Extended Abstracts of the 17th Conference on Solid State Devices and Materials, Tokyo, 1985, pp. 313-316

Invited

Ionized Cluster Beam Epitaxy of Single Crystalline Aluminum Films on Semiconductors and Insulators as an Approach to New Device Structures

I. Yamada, H. Usui, H. Inokawa, and T. Takagi

Ion Beam Engineering Experimental Laboratory, Kyoto University

Sakyo, Kyoto 606, Japan

Ionized Cluster Beam (ICB) deposition offers the capability of introducing useful kinetic energy and chemical activity during the film formation process. Epitaxial Al films on Si substrates were obtained at room temperature by ICB. Experimental results showed that ICB deposition leaves hardly any damages on the substrate. The epitaxial Al films have excellent thermal and electrical stability. No degradation in morphological, crystallographical and electrical characteristics was found by annealing to temperatures up to 550°C. ICB was used to form epitaxial Al on GaAs, sapphire and also for epitaxial CaF_2 on Si. The deposition of epitaxial Al on epitaxial CaF_2 on Si substrates by ICB demonstrates the possibility of constructing all epitaxial metal/insulator/semiconductor system, which will bring about high performance solid state devices.

1. Introduction.

The possibility for the advancement of solid state devices comes mainly from the new device fabrication technologies that have evolved in the last few years. These technologies include not only those that can change the device size and the quality of the materials, but those that can modify the chemical, structural and physical properties of the material. The emerging film deposition technologies that involve ions have been preparing new solid materials and improving the properties of established materials by controlling deposition processes. These film deposition technologies offer higher energy for the arriving species but have, at present time, limitations for many applications. The energy an ion beam should have to enhance crystal growth and to promote chemical reactions is in the range from a few tenths of an eV per atom, i.e. above-thermal energy, to a few tens of eV per atom [1].

Ionized Cluster Beam (ICB) offers the capability of introducing useful energy into the film formation process at low substrate temperature without limiting the deposition rate due to space charge effects. The effects of acceleration voltage upon film formation through the influence of adatom migration, nucleation center density, sticking coefficient and enhancememt of chemical reaction have already been demonstrated [2]. Experimental results have also clearly shown that the kinetic energy of the accelerated clusters can be sufficiently low to leave no damage on the substrate surface [3]. In ICB deposition the beam provides tight control of the kinetic energy and ion content which distinguishes this technique from other deposition methods, such as MBE, VPE and LPE, which involve neutral atomic vapors. Unique advantages of ICB can be demonstrated by low temperature epitaxy and deposition of many kinds of materials. Examples include Al epitaxy on Si [4].

Aluminium metallization has been widely used for contact electrodes and interconnects in silicon semiconductor devices. Many problems associated with electromigration lifetime, contact stability, corrosion resistance, and multilevel capability have frequently arisen [5,6]. Hence, it is important to establish the quantitative limits of the Al/Si systems.

This paper will emphasize characteristics of epitaxial Al films on Si and the formation of epitaxial Al films on GaAs and dielectric materials. Our experimental results show that Al metallization problems depend critically upon the film structure (primarily grain size and orientation) and the interface. These properties are not precisely controlled under any conventional deposition technique. Study of epitaxial Al permits separating those limits intrinsic to Al itself from the ones related to a specific deposition technique. Achieved results lead toward realization of device miniaturization, to fabrication of high performance submicron ICs and to three dimensional device structures.

2. Operating Principle.

The basic deposition system construction is shown in Figure 1. Source material is vaporized through the nozzle of a crucible at high temperature into a high vacuum chamber. Operating conditions are selected so that vapor emerging through the crucible nozzle undergoes adiabatic expansion and subsequent cooling to a supersaturated condition. This leads to the formation of atomic aggregate clusters. Energetic electrons injected into the transport stream cause impact ionization of some of the clusters; these positively charged clusters can then be accelerated toward the deposition surface of the substrate by an applied potential [7].

Our simple calculation of the cross-section for displacement of the surface atoms by low energy ion bombardment has shown that the displacement begins to occur at an energy range of 20-100eV. Since displacements of surface atoms affects the film growth process and the interface properties, it is important to quantify the ion beam induced damage. The surface disorder caused by Al-ICB bombardment has been measured in connection with Al-single crystal growth on Si single crystal substrates. After the Al film was chemically etched from the Si substrate, the surface disorder of the substrate was measured by 0.925 MeV He^+ channeling. As a standard for comparison, Ar^+ bombarded Si substrates at 0.5kV to a dose of $2x10^{15}$ ions/cm^2 were also prepared. Figure 2 shows the comparison of the channeling spectra for 5kV and 0.2kV ICB Al bombarded surface, and 0.5kV atomic Ar ion bombarded surface. The spectrum representative of the unbombarded Si substrate is also shown in the figure. The results show that Si surface disorder (proportional to the area of the surface peak at about channel 105) produced by Al-ICB bombardment at the energy of 5keV and at 0.2keV is much smaller than that caused by the 0.5keV Ar ion beam. These experimental results suggest that the kinetic energy of the individual atoms in an accelerated Al cluster as it impacts a surface appears no larger than the order of 10's of eV.

One of the important bombarding effects of ICB at energies lower than that which would cause disorder is the enhancement of adatom migration. The migration effect was demonstrated directly by using a mask in front of depositing metal clusters and studying the migration length under the mask as a function of acceleration voltage. The experiment showed that a useful lateral energy is liberated as the cluster impacts the substrate and the adatoms roll off the cluster. During Al-epitaxial growth on Si, we induced at early stages a change from three dimensional to nearly layer-by-layer growth by increasing the acceleration voltage [8]. We presently understand that this phenomena might be mainly due to the enhanced surface migration of adatoms, but a change in nucleation site density could also contribute.

3. Characteristics of Epitaxial Films.

A necessary condition for obtaining good epitaxial films is generally known that the matching of lattice parameters should be less than a few percent. In Al deposition on Si(111) substrate the lattice misfit is -25 % at least in one direction. But ICB permitted epitaxial growth of Al on both Si(111) and Si (100) substrates.

Single crystal Al films of 400 nm thick were grown on Si (111) and Si (100) substrates at room temperature. Figure 3 shows a typical 75keV reflection electron diffraction (RED) pattern from Al films on Si(111) (a) and Si(100) (b) substrates. The crystalline orientation of the Al films on the Si(111) substrate was determined as Al(111)//Si(111), Al[$\bar{1}$10]//Si[$\bar{1}$10]. Two orthogonal orientations, Al(110)//Si(100), Al[001]//Si[011] denoted as Al(110) and Al(110)//Si(100), Al[$\bar{1}$10]//Si[011] denoted as Al(110)R were observed in the film on the Si(100) substrate. Crystalline quality and epitaxial relation were determined by RBS and RHEED. After the films were annealed for 30 minutes at temperatures in the 450-550°C range there were no hillocks and valleys as normally observed in Al films prepared by conventional vacuum deposition. Figure 4 shows the SEM images of the surface and the interface, revealed by phosphoric acid etching after annealing at 450°C, for ICB and conventional vacuum deposited films. AES measurements of the ICB films also showed that the interface was very abrupt and RBS measurements showed that the crystalline quality remained stable after annealing.

Most encouraging were electromigration results for 10 micron wide, 1000 micron long strips, 400 nm thick. When they passed current at a density of 10^6 A/cm^2 at 250°C, there was no change in resistance after 400 hours of operation, as opposed to sputtered Al films that normally fail at 10^6 A/cm^2 after 20 hours [9].

Thermal stability of the electrical characteristics was evaluated by fabricating Schottky barriers. The barrier height and the n value are 0.75eV and 1.17, respectively and the values showed little change after anneals to temperatures up to 550°C, as shown in Figure 5. This behavior is in stark constrast to that of the films made by sputtering or evaporation which show changes of more than 0.1eV in barrier height and 0.1 in n value. The electrical resistivity of 2.7 μohm-cm for the 400 nm thick film is comparable to the bulk value [10].

Oxidation resistance of the epitaxial Al films was examined by exposing them to high temperature steam at 550°C for 2 hrs. RBS measurement shows that the concentration of oxygen was 1.6 x 10^{16}atoms/cm^2 before and 3.4 x 10^{16}atoms/cm^2 after the treatment. The oxide thickness increases only by twice after the exposure, which means that the epitaxial film showed remarkable oxidation resistance.

4. Applications of Heteroepitaxy

Heteroepitaxial depositions of Al on various kinds of substrates could be applied to form single crystalline metal/insulater/semiconductor and metal/semiconductor device structures. For this purposes epitaxial depositions of Al on GaAs, CaF_2 and sapphire substrate have been performed.

Deposition of Al films on GaAs(100) substrate was tried by ICB at room temperture. It was found that epitaxial Al films can be grown with the orientation of Al(100)//GaAs(100), Al[011]//GaAs[010]. Heteroepitaxial depositions of Al on insulating substrates have also been carried out. Epitaxial CaF_2 films were prepared on Si substrate by ICB, on which Al films were epitaxially grown at room temperature. Figure 6 shows the 2.0 MeV He^+ channeling spectra of the CaF_2 film epitaxially grown on Si(111) at substrate temperature of 700°C and Va = 1kV.

The minimum yield for CaF_2 <110> axis was 2.0%, which shows the excellent crystallinity of the film. It was found that the Al grew with (111) plane parallel to the substrate. Al epitaxy on sapphire (0001) substrate was also performed. Figure 7 shows the RED patterns of the Al film deposited by ICB at room temperature and Va = 5kV. The incidence direction of electron beam was sapphire $[2\bar{1}\bar{1}0]$ (a) and $[10\bar{1}0]$ (b). It indicates that Al grows with the epitaxial orientation of Al(111)//sapphire(0001), Al$[\bar{1}10]$// sapphire$[2\bar{1}\bar{1}0]$ and Al(111)//sapphire(0001), Al$[1\bar{1}0]$//sapphire$[2\bar{1}\bar{1}0]$.

5. Conclusions

ICB application has been directed toward the area of semiconductor metallization. Other applications exist where there is a crucial need for semiconductor and insulator films with well-controlled crystalline structure. Development of ICB techniques for electronic device, opto-electronic and magneto-optical device fabrications is in progress.

References

1) T.Takagi, J. Vac. Sci. Technol. A2 (1984) 382.
2) I.Yamada, Proc. Int. Ion Eng. Cong. -IAIAT'83 & IPAT'83-, Kyoto (IEEJ, Tokyo, 1983) p.1177.
3) I.Yamada, C.J.Palmstrøm, E.Kennedy, J.W.Meyer, H.Inokawa and T.Takagi, Mat. Res. Soc. Proc. 37 (1985) 401.
4) I.Yamada, H.Inokawa and T.Takagi, J. Appl. Phys. 56 (1984) 2746.
5) D.Pramanik and A.N.Saxena, Solid State Technol. 26 (1983) 127. and 26 (1983) 131.
6) P.S.Ho and T.Kwok, Extd. Abs. 16th (1984 International) Conf. Solid State Device Mater., Kobe (1984) p.55.
7) T.Takagi, I.Yamada, M.Kunori and S.Kobiyama, Proc. 2nd Int. Conf. Ion Sources, 1972, Vienna (Österreichische Studiengesellschaft für Atomenergie, Vienna, 1972) p.790.
8) I.Yamada, H.Inokawa and T.Takagi, Thin Solid Films 124 (1985), 179.
9) G.Chen (EATON), private communication.
10) D.Casey (GET), private communication.

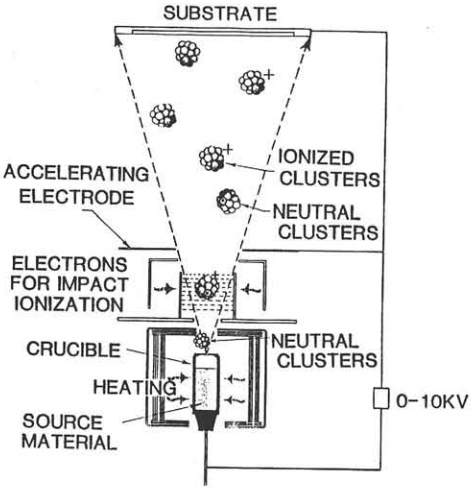


Figure 1. Schematic diagram of a laboratory ICB source.

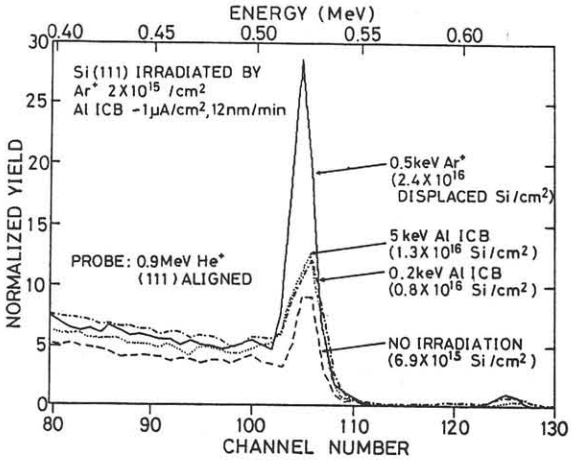
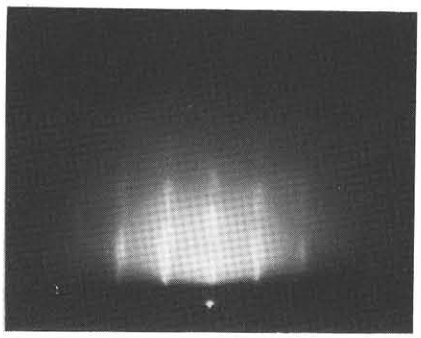


Figure 2. 0.925 MeV He^+ channeling spectra for Si bombarded by Ar^+ and Al cluster beams (Al has been etched off).

(a)



(b)

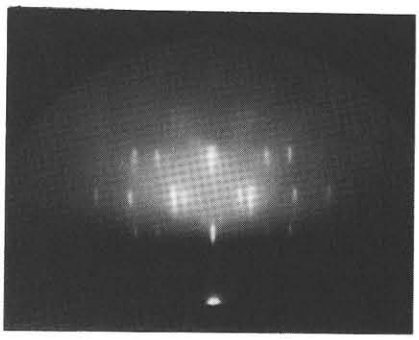


Figure 3. 75 keV RED patterns from Al films deposited by ICB. (a) Al film on Si(111) substrate. Incident electron is parallel to Si$[\bar{1}10]$. (b) Al film on Si(100) substrate. Incident electron is parellel to Si[011].

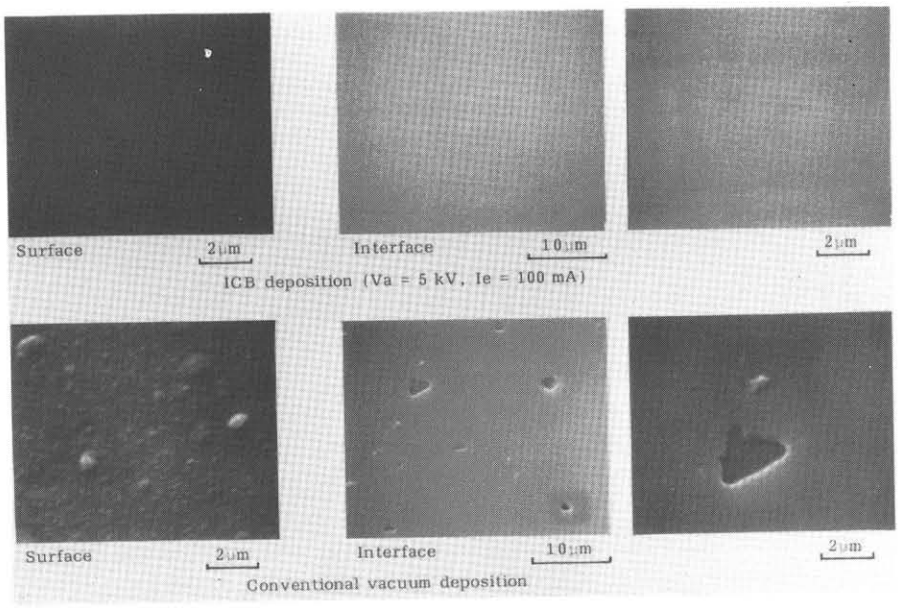


Figure 4. Surface and interface conditions of Al films on Si(111) formed by ICB deposition and conventional vacuum deposition after annealing at 500°C for 30 min.

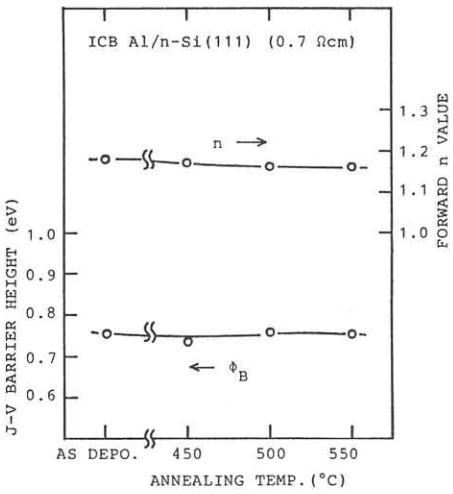


Figure 5. Schottky barrier height ϕ and ideality factor n after anneals to successively higher temperatures.

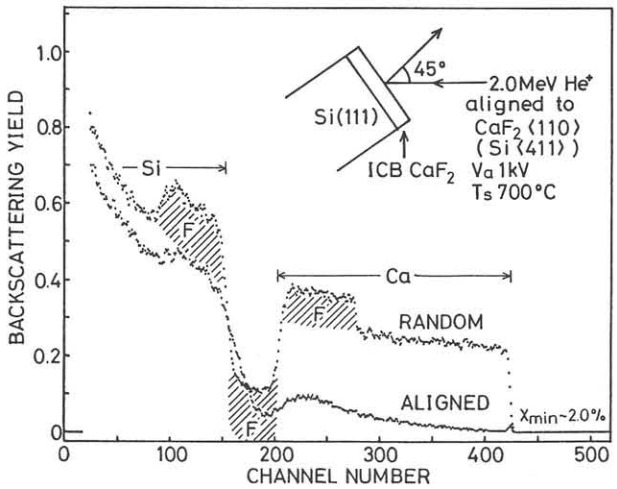


Figure 6. 2 MeV He^+ backscattering spectra of epitaxial CaF_2 film deposited on Si(111) substrate by ICB (Va=1kV, Ts=700°C).

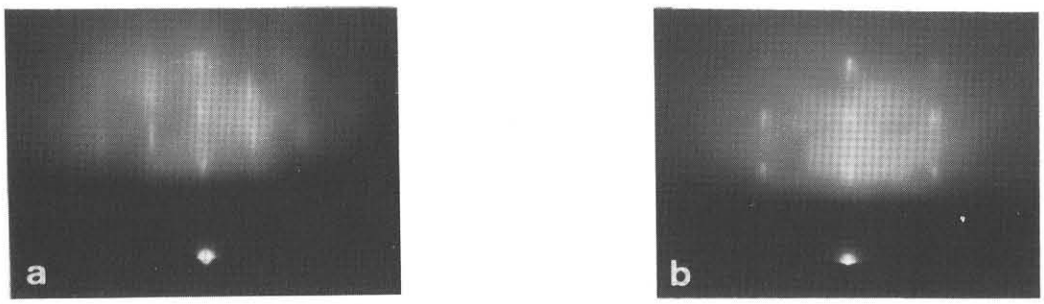


Figure 7. RED patterns of epitaxial Al film deposited on sapphire substrate by ICB. Incident electron beam is parellel to sapphire $[2\bar{1}\bar{1}0]$ (a), and $[10\bar{1}0]$ (b).