Films

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(Received 18 June 2024; revised manuscript received 22 October 2024; published online 30 October 2024)

Vanadium dioxide (VO_2) is a typical metal-insulator transition material, which changes from room-temperature monoclinic insulating phase to high-temperature rutile metallic phase. The phase transition of VO_2 is accompanied by sudden changes in conductance and optical transmittance. Due to the excellent phase transition characteristics of VO_2, it has been widely studied in the applications of electric and optical devices, smart windows, sensors, actuators, etc. The dispersion properties of thin vanadium dioxide films were researched using the modulation polarimetry (MP) technique. VO_2 thin films were deposited on quartz glass substrates by magnetron sputtering of the VO_2 target. The films had different modifications of composition, structure, morphology, and optical properties due to the manufacturing technology. The main measured parameter of the MP technique is the polarization difference between the internal reflection coefficients of linearly polarized light with an electric field azimuth perpendicular and parallel to the plane of incidence. The spectral dependences of the polarization reflection difference were measured at different angles of light incidence for several samples. Spectral dependences of the polarization absorption difference were shown. Also, the dispersion characteristics of surface plasmonic polaritons appearing at the film-air interface were shown. The value of the group velocity of excitation of surface plasmons for all films was determined from the slope of the frequency depending on the wave vector. The proposed method makes it possible to conclude the film morphology. The crystal structure of the films was investigated using an X-ray diffractometer. The surface nanorelief of annealed vanadium dioxide films were studied using atomic force microscopy.

Keywords: Modulation polarimetry technique, Polarization, VO_2 thin films, Dispersion properties, Gaussian functions.

DOI: 10.21272/jnep.16(5).05001 PACS numbers: 42.25.Ja, 42.68.Ay

1. INTRODUCTION

The fascinating thermochromic material vanadium dioxide is currently attracting much attention for both scientific and technological applications. VO_2 exhibits a reversible, solid-solid semiconductor-to-metallic phase transition at a relatively low temperature ($T_{trans} \approx 68$ °C) [1-4]. The phase transition can be initiated by several external stimuli, including temperature, pressure, electric or magnetic fields, photoexcitation, and carrier injection. A colossal drop in resistivity by more than four orders of magnitude and other sharp changes in properties occur in this case. Therefore, VO_2 has attracted a lot of attention for its use in highly sensitive smart devices that can react sharply to various external stimuli. All of these characteristics make VO_2 an ideal candidate for use in bolometers, coatings in various fields, optical and electrical switches, RF microwave switches, tuneable plasmonic and metamaterial systems, and smart windows [5-11]. However, the mechanism of the phase transition is still controversial over the past few decades.

In this paper, we demonstrate the further development of the information capabilities of the modulation polarimetry technique (MP technique) by studying the synthesized thin VO_2 films of a thickness suitable for demonstrating the plasmon-polariton resonance [12-13]. The work aims to determine the plasmon properties of films and obtain the dispersion characteristics $\omega(k)$ of plasmon interactions (ω is the frequency, k is the wave vector). The main measured parameter of the MP technique is the polarization difference between the internal reflection coefficients of linearly polarized light with an electric field azimuth perpendicular (R_s^2) and parallel (R_p^2) to the plane of incidence $\rho = R_s^2 - R_p^2$. The spectra of the polarization difference $\rho(\omega)$ were measured at various angles of light incidence. The dispersion characteristics $\omega(k)$ were created from the results of the decomposition of the experimental spectra into elementary functions, each of which corresponds to a certain resonant interaction of radiation with the film.

Successful expansions of functions similar to $\rho(\omega)$ into spectrum components by Gaussian functions are shown in [14-18]. The modification of the parameters of the VO_2 films was achieved by changing the substrate temperature during their deposition.

2. EXPERIMENT AND SAMPLE PREPARATION

2.1 Physical Principle of the Method

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Cite this article as: I. Matyash et al., *J. Nano- Electron. Phys.* 16 No 5, 05001 (2024) https://doi.org/10.21272/jnep.16(5).05001

The phenomenon of internal reflection is accompanied by a system of polarization-dependent effects. In general, the reason for this fact is the inequality according to Fresnel's formulas of the reflection coefficients of polarized radiation. This inequality or polarization difference is essentially the amplitude anisotropy of internal reflection. The polarization difference between the internal reflection coefficients of linearly polarized light with wave field azimuths perpendicular R_s^2 and parallel R_p^2 to the plane of incidence is $\rho(\lambda) = R_s^2 - R_p^2$.

Spectral polarization characteristics of VO_2 thin films were measured in Kretschmann geometry using the MP technique. in the wavelength range $\lambda = 0.45 - 1$ µm at light incidence angles $\theta = 45°$, 50°, and 60°. The setup scheme is described in detail in [19].

2.2 Sample Preparation

For VO_x thin film deposition the two-step method developed earlier was used [20-23]. VO_2 films were grown on quartz glass substrates by magnetron sputtering of the VO_2 target. The residual gas pressure was ~ $2 \cdot 10^{-6}$ Torr. During the sputtering process, the Ar/O_2 mixture pressure was kept at $3 \cdot 10^{-3}$ Torr. The power on magnetron was ~ 10…12 W. The substrate temperatures for all samples were different, °C: for 1 – 210, 2 – 240, 3 - 150, and 4 – 180. Obtained layers' thickness was 180 ± 10 nm. After deposition, the samples were annealed at 400 °C for 15 min in Ar ambient was performed. Such technological parameters of growing allow obtaining films with reduced to ~ 50 ºC phase transition temperature.

For film structure modification the Ar^{2+} ion implantation was used. The energy was 180 keV and doses 0.2…5 × 10^{14} cm^{-2}. The choice of energy was made so that Ar ions create a uniform distribution of defects throughout the whole film thickness. In this paper, we present the results for samples implanted with dose 1.5 × 10^{14} cm^{-2} where the best results were obtained. At higher doses, there is a significant deterioration in the crystallinity of the film and the metal-insulator-transition parameters.

3. METHODS

The crystal structure of the films was investigated using an X'Pert Pro MPD X-ray diffractometer. The surface nano relief of annealed vanadium dioxide films were studied using atomic force microscopy (AFM) with a scanning probe microscope NanoScope IIIa Dimension 3000TM. Measurements were performed in the tapping mode by using the ultrasharp silicon probes with a nominal tip radius of 8 nm.

4. RESULTS AND DISCUSSION

The spectral dependences of the polarization reflection difference are shown in Fig. 1. They were measured at an angle of incidence of 50° for all VO_2 films used in the paper. Films deposited on substrates with a temperature of 180 °C (sample 4) and 210 °C (sample 1) have similar reflection characteristics, as can be seen from Fig. 1. At the same time, an increase or decrease in temperature leads to a short-wave shift of the extrema. These shifts can be caused by changes in the structure or composition of the film. Additional research has shown that the second option is correct. Samples 2 and 3 contained inclusions of other types of vanadium oxides. In this case, all dependencies demonstrate the presence of several mechanisms of resonant interaction of radiation with the film: one in the range of 400-500 nm, the second in the range of 600-800 nm, and the third at a wavelength greater than 1000 nm (especially noticeable in the dependence of sample 3).

The spectra of the polarization reflection difference $\rho(\lambda)$ at different angles of light incidence were obtained to accurately determine the nature of resonant interactions of radiation with films. The result is shown in Fig. 2 (sample 4) and is typical for all the studied films. We observe in the figure a short-wavelength shift of extrema ($\lambda \approx 750$ nm and $\lambda > 1000$ nm) with an increase in the angle of incidence of light and large amplitudes of these extrema compared to high-energy ones ($\lambda \approx 490$ nm). This circumstance makes it possible to unambiguously classify these films as continuous structures with insignificant roughness. Thus, the high-frequency extremum is due to the resonant excitation of local surface plasmons at the inhomogeneities of the film surface. The localization of these extrema practically does not change depending on the energy for different angles of light incidence. This indicates the correct hemispherical shape of the inhomogeneities or their chaotic arrangement. The AFM study confirmed the second assumption. Two low-frequency extrema are caused by two modes of surface plasmon-polaritons at the interfaces: film-air ($\lambda \approx 750$ nm) and glass-film ($\lambda > 1000$ nm).

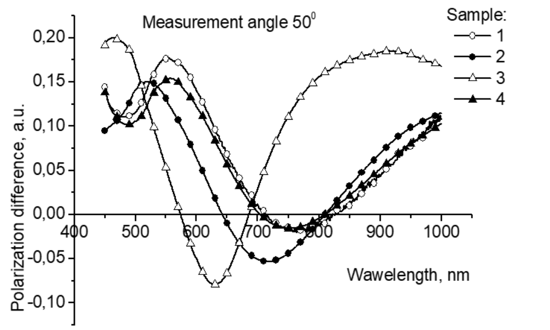

Fig. 1 – Spectral dependencies of the polarization difference $\rho(\lambda)$ at $\theta = 50°$ for samples 1-4

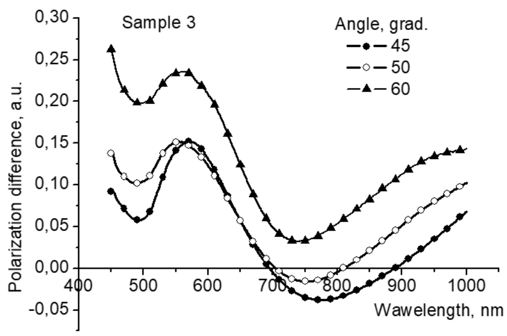

Fig. 2 – Spectral dependencies of the polarization difference $\rho(\lambda)$ for sample 4 at $\theta = 45°$, 50°, 60°

The paper also analyzed the spectral dependences of the polarization absorption difference $\Delta\alpha$. The thickness of the films used (d = 170 nm) allows us to neglect the transmission $\Delta\alpha = 1 - \rho$. For analysis, the curves were plotted depending on the frequency ($\omega = 2\pi c/\lambda$). Fig. 3 shows the spectrum obtained at a light incidence angle of 60° for film 4 and its approximation by elementary Gaussian functions. The almost complete agreement between the experimental and calculated curves became possible using at least three Gaussians. These Gaussians correspond to a certain resonant interaction of the radiation with the film.

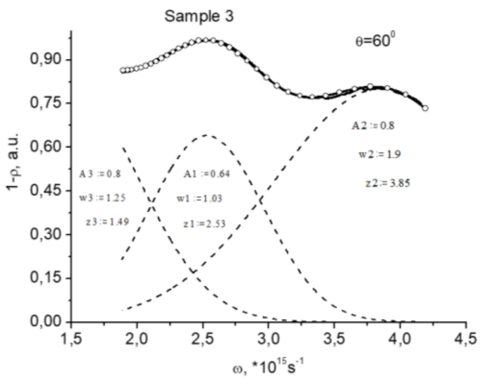

Fig. 3 – Spectral dependences of the polarization absorption difference $\Delta\alpha$ for sample 4 at θ = 60°

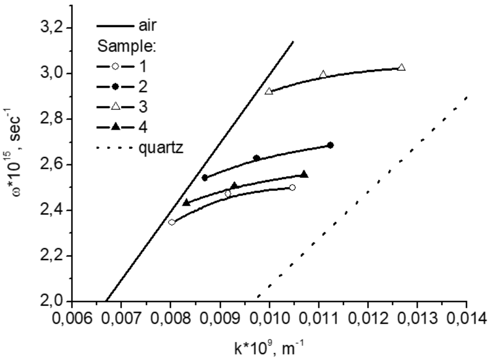

Fig. 4 – Dispersion characteristics of surface plasmon polaritons that appear at the film-air interface

A similar analysis was performed for all films (1-4) and different angles of light incidence. The results obtained were used to determine the dispersion dependences of the resonant modes of surface plasmons for each film. The position of the maximum of the Gaussian on the energy scale gives the frequency data. The wave vector of a plasmon can be determined if the angle of incidence of light on the film is known. The resonant interaction of light with a film is possible when the projection of the radiation wave vector onto the film plane is equal to the plasmon wave vector k. In turn, $k = \frac{2\pi sin\theta}{\lambda/n}$, where n is the refractive index of the glass.

Fig. 4 shows the dispersion characteristics of surface plasmon polaritons that appear at the film-air interface. The slope of the dependences indicates a positive value of the group velocity of surface plasmon excitations for all films. This conclusion unambiguously confirms their continuous morphology.

5. SUMMARY

In this paper, some optical properties of VO_2 were introduced. Almost all the application prospects are based on the phase transition characteristic of VO_2. The electrical changes caused by metal-insulator transition make VO_2 promising in applications such as electronic switches, field-effect transistor and memory devices. Due to the change of optical properties caused by metal-insulator transition and the electrical properties of VO_2 being sensitive to light, VO_2 can be used to develop smart windows, photodetectors, etc. The metal-insulator transition of VO_2 is sensitive to strain, temperature and surrounding gas environment, which makes VO_2 as a candidate material in multiple types of response sensors. The strain produced by the metal-insulator transition of VO_2 makes it useful for micro or nanoactuators. Moreover, VO_2 has great potential for developing infrared camouflage and thermal regulation.

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Дисперсійні властивості поверхневого плазмону плівок оксиду ванадію

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Діоксид ванадію (VO_2) є типовим перехідним матеріалом метал-діелектрик, який змінює моноклінну фазу діелектрика при кімнатній температурі на металеву рутилову фазу при високій температурі. Фазовий перехід VO_2 супроводжується різкими змінами провідності та оптичного пропускання. Завдяки чудовим характеристикам фазового переходу VO_2 його широко вивчають в застосуванні електричних і оптичних пристроїв, розумних вікон, датчиків, приводів тощо.

Методом модуляційної поляриметрії (МП) досліджено дисперсійні властивості тонких плівок діоксиду ванадію. Тонкі плівки VO_2 були нанесені на підкладки з кварцового скла шляхом магнетронного розпилення мішені VO_2. Плівки мали різні модифікації складу, структури, морфології та оптичних властивостей, зумовлені технологією виготовлення. Основним вимірюваним параметром техніки МП є різниця поляризацій між коефіцієнтами внутрішнього відбиття лінійно поляризованого світла з азимутом електричного поля, перпендикулярним і паралельним площині падіння. Виміряно спектральні залежності різниці коефіцієнтів внутрішнього відбиття при різних кутах падіння світла для кількох зразків. Показано спектральні залежності поляризаційної різниці коефіцієнтів поглинання. Також показано дисперсійні характеристики поверхневих плазмонних поляритонів, що виникають на межі розділу плівка-повітря. Значення групової швидкості збудження поверхневих плазмонів для всіх плівок визначали за нахилом частоти в залежності від хвильового вектора. Запропонований метод дозволяє зробити висновок про морфологію плівки. Кристалічна структура плівок досліджувалась на рентгенівському дифрактометрі. Методом атомно-силової мікроскопії досліджено нанорельєф поверхні відпалених плівок діоксиду ванадію.

Ключові слова: Метод модуляційної поляриметрії, Поляризація, Тонкі плівки VO_2, Дисперсійні властивості, Функції Гауса.