Optical evidence for a self-propagating molten buried layer in germanium films upon nanosecond laser irradiation

F. Vega, N. Chaoui, J. Solis, J. Armengol, and C. N. Afonso

Citation: *J. Appl. Phys.* **97**, 103519 (2005); doi: 10.1063/1.1900932

View online: <http://dx.doi.org/10.1063/1.1900932>

View Table of Contents: <http://jap.aip.org/resource/1/JAPIAU/v97/i10>

Published by the [American Institute of Physics](#).

Additional information on J. Appl. Phys.

Journal Homepage: <http://jap.aip.org/>

Journal Information: http://jap.aip.org/about/about_the_journal

Top downloads: http://jap.aip.org/features/most_downloaded

Information for Authors: <http://jap.aip.org/authors>

ADVERTISEMENT

The advertisement banner for AIP Advances features a green and yellow background with abstract wavy lines. The AIP Advances logo is centered, with a series of orange dots forming a curved path above the word "Advances". To the right, a circular seal states "Now Indexed in Thomson Reuters Databases". Below the logo, the text "Explore AIP's open access journal:" is followed by a bulleted list of features.

AIPAdvances

Now Indexed in
Thomson Reuters
Databases

Explore AIP's open access journal:

- Rapid publication
- Article-level metrics
- Post-publication rating and commenting

Optical evidence for a self-propagating molten buried layer in germanium films upon nanosecond laser irradiation

F. Vega^{a)}

Departament d'Òptica i Optometria, Universitat Politècnica de Catalunya, C/Violinista Vellsolà 37, 08222 Terrassa, Spain

N. Chaoui^{b)} and J. Solis

Instituto de Óptica del Consejo Superior de Investigaciones Científicas, C/Serrano 121, 28006 Madrid, Spain

J. Armengol

Departament d'Òptica i Optometria, Universitat Politècnica de Catalunya, C/Violinista Vellsolà 37, 08222 Terrassa, Spain

C. N. Afonso

Instituto de Óptica del Consejo Superior de Investigaciones Científicas, C/Serrano 121, 28006 Madrid, Spain

(Received 25 October 2004; accepted 10 March 2005; published online 10 May 2005)

This work describes the phase transitions occurring at the film-substrate interface of amorphous germanium films upon nanosecond laser-pulse-induced melting of the surface. Films with thickness ranging from 50 to 130 nm deposited on glass substrates were studied. Real-time reflectivity measurements with subnanosecond time resolution performed both at the air-film and film-substrate interfaces were used to obtain both surface and in-depth information of the process. In the thicker films (≥ 80 nm), the enthalpy released upon solidification of a shallow molten surface layer induces a thin buried liquid layer that self-propagates in-depth towards the film-substrate interface. This buried liquid layer propagates with a threshold velocity of 16 ± 1 m/s and causes, eventually, melting at the film-substrate interface. In the thinnest film (50 nm) there is no evidence of the formation of the buried layer. The presence of the self-propagating buried layer for films thicker than 80 nm at low and intermediate laser fluences is discussed in terms of the thermal gradient in the primary melt front and the heat released upon solidification. © 2005 American Institute of Physics. [DOI: 10.1063/1.1900932]

I. INTRODUCTION

Laser-induced melting and rapid solidification phenomena in semiconductor thin films is currently receiving renewed attention as a cost-effective method to obtain polycrystalline thin films.^{1–4} In the particular case of germanium, the interest for studying nanosecond laser melting of amorphous Ge (*a*-Ge) films relies on their potential for the fabrication of infrared detectors as well as substrates for GaAs solar cells. Although this technique for producing polycrystalline films is in general considered to show a great ability to preserve abrupt interfaces and junctions, this aspect has to be considered with care when dealing with high-pulse fluences⁵ or films deposited on insulating substrates.

Real-time reflectivity (RTR) measurements at the film surface have widely been used to monitor the laser-induced melting and rapid solidification processes in semiconductors. However, as long as the semiconductor becomes molten, the metallic nature of the liquid phase reduces the optical penetration depth to a few nanometers, and the reflectivity measurements performed at the film surface provide no further

in-depth information about the process dynamics. Transient conductance measurements have been combined with reflectivity measurements to overcome this problem.^{6,7} However, they require complex patterning of the films to form the metallic contacts. A much simpler alternative when dealing with films deposited onto transparent substrates is to perform RTR measurements both at the front and back interfaces of the film. With this experimental approach, it is possible to obtain very sensitive time and in-depth information on the structural state of the film.^{8,9}

In the case of *a*-Ge films deposited on low thermal conductivity substrates such as glass or quartz, measurements of this type have shown that at large laser fluences solidification can occur throughout the whole molten volume (*bulk solidification process*). In this case, the large amount of solidification enthalpy released can substantially reduce the initial supercooling, in a process usually referred to as *recalescence*, which can lead eventually to the partial remelting of the initially solidified material.¹⁰ In relatively thick films (80, 100, and 130 nm), RTR measurements at the back film surface have also allowed the identification of a melting process occurring at the vicinity of the film-substrate interface several nanoseconds after the solidification of the primary melt front has already started.¹⁰ The origin of this *delayed melting process* was not fully understood even when its occurrence was

^{a)}Electronic mail: fvega@oo.upc.es

^{b)}Permanent address: Laboratoire de Chimie et Application, Institut Universitaire de Technologie de Metz, Département Chimie Rue Victor Demange, 57500 Saint-Avold, France.

clearly related to the slow transfer ratio of the solidification heat to the substrate. The limited time resolution of the reflectivity measurements earlier performed did not allow us to measure accurately the time shift between the primary (occurring at the film surface) and secondary (occurring at the film-substrate interface) melting processes. This limited time resolution was especially critical for the thinnest film (50-nm thick) studied in Ref. 10, in which the occurrence of the delayed melting process could not even be established.

The aim of this work is to determine, by means of front and back RTR measurements with improved time resolution, the origin of the delayed melting process occurring at the film-substrate interface upon nanosecond laser-pulse melting of *a*-Ge films and whether it also occurs in 50-nm thick films.

II. EXPERIMENTS AND NUMERICAL SIMULATIONS

The samples were amorphous Ge (*a*-Ge) films grown by dc magnetron sputtering from pure (99.999 9%) Ge targets on glass substrates at room temperature. Their thicknesses were 50, 80, 100, and 130 nm. The real and imaginary parts of the refractive index of the films have been reported elsewhere.¹¹

The experimental setup used to irradiate the films and to perform the real-time reflectivity (RTR) measurements has extensively been described elsewhere.^{10,12} A dye laser amplifier (sulforhodamine 6), pumped with the second harmonic of a *Q*-switched Nd:yttrium-aluminum-garnet laser ($\lambda = 532$ nm), was seeded with the output of a continuous-wave (cw) dye laser (rhodamine 6G) tuned at $\lambda = 584$ nm and pumped with an Ar⁺ laser operating at $\lambda = 514$ nm. The amplified output beam [$\lambda = 584$ nm, pulse duration $\tau = 6$ ns full width at half maximum (FWHM)] was spatially filtered before being focused at normal incidence onto the sample surface to a spot size of several hundreds of microns.

The structural transformation dynamics was studied by measuring in real time the reflectivity changes induced by the pump pulse by means of a single-mode, low-power, cw argon-ion laser ($\lambda = 514.5$ nm) at both the air-film and film-substrate interfaces. They will be referred from now on as front and back reflectivity transients, respectively. This probe beam was *s*-polarized and was focused to a 50- μ m-diameter spot onto the center of the irradiated area, the incidence angle being of $\approx 15^\circ$. The specular reflection of this beam was collected by a single-sweep streak camera,¹³ which provides a subnanosecond time resolution of typically 350 ps over a time window of 50 ns. Each area was irradiated only once and for a given fluence value, at least one front and one back RTR transient were recorded.

In order to correlate the time evolution of the reflectivity at the probe wavelength with the characteristics of the melting process, optical simulations of the front and back reflectivity changes expected upon film melting have been carried out for all films. For this purpose, a home-developed computer program based on the theory of the optical reflection of thin multilayer films has been used.^{14,15} The program takes into account both the incidence angle and polarization of the probe beam and calculates the reflectivity evolution both at

FIG. 1. Front and Back RTR transients recorded upon nanosecond laser-pulse irradiation of a 50-nm *a*-Ge film on glass. The transients have been vertically shifted for the sake of clarity, but preserving their relative scales. The laser fluence used for each transient is given in mJ/cm² on its right. The zero on the X axis corresponds to the temporal position of the maximum of the irradiation pulse (6-ns FWHM).

the front and back film interfaces as a film having the optical constants of *a*-Ge transforms layer by layer into a film having the optical constants of liquid Ge (*l*-Ge).

III. RESULTS

Figure 1 shows the time evolution of the reflectivity at the air and substrate interfaces upon irradiation of the 50-nm-thick film. For each front reflectivity transient (air-film interface), we have plotted the corresponding back reflectivity transient (film-substrate interface) obtained at the same fluence. To ease the comparison of these results to those obtained in thicker films, the reflectivity has been normalized to its initial value at the corresponding interface. All the transients are plotted in the same vertical scale (but shifted for the sake of clarity), so that the front and back RTR measurements can directly be compared. For the lowest fluence shown in Fig. 1 (88 mJ/cm²), the front RTR transient exhibits a reflectivity increase up to a maximum level related to the formation of a primary liquid Ge layer at the surface. The reflectivity remains at this value (a *plateau*) during a few nanoseconds and decreases afterwards due to the solidification and cooling processes. The reflectivity at the plateau (R_{fmax}) remains constant, while the plateau width increases as fluence is increased. For the higher fluences (154–175 mJ/cm²), a second reflectivity maximum (R_{f2}) is clearly seen at the cooling tail of the transients. This second maximum has earlier been observed upon irradiation of *a*-Ge films with both picosecond¹⁶ and nanosecond¹² pulses, and it has been related to the recalescence process, i.e., reheating and remelting produced by the release of the solidification heat during the bulk solidification of the primary melted layer.^{6,12,17} It is worth noting that whenever R_{f2} appears in the front transients, the final reflectivity measured in a longer time scale once the cooling process has been completed is lower than the initial reflectivity of the amorphous film. This result has earlier been reported in Ref. 12 and related to the

FIG. 2. Front and Back RTR transients recorded upon nanosecond laser-pulse irradiation of a 100-nm a -Ge film on glass. The transients have been vertically shifted for the sake of clarity, but preserving their relative scales. The laser fluence used for each transient is given in mJ/cm^2 on its right. The zero on the X axis corresponds to the temporal position of the maximum of the irradiation pulse (6-ns FWHM).

formation, upon solidification, of a mixture of amorphous and polycrystalline material.

The back RTR transient recorded at $88 \text{ mJ}/\text{cm}^2$ is essentially featureless, thus indicating that the primary melt front is too thin to be detected from the backside. At higher fluences, new features appear in the back RTR transients. For the transient recorded at $140 \text{ mJ}/\text{cm}^2$, a decrease in reflectivity is observed at the beginning of the transient and is followed by a neat maximum occurring at a time comparable to that of the plateau in the front transient. This maximum will be referred to from now on as $R_{b\text{max}}$, and its width increases as fluence is increased. In addition, a second well-resolved maximum with smaller amplitude is observed at longer times (this maximum will be referred to from now on as R_{b2}). It is worth noting that the lower temporal resolution used in Ref. 10 made it impossible to determine the time separation between the peaks of $R_{b\text{max}}$ and R_{b2} in the 50-nm-thick a -Ge film.

Since the RTR transients recorded in the thicker films (80, 100, and 130 nm) showed very similar characteristics as a function of fluence, representative transients obtained in the 100-nm-thick film have been selected and plotted in Fig. 2. In the case of the front RTR transients, their shape and evolution as a function of laser fluence are very similar to that observed in the 50-nm-thick film, although the second peak in the front transients, R_{12} , now becomes more evident. The final reflectivity of the front transients is always smaller than the initial value whenever R_{12} is seen. Nevertheless, in the case of the back transients, only the one obtained at the highest fluence ($220 \text{ mJ}/\text{cm}^2$ in Fig. 2) is similar to those induced in the 50-nm-thick film for high fluences. In this transient we can observe a slight decrease in reflectivity that precedes a first reflectivity maximum in the form of a plateau ($R_{b\text{max}}$) and a second maximum with a weaker intensity. We have also labeled this second reflectivity maximum appearing in the back transients at high fluences as R_{b2} .

The back RTR transients obtained at low and intermedi-

FIG. 3. Left: maximum (○) front R_{max} and (□) back $R_{b\text{max}}$ transient reflectivity as a function of the laser fluence in the 50- and 100-nm-thick a -Ge films. The lines are included to guide the eye. Right: simulated (-○-○-) front and (-□-□-) back reflectivity at the probe wavelength ($\lambda=514 \text{ nm}$) as a function of the melt depth in 50- and 100-nm-thick films.

ate fluences show instead significant differences with respect to those obtained in the 50-nm-thick film. The back transients obtained at 130 and $149 \text{ mJ}/\text{cm}^2$ are featureless for short times, but exhibit a clear reflectivity maximum at a time that is delayed in respect to the second reflectivity maximum R_{b2} observed in the transient at $220 \text{ mJ}/\text{cm}^2$. It is worth noting that this second peak, unlike R_{b2} (Fig. 1 all fluences and Fig. 2 highest fluence), is always preceded by a minimum with a level which is below that of the nonirradiated film and which shifts to shorter times as fluence is increased. This peak will be referred to from now on as *delayed peak* $R_{b\text{delay}}$. Although the observation of this delayed reflectivity peak was already reported in Ref. 10, the low time resolution of the reflectivity measurements in this earlier work made it impossible to resolve the decrease in reflectivity that precedes the delayed peak, and thus to identify that there were two different kinds of secondary maxima in the back reflectivity transients, namely, R_{b2} and $R_{b\text{delay}}$.

The dependence of the reflectivity of the primary maximum on fluence in both front and back transients, namely, R_{fmax} and $R_{b\text{max}}$, has been plotted in the left-hand side of Fig. 3 for the case of the 50- and 100-nm-thick films. On the right-hand side, the front and back reflectivity changes calculated from optical simulations, assuming that melting starts at the film surface and propagates in depth, have been plotted as a function of the primary melt depth. The complex refractive indices ($n-ik$) used for the simulations were $n_{a\text{-Ge}}=4.73$, $k_{a\text{-Ge}}=2.21$, $n_{l\text{-Ge}}=2.51$, $k_{l\text{-Ge}}=5.12$, and $n_{\text{glass}}=1.52$ (see Refs. 11, 18, and 19, respectively). The comparison of the experimental results (left-hand side) and the calculated ones (right-hand side) shows a very good agreement, and thus confirms that the primary melt front is responsible for the reflectivity maxima (R_{fmax} and $R_{b\text{max}}$) in the initial part of the front and back RTR transients. Actually, the initial decrease in reflectivity followed by $R_{b\text{max}}$ in the back transients can be understood as a consequence of optical interference effects associated with the layer that remains solid between the melted liquid layer and the substrate. A more detailed analysis of the experimental and calculated results shows that regardless of the film thickness the reflectivity minimum that precedes $R_{b\text{max}}$ occurs once the primary melt front gets closer to the substrate than approximately 15 nm.

Figure 3 clearly shows also that while the front reflec-

FIG. 4. Calculated melt depth as a function of laser fluence in (▲) 50-, (○) 80-, (□) 100-, and (◇) 130-nm-thick *a*-Ge films.

tivity measurements are very sensitive at low fluences and shallow melt depths, the back ones become extremely sensitive to melt depth variations once the primary front melt depth approaches the substrate. Using both sets of data, the dependence of melt depth on fluence can be obtained straightforward from Fig. 3. The corresponding results are shown in Fig. 4, where it is seen that, as expected, the melt depth increases as the fluence is increased. Moreover, it is also seen that for a given fluence, melt depth decreases as thickness is increased, the 50-nm-thick film being an exception to this behavior.

Figure 5 shows the reflectivity values at the different maxima that appear in the representative 100-nm-thick film as a function of fluence: (a) R_{fmax} and R_{f2} in the front transients and (b) R_{bmax} , R_{b2} , and R_{bdely} in the back transients. To ease the discussion, the dependence of the melt depth on fluence has been included in (c). Similar results were obtained for the 80- and 130-nm-thick films. Figure 5(b) shows that the reflectivity at R_{bdely} peak increases rapidly with fluence up to a saturation value. The comparison of Figs. 5(a) and 5(b) shows that R_{bdely} appears at fluences slightly below

FIG. 5. Reflectivity at the representative peaks labeled in Fig. 2, for the 100-nm-thick *a*-Ge film as a function of fluence for (a) the front transients, (●) R_{fmax} and (○) R_{f2} , and (b) the back transients, (■) R_{bmax} , (△) R_{bdely} , and (□) R_{b2} reflectivity, (c) Melt depth as a function of fluence deduced from Fig. 4.

the threshold to observe the second peak R_{f2} in the front transients and much lower than the fluence necessary to observe the second peak R_{bmax} in the back transients. Comparing Figs. 5(b) and 5(c), it is clearly seen that it is necessary that the primary melt front reaches a depth around 30–35 nm to obtain a measurable R_{bdely} peak. In other words, R_{bdely} in the back transients appears when the primary melt front is still too thin (only 30–35 nm) to be detected from the backside.

Finally, when the whole film is melted, the R_{bdely} peak is no longer observed in the back transients (see Fig. 2), and the secondary reflectivity maximum R_{b2} appears instead. The transition between the occurrence of R_{bdely} and R_{b2} is seen in Fig. 5 by a sharp decrease of the reflectivity value at R_{bdely} . This happens when the primary melt front becomes very close to the substrate (around 10–15 nm away from it).

IV. DISCUSSION

The second reflectivity maxima R_{f2} and R_{b2} appearing at the cooling tail of both front and back RTR transients have earlier been observed upon irradiation of *a*-Ge films with both, picosecond^{20,21} and nanosecond¹² pulses, and it has been related to the recalescence process, i.e., the reheating and remelting processes produced by the release of the solidification heat during the bulk solidification of the primary melted layer.^{6,12,16} Therefore, the existence of R_{f2} and R_{b2} can be considered as a direct evidence of the recalescence processes, as seen from either side of the film. Further support to this reasoning comes from the fact that recalescence gives rise to the formation of a crystalline material upon solidification,²⁰ with optical properties different from those of the initial amorphous film. The latter is in agreement with the lower final reflectivity observed in our experiments whenever R_{f2} was present in the front RTR transients.

The results achieved in the thicker films (80, 100, and 130 nm) clearly evidence the existence of a delayed R_{bdely} peak with new features. The value of the reflectivity at R_{bdely} increases with fluence (see Fig. 5) and saturates for a reflectivity change around 0.60 which, according to optical simulations, is consistent with an 8-nm-thick Ge liquid layer located at the film-substrate interface. We can thus conclude that R_{bdely} is associated with the existence of a thin melted layer in the neighborhood of the substrate-film interface, this layer being referred from now on as *buried melted layer*.

The optical simulations allow us to estimate that R_{bdely} is observed for primary melt depths as shallow as 30–35 nm. Furthermore, the comparison of the times at which R_{bdely} and R_{fmax} occur clearly shows that the buried liquid layer appears once the primary melted layer at the front surface has already solidified. This is further supported by two experimental observations: (i) R_{bdely} appears in back RTR transients that are essentially “featureless” since a 30–35-nm-thick primary melted layer at the film surface cannot be detected from the backside (see back transients in Fig. 2 obtained at 130 and 149 mJ/cm²) and (ii) when R_{bdely} is observed, the corresponding front transient does not show recalescence since the melt depth is not enough for bulk solidification to occur.^{17,22}

FIG. 6. Scheme of the melting-solidification process that leads to the formation of the buried liquid layer. (a) The solid a -Ge film on glass substrate prior to irradiation, (b) The film upon irradiation with a nanosecond laser pulse that leads to surface melting while a X_{a-Ge} thick a -Ge layer remains, (c) The film once the primary melt front moves backwards to the surface and the buried liquid layer self-propagates in depth towards the glass substrate.

In order to understand the observed behavior, there are thus two questions to consider: Which is the origin of this buried liquid layer located at the film-substrate interface? Why is it delayed in respect to the primary melt front? To answer the first question, it must be taken into account that before the buried liquid layer appears, there must be a primary liquid layer at the film surface that is at least 30–35-nm thick. Figure 6 illustrates the sequence that leads to the appearance of the buried layer: (a) the starting film configuration, (b) the film configuration once the primary surface melt layer has achieved its maximum depth, and (c) the film configuration once solidification of the surface melt layer has started, showing the coexistence of a liquid front moving backwards to the surface and a buried layer moving towards the film-substrate interface. Since the buried layer can be formed with an initial melt depth that is relatively shallow, a remnant solid a -Ge layer must exist between the primary liquid layer and the glass substrate. This solid a -Ge has a thermal conductivity higher than that of the glass substrate,²³ promoting a significant thermal gradient across the depth of the primary liquid layer and a severe undercooling. Under these conditions, the resolidification of the primary melted layer leads to the formation of a polycrystalline material (p -Ge) that has a melting point higher than that of the amorphous phase. The latent heat released by the solidification process would be able to melt a thin layer of the underlying solid a -Ge. Upon solidification of this buried liquid layer into the crystalline phase, the process restarts, thus leading to the self-propagation of a melted layer towards the film-substrate interface even once the surface melted layer has completely solidified.^{24–26} The fact that our front RTR transients do not show the characteristic interference maxima and minima^{27,28} can easily be understood by taking into account that the buried liquid layer is always located at a depth for which the front reflectivity probe is saturated (see Fig. 3). Once the primary melt front is solidified the front probe could detect the buried layer. However, when this happens, the buried liquid layer is already too deep to be probed from the front side due to the absorption of the 514-nm light by the resolidified Ge. The existence of such self-propagating in-depth buried layer and its relation to R_{bdely} are further supported by the decrease in reflectivity that always precedes R_{bdely} (see back RTR transients recorded at 130 and

FIG. 7. Distance X_{a-Ge} as a function of Δt_{a-Ge} (see text for definitions) for the 80-nm-thick a -Ge film. The horizontal dot line marks the approximate depth for which the slope change occurs. The solid line is a linear fit of the experimental data for $X_{a-Ge} > 10$ nm.

149 mJ/cm² in Fig. 2). The optical simulations show that this decrease is caused by interference effects between the light reflected at the buried liquid layer/remnant a -Ge interface and the light reflected at the remnant a -Ge/glass substrate interface.

The propagation velocity of the buried liquid layer can be calculated straightforward as follows. Figure 4 provides the primary melt depth induced as a function of fluence. The film thickness minus this primary melt depth gives the distance that the buried liquid layer must travel across the remnant a -Ge to reach the substrate. This distance is indicated in Fig. 6(b) as X_{a-Ge} . The time interval the buried liquid layer needs to reach the substrate is easily obtained from the back RTR transients as the temporal separation between R_{bmax} (the time at which the primary melt front achieves its maximum depth) and the position of the second maximum (either R_{bdely} or R_{b2}), that is, the time at which the buried layer reaches the substrate. This time interval will be referred from now on as Δt_{a-Ge} . Figure 7 shows the dependence of X_{a-Ge} on Δt_{a-Ge} for the case of the 80-nm-thick film. It is seen that the data follow a linear trend with a clear slope change at the substrate distance of ≈ 10 nm, i.e., when the primary melt front is very close to the glass substrate. This change of slope occurs when R_{bdely} converts into R_{b2} , and thus provides further support to our interpretation that the two secondary reflectivity peaks in the back transients (R_{b2} for thick primary melt depths or R_{bdely} for shallower ones) have a different origin. The linear evolution of the results plotted for X_{a-Ge} for distances higher than 10 nm indicates that the buried layer propagates with a constant velocity of 16 ± 1 m/s. This value is of the same order of magnitude as those reported earlier for the propagation of a buried layer in amorphised Si by ion implantation.^{24,28}

The results obtained in the thinnest 50-nm film show no evidence of a self-propagating buried liquid layer. Instead, the results show “symmetric” front and back recalescence processes above a fluence threshold (front and back RTR transients obtained at fluences higher than 126 mJ/cm² in Fig. 1). To explain this behavior, it has to be considered that the skin penetration depth in a -Ge at the irradiation wavelength ($\lambda = 584$ nm) is as high as 26 nm.^{11,12} So, the lower the film thickness, the lower the expected thermal gradient across the film depth related to the laser absorption. In addition, the lower effective thermal conductivity of the 50-nm-thick film-glass substrate system compared to the case of the thicker films contributes further in reducing the temperature

gradient induced upon irradiation at a given fluence.²⁹ Both conditions favor a solidification scenario, with a reduced thermal gradient across the film thickness that prevents the propagation of a buried liquid layer. Therefore, the thermal response conditions achieved in this work for a film thickness of 80 nm represent the threshold for the buried layer to occur and the 16 ± 1 -m/s velocity determined is thus the minimum velocity for such a self-propagating buried layer.

In the case of the thicker films (80, 100, and 130 nm), when the laser fluence is increased and the primary melted layer becomes thicker, the front RTR transients provide evidence for the occurrence of recalescence (see front transients in Fig. 2 obtained at 149–193 mJ/cm²). It occurs for large melt depths and low thermal response as discussed in Refs. 12 and 16, and is consistent with a bulk solidification scenario that occurs once the thermal gradient through the primary melted layer is significantly reduced. Once the laser fluence is high enough to induce melting across the whole film (i.e., the primary melted layer reaches the glass substrate) the front and back transients become quite similar, both exhibiting secondary reflectivity peaks (R_{f2} and R_{b2}) in the cooling tail of the transients due to the recalescence process. Under these conditions, there is obviously no chance for a buried liquid layer to be formed. Further support to this interpretation comes from the fact that following both R_{f2} and R_{b2} , the reflectivity remains decreasing with a smooth slope, thus indicating that the solidification process is still in progress.

V. CONCLUSIONS

We have demonstrated the existence of melting at the film-substrate interface due to a buried liquid layer that self-propagates in depth when *a*-Ge films deposited on low thermal conductivity substrates are irradiated with nanosecond laser pulses. This buried liquid layer propagates independently of the primary liquid layer formed at the film surface and leads to melting at the film-substrate interface once the film surface has completely solidified. The existence and propagation features of this buried liquid layer are laser fluence and film thickness dependent since it is only observed for the thicker films studied (80, 100, and 130 nm) and for low and intermediate fluences. Once the surface liquid layer starts to solidify into a crystalline phase, the solidification enthalpy released melts the amorphous material underneath, thus forming a buried liquid layer that self-propagates in depth towards the film-substrate interface. Such a buried layer appears once a primary melted layer of at least 30–35 nm is induced, and propagates with a threshold velocity of 16 ± 1 m/s. For the higher fluences studied, the primary melt front approaches the film-glass interface and bulk solidification occurs instead, with a release of solidification enthalpy sufficient to eventually remelt the primary solidified mate-

rial. For the thinnest film studied (50-nm thick), the induced thermal gradient across the film is much lower favoring a “symmetric” bulk solidification scenario.

ACKNOWLEDGMENTS

This work has been partially supported by the MCYT through Project No. DPI: DPI2002-00151 and by the European Union in the framework of the TMR ERB FMRX Contract No. CT098-0188 “Laser Cleaning” Network. N. C. acknowledges the funding of the European Union through the latter contract.

- ¹M. Lee, S. Moon, M. Hatano, K. Suzuki, and C. Grigoropoulos, *J. Appl. Phys.* **88**, 4994 (2000).
- ²L. Brambilla, L. Colombo, V. Rosato, and F. Cleri, *Appl. Phys. Lett.* **77**, 2337 (2000).
- ³P. V. Santos, A. Trampert, F. Doudeo, D. Comedi, H. J. Zhu, K. H. Ploog, A. R. Zanatta, and I. Chambouleyron, *J. Appl. Phys.* **90**, 2575 (2001).
- ⁴Y. F. Chong, K. L. Pey, A. T. S. Wee, M. O. Thompson, C. H. Tung, and A. See, *Appl. Phys. Lett.* **81**, 3786 (2002).
- ⁵J. Siegel, J. Solis, C. N. Afonso, F. Vega, J. Bankmann, O. Martinez-Sacristan, and K. Sokolowski-Tinten, *J. Appl. Phys.* **89**, 3644 (2001).
- ⁶S. R. Stiffler, M. O. Thompson, and P. S. Peercy, *Phys. Rev. Lett.* **60**, 2519 (1988).
- ⁷S. R. Stiffler, M. O. Thompson, and P. S. Peercy, *Phys. Rev. B* **43**, 9851 (1991).
- ⁸K. Murakami, O. Eryu, K. Takita, and K. Masuda, *Phys. Rev. Lett.* **59**, 2203 (1987).
- ⁹J. Boneberg, J. Nedelcu, H. Bender, and P. Leiderer, *Mater. Sci. Eng., A* **173**, 347 (1993).
- ¹⁰F. Vega, J. Solis, J. Siegel, and C. N. Afonso, *J. Appl. Phys.* **88**, 6321 (2000).
- ¹¹J. C. G. de Sande, C. N. Afonso, J. L. Escudero, R. Serna, F. Catalina, and E. Bernabeu, *Appl. Opt.* **31**, 6133 (1992).
- ¹²J. Armengol, F. Vega, N. Chaoui, J. Solis, and C. N. Afonso, *J. Appl. Phys.* **93**, 1505 (2003).
- ¹³J. Solis, J. Siegel, and C. N. Afonso, *Rev. Sci. Instrum.* **71**, 1595 (2000).
- ¹⁴H. A. Macleod, *Thin Film Optical Filters*, 2nd ed. (Hilger, London, 1986).
- ¹⁵F. Vega, J. Solis, J. Siegel, and C. N. Afonso, *J. Appl. Phys.* **88**, 6321 (2000).
- ¹⁶J. Siegel, J. Solis, and C. N. Afonso, *Appl. Phys. Lett.* **75**, 1071 (1999).
- ¹⁷T. Sameshima and S. Usui, *J. Appl. Phys.* **74**, 6592 (1993).
- ¹⁸G. E. Jellison and D. H. Lowndes, *Appl. Phys. Lett.* **51**, 352 (1987).
- ¹⁹E. D. Palik, *Handbook of Optical Constants of Solids* (Academic, New York, 1985).
- ²⁰J. Siegel, J. Solis, and C. N. Afonso, *J. Appl. Phys.* **84**, 5531 (1998).
- ²¹J. Siegel, J. Solis, and C. N. Afonso, *Appl. Phys. Lett.* **75**, 1071 (1999).
- ²²F. Vega, C. N. Afonso, W. Szyszko, and J. Solis, *J. Appl. Phys.* **82**, 2247 (1997).
- ²³W. Szyszko, F. Vega, and C. N. Afonso, *Appl. Phys. A: Mater. Sci. Process.* **61**, 141 (1995).
- ²⁴M. O. Thompson, G. J. Galvin, J. W. Mayer, P. S. Peercy, J. M. Poate, D. C. Jacobson, A. G. Cullis, and N. G. Chew, *Phys. Rev. Lett.* **52**, 2360 (1984).
- ²⁵W. Sink and F. W. Saris, *Phys. Rev. Lett.* **53**, 2121 (1984).
- ²⁶R. F. Wood and G. A. Geist, *Phys. Rev. Lett.* **57**, 873 (1986).
- ²⁷D. H. Lowndes, G. E. Jellison, Jr., S. J. Pennycook, S. P. Withrow, and D. N. Mashburn, *Appl. Phys. Lett.* **48**, 1389 (1986).
- ²⁸J. J. P. Bruines, R. P. M. van Hal, H. M. J. Boots, A. Polman, and F. W. Saris, *Appl. Phys. Lett.* **49**, 1160 (1986).
- ²⁹M. von Allmen and A. Blatter, *Laser-Beam Interactions with Materials*, 2nd ed., Springer Series in Material Science Vol. 2 (Springer, Berlin, 1987), pp. 44–47.