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Optical constants of gold-silver-copper alloy system

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1. Introduction

Plasmon resonance using gold – silver – copper alloy system is promising materials in the field of enhanced fluorescence, surface enhanced Raman scattering (SERS) and so on. The resonance wavelength and electro-magnetic field enhancement of plasmon resonance depends on the permittivity of the metal. We have demonstrated that the optical constants of gold – silver alloy system become different features from the linear combination of each optical constant [1], and revealed the importance of experimentally determination of optical constants. In this study, we determined the optical constants of gold-silver, gold-copper, silver-copper alloy system and explained the relationship between the optical constants and the Drude parameters (ω_p; plasma frequency and τ; relaxation time)

2. Experiments

Gold – silver – copper alloy thin film was prepared using thermal evaporation (ULVAC, VPC – 410S) and alloying by alternately deposition method. The alloy film was evaporated on a glass substrate. Composition of the alloy was controlled by measuring the thickness of metals by quartz crystal microbalance. The thickness of each layer was designed to less than 1 nm and the total alloy thickness was designed to 20 nm, 30 nm, and 50 nm. We determined the optical constants by measuring the transmission and reflection spectra of these three alloy films and fitting the least square method to the Lorentz – Drude model shown in eq. 1.

$$\varepsilon(\omega)=\varepsilon(\infty)-\frac{\omega_p^{\ 2}}{\omega^2+i\omega\Gamma}+\sum_j\frac{A_j\hbar\omega_{0,j}}{(\hbar\omega_{0,j})^2-(\hbar\omega)^2-i\Gamma_j\hbar\omega} \qquad (1)$$

Where $\varepsilon(\infty)$ represents permittivity at the infinite frequency, ω_p is plasma frequency, Γ is decay constant (=1/τ), j is number of Lorentz oscillator (in this work, we assumed j = 15), A_j is oscillator amplitude, $\omega_{0,j}$ is oscillator resonant frequency, Γ_j is oscillator decay constant, $\hbar$ is reduced Planck constant, i is imaginary unit, respectively. This model consists of the flee electron and bound electron behaviors. Therefore this can be applied to full wavelength range of transmission and reflection spectra.

3. Results and Discussion

Plasmon resonance is the phenomena, which give raised by the free electron oscillation. The parameter of Drude part (ω_p and τ) plays important role for the optical properties of plasmon resonance. Figure 1. shows experimentally determined ω_p and τ. The values obtained from pure metals are well matched to the references of Johnson and Christy's results [2].

We noticed that the density of free electron decreased and the relaxation time increased around the 50% two - component alloy system. This result indicated that the bonding strength in the alloy crystal got stronger and then decreased the amount of free electron so that the remaining free electron could vibrate longer time in the case of gold-silver, gold-copper, and silver-copper alloy system.

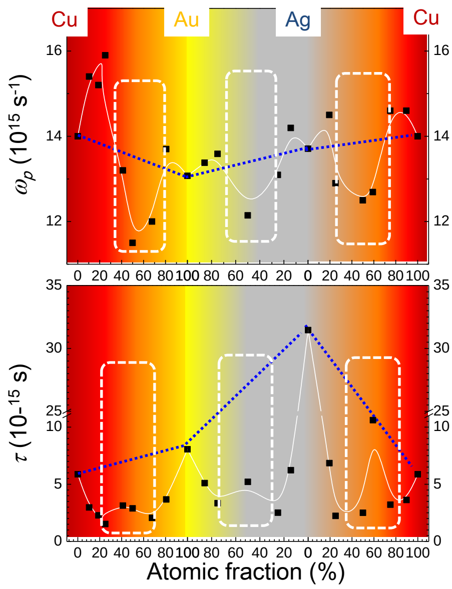

Figure 1. Plasma frequency and relaxation time of the Gold - silver – copper alloy system.

References

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[2] P. B. Johnson and R. W. Christy, Phys. Rev. B vol. 6, no. 12, pp. 4370–4379, 197