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Effect of film thickness on the optical behavior of silver nanoparticles/polymer nanocomposite films

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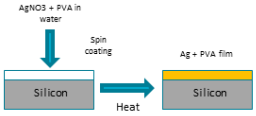
Introduction

- **Optical properties of metal nanoparticles** (NPs) determined by a **collective oscillation of the conduction electrons : surface plasmon polariton resonance (SPPR).**
- **Influence of the matrix on the SPPR :** $\epsilon_{Ag}(\omega) = -2\epsilon_{PVA}(\omega)$
- ***In situ* synthesis of the NPs in a poly(vinyl alcohol) (PVA) matrix** by chemical reduction of the metal salt during thermal annealing of the film
- **Preparation of Ag-PVA films** with high (25% w:w) and low doping levels (2.5% w:w) **by spin-coating** of the doped polymer solution on silicon wafers



- Study of the **film topography by AFM** in true non-contact mode
- Study of the **optical properties by spectroscopic ellipsometry** to simultaneously access to the thickness of the film and to the frequency-dependent dielectric function $\epsilon(\omega) = N^2(\omega)$

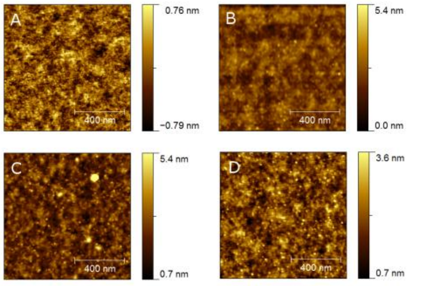


Figure 1 : AFM images of the polymer films topography (1 µm x 1 µm): A, pure PVA film; B, 25 nm thick doped film (Ag/PVA ratio: 2.5%); C, 25 nm thick doped film (Ag/PVA ratio: 25%); D, 290 nm thick doped film (Ag/PVA ratio: 25% w: w).

Spectroscopic ellipsometry

Spectroscopic ellipsometry : Non-destructive optical analysis technique based on the relative change of polarization of the *p*- and *s*- components of the light at the interface between two media characterized by different optical properties

$$\rho = \frac{r_p}{r_s} = \tan\Psi\, e^{i\Delta} \qquad \alpha = \cos(2\Psi) \qquad \beta = \sin(2\Psi)\cos(\Delta)$$

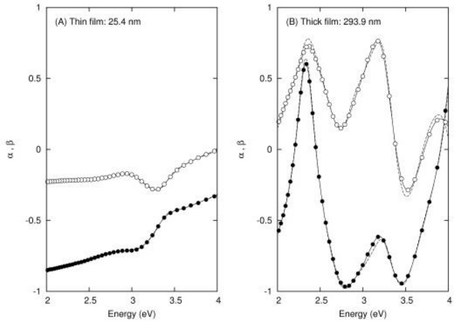


Figure 2 : Ellipsometric spectra of Ag -PVA films (Ag/PVA ratio: 25% w: w): A,thin films (thickness: 25.4 nm); B, thick film (thickness: 293.9 nm). Experimental data: α (filled circles) and β (open circles). Dashed lines: optimized results from the optical model.

Table 1: Parameters of the plasmon absorption peak (A_0: amplitude of the absorption peak; λ_0: position of the resonance; Γ_0: width of the resonance) as a function of the film thickness for highly doped PVA films (Ag/PVA ratio: 25% w: w). Data correspond to the optical properties presented in Fig. 3.

Sample	Thickness (nm)	A_0	λ_0 (nm)	Γ_0 (nm)
Thin films	23.4 ± 0.2	0.145 ± 0.6	414.2 ± 0.7	67.6 ± 2.9
	25.4 ± 0.3	0.133 ± 0.5	415.6 ± 0.6	69.0 ± 2.6
Thick films	305.9 ± 1.7	0.117 ± 0.2	405.4 ± 0.7	47.3 ± 1.6
	293.4 ± 1.7	0.118 ± 0.2	409.5 ± 0.6	49.2 ± 1.5

Optical properties of thick and thin films

Significant difference in the refractive index of thin and thick films at constant doping level

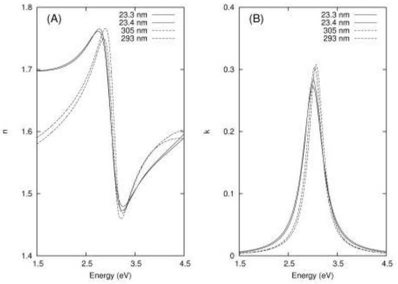


Figure 3 : Optical properties of thin (solid lines) and thick (dashed lines) silver NPs-doped PVA films (Ag/PVA ratio: 25% w: w): A, refractive index n; B, extinction coefficient k.

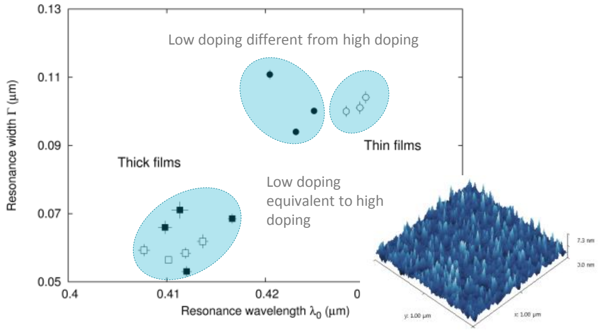


Figure 4 : Resonance width versus resonance wavelength for PVA films with high (open symbols) and low Ag doping levels (plain symbols)

Conclusion and acknowledgements

- **In situ synthesis of silver NPs by thermal annealing of a polymer film containing metal salt**
- **Simultaneous determination of thickness and optical properties : SPPR localized at ~ 3 eV**
- **At high doping level (25% Ag) : different behavior between thin and thick films (2D → 3D)**

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