論 文 の 内 容 の 要 旨

論文題目　Combined Surface Activation Approaches to Low-Temperature Wafer Bonding for Three-Dimensional Integration
（三次元集積化のための複合表面活性化手法による低温ウェハ接合）

氏　名　赫　然

As one of the key enabling technologies for the emerging three-dimensional (3D) integration applications, wafer bonding faces a major challenge to improve bonding quality at low temperatures. This doctoral dissertation explores a newly proposed combined surface activated bonding (SAB) technique for wafer bonding at no more than 200 °C. The scope of this dissertation includes investigation of various combined activation approaches for different bonding schemes, including dielectric (SiO_2–SiO_2 and SiO_2–SiN_x) bonding and Cu/dielectric (SiO_2, SiN_x, and polymer adhesive) hybrid bonding, which are promising to offer high-quality and seamless bonding for 3D integrated systems. In more detail, the following four kinds of combined surface activation approaches are studied.

Firstly, Si deposition is combined with Ar beam irradiation to form a Si capping layer on clean SiO_2, followed by hydrophilic SiO_2–SiO_2 bonding in vacuum. Measurement results show the bonding strength increases to 1.3 J/m^2 from 0.6 J/m^2 of bonding without the Si capping, with postbonding annealing at 200 °C. Transmission electron microscopes (TEM) and energy-dispersive X-ray spectroscopy (EDS) results indicate that the SiO_2 films are bonded via an oxide interlayer with thickness around 15 nm. The bonding mechanism is suggested that the capping layer, containing ≡Si–Si≡ and ≡Si– dangling bonds, acts as an OH-adsorption layer (increasing the number of ≡Si–OH bonding sites) and H_2O-consumption layer (converting excess H_2O into H_2) for improvement of the hydrophilic SiO_2–SiO_2 bonding.

Secondly, Si-containing Ar beam that generated by a modified Si-walled beam source is used for bonding of dielectrics (SiO_2–SiO_2 and SiO_2–SiN_x) and Cu–Cu in ultrahigh vacuum (UHV) at room temperature. Bonding strength of the dielectric films is increased (to 0.8-1 J/m^2) compared to using the standard Ar beam. The improvement is suggested to be owing to the increased number of ≡Si– dangling bonds on the surfaces after the Si-containing Ar beam irradiation, which enables simultaneous Si subplantation and surface sputtering. In case of Cu–Cu bonding, bonding strength greater than Si–Ti adhesion strength and the Si bulk fracture strength (2.5 J/m^2) has been achieved. TEM/EDS and X-ray photoelectron spectroscopy (XPS) results show little Si impurities at the Cu–Cu bonding interface and on the as-activated Cu

surface, respectively. This can be explained by re-sputtering of the Si atoms from the Cu surface during beam irradiation. Thanks to the selective surface activation on dielectrics and Cu, this approach holds promise for Cu/SiO_2 and Cu/SiO_2/SiN_x hybrid bonding even at room temperature without any elevated-temperature processes and without concerns of impurities such as O and Si at the Cu–Cu interface.

Thirdly, a method called prebonding attach-detach is proposed and combined with the Si-containing Ar beam irradiation for further improvement of hydrophilic bonding of SiO_2–SiO_2 and SiO_2–SiN_x. Bonding strength close to the Si bulk fracture strength has been achieved with annealing at 200 °C. The effects of the prebonding attach-detach include increase in OH sites adsorbed (from trapped H_2O) on the surfaces during prebonding attach in air and H_2O removal by prebonding detach in vacuum. This mechanism also explains that the low SiO_2–SiO_2 bonding strength obtained by existing hydrophilic bonding methods conducted in vacuum is mainly because of the insufficient number of OH bonding sites. The same surface activation is also investigated for Cu–Cu bonding at 200 °C, resulting in high bonding strength greater than the Si bulk fracture strength. TEM confirms that the bonding interface mainly consists of O-less and ultrathin (~6 nm) O-rich ($CuO_{0.78}$) portions after postbonding annealing at 200°C. This combined surface activation approach offers improved dielectric bonding strength for the sequential 3D integration application, and Cu/dielectric (SiO_2, SiN_x) hybrid bonding for the parallel 3D integration.

Finally, surface treatment using hydrogen(H)-containing formic acid (HCOOH) vapor is investigated for Cu/adhesive and Cu/SiO_2/adhesive hybrid bonding at sub-200 °C. This chemical treatment approach avoids the Cu contamination issue on adhesive surfaces, which is found in cases using physical irradiation by Ar beam and Ar plasma. It is found that the Cu oxide reduction effect of the H-containing HCOOH vapor treatment begins at 160 °C, while the necessary bonding temperature is in range of 180-200 °C, which results in Cu–Cu bonding with shear strength in range of 40-160 MPa depending on the treatment temperature/time and bonding temperature/pressure. Despite thermal-induced prebonding curing, no negative effects of the H-containing HCOOH vapor treatment were found on bonding of the cyclic olefin polymer adhesive (bonding strength >6 MPa). By controlling the temperature (160-200 °C) and time (300-1800s) of the H-containing HCOOH vapor treatment, Cu/adhesive and Cu/SiO_2/adhesive hybrid bonding can be realized through a Cu–first bonding fashion at bonding temperatures of 180-200 °C.

The above combined surface activation approaches investigated in this dissertation have successfully shown their improvements of bonding quality at no more than 200 °C. The involved mechanisms are helpful for better understanding of the nature of low-temperature wafer bonding, while it also provides more technical options for development of novel low-temperature bonding techniques and for 3D integration applications in future.