KrF excimer laser induced formation of thin crystalline silicon carbide layers

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Thin silicon carbide layers have been the subject of numerous works. Crystalline silicon carbide (SiC) is characterized by a refractory nature, a high thermal conductivity, a high electron mobility and a large band gap. Actually, SiC is one of the promising materials for high-temperature device applications. In this work, we investigate the excimer laser crystallization of thin amorphous silicon carbide films deposited by sputtering method onto single crystalline silicon. The crystallized samples were characterized with grazing incidence X-rays diffraction and infrared spectroscopy. The obtained results show the formation of α-SiC. The nature of SiC poly-type depends on the laser irradiation energy. The epitaxial growth of the α-SiC has been observed.

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1. Introduction

During last years numerous works have been concentrated on the synthesis of silicon carbide (SiC). This material is characterized by a refractory nature, a high thermal conductivity, a high electrons mobility and a large band structure gap. All these properties make silicon carbide an excellent and promising material for devices operating at high temperature, high power, and high frequency.

In order to elaborate thin amorphous silicon carbide films, several techniques have been proposed besides CVD which is the most used one [1-4]. The crystallization of these layers needs heat treatments at high temperature and longer time which are performed by conventional annealing.

Since 1978, numerous works have been devoted to laser crystallization of amorphous silicon thin layers. It is very attractive to extend a similar technology to the amorphous silicon carbide. A particular interest is related to the use of excimer laser to crystallize thin amorphous SiC layers [5,6].

In this work, we study the crystallization of amorphous silicon carbide films, deposited by DC magnetron onto single crystalline silicon, using a KrF excimer laser as source of thermal annealing. Samples have been analyzed by both grazing incidence X-rays diffraction and infrared spectroscopy. We show that crystallization has strong dependence on laser annealing conditions, such as laser energy and the number of laser pulses.

2. Experimental procedure

Amorphous silicon carbide thin films, of 1000 Å thickness, were deposited using a DC magnetron sputtering system onto (100) p-type silicon substrate heated at 300°C. Some of the deposition parameters were as follows: input power, 80W (power density, 1.8 $W.cm^{-2}$), the argon and hydrogen pressures were kept constants at respectively 10^{-3} and 2.10^{-3} mbar (0.2Pa).

The as-deposited samples were irradiated with KrF excimer laser (λ = 248 nm) with 20 ns pulse duration and fluence ranging from 160 to 720 mJ/cm^2. They were analyzed by means grazing incidence X-rays diffraction (XRD) and infrared spectroscopy. Crystal structure analysis was performed with D8 Advance Bruker AXS X-ray diffractometer with Cu Kα radiation. Infrared measurements were carried out using Thermo-Nicolet-Nexus spectrometer.

3. Results and discussion

3.1. XRD analysis

The structure of the laser irradiated amorphous SiC thin layers, onto (100) p-type silicon substrate, was characterized by grazing incidence X-rays diffraction. We note that the crystalline growth of the amorphous material depends on the nature and the orientation of substrate, and SiC exhibits a better crystalline order in the case of Si (100) single crystal [7-9]. From the literature [10,11], we can suggest that we have two types of crystallization:

a) solid phase crystallization for 160, 200 and 280 mJ/cm^2: these laser fluences are lower than melting threshold energy

b) liquid phase crystallization for 500 and 720 mJ/cm^2

3.1.1 Solid phase crystallization

Fig. 1 shows three XRD spectra corresponding to both the as-deposited SiC layer and the laser annealed samples with 160 and 200 mJ/cm^2 for ten laser shots. The as-deposited layer is amorphous; XRD spectrum exhibits no SiC diffraction peak. From X-ray spectra of the irradiated samples, one can see that crystallization has occurred. The presence of only one peak, at 2θ = 37.33°, is probably due to the partial crystallization. The used laser energies are not sufficient to crystallize all the amorphous silicon carbide. We note that from this result the type of SiC cannot be determined.

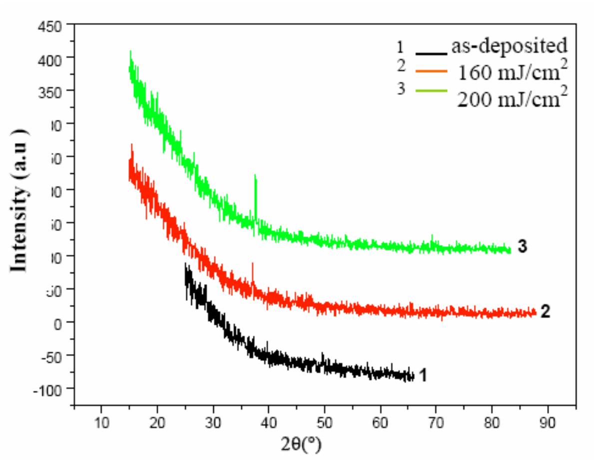

Fig. 1. XRD spectra of the as-deposited and irradiated samples with 160 and 200 mJ/cm^2 for 10 shots.

Increasing the laser energy to 280 mJ/cm^2 for 10 shots leads to an important disorder - order transition. Indeed, the X-ray spectrum (Fig. 2) indicates the presence of two peaks at 2θ = 49.75° and 2θ = 65.65°, corresponding to reflection plan (102) and (103) of crystalline 2H-SiC (JCPDS card 29-1126) or reflection plan (104) and (106) of crystalline 4H-SiC (JCPDS card 29-1127), which can be attributed to the increase in the crystallinity degree. We note that for the used laser energies (160, 200 and 280 mJ/cm^2) and for one laser shot, the irradiated samples remain amorphous. The solid phase crystallization of a-SiC layers takes place only after many shots. These results are very consistent with those obtained by P. Baeri et al. [10]. Indeed, P. Baeri has observed an amorphous–crystal solid phase transition after laser irradiation but with more than one order of magnitude in the time duration of the laser.

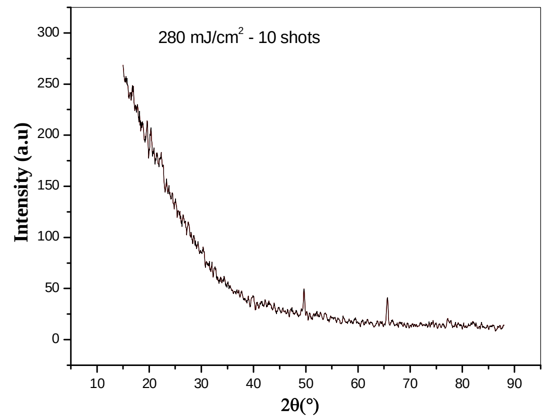

Fig. 2. XRD spectrum of the irradiated samples with 280 mJ/cm^2 for 10 shots.

3.1.2. Liquid phase crystallization

To crystallize thin amorphous silicon carbide layers, via liquid phase crystallization, necessitates higher laser energies. In our case, we have irradiated the a-SiC films with two laser energies 500 and 720 mJ/cm^2 for only one shot.

Fig. 3 shows the XRD spectrum of the sample annealed with 500 mJ/cm^2. One can observe an intense peak and very weak one at 2θ = 65.65° and 2θ = 49.75°, respectively. This indicates that the a-SiC layer was crystallized. From this XRD pattern, we can suggest that the crystallized SiC film is epitaxial. Increasing laser energy to 720 mJ/cm2 leads to highly textured polycrystalline SiC layer as can be observed in Fig. 4. XRD spectrum shows an intense peak at 2θ = 66.96° and two weak peaks at 2θ = 50.71° and 2θ = 78.91° corresponding to reflection plan (009), (007) and (204) of crystalline 5H-SiC (JCPDS card 42-1360). The formation of this poly-type is unexpected. To our knowledge 5H-SiC is not mentioned by the literature concerning the synthesis of crystalline SiC thin layers.

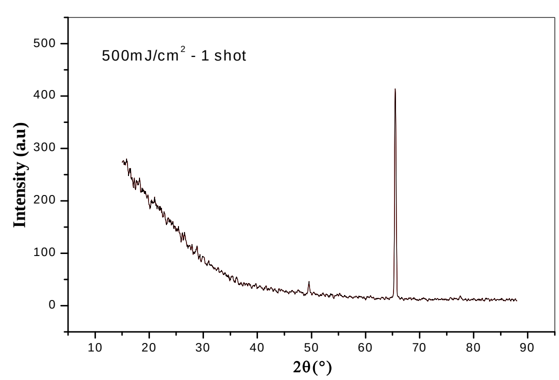

Fig. 3. XRD spectrum of the irradiated samples with 500 mJ/cm^2 for one shot.

At this stage of characterization and from these results, one can conclude that the formed crystalline SiC is stoechiometric. R. Reitano and al. [12] have shown that SiC exhibits a peritectic decomposition at 2840 K. We have obtained this peritectic decomposition in the case of our samples irradiated with laser fluence higher than 1 J/cm^2.

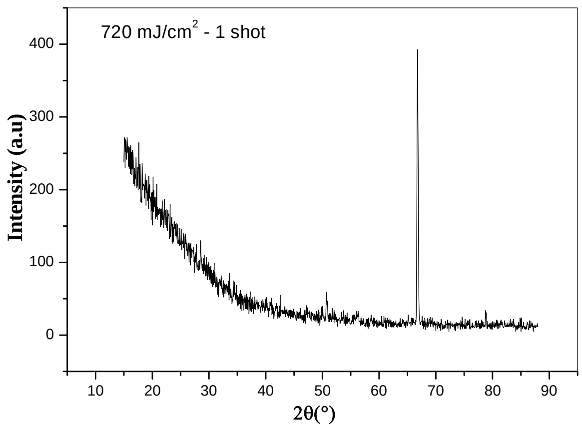

Fig. 4. XRD spectrum of the irradiated samples with 720 mJ/cm^2 for one shot.

3.2. Infrared analysis

The spectrum of the as-deposited sample (Fig. 5) exhibits broad peak at around 730 cm^{-1} related with the Si-C stretching vibration mode [13], due to the fluctuations in the bond length and bond angle in the amorphous phase. This spectrum is of Gaussian type, characteristic of the amorphous state. Some works consider that the absorption band at around 730 cm^{-1} corresponds to Si-C transitional phase in the case of $Si_{1-x}C_x$ alloy (x<0.5) [14,15].

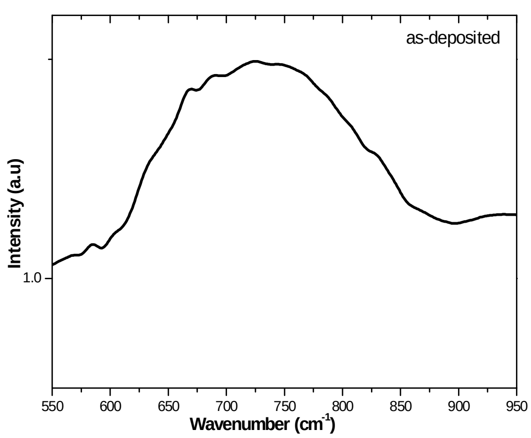

Fig. 5. Infrared spectrum of the as-deposited layer.

After laser annealing, the peak assigned to Si-C bond shifts towards 840 cm^{-1} (Fig. 6). This value is close to the one reported for the stretching mode of Si–C bonds in crystalline SiC [13,16-18].

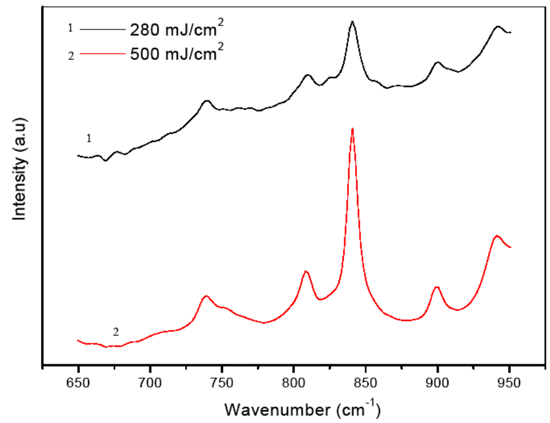

Fig. 6. Infrared spectra of the laser irradiated samples with 280 and 500 mJ/cm^2 for 10 shots and one shot, respectively.

One can see that the peak width is considerably decreased and peak becomes more intense in the case of 500 mJ/cm^2 laser energy. The corresponding infrared spectrum is of Lorentzian type indicating that laser irradiation has induced crystallization of the amorphous silicon carbide layer [19-21]. For 500 mJ/cm^2 narrowing of the band at 840 cm^{-1} and the increase in intensity attest that an important crystallization has occurred.

4. Conclusions

We have investigated the KrF laser crystallization of amorphous silicon carbide thin layers deposited by DC magnetron onto (100) p-type silicon substrate.

The most important results are as follows:

- For the lower laser fluence and after one shot the sample is still amorphous
- The increase of shots number (10 shots) induces the transition from amorphous to crystalline phase.
- Important crystalline growth of SiC is observed after liquid phase crystallization
- Nature of SiC poly-type depends on the laser fluence.

References

[1] A. Feng, Z. A. Munir, J. Appl. Phys. **76**, 1927 (1994).
[2] H. Matsumani, Mater. Sci. Forum **338–342,** 125 (2000).
[3] K. Fujihira, T. Kimoto, H. Matsunami, J. Cryst. Growth **255**, 136 (2003).
[4] T. Takahashi, Y. Ishida, H. Tsuchida, I. Kamata, H. Okumura, S.Yoshida, K. Arai, Mater. Sci. Forum **389–393,** 323 (2002).
[5] H. Hobert, H. H. Dunken, S. Urban, F. Falk, H. Stafast, Vibra. Spectrosc. **29**, 177 (2002).
[6] A. Hedler, S. Urban, F. Falk, H. Hobert, W. Wesch, Appl. Surf. Sci. **205**, 240 (2003).

[7] P. Zanolaa, E. Bontempi, C. Ricciardi, G. Baruccac, L.E. Deperoa, Mater. Sci. Eng. B **114–115**, 279 (2004)
[8] H. Schmidt, E.R. Fotsing, G. Borchardt, R. Chassagnon, S. Chevalier, M. Bruns, Appl. Surf. Sci. **252**, 1460 (2005).
[9] K.A. Jones, J. Cryst. Growth **310**, 2417 (2008)
[10] P. Baeri, C. Spinella, R. Reitano, Inter. J. Thermophys. **20**(4), 1211 (1999)
[11] C. Dutto, E. Fogarassy, D. Mathiot, Appl. Surf. Sci. **184**, 362 (2001)
[12] R. Reitano, P. Baeri, Inter. J. Thermophys. **17**, 1079 (1996).
[13] J. A; Boarders, S. T; Picraux, W. Beezhold, Appl. Phys. Lett. **18**, 509 (1971).
[14] C. Than, X. L. Wu, S. S. Deng, G. S. Huang, X. N. Liu, X. M. Bao, Phys. Lett. A **310**, 236 (2003)
[15] Z. D. Sha, X. M. Wu, L. J. Zhuge, Vacuum **79**, 250 (2005).
[16] T. Kusumori, H. Muto, M.E. Brito, Appl. Phys. Lett. **84**, 1272 (2004).
[17] W. G. Spitzer, D. Kleinman, D. Walsh, Phys. Rev. **113**, 127 (1959)
[18] W. G. Spitzer, D. Kleinman, C. J. Frosch, Phys. Rev. **1113**, 133 (1959)
[19] T. Rajagopalan, X. Wang, B. Lahlouh, C. Ramkumar, P. Dutta, S. Gangopadhyay, J. Appl. Phys. **94**(8), 5252 (2003)
[20] M. Dkaki, L. Calcagno, A.M. Makthari, V. Raineri, Mater. Sci. Semicond. Process. **4**(1–3), 201 (2001).
[21] D. Gracin, K. Juraic, P. Dubcek, A. Gajovic, S. Bernstorff, Vacuum **80**(1-3), 98 (2005)

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