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Nanopaste sinter-bonding for thin-film transfer and integration

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Abstract

Over the last decades, the development of micro- and nanoparticle pastes ("nanopastes") for sinter-bonding has been a major research effort in the field of micro- and nanojoining, in particular of Ag-based pastes, and more recently also of Cu-based pastes. One classical application scenario is die attach for power electronics, where sinter-bonding can provide bonds with superior temperature stability as well as thermal and electrical conductivity. A recently proposed novel application case is the use of sinter-bonding for transfer and integration of functional thin films: Thin films and 2D materials can offer unique properties, e.g. for sensing, catalysis, and electronics. However, film manufacturing often requires distinct growth conditions. For subsequent film integration into a functional device, the films must be transferred and bonded to a suitable host structure. Current transfer methods often lead to film damage, and/or insufficient bond qualities. We show that sinter-bonding with nanopastes represents as a promising new method for high-quality thin-film transfer.

Keywords

Nano-microjoining, Sintering, Thin Films, Microelectronics

1. Introduction

The realisation of technological advances from microelectronics to large-scale structures often also requires novel joining approaches to fulfil increased demands on joint performance. This makes joining a key enabling technology for advanced manufacturing. The use of metal-particle sintering for low-temperature joining was first proposed in the late 1980s for the manufacturing of power electronics devices in order to realise joints with higher thermomechanical stability and better thermal and electrical performance compared to conventional soldering [1].

Joining by particle sintering utilises the high sintering activity of metallic micro-flakes and nanoparticles (NPs) to create a joint at comparatively low temperatures, for instance in case of Ag NPs at about 180 °C - 250 °C, i.e. well below the bulk melting point of Ag at 961.78 °C. The solid-state sintering process is driven by the reduction of the surface area of the particles (minimisation of surface energy), and is facilitated by the high mobility of the metal atoms on the particle surfaces (large surface area + fast surface diffusion). As is generally the case for sintering processes, particle sintering leads to residual porosity, which can be minimised by application of an external bonding pressure. For practical use, the particles are often applied as a paste that contains different organic additives: surfactants, to stabilise the paste and avoid premature sintering, as well as binders (organic macromolecules) and thinners (organic or inorganic solvents) to adjust the workability and the thermal behaviour of the paste. Upon heating, the organic additives decompose and evaporate, thus allowing the remaining NPs to sinter together and sinter to the surfaces of the bonding partners, thereby creating a solid bond.

Recent years saw intense research efforts in sinter-paste development to optimise process parameters (temperature, time, pressure), reduce materials costs, and increase the application range, leading e.g. to the development of silver (Ag) nanoparticle pastes and later of (more oxidation-sensitive) Cu- and Ni-pastes, of pastes with in-situ production of metallic nanoparticles by metal-oxide reduction, and of particle-free inks and pastes, e.g. for fine-pitch printing. Several recent reviews provide extensive overviews of the development and use of Ag- and/or Cu-pastes [2]-[4]; some examples of Ni-paste usage are given in [5]-[8].

Up to now, die attach for power electronics and related fields are still the dominating application areas for Ag- and Cu-sinter pastes, with focus on the superior thermomechanical stability and thermal performance of the sintered joints. Recently, we proposed the use of nanopaste sinter-bonding as novel solution for the transfer and integration of thin films [9]. Thin films and 2D materials can offer unique functional properties, e.g. in sensing, catalysis, and electronics. However, film synthesis often requires distinct growth conditions that are not compatible with the envisioned operation conditions. For instance, film growth may only be possible with special growth substrates, atmospheres, and high temperatures. For integration into a functional device structure, the films must then be removed from the parent substrate, and transferred and bonded to the desired host structure. Typically employed film transfer methods often involve dissolution of the substrate or of a sacrificial interlayer followed by fishing of the free-floating film, or involve the use of adhesives (e.g. [10]-[12]). Such techniques can lead to film damage, incomplete film transfer, and/or insufficient bond qualities. On the other hand, transfer methods based on wafer bonding (e.g. direct bonding, anodic bonding, or SLID/TLP, [13]) require ultra-flat and even surfaces, and are therefore limited in their application range. Here, we present the thin-film transfer based on sinter-bonding with nanopastes as a flexible and easy-to-use method to realise high-quality thin-film transfers.

2. Sinter-bonding procedure

2.1. Overview

The proposed nanopaste thin-film transfer procedure involves three main steps:

1. deposition of the thin film on a parent substrate
2. bonding of the deposited film to a host structure using a nanopaste sinter-bonding process
3. separation of film on host structure from the parent substrate and further film processing

If required, a number of intermediate process steps can be included, e.g. deposition of a sacrificial interlayer to facilitate film separation, or metallisation of the film surface to enhance bond formation. Figure 1 shows the transfer procedure for the example case of epitaxial perovskite thin films, as presented in [9].

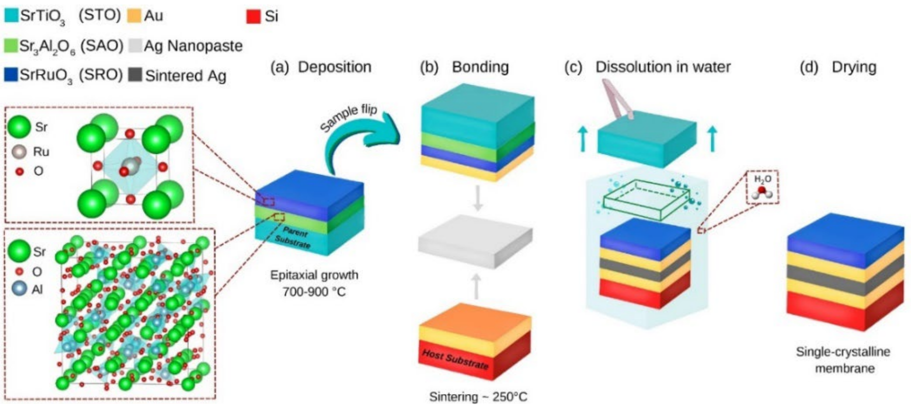

Figure 1: Proposed thin-film transfer procedure for the case of epitaxial perovskite thin films involving a sacrificial SAO interlayer [9].

2.2. Transfer procedure steps

Step 1: Thin-film deposition

Deposition of the thin films onto the parent substrate can performed according to the respective requirements, e.g. by physical or chemical vapour deposition (PVD/CVD), or other thin-film deposition techniques (spin-coating, printing,…). Here, also additional film processing steps (e.g. doping, structuring, annealing, etc.) can be applied. If necessary, a sacrificial interlayer can first be deposited onto the parent substrate in order to facilitate separation after bonding (see Step 3). The interlayer material can be selected according to two base criteria: (i) temperature stability up to the sinter-bonding temperature (180 °C – 250 °C for typical Ag-nanopastes), and (ii) selective degradability, e.g. by dissolution in a suitable solvent. Depending on the application case, interlayers may consist of selectively dissolvable salts, polymers, or metals.
In the presented example case (Figure 1), epitaxial perovskite thin films were deposited by pulsed laser deposition (PLD) using single-crystal $SrTiO_3$ (STO) substrates as parent substrate. First, a water-soluble sacrificial $Sr_3Al_2O_6$ (SAO) interlayer of 20 nm to 40 nm thickness was deposited onto the STO at a substrate temperature of 800 °C. Under these conditions, SAO shows full epitaxy with STO and can therefore serve as a template for further epitaxial growth of perovskites [10]. Subsequently, $SrRuO_3$ (SRO) and/or STO perovskite thin films with various thickness (here 20 nm to 180 nm) and area up to 5 mm x 5 mm were deposited at 725 °C. After deposition, the SRO and STO films indeed exhibited high-quality epitaxy, as shown by X-ray diffraction (XRD, [9]).

Step 2: Thin-film transfer by nanopaste sinter-bonding

In a second step, the deposited film, while still attached to the parent substrate, is bonded to the host structure by nanopaste sinter-bonding. To this end, the nanopaste is applied onto either the film or the host side, followed by assembly and bonding. If necessary, the film surface can first be metallised in order to improve bond formation with the nanopaste material.
In the present case, the SRO and STO film surfaces were metallised with Au by sputtering. Subsequently, a commercial Ag nanopaste (NanoTach-X, NBETech) was stencil-printed onto the metallised film (typical thickness: 20 µm, typical area: 4 mm x 4 mm). Assembly and bonding to a metallised Si parent substrate was done using a manual flip-chip bonder (Fineplacer Pico Ma, Finetech; Figure 2). The sinter-bonding process was performed in air at 250 °C for maximum 30 min with a bonding force of ≥ 2 N (corresponding to ≥ 0.125 MPa for a bonding area of 16 mm2; for further details see [9]).

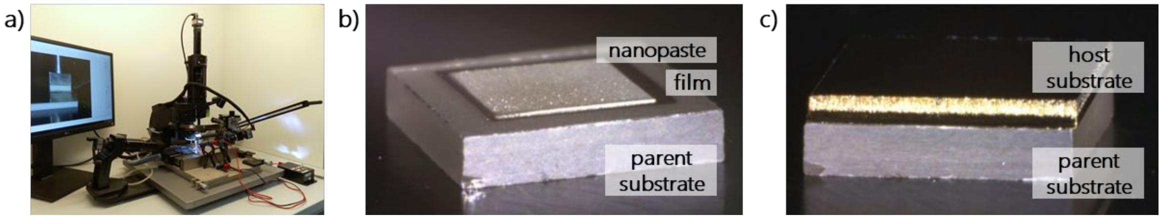

Figure 2: a) Manual flip-chip bonder used for the nanopaste sinter-bonding process, b) perovskite thin film on the STO parent substrate with a nanopaste layer printed on top the film, c) parent substrate with the host substrate sinter-bonded to the film.

Step 3: Separation from the parent substrate

As final step of the transfer procedure, the film, which is now firmly bonded to the host structure, can be separated from the parent substrate. This can e.g. be achieved by mechanical cleavage in case of a weak bond between parent substrate, by dissolution of the sacrificial interlayer (cf. Step 1), or by other separation methods.
In our application case, the perovskite thin films were separated from the STO parent substrate by dissolution of the sacrificial SAO interlayer in ultrapure water, followed by a drying step. This way, complete film transfer of the bonded areas could be achieved (Figure 3 a)).

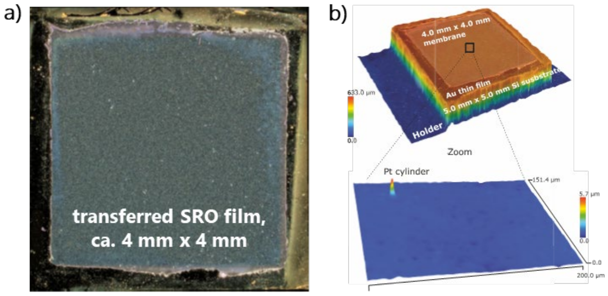

Figure 3: a) Optical microscopy image of a transferred SRO film on a metallised Si-substrate. b) Confocal microscopy image. The film surface is defect-free, and shows very low roughness and high planarity with the Si host substrate (the Pt cylinder was deliberately added to visualise the height scale; images modified after [9]).

2.3. Further processing and analysis

Once the transfer to the host structure is completed, the flipped film can be further processed, e.g. by performing additional integration steps such as deposition of metallisation pads and top electrodes for electrical contacting.
For the showcase of epitaxial perovskite thin films, an extensive characterisation of the post-transfer properties was performed to assess the quality of the transfer process. The transferred films exhibited very high quality, i.e. high surface planarity and low roughness over large areas (Figure 3 b; confocal microscopy, profilometry, atomic force microscopy), low degree of surface contamination (X-ray photoelectron spectroscopy), and a high degree of post-transfer epitaxy (XRD, for details see [9]). Cross-sections showed formation of a homogeneous bond line with low defect density along the bond interface (scanning electron microscopy, Figure 4). Bond formation between the Au-metallisation layer of the film and the Ag-sinter-layer was found to occur by solid-state interdiffusion of Au and Ag.

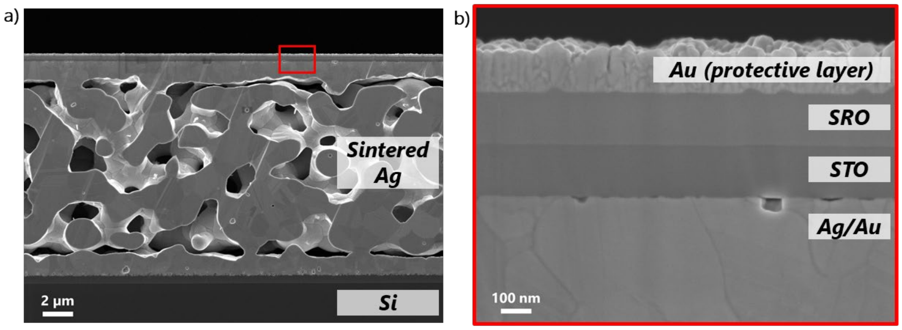

Figure 4: a) Scanning electron microscopy images of a transferred SRO-STO double-layer film in cross-section bonded with a low pressure of 0.125 MPa. Left: overview of the cross-section; the sintered Ag-layer shows grain sizes of 1-2 µm and a relatively high porosity due to the low sintering pressure. b) Close-up of the SRO-STO double layer and the Au-Ag-bond layer formed by the interdiffusion reaction of the Ag-nanopaste with the Au-metallisation (images modified after [9]).

3. Discussion

For the presented example of perovskite thin films, a high transfer efficiency and high film quality after transfer was found compared to conventional methods, which highlights the benefits of the novel transfer method. This holds in particular for the case of oxide thin films, which are general prone to film cracking and wrinkling. Moreover, the purely metallic bond between transferred film and host structure ensures a high thermal stability as well as high thermal and mechanical stability. The high quality of thin-film transfer realised by use of nanopaste sinter-bonding can be attributed to a combination of several factors: First, due to the transfer sequence of deposition-bonding-separation, the film is always mechanically supported throughout the transfer process. Hence, mechanical damages such as wrinkling and cracking due to film manipulation can be minimised. Second, the sinter-bonding process itself represents a comparatively benign bonding process, because the build-up of stress during the bonding process is comparatively small for sinter-bonding with regard to e.g. the use of solders or adhesives: while the latter processes general show a considerable, and sometimes relatively abrupt, volume shrinkage due to solidification, respectively curing, the sinter-bonding process can compensate for the volume change upon paste decomposition by particle rearrangements and atomic diffusion processes which reduce the stress exerted onto the film. In addition, the bond-formation by solid-state interdiffusion instead of liquid-phase wetting also reduces the risk of film attack and dissolution. Due to the adaptability of the nanopaste during the bonding process, the requirements on surface flatness, roughness and overall cleanness of the bonding partners are comparatively low (cf. [13]), which overall makes the proposed thin-film transfer method a flexible and easy-to-use new technology for the integration of thin films.

4. Conclusion

In this study, a novel procedure for thin-film transfer by use of nanopaste sinter-bonding is presented. As shown for the application case of epitaxial perovskite thin films, the proposed procedure represents a comparatively benign, flexible and easy-to-use method for the integration of thin films.

5. References

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