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Intrinsic stress analysis of sputtered carbon film

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Intrinsic stresses of carbon films deposited by direct current (DC) magnetron sputtering were investigated. The bombardments of energetic particles during the growth of films were considered to be the main reason for compressive intrinsic stresses. The values of intrinsic stresses were determined by measuring the radius of curvature of substrates before and after film deposition. By varying argon pressure and target-substrate distance, energies of neutral carbon atoms impinging on the growing films were optimized to control the intrinsic stresses level. The stress evolution in carbon films as a function of film thickness was investigated and a void-related stress relief mechanism was proposed to interpret this evolution.

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Carbon films are ideal total-reflection mirrors for energies below 200 eV[1] with more than 90% reflectivity at grazing incident angle of 4°. They are widely used in synchrotron radiation beam-lines as order sorters to suppress higher order light[2]. However, the high level intrinsic stresses developed in sputtered carbon films degrade the mechanical properties, limit monolayer's maximum thickness, cause poor adhesion to substrate, and lead to the main limitation for their practical applications. Numerous models and theories have been explored to explain the structure-related compressive stresses in sputtered carbon films. Atom-peening model proposed by d'Heurle[3,4] appears to be appropriate for carbon films deposited at relatively low argon pressure. It proposes that energetic particles impinge on the surface of the growing films and insert into the films, displace atoms from equilibrium positions through a series of primary and recoil collisions, increase packing density, change microstructures and cause volumetric distortion, at last these processes result in compressive stresses in films[5,6].

It is well known that atom-peening effect depends considerably on deposition parameters[7]. In this letter, the residual stresses in carbon films were investigated as functions of sputtering gas pressure and target-substrate distance. Optical performance of the fabricated carbon films was measured at Hefei National Synchrotron Radiation Laboratory (NSRL). The simulated packing densities match our prediction according to atom-peening model. Then the stress which was dependent on film thickness was investigated and explained with self-releasing mechanism.

Intrinsic stress was calculated by Stoney's equation, when $t_{\mathrm{s}} \ll t_{\mathrm{f}}$:

$$\sigma = \frac{E_{\mathrm{s}}}{6(1-\nu_{\mathrm{s}})}\left(\frac{t_{\mathrm{s}}^2}{t_{\mathrm{f}}}\right)\left(\frac{1}{R}-\frac{1}{R_0}\right), \qquad (1)$$

where E_{s} and ν_{s} are Young's modulus and Poisson's coefficient of the substrate, t_{s} and t_{f} are the thicknesses of the substrate and film, and R_0 and R are the radius of curvature of the substrate before and after film deposition, respectively. The radius of curvature was measured with a Dektak 6M stylus profiler produced by Veeco Company. All the substrates were measured twice in orthogonal directions (C-direction 1,2).

Carbon films were deposited on grounded glass substrates at room temperature by direct current (DC) magnetron sputtering[8,9] from a graphite target in pure argon discharges. The argon pressure varied from 1 to 5 mTorr and the target-substrate distance changed from 8 to 12 cm at fixed sputter power of 120 W. Stresses of 30-nm-thick carbon films deposited at different argon pressures are shown in Fig. 1. We can see that the stress is strongly dependent on argon pressure. But the variation of target-substrate distance has little influence on intrinsic stress that just changes from 1.87 to 1.65 GPa as the distance increases from 8 to 12 cm. The energetic particles bombarding the surface of growing films are essentially neutral carbon atoms ejected from the target and argon ions from the plasma. The contribution of neutral carbon atoms to the peening effect is significantly higher than that of argon ions, in particular at low argon pressures[10]. The level of compressive stresses is determined by flux and energies of neutral carbon atoms. The kinetic energies of ejected carbon atoms decrease progressively because of the inter-atomic collisions in the gas phase during the movement from target to substrate. As the argon pressure increases, the mean free path of atoms decreases and the collision possibilities increase,

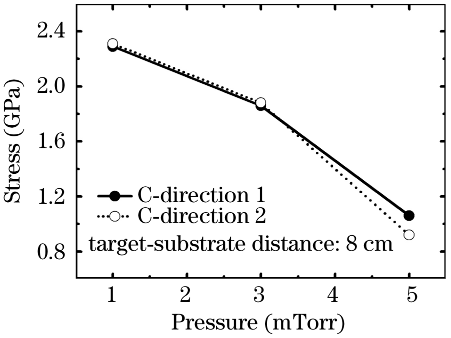

Fig. 1. Effect of argon pressure on intrinsic stress.

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and the sputtered carbon atoms lose more kinetic energies. As a result, the compressive stresses in carbon films decrease. Increasing target-substrate distance also leads to the increase of collisions to some extent, but it is not so effective in limited region.

Based on the atom-peening model, the packing densities have close relationship with intrinsic stresses in carbon films. The bombardments of higher energetic carbon atoms simply result in higher intrinsic stress and higher packing density. We can compare these values to get more reliable results. We firstly calculated the reflectivity of carbon films at different densities with IMD software[11], and then compared the measured reflectance curves with the calculation results. Films fabricated at argon pressure of 1, 3, and 5 mTorr correspond to packing densities of 2.05, 1.9, and 1.85 g/cm^3, as shown in Fig. 2. It is shown that lower compressive stresses are in accordance with lower packing densities. By increasing pressure, the intrinsic stress can be reduced but on cost of optical performance.

In order to study the stress evolution with the film thickness, a series of carbon films were fabricated with the thicknesses from 5 to 100 nm. Figure 3 shows the dependence of intrinsic compressive stress on film thickness. There are two distinct stress variation regions for carbon films. When the thickness is less than 30 nm, compressive stress increases rapidly. Then the stress keeps almost constant around 2.2 GPa with the thickness increasing from 30 to 100 nm. Atom-peening effect is the main contribution to compressive stresses in carbon films, while the existence of voids in amorphous carbon films plays an important role in releasing the compressive stress[12]. Highly porous structures lead to tensile stresses[13], and a lower density of voids may act as a stress-relief mechanism to a compressively stressed film. The stress evolution in carbon films is the interaction of these two effects. At the first region, the film is not thick enough to contain significant stress-relieving voids. Atom-peening effect contributes to the progressive increase of compressive stress. As the film grows thicker, the chance of creating voids increases a lot. When the void-related stress releasing balances the stress increased by atoms bombarding, the compressive stress keeps almost constant, as shown in Fig. 3.

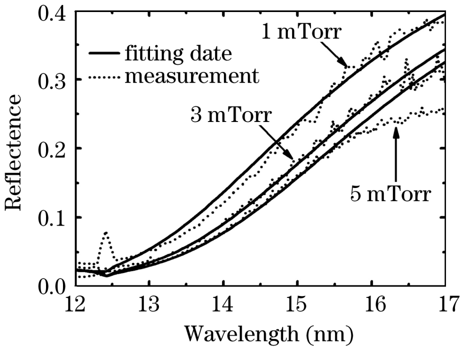

Fig. 2. Reflectance of carbon films at grazing incident angle of 5° measured at NSRL.

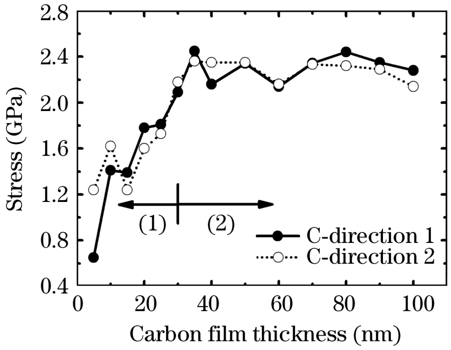

Fig. 3. Variation of intrinsic stress as a function of film thickness.

In summary, intrinsic stresses of carbon films depend on the sputtering parameters such as sputtering gas pressure and target-substrate distance. Intrinsic stress can be greatly reduced with the increase of argon pressure. According to the reflectivity curves measured at NSRL, the relationship between intrinsic stress and film density is established. The stress evolution in carbon films is investigated as the function of film thickness. The interaction of the atom-peening effect with void-related stress relief mechanism determines the stress level in carbon films. To fabricate durable carbon films without degrading too much optical performance, a thin interlayer will be used to improve the poor adhesion between the carbon film and substrate in future work.

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References

1. S. Jacobi, B. Steeg, J. Wiesmann, M. Störmer, J. Feldhaus, R. Bormann, and C. Michaelsen, Proc. SPIE **4782,** 113 (2002).
2. http://www-cxro.lbl.gov/als6.3.2/.
3. F. M. d'Heurle, Metall. Teans. **1,** 725 (1970).
4. F. M. d'Heurle and J. M. E. Harper, Thin Solid Films **171,** 81 (1989).
5. H. Windischmann, J. Appl. Phys. **62,** 1800 (1987).
6. C. A. Davis, Thin Solid Films **226,** 30 (1993).
7. E. Mounier and Y. Pauleau, J. Vac. Sci. Technol. A **14,** 2535 (1996).
8. F. Wang, Z. Wang, J. Zhu, Z. Zhang, W. Wu, S. Zhang, and L. Chen, Chin. Opt. Lett. **4,** 550 (2006).
9. Z. Wang, S. Zhang, W. Wu, J. Zhu, H. Wang, C. Li, Y. Xu, F. Wang, Z. Zhang, L. Chen, H. Zhou, and T. Huo, Chin. Opt. Lett. **4,** 611 (2006).
10. E. Mounier and Y. Pauleau, Diamond. Relat. Mater. **6,** 1182 (1997).
11. D. L. Windt, Computers in Physics **12,** 360 (1998).
12. D. Henderson, M. H. Brodsky, and P. Chaudhari, Appl. Phys. Lett. **25,** 641 (1974).
13. O. Durand-Drouhin and M. Benlahsen, Solid State Commun. **131,** 425 (2004).