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Ellipsometric study of the Cu/V/mica system

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ABSTRACT. We have grown Cu (V) thin films on V (Cu) films deposited in high vacuum on crystalline mica substrates (muscovite). The optical properties of this system have been determined using spectroscopic ellipsometry. The dielectric functions are analyzed theoretically using the Drude, the classical oscillator and Lorentz models. In the case of a 3.5 nm vanadium film on copper, the optical constants of the composite system were as expected from the optical constants the individual components at the apropriate ratio. However, for 3.5 nm of copper on vanadium, a strong screening effect of the copper electrons on the vanadium optical response is observed.

RESUMEN. Se han depositado películas delgadas de Cu (V) sobre películas de V(Cu) en muy alto vacío sobre sustratos de mica cristalina (moscovita). Las propiedades ópticas de este sistema se determinaron utilizando elipsometría espectroscópica. Las funciones dieléctricas se analizaron teóricamente utilizando los modelos de Drude, osciladores clásicos y Lorentz. En el caso de una película de 3.5 nm de vanadio sobre cobre, las constantes ópticas del sistema compuesto resultaron como se esperaban de acuerdo a las constantes ópticas de cada una de las las componentes del sistema en la proporción apropiada. Sin embargo, para el caso de 3.5 nm de cobre sobre vanadio, se observó un fuerte efecto de apantallamiento de los electrones del cobre sobre la respuesta óptica del vanadio.

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1. INTRODUCTION

The optical properties of metallic superlattices have been studied extensively in the past few years. In particular, artificial superlattices are useful samples to investigate electromagnetic anomalies at interfaces and properties of two-dimensional systems. Recently, monolayers and multilayers of V on noble metal (Ag and Au, for example) surfaces have been investigated by several groups [1–7]. Magnetic behavior has been predicted theoretically and observed experimentally.

In an effort to understand the effect that the presence of a certain material can have in the properties of the other, as the coverage of one on to the other goes from 0 to 1, in this work we study the optical properties of a composite system of copper and vanadium. The optical responses of a Cu/V/mica and a V/Cu/mica systems are measured using spectroscopic ellipsometry. Mica substrates were selected as a mean to induce a preferential direction growth which may be useful in the superlattice application.

X-ray diffraction studies of the films showed a random oriented polycristalline growth with a slight preferential orientation for the copper in the (111) direction and in the (110) direction the vanadium film. Even though mica is an anisotropic material [8], the copper and vanadium films in contact with the substrate were thick enough to be opaque to the incident light probe of the ellipsometer therefore no information about the substrate was recorded in the analyzed beam. Witness samples on glass were produced in every deposit on mica and the optical characterization showed a similar behavior. The theoretical determination of the dielectric functions is achieved by fitting the experimental curves using Drude, classical oscillator and Lorentz models [9]. Because of the intrinsic tendency of Cu and V to react with the ambient oxygen as soon as they are exposed to the atmosphere, oxide layers for Cu and V must be considered. To minimize this effect, high deposition rates of the order of $r > 2$ nms^{-1} are used.

2. EXPERIMENTAL

2.1. SAMPLE PREPARATION

The samples were prepared by argon ion RF magnetron sputtering of vanadium (99.7% purity) and copper (99.9% purity) commercial targets on mica (muscovite) substrates cleaved in air. The base pressure was 3×10^{-6} Torr and the argon work pressure was 3 mTorr. The mica substrates were heat treated firstly in vacuum at 200°C to eliminate water. At room temperature, a 100 nm Cu (V) was deposited on the mica substrate, and then, without breaking vacuum, a 3.5 nm V (Cu) overlayer. The thicknesses were monitored with a quartz microbalance. The optical characterization of the samples was performed *ex situ* with a phase modulated spectroellipsometer (UVISEL by Jobin-Yvon) in the range of 1.5 to 5.5 eV photon energy (225 to 825 nm) at 70° angle of incidence. The experimental data thus obtained were used to obtain the optical properties of the system by curve fitting, based on a Drude plus a classical oscillator and Lorentz models, starting with the properties of the individual components. A set of twenty samples was fabricated to test repeatability and for statistical purposes to visualize the tendencies of the optical properties.

2.2. DETERMINATION OF THE OPTICAL PROPERTIES: CURVE FITTING

The dielectric functions ϵ of the CuO/Cu/mica, V/mica, Cu/V/mica and V/Cu/mica systems were modeled by the general expression

$$\epsilon = \epsilon_\infty + \frac{(\epsilon_s - \epsilon_\infty)\,\omega_t^2}{(\omega_t^2 - \omega^2) - i\Gamma_0\,\omega} - \frac{\epsilon_s\,\omega_p^2}{\omega^2 + i\Gamma_D\,\omega} + \sum_j \frac{f_j\,\omega_{0j}^2}{(\omega_{0j}^2 - \omega^2) - i\Gamma_j\,\omega}, \tag{1}$$

where ω is the frequency of the incident field, ϵ_∞ and ϵ_s represent the high frequency and static dielectric constants, respectively. The second term is a lattice contribution to ϵ, where ω_t is the transverse mode frequency and Γ_0 is the corresponding linewidth. The third is a Drude term where ω_p is the plasma frequency and Γ_D is the associated linewidth. The last term is a summation of Lorentz oscillators where f_j, ω_{0j} and Γ_j, represent the strength, frequency and linewidth of the jth oscillator respectively.

TABLE I. Fitting parameters used in this work.

	CuO	Cu	V
ϵ_∞	0.958	2.007	0.847
ϵ_s	0.825	7.428	
ω_T	1.736		
ω_p		6.78	11.067
Γ_0	0.844		
Γ_D		0.0524	4.001
f_1		1.221	2.969
f_2		3.582	1.304
ω_{01}		2.699	2.544
ω_{02}		5.690	8.751
Γ_1		1.264	1.739
Γ_2		6.352	3.940

Copper on mica and vanadium on mica were modeled using the third and fourth terms. The second term was relatively important for copper with its oxide layer. A curve fitting process based on the Levenberg-Marquardt method [9] was performed and values of $\chi^2 < 1$ were obtained for copper and $\chi^2 < 0.04$ for vanadium. Using the dielectric functions thus obtained, the Cu/V/mica system was calculated considering a 100 nm vanadium film on mica plus a 2 nm continuous Cu layer with 80% coverage of 2 nm diameter, 2nm height copper islands. The same model was used for the V/Cu/mica system but now we had 100 nm copper on mica plus 0.5 nm continuous vanadium film with 70% coverage of 2 nm diameter, 0.5 nm height vanadium islands. The dielectric function of the air/metal overlayer was obtained through Bruggeman's effective medium approximation which performed better than a Maxwell Garnett approximation. Both possibilities are included in the ellipsometer software. The fitting parameters used in this work are shown in Table I.

3. RESULTS

The results of the above mentioned process are summarized in Figs. 1 to 4. Figure 1 shows the best possible fit in the 1.5 to 5.5 eV energy interval for copper using Eq. (1) and taking into account the presence of a copper oxide (CuO) layer. Previously published data [11] was included for comparison. Figure 2 shows the results obtained for vanadium. In this case the oxide layer was of negigible thickness since no noticeable improvement in the fit was observed as the thickness was varied. We found that, using a Drude model, vanadium is better modeled than copper in the 1.50–5.5 eV energy interval. Nevertheless, a correction term based on the classical oscillator model had to be added for a better fit. Experimental values for the optical properties of vanadium are reported by Guizzetti and

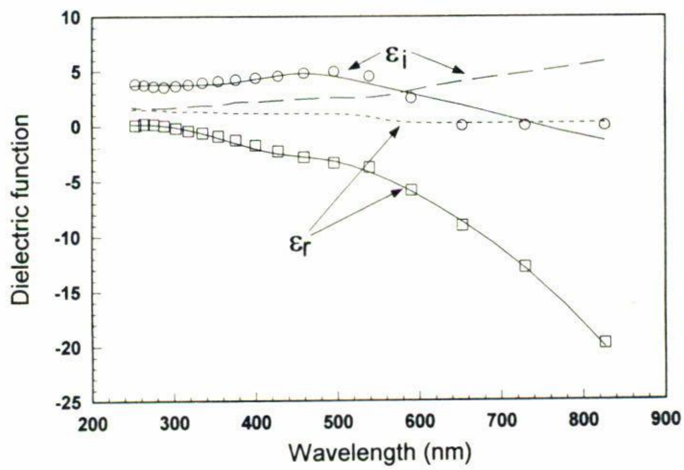

FIGURE 1. Real and imaginary parts of the dielectric function of a 100 nm copper film with a 10 nm copper oxide layer on top. The substrate is mica. Circles and diamonds are experimental, continuous lines are fitted data and dashed lines are published data from Ref. 11.

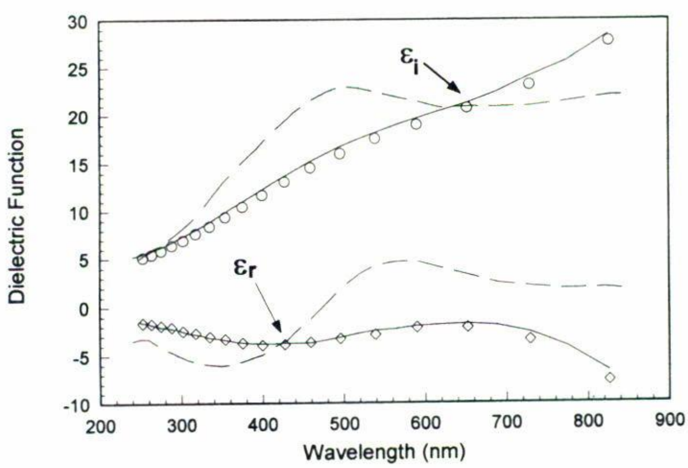

FIGURE 2. Real and imaginary parts of the dielectric function of vanadium. Discrete points are experimental, continuous lines are fitted curves and dashed lines are data from Ref. 12.

Piaggi [12] and are also presented for comparison. The copper layer optical performance in the visible region of the spectrum is not close to that of a Drude metal and the classical oscillator contribution must be taken into account. We must say that in the infrared (0.13 to 0.60 eV), a very good Drude fit was obtained. The plasma frequency ω_p in Table I was obtained from a fit in this energy interval.

Figures 3 and 4 show the result of using the dielectric functions of copper and vanadium as obtained from the single layers in the designs explained in the previous section. In the case of the 3.5 nm vanadium film on copper, the change in the optical constants of copper were as expected from the optical constants of the individual components. How-

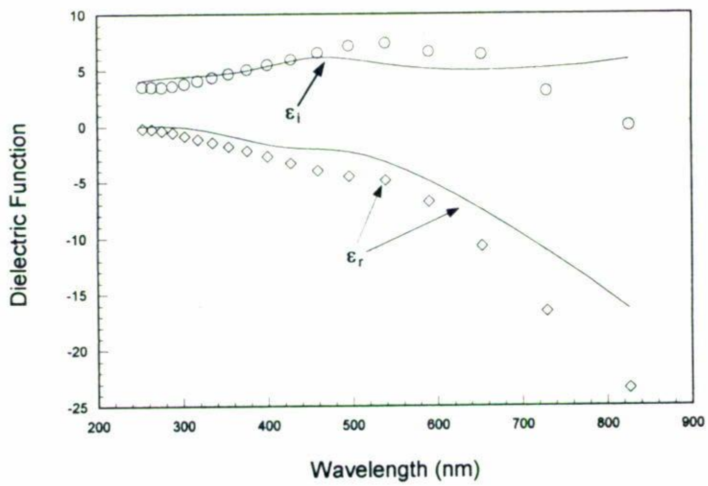

FIGURE 3. Real and imaginary parts of the dielectric function of a 3.5 nm vanadium film on a 100 nm copper layer deposited on mica. Discrete points are experimental and continuous lines are fitted curves.

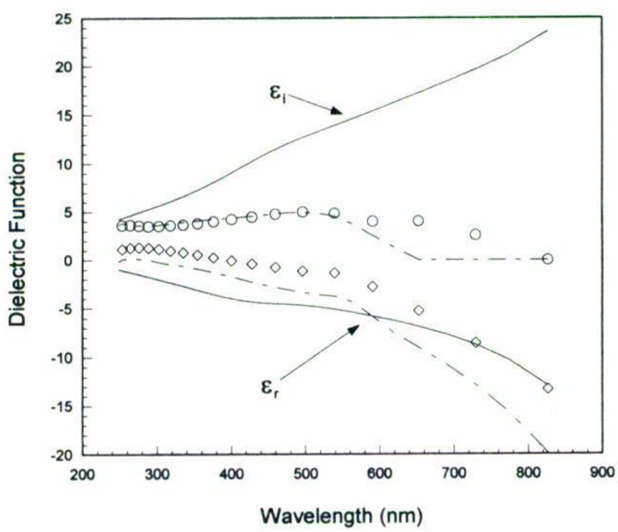

FIGURE 4. Real and imaginary parts of the dielectric function of a 3.5 nm vanadium film on a 100 nm copper layer deposited on mica. Discrete points are experimental, continuous lines are fitted curves and dashed lines are experimental data for copper on mica.

ever, for 3.5 nm of copper on vanadium, the dielectric constants were closer to those of copper covered with an oxide layer than to those of the Cu/V/mica system at the corresponding ratio. This behavior may be atributed in part to the interband transitions of the copper electrons in the studied spectral range and, in a lesser extent to the nonlocal nature of the response of copper such as the anomalous skin effect [11].

4. CONCLUSIONS

Using experimental ellipsometric measurements, we have evaluated the optical performance of the Cu/V/mica and V/Cu/mica systems deposited at room temperature by RF sputtering on mica substrates cleaved in air. The reproducibility is very good under the same conditions. Because of the contribution of the interband transitions in the copper, the theoretical fit was better for the pure vanadium than for the pure copper by an order of magnitude in the visible range of the spectrum. Oxide layers of copper and vanadium were taken into account.

For the 3.5 nm vanadium film on copper, the change in the optical constants of the composite system was as expected from the optical constants of the individual components at the appropriate ratio. However, for 3.5 nm of copper on vanadium, the electrons in copper act to screen the external fields from the vanadium film resulting in a strong influence of the copper properties in the vanadium. It is found that interband transitions and the nonlocality of the cover metal play an important role in the optical characterization of the composite system.

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