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Engineering inclined orientations of piezoelectric films for integrated acoustofluidics and lab-on-a-chip operated in liquid environments

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Different acoustic wave modes are required for effective implementation of biosensing and liquid actuation functions in an acoustic wave-based lab-on-a-chip. For efficient sensing in liquids, shear waves (either a thickness-shear bulk wave or an shear-horizontal surface acoustic wave) can achieve a high sensitivity, without significant loss of acoustic wave energy. On the other hand, longitudinal bulk waves or out-of-plane displacement waves (such as Rayleigh waves) enable efficient sampling functions and liquid manipulation. However, there are significant challenges involved in generating multiple wave modes and performing both these functions on a single piezoelectric substrate, especially when a single crystalline orientation is available. This paper highlights the latest progress of the theories and techniques to deliver both sensing and microfluidic manipulation functions using engineered inclination-angled piezoelectric films, allowing for the simultaneous generation of longitudinal (or Rayleigh) and thickness-shear bulk (or shear-horizontal surface acoustic) waves. Challenges and theoretical constraints for generating various wave modes in the inclined films and techniques to efficiently produce inclined columnar and inclined crystalline piezoelectric films using sputtering deposition methods are presented. Applications of different wave modes in the inclined film-based lab-on-chips with multiple sensing and acoustofluidic functions are also discussed.

1 Introduction

Piezoelectric materials have unique properties of generating electricity when they are subjected to external deformation/pressure, and vice versa, they are able to perform actuation functions through geometrical changes when external electrical fields are applied. These materials include: (1) bulk crystals of quartz (SiO₂), lithium tantalite (LiTaO₃), lithium niobate (LiNbO₃), sapphire (Al₂O₃), lead zirconate titanate (Pb(Zr,Ti)O₃, PZT), barium strontium titanate (BaSrTiO₃), etc.; and (2) piezoelectric thin films such as PZT, zinc oxide (ZnO), aluminium nitride (AlN), LiNbO₃, gallium nitride (GaN), boron nitride (BN); and (3) piezo-polymers such as polyvinylidene fluoride (PVDF) and polymer-ceramic composites, etc. By depositing metal electrodes on top and bottom surfaces of a piezoelectric material, a bulk acoustic wave (BAW), in either a longitudinal mode or a thickness-shear mode (TSM), can be generated when electrical signals are applied to these two electrodes as illustrated in Figs. 1a and 1b. A common example is quartz crystal microbalance (QCM), with a TSM wave mode generated in a quartz crystal sandwiched between two electrodes, which have been commercially used in various sensing applications. Film bulk acoustic resonators (FBARs, with a piezoelectric film on top of a membrane structure) and solidly mounted resonators (SMRs, with a piezoelectric film on stacks of alternate layers of low and high acoustic impedance forming a Bragg acoustic reflector) have recently been extensively studied. They have structures similar to those of BAW devices, but their dimensions (both size and film thickness) are much smaller (in micron or sub-micron scale), thus achieving operation frequencies from sub-GHz to tens of GHz, suitable for high precision sensor applications. Similarly, FBARs and SMRs can have either longitudinal or TSM waves, similar to those shown in Figs. 1a and 1b, depending on the crystal orientation of the film.

Lamb waves are commonly generated when the substrate thickness is smaller than or comparable to the wavelength of the propagating waves on the substrate (see Figs. 1c and 1d). Lamb waves include: antisymmetric mode (A mode), also called flexural plate waves (FPWs), and symmetrical mode or extensional mode (S mode).
mode). Lamb waves were recently reported that can be generated using thin film piezoelectrics on a membrane structure.\(^5^,\)\(^6\)

Figure 1. Illustrations of different types of commonly observed acoustic waves: including (a) longitudinal bulk acoustic waves (BAWs) generated between a top electrode and bottom electrode; (b) thickness-shear mode (TSM) waves generated between two electrodes; (c) Lamb waves, antisymmetric mode (A mode, or flexural plate waves-FPWs); (d) Lamb waves, symmetrical mode or extensional mode (S mode); (e) Rayleigh surface acoustic wave (SAW) generated on a piezoelectric materials using interdigital transducer (IDTs); (f) shear horizontal SAWs (SH-SAWs) generated on piezoelectric materials using IDTs; (g) Love-mode wave modes generated on the surface of an SH-SAW device coated with a thin wave-guide layer.

In a surface wave configuration, interdigitated transducers (IDTs) are commonly deposited onto the piezoelectric material, and surface acoustic waves (SAWs) can be generated after resonant frequency signals are applied to these IDTs. The SAWs propagate on the surface of the material in either an out-of-plane (e.g., Rayleigh) mode or an in-plane (e.g., shear horizontal) mode, as shown in Figs. 1e and 1f. Rayleigh SAWs can be generated on specifically oriented bulk piezoelectric substrates such as ST cut and Y-cut quartz, \(128^\circ\) Y-X-cut LiNbO\(_3\) and X-cut LiTaO\(_3\) substrates, as well as vertically aligned or (0002) oriented films (often called c-plane or r-plane oriented) of ZnO, AIN, PZT or LiNbO\(_3\). Shear-horizontal SAWs (SH-SAWs) are generated on ST cut quartz, \(64^\circ\) Y-X-cut LiNbO\(_3\) and \(36^\circ\) Y-X-cut LiTaO\(_3\), and the waves propagate via a shear-horizontal mode on the substrate surface.\(^6^,\)\(^7\) Figure 1f. SH-SAWs can also be generated on many piezoelectric thin films with in-plane textures such as (10\(^1\)0) (commonly called m-plane) or (11\(^\pm\)2) (commonly called a-plane), or (10\(^2\)2) orientations (commonly called r-plane).\(^8^\)\(^-\)\(^1^3\)

Another commonly investigated SAW is Sezawa mode, which are excited in SAW devices that harbour a layered structure where the phase velocity of the SAWs in the substrate is much larger than that of the overlaid piezoelectric film.\(^5^,\)\(^6\) They are intrinsically guided waves, which propagate at the interfaces between the film and substrate. The ratio of the wavelength to the piezoelectric film thickness plays a critical role on the properties of Sezawa waves. As the waves propagate along the interface, the influence of the structure of the thin film may be limited, although the detailed effects have not been studied.

Generally, bulk piezoelectric materials are much thicker and have better piezoelectric parameters in large scale, whereas piezoelectric thin films have advantages in microscale devices for MEMS and lab-on-chip applications.\(^2^6^,\)\(^2\) For a better understanding of the different film orientations of piezoelectric film materials, Fig. 2 shows four typically observed crystal orientations, i.e., (0001) c-plane; (10\(^1\)0) m-plane; (11\(^\pm\)2) a-plane and (10\(^\pm\)2) r-plane. Love mode SAWs, one of the key guided acoustic waves, can be generated within a thin wave-guide layer (such as SiO\(_2\), ZnO or polymers with sub-micron thickness) coated onto an SH-SAW device as shown in Fig. 1e.\(^2\)\(^1\) The Love mode waves have been regarded as optimal for biosensing in liquid conditions, mainly because the acoustic velocity of the wave-guide layer is lower than that of the substrate; thus the SH-SAW generated are mostly trapped within this thin wave-guide layer, leading to a high sensitivity to signal changes at the surface.\(^2\)\(^2^,\)\(^2\)\(^3\)

Both the longitudinal bulk waves and out-of-plane (Rayleigh) SAWs are significantly damped when they come in contact with a liquid, because a large portion of their wave movements on the surface is vertical. Therefore, they are subjected to extensive damping, with significant deformations up to a few wavelengths at the solid-liquid interface. Therefore, they are not recommended for sensing applications in a liquid or highly humid environment, but instead have recently been extensively explored for acoustofluidics (or bio-sampling) functions such as mixing, jetting, nebulisations, particle/cell manipulations and acoustic heating.\(^2\)\(^4^,\)\(^2\)\(^7\) TSM waves, SH-SAWs and Love mode waves have been applied for liquid sensing due to their limited dissipation of wave energy during their propagations at the solid-liquid interface, but for the same reason, they are less commonly used in acoustofluidics.\(^2\)\(^8^,\)\(^2\)\(^9\)

Recently acoustic wave devices have been extensively explored for detection of biomolecules, viruses, DNA, bacteria and cells\(^5,\)\(^6,\)\(^3\)\(^0\). Most of these devices are required to work in a liquid environment. Therefore, there is a need to generate both a shear wave (either TSM or SH-SAWs) for efficient bio-sensing, and longitudinal or Rayleigh waves for bio-sampling and liquid sample manipulation processes, in one device platform. However, there are significant challenges to generate both these functions onto one single piezoelectric substrate with a given crystal orientation.
For the BAW devices, it is possible to generate both longitudinal and shear wave modes by fabricating devices on a bulk piezoelectric material with a suitably chosen crystal cut or orientation (e.g., non-vertically aligned ones). Similarly, thin-film FBARs based on specially designed inclined angled piezoelectric ZnO and AlN thin films have been extensively investigated to generate both longitudinal (for microfluidics) and BSM waves (for liquid sensing) and thus have been used in liquid conditions as shown in Fig. 3a. Generally the longitudinal wave has a higher frequency than that of the corresponding bulk shear mode due to their differences in acoustic wave velocities with an illustration as shown in Fig. 3b.

Another method is to modify electric fields generated from standard IDTs (as shown in Figs. 4b and 4c). In Fig. 4b, a bulk shear acoustic wave can be generated using a standard IDTs on a c-axis vertically aligned piezoelectric film deposited on dielectric reflector stacks of AlN/SiO₂, thus resulting in the electric fields mainly aligned along the x-axis of the film plane. Whereas in Fig. 4c, the top and bottom electrodes are not perfectly aligned in z-direction, thus the asymmetrical fields will generate both vertical and shear waves, although the film’s crystal structure is vertically aligned.

Another method explored to a lesser extent is called off-axis excitation, where the top electrode is relatively smaller than the bottom electrode thus both pseudo-shear and pseudo-longitudinal waves can be generated by this method as shown in Fig. 4d. A similar idea using two top electrodes with different areas has also been proposed for a ZnO SMR sensor, which can generate both shear and longitudinal modes for biosensing in liquid conditions. This design is actually often reported in standard pulse-echo ultrasonic tests that a mixture of shear and longitudinal waves can be easily generated when using a small area top electrode on thin-film coated metallic substrates (see Fig. 4e).

There is another reported method in which a periodically patterned ZnO film was deposited between fingers of IDT to fill their gaps and implement the SAW resonator as shown in Fig. 4f. Both the longitudinal and transverse wave modes can be generated.

However, the efficiency in generating strong and effective shear mode waves for all the above-mentioned unique designs are debatable.

For quartz-crystal based SAW devices, Rayleigh waves can be excited and then propagate along the X-cut direction on the 37° Y-cut quartz wafer, whereas the SH-SAW can be excited normal to the above direction. For thin film-based SAW devices, if the electrodes of the interdigitated transducers (IDTs) are deposited on top of inclined angled piezoelectric films; (d) A schematic illustration to indicate the differences in the frequencies of longitudinal and bulk shear waves; (c) Rayleigh SAW and SH-SAW generated with IDTs deposited on top of inclined angled piezoelectric films; (d) A schematic illustration to indicate the differences in the frequencies of Rayleigh SAW and SH-SAW.

For quartz-crystal based SAW devices, Rayleigh waves can be excited and then propagate along the X-cut direction on the 37° Y-cut quartz wafer, whereas the SH-SAW can be excited normal to the above direction. For thin film-based SAW devices, if the electrodes of the interdigitated transducers (IDTs) are deposited on top of inclined angled piezoelectric films as shown in Fig. 3c, both the Rayleigh SAW and SH-SAW can be generated. It is generally agreed that the SH-SAW has a higher frequency than that of the Rayleigh one as illustrated in Fig. 3d.

Apart from using the inclined-angled or non-vertically aligned films, other potential methods have been proposed to generate waves for sensing in liquid conditions on vertically oriented (0002) materials or films. For example, one key and widely studied method is to use a lateral field electrode (LFE) design as illustrated in Fig. 4a. This establishes laterally excited resonators to generate shear, or mostly pseudo-shear waves, which can be used for biosensing in a liquid environment, along with vertical waves for acoustofluidics functions to form an integrated lab-on-a-chip based on thin films. These types of LFE designs can be easily combined with FBAR or SMR designs, thus generating both longitudinal and shear thin film bulk modes. For the LFE mode, the shape, dimension and distance of two electrode pads are not critical, whereas the thickness of the piezoelectric material below is critical to determine the generation of the wave frequencies.
So far, using inclined angles thin films has been shown to generate both longitudinal (or Rayleigh waves) and TSM (or SH-SAW) waves. The produced inclined angled thin film devices can be used as the building blocks of integrated Microsystems for microfluidic lab-on-a-chip, with possibility of using separate wave modes for the functions of liquid transportation, biosampling and sensing. However, there are many contradictory views in the literature about the inclined angles of films (either columnar or crystal ones), the fundamental formation mechanisms of inclined crystals and their key growth parameters. Many different techniques have been developed but there are a lot of issues on uniformity of inclined angles, film thickness and acoustic wave properties. It is still a great challenge to successfully generate both these types of waves for SAW and FBAR devices and realise all the different functions of an integrated lab-on-a-chip. In this paper, we address all of the above issues and discuss the following key questions:

(1) What are fundamental and theoretical issues in the use of inclined films to generate both vertical and shear waves?

(2) How can inclined columnar and inclined crystal piezoelectric films be produced efficiently using film deposition methods and what are the key processing parameters?

(3) How can both shear waves and longitudinal/Rayleigh waves be generated?

(4) How can these waves be effectively integrated into one platform and applied for lab-on-chip applications?

2 Fundamentals of angles of inclination and wave modes in inclined films

SAW and FBAR devices based on inclined films have recently been extensively studied and have shown improved mass sensitivities and good performance under liquid conditions\textsuperscript{57,58}. For these devices, the inclination angles have shown significant influences on the generation of acoustic wave modes.

2.1 Definitions of three angles of inclination

Before discussing the details of these mixed wave modes, three different definitions of the angles of inclination must be clarified, which have been previously discussed in Refs.\textsuperscript{4,5}:

a. **Substrate inclination angle** $\alpha$. The first type of inclination angle has been defined as the substrate tilting angle. During deposition, the substrate can be tilted or inclined to the deposition source at a desired angle, which can be varied from 0° to 90°. The angle $\alpha$ is sometimes also called the target-tilting angle if the target is adjusted into a certain angle when the substrate position is fixed. This angle is easily adjusted by the experimental design using deposition methods such as oblique angle deposition (OAD, see Fig. 5a) or glancing angle deposition (GLAD, see Fig. 5b)\textsuperscript{59}. During the OAD process, inclined columnar structures can be readily achieved by tilting/fixing the substrate at a large angle (normally much larger than 45°) in the direction of the incident flux. In the GLAD process, the substrate movements are precisely controlled (tilting/rotating in a 3D scheme at very large tilting angles) during the deposition, and the deposited films often show large inclined columnar angles with nanopillars, zig-zag or helical nanoscale patterns\textsuperscript{60-66}.

b. **Columnar inclination angle** $\beta$. The second definition of inclined angle is the film’s columnar inclined angle, and the exact values of the angle $\beta$ can be obtained from morphological analysis based on cross-section images of optical or electron microscopes. The columnar inclined angle $\beta$ is generally smaller than the substrate tilted angle $\alpha$, as illustrated in Fig. 5c\textsuperscript{59}. In some cases, there is no apparent inclined columnar morphology (probably due to the formation of smooth and dense film microstructures) even after the substrate has been tilted significantly.

c. **Crystal inclination angle** $\gamma$. The third angle is defined as the film’s crystal inclination angle, $\gamma$, which specifies that the crystal orientation is inclined at a certain angle to the(0002) plane of the films (see Fig. 5c). The crystal inclination angle $\gamma$ can be determined using texture analysis methods such as X-ray diffraction (XRD) through pole figures or psi ($\psi$) scan\textsuperscript{68} (with examples of illustration shown in Fig. 5d for a (0002) vertically aligned AlN film and Fig. 5e for a 30° crystal inclined AlN film), or using a transmission electron microscope (TEM) with the help of electron beam scattering diffraction (EBSD). Another less commonly used method to characterise the tilting angle (mainly for AlN films) is to analyse the Raman peak position of longitudinal optic (LO) quasi-mode at ~890 cm$^{-1}$, whose shifts are linked to the c-axis crystal angle tilting at the surface of the film\textsuperscript{59}.

Generally, the crystal inclination angle $\gamma$ is much smaller than the columnar tilted angle $\beta$ (see Figs. 5c and 5f). In many cases, even though the film shows a large columnar inclined angle, the crystal inclination angle is actually near zero, meaning that the crystals in
2.2. Relationships among angles of inclination

Empirical relationships have been proposed to analyse the links between the substrate tilting angle $\alpha$ (or incident angle of the vapour flux) and columnar inclination angle $\beta$ using the OAD method. For example, the tangent rule$^{72,73}$, cosine rule$^{74,75}$ and other empirical equations are commonly used relationships:

$$\tan(\alpha) = 2\tan(\beta)$$

$$\beta = \alpha - \arcsin\left(1 - \cos(\alpha)\right)/2$$

$$\tan(\beta) = 2\tan(\alpha)/(3[1 + \beta\tan(\alpha)\sin(\alpha)])$$

Fig. 5(f) shows the curves of substrate tilting angle $\alpha$ as a function of columnar inclination angle $\beta$, predicted by both tangent rule and cosine rule. Clearly both of these rules could not precisely predict the experimental data of sputtered deposited ZnO thin films especially at large substrate tilting angles (although it seems the cosine rule shows a better prediction of data obtained at a lower tilting angles). The reason for this difference is mainly because these proposed rules were developed based on the measurement data of thin films deposited using evaporation processes (in which a good “line of sight” property of vapour flux and a long distance between evaporation source and substrate can be achieved due to low chamber pressures in the deposition process), while most of the piezoelectric films for the SAW and FBAR devices are prepared using sputtering methods. During sputtering processes, there are more random scattering effects$^{4,5}$ due to: (1) the shorter distance between target and substrate; (2) varied target geometries (e.g., rectangular or circular); (3) significant angular distribution of plasma sources. Therefore, the columnar inclination angles of the sputtered films may not be uniform or may not exactly follow the empirical rules developed from the data of evaporation processes, such as those obtained using equations 1 to 3. It was also reported that the columnar inclined angles are also dependent on the film materials/chemistry$^{76}$ and substrate conditions (for example, texture or roughness of the substrate), which makes this a complex issue.

To the best of our knowledge, there are no available analytical relationships between the substrate tilting angle and crystal inclination angle $\alpha$ vs. $\beta$, or between the columnar inclination angle and crystal inclination angle $\beta$ vs. $\gamma$. This is mainly due to the crystal orientation/angles being influenced by a wide range of parameters, including process parameters, thin film material properties, target and substrate conditions and properties. Fig. 5(f) shows some data of crystal inclination angle $\gamma$ as a function of substrate tilting angles $\alpha$ obtained from inclined angled ZnO and AIN films. Another challenge is that the XRD or TEM/EBSD methods are needed to precisely determine crystal orientations. However, these data are not widely available, partly because of the complexity of the geometry of the sample preparation and the speciality of the characterisation techniques. It is generally agreed that the crystal inclination angles are much smaller than those of the columnar angles, and in many cases, c-axis crystal orientation (0002) is still dominant for most of the columnar inclined films.

2.3. Key mechanisms determining columnar and crystal inclination angles

The columnar inclination angles are pre-dominantly controlled by deposition process parameters and substrate conditions. To generate columnar inclination angles in the films, the main controlling mechanisms include:

1. Direction and energy of the incoming adatoms on the deposited film;
2. Surface micro-/nano-scale features, and their shadowing and self-shadowing effects;
3. Migration/diffusion of surface adatoms;
4. Channelling effect or momentum/bombardment of arriving adatoms.

On the other hand, crystal inclination angles $\gamma$ in the deposited films are intrinsically determined by competing factors among the growth rates of different crystal orientations or crystal planes on the film surface$^{77,78}$, which are controlled not only by process parameters, but also by material properties of both the film and the substrate. In the following, we will discuss a few key factors.

Figure 6. (a) Illustration of the shadowing effect during film deposition; (b) Cosine law distribution of the sputtered adatoms on the substrate surface; (c) channelling effects during sputtering process; (d) A circular-shaped target produces a ring distribution of inclined columnar angles (dark area) in the films, which can be revealed clearly from the illustration of cross-section morphology; (e) An interlayer can promote the growth of inclined angled films; (f) Pre-treatment of surface to generate the desired surface roughness or surface topography and enhance shadowing effect for growth of inclined angled films; (g) An additional blind/shutter positioned between the target and the substrates can...
change the plasma distribution onto the deposited films, thus promoting the inclined film growth; (g) An external oxygen or nitrogen ion or plasma source can control the oxygen/nitrogen ion bombardment to the deposited film during its inclined angled film growth.

- **Shadowing effect** (mostly determine the columnar inclined angles)
  During the OAD processes, the newly generated crystal nuclei on the substrate can form shadows behind them, which prevents the further formation of microstructures in these "shadowed" regions (see Fig. 6a), thus forming columnar inclined angle structures. This so-called shadowing effect is strongly linked to the directionality of the incoming particles. The inclined angled crystal structure is easily generated when the arriving particles have a good line of sight such as in an evaporation process, and also if the substrate titled angle is large enough. On the contrary, during the sputtering process, the plasma contains energetically charged ions, reactive species and sputtered atoms, which are all attracted onto the surface of the deposited film with different impinging angles. There is a well-known cosine law to describe the distribution of the sputtered adatoms as below (also illustrated in Fig. 6b)\(^\text{[7]}\):

\[
J(\theta) = Y(\cos\theta)\pi
\]

where \(J(\theta)\) is the emission flux angular distribution as a function of the tilting angle \(\theta\) (vs. the vertical direction), \(Y\) is the sputter yield emitted from the surface, and \(\phi\) is the local ion flux incident onto the surface. This cosine law distribution of sputtered adatoms will surely cause a more randomly distributed flux of ions/adatoms (compared with a conventional evaporation process) through the discharge plasma, and thus less line-of-sight effect when the adatoms reach to the substrate surface. These, unfortunately, may not contribute to generating a strong shadowing effect as preferred.

- **Bombardment and channelling effects** (for both columnar and crystal inclination angles)
  During sputtering processes of ZnO, AlN and PZT films, various reactive species (such as O\(_2\), O, N, N\(_2\), N\(_2^+\), etc.) and energetic species (such as Ar or Ar\(^+\) ions) in the plasma have strong influences on the energy transfer to the deposited films. During a standard magnetron sputtering process, the kinetic energy of sputtered atoms/ions arriving to the substrate could vary from sub-ev to hundreds of ev, which generate various effects (such as deposition, bombardment, etching, and implantation) and have a significant effect on the formation of columnar structures. As will be explained later, process parameters, such as a higher plasma power or a lower pressure, will result in an increase of the kinetic energy of these impinging atoms onto the substrate, which is one of the key reasons that fully crystalline films and also piezoelectric effects of ZnO, AlN and PZT can be easily achieved in sputtering process operated without any external heating, or post-polishing processes\(^\text{[8,9]}\). Similarly, this effect would significantly influence the columnar inclined angles of the deposited film.

It is well-known that bombardment of the adatoms during sputtering will generate so-called channelling effects (see Fig. 6c), which enhance the formation of columnar inclined structures, especially at a low incident flux and low temperature/pressure environment. At such circumstances, free movement of adatoms (deposited onto the substrate surfaces) can be restricted in a certain direction, thus promoting film growth along a desired orientation. For example, (10\(\overline{1}0\)) plane was found to be easily generated when the film was exposed to ion irradiation during electron beam evaporation due to its line-of-sight effect\(^\text{[10]}\). During deposition of ZnO and AlN films, the film texture was changed from (0002) into (10\(\overline{1}0\)) or (11\(\overline{2}0\)) by using a directional oxygen or nitrogen ion beam placed at an angle to the substrate surface, which is mainly due to this ion channelling effects\(^\text{[11]}\).

- **Surface migration/diffusion** (for both columnar and crystal inclination angles)
  Irradiation or bombardment of the arriving adatoms could contribute to the densification of film’s microstructure by enhancing mobility of the deposited adatoms through the angular distribution of the neutral species\(^\text{[12]}\). During the sputtering process, ion bombardment (mainly Ar, as well as O\(_2\) or N\(_2\)) will significantly influence the structures of the crystals, as well as the development of large compressive stress\(^\text{[8,13]}\), and could result in either formation of dense microstructures (due to the enhanced mobility of the deposited adatoms), or porous structures (due to the channelling effect or bombarding/damaging effects). In many cases, significant surface migrations of adatoms deposited onto the film surface is not effective to promote the crystal inclined angle formation.

- **Nature of film material and bonding probability** (mostly for crystal inclination angles)
  Crystal angles of films are linked with the nature of film materials and also different bonding probabilities of atoms during growth. For example, for an AlN film, this factor could link to the formation of bonds such as (N, Al) or dimers (Al–N)\(^\text{[8]}\). It was reported that a higher chamber pressure promotes the generation of more (Al–N) dimers thus resulting in the growth of AlN (10\(\overline{1}0\)) planes. Whereas at a low pressure, atoms of N and Al are dominant, rather than the dimers of Al-N, and thus the (0002) growth is promoted\(^\text{[13]}\). Effects of pressure will be discussed in the next section.

### 2.4. Effects of sputter deposition process parameters

As discussed above about the physical mechanisms, energy/angular distribution of the incoming material flux and mobility of the adatoms during the inclined angled deposition process are critical for the formation of film microstructures. All sputter deposition process parameters will influence the film growth on different orientations and the surface mobility of adatoms and thus will influence the inclination angle of the film growth\(^\text{[14]}\). These key process parameters include\(^\text{[4,5]}\): the nature of the substrate (such as phases or chemical composition, crystalline orientation and roughness, etc) and process parameters (such as gas pressure, target plasma power, position and distance of substrate vs. target, deposition time, bias potential, substrate temperature, gas ratio and flow rates, external ion/electron beam source, plasma shields)\(^\text{[15]}\).

#### 2.4.1. Plasma power and substrate bias voltage

In sputtering processes, target power affects the energy and mobility of the adatoms when they arrive at the substrate. Increasing target power will increase the kinetic energy of the adatoms arriving at the substrate, thus enhancing both the channelling effect and surface migration/diffusion effect (see Section 2.3). However, the effect of target power is quite complex. For example, an increase of the target power increases the deposition rate, and the fast arrival of the adatoms on the film surface quickly buries the previously deposited ones, causing the formation of loose or columnar microstructures\(^\text{[16]}\). Excessive target power causes structural damage, resulting in the formation of...
coarse columnar structures in the films. Whereas a high target power can also generate high kinetic energy, thus bombarding the film surface and enhancing the surface diffusion, which might densify the deposited film and prevent the formation of columnar structure.

When an external bias voltage/power source is applied to the substrates during the sputtering deposition, the film orientation along (0002) is generally enhanced, but not the other orientations due to the normal direction of the energetic Ar ions bombarding onto the substrate surface. During the growth of ZnO and AlN films, a positive bias has been shown to increase the kinetic energies of oxygen or nitrogen ions, and enhance the mobility of adatoms arriving onto the film surface, causing significant bombardment/cleaning effects, large compressive stress and increase in roughness. A negative bias decreases bombardment but still provides sufficient mobility of adatoms arriving onto the film surface, thus enhances (0002) texture growth.\(^2,3\)

2.4.2. Target geometry

As mentioned, the sputtering target is normally placed close to the substrate (compared with evaporation process, for example), thus the atoms/ions which are sputtered away from the target reach the substrate along a wide range of directions due to the angular distributions of ions or scattering in the plasma. Positioning the substrate away from the sputtering target could produce clearer columnar inclined films, but at the expenses of reduced energy of arriving adatoms on the substrates and poor crystallinity.\(^4,5\) The shape of the target also shows a significant influence on the uniformity of the inclined angled films. For example, a circular-shaped target produces a circular ring region of inclined columnar angles in the films (as illustrated in Fig. 6d), but the central areas directly below the target and the areas far away from the targets could still show the vertical columnar structures due to the energy distribution of the arriving deposited adatoms, which we will discuss in further section.

2.4.3. Gas pressure and gas flow rate

Gas pressure and gas flow rate have been shown to not only determine the mean free path of sputtered adatoms, but also influence the plasma density on the target.\(^6,7,8,9,10\) A low chamber pressure (for example, below 1 mTorr) is often required to generate significant ion bombardment from atoms/ions of argon, oxygen and nitrogen) and provide a longer mean free path to preserve adatoms energies and reduce gas-particle scattering.\(^11\) This enhances the channelling effects and promotes the formation of inclined columnar angles. At a high chamber pressure (for example, tens of mTorr), the sputtered particles can experience a large number of collisions in the chamber before arriving at the substrate and this reduces the kinetic energy and momentum (including the direction of arrival angles) of the sputtered particles upon reaching the film surface. Therefore, a high chamber pressure could decrease the adatom mobility and increase the shadowing effect, thus could also be beneficial for the formation of columnar inclined films. For example, it was reported that the inclined crystal angle was increased by up to 10° with the increase of gas pressure.\(^8,9,12\)

O\(_2\)/Ar or N\(_2\)/Ar gas ratios also have significant effects on the film texture.\(^9,13\) For example, AlN films with (10\(\overline{1}\)0) or (1\(\overline{1}\)20) orientation were obtained on 3C-SiC-on-Si substrates by simply increasing the nitrogen to argon ratio from 40% to 80% at a total gas flow of 50 sccm.\(^14\) In this case, the variations of gas ratio would change the kinetic energy of the atoms deposited onto the films.

2.4.4. Substrate temperature

Substrate temperature during deposition has been shown to provide a way to control both columnar and crystal inclination angles. Heating the substrate changes the microstructures of the deposited film due to an increase in the mobility of adatoms, which could promote (0002) oriented film growth rather than crystal inclined film growth such as reported for AlN film.\(^15\) Denzi et al.\(^16\) reported that there is a temperature threshold, above which surface diffusion of the adatoms becomes dominant over surface shadowing, thereby preventing the formation of a typical inclined angled columnar feature.

2.4.5. Substrate effect: composition, orientation, interlayer, and surface features

Epitaxial growth of films with various orientations on different substrates have previously been explored using methods such as molecular beam epitaxy (MBE), metal organic vapour phase epitaxy (MOVPE), or laser processing,\(^16-100\) on certain orientations of common substrates such as quartz, sapphire, LiNbO\(_3\), SrTiO\(_3\), diamond, MgO, CaF\(_2\), GaAs, GaN, GaN/Sapphire and SiC. Examples include ZnO(11\(\overline{2}\)0)/LaAlO\(_3\)(100),\(^101\) ZnO(11\(\overline{2}\)0)/r-plane (10\(\overline{1}\)2)\(_0\),\(^102\) ZnO(11\(\overline{2}\)0)/sapphire Al\(_2\)O\(_3\)(01\(\overline{1}\)2)\(_0\).\(^103\) However, these epitaxial growth methods are expensive and the orientations are significantly dependent on the quality of the substrate materials and deposition conditions.\(^104,105\)

Another commonly used method for changing the film orientation is to deposit an interlayer or a seed-layer (as illustrated in Fig. 6e), which could provide a template and promote the inclined angle growth of the piezoelectric films.\(^104,105\) For example, a thin AlN buffer of tens of nanometres has significantly facilitated the crystal quality of \(\alpha\)-plane ZnO films and formation of SH-SAWs.\(^106\) This type of interlayer does not need to be a piezoelectric material. This interlayer can be further pre-treated to generate the desired surface roughness or surface topography and enhance shadowing effect in order to grow columnar inclined angled films (see Fig. 6f for some examples of these patterns). For example, ZnO film with an inclined angle of 45° was obtained on a diamond substrate\(^107\) with pre-deposited pyramidal-shaped diamond features,\(^108\) mainly due to shadowing effects. Pre-patterned substrates can be prepared using a variety of methods, including photolithography, ion-beam milling, laser beam writing, nano-imprinting, embossing and self-assembly of nano-structured array patterns.

3 Sputtering techniques and technical challenges

In this section, we will discuss further about the sputtering techniques and challenges to realise different types of inclined films.

3.1 Substrate tilting or target tilting

As mentioned, inclined ZnO and AlN films are commonly prepared using the OAD method. However, high-performance SAW and FBAR devices could promote (0002) oriented film growth rather than crystal inclined film growth such as reported for AlN film.\(^15\) Denzi et al.\(^16\) reported that there is a temperature threshold, above which surface diffusion of the adatoms becomes dominant over surface shadowing, thereby preventing the formation of a typical inclined angled columnar feature.
3.2 Locating samples at off-centre positions\(^{115}\); In the sputtering process, if the target is small compared with the substrate, a deposited film has a vertically columnar structure and (0002) orientation at its centre area as shown in Fig. 6d as an example. At a certain distance away from the target (or at off-centre positions from the plasma source as shown in Fig. 6d), inclined angle columnar structures with tilted angles up to 10° to 20° can be obtained, mainly due to ion channelling and shadowing effects\(^{116}\). However, at locations far away from the plasma source, the (0002) orientation becomes dominant again as illustrated in Fig. 6d, mainly due to the limited ion/electron/atom bombardment energy and insignificant channelling effects, leading to the dominant self-orientation along (0002) orientation. This simple technique is believed to avoid the need for any special re-configuration of deposition setup. However, the window area over which a uniformly inclined angle is produced is quite small (for example, a circular and ring area as shown in the dark area in Fig. 6d). This surely poses a challenge for the mass production of acoustic wave devices.

3.3 Epitaxial growth on a specific substrate Generally, by selecting a suitable substrate with a preferred orientation, inclined crystal films can be deposited using specific methods. However, these process methods are generally expensive, complicated, and difficult to control, as already discussed in Section 2.4.5.

3.4 A suitable seed layer with inclined angles Inclined films can be obtained when they are grown onto a pre-deposited seed layer with a texture or pre-fixed inclined angle on the substrate. For example, using a textured Al film as a seed layer, tilted crystal ZnO films have been developed for liquid sensors\(^{124}\). For example, using a textured Al film as a seed layer, deposited seed layer with a texture or pre-fixed inclined angle on the substrate, can have a significant effect on the orientation of the growing films based on ion channelling effects\(^{37}\), as discussed in Section 2.4.1. This generally makes the deposition process complicated and high cost, and the treated area of the film is not large, which is mainly dependent on the size of ion beam sources.

3.7 Summary of technical challenges Even though inclined angled films as thick as a few microns (see Fig. 7a for an example) with a size of four-inch wafer have been successfully prepared using different techniques during the sputtering deposition, there are still many critical issues to be solved:

- (1) Non-uniformity of film thickness and inclination or tilting angles over a large planar scale. As mentioned, most of the methods used to generate the inclination angle are not able to obtain a significant uniformity over large areas (for example, larger than 2 or 3 inches)\(^{125}\).
- (2) Variation of inclination angles along the film’s cross-section (e.g., thickness) direction. It is a challenge to maintain a constant inclined angle along the thickness direction. Studies showed that the columnar inclination angles of films would often be gradually changed during deposition, or sometimes even change to a vertical orientation during deposition (see Fig. 7b for an example)\(^{117,126}\). These phenomena can be explained by the inevitable minor changes in the deposition conditions occurring over a lengthy deposition process (in most cases tens of hours), which could change the mobility of deposited species.
- (3) Significant variations in acoustic wave propagation characteristics in planar directions. The large variation in c-axis tilt angles over a large area causes difficulties in obtaining consistent resonant frequencies and repeatable performance when a number of resonator chips are fabricated over this area. The acoustic wave velocities and frequencies of the devices decrease with increasing angles of inclination.
- (4) Mixture of c-axis and inclined crystals in the deposited film. It is commonly to see that for the prepared inclined films, XRD analysis reveal that the film has actually consisted of both c-axis and crystal inclined films. This is probably the reason why many characterisation data of crystal inclined angles are confused. However, identification of the precise percentages or ratios between them would be a challenging work.
- (5) Formation of loosely columnar, porous or nanofibre structures due to self-shadowing effects. When the film is deposited at very large substrate tilting angles (e.g., above 60° or 70°), the formed columnar microstructures are normally quite porous and loose (see Fig. 7c). Indeed, both the OAD and GLAD methods have been mainly used to generate nano-pillar, nanorods or 3D spiral porous structures (see Fig. 7e for an example). Tait\(^{1}^{\text{1}}\) derived an equation about the density of the film, \(\rho\), as a function of substrates tilting angle, \(\alpha\), for example).

\[
\rho = 2\cos \alpha / (1 + \cos \alpha)
\]

The above equation clearly shows that the density of the deposited film decreases significantly with an increase of the tilting angle. The resulting rough surfaces and low density for the inclined angled films significantly reduce the Q factor of the resonant peaks of the devices due to the formation of surface defects such as etch pits, pyramids, ridge, and ledge like and elongated grains structures. The potential spaces between columnar structures will cause elastic abnormality and anisotropy of the films\(^{1}\). Therefore, acoustic wave devices made of these films with very large columnar inclined...
angles often exhibit weak signals and low Q factors, i.e., poor reflection and transmission performance\cite{128}.

It should also be noted that one of the key advantages using the piezoelectric films for acoustic wave devices is to generate in-plane isotropic acoustic wave properties\cite{133}, so the patterning positions or directions of IDTs or electrodes will not affect the wave mode generation and acoustic wave properties (such as velocities or frequencies). However, if columnar and crystal inclined films are used, the anisotropic acoustic wave properties within the film plane negate this benefit, e.g., becoming significantly anisotropic.

Inclination angles (both columnar and crystal lattice) have significant effects on peak amplitude ratios between shear and longitudinal wave modes and their electromechanical coupling coefficients\cite{134}. However, it should be noted that for most theoretical studies, crystal inclined angles \(\beta\) was used in the analysis, whereas for most of experimental studies, columnar inclined angle \(\beta\) was often used in the data analysis.

Figs. 8a and 8b shows theoretical values of wave velocities of both longitudinal and bulk shear modes of AIN film as a function of crystal inclined angles, respectively, which follows the results proposed in references\cite{134}. As shown in Fig. 8a, the velocity of longitudinal wave decreases quickly with the increase of inclination angle to \(~45^\circ\) above which it increases slightly until \(90^\circ\). Whereas as shown in Fig. 8a, the velocity of bulk shear waves increases continuously with the increase of inclination angle to \(35^\circ\), above which it gradually decreases until \(90^\circ\). Figure 8b shows electromechanical coupling coefficients of AIN film as a function of c-axis inclination angle for the bulk acoustic waves (including FBARs). When the inclination angle is increased from 0 to 45\(^\circ\), the ratio of shear wave mode (to longitudinal one) and its \(k^2\) values gradually increase, whereas those of the longitudinal mode gradually decrease. The values of \(k^2\) for the shear and longitudinal waves become similar at an angle of \(~10^\circ\). The \(k^2\) values of the shear mode become larger than those of the longitudinal values with the angle increased from \(~10^\circ\) to \(~60^\circ\)\cite{135,137}. The maximum \(k^2\) value of shear mode resonance occurs at a c-axis angle of inclination of about 25-38\(^\circ\). Theoretically analysis show that pure thickness-longitudinal modes for ZnO (or AIN) FBARs are generated at c-axis angular of 0\(^\circ\) (0\(^\circ\)) and 67.1\(^\circ\) (65.4\(^\circ\)), whereas pure thickness-shear modes for ZnO (or AIN) FBARs are generated at 43\(^\circ\) (46.1\(^\circ\)) and 90\(^\circ\) (90\(^\circ\))\cite{134,138}.

Pang et al. studied the effect of inclination angles using a pulse-echo ultrasonic method\cite{141,142} for inclined ZnO films with different columnar inclination angles on steel substrates. Results showed that both the longitudinal echoes and shear echoes from the back wall of the steel plate could be obtained. A dual mode wave pattern (i.e., with both components of longitudinal and shear waves, mainly due to the small size of the top electrode compared to that of the
metal substrate) was detected for the ZnO film with a crystal inclined angle of 0°. When the inclined columnar angle was increased, the amplitude ratios of the shear/longitudinal wave components were increased. When the inclined angle was increased to 31°, a nearly pure shear wave was obtained whilst the longitudinal wave excitation was significantly suppressed141. For further increases of the inclination angle up to 35°, the shear wave was still dominant but the longitudinal component re-appeared141.

3.2. On SAWs

Reports on effects of inclination angle on the generation of Rayleigh mode and SH-SAWs are less readily available. This is mainly because a relatively thicker inclined film with a larger scale of uniform inclination angle is required for generation of these SAW modes, and the IDTs for the SAW devices generally have larger sizes (for example, in many cases the dimensions of their reflectors are added). Fig.8c shows velocity results of Rayleigh and SH-SAW modes as a function of the inclined angle vs. X-axis in bulk LiNbO3 SAW devices. When the inclined angle is increased from 0° to 90°, the theoretically calculated velocity of the Rayleigh mode is much lower than that of the SH-SAW mode139,140. A similar dispersive behaviour of the Rayleigh and SH-SAW modes exist in the inclined piezoelectric films136. Fig. 8d illustrates that when the crystal inclined angle is 0° (e.g., (0002) orientation) for the ZnO SAW devices, the Rayleigh wave is dominant and has the largest value of k2. With an increase in crystal inclined angle, the value of k2 for the Rayleigh wave gradually decreases until becoming negligible as the crystal inclined angle approaches 90°141. Conversely, the amplitude ratio and k2 value of the SH-SAW peak become zero for vertically aligned films, but they increase with the crystal inclined angles up to 45°, and then gradually decrease up to 90°.

Yanagitani and Suzuki142 reported that c-axis inclined AlN films showed significant decreases in shear wave acoustic impedance, but not in that of vertical Raleigh type of waves. They have attributed this effect to significant shear elastic softening caused by the weak inter-columnar connection. This elastic softening effect could also explain the difficulties in achieving the SH-SAWs in the inclined films, and there are few papers about the successful generation of both wave modes in the inclined films.

5. Sensing, acoustofluidics and integrated lab-on-a-chip using inclined films

5.1. FBAR liquid sensors and microfluidic devices based on inclined angled films

Recently, FBAR or SMR sensors with the inclined angled piezoelectric films have been extensively studied for biosensing in liquid. These sensors could be made from both ZnO and/or AlN films with columnar inclination angles and crystal inclination angles to generate shear waves for sensing in liquid144-148. The inclined angled films used in the fabrication of the FBAR or SMR devices are easily obtained as they are generally very thin (sub-microns to one or two microns) and small (e.g., micron scale). Within such a thickness range and such an area range, a uniform inclined angled film in both thickness and planar directions are readily obtained, enabling a huge number of successful reports into their applications. Both ZnO and AlN films have been extensively used in these FBAR or SMR devices for biological applications in liquid environments as summarized in Table 1. Due to its much higher wave propagation velocities for AlN films compared with those of the ZnO films, AlN FBARs or SMRs have their advantages in comparison with those of the ZnO devices, but ZnO films have its advantages with relatively higher electro-coupling coefficients149. To further enhance the sensitivity and performance of FBAR devices, new materials, such as carbon nanotubes and graphene, were also deposited onto the inclined AlN films150.

Table 1. Summary of film bulk acoustic resonators (FBAR) and solidly mounted resonators (SMR) based on inclined angled ZnO and AlN films for biosensing applications

<table>
<thead>
<tr>
<th>Piezoelectric material</th>
<th>Struc tured angles</th>
<th>Working frequency</th>
<th>Sensitivity</th>
<th>Key applications</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZnO FBAR</td>
<td>2 GHz</td>
<td>2.4 kHz·cm/²/ng</td>
<td>detect DNA and protein molecules</td>
<td>151</td>
<td></td>
</tr>
<tr>
<td>ZnO FBAR 16°</td>
<td>830 MHz</td>
<td>1 kHz·cm/²/ng</td>
<td>water-glycerol solution</td>
<td>152</td>
<td></td>
</tr>
<tr>
<td>ZnO FBAR 158</td>
<td>1.04 GHz</td>
<td>500-1358 Hz·cm/²/ng</td>
<td>blood proteins (e.g. Alb, IgG and Fibrinogen)</td>
<td>153</td>
<td></td>
</tr>
<tr>
<td>ZnO SMR C-axis inclining angle 25°</td>
<td>1.1 GHz</td>
<td>4.9 kHz·cm/²/ng</td>
<td>rabbit immunoglobulin G (IgG) molecule</td>
<td>104</td>
<td></td>
</tr>
<tr>
<td>ZnO FBAR 141</td>
<td>1.4 GHz</td>
<td></td>
<td>human blood coagulation process</td>
<td>154-156</td>
<td></td>
</tr>
<tr>
<td>ZnO FBAR 136.6° tilted c-axis</td>
<td>1.28 GHz</td>
<td></td>
<td></td>
<td>158</td>
<td></td>
</tr>
<tr>
<td>ZnO FBAR c-axis inclined angle of 24°</td>
<td>2.26 GHz</td>
<td>20 pg/ml</td>
<td>cardiac biomarkers in the clinical serum</td>
<td>159</td>
<td></td>
</tr>
<tr>
<td>AIN FBAR c-axis 23°</td>
<td>800 Hz</td>
<td></td>
<td>human IgE</td>
<td>160</td>
<td></td>
</tr>
<tr>
<td>AIN FBAR 30°</td>
<td>700 MHz to 1.5 GHz</td>
<td>1.5 to 2.9 kHz·cm/²/ng</td>
<td>streptavidin and biotinylated BSA, and crosslinked fibrinogen</td>
<td>161, 162</td>
<td></td>
</tr>
<tr>
<td>AIN SMR crystalline inclined angle of 30°</td>
<td>1.2 GHz</td>
<td></td>
<td></td>
<td>163</td>
<td></td>
</tr>
<tr>
<td>AIN FBAR 23° tilted colu</td>
<td>1.175 GHz</td>
<td>1.425×10⁵ cm³/g</td>
<td>IgE</td>
<td>165</td>
<td></td>
</tr>
</tbody>
</table>
As discussed above, inclined angled FBAR devices have been widely used in liquid biosensing, whereas so far, there are no reports for liquid actuation applications using the inclined angled FBAR devices. The integrated functions for both microfluidics and biosensing on one platform have never been demonstrated. In principle, bulk acoustic waves are able to generate liquid streaming and create flow patterns. For example, a circularly self-focused bulk wave acoustic transducer capable of generating focused waves on (0002) oriented ZnO film on silicon, so-called “liquid needles”, was used to generate a focused acoustic wave in a liquid chamber and to produce a needle-shape liquid column on the free surface of the liquid tank \(^{169-172}\). Whereas for many microfluidics functions, the electrical power required is quite high, thus the devices could be easily damaged, unless this has been controlled using methods such as compensation circuits.

5.2. Inclined angled SAW liquid sensors and microfluidic devices

Pure SH-SAW devices using films of (10 \(\overline{1}0\)) m-plane, (11 \(\overline{2}0\)) a-plane and (1 \(\overline{1}0\) \(\overline{2}0\)) r-plane, as well as Love mode thin film SAWs have been obtained in many configurations. However, there are few reports for successful achieving both Rayleigh SAW and SH-SAWs using one inclined angled piezoelectric film. It is still a great challenge to enable the integrated SH-SAW and Rayleigh waves for both microfluidics and biosensing by using inclined ZnO and AlN films, and this has never been reported so far.

Fardeheb-Nammeriet et al. \(^{173}\) previously reported SAW devices made from c-axis inclined AlN film (with a maximum crystal inclined angle of 10\(^\circ\)) achieved shear waves with a velocity of 5832 m/s, and Rayleigh waves with much lower velocity of 5082.72 m/s. SH-SAW devices were also demonstrated using c-axis tilted Sc doped (44%) AlN films (c-axis inclined 5.5\(^\circ\)) over the whole sample surface) fabricated on sapphire \(^{174}\). Within these limited reported references, the SH-SAWs from the inclined film SAW devices have been applied for liquid sensing demonstrations, and the Rayleigh SAW components have never been used for microfluidics or biosampling demonstrations.

As explained in Section 3.7, the specific challenge related to the generation of both Rayleigh waves and SH-SAWs using the inclined angled films is the need for thick (a few microns) and large-area films (2 or 3 inch and above) in order for patterning the IDTs and precisely controlling waves. Most of the films deposited using the OAD method have a mixed c-axis and inclined crystals, and often the c-axis orientation is more dominant for the film texture. In most cases, due to the formation of loosely columnar, porous or nanofiber structures, the SH-SAWs in the inclined angled film devices show weak signals.

6 Conclusions and future outlook

This paper reviews the mechanisms, techniques and problems about incline-angled piezoelectric films to generate both longitudinal (or Rayleigh) waves and thickness-shear bulk (or shear-horizontal surface acoustic) waves. This approach is advantageous, yet challenging, for simultaneously enabling molecular sensing and liquid actuation. So far, the generation of different wave modes has been demonstrated using the inclined piezoelectric thin films with FBAR and SAW structures using three different types of inclinations: substrate tilting, columnar tilting, and crystal tilting. Effects of the inclination angle design should be evaluated carefully by identifying the specific definitions of these different angles. Applications of different types of waves integrated into lab-on-chips for multifunctionality have been explored. However, there are still technical challenges such as non-uniformity of the inclination angle and non-uniformity of the film thickness that limits their widespread adoption. The inclined films are generally relatively porous, have rough surfaces with poor adhesion, and generally do not retain usable acoustic wave properties for their application to real-life systems.

Recently there are also many studies for the wearable and flexible SAW devices, in which thin foils of metal and polymers are used. When inclined films are deposited on thin substrates and foils, the control of foil deformation and the generation of different waves become challenging. In that case, Lamb waves or flexural waves will be dominant and co-existing with shear waves, thus the modes will become hybridized.

The acoustic devices of the inclined-angle piezoelectric films, including SAW and FBAR devices, will be used as a critical part to be integrated with electrowetting-on-dielectrics (EWOD), electrophoresis, and dielectrophoresis (DEP), into a complex system, which may be developed to control and manipulate the liquid sample or cells, detect the toxic and poisonous gas and sensitively response to the environmentally slight changes such as the variation of the ultraviolet light, pressure and humidity. \(^{175-178}\) They are also promising to embed in 3D printed microfluidics to enhance the capacity of pumping and sensing the biological liquid in the desired tubes and chambers. \(^{177}\) Considering the functionalities of acoustic waves, technical developments and the challenges, our projections for the further development in the area of engineered inclined-angle piezoelectric films are as follows:

- Different electrode structures can be patterned on a single substrate that can be used for the efficient generation of different wave modes. For example, anisotropy due to inclined-angle piezoelectric films allows the generation of longitudinal waves in one axis and shear waves in the perpendicular axis. Longitudinal waves can be utilized for efficient liquid pumping, while the shear waves enable sensitive detection. This configuration is especially advantageous in a microfluidic channel where several detectors can be placed in an array formation as shown in Fig. 9a. In this architecture, the longitudinal wave could be coupled along the channel and the shear wave transducers placed perpendicular to the channel. The detection area for these transducers can be functionalised with individual types of molecules for specific detection. An array of SAW devices can be patterned and fabricated, and then used for multiple sensing purposes.
(2) A single structure can be designed as a multimode transducer. The difference in acoustic velocities associated with different wave modes allows simultaneous optimisation of a single structure at distinct wavelengths favouring selected wave modes. In an operation scenario, this enables liquid pumping using longitudinal waves at one frequency and sensing using shear waves at another frequency.

(3) A particular configuration to simultaneously optimise the sensing and liquid manipulation functions is the integration of Rayleigh SAW and shear wave FBAR devices on a single platform. The best inclination angle should be optimised so that a single structure can be used to generate Rayleigh SAW for liquid manipulation and shear waves in FBAR configuration for sensitive detection.

(4) Despite its benefits in terms of multifunctionality, improved performance and high level of integration, there are technical challenges to realise this method such as the difficulty in mass production of inclined films with a uniform thickness due to the difference of the deposition rate among others. Precise control in deposition processes can provide a breakthrough point to design and develop various acoustic devices for efficient acoustofluidic manipulation and highly sensitive detection on a single platform.

(5) More studies can be focused on the mechanisms of interference of different types of wave modes and enable beam steering on a single substrate. The ability to generate different wave types on a single substrate opens the possibility of applying new techniques designed for Lamb waves, Love waves, and bulk waves into SAW devices.

Fig. 9b demonstrates that a novel, smart and integrated system with multiple functions could be developed, based on the flexible control of the inclination angle and easy steering of the propagation of the acoustic waves on different substrates. The liquid from a reservoir will be efficiently and remotely transported to the detection area where the biobinders are located (i.e., on top of the FBAR) through conventional microchannel designs. The following SAW induced mixing will speed up the reaction and reduces the immuno-reaction time, which can minimize non-specific bonding and thus reduce faulty detection. Pairs of FBAR devices will be fabricated in order to minimize the temperature drifts and other interferences on the FBAR sensors. This can also be extended by fabricating multiple FBAR sensor arrays to provide multiple disease detection capabilities. In the MEMS process, integration is more challenging as both the SAW and FBAR device elements involve several process steps, and their process compatibility as well as single interferences will be the main focus. One inherent advantage of this design is that SAW devices and FBAR sensor arrays on the same chip can be fabricated using similar fabrication steps. A judicious selection and deposition of different immobilised biobinders on the FBAR arrays enables the detection of different types of DNA or proteins, providing a basis for multiple disease detection. Once the reaction completes, the SAW device can generate a wave to drive the liquid waste away. In this proposed integrated system, the droplet transportation, immobilization and detection are implemented in sequence, to minimise cross-talk and interference.

Author contributions
Y. Q. Fu: Conceptualization, Resources, Methodology, Writing-original draft, Supervision, Project administration, Funding acquisition. H. F. Pang: Investigation, Visualization, Data curation, Writing-original draft, Funding acquisition. H. Torun: Visualization, Data curation, Validation, Writing-review & editing. R. Tao: Investigation, Formal analysis, Data curation. G. McHale: Funding acquisition. Validation, Writing-review & editing. J. Rebound: Investigation, Validation, Writing-review & editing. K. Tao: Investigation, Data curation. J. Zhou: Investigation, Data curation. J. Luo: Investigation, Funding acquisition. D. Gibson: Validation, Writing-review & editing. J.K. Luo: Funding acquisition, Writing-review & editing. P. Hu: Validation, Writing-review & editing.

Conflicts of interest
There are no conflicts to declare.

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