Supplementary Information

Design rules for highly transparent electrode using dielectric constant matching of metal oxide with Ag film in optoelectronic devices **

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Optical simulation

Optical simulation involving a multilayer structure is performed by using commercial software (The Essential Macleod, Thin Film Center, Inc.) based on what is known as the characteristic matrix method.^{1,2} Providing the incidence medium and the emission medium is a half-infinite non-absorption medium, its characteristic matrix can be formulated as followed,

$$\begin{bmatrix} B \\ C \end{bmatrix} = \left\{ \prod_{r=1}^{k} \begin{bmatrix} \cos \overline{\delta_r} & j \sin \overline{\delta_r} / \overline{\eta_r} \\ j \overline{\eta_r} \sin \overline{\delta_r} & \cos \overline{\delta_r} \end{bmatrix} \begin{bmatrix} 1 \\ \eta_{k+1} \end{bmatrix}$$

where B, C, and δ are electrical field, magnetic field, and the phase thickness as follows:

$$\overline{\delta_r} = \frac{2\pi}{\lambda} N_r d_r \cos\overline{\theta_r} = \frac{2\pi}{\lambda} (n_r - jk_r) d_r \cos\overline{\theta_r}$$

and is the complex admittance in the horizontal and vertical directions, respectively, as follows:

$$\overline{\eta_r} = N_r \cos \overline{\theta_r} = (n_r - jk_r) \cos \overline{\theta_r}$$

So, the transmittance of the general multilayer film system is:

$$T = \frac{4\eta_0\eta_{k+1}}{(\eta_0 B + C)(\eta_0 B - C)}$$



Figure S1. The optical constants (refractive index, extinction coefficient) of (a) Ag and (b)-(f) various metal oxides used for optical analysis.

Figure S1 shows the refractive index and extinction coefficient of Ag and various metal oxides as a function of wavelength. The optical constants were measured by using spectroscopy ellipsometry. These values were used for optical simulation.

Frequency dependent relative permittivities of various kinds of metal oxide materials



Figure S2. Measured relative permitivities for various kinds of metal oxides.

Figure S2 shows the measured relative permitivities (ε) for various kinds of metal oxides. The WO₃, CaO, and Ga₂O₃ showed relatively large values of 35.2, 21.8, and 10.7, respectively. In the meanwhile, the glass, SiO₂, and MgO showed relatively small values of 5.7, 5.2, and 5.4, respectively.





Figure S3. Dark field reflectance images of (a) glass/Ag, (b) WO_3/Ag , (c) glass/Pt, and (d) WO_3/Pt samples. (d) UV-vis absorbance spectra and (e) optical transmittance spectra of four kinds of samples.

To clarify the effect of plasmon frequency on SPs in metal films, we observed dark-field reflectance images of Ag-based double layers (glass/Ag and WO₃/Ag) and Pt-based double layers (glass/Pt and WO₃/Pt) (Figs. S3(a)-(d)). The brightness of dark-field images can be enhanced in the glass/Ag sample, due to the collection of the scattered light, via, SPs coupling. However, the WO₃/Ag sample showed relatively dark image (Fig. S2(b)) due to the large dielectric constant of WO₃.

In the meanwhile, both of glass/Pt and WO₃/Pt samples showed relatively dark images, irrelevant to dielectric layers underneath due to the small plasmon frequency (<2 eV) of Pt. In UV-visible absorbance spectra, glass/Ag sample showed broad peaks induced by SP absorption in the visible region which were not seen in the WO₃/Ag and Pt-based samples (Fig. S3(e)).

The measured transmittance spectra of glass/Ag sample showed low transmittance (< 50 %) in visible region (Fig. S3(f)). When we employed the WO₃ layer between glass and Ag layers, the WO₃/Ag sample showed enhanced optical transmittance (~70 %). In the meanwhile, both of glass/Pt and WO₃/Pt samples showed low optical transmittance due to the low plasmon frequency and large *k* value of Pt. These results indicate that the Ag has the plasmon frequency in visible range and we can control the SPs coupling and optical property of Ag-based films.

References

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- 2 H. A. Macleod, Thin-film Optical Filters. New York: Taylor & Francis, 2001.