



Thickness and microstructure effects in the optical and electrical properties of silver thin films

Guowen Ding, César Clavero, Daniel Schweigert, and Minh Le

Citation: [AIP Advances](#) **5**, 117234 (2015); doi: 10.1063/1.4936637

View online: <http://dx.doi.org/10.1063/1.4936637>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/adva/5/11?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Electrical and microstructural properties of N + ion-implanted ZnO and ZnO:Ag thin films](#)

J. Vac. Sci. Technol. A **29**, 03A108 (2011); 10.1116/1.3554836

[Effect of Ag thickness on electrical transport and optical properties of indium tin oxide–Ag–indium tin oxide multilayers](#)

J. Appl. Phys. **105**, 123528 (2009); 10.1063/1.3153977

[Microstructure and magnetic properties of Co Pt - Si N x/Ag thin films](#)

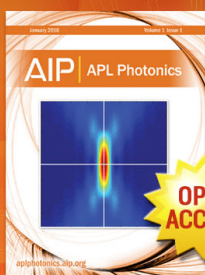
J. Appl. Phys. **103**, 07F514 (2008); 10.1063/1.2832342

[Structural, electrical, and optical properties of p -type ZnO thin films with Ag dopant](#)

Appl. Phys. Lett. **88**, 202108 (2006); 10.1063/1.2203952

[Effects of Ag underlayers on the microstructure and magnetic properties of epitaxial FePt thin films](#)

J. Appl. Phys. **89**, 7068 (2001); 10.1063/1.1360683



Launching in 2016!

The future of applied photonics research is here

OPEN
ACCESS

AIP | APL
Photonics

Thickness and microstructure effects in the optical and electrical properties of silver thin films

Guowen Ding,^a César Clavero, Daniel Schweigert, and Minh Le
Intermolecular, Inc., 3011 North First Street, San Jose, CA 95134 USA

(Received 1 September 2015; accepted 12 November 2015; published online 23 November 2015)

The optical and electrical response of metal thin films approaching thicknesses in the range of the electron mean free path is highly affected by electronic scattering with the interfaces and defects. Here, we present a theoretical and experimental study on how thickness and microstructure affect the properties of Ag thin films. We are able to successfully model the electrical resistivity and IR optical response using a thickness dependent electronic scattering time. Remarkably, the product of electronic scattering time and resistivity remains constant regardless of the thickness ($\tau\rho=C$), with a value of $59\pm2\ \mu\Omega\text{ cm}\cdot\text{fs}$ for Ag films in the investigated range from 3 to 74 nm. Our findings enable us to develop a theoretical framework that allows calculating the optical response of metal thin films in the IR by using their measured thickness and resistivity. An excellent agreement is found between experimental measurements and predicted values. This study also shows the theoretical lower limit for emissivity in Ag thin films according to their microstructure and thickness. Application of the model presented here will allow rapid characterization of the IR optical response of metal thin films, with important application in a broad spectrum of fundamental and industrial applications, including optical coatings, low-emissivity windows and semiconductor industry. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4936637>]

I. INTRODUCTION

The study of electric conduction and optical response in metals has now more than a hundred years of history.¹⁻³ Drude postulated the existence of a gas of free electrons traveling among the metal ions.⁴ According to this model, two main parameters govern the electronic response: (i) the electron collision time τ , a statistical parameter describing the mean time between collisions (electron-electron, electron-phonon, etc.), and (ii) the plasma frequency ω_p , mainly determined by the concentration of carriers. Lorentz subsequently analyzed the electronic behavior using the dynamical theory of gases.⁵ The Drude-Lorentz theory was able to successfully predict the law connecting the electrical and the thermal conductivities. Further contributions by Pauli and Sommerfeld allowed application of some aspects of the quantum theory leading to the Drude-Lorentz-Sommerfeld model. The Drude model itself describes extremely well the free electron contribution to the optical response of metals such as Ag, Au and Cu,⁶ which dominates the near infrared and infrared response. However, surface electronic scattering effects are extremely important when the dimension of the metal system approaches the electron mean free path and need to be carefully considered. It has been shown how the optical response of metal nanoparticles with diameters in the few nanometers range can be described using the Drude model by considering a size-dependent reduction of the electron scattering time due to electron scattering with the interfaces.⁷⁻⁹ For the case of metal thin films, the applicability of the Drude model still remains largely unexplored. Here, we show how the optical response of Ag thin films can also be described using the Drude model by introducing a thickness and microstructure dependence in the electron scattering time.

^agding@intermolecular.com

Ag is one of the best conducting metals and is widely used in the modern world for electrical conduction in multiple industries and applications, including light-emitting diodes (LED), and flat-panel displays (FPD) to name a few. Ag thin films have also been widely applied in optical applications, mainly due to their high transparency and neutral color in the visible range. They also exhibit extremely high reflectivity in the near infrared (IR) and IR parts of the spectrum.¹⁰ These properties make Ag thin films ideal candidates for low-thermal-emissivity (Low-E) coatings on glass panels, leading to energy-efficient windows for residential and commercial buildings. For the last twenty years, low-E coatings have motivated an intense research activity to improve the growth mode and optical properties of Ag thin films, ultimately leading to substantial energy savings.

For this study, two kinds of Ag systems were investigated. Ag was directly deposited on glass in the case of the first series. This typically leads to inferior Ag performance owing to high roughness, lack of continuity and small grain size. Due to this, only films with thicknesses higher than 12 nm are representative. However, the simplicity of this system allows for optical modeling with minimum assumptions, providing more confidence and simplicity to the theoretical study. In a second series, samples comprising a more optimized seed/capping layer system known to lead to superior Ag performance were deposited. ZnO acting as seed layer for Ag^{11,12} combined with NiCr or Ti oxide¹³ as top barrier leads to a more continuous Ag growth, smoother interfaces, larger grain size and ultimately a lower resistivity. This approach needs more assumptions during optical modeling to study the silver film, but provides a method to study silver thickness down to 3 nm.

Our studies show the theoretical lower limit for emissivity in Ag thin films according to their microstructure and thickness, which will allow prediction of the maximum energy savings achievable by these kinds of devices. We also provide a method for rapid characterization of the optical response of metal thin films from resistivity measurements.

II. EXPERIMENTAL

Ag thin films were deposited on glass substrates using Intermolecular's Tempus™ physical-vapor deposition (PVD) chamber, which has four magnetron sputtering guns and an aperture above the substrate to provide uniform depositions within the site-isolated region (Figure 1). This chamber configuration supports simultaneous deposition of up to four types of materials, and allows for dozens of experiments to be performed on a single substrate in a single vacuum pump-down, thus it accelerates the cycles-of-learning in materials R&D. The chamber base pressure was typically kept at 5×10^{-8} Torr or below.

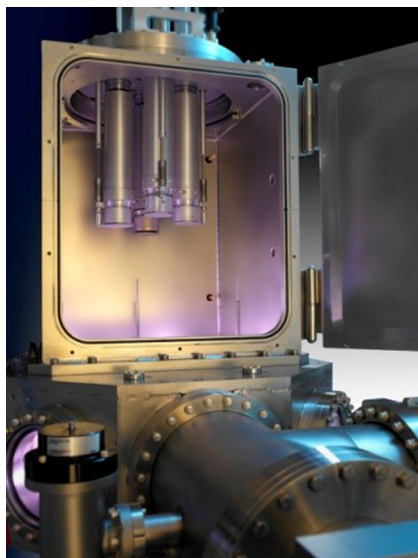


FIG. 1. Intermolecular Tempus™ physical-vapor deposition (PVD) chamber capable of depositing four different materials in site-isolated regions without vacuum break.

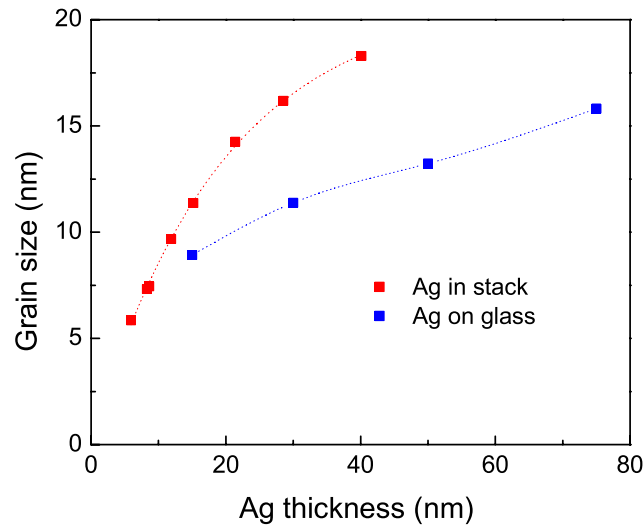


FIG. 2. Ag grain size measured for the case of films deposited on glass and in stack configuration.

Two kind of samples were prepared. In a first series, Ag thin films with thicknesses between 15 and 75 nm were sputter deposited in DC mode on glass substrates using an Ar pressure of 2 mT. In a second series, stacks with the structure glass/10nm TiO₂/10nm ZnO/x nm Ag/2nm TiO₂/10nm ZnO were deposited, with only the Ag thickness varying from 3 to 40 nm. The initial TiO₂ layer on glass is used to avoid glass surface defects impacting the performance. The 10 nm TiO₂ films were deposited by reactive magnetron sputtering at 2 mT with 300 W pulsed DC power. Subsequently, 10 nm ZnO was deposited by reactive magnetron sputtering at 2 mT with 150 W pulsed DC power. It is well known that ZnO as seed layer enhances the Ag optical and electrical performance.^{11,16} After the Ag deposition with 90 W DC power, a 2 nm thick TiO₂ barrier layer followed by a 10 nm ZnO layer was deposited. This type of multilayer stacks are a common approach used by the low-E industry for excellent transmission and emissivity performance.¹⁴ Figure 2 shows the grain size measured for the Ag films deposited on glass, and also for those in the described stacks. Larger grains are obtained in the stack systems for each Ag thickness, which is directly related to a better epitaxial growth of Ag on ZnO and also the protection provided by the top layers. The thickness of the films was measured by ellipsometer and confirmed by XRR and TEM.

The optical characterization was carried out by measuring spectral ellipsometry and transmittance using a Woollam M2000 ellipsometer and a Perkin-Elmer Lambda 1050 spectrometer, respectively. The spectral ellipsometry parameters (Ψ and Δ) measured at three different angles (60, 65 and 70°), along with the spectral transmittance, were used to uniquely calculate the three unknowns: n , k and thickness of the single silver layer on glass. The optical constants were calculated assuming that the Ag film surface is smooth and uniform on glass, which reflects the experimental observations in PVD deposition with a 2% thickness standard deviation. For the optical analysis of the Ag stack systems, the optical constants for the non-Ag layers were obtained by measuring different single layer depositions. In these calculations, we assumed that the effect of the interface between layers is negligible. In fact, the transmittance and reflectance spectra simulations for such stacks showed very good agreement with the experimental observations, indicating that those assumptions are fairly accurate.

Sheet resistance measurements were carried out using a four point probe. Resistivity values were obtained using the thicknesses accurately calculated with ellipsometry and XRR. All the experimental measurements were completed within 30 minutes after the sample was exposed to atmosphere to avoid contamination effects.

III. THICKNESS EFFECT IN THE ELECTRICAL PROPERTIES

The electrical and optical properties of Ag thin films exhibit a marked dependence with thickness when this is comparable to the electronic mean free path. Figure 3 shows experimental resistivity

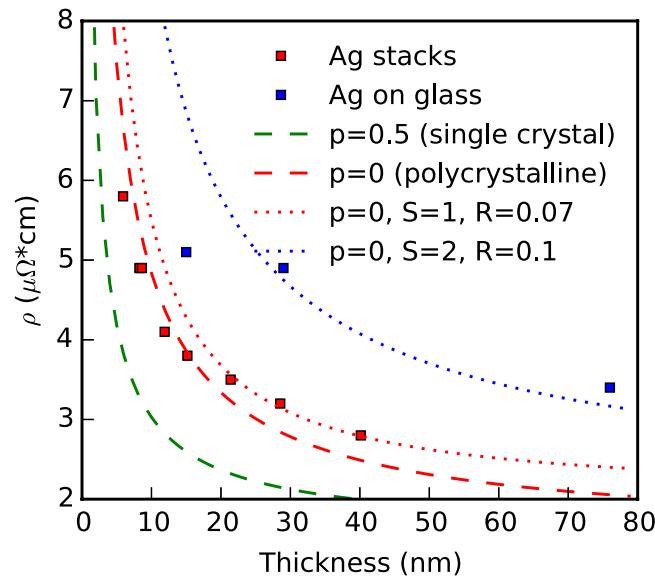


FIG. 3. Experimental electrical resistivity for Ag stacks (red dots), Ag on glass (blue dots) and calculated values using Fuchs-Sondheimer's theory for polycrystalline films ($p=0$) and single crystal films ($p=0.5$). The dotted lines show how roughness and inter-grain scattering affect resistivity using the Rossmagel and Kuan formalism.

values (ρ) for Ag films with thicknesses ranging from 3 to 74 nm and deposited using the above mentioned two methods, *i.e.* Ag on glass and multilayer Ag stack. The Ag stacks show lower resistivity for similar Ag thickness. An exponential increase in resistivity is observed for the thinner films. Such evolution can be explained using the Fuchs-Sondheimer theory,⁵ which extends the concept of scattering in bulk to thin films, and solves Boltzmann's equation considering electronic scattering by the interfaces. The resistivity of metal thin films is given by

$$\rho = \rho_i \left[1 - \frac{3}{2\kappa} (1-p) \int_1^\infty \left(\frac{1}{t^3} - \frac{1}{t^5} \right) \frac{1 - e^{-\kappa t}}{1 - p e^{-\kappa t}} dt \right]^{-1} \quad (1)$$

where $\kappa=d/l$, with d the thin film thickness, l the electronic mean free path, ρ_i the bulk resistivity, t is an integration parameter and p the probability that an electron will be specularly reflected upon scattering from one of the surfaces. Typical values for p are 0 for polycrystalline films and 0.5 for single crystal films. Characteristics such as roughness can be considered to be included in the parameter p , since they directly affect the way electrons are scattered. Using equation (1) we were able to fit our experimental resistivity data as shown in Figure 3. A model considering polycrystalline Ag thin films ($p \approx 0$), and a bulk resistivity of $1.6 \mu\Omega \text{ cm}$, achieves a good agreement with our Ag stacks data. This result matches our expectations considering the growth mode of Ag thin film stack on glass. The trend expected for single crystal thin films ($p \approx 0.5$) is also shown for comparison. On the other hand, the single layer thin Ag films deposited directly on glass deviate from the polycrystalline prediction. The non-optimum growth of Ag on glass leads to rougher films and agglomeration in the lower thickness limit. The Fuchs-Sondheimer's model does not consider the surface roughness and grain boundaries contributions. However, in their studies Mayadas *et al.*^{15,16} extended Fuchs-Sondheimer theory to consider electronic scattering by grain boundaries. Furthermore, Rossmagel and Kuan¹⁷ revised this model and extended it to take into account surface roughness and grain size as

$$\rho = \rho_i \left[1 + \frac{0.375 (1-p) S l}{d} + \frac{1.5 R l}{(1-R) g} \right] \quad (2)$$

where S is a roughness parameter that equals 1 in perfect, atomically flat interfaces, and increases as roughness does so. R is the scattering coefficient, illustrating the scattering of electrons at the grain boundaries. Finally g is the average grain size. The grain sizes obtained with XRD and showed

in Figure 2 were used in the calculations. As shown in Figure 3, a better fit was obtained for the Ag films in stacks by considering $S=1$ and a scattering coefficient of $R=0.07$. For the case of the rougher Ag films deposited on Ag, an S value of 2 and R of 0.1 had to be used. These parameters clearly illustrate how thickness, grain size and surface roughness affect the films reflectivity.

IV. THICKNESS EFFECT IN THE OPTICAL PROPERTIES

Figure 4(a) shows the refractive index (n) and extinction coefficient (k) for Ag thin films deposited on glass with thickness ranging from 15.3 to 74.3 nm. A progressive reduction in n is observed as the Ag films get thicker, achieving values close to bulk for thicknesses around 74.3 nm.¹⁰ On the other hand, the extinction coefficient (k) was found to remain almost identical for all the thicknesses. The overall optical response of Ag films is strongly determined by two main components: intra-band transitions and the free electrons contribution. The UV and visible regions are dominated by inter-band transitions at the d electronic bands¹⁸ and agglomerated particle absorption. Such contribution can be described using a Lorentz oscillator given by the expression:

$$\epsilon_{Lorentz} = 1 + \frac{4\pi e^2}{m} \frac{N}{(\omega_0^2 - \omega^2) - j\Gamma\omega}. \quad (3)$$

where e and m are the charge and mass of the electron, N is the number of Lorentz oscillators, ω_0 is the resonance frequency and Γ is the damping parameter. On the other hand, the optical response is uniquely determined by a free electrons contribution in the infrared range.¹⁸ Such contribution can be modeled using the Drude model combined with an offset ϵ_∞ :

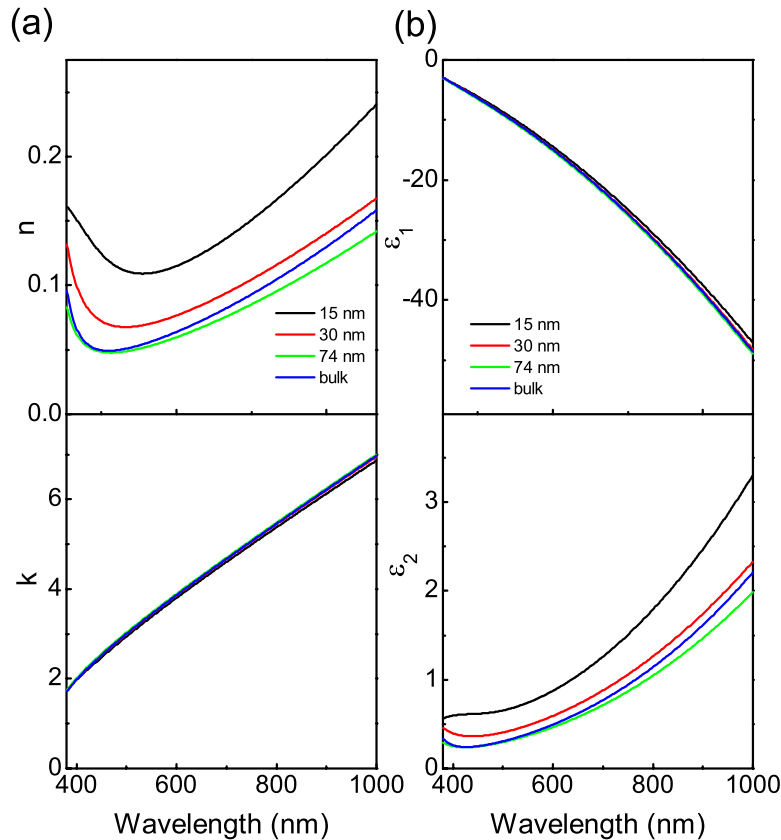


FIG. 4. (a) Refractive index (n) and extinction coefficient (k) and (b) real and imaginary parts of the dielectric function for Ag thin films deposited on glass.

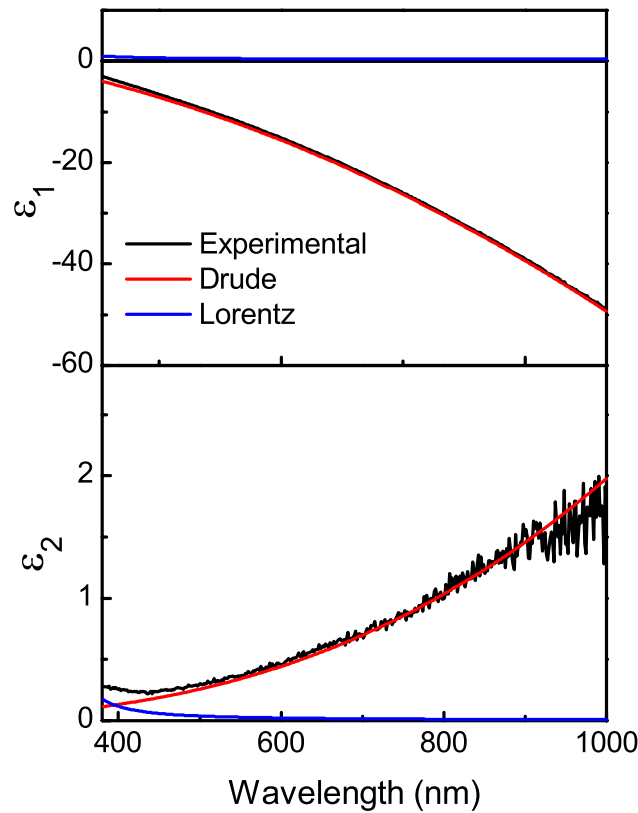


FIG. 5. Experimental real (ϵ_1) and imaginary (ϵ_2) part of the dielectric constant along with model fitting results considering the Drude and Lorentz contributions for a 74 nm silver film.

$$\epsilon_{Drude} = \epsilon_{\infty} - \frac{\omega_p^2}{\omega^2 - i\frac{\omega}{\tau}} = \epsilon_{\infty} - \frac{\omega_p^2 \tau^2}{1 + \omega^2 \tau^2} + i \frac{\omega_p^2 \tau^2}{\omega \tau (1 + \omega^2 \tau^2)} \quad (4)$$

where, as previously mentioned, ω_p is the plasma frequency and τ is the electron scattering time. The parameter ϵ_{∞} represents an offset of the real part of the dielectric constant. In Figure 5, a fitting for a selected 74.3 nm thick Ag films using equations (2) and (3) is presented considering both the Lorentz and Drude contributions. The Drude contribution clearly dominates for wavelengths higher than 600 nm, and thus will fully determine the response of Ag in the infrared. From this fitting, it is possible to extract the scattering times τ and plasma frequencies ω_p for the films.

Using this description of the optical constants, the electron scattering times τ for the Ag films deposited on glass and also in stacks were extracted as shown in Figure 6(a). Lower scattering times are found in the samples deposited directly on glass, which is consistent with their higher resistivity as shown in Figure 3. The more optimized growth of Ag in the stacks leads to lower scattering values, with a progressive decrease as the Ag films get thinner.

As previously discussed, it still remains unclear whether the optical Drude model can be used in systems with dimensions comparable or lower than the electronic free mean path. For the case of metal thin films, a progressive decrease of the electron scattering time τ is certainly observed when the electronic mean free path approaches the thickness of the film due to scattering with the interfaces, as shown in Figure 6(a). For the case of an infinite medium within the Drude framework, the relationship between electronic scattering time and resistivity is given by

$$\tau \rho = \frac{m_0}{n e^2} \quad (5)$$

where m_0 is mass of the electron and n is the carrier concentration. This product is constant in bulk materials as long as the carrier concentration is constant. Interestingly, for the Ag thin films

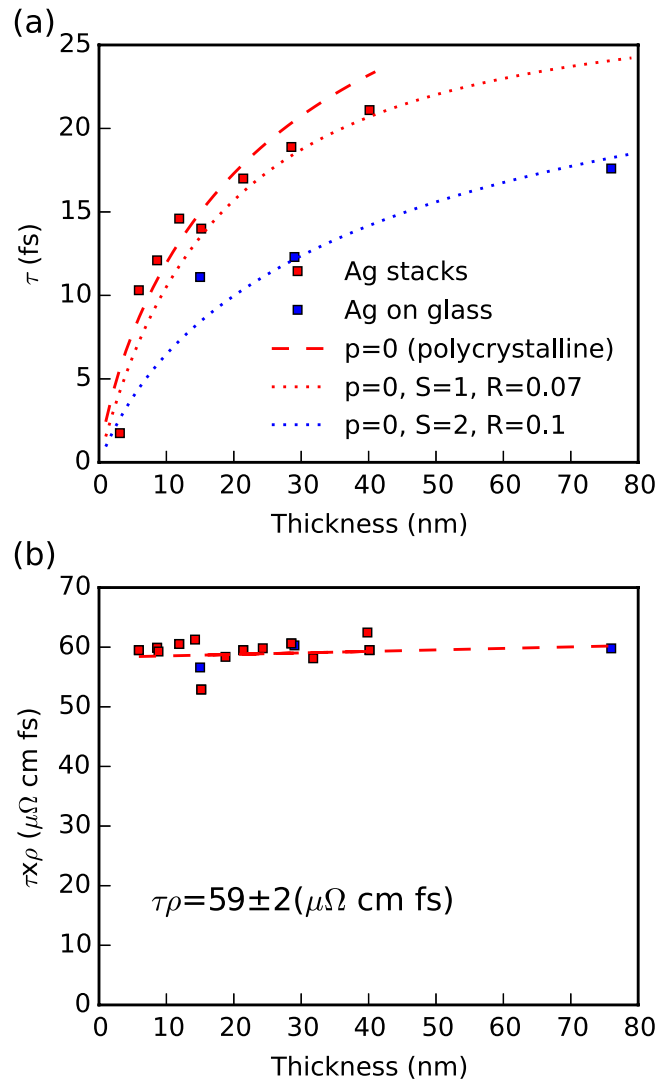


FIG. 6. (a) Electron scattering time τ vs. thickness for Ag thin films. (b) The product $\tau \times \rho$ remains constant in the thicknesses range studied.

investigated here, we observe that the product $\tau \times \rho$ also remains constant in the thicknesses range studied for both the Ag thin film deposited on glass and for the Ag stacks [Figure 6(b)]. This observation implies that the Ag thin films in both cases follow the same physical mechanisms, which is dominantly controlled by the free carrier scattering time and resistivity. Thus, we can conclude that the electron scattering time should follow the same dependence with thickness, grain size and roughness described for resistivity by Rosnagel and Kuan¹⁷ model [Eq.(2)], and therefore

$$\tau = \tau_0 \left[1 + \frac{0.375(1-p)Sl}{d} + \frac{1.5 Rl}{(1-R)g} \right]^{-1} \quad (6)$$

where τ_0 is the bulk electron scattering time. More importantly, the Drude model can still be applied to calculate the optical response of metal thin films provided that the electron scattering time τ is corrected with thickness and microstructure considering the effect of scattering with the interfaces and grain boundaries using the Rosnagel and Kuan¹⁷ model, as shown in Figure 6(a). Remarkably, the same parameters S and R used to describe the dependence of resistivity with surface roughness and inter-grain scattering in Figure 3 are able to fit very accurately the evolution of scattering time with thickness. This, once more, demonstrates the validity of the constant $\tau \times \rho$ relationship.

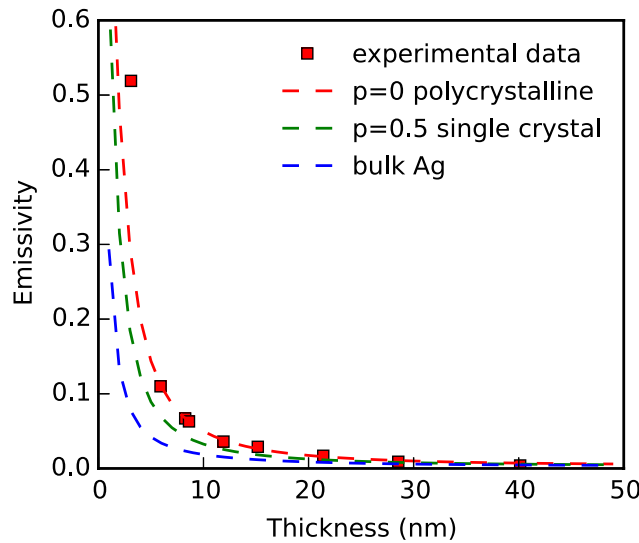


FIG. 7. Emissivity vs. thickness for Ag thin films. Calculations correspond to bulk Ag optical constants considering no dependence with thickness (blue line), considering the evolution of the electron collision time predicted for polycrystalline films $p=0$ (red line) and single crystal films $p=0.5$ (green line).

V. INFRARED OPTICAL RESPONSE: EMISSIVITY DEPENDENCE WITH THICKNESS

In order to validate our model, we analyze here a physical property mainly determined by the optical response of the thin films in the near-infrared and infrared range, where the free-electron contribution is the sole factor. The emissivity ϵ of the surface of a material is given by its effectiveness in emitting energy as thermal radiation. A blackbody has an emissivity of 1 while a perfect reflector would have a value of 0. Metal thin films exhibit extremely low emissivity values, as most of the incident infrared radiation is reflected by their free-electrons. Low-emissivity coatings have important application in energy efficient windows and thermal coatings. In particular, Ag is so far regarded as one of the highest performing materials for low-emissivity windows. Ag growth mode and quality strongly affects the optical response of the films and, for hence, their emissivity. Figure 7 shows experimental emissivity values for Ag thin films with thicknesses ranging from 3 to 40 nm. The emissivity values are obtained by integrating $\epsilon = 1 - R$ over wavelength λ weighted by the Planck emissive power at room temperature in the spectral range from 5 to 50 μm ,^{19,20}

$$\epsilon = \frac{\sum_{i=1}^m (1 - R_{\lambda_i}) E_{bi} \Delta\lambda_i}{\sum_{i=1}^m E_{bi} \Delta\lambda_i} \quad (7)$$

where R_{λ} is reflectivity at wavelength λ and $E_{b\lambda}$ is the radiation emitted by blackbody at wavelength λ .

A progressive increase in emissivity is observed as the films get thinner, meaning their ability of reflecting infrared radiation decreases. Emissivity was modeled here by calculating the spectral reflectivity of the system between 5 and 50 μm using the matrix transfer formalism²¹ and applying eq. (7). Considering bulk optical constants for Ag with no thickness dependence (dashed blue line in Figure 7) fails to fit the evolution observed. However, we are able to accurately reproduce the observed emissivity evolution considering a thickness dependent electron collision time τ as shown in eq. (6) and a p value of 0, which corresponds to polycrystalline films. Interestingly, the curve corresponding to single crystal films ($p=0.5$) describes the minimum achievable value for emissivity as a function of thickness in Ag thin films.

VI. RELATIONSHIP BETWEEN IR OPTICAL REFRACTIVE INDEX AND ELECTRICAL RESISTIVITY

As shown in Figure 5, Ag thin films accurately follow the Drude model in the infrared range considering a modified electron collision time. In addition, we have shown that electric conductivity

is closely related to the optical response of these films. In this section, we will develop a method to predict the infrared optical response of metal thin films in the near IR region based on their electrical resistivity and thickness. We also evaluate its validity in different scenarios.

Figure 4(b) shows the dielectric constants of Ag thin films deposited directly on glass. Upon close inspection, some trends are easily identified: (1) $|\varepsilon_1| \gg |\varepsilon_2|$ with $\varepsilon_1 < 0$, and (2) ε_1 and extinction coefficients k are almost identical for the three films. Taking into account the relationship between the optical constants n and k and the dielectric function $\varepsilon = \varepsilon_1 + i\varepsilon_2$ [Eq. (8) and (9)], the two first terms of the Taylor series of $\sqrt{1+x}$ and that $|\varepsilon_1| \gg |\varepsilon_2|$ and $\varepsilon_1 < 0$, we obtain the following relationships:

$$n = \sqrt{\frac{1}{2} [(\varepsilon_1^2 + \varepsilon_2^2)^{\frac{1}{2}} + \varepsilon_1]} = \sqrt{\frac{1}{2} |\varepsilon_1| \left\{ \left[1 + \left(\frac{\varepsilon_2}{\varepsilon_1} \right)^2 \right]^{\frac{1}{2}} - 1 \right\}} \approx \frac{1}{2} \left| \frac{\varepsilon_2}{\varepsilon_1} \right| \sqrt{|\varepsilon_1|} \quad (8)$$

$$k = \sqrt{\frac{1}{2} [(\varepsilon_1^2 + \varepsilon_2^2)^{\frac{1}{2}} - \varepsilon_1]} = \sqrt{\frac{1}{2} |\varepsilon_1| \left\{ \left[1 + \left(\frac{\varepsilon_2}{\varepsilon_1} \right)^2 \right]^{\frac{1}{2}} + 1 \right\}} \approx \sqrt{|\varepsilon_1|}, \quad (9)$$

Including the Drude dielectric function from eq. (4), and considering that $\tau^2 \omega^2 \gg 1$ for wavelengths $< 5 \mu\text{m}$, and the relationship between resistivity, plasma frequency and electron scattering time $\omega_p^2 = 1/\varepsilon_0 \rho \tau$, we obtain:

$$n \approx \frac{1}{2} \frac{\omega_p^2}{\omega^3 \tau \sqrt{\frac{\omega_p^2}{\omega^2} - \varepsilon_\infty}} \approx \frac{1}{2 \varepsilon_0 \rho \tau^2 \omega^3 \sqrt{\frac{1}{\varepsilon_0 \rho \tau \omega^2} - \varepsilon_\infty}} \quad (10)$$

$$k \approx \sqrt{\frac{\omega_p^2 \tau^2}{1 + \omega^2 \tau^2} - \varepsilon_\infty} \approx \sqrt{\frac{1}{\varepsilon_0 \rho \tau \omega^2} - \varepsilon_\infty} \quad (11)$$

where ε_0 is the vacuum dielectric constant. We can thus describe the spectral optical constants n and k as a function of resistivity ρ ($\Omega\text{-cm}$) and electron scattering time τ (sec). In addition, since we have shown that the product $\tau \times \rho$ is constant in the investigated thicknesses range, equations (10) and (11) can be simplified as

$$n \approx \frac{\rho}{2 \varepsilon_0 (\rho \tau)^2 \omega^3 k} = \frac{\rho}{2 \varepsilon_0 C^2 \omega^3 k} \quad (12)$$

$$k \approx \sqrt{\frac{1}{\varepsilon_0 C \omega^2} - \varepsilon_\infty} \quad (13)$$

where C is the product $\rho \times \tau$, with the value $59 \pm 2 \mu\Omega \text{ cm fs}$, independent of the wavelength. This relationship between n and k spectra and resistivity ρ agrees with the experimental observations shown in Figure 4. There exists a linear relationship between n and ρ , and also the coefficient k is independent of film thickness and resistivity, as shown in Figure 4.

In what follows, we will evaluate the validity of Eq. (11) and (12) for Ag thin films with different thicknesses by comparing experimental data with calculations. Considering that, as shown in Eq. (12), k shows no dependence with thickness and thus it can be estimated from bulk values, we focus our attention on n . Figure 8 shows the spectral dependence of n for Ag thin films with thicknesses of (a) 29.5 nm and (b) 74.3 nm. The measured resistivity for these films was $4.9 \mu\Omega\text{cm}$ and $3.4 \mu\Omega\text{cm}$ respectively. We carried out calculations using Eq. (11) and considering a C constant as calculated experimentally $59 \pm 2 \mu\Omega \text{ cm fs}$ as shown in Figure 8. Since an error bar of $\pm 2 \mu\Omega \text{ cm fs}$ was estimated ($\pm 3\%$), calculations were performed using $59 \mu\Omega \text{ cm fs}$, and also the upper and lower limits of 61 and 57 $\mu\Omega \text{ cm fs}$, respectively, in order to evaluate the confidence range. We find a very good agreement between experimental and theoretically calculated n in the IR range ($\lambda > 800 \text{ nm}$). However, as previously discussed, this model cannot reproduce the response in the blue and UV ranges since interband transitions dominate the optical response.

In summary, the near IR optical response of Ag thin films can be estimated by using Eq. (11) and (12), the electrical resistivity of the films, their thickness and the experimentally measured

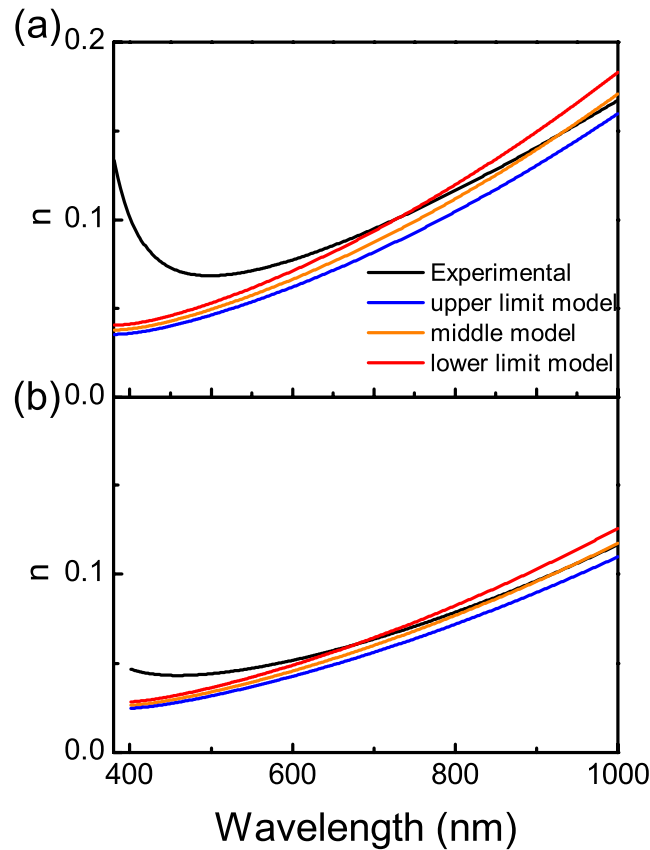


FIG. 8. Experimental refractive index (n) spectra for Ag films (a) 29 nm and (b) 74 nm thick along with calculations using three values of the constant C , i.e. $59 \mu\Omega \text{ cm fs}$ as middle point, and 57 and $61 \mu\Omega \text{ cm fs}$ as lower and upper limits respectively.

constant C . On the other hand, k remains unaltered with thickness and the bulk value can be used. This theoretical method is valid based when the following conditions are met: (1) $\tau \times \rho = \text{constant } C$, (2) $|\epsilon_1| \gg |\epsilon_2|$, and (3) $\tau^2 \omega^2 \gg 1$. As shown, Ag is a clear example of metal satisfying all these conditions in the IR region (wavelengths between 700nm and $5\mu\text{m}$.)

VII. SUMMARY

The optical and electrical response of silver thin films approaching thicknesses in the range of the electron mean free path were studied theoretically and experimentally. A correlation on how thickness and microstructure affect the properties of Ag thin films is presented. We also show how the optical refractive index of silver thin films in near IR range can be theoretically estimated considering their thickness and resistivity, provided that the three following conditions are satisfied: (1) $\tau \times \rho = \text{constant } C$, (2) $|\epsilon_1| \gg |\epsilon_2|$, and (3) $\tau^2 \omega^2 \gg 1$. This study also shows the theoretical lower limit for emissivity in Ag thin films according to their microstructure and thickness. Application of the model presented here will allow rapid characterization of the optical response of metal thin films with application in a broad spectrum of fundamental and industrial applications, including optical coatings, low-emissivity windows and semiconductor industry.

ACKNOWLEDGMENTS

The authors would like to thank Yu Wang and Zhiwen Sun for valuable discussions, Vince Nguyen for experimental measurements, and Ed Korczynski for advice on the manuscript.

Intermolecular is a registered trademark, and Tempus and HPC are trademarks of Intermolecular, Inc., all rights reserved.

- ¹ M. Fox, *Optical Properties of Solids* (Oxford University Press, 2001).
- ² D. E. Aspnes, in *Handbook of Optical Constants of Solids*, edited by E. D. Palik (Academic Press, Burlington, 1997), pp. 89-112.
- ³ L. Azaroff, *Introduction to Solids* (TMH Edition, Tata McGraw-Hill Publishing Company Limited).
- ⁴ P. Drude, *Ann. Phys.* **306**(3), 566-613 (1900).
- ⁵ E. H. Sondheimer, *Adv. Phys.* **1**(1), 1-42 (1952).
- ⁶ P. B. Johnson and R. W. Christy, *Phys. Rev. B* **6**(12), 4370-4379 (1972).
- ⁷ U. Kreibig and M. Vollmer, *Optical properties of metal clusters* (Springer-Verlag, Berlin, 1995).
- ⁸ C. Clavero, A. Cebollada, G. Armelles, Y. Huttel, J. Arbiol, F. Peiro, and A. Cornet, *Phys. Rev. B* **72**(2), 024441 (2005).
- ⁹ C. Clavero, Ph.D. Thesis, Universidad Autónoma de Madrid, 2007.
- ¹⁰ P. Grosse, R. Hertling, and T. Muggenburg, *J. Non-Cryst. Solids* **218**(0), 38-43 (1997).
- ¹¹ Y. Tsuda, H. Omoto, K. Tanaka, and H. Ohsaki, *Thin Solid Films* **502**(1-2), 223-227 (2006).
- ¹² K. Kato, H. Omoto, T. Tomioka, and A. Takamatsu, *Sol. Energy Mater. Sol. Cells* **95**(8), 2352-2356 (2011).
- ¹³ O. Treichel, V. Kirchhoff, and G. Bräuer, in *43rd Annual Technical Conference 2000* (Society of Vacuum Coaters -SVC-, Albuquerque/NM, Denver, Colorado, 2000), Vol. Proceedings : April 15-20, 2000, pp. 121-126.
- ¹⁴ H. J. Glaser, *Large Area Glass Coating* (Von Ardenne Anglagenteknik GmbH, Dresden, Germany, 2000).
- ¹⁵ A. F. Mayadas, M. Shatzkes, and J. F. Janak, *Appl. Phys. Lett.* **14**(11), 345-347 (1969).
- ¹⁶ A. F. Mayadas and M. Shatzkes, *Phys. Rev. B* **1**(4), 1382-1389 (1970).
- ¹⁷ S. M. Rossnagel and T. S. Kuan, *Journal of Vacuum Science & Technology B* **22**(1), 240-247 (2004).
- ¹⁸ G. Fuster, J. M. Tyler, N. E. Brener, J. Callaway, and D. Bagayoko, *Phys. Rev. B* **42**(12), 7322-7329 (1990).
- ¹⁹ M. Rubin, D. Arasteh, and J. Hartmann, *Int. Commun. Heat Mass Transfer* **14**(5), 561-565 (1987).
- ²⁰ NFRC 301-2014 Standard Test Method for Emittance of Specular Surfaces Using Spectrometric Measurements, 2013.
- ²¹ M. Schubert, *Phys. Rev. B* **53**(8), 4265 (1996).