Laser-induced dewetting of silver-doped chalcogenide glasses

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ABSTRACT

We report the observation of laser-induced dewetting responsible for the formation of periodic relief structures in silver-based chalcogenide thin-films. By varying the concentration of silver in the Ag\textsubscript{x}(As\textsubscript{20}S\textsubscript{80})\textsubscript{100-x} system (with x = 0, 4, 9 and 36), different surface relief structures are formed. The evolution of the surface changes as a function of laser parameters (power density, duration of exposure, and polarisation) as well as film thickness and silver concentration has been investigated. The scanning electron microscopy and atomic force microscopy images of irradiated spots show periodic ripples aligned perpendicularly to the electric field of incident light. Our results show that addition of silver into sulphur-rich chalcogenide thin-films improves the dewetting when compared to pure As\textsubscript{20}S\textsubscript{80} thin-films. The changes in surface morphology were attributable to photo-induced chemical modifications and a laser-driven molecular rearrangement.

1. Introduction

The formation of periodic patterns is a fundamental and technologically significant topic with great interest over the years. This process can be achieved using several methods such as lithography or focused ion beam (FIB). Despite their effectiveness, these techniques are either time consuming and costly (i.e., FIB) or need the use of other materials and extra steps during the process such as lithography, which can be inconvenient taking into account the compatibility of the different components.

To supersede these drawbacks, an alternative technique to produce relief periodic patterns would be the use of the dewetting phenomenon in thin-films. In fact, the reduction of the total free energy when the interfacial area between the film and its substrate is reduced by agglomeration of the film into islands [1] is accomplished using a thermal source or via photo-induced effects using a laser. In a previous study, we reported the use of thermal dewetting on chalcogenide thin-films to fabricate microlenses transparent in the infrared region [2].

While the pattern formation via thermal dewetting of metal and polymer thin-films has been studied in detail, few works have been reported in the literature on laser-induced dewetting. For instance, dewetted structures following laser irradiation have been used to produce nano and micro-sized particles of noble metals [3–5] for plasmonic applications. In the case of Ag thin-films the dewetting is explained by capillary instabilities due to the high temperature reached by a pulsed laser [6].

In this paper we report the formation of laser-induced surface relief structures of glassy Ag-based chalcogenide thin-films using a continuous wave (CW) laser which are created as a result of dewetting. In contrast to the literature, we demonstrate here that Ag–As–S thin-films can be completely dewetted by a laser beam via photonic effects rather than thermal effects. Silver migration and mass transport due to an electric field gradient force was suggested to be at the origin of the observed phenomenon. In fact, the real mechanism is mainly dependant on the glass composition and conditions of processing. In this study, we will show the possibilities of tuning the period and shape of the structures by varying power density of the incident light, and the silver concentration and thickness of the Ag–As–S thin-films.

2. Materials and methods

The As\textsubscript{20}S\textsubscript{80} glass was prepared by the usual melt-quenching technique, from high purity arsenic (7 N) and sulphur (5 N), sealed in an evacuated silica tube, which was then heated at 675 °C for 12 h and air-quenched. The glass was annealed at 70 °C for 8 h to prevent internal stresses.
Thin-films of the ternary system $\text{Ag}_x(\text{As}_{20}\text{S}_{80})_{100-x}$ ($x = 0, 4, 9,$ and $36$) (respectively denoted $\text{Ag}_{00}, \text{Ag}_4, \text{Ag}_9$ and $\text{Ag}_{36}$) were deposited on a glass substrate (microscope slides) by the co-evaporation technique, from pure Ag (4 N) and $\text{As}_{20}\text{S}_{80}$ glass. A Nanochrome device (IntiVac), equipped with one current induced heated source and an electron beam evaporator was used. The current induced heated source was used to evaporate Ag, whereas the electron beam was used to evaporate the $\text{As}_{20}\text{S}_{80}$ glass. Before the deposition, the chamber was evacuated down to approximately $10^{-6}$ Torr. The thickness of the thin-films varies from 110 nm to 2.7 µm. During the co-evaporation, the substrate holder is rotated at 80 rpm providing a highly homogeneous and uniform thin-film deposited on the substrate, thus, preventing initial thickness or composition inhomogeneities.

We performed the irradiations using a linearly polarised CW argon ion laser (operating at 488 nm) whose power ranges from 0 to 900 mW. The diameter of the beam can be modulated by varying the mechanical aperture of the laser. In our case, we used an aperture of 2.26 mm, giving us a power density ranging from 0 to 22.44 W cm$^{-2}$. For our experiments we irradiated the samples for a duration varying between 5 and 90 min. To study the laser-induced changes of chalcogenide thin-films, the samples were placed in the vertical position (90°) and the laser beam is directed perpendicularly to the samples (normal incidence from the chalcogenide thin-film side). The laser beam profile was characterized by a beam shape profile analyser (Thorlabs’ Dual Scanning Slit Beam Profillers BP209-VIS/M) to ensure the beam was Gaussian.

A Scanning Electron Microscope (SEM) (FEI, model Quanta 3D FEG) coupled to Energy Dispersive X-ray spectroscopy (EDX) was used to analyse the surface morphology and to provide quantitative compositional information, respectively. Both SEM images and EDX measurements were performed under low vacuum (with $\text{H}_2\text{O}$ vapour). EDX scans were executed with a voltage of 30 kV. The corresponding quantitative EDX analysis result is presented in Table 1.

The topographic features such as characteristic length, width and height of the dewetted structures were measured with the help of Atomic Force Microscopy (AFM, diDimension V SPM model from Veeco) in tapping mode.

3. Results and discussion

3.1. Laser-induced dewetting evolution with the silver concentration

Fig. 1 presents the scanning electron microscopic (SEM) images of the irradiated area of the $\text{Ag}_x(\text{As}_{20}\text{S}_{80})_{100-x}$ ($x = 0$–36 at.% Ag) thin-films at 4.64 W cm$^{-2}$ during 60 min. As can be seen, three distinct types of surface features are formed depending on the silver concentration. The first type of laser structural change is observed in the Ag$_0$ sample, that is the As$_{20}$S$_{80}$ matrix without silver. For this concentration, the irradiation induced the formation of small crystals on the surface which can be attributed to As$_2$O$_3$. This is a well-known phenomenon for As-S chalcogenide glasses, and many authors have reported the photocristallisation of As$_2$S$_3$ resulting in As$_2$O$_3$ crystals [7,8].

The second type of the surface modification is formed on the samples with a silver concentration in the range of 4% < Ag% < 19%. In this interval of concentration, relief periodic structures are formed. For a silver content of 4%, the formation of the relief structures is not total and we can observe two distinct phases (the ripples-like features and the remaining matter around them), as presented on Fig. 1b. With an increase in silver concentration up to 19% we observed the amplitude of the surface relief increases; and finally, the relief height grows forming totally dewetted patterns, as illustrated in Fig. 1c. Here the term “dewetting” refers to the process by which a fluid moves to uncover a substrate. The film is still in solid state and the dewetting process occurs via diffusion rather than fluid convection. The periodicity of the structures is approximately 10 µm and the entire laser modified region is approximately 2 mm in diameter. Interestingly, it was found that the direction of the ripples depends upon the direction of laser polarisation.

The third type of surface change that occurs under laser irradiation is observed for concentrations of silver greater than 36%. At high concentrations of silver, we observed a rough texture due to photocristallisation of the thin-film (Fig. 1d). The photo-induced crystallisation process in chalcogenide glasses occurs at temperatures lower than the thermal crystallisation temperature [9]. The presence of crystallites of acanthite (Ag$_2$S) has been verified by the X-ray diffraction technique.

The laser-induced periodic surface structures – as seen in the $\text{Ag}_x(\text{As}_{20}\text{S}_{80})_{100-x}$ sample exposed at 4.64 W cm$^{-2}$ – were also reported on chalcogenide materials irradiated by both femtosecond [10] and CW-laser [11]. The observed ripples on the surface of thin-films have been assigned to two different processes. In the case of femtosecond irradiation, the period of the observed ripples is close to the wavelength of the incident radiation and the mechanism relies on the optical interference between the incident laser and a scattered surface optical wave [12,13]. In the case of CW irradiation [11] much experimental and theoretical work has been carried out to explain the mechanism of this phenomenon. Models have been proposed to describe the formation of surface relief, where mass transport [14], photofluidity [15], and interaction amongst dipoles [16] were considered as the driving forces for deformation. However, a vast majority of studies on the laser induced periodic surface structures have been devoted to stoichiometric As$_2$S$_3$ [17] or Se-rich arsenic glasses [18] because their properties are well-known.

As shown in our previous work, addition of Ag in the As$_{20}$S$_{80}$ matrix changes the As-S glass backbone structure itself leading to a phase separation [2] and it is well-known that in silver-doped chalcogenide films, silver migration is induced toward the irradiated region under light exposure. We should take into account, for all experimental observations, the competing effects that arise due to the silver addition.

Given the results presented in Fig. 1, it is clear that the surface relief formation is more pronounced in the Ag$_0$ thin-film. Thus, we systematically investigated the laser-induced periodic surface structures for this concentration of silver as a function of power density, polarisation and thickness.
3.2. Laser-induced surface structures dependence on laser power density

We also investigated the relief formation at different laser power density ranging from 1.7 to 15Wcm\(^{-2}\) as shown in the inset of Fig. 2. We found that the structures’ height increased with the power density, whereas the ripples’ period remains constant.

As it was reported in our previous study [2], dewetting occurs when silver-doped chalcogenide thin-films are heated and it is due to phase separation of the film. The formation of organised structures obtained by thermal dewetting was observed within a processing window defined by a temperature ranging between 200 °C and 280 °C. We have employed thermal dewetting in the fabrication of transparent IR chalcogenide micro-lenses. On that basis, it becomes important to evaluate the role of the temperature due to the local heating induced by laser irradiation. An estimated value can be obtained by the following approximate relation [19]:

\[
\Delta T = \frac{P}{k w \sqrt{2\pi}}
\]

where \(\Delta T\) is the temperature variation, \(P\) is the laser output power, \(w\) is the beam diameter, and \(k\) is the thermal conductivity of the glass.

The surface temperature of the sample was monitored during the exposure using an infrared camera (Telops Hyper-Cam TEL-3306) with a spatial resolution of 640 x 512 pixels and acquisition delay of 1 s between each measurement (Fig. 2).

We observed that the temperature of the sample increased for higher power density exposures. For example, at a power density of 15 W cm\(^{-2}\), a temperature of 95 °C is reached after 10 s, but the laser-induced dewetted experiment was performed for a power density of 4.64 W cm\(^{-2}\), which corresponds to a measured temperature below 60 °C. Bearing in mind that in similar As–S–Ag glass compositions, the glass transition temperature is around 150 °C and the crystallisation temperature around 240 °C [20], a rise to 60 °C is below the glass transition temperature of the thin-film and is not sufficient enough to induce any significant structural changes in the glass through thermal processes. Hence, the observed photoinduced changes cannot be accounted for, based solely on sample heating. Moreover, it is known that chalcogenide glasses typically have low thermal conductivity and diffusivity. These properties slow the rate at which heat is diffused in the material and at which it can be dissipated. Thus, during long irradiation times, one would expect some heat accumulation which could result in glass softening. However, additional measurements showed that even for longer irradiations (not presented here) the temperature stays constant and decays suddenly when the laser beam is turned off.

The formation of ripples was actually observed within a processing window defined by a laser power density ranging between 3.5 and 7.5 W cm\(^{-2}\). Above 10 W cm\(^{-2}\) the periodic structures begin to crystallise. The insets in Fig. 2 show the evolution of the surface morphology as a function of the power density. With only 1.7Wcm\(^{-2}\) (Fig. 2a), a few ripples can be observed inside the irradiated spot. For 3.5Wcm\(^{-2}\) (Fig. 2b) a series of periodic ripples that broaden as the power density increases appear on the entire laser spot (laser beam spot is about 2 mm in diameter). It was previously reported that ripples in silver-free chalcogenide thin-films under CW laser illumination can also be observed [11,14,15] and the mechanism of their formation was explained by mass transportation induced by light. Nevertheless, the mass transfer detected on As\(_2\)S\(_3\) glasses allows the extraction, upon exposure to intense light, of some arsenic atoms from the As\(_2\)S\(_3\) network. The remaining sulphur atoms interact strongly with Ag and form a binary compound: Ag\(_2\)S.

3.3. Elemental analysis

Within the appropriate laser conditions described above, laser-dewetted periodic ripples are observed on the surface of the Ag–As–S thin-film samples. We analysed the change of elemental compositions for different points across the surface using energy dispersive X-ray spectroscopy (EDX). Fig. 3a shows the SEM micrograph of the irradiated spot (4.4Wcm\(^{-2}\)). Fig. 3b presents the laser-induced variation of silver concentration as a function of the position in the irradiated spot.

As can be noted, the silver concentration in the irradiated region rises up to 3%. Additional measurements (not presented here) reveal that the silver concentration increases with increasing exposure time and power density. It is known that Ag\(^{+}\) ions in AgAs\(_2\)S\(_2\) glasses are likely to move toward illuminated regions, and the photo-induced silver migration process is assumed to proceed as follows: When a silver-doped chalcogenide glass is exposed to near band gap light, electron-hole pairs are created, and the holes diffuse toward unilluminated regions, since holes are more mobile than Ag\(^{+}\) ions. The illuminated region then becomes negatively charged, and in response, Ag\(^{+}\) ions in the unilluminated region migrate toward the illuminated region. Tanaka et al. report that this migration gives an Ag concentration increase of 5 at.% in AgAs\(_2\)S\(_2\) [21] which is a similar amount to the present result (3%) as seen in Fig. 3b. It seems possible to conclude, therefore, that the Ag atoms come from the unilluminated area around the laser beam irradiation spot. This fact can be corroborated by the decrease of silver amount (0.5% at.) at the peripheral region around the laser spot shown in Fig. 3b. This EDX quantification can be accounted for as a manifestation of the Ag depletion.

According to Dwivedi et al. [22–24], since there are a large number of dangling bonds in chalcogenide glasses, one would eventually expect a new equilibrium to occur after migration [25], in which dipoles of Ag\(^{+}\) coupled to dangling bonds would form, causing a rearrangement of atomic structures. Popescu et al. [26] indicates that the introduction of Ag into As\(_2\)S\(_3\) glasses allows the extraction, upon exposure to intense light, of some arsenic atoms from the As\(_2\)S\(_3\) network. The remaining sulphur atoms interact strongly with Ag and form a binary compound: Ag\(_2\)S.

At present, we suggest that laser-dewetted periodic ripples arise from two well-known processes of the laser-induced phenomena in chalcogenide glasses: photo-induced chemical modifications and photo-induced mass transport.
3.4. Effects of laser polarisation

The dependence of the incident polarisation of the laser on Agx thin-films (1.35 lm) was performed by using the s-polarised beam of the CW argon ion laser. Its polarisation was oriented by means of a half-wave plate (\(\lambda/2\)) to obtain a linear polarised illumination and a quarter wave plate (\(\lambda/4\)) to obtain a circular polarisation. Samples were exposed during 60 min at 4.64 W cm\(^{-2}\).

Variations in the polarisation of the incident illumination altered the direction of ripples that developed during laser dewetting of the As–S–Ag thin-films as it is showed in Fig. 4(a–c). Under linearly perpendicular polarised illumination, the ripples appear to be aligned perpendicularly to the electric field vector of the incident illumination (Fig. 4a). When the polarisation is changed for a linear parallel one, ripples are developed with an orientation still perpendicular to the direction of the electric field (Fig. 4b). Circular polarisation produced spiral structures. These results show that the appearance of As\(_{20}\)S\(_{80}\) thin-films as it is showed in Fig. 4(a–c). Under linearly perpendicular polarised illumination, the ripples appear to be aligned perpendicularly to the electric field vector of the incident illumination (Fig. 4a). When the polarisation is changed for a linear parallel one, ripples are developed with an orientation still perpendicular to the direction of the electric field (Fig. 4b). Circular polarisation produced spiral structures. These results show that the direction of the ripples depends on the direction of light polarisation and confirm that the laser dewetting was not induced by thermal effects caused by light absorption. Similar results were observed on As\(_{20}\)S\(_{80}\) thin-films [27]. The mechanism of such process is still unclear and other studies are in progress.

3.5. Effects of the thickness

Experiments are conducted on four different thicknesses for the Agx sample: 0.26, 0.60, 1.35 and 2.70 µm. The primary objective is to make a correlation between the features’ sizes and the initial thickness of the thin-film. Thus, AFM measurements were carried out to quantify the geometrical characteristic profile parameters such as height (h), space between the structures (\(\lambda\)), and width (\(\omega\)), see Fig. 5 (left). The results were then correlated to the starting thicknesses of the thin-films with linear regressions presented in Fig. 5 (right).

AFM studies show that increasing the thickness from 0.26 µm to 2.7 µm, the space between the structures can increase up to seven times with an homogeneous distribution. Our investigation reveals that the directly proportional features of dewetting to the thickness can be assigned to a novel mix between chalcogenide and silver. Also, the interaction with laser light makes the dewetting a more complex process. Based on the dynamic response of film parameters it is possible to either predict the size of the structures from the starting thickness or choose the thickness depending on the desired size for the dewetted features.

In order to explain the above results, two well-known processes of the laser-induced phenomena in silver-based chalcogenide glasses are used. The first mechanism is the scalar photo-induced chemical modifications (PCM) observed in silver-based chalcogenide glasses. It is widely admitted that silver-doped chalcogenide thin-films induce the silver migration toward the irradiated region under light exposure [28,29]. As previously mentioned, Tanaka et al. [30] explain the PCM as follow: when an Ag– As–S system is illuminated with light of energy near the band gap, the lone pair of electrons is excited forming electrons and holes. The holes diffuse toward the unilluminated region making it positively charged. The Ag\(^{+}\) ions contraflow the hole current [31] to maintain charge neutrality and a variation of the Ag content with light illumination arise [21]. Fig. 6a–d, which illustrates this process, is adapted from Jain’s schematics [32].

The second process is connected to the increase of Ag due to photoinduced migration. When a high quantity of silver migrates to the exposed area, the relatively large immiscibility between the two limiting materials, As–S and Ag, may increase segregation of the silver and lead to phase separation [33]. In our previous work [2], we assume that these separated phases (Ag-rich and Ag-poor) have a different surface tension from the original thin-films which will induce a solid dewetting as observed in Fig. 4.

These laser-dewetted periodic ripples only appear in the unstable Ag\(_{x}\)(As\(_{20}\)S\(_{80}\))\(_{100-x}\) system (\(x > 0\)). There is not any structure formed for the As\(_{20}\)S\(_{80}\) thin-films, which implies that Ag incorporation plays an important role in pattern formation. However, the above mechanism, of silver migration and phase separation are scalar effects that are isotropic and independent of the laser beam polarisation and could not explain the polarisation dependence in Fig. 4 and the periodic relief formation.

Recently, some publications have been dedicated to studies laser-induced relief structure which are polarisation dependent in chalcogenide materials, in particular for holographic recording [34]. Several models have been proposed [14,35–37] to explain the formation of such structures, where interaction among dipoles, optical gradient forces, and photoinduced dielectrophoresis were considered as driven forces for such surface deformation. Salaminia et al. [14] detected a formation of a polarised relief photomodulation of As\(_2\)S\(_3\) films, namely, light induced mass transport. They suggested that the mechanism of mass transport lies on an electric field gradient of the writing laser along the grating vector and a force on dipoles (dipolar defects leading to mass transport due to their interaction and/or rearrangement [38]).

In our recent work [39], it was demonstrated that due to the incorporation of Ag in the As–S thin-film, a structural rearrangement favouring the appearance of AgS\(_{12}\) pyramids and As\(_8\)S\(_{18}\) molecules are formed. In order to explain the polarisation dependence of the laser-dewetted surface relief structure, we can assume that at the early stage of irradiation by a linearly polarised laser, structural As\(_8\)S\(_{18}\) cages units are reoriented and serve as anisotropic sensitive structural units. Indeed, As\(_8\)S\(_{18}\) units have a cage-like structure, which includes two planar homopolar As-As bonds that are heavily polarised. It is supposed that such As\(_8\)S\(_{18}\) cage molecules whose major axis is parallel to the electric field vector of the incident light after excitation and subsequent relaxation became orthogonal to the polarisation vector [40].

Such laser-driven molecular rearrangement – or mass transport – takes place and follows the direction perpendicular to the polarisation of the incident light, leading to the creation of stable surface relief formation inside the irradiated spot. These steps are schematically illustrated in Fig. 6.

A fingerprint of this phenomenon is the conservation of volume [41]: peaks and valleys appearing on the sample surface preserve the irradiated spot volume on the mesoscopic length scale. The complete laser-induced dewetting is achieved through the surface tension different from the original thin-films due to silver migration and subsequently phase separation.
A detailed microscopic mechanism of the observed phenomenon is under study and in situ birefringence and Raman scattering measurements are currently being performed, and will be reported on a subsequent publication.

4. Conclusion

Ag\(_x\)(As\(_{20}\)S\(_{80}\))\(_{100-x}\) (\(x = 0–36 \text{ at.}\% \text{Ag}\)) chalcogenide thin-films were prepared by the co-evaporation technique. This method enlarges the vitreous domain, allowing a wide range of new metastable compositions. Depending on the silver concentration, periodic relief structures have been formed on the surfaces of silver-doped chalcogenide thin-films when exposed to a 488 nm CW argon laser. Those features were the result of the laser-induced dewetting phenomenon, which can be tuned by adjusting the laser parameters (power density, exposure time) and the initial thickness of the thin-films.

Our analyses indicate that different mechanisms contribute to the formation of laser-dewetted periodic ripples. Initially a photo-induced migration of silver from an unexposed area to an illuminated one leads to a phase separation. Differences between surface tension in silver-poor and silver-rich regions lead to a complete dewetting of the film into periodic microstructures.

A laser-induced mass migration process may be generated, leading to the formation of relief patterns on the silver-based chalcogenide thin-films. It was found that the direction of the laser-dewetted structures depends on the direction of light polarisation.

Due to their predictable sizes and shapes and their chalcogenide nature, the surface relief features could potentially be used in holographic memories, infrared waveguides, and diffraction gratings.

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References


Table 1
Evaporation rates used for the deposition of the thin-films and their corresponding compositions.

<table>
<thead>
<tr>
<th>Codename</th>
<th>Evaporation rate (Å s⁻¹)</th>
<th>Composition (At%)</th>
<th>Chemical formula</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ag</td>
<td>As₃₀Sr₀</td>
<td>Ag</td>
</tr>
<tr>
<td>A₈₀</td>
<td>0</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>A₈₁</td>
<td>0.5</td>
<td>10</td>
<td>3.5</td>
</tr>
<tr>
<td>A₈₂</td>
<td>1</td>
<td>10</td>
<td>9.2</td>
</tr>
<tr>
<td>A₈₅₀</td>
<td>4</td>
<td>10</td>
<td>36.1</td>
</tr>
</tbody>
</table>
Fig. 1. SEM images of the thin-film surfaces after laser irradiation at 4.64 W cm$^{-2}$ for 60 min. (a) $\text{As}_2\text{S}_8$. (b) $\text{Ag}_4(\text{As}_2\text{S}_8)_9$. (c) $\text{Ag}_9(\text{As}_2\text{S}_8)_3$. (d) $\text{Ag}_{36}(\text{As}_2\text{S}_8)_1$. The arrowed E indicates the laser polarisation direction.
Fig. 2. Measured temperature dependence of the Ag thin-film surface for different laser power densities. (a) Initial stage of dewetting for power densities below 1.7 W cm$^{-2}$ (zone a). (b) A series of ripples with periodic distribution not totally dewetted from zone b. (c) Totally dewetted structures form in zone c. (d) Crystallisation of the dewetted features occurs due to high laser power densities in zone d. A fixed exposure time of 20 min was used in each case.
Fig. 3. (a) SEM micrograph of the exposed region. The diameter of the affected area is around 2 mm. (b) EDX analysis of the thin-film composition across the surface after irradiation: the solid black squares represent the measured data, and the open squares are the extrapolated data.
Fig. 4. SEM images of Ag$_x$ exposed to the same laser power density, 4.64 W cm$^{-2}$, for different polarisations: (a) vertical, (b) horizontal and (c) circular. The arrows indicate the direction of the laser polarisation.
Fig. 5. (Left) AFM 3D scan of Ag (263 nm) showing the three characteristic lengths we measured: (1) spacing between structures (\( \lambda \)), (2) width of the features (\( \omega \)), and (3) height of the structures (\( h \)). (Right) \( \lambda \), \( \omega \), and \( h \) as a function of the thickness, and the linear regression equations corresponding.
Fig. 6. Schematic representation of the laser-induced structural changes in Ag$_{y}$(A$\delta_{2}$S$_{90}$)$_{100-y}$ thin films, where the Ag$^{+}$ circles, the open white circles and the solid black circles represent Ag$^{+}$ ions, holes, and electrons, respectively. (a) thin-film before exposure, (b) creation of electron-hole pairs in the laser-illuminated volume, (c) migration of holes from the illuminated to the dark area, (d) diffusion of silver to the illuminated region, (e) illuminated region where mass transport and phase separation lead to dewetting of the thin-film, and (f) complete dewetting of the illuminated area.