Local Si-H bonding environment in hydrogenated amorphous silicon films in relation to structural inhomogeneities

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The hypothesis of the relation between the local environment of polyhydride $[(SiH_2)_n]$ groups and structural inhomogeneities (due to the presence of voids) of an amorphous silicon network has been tested in hydrogenated amorphous silicon films prepared by rf magnetron sputtering (RF MS). Comparative infrared (IR) absorption, Raman spectroscopy and optical transmission measurements have been performed on samples deposited under the same plasma conditions at various substrate temperature (T). The results of the IR studies show that all samples exhibit the same hydrogen content incorporated as isolated monohydride (SiH) groups responsible for the absorption band at 2004 cm^{-1}. As a consequence, the gradual decrease in the total bonded hydrogen content as T increases is essentially due to the decrease in the amount of hydrogen bonded as polyhydride $[(SiH_2)_n]$ groups which absorb around 2086 cm^{-1}. A direct relationship is established between the changes in the IR spectra and the variation of structural inhomogeneities observed by Raman spectroscopy. The linewidth in the low-frequency side of the transversal acoustic (TA) -like band in the Raman spectra decreases monotonously as T increases. This decrease is attributed to a decrease in density of quasilocalized vibrational modes in structural inhomogeneities (disordered domains around the surface of voids) in the amorphous network. The good correlation between the IR and Raman results indicates that the $(SiH_2)_n$ groups are located in structural inhomogeneities due to the presence of voids in the amorphous network where the formation of these complexes is favorable. This interpretation is corroborated by the variation of both the static refractive index, which gives information about the density of material, and the dispersion energy, which measures the mean coordination number of Si atoms and the effective number of valence electron per Si atom in the amorphous network. [S0163-1829(99)02608-9]

I. INTRODUCTION

The distribution of silicon-hydrogen (Si-H) bonding in hydrogenated amorphous silicon (a-Si:H) films has been the subject of considerable studies.[1–5] This interest has been guided not only by the role played by the hydrogen in saturating the Si dangling bonds in the amorphous silicon network, which leads to a decrease in the density of states in the gap, but also it reduces the structural disorder, which results in a narrowing of the band tails.[6,7] In addition, it has also been shown that a-Si:H samples cannot be schematized by a homogeneous structure. Several studies show that this material exhibits structural inhomogeneities[8–10] which affect the optoelectronic properties.[11,12] The study of the vibrational modes of Si-H bonding, using infrared (IR) absorption measurements especially in the stretching and bending modes regions, gives information about the structural inhomogeneities in the amorphous network, because the vibrational frequencies of Si-H bonding depends strongly on both their configurations and their local environment. Three groups of structures are found in IR spectra corresponding to (i) stretching band modes located at 2000 and 2090 cm^{-1}, which have been associated with isolated monohydride (SiH) groups and polyhydride $(SiH_2)_n$, and/or clustered monohydride $(SiH)_n$ groups, respectively, (ii) doublet bending modes at 840 and 890 cm^{-1} which have been associated with $(SiH_2)_n$ groups, and (iii) waging band modes around 640 cm^{-1} which is attributed to all configurations. In fact, if the band at 2000 cm^{-1} is undisputed, the one located at about 2090 cm^{-1} has been, on the contrary, the subject of large controversies.[3–5,11,13,14] The center of these controversies is mainly the shift of this band due to different structural and/or chemical environment. It has also been suggested in particular that, within the "island and tissue" model, the region between the islands in a-Si:H, where the density is lower than in the islands, includes $(SiH_2)_n$ groups.[15,16]

In order to correlate the modes of incorporation of hydrogen in the amorphous network with structural inhomogeneities, we investigate the effect of the substrate temperature at increasing temperature on both the IR absorption and Raman spectra in rf magnetron sputtering (RFMS) films. It is important to notice that the Raman spectroscopy is a good tool not only to study the local structural order[17,18] but also to obtain information about the structural inhomogeneities (linked with the presence of voids) in the amorphous network.[19] We
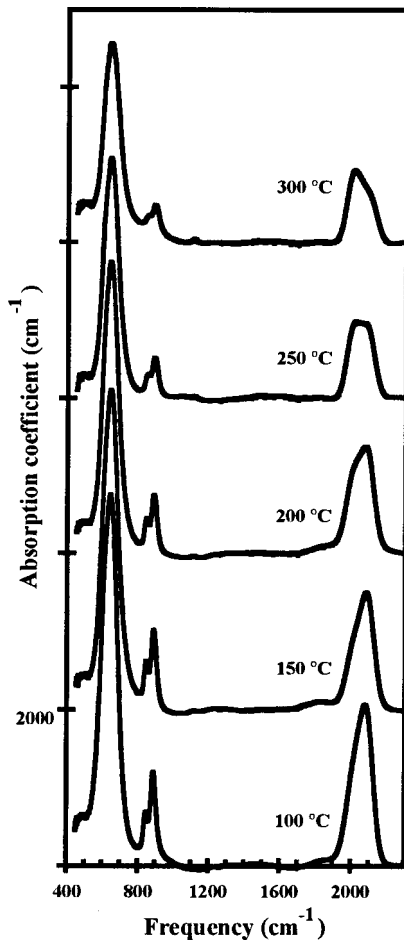
FIG. 1. IR absorption spectra obtained for series 1–5 deposited at increasing substrate temperature as indicated.

also studied the Si-Si bonding by optical transmission experiments, within the chemical bonding spectroscopy, using the model of signal-effective oscillator.[20] In this model the refractive-index dispersion $n(E)$ below the interband absorption edge is linked to the dispersion energy E_d, which depends on the strength of interband optical transition, by $n^2(E) - 1 = E_m E_d / E_m^2 - E^2$, where E is the photon energy and E_m is the single oscillator energy. The parameter E_d follows the the empirical relation $E_d = \beta N_c Z N_e$, where β depends on the ionicity of atoms, N_c is the mean coordination number of the atoms, Z is the formal chemical valency of atoms, and N_e is the effective number of valence electron per atom. Finally, we will attempt to give a quantitative description of the growth process of the studied a-Si:H films.

II. EXPERIMENT

Five series of hydrogenated amorphous silicon films (labeled 1–5), 2 μm thick, have been deposited on both crystalline silicon and Corning 7059 substrates by rf magnetron sputtering in an Ar+H$_2$ gas mixture ([H$_2$]/[Ar]= 15%). The target-substrate distance, the gas pressure, and the rf power were fixed at 60 mm, 5.5×10^{-3} mbar and 500 W, respectively. The only varying parameter was the substrate temperature from 100 to 300 °C by steps of 50 °C. The resulting deposition rate is relatively high and increases from 7 to 10

Å/s, when T increases from 100 to 300 °C.

The hydrogen content and bonding configurations are deduced from IR absorption measurements. The IR spectra are obtained at room temperature using a Bomem DA 3.01 Fourier transform spectrometer. Using the proportionality constants previously reported,[21] $A_{640} = 2.1 \times 10^{19} \, \text{cm}^{-2}$, $A_{2000} = 9 \times 10^{19} \, \text{cm}^{-2}$, and $A_{2090} = 2.21 \times 10^{20} \, \text{cm}^{-2}$, we can estimate from the integrated intensity (i) of the wagging band, the total content of the bonded hydrogen C_{640}, (ii) of the 2000 cm^{-1} stretching band, the concentration of hydrogen bonded as isolated monohydride groups C_{2000}, and (iii) of the 2090 cm^{-1} stretching band, the concentration of polyhydride and/or clustered monohydride groups C_{2090}, respectively.

The Raman spectra were recorded at room temperature with a double monochromator spectrometer (Jobin Yvon U 1000), equipped with a cooled photomultiplier and a photon counting system. The measurements were performed in the usual quasi-back-scattering configuration, in vacuum to prevent air scattering, with the 476.5-nm line of an argon laser as the exciting source. At this wavelength, the laser light is entirely absorbed by the a-Si:H layer, giving information about the bulk layer. The laser spot diameter at the sample surface was on the order of 80 μm, with a power density of $\cong 10^{+3} \, \text{W/cm}^2$ in order to minimize heating effects.

The optical constants were determined by optical transmission experiments using a Cary Model 05 double beam spectrometer at room temperature. For the determination of the refractive-index dispersion below the interband absorption edge from these measurements, we used the method presented in Chahed's thesis.[22] This method leads to the small errors for thick films ($\cong 2 \, \mu$m).

III. RESULTS AND DISCUSSION

A. Silicon-hydrogen bonding

The IR absorption spectra presented in Fig. 1 show the different hydrogen bonding configurations for 1 to 5 films deposited at different substrate temperatures. For all the samples, even those deposited at high substrate temperature, the stretching band modes can be decomposed into two Gaussian components centered at fixed frequencies 2004 and 2086 cm^{-1}, as evidenced by Fig. 2. The 2086 cm^{-1} component is systematically accompanied by a well-resolved doublet in the bending region located also at fixed frequencies 845 and 890 cm^{-1} (Fig. 1). These doublets still remain detectable even for films deposited at substrate temperature as high as 300 °C. These result suggest clearly that all samples exhibit, in addition to the isolated monohydride (SiH) groups (corresponding to the 2004 cm^{-1} component), a large proportion of polyhydride (SiH$_2$)$_n$ groups (corresponding to the 2086 cm^{-1} component).

In order to test if our samples contain clustered monohydride (SiH)$_n$ groups which do not give any bending modes, we plotted in Fig. 3 the integrated absorption of the bending (840 and 890 cm^{-1}) bands as a function of that of the stretching band at 2086 cm^{-1}. This figure clearly shows, on the one hand, a linear behavior between the integrated absorption of the bending bands and that of the stretching band at 2086 cm^{-1}, and, on the other hand, the extrapolation of this linearity shows that the integrated absorption of the 840 and 890 cm^{-1} bands tends towards zero when that of the 2086 cm^{-1} band tends to zero. This suggests that the component at
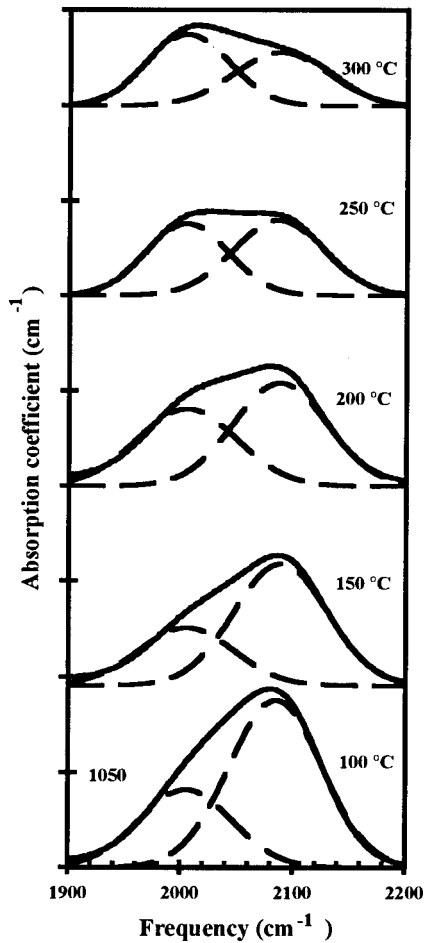
FIG. 2. IR absorption spectra in the region of the stretching band modes corresponding to the a-Si:H samples of Fig. 1. The decomposition of this band into two Gaussian components corresponding, respectively, to the SiH groups (at 2004 cm^{-1}) and $(SiH_2)_n$ groups (at 2086 cm^{-1}) are also shown (dashed curves).

FIG. 3. Variation of the integrated absorption of the bending bands (at 840 and 890 cm^{-1}) as a function of that of the stretching band (at 2086 cm^{-1}), deduced from the decomposition of Fig. 2, for the series 1–5. The integrated absorption of the bands increases in going from series 5 ($T = 300\,°C$) to series 1 ($T = 100\,°C$).

2086 cm^{-1} is essentially associated with the presence in the films of polyhydride $(SiH_2)_n$ groups rather than interacting monohydride complexes $(SiH)_n$.

The bonded hydrogen concentrations deduced from the analysis of IR spectra, i.e., the total bonded hydrogen content C_{640}, the concentration C_{2000} of the hydrogen bonded as isolated monohydride groups, and the concentration C_{2090} of hydrogen bonded as polyhydride groups are reported in Fig. 4 as a function of the substrate temperature.

Surprisingly enough, Fig. 4 shows that all the samples exhibit almost the same content ($C_{2000} \cong 6$ at %) of hydrogen incorporated as isolated monohydride groups. Consequently, the gradual decrease of total bonded hydrogen concentration C_{640} (from 38.6 to 20.5 at %) as the substrate temperature increases is essentially due to the decrease of the amount of hydrogen incorporated as polyhydride groups C_{2090}. In addition, these complexes are still present in the films in a large proportion even at substrate temperature as high as 300 °C. This suggests that the substrate temperature limits the incorporation of hydrogen atoms bonded in the shape of $(SiH_2)_n$ groups without any effect on the incorporation of hydrogen in isolated monohydride groups. Such a behavior is very useful to study the effect of the polyhydride groups on the prop-
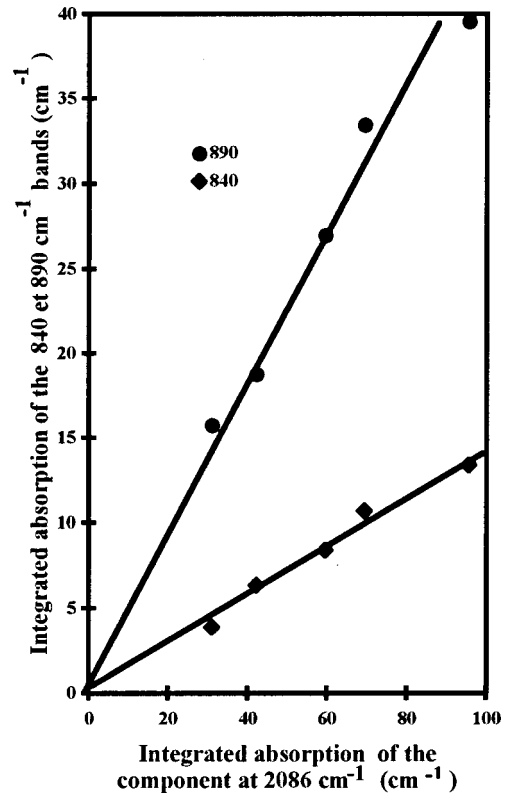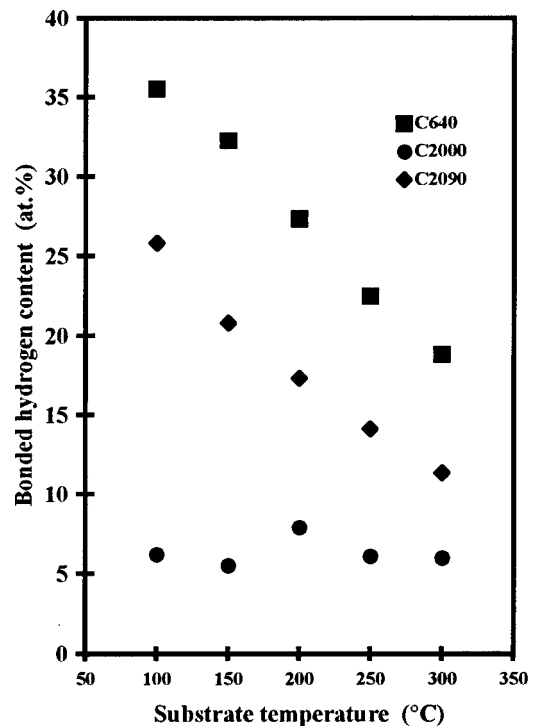
FIG. 4. Variation, as a function of the substrate temperature, of the total bonded hydrogen content C_{640}, the concentration of hydrogen bonded as monohydride SiH groups C_{2000}, and the concentration of hydrogen bonded as polyhydride $(SiH_2)_n$ groups C_{2090}, obtained for series 1–5.
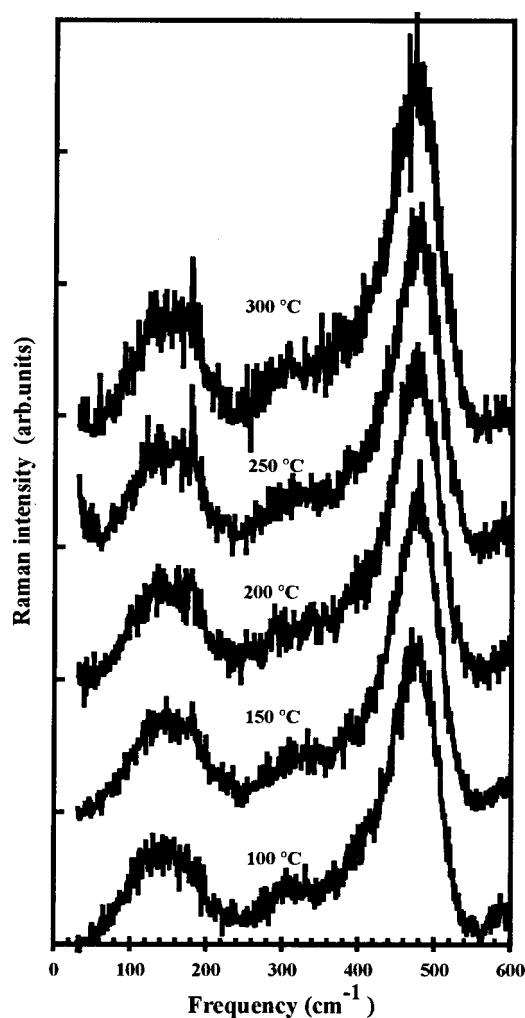
FIG. 5. Raman spectra obtained for films 1–5 deposited at different substrate temperatures in the range 0–600 cm^{-1}, showing the different bands. The areas under each spectrum are normalized to the same value.

erties of the material. It is worth noticing that similar results have been also observed in a-Si:H films obtained by plasma enhanced chemical vapor decomposition (PECVD) of pure silane.[23] These results can be interpreted as an effect of the substrate temperature which reduces the proportion of structural inhomogeneities, linked with the presence of voids, where the formation of the $(SiH_2)_n$ configurations is favorable, and will be corroborated by the Raman spectra analysis.

B. Structural inhomogeneities

Let us now discuss the results from the point of view of structural inhomogeneities of the amorphous network. We present in Fig. 5 the first-order Raman spectra in the 0–600 cm^{-1} region, obtained for different substrate temperatures 100–300 °C, corresponding to the same a-Si:H films (1–5). These spectra are corrected to the background caused by higher-order Raman scattering processes[24] and the areas under each spectrum are normalized to the same arbitrary value. Particular attention has been paid to the low frequencies of the transversal acoustic (TA) -like structure. The low part ($\omega < 160$ cm^{-1}) of the Raman spectra can be fitted by a
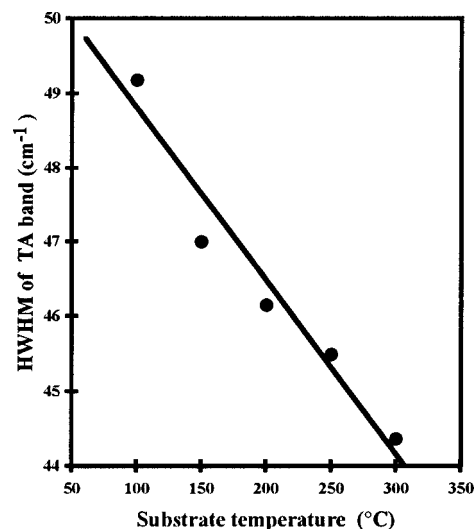

FIG. 6. Variation of the half-width at half maxima (HWHM) of the TA-like band as a function of the substrate temperature for the same series of films (1–5).

Gaussian line shape centered at about 140 cm^{-1}. A remarkable result is gained from the data obtained for the half-width at half maxima (HWHM) towards the low frequencies of the TA-like structure, which increases from 44.3 to 49.2 cm^{-1} as the substrate temperature decreases from 300 to 100 °C, as is shown in Fig. 6. This increase may be interpreted by an increase in the density of phonons and therefore leads to a divergence to Debye's law characteristic of well connected structure such as crystalline silicon and fully coordinated a-Si models, where the density of phonons is proportional to the square of the frequency ($\cong \omega^2$) in the low-frequency region. In the context of the calculations of the density of phonons previously proposed by Chehaidar et $al.$,[19] our results would suggest an increase of the size and concentration of voids in the amorphous network as the substrate temperature decreases. Indeed, these calculations[19] have been made using various models of an amorphous silicon network, involving different sizes and concentrations of voids, and which show that the linewidth of the TA-like band towards the low frequencies increases systematically as the size and the concentration of the voids increase. The class of a-Si models, generated by Chehaidar et $al.$,[19] where various concentrations and sizes of voids have been introduced, suggests the presence of two domains in the amorphous network: (i) the first one, which is more disordered (i.e., inhomogeneity), is located around the surface of the voids; (ii) the second one, which is more relaxed, is located between the inhomogeneities. Such domains have been detected recently in both unhydrogenated and hydrogenated amorphous silicon using x-ray diffraction spectroscopy.[10] The effect of the structural inhomogeneities on the TA-like band towards the low frequencies in the amorphous network is a universal feature of amorphous solids with high disorder, which can be interpreted as due to the coexistence of the extended acoustic phonon (modes of Debye) with the quasilocalized modes in the structural inhomogeneities.[19,25,26] These inhomogeneities have been defined by disorder domains located around the surface of the voids,[19] by domains containing coordination
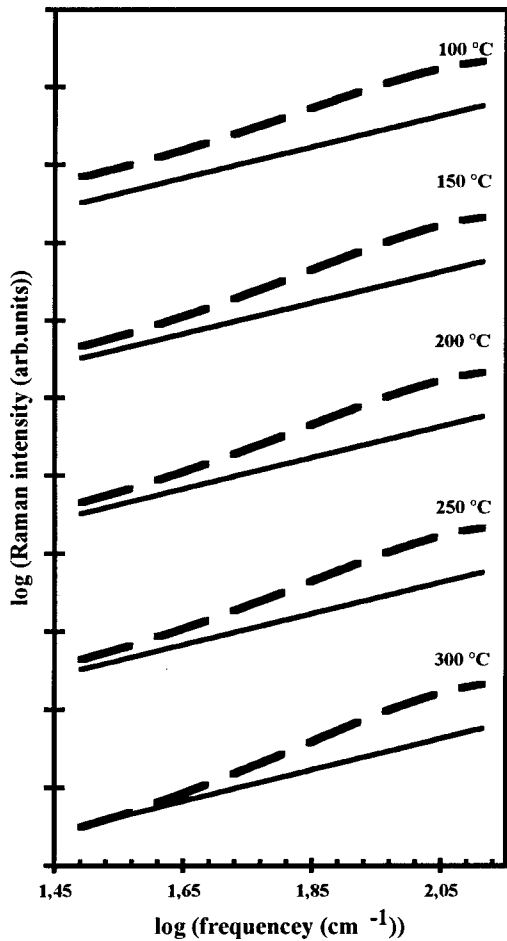
FIG. 7. Double-logarithm plot of the the fitted spectra of Fig. 5 in the low-frequency range $(30-130 \text{ cm}^{-1})$ (dashed curves) for samples 1–5. The straight line of slope equal to 2 corresponds to Debye's law.

defects,[25] or by domains which differ significantly from the average medium environment.[26]

Therefore, more information about such behavior in our samples can be gained from a more detailed analysis of the Raman spectra and consequently the vibrational densities obtained for the samples 1–5 in the low-frequency range $(30 < \omega < 130 \text{ cm}^{-1})$. The data of Fig. 5 in the $30-130 \text{ cm}^{-1}$ range are reported in Fig. 7 in a double-logarithmic plot. The spectra are replaced by their best fit with Gaussian line shapes and shifted by the same quantity for more clarity. In this figure we fitted the low-frequency part $(\omega < 40 \text{ cm}^{-1})$ of the spectrum corresponding to the more dense sample (deposited at 300 °C) with a straight line of slope 2, which presents Debye's law. This straight line is then translated by the same value separating the spectra of the other samples (deposited at 250, ... ,100 °C). The analysis of this figure shows that the relative divergence from the law of Debye becomes more important as the substrate temperature decreases, i.e., as the fraction of inhomogeneity increases. This behavior can be, as already emphasized, interpreted with the increase in density of the quasilocalized phonons at structural inhomogeneities.[19,25,26] Indeed, this divergence as a function of the inhomogeneities fraction, generated in the models of the amorphous network, is clearly evidenced by the calculations of the vibrationnel density in the 10–90 cm^{-1} range.[19]
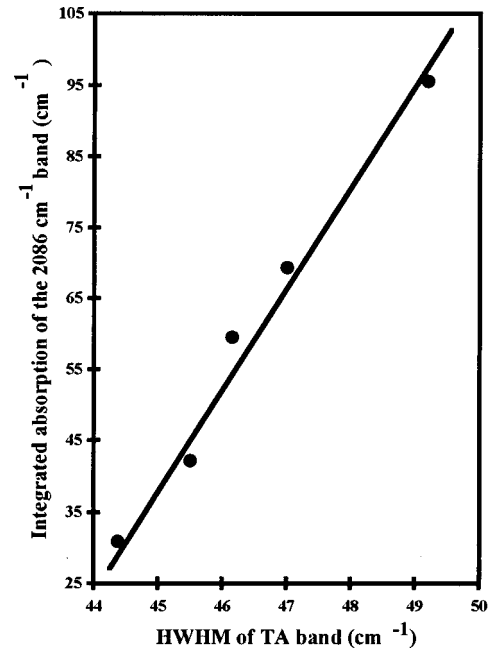

FIG. 8. Variation of the integrated absorption of the stretching band at 2086 cm^{-1} as a function of the HWHM of the TA-like band for series 1–5. Note that the values increase in going from series 5 to series 1.

Let us now return to the Si-H bonding results obtained from the IR absorption experiments and discussed in the above section. We present in Fig. 8 the integrated absorption of the 2086 cm^{-1} component as a function of the HWHM of the TA-like band, determined for each sample 1 to 5. This figure clearly shows that the hydrogen content bonded as polyhydride complexes increases linearly with the HWHM of a TA-like band (i.e., structural inhomogeneities) in going from sample 5 (deposited at the highest $T = 300$ °C) to sample 1 (deposited at the lowest $T = 100$ °C). The clear correlation obtained between the variation of the proportion of the structural inhomogeneities, through the HWHM of the TA-like band, and that of the integrated absorption of the 2086 cm^{-1} component, suggests that the hydrogen bonded as polyhydride groups, which give rise to this absorption band, might be attributed to hydrogen atoms located in structural inhomogeneities (around the voids and their surfaces), and therefore gives a satisfactory interpretation of the origin of the 2086 cm^{-1} component of the IR stretching band. This result is in good agreement with the comparative small-angle x-ray scattering and IR absorption studies performed on a-Si:H films obtained by the PECVD method, which indicate that the polyhydride groups reside on the void surfaces.[27]

As already mentioned above, the concentration of hydrogen bonded as isolated monohydride groups (SiH) is close to 6 at % and about the same for all the samples 1–5 (Fig. 4). On the contrary, the concentration of polyhydride $(SiH_2)_n$ complexes is clearly higher than that of the SiH groups by a factor varying from 2 to 4.5 in going from the the sample deposited at the highest $T = 300$ °C (series 5) to the sample deposited at the lowest $T = 100$ °C (series 1) (Fig. 4). The increase in the polyhydride groups from series 5 to series 1 is due to the increase in the concentration and size of the voids

and therefore to the increase in the fraction of disordered domains in the corresponding a-Si:H samples. It is only the fraction of these disordered domains in the amorphous network, which depends on the deposition conditions and especially the substrate temperature, that controls the concentration of bonded hydrogen as polyhydride and/or clustered monohydride groups. This important proportion of the $(SiH_2)_n$ groups in the amorphous network can be interpreted within the heterogeneous growth process where the layer during deposition can be represented by local disorder inhomogeneities (disordered domains around the surface of the voids and relaxed domains), as in the a-Si model proposed by Chehaidar et al.[19] From the point of view of solubility of hydrogen atoms in the amorphous network, the proportion of bonded hydrogen as $(SiH_2)_n$ and/or $(SiH)_n$ groups is the result of the presence of the fraction of disordered domains in the material. In relaxed domains, the more important role of the hydrogen atoms is to passivate Si dangling bonds, the concentration of which is small. This can explain the low hydrogen content bonded as isolated SiH groups. On the contrary, in the disordered domains, the hydrogen passivates not only the Si dangling bonds, for example at the surface of voids, the concentration of which is higher than in the relaxed domains, but also can break strained Si-Si bonds and then passivates the created Si dangling bonds. This leads to the formation of the polyhydride and/or clustered monohydride complexes, which may explain the higher solubility of the hydrogen atoms in the disordered domains than in the relaxed ones.

It is important to notice that it has been suggested previously that the fraction of the hydrogen bonded as $(SiH_2)_n$ and/or $(SiH)_n$ groups is controlled only by the total content of bonded hydrogen.[2,5] These authors assume that the sites in the amorphous network which are susceptible to giving rise to the Si-H bonding are distributed homogeneously whatever the preparation conditions are, which control only the amount of hydrogen bonded in the layer. This hypothesis is far from that of the real situation because, on the one hand, the amorphous network of a-Si:H layers is generally inhomogeneous, and, on the other hand, these inhomogeneities are strongly correlated with the deposition conditions.

C. Silicon-silicon bonding

Let us now consider the results of the analysis of the data obtained for the refractive index dispersion below the interband absorption edge, within Wemple-Didominico's model,[20] which depends on the nearest-neighbor atom and gives the complementary information of the results of the Raman scattering, which does appeal to the collective vibration of all atoms. We first start with the analysis of the results obtained for dispersion energy E_d $(E_d = \beta N_c Z N_e)$ for the series 1–5. The variation of E_d as a function of the substrate temperature is presented in Fig. 9(a). It clearly shows an increase of E_d as the substrate temperature increases (i.e., in going from series 1 to series 5). This behavior can be due to an increase of both the mean coordination number of the Si atoms in the amorphous network (i.e., increase in N_c) and the density of the films (i.e., increase of the effective number of valence electrons per Si atom N_e) when the substrate temperature increases. This result therefore suggests that increasing the temperature of deposition leads to more dense films. Indeed, this conclusion is clearly supported by the analysis of the variations with substrate temperature of the static refractive-index n_0 (refractive index at 0 eV) presented in Fig. 9(b), which shows an increase of n_0 with increasing substrate temperature. This result is in good agreement with previous studies showing that the increase of the density of the material is one of the parameters that leads to an increase of the static refractive index.[28] It is worth noticing at this stage that the increase of E_d and n_0 with increasing the temperature of deposition [Figs. 9(a) and 9(b), respectively] is well correlated with the decrease of the HWHM of the TA-like band. Indeed, this correlation is clearly suggested by Figs. 10(a) and 10(b), which show a linear variation of both the dispersion energy and the static refractive index, respectively, with the HWHM of the TA-like band. This comparison confirms clearly the effect of the porosity of the material on the HWHM of the TA-like band, as already emphasized above.
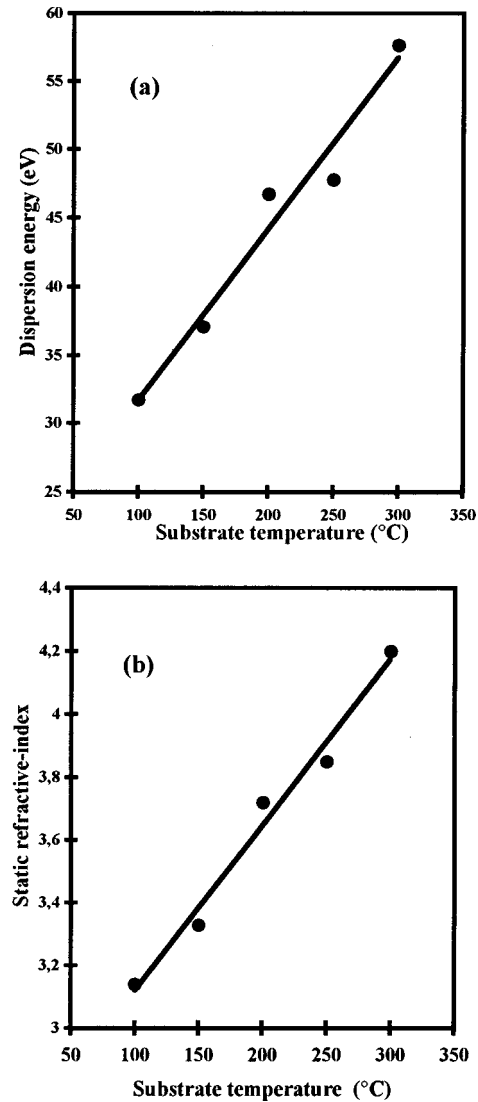

FIG. 9. Variation of the dispersion energy E_d (a) and the static refractive index n_0 (b) as a function of the substrate temperature for the same series 1–5.
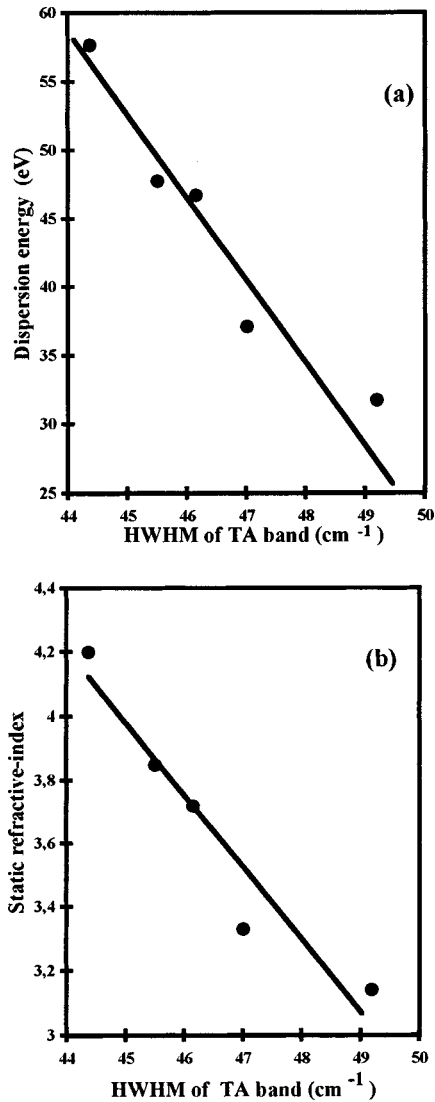
FIG. 10. Variation, as a function of the HWHM of the TA-like band, of the dispersion energy E_d (a) and the static refractive index n_0 (b) for series 1–5. Note that the lowest values of E_d and n_0 correspond to series 1 ($T = 100\,°C$) and the highest ones correspond to series 5 ($T = 300\,°C$).

D. Quantitative description of the growth process of a-Si:H

The last point we would like to discuss is the growth process of the films with respect to the whole results obtained for our particular samples. Whatever is the method used for the deposition of a-Si:H films, in which the hydrogen atoms are incorporated during deposition, the effect of the substrate temperature seems to be the most important parameter which controls the incorporation of hydrogen and therefore the density of states in the gap in a-Si:H.[29–31] Generally, high substrate temperature limits the hydrogen content incorporated, favors the formation of isolated monohydride groups, and leads to weak structural inhomogeneities. On the contrary, the deposition at low temperature favors the formation of polyhydride and/or cluster monohydride groups and leads generally to material involving high structural inhomogeneities.[23,32] The amorphous silicon network is indeed a metastable state from the thermodynamical the point of view. This metastability is due to the presence of the dangling bonds, the structural inhomogeneities, and the weakly bonded hydrogen such as $(SiH_2)_n$ and/or $(SiH)_n$ groups.[33] When the temperature of deposition increases, the amorphous network looks for another more stable state (but it remains nevertheless metastable) due to the collective motion of the number of atoms over a very short distance in the layer during the growth process. This process may reduce the density of the dangling bonds by the bond reconstruction mechanism as well as the concentration of the weakly bonded hydrogen. This appears therefore to decrease the porosity of the material. Despite the high hydrogen content incorporated in our films even at substrate temperature as high as 300 °C, this quantitative description of the growth process of a-Si:H layers accounts for the whole of our results.

IV. CONCLUSION

Infrared (IR) absorption measurements are combined with Raman scattering and optical transmission experiments to investigate in detail the direct relationship between the chemical heterogeneity of Si-H bonding and the structural inhomogeneities (linked with the presence of voids) of an amorphous silicon network in hydrogenated amorphous silicon films prepared by rf magnetron sputtering at increasing substrate temperature (T) from 100 to 300 °C. The results obtained from the IR experiments indicate that all the samples exhibit almost the same hydrogen content (about 6 at. %) incorporated as isolated monohydride (SiH) groups, as illustrated by the presence of the 2004 cm^{-1} component. As a consequence, the gradual decrease in the total hydrogen concentration (from 38.6 to 20.5 at. %) as T increases is essentially due to the decrease in the amount of polyhydride $(SiH_2)_n$ groups which absorb around 2086 cm^{-1}. A clear relationship is established between the changes in the Si-H bonding configurations related to the 2086 cm^{-1} component and the variation of structural inhomogeneities, as evidenced by Raman spectroscopy. The linewidth towards the low frequencies of the transversal acoustic (TA)-like band in Raman spectra decreases (from 49.2 to 44.3 cm^{-1}) as T increases. This decrease in the linewidth of the TA-like band is attributed to the decrease of the density of quasilocalized phonons in structural inhomogeneities (disordered domains around about the surface of voids) in the amorphous network. The correlation between IR and Raman results leads us to conclude that the $(SiH_2)_n$ groups are mainly located in structural inhomogeneities due to the presence of voids, where the formation of these complexes is favorable. This interpretation is corroborated by the variation of both the static refractive index, which measures the density of material, and the dispersion energy, which measures the mean coordination number of Si atoms and the effective number of valence electron per Si atom in the amorphous network. We observe an increase of both the static refractive index and the dispersion energy as T increases.

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