IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

Electroluminescence from an Au - extra-thin silicon oxynitride film - Si structure

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1996 J. Phys.: Condens. Matter 8 L223

(http://iopscience.iop.org/0953-8984/8/14/001)

View the table of contents for this issue, or go to the journal homepage for more

Download details:
IP Address: 171.66.16.208
The article was downloaded on 13/05/2010 at 16:28

Please note that terms and conditions apply.

J. Phys.: Condens. Matter **8** (1996) L223–L228. Printed in the UK

LETTER TO THE EDITOR

Electroluminescence from an Au–extra-thin silicon oxynitride film–Si structure

A P Li†, L D Zhang†, Y X Zhang†, G Qin‡ and G G Qin†§

† Department of Physics, Peking University, Beijing 100871, People's Republic of China
‡ CCAST (World Laboratory), PO Box 8730, Beijing 100080, People's Republic of China, and Department of Physics, Nanjing University, Nanjing 210008, People's Republic of China
§ International Centre for Materials Physics, Academia Sinica, Shenyang 110015, People's Republic of China

Received 2 February 1996

Abstract. The electroluminescence (EL) at room temperature from Au–extra-thin silicon oxynitride (ETSON)–p-Si and Au–ETSON–n-Si structures is reported. The ETSON films (∼80 Å) were deposited by the rf magnetron sputtering technique, and Si_3N_4–Si composite targets were used (the area ratio of Si to Si_3N_4 was ∼6%). The EL spectra were measured under forward bias $\geqslant$ 4 V after the ETSON films had been annealed at 300, 600, 800, and 1000 °C in N_2 ambient for 30 min. The Au–ETSON–p-Si structure has a dominant EL band with peak wavelength around 680 nm, and its integrated EL efficiency is an order of magnitude higher than that from a Au–extra-thin Si-rich SiO_2 (ETSSO) film (∼80 Å)–p-Si structure. Distinguishing it from the Au–ETSSO–Si structure, which emits visible EL only when it is fabricated on p-Si, EL can also be observed in the Au–ETSON–n-Si structure under forward bias, and the two dominant EL bands peak one at around 700 nm and one at around 800 nm. Electron beam irradiation induces a new EL band peaked at 500 nm in the EL spectrum from an Au–ETSON–p-Si structure. From the experimental results it is suggested that electrons and holes from opposite sides tunnel into the ETSON layer and recombine radiatively at luminescence centres there.

The visible electroluminescence (EL) from Au–extra-thin Si-rich SiO_2 (ETSSO)–Si structures has been reported in our previous works [1]. Therein we studied the EL dependence on the forward bias and the effects of thermal annealing, and attributed light emission to radiative recombination of electrons and holes which tunnel from opposite sides into the SiO_2 layers and recombine radiatively at the luminescent centres there. Besides luminescent centres, the nanosize Si particles also play a key role, since they shorten the distance of carrier tunnelling and affect the band offsets on the interface of nanosize Si particles and SiO_2. We consider that the replacement of the silicon oxide by silicon nitride in the MIS structure may lead to EL with different emission efficiency or different peak energies because the concentration and the types of luminescent centres and the size distribution of nanosize Si particles have been changed. Amorphous silicon nitride films are traditionally used in the microelectronics industry [2–6] as oxidation masks, dopant diffusion barriers, gate dielectrics in field effect and thin-film transistors, charge storage layers in nonvolatile metal–nitride–oxide–semiconductor memory devices, and as final passivation layers for device packaging. All these applications are based on the excellent insulating properties of silicon nitride. There are some reports on optical property studies of amorphous silicon nitride, including photoluminescence (PL) [7–12], photoluminescence excitation (PLE) [13, 14], and luminescence decay [15]. However, few studies have been reported on its EL characteristics and mechanism. Boonkosum *et al* [16]

0953-8984/96/140223+06\$19.50 © 1996 IOP Publishing Ltd

applied hydrogenated amorphous silicon nitride (a-SiN:H) for the first time as a luminescent active layer in a light-emitting diode (LED) with a structure of glass–ITO–p a-SiC:H–i a-SiN:H(250–1000 Å)–n a-SiC:H–Al. Recently, Deshpande *et al* [12] have reported weak EL from a structure of ITO–a-SiN:H(1000 Å)–Si during their extensive studies on the optical properties of a-SiN:H films deposited by hot-filament chemical vapour deposition.

In this letter, we report the visible EL from Au–extra-thin silicon oxynitride (ETSON)–Si structures, where the thicknesses of ETSON films are only ~80 Å, far less than those reported in [12] and [15], compare the EL behaviour from Au–ETSON–Si and Au–ETSSO–Si structures, study the effects of electron beam irradiation on EL spectra, and discuss the EL mechanism of the structures.

The substrates used in this experiment were (100) oriented, 5–9 Ω cm, p-type or n-type Si wafers. After the Si wafers had been cleaned carefully, the native oxide layers on Si were removed by HF solution. Before the deposition of dielectric films, an ohmic contact on the back side was formed by evaporating thin Al film and alloyed at 530 °C. The ETSON films were deposited by an rf magnetron sputtering technique which is similar to the deposition of ETSSO films reported in our previous paper [1]. The base vacuum was around 5×10^{-5} Pa and the deposition of the film was carried out in argon (~2 Pa). The only alteration is that Si–Si_3N_4 composite targets instead of Si–SiO_2 were used. The area ratio of Si to Si_3N_4 was about 6%. The thickness of deposited films was around 80 Å and they were annealed at 300, 600, 800, and 1000 °C in N_2 ambient for 30 min. Finally, semitransparent Au films were evaporated onto ETSON films to form Au–ETSON–p-Si and Au–ETSON–n-Si structures. The area of Au film for each diode is about 3.14 mm^2.

The chemical composition analysis by x-ray photoelectronic spectrometry (XPS) and Auger electronic spectrometry (AES) shows that besides N and Si, the ETSON films also contain O and C atoms. The atom number ratio of N, O, and Si is about 1:2:3. The large amounts of O introduced into the ETSON film may be attributed to the fact that the base vacuum (~5 $\times 10^{-5}$ Pa) in the magnetron sputtering system is not high enough, and the thickness of deposited films is extra-thin. Though our purpose is to deposit extra-thin Si-rich Si_3N_4 films, the dielectric films obtained are actually extra-thin silicon oxynitride films.

The I–V measurements indicate that all the diodes show good rectifying junction behaviour for both the Au–ETSON–p-Si and Au–ETSON–n-Si structures, but the series resistance is around 1–2 kΩ, which is around two to four times that of Au–ETSSO–Si diodes.

It has been observed that after annealing ETSON films at 300–1000 °C both the Au–ETSON–p-Si and Au–ETSON–n-Si structures can emit visible light under forward bias $\geqslant$ 4 V (for Au–ETSON–p-Si structures positive voltage is applied to the p-Si substrate, and for Au–ETSON–n-Si structures positive voltage is applied to the Au film). Figure 1 shows the EL spectra from Au–ETSON–p-Si structures after annealing at different temperatures, 300, 600, 800, and 1000 °C, in N_2 ambient. If the ETSON film is not exposed to any thermal annealing, no EL can be observed in Au–ETSON–p-Si structures; however, the EL from an Au–ETSSO–p-Si structure without any annealing can be clearly observed by the naked eye and its EL intensity is higher than those of the annealed structures. An EL spectrum from an Au–ETSSO(80 Å)–p-Si structure without annealing is also shown in figure 1 for comparison. It can be seen that the EL spectra from the Au–ETSON–p-Si structure have a dominant peak at ~680 nm accompanied by a shoulder at around 820 nm. The ~680 nm peak occurs also in the Au–ETSSO–p-Si structure. With further increase of annealing temperature from 300 to 600 °C, the EL intensity increases but the peak energy has no evident shift. However, when the annealing temperature increases from 600 to 800 and 1000 °C, the EL intensity decreases rapidly, and the peak energy has a slight blue shift.

Figure 2 shows the EL dependence on forward bias for the Au–ETSON–p-Si structure where the ETSON film has been annealed at 300 °C. With increasing forward bias, the EL

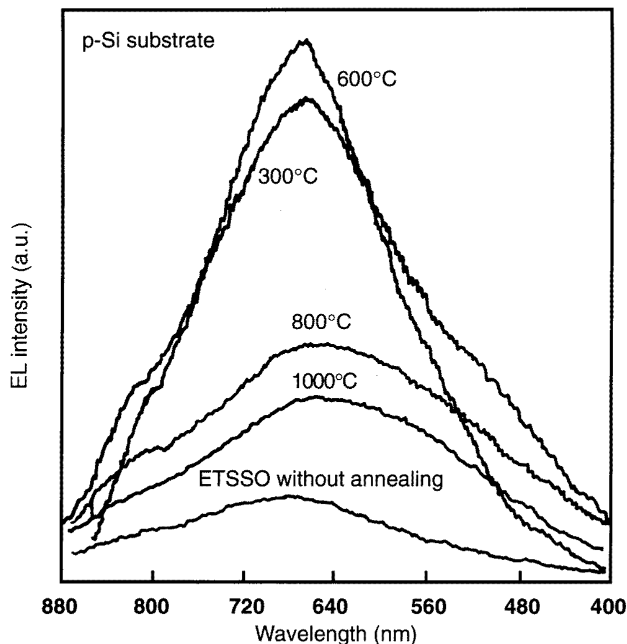

Figure 1. EL spectra of Au–ETSON–p-Si diodes after thermal annealing at the different temperatures indicated, and an Au–ETSSO–p-Si diode without any thermal annealing measured under a forward bias of 8 V. The thicknesses (∼80 Å) and Si area ratios in the composite targets (6%) are almost the same for ETSON and ETSSO films.

intensity increases but the peak wavelength has no evident shift.

Figure 3 gives the relationship of the EL integrated intensity from an Au–ETSON–p-Si diode to input electric power, and its comparison with that from an Au–ETSSO–p-Si diode, where both the ETSON and ETSSO films have been annealed at 300 °C. It can be found that the EL integrated intensity increases almost linearly with input electric power, but the efficiency for an Au–ETSON–p-Si structure is an order of magnitude higher than that for an Au–ETSSO–p-Si structure.

Figure 4 shows the EL spectra from an Au–ETSON–n-Si structure under forward bias, where the ETSON film has been annealed at 300 °C. It is evident that the spectra comprise at least two bands peaked one at around 700 nm and the other at around 820 nm. On the other hand, no EL has been observed in the Au–ETSSO–n-Si structure under either forward bias or reverse bias.

The dependence of EL peak energy on the annealing or bias is similar for both Au–ETSON–p-Si and Au–ETSSO–p-Si structures. This implies that the EL mechanisms are the same for these two types of structure. In our previous paper [1], we proposed that the electrons and the holes from opposite sides tunnelled into the dielectric layers and radiatively recombined via the luminescent centres there. In order to examine whether the present EL can be attributed to radiative recombination via the luminescent centres in the dielectric layers and clarify the different effects between silicon nitride and silicon oxide, we have studied the effects of electron beam irradiation on the EL spectra.

The electron beam irradiation experiments were carried out in a Hitachi S-450 SEM. The electron beam current is around 10 nA and the accelerating voltage is 30 keV. The vacuum in the chamber during irradiation was maintained at $\sim 10^{-4}$ Pa. Figure 5 shows

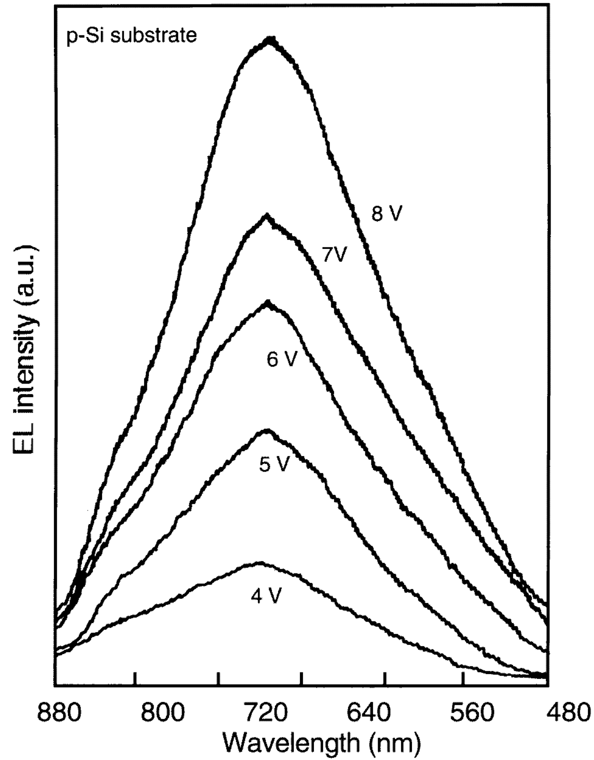

Figure 2. The EL dependence on forward bias for a diode with an Au–ETSON–p-Si diode, where the ETSON film has been annealed at 300 °C.

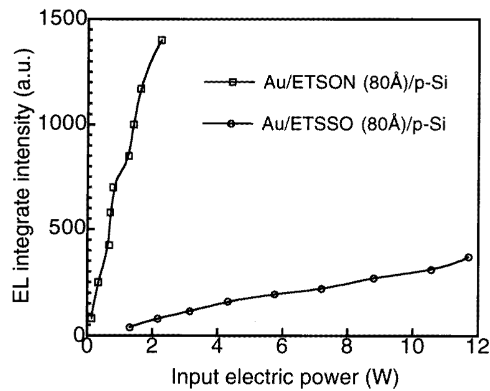

Figure 3. The variation of EL integrated intensity with input electric power for Au–ETSON(∼80 Å)–p-Si and Au–ETSSO(∼80 Å)–p-Si structures, where both the ETSON and ETSSO films have been annealed at 300 °C.

the electron beam irradiation time dependence of the EL spectra from an Au–ETSON–p-Si structure, where the ETSON film has been annealed at 300 °C. The intensity of the 680 nm band has no evident increase after the film has been irradiated for 10 or 20 min; however,

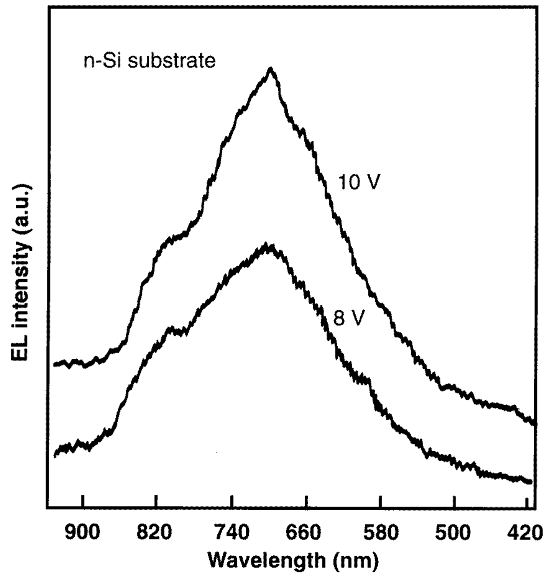

Figure 4. EL spectra of an Au–ETSON–n-Si diode with the ETSON film annealed at 300 °C measured under forward biases of 8 and 10 V.

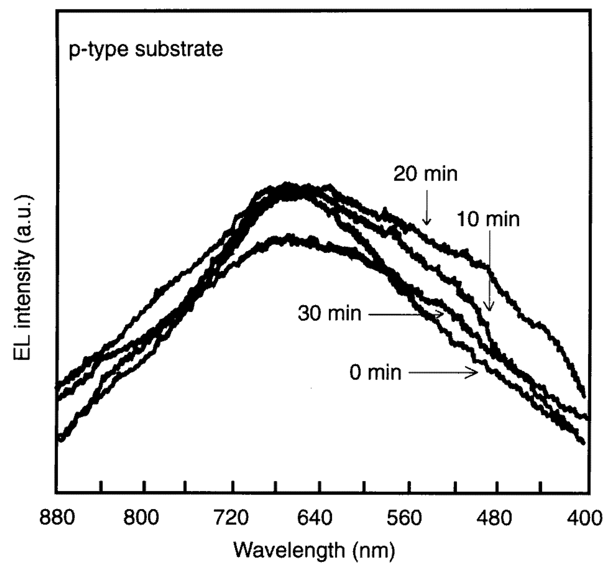

Figure 5. EL spectra from an Au–ETSON–p-Si diode having been irradiated by an electron beam (30 keV) for various irradiation times, where the ETSON film has been annealed at 300 °C.

it decreases obviously after irradiation for 30 min. On the other hand, the irradiation leads to the appearance of a 500 nm band, which reaches the maximum intensity after 20 min irradiation.

It is well known that electron bombardment can induce defects: for example, it causes the nonbridging oxygen hole centre (NBOHC) in silicon oxide [17, 18]. The NBOHC may

be responsible for the red PL emission band in porous silicon [19, 20]. Suzuki *et al* [18] reported that electron beam irradiation caused an increase of the 1.9 eV cathodoluminescence band in porous silicon, and our work which will be reported elsewhere has shown that electron beam irradiation causes an increase of the 1.9 eV EL band in the Au–ETSSO–Si structure. However, in the present work, a new 500 nm (2.5 eV) band appears instead of an increase of the 680 nm (1.82 eV) peak after irradiation. Therefore, it is thought that both the 680 and 500 nm bands presented in this paper are attributed to the defects in silicon oxynitride rather than those in SiO_2.

The PL bands at around 1.76 eV (700 nm) and 1.82 eV (680 nm) have been observed by Lin and Lee [11] in a-SiN:H films deposited by plasma-enhanced chemical vapour deposition. A 1.8 eV (690 nm) PL peak and a broad band peaked at 2.5 eV (500 nm) were observed by Deshpande *et al* [12] in their silicon nitride films prepared by hot-filament chemical vapour deposition. They regarded the light emission as being related to the optically active defects. The present EL peaks at $\sim$680 nm (1.82 eV) and 820 nm (1.56 eV) in the Au–ETSON–p-Si structure, or 700 nm (1.76 eV) and 820 nm (1.56 eV) in the Au–ETSON–n-Si structure, are very close to the above PL peak wavelengths, but the preparation methods of silicon nitride films are very different from each other. Combining this with the fact that the 2.5 eV band in the Au–ETSON-p-Si structure induced by electron beam irradiation has also its corresponding PL band in the silicon nitride film [12], we consider that both the EL of Au–ETSON–Si structures and the PL of silicon nitride originate from the luminescent centres in dielectric films, though their carrier injection processes are different.

In summary, the visible EL spectra have been observed in Au–ETSON–Si structures. The EL efficiency of the Au–ETSON–p-Si structure is an order of magnitude higher than that of the Au–ETSSO–p-Si structure. The EL behaviour and its dependence on forward bias and electron beam irradiation indicate that the EL emissions originate from the radiative recombination in the luminescence centres of silicon oxynitride.

References

[1] Qin G G, Li A P, Zhang B R and Li B C 1995 *J. Appl. Phys.* **78** 2006

[2] Taft E A 1971 *J. Electrochem. Soc.* **118** 1341

[3] Watanabe H, Katoh K and Yasui M 1982 *Japan. J. Appl. Phys.* **21** L341

[4] Lucovsky G, Yang J, Chao S S, Tyler J E and Czubaty W 1983 *Phys. Rev.* B **28** 3234

[5] Tsu D V, Lucovsky G and Mantini M J 1986 *Phys. Rev.* B **33** 7069

[6] Yin Z and Smith F W 1990 *Phys. Rev.* B **42** 3666

[7] Austin I G, Jackson W A, Searle T M, Bhat P K and Gibson R A 1985 *Phil. Mag.* B **52** 271

[8] Chen D, Viner J M, Taylor P C and Kanicki J 1992 *Mater. Res. Soc. Symp. Proc.* vol 258 (Pittsburgh, PA: Materials Research Society) p 661

[9] Pundur P A, Shavaigm J G and Gritsenko V A 1986 *Phys. Status Solidi* a **94** K107

[10] Vasilev V V and Mikhailovskii J P 1985 *Phys. Status Solidi* a **90** 355

[11] Lin K C and Lee S C 1992 *J. Appl. Phys.* **72** 5474

[12] Deshpande S V, Gulari E, Brown S W and Rand S C 1995 *J. Appl. Phys.* **77** 6534

[13] Vasilev V V, Mikhailovskii J P and Svitashev K K 1986 *Phys. Status Solidi* a **95** K37

[14] Jackson W A, Searle T M, Austin I G and Gibson R A 1985 *J. Non-Cryst. Solids* **77&78** 909

[15] Palsule C, Gangopadhyay S, Yi S, Shen J M, Naseem H A, Kizzar S and Goh F H C 1993 *Mater. Res. Soc. Symp. Proc.* vol 284 (Pittsburgh, PA: Materials Research Society) p 83

[16] Boonkosum W, Kruangam D and Panyakeow S 1993 *Japan. J. Appl. Phys.* **32** 1534

[17] Koyama H 1980 *J. Appl. Phys.* **51** 2228

[18] Suzuki T, Sakai T, Zhang L and Nishyama Y 1995 *Appl. Phys. Lett.* **66** 215

[19] Qin G G and Jia Y Q 1993 *Solid State Commun.* **86** 559

[20] Prokes S M 1993 *Appl. Phys. Lett.* **62** 3244