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Surface plasmon resonance properties of CuAg films prepared by laser dewetting process

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ABSTRACT

Single-layer Cu and Ag films, as well as bilayer Cu/Ag films, each with a thickness of 6 nm, were deposited on glass substrates by high-vacuum thermal evaporation. Metal nanoparticles were fabricated through laser-induced dewetting using a 1064 nm laser operated at various power levels. The effects of laser power and Ag content on the size of the synthesized nanoparticles and the localized surface plasmon resonance (LSPR) behavior of the films were systematically investigated. The LSPR peak wavelength showed a blue shift with increasing Ag content and laser power, enabling a tunable range from 659 to 406 nm. Overall, the results demonstrate that the LSPR peak position can be effectively tuned by adjusting the laser power and material composition of the nanoparticles. This approach shows strong potential for tailoring the optical properties of thin films for advanced photonic and plasmonic applications.

1. Introduction

Nanoparticles (NPs) exhibit many unique properties, such as enhanced surface plasmon resonance (SPR) [1] and strong antimicrobial activity [2]. The resonant properties of NPs are highly dependent on their composition, shape and size [3]. Compared to their bulk counterparts, nanomaterials possess distinct optical, magnetic, electronic, and mechanical properties, which have led to their use in diverse applications, including optoelectronics [4], biomedicine [5], antimicrobial treatment [6], and Raman detection [7]. Among the various metals used in NP synthesis, silver (Ag) and gold (Au) are the most commonly used. Gold NPs have excellent biocompatibility and chemical stability, while silver NPs exhibit excellent localized surface plasmon resonance (LSPR). Leveraging these advantages, many researchers have developed bimetallic Ag—Au NPs to broaden their application beyond that of the respective pure metal NPs. These NPs are generally produced by thermal annealing or laser-induced dewetting [8]. Both processes yield an effective widening of the LSPR region [9] by altering the shape and size of the grains in the metal films.

The optical extinction spectra of Ag, Cu, CuZr, Au and AgCuAl [7,10–12] nanostructures occurs at wavelengths of 400–450 nm, 700 nm, 580 nm, 560 nm, and 600–620 nm, respectively. However, Ag and Au are expensive, which limits their potential for widespread use. Consequently, copper (Cu) is regarded as a more practical and cost-effective alternative. Although Cu NPs have an LSPR behavior comparable to that of noble metals, their plasmonic performance is generally weaker. Nonetheless, Cu has many advantageous properties for practical applications, such as high electrical conductivity, strong catalytic activity, good antibacterial efficacy, and suitability for drug delivery. Consequently, Cu NPs are widely applied across various fields, including electronics, chemistry, and biomedicine. Wang [13] reported that the island structures of as-sputtered Ag films transformed into NPs under high-temperature annealing, with the NP size decreasing with increasing laser energy.

In this study, single-layer (Cu and Ag) and bilayer (Cu/Ag) thin films were deposited on glass substrates [14]. Laser dewetting processes were employed to fabricate NPs in the deposited films by laser dewetting using a 1064 nm laser at varying power levels. The nanostructures, NP size distributions and LSPR behaviors of the resulting films were systematically explored. The findings indicated that the laser dewetting process led to an effective reduction in the NP size. Moreover, the addition of Ag to the Cu film in the bilayer configuration significantly influenced the LSPR response, leading to a pronounced blue shift in the absorption spectrum.

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2. Experiment

Four types of metal films were deposited on glass substrates with a thickness of 0.7 mm using a high-vacuum evaporation system: (1) pure Cu with a thickness of 6 nm, (2) Cu (4 nm) and Ag (2 nm), (3) Cu (2 nm) and Ag (4 nm), and (4) pure Ag with a thickness of 6 nm. For convenience, the four films were designated as Cu, Cu4/Ag, Cu2/Ag, and Ag, respectively.

The laser dewetting process was conducted using a fiber laser with a wavelength of 1064 nm, a spot size of 40 μm and a pulse duration of 200 ns. The laser operated at a repetition rate of 100 kHz and a fixed scanning speed of 200 mm/s, with power levels of 4, 6, 8, and 10 W. The optical properties of the thin films were measured over the wavelength range of 200–1100 nm using a UV–Vis spectrophotometer (Lambda 35). The microstructures of the thin films were characterized using a scanning electron microscope (SEM, JSM-7600F) equipped with an energy-dispersive X-ray spectroscope (EDS, X-MaxN50TLE). The NP size of the dewetted films were analyzed using ImageJ software (National Institutes of Health, USA).

3. Results and discussions

The as-deposited Cu, Cu4/Ag, Cu2/Ag, and Ag films exhibited smooth and continuous surfaces (Fig. 1). The EDS analysis results confirmed that the Cu and Ag elements were evenly distributed in the Cu/Ag films. Moreover, the absorbance spectra of the four films showed no distinct absorption peaks, indicating that the unprocessed films did not contain NPs.

Fig. 2 shows SEM images of Cu, Cu4/Ag, Cu2/Ag, and Ag films dewetted using laser powers ranging from 4 to 10 W. The Cu film underwent effective dewetting, forming uniform and densely packed NPs. Particle diameter trend of the four films dewetted using laser powers ranging from 4 to 10 W in Fig. 3. As the laser power increased from 4 to 10 W, the average particle size decreased from 70 to 47 nm. For the Cu4/Ag film, uniform and dense NPs were also produced, with the particle size decreasing from 58 to 49 nm as the laser power increased from 4 to 8 W. The Cu2/Ag film similarly underwent successful dewetting, forming NPs with sizes ranging from 61 nm (4 W) to 53 nm (10 W).

Fig. 4(a–d) shows that the LSPR peak wavelength underwent a significant blue shift with an increasing Ag content and laser power. For the Cu, Cu4/Ag, Cu2/Ag, and Ag films, the LSPR peak shifts induced by increasing the laser power from 4 to 10 W were 659–622 nm, 587–574 nm, 528–523 nm, and 408–406 nm, respectively. In other words, increasing Ag content, and laser power resulted in a progressive blue shift of the LSPR peak, with the overall range spanning from 659 nm to 406 nm. While the LSPR peak position decreased progressively with an increasing Ag content, the NP size remained relatively constant unless laser dewetting parameters were changed. Fig. 4(e-h) summarizes the variations in the particle size and LSPR peak wavelength with the laser power for the four films. The results suggest that a higher laser power increased the heat accumulation in the film, which promoted the formation of smaller NPs. As the laser power increased from 4 to 10 W, the average particle size decreased from 70 to 47 nm. The smaller NP size, in turn, prompted a shift in the LSPR peak toward a shorter wavelength. A tunable LSPR peak wavelength is from 659 to 406 nm. Raveendran et al. [15] used the relative compositions of Au and Ag to tune the LSPR peak over a range of 410 to 540 nm. In contrast, the present study synthesized Cu—Ag NPs and achieved a broader tunable range of 406 to 659 nm, offering both a wider spectrum and a lower material cost. Overall, after laser dewetting processing, the Cu sample exhibited a notable blue shift in the LSPR peak, indicating a reduction in the NP size. The Cu4/Ag and Cu2/Ag sample again demonstrated a clear blue shift owing to a smaller NP size. For the pure Ag sample, the LSPR peak position remained relatively unchanged.

4. Conclusions

This study examined the effects of laser dewetting methods on the formation of NPs in single-layer (Cu and Ag) and bilayer (Cu2/Ag and Cu4/Ag) thin films. The investigation focused on the effects of laser power, and Ag content on the size of the NPs formed in the films and the resulting LSPR behavior. For laser irradiation, the NP size decreased from approximately 70 nm to 47 nm as the laser power increased from 4 to 10 W, and caused a blue shift in the LSPR peak. When Ag was added to the Cu film, the LSPR peak exhibited a significant blue shift, ranging from 659 nm to 406 nm, as the Ag content increased. Overall, the results of this study demonstrate that the LSPR peak position can be effectively controlled by adjusting the laser power and Ag content through the laser dewetting process. Therefore, laser dewetting using a bilayer configuration offers a promising strategy for tuning the optical properties of thin films for advanced plasmonic applications.

CRediT authorship contribution statement

Hsuan-Kai Lin: Writing – review & editing, Writing – original draft, Supervision, Project administration, Funding acquisition, Conceptualization. **Cheng-En Zhong:** Methodology, Investigation, Formal analysis, Data curation. **Chao-En Yu:** Visualization, Methodology, Formal

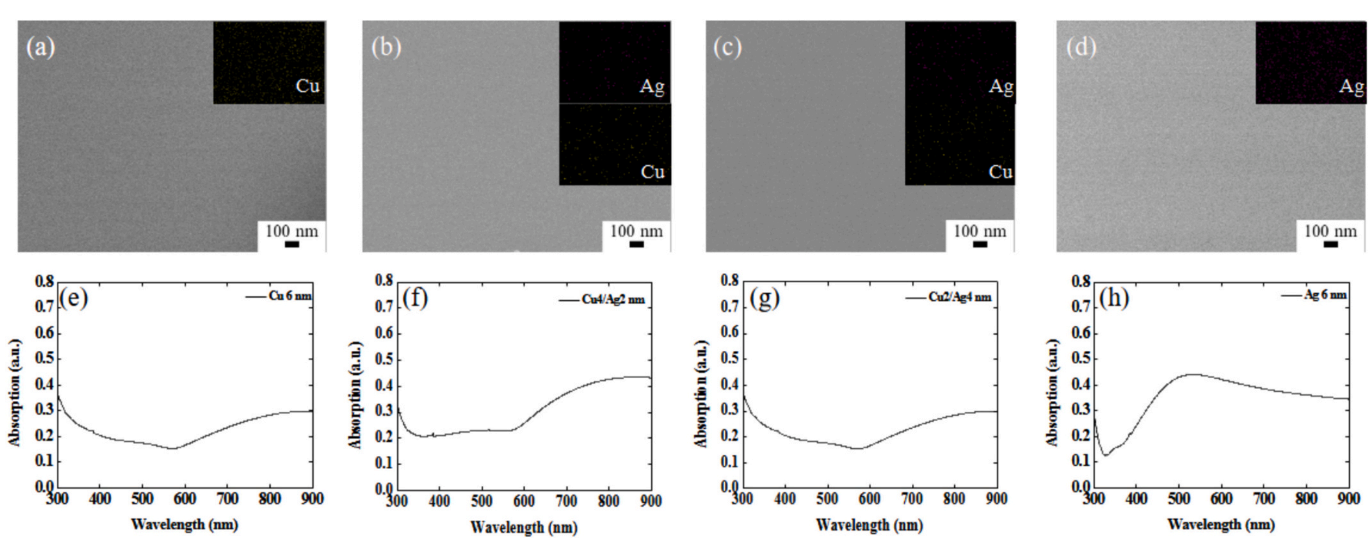

Fig. 1. SEM images and corresponding absorption spectra of the as-deposited films: Cu (a, e), Cu4/Ag (b, f), Cu2/Ag (c, g), and Ag (d, h).

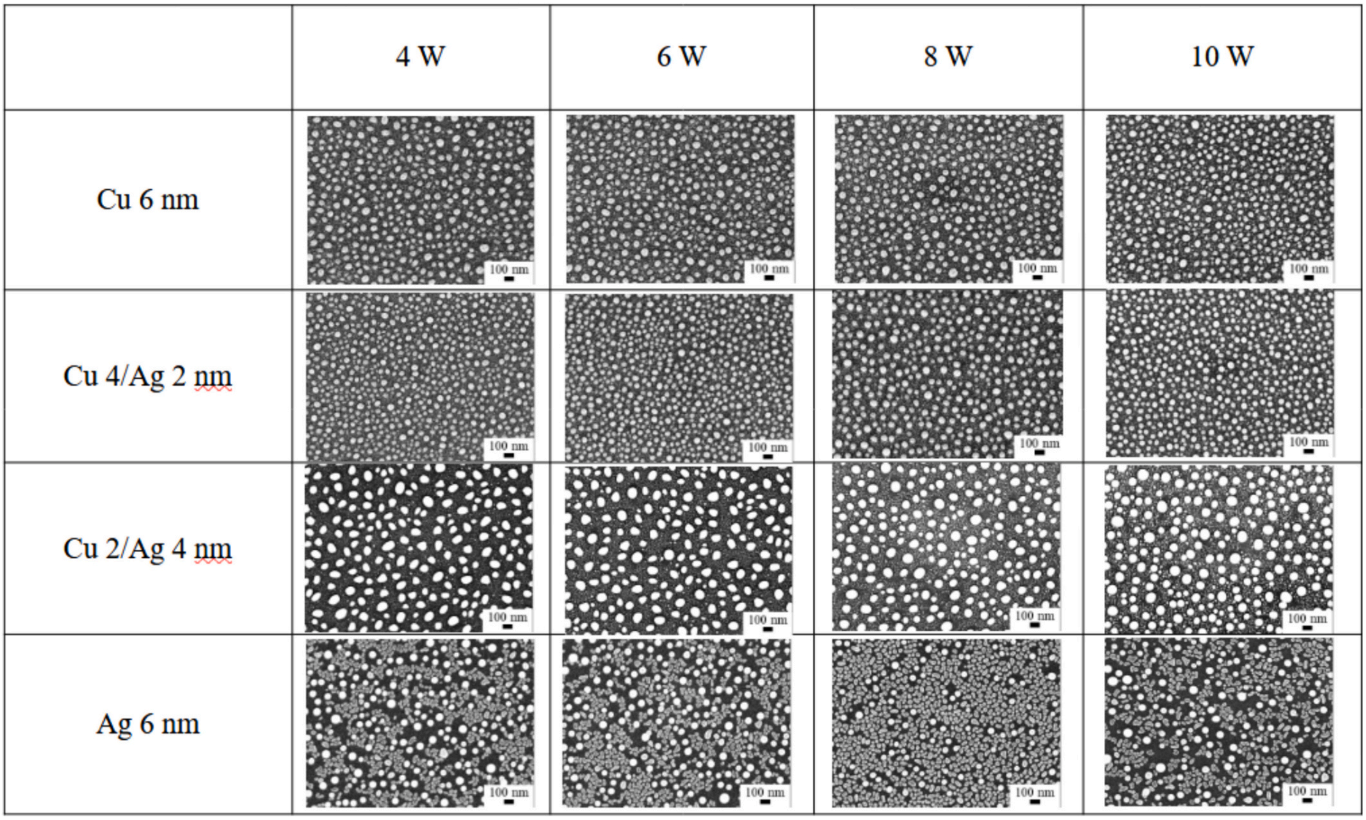

Fig. 2. SEM images of Cu, Cu4/Ag, Cu2/Ag, and Ag films dewetted using laser powers ranging from 4 to 10 W.

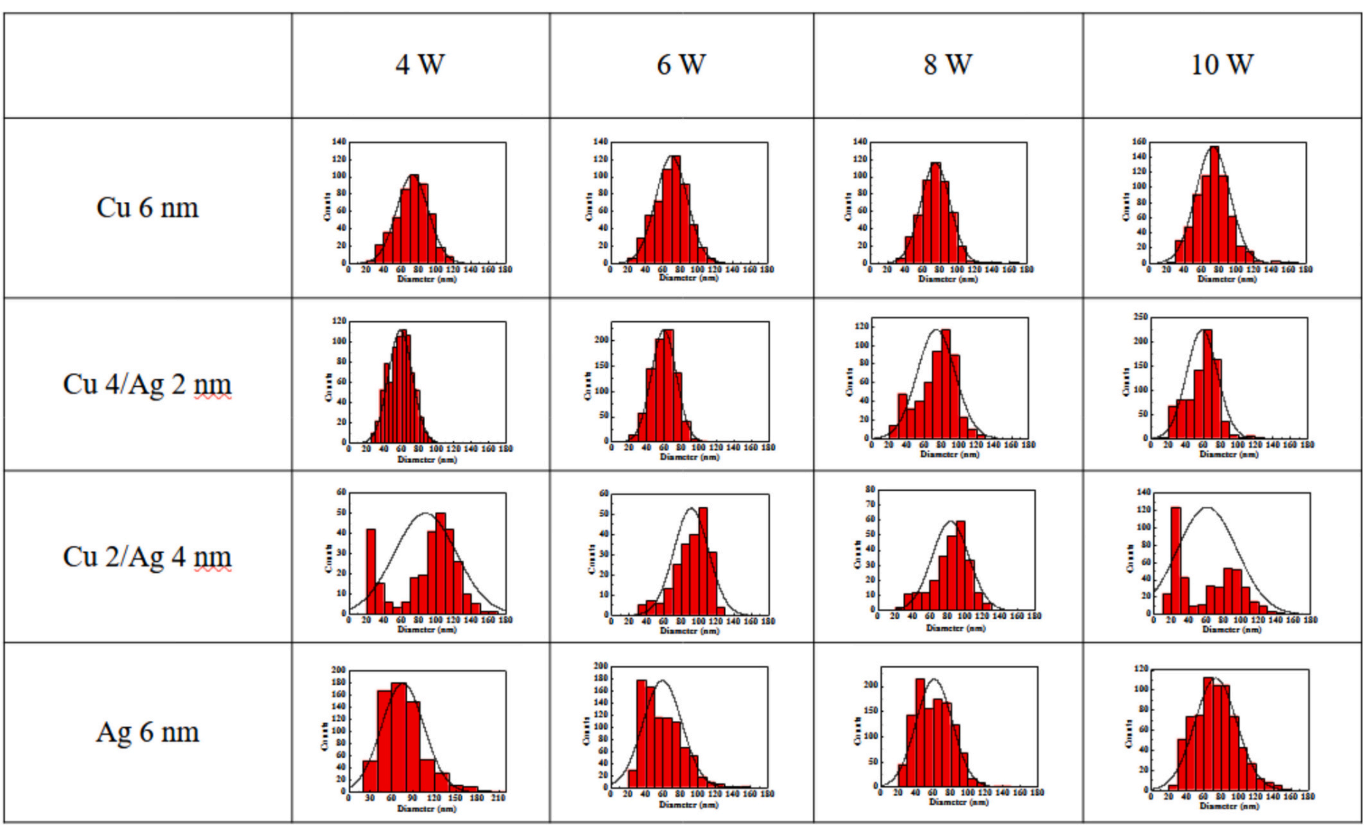

Fig. 3. Particle diameter trend of Cu, Cu4/Ag, Cu2/Ag, and Ag films dewetted using laser powers ranging from 4 to 10 W.

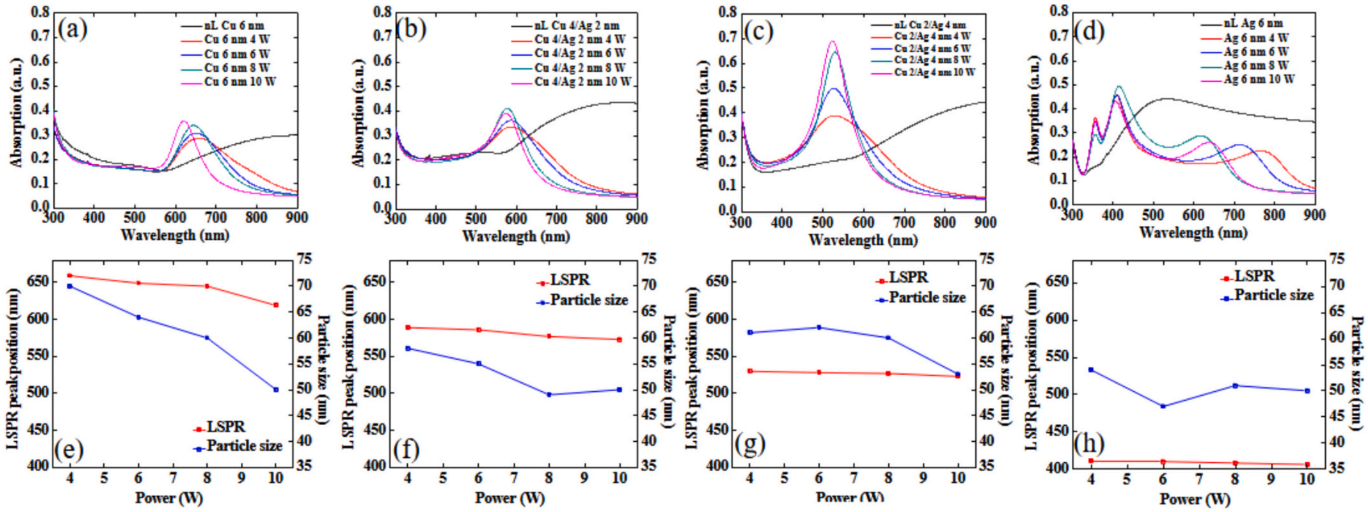

Fig. 4. Variation in particle size, absorption spectra, and LSPR peak positions of Cu (a, e), Cu4/Ag (b, f), Cu2/Ag (c, g), and Ag (d, h) films dewetted at laser powers ranging from 4 to 10 W.

analysis, Data curation. **Chien-Hsing Chen:** Writing – original draft, Resources, Methodology, Investigation. **J.Y. Cheng:** Resources, Methodology, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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Data availability

Data will be made available on request.

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