Lateral field excitation properties of langasite single crystal*

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In this work, bulk acoustic wave propagation properties of langasite single crystal excited by lateral electric field have been investigated. Three important crystal cuts have been identified for different operational modes of lateral field excitation (LFE) on langasite substrate, namely the (yxl)65° (pure-LFE mode), (yxl)45° (quasi-LFE mode), and (yxl)0° (pseudo-LFE mode). Devices on langasite substrate with the above cuts were fabricated and tested, and the experimental results agree well with the theoretical analysis. It is found that a pure thickness shear mode exists in the (yxl)65° langasite LFE device with the bare side facing liquid, and no spurious mode is found due to its moderately large piezoelectric coupling factor. In addition, (yxl)0° langasite LFE device is also found suitable for liquid phase sensing applications.

Keywords: lateral field excitation, langasite, bulk acoustic wave
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1. Introduction

Bulk acoustic wave (BAW) devices may be classified into two groups according to different excitation mechanisms. One is thickness field excitation (TFE) where the thickness shear mode (TSM) wave is excited by electric field along the plate-thickness direction. This is realized by laying two electrodes on opposite plate faces, as shown in Fig. 1(a). The other is lateral field excitation (LFE) where the TSM is excited by the electric field along the plane of the plate, and is realized by laying two electrodes on the same plate surface, as shown in Fig. 1(b).

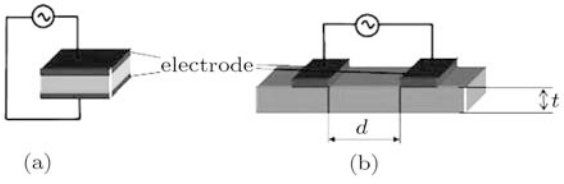


Fig. 1. Field excitation mode: (a) TFE mode and (b) LFE mode.

The LFE was first used by Atanasoff and Hart[1] to determine the elastic constants of quartz in the 1940s. Since then, some researchers have carried out studies on the LFE devices used as high precision frequency control elements, such as resonators.[2] Ballato *et al.*[3] have shown that LFE devices have a number of advantages over TFE devices. There is no electrode on the region of maximum displacement, which results in higher Q values, better frequency stability, and reduced aging of the device. By advisable placement of the electrodes that results in a particular orientation of the exciting electric field, undesirable modes of certain orientations in the crystal could be totally eliminated. Besides, due to the particular electrode configuration, LFE devices have been found sensitive to both mechanical and electrical properties of the target analyses.[4] Based on the advantages mentioned above, LFE devices have been applied in liquid phase sensing, such as the detection of biomolecules.[5]

Recently, Wang *et al.*[6] found three operational modes of LFE devices, namely pure-LFE operational mode, pseudo-LFE operational mode and quasi-LFE operational mode. According to Wang's theory, for the pure-LFE mode, the TSM wave is excited only by the lateral electric field. For the pseudo-LFE mode device, when in air, there is no resonance peak. While

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in liquids, a resonance peak appears. This resonance peak is generated not by the LFE but by the TFE with the liquid acting as a virtual electrode. For the quasi-LFE mode device, when in air, there is one resonance peak generated by LFE, while two resonance peaks appear when in liquids. One is generated by LFE and the other is generated by TFE due to the role of the liquid virtual electrode. In resonator and sensing applications, it is important to have only a single thickness shear wave mode active to obtain good energy trapping.[7] Therefore, the pure-LFE mode device can operate not only in air but also in a liquid environment. The pseudo-LFE mode device can operate only in a liquid environment, and the quasi-LFE mode can operate only in an air environment. Wang showed that the three operation modes of LFE devices can be realized through appropriate selection of the substrate material, cuts, and the exciting electric field direction. This research result has provided great help in applying more unknown cuts of piezoelectric crystal material in different environment of sensing application. Moreover, this also makes it possible to fully excavate potentialities of LFE on other crystal material besides quartz.

Especially, langasite single crystal (for convenience, "langasite single crystal" is substituted by "langasite" in the following), which can be produced by the conventional Czochralski melt pulling technique using La_2O_3, Ca_2O_3, and SiO_2, belongs to the trigonal 32 crystal class and has moderately large piezoelectric coupling factors, moderate dielectric permittivity, very small acoustic loss and good temperature characteristic.[8,9] Therefore, a study on LFE of langasite is highly desirable. Several years ago, Khan and Ballato[10,11] studied the LFE properties of langasite theoretically. They found that high LFE coupling factors can be achieved in some special cuts of langasite. However, the LFE coupling factors for all different cuts were not presented and the relevant cuts for the three operational modes are still unknown, which has hindered the application of langasite LFE devices in sensing.

For the LFE devices in liquid phase sensing, since the liquid can act as a virtual electrode, the TFE and LFE may be effective simultaneously.[6] Therefore, it is needed that only one mode is active among the four thickness shear modes, namely LFE-b, LFE-c, TFE-b, and TFE-c modes (As a quasi-extensional wave, the a-mode acoustic wave attenuates rapidly in liquid and does not influence the wave of other modes). Only in this way, can the TSM be excited by a single mode. Up to now, nobody has presented the cuts of langasite suitable for liquid environment so that the TSM is excited by a single LFE shear mode. Besides, no experimental result has been given for verifying the LFE properties of langasite.

In the present paper, the LFE properties of langasite are studied theoretically and experimentally. Wave propagation properties of LFE shear wave are investigated for all cuts of langasite and lateral field directions, based on which, the cuts for three different operational modes of langasite are presented and the cuts of langasite suitable for liquid environment may be found. Several devices on different operational modes are fabricated and tested for verifying the theoretical results.

2. Theory

2.1. Piezoelectric coupling factor

The extended Christoffel–Bechmann method[12] was used to calculate the LFE coupling factors of the langasite in all different cuts and lateral field directions. The calculations were made using the material constants of Sakaharov *et al.*[13]

The IEEE standard[14] is used to define the crystal axial set and the notation (yxwl)ϕ/θ, as shown in Fig. 2(a), specifying the orientation of a substrate aligned with a rotated coordinate set of axes. Fig. 2(b) shows the E-field direction relative to crystallographic axes.

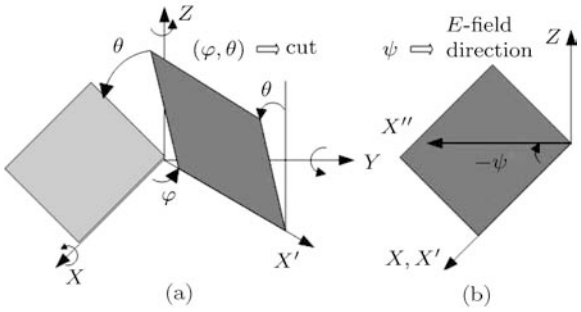


Fig. 2. Notation system used for calculating: (a) crystallographic orientation of a given substrate, and (b) E-field direction relative to crystallographic axes.

The LFE coupling factors for a mode (quasi-extensional mode), b mode (fast quasi-shear mode),

and c mode (slow quasi-shear mode) of langasite in all different cuts and lateral field directions, namely as functions of the rotation angles (ϕ, θ, ψ), have been achieved. As the thickness shear modes are commonly used in bulk acoustic wave resonators, only b mode and c mode are discussed here. The four-dimensional graphs of langasite LFE coupling factors for c mode and b mode are plotted in Fig. 3. Based on the calculated results shown in Fig. 3, the cuts for three operational modes can be found.

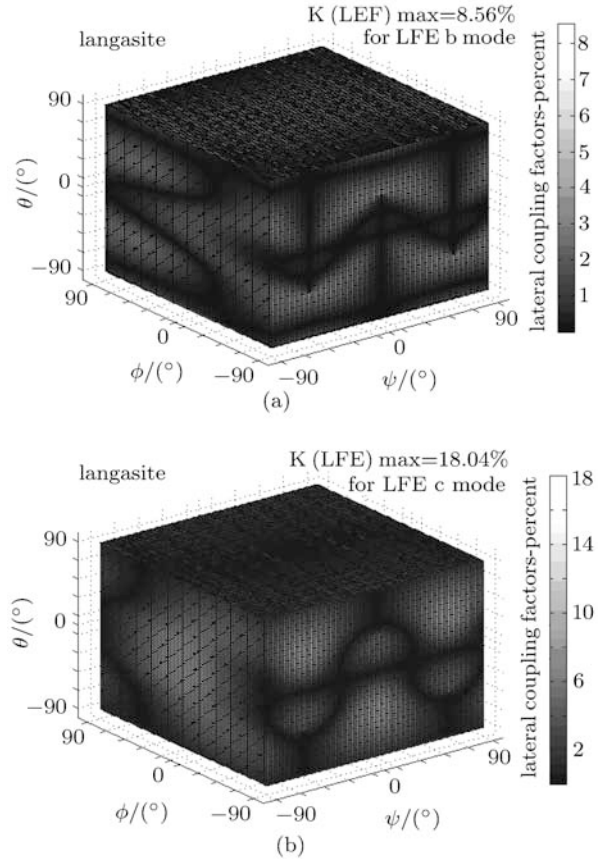


Fig. 3. Lateral coupling factor versus the rotation angles (ϕ, θ, ψ): (a) LFE-b mode and (b) LFE-c mode.

According to the conditions of pure-LFE mode, it is found that for langasite, the cut of (yxl)65° of langasite supports the pure-LFE mode theoretically. The lateral coupling factors for the acoustic wave modes (a, b, c) of (yxl)65° langasite plate are plotted as a function of the electric field angle ψ. The TFE coupling factors of (yxl)65° langasite were also calculated. These results are shown in Fig. 4. These plots are symmetric around $\psi = 0°$. It is found that when $\psi = 0°$, the LFE coupling factor of c mode reaches its maximum value of 8.78% and that of b mode is 0. Besides, the TFE coupling factors of c mode and b mode are both constant of 0. Therefore, when $\psi = 0°$ the pure TSM wave launched by LFE-c mode can be excited; and when $\psi = 0°$, the (yxl)65° langasite device works on pure-LFE mode.

It is found that for langasite, the (yxl)45° cut can satisfy the conditions of quasi-LFE operational mode. The LFE and TFE coupling factors of the acoustic wave modes (a, b, c) of (yxl)45° langasite plate are plotted as functions of the electric field angle ψ, shown in Fig. 5. It is found that when $\psi = \pm 90°$, the LFE coupling factor of c mode is 13.1% and that of b mode is 0. Besides, the TFE coupling factor of c mode is constant of 8.35% and that of b mode is constant of 0. Therefore, when $\psi = \pm 90°$, the TSM can be launched by both LFE-c mode and TFE-c mode simultaneously. When $\psi = \pm 90°$, (yxl)45° LFE devices work on quasi-LFE mode in liquid phase. In gas phase, the TFE-c mode is not effective for no liquid being there can act as a virtual electrode, therefore the TSM can still be excited by a single mode, namely LFE-c mode.

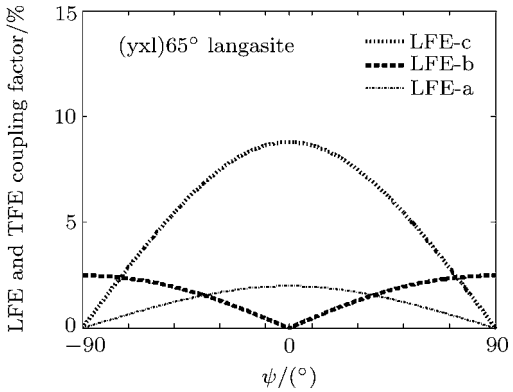


Fig. 4. LFE and TFE piezoelectric coupling factors of (yxl)65° langasite (the TFE-c, TFE-b and TFE-a coupling coefficients are zero).

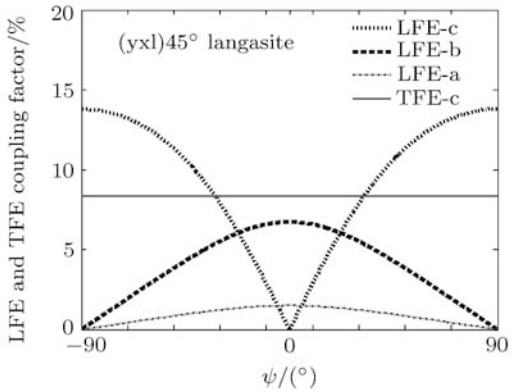


Fig. 5. LFE and TFE piezoelectric coupling factors of (yxl)45° langasite (the TFE-b and TFE-a coupling coefficients are zero).

According to conditions of pseudo-LFE operational mode, it is found that the cut of (yxl)0° of langasite supports the pseudo-LFE operational mode theoretically. The LFE and TFE coupling factors of the acoustic wave modes (a, b, c) of (yxl)0° langasite plate are plotted as functions of the electric field angle ψ as shown in Fig. 6. It is found that the TFE coupling factor of c mode is a constant of 16.0% and that of b mode is a constant of 0. When $\psi = \pm 90°$, the LFE coupling factor for c mode and that for b mode reach to 0. Therefore, when $\psi = \pm 90°$, the pure TSM launched by TFE coupling factor of c mode can be excited. As the effective electrical field of pseudo-LFE mode is a thickness field, the (yxl)0° langasite device works on pseudo-LFE mode under this condition.

2.2. Acoustic wave phase velocity

According to the piezoelectrically stiffened Christoffel equation (1), the phase velocity $V_{\rm a}$ of the TSM launched by the LFE or TFE mode can be achieved[15]

$$k^2 \left(l_{iK} \left\{ c_{KL}^{\rm E} + \frac{(e_{Kj} l_j)(l_i e_{iL})}{l_i \varepsilon_{ij}^{S} l_j} \right\} l_{Lj} \right) v_j = \rho \omega^2 v_i. \quad (1)$$

Here k is the wave vector, l_{iK} is the wave propagating direction matrix, $c_{KL}^{\rm E}$ is the stiffness constants matrix under the condition of constant electric field, l_j is the field direction matrix, l_i is the transpose of l_j, e_{Kj} is the piezoelectric stress constants matrix, e_{iL} is the transpose of e_{Kj}. $\varepsilon_{ij}^{\rm S}$ is the permittivity constants matrix under the condition of constant strain, v_i is the acoustic wave phase velocity, ρ is the material density, and ω is the angular frequency.

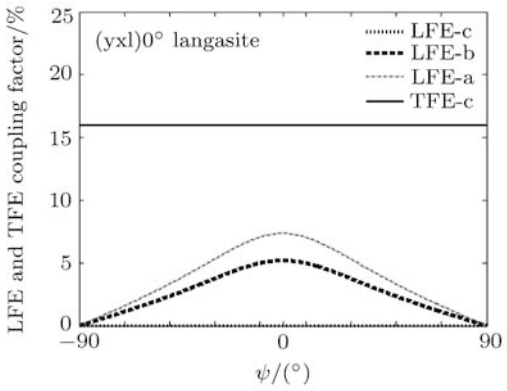


Fig. 6. LFE and TFE piezoelectric coupling factors of (yxl)0° langasite (the TFE-b and TFE-a coupling coefficients are zero).

For (yxl)65° langasite, as the effective wave mode of pure-LFE is LFE-c mode, the phase velocity ($V_{\rm a}$) for this mode is calculated. The result is plotted in Fig. 7. It can be seen that the phase velocity of LFE-c mode TSM is a function of the electric field angle ψ. When $\psi = \pm 90°$, the LFE phase velocity drops to its minimum value of 2942 m/s. When $\psi = 0°$, it reaches to its maximum value of 2953 m/s.

In (yxl)45° langasite, the phase velocities ($V_{\rm a}$) for LFE-c mode and TFE-c mode have ben calculated. The results are plotted in Fig. 8. It can be seen that when $\psi = \pm 90°$, the phase velocity of LFE-c mode drops its minimum value of 3245 m/s; when $\psi = 0°$, it reaches to its maximum value of 3256.6 m/s. However, for TFE-c mode due to the unchanged thickness electric field direction, the phase velocity is a constant value of 3245 m/s.

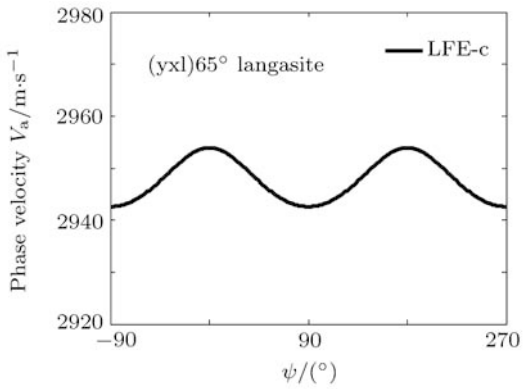


Fig. 7. Phase velocity in (yxl)65° langasite for LFE-c mode.

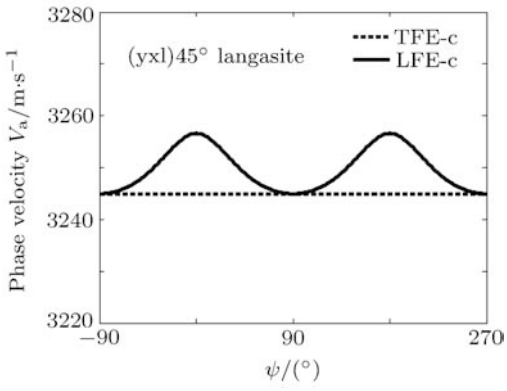


Fig. 8. Phase velocity in (yxl)45° langasite for LFE-c mode and TFE-c mode.

In (yxl)0° langasite, as the effective wave mode of pseudo-LFE is the TFE-c modes, the phase velocity ($V_{\rm a}$) for TFE-c mode has been calculated. The result is plotted in Fig. 9. It can be seen that due to the

unchanged thickness electric field direction, the phase velocity is a constant value of 2739.6 m/s.

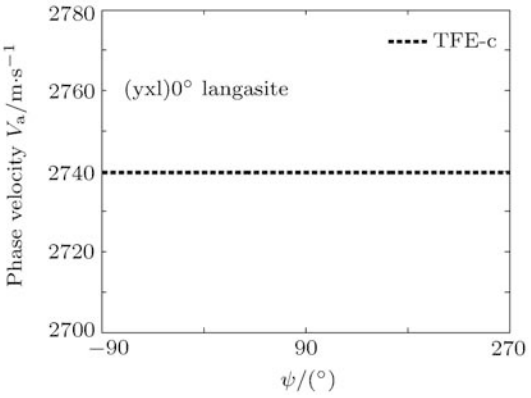


Fig. 9. Phase velocity in (yxl)0° langasite for TFE-c mode.

3. Experimental

For a TSM device, the resonance will take place when a standing wave is formed between the two opposite surfaces of its plate. The wavelength of the standing wave is twice the plate thickness at the fundamental resonant frequency, forming a sequence of resonance frequencies as

$$f_N = \frac{NV_a}{2d}, \tag{2}$$

where N, V_a, and d are positive odd number, acoustic wave phase velocity, and the plate thickness respectively. When $N = 1$, f_1 is a fundamental resonance frequency. Using Eq. (2), the plate thickness d of the TSM device can be determined by the acoustic wave phase velocity V_a in the TSM and the desired resonance frequency f_N.

The thin crystal plate of (yxl)65° langasite was machined to 0.3244 mm thickness, which results in a 4.5515-MHz fundamental resonance frequency for LFE-c mode. For (yxl)45° langasite, the crystal plate was machined to a thickness of 0.2673 mm, which results in a 6.07-MHz fundamental resonance frequency for LFE-c mode and TFE-c mode. In addition, for (yxl)0° langasite, the thin crystal plate was machined to 0.2786 mm in thickness, which results in a 4.917-MHz fundamental resonance frequency for TFE-c mode.

LFE devices for three kinds of different operational modes were designed and fabricated, as shown in Fig. 10. The sensing surface of the devices has no electrode. The devices' plate is a thin disk with a diameter of 25.4 mm. The two electrodes are of semi-circle shape and have a diameter of 12.7 mm,

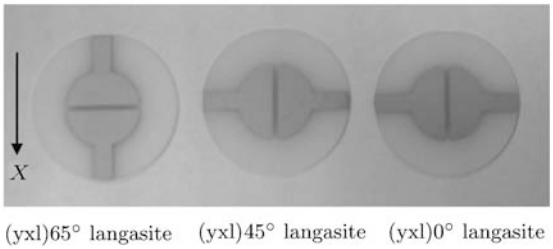


Fig. 10. Photos of three kinds of LFE devices with different cuts.

separated by a gap of 1 mm. For (yxl)65° device, to obtain an electric field angle ψ of 0°, the gap direction is normal to the X axis. For (yxl)45° and (yxl)0° devices, to obtain an electric field angle ψ of 90°, the gap direction is parallel to the X axis. The electrodes were deposited by vacuum evaporation, consisting of about 2-nm thickness adhesion layer of chromium covered by 200-nm thickness of gold.

A crystal sensor holder was used for installing the LFE device and forming a liquid sensing cell. The two electrodes of the device were connected to external electric field source applied through the two contract springs inside the crystal holder. The devices' responses were measured using the impedance analyser Agilent4294A. All experiments were performed at room temperature. During experiment, the experimental results were depicted using impedance characteristic curves.

4. Results and discussion

The measured impedance curve for (yxl)65° langasite in air is shown in Fig. 11(a), where a significant major resonance peak appears. It can be seen that the resonance frequency excited by the lateral electric field is 4.55065 MHz approximately in agreement with the calculated LFE-c mode resonance frequency 4.5515 MHz. The measured impedance curve of (yxl)65° langasite in water is shown in Fig. 11(b), where also a significant major resonance peak appears. The resonance frequency is 4.55035 MHz, which is excited by the same mode as that in air. The lower resonant frequency than that in air can be due to the effect of liquid loading impedance. It is believed that the TSM is launched only by LFE-c mode. Therefore, (yxl)65° langasite LFE device works in pure-LFE

mode, which is in agreement with the calculated result.

From Fig. 11, it can be seen that when the device works in air or water, the TSM can be excited only by LFE-c mode, and no spurious mode emerges due to its moderately large piezoelectric coupling factor. Therefore, besides gas phase, the (yxl)65° langasite LFE device is also suitable for operating in liquid phase. This is greatly helpful for developing liquid sensor using the langasite LFE device.

The measured impedance curve for (yxl)45° langasite in air is shown in Fig. 12(a), where a significant major resonance peak appeares. It can be seen that the resonance frequency excited by lateral electric field is 6.057 Hz approximately in agreement with the calculated value for LFE-c mode, namely 6.07 MHz. Therefore, the frequency of 6.057 MHz can be believed to be that for LFE-c mode. The impedance curve of (yxl)45° langasite in water is shown in Fig. 12(b), where there are two significant major resonance peaks observed. The measured resonance frequency for LFE-c mode decreases to 6.056225 MHz due to the effect of liquid loading impedance. The frequency of the new peak, namely 6.064 MHz, approximately agrees with the calculated TFE-c mode resonance frequency of 6.07 MHz, and can be believed to be that for TFE-c mode. It can be seen that the TSMs are excited by LFE-c mode and TFE-c mode simultaneously, thus, (yxl)45° langasite LFE device works in quasi-LFE mode.

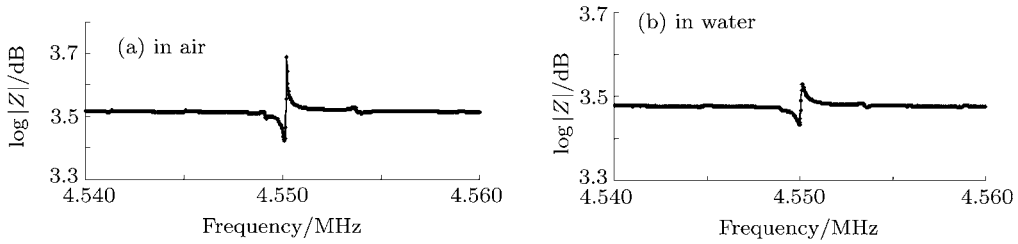


Fig. 11. The measured impedance curves for (yxl)65° langasite LFE device in air (a) and in water (b).

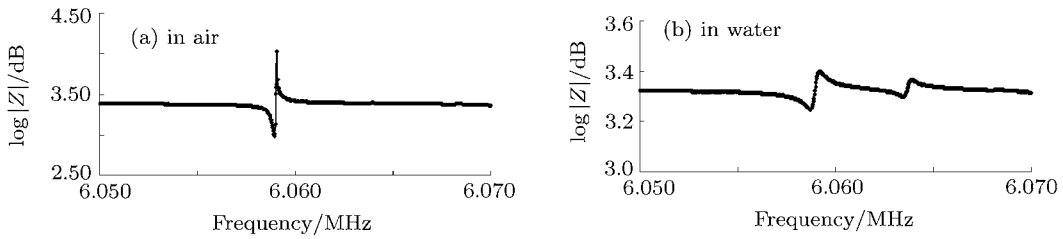


Fig. 12. The measured impedance curves for (yxl)45° langasite LFE device in air (a) and in water (b).

From the above analyses, it is shown that when the device operates in air, the TSM can be excited only by LFE-c mode. Therefore, the (yxl)45° langasite LFE device is only suitable for operating in gas phase.

The measured impedance curve for (yxl)0° langasite in air is shown in Fig. 13(a), where there is no resonance peak observed. However, when only the bare crystal surface of the device is exposed to de-ionized water, there appears a resonance peak at the frequency of 4.9152 MHz, as shown in Fig. 13. The reason for the phenomena is that the de-ionized water acts as a virtual electrode and then a serial configuration of two TFEs is formed. The measured resonance frequency of 4.9152 MHz is near the calculated b-mode TFE resonance frequency of 4.9170 MHz. But the resonance frequency is a little lower than the calculated resonance frequency as a result of the liquid loading impedance's influence. Therefore, the device is considered working in pseudo-LFE mode. This is in agreement with the calculated result.

From the above analysis, it can be seen that when the device operates in water, the TSM can be excited only by TFE-c mode. Therefore, the (yxl)0° langasite LFE device is suitable for operating in liquid phase. This

cut is valuable for developing liquid sensor using langasite LFE device operating in pseudo-LFE mode.

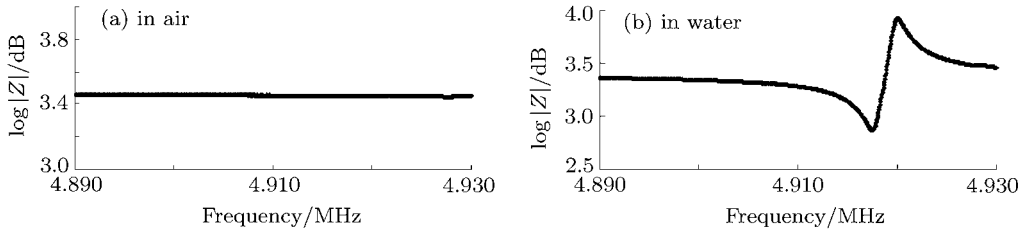


Fig. 13. The measured impedance curves of (yxl)0° langasite LFE device in air (a) and in water (b).

5. Conclusion

Bulk acoustic wave propagation properties of langasite excited by lateral electric field have been investigated. The theoretical analysis shows that (yxl)65°, (yxl)45°, and (yxl)0° support pure-LFE mode, quasi-LFE mode, and pseudo-LFE mode of langasite crystal respectively. Several devices, by using the three different cuts, were fabricated and tested. The experimental results agree well with the theoretical prediction. The results provide the cuts of langasite for LFE devices suitable for different operating environments. Especially, it is shown that when in liquid, the (yxl)65° langasite LFE device can achieve pure TSM wave excited by lateral field, and no spurious mode emerges due to its moderately large piezoelectric coupling factor. In addition, (yxl)0° langasite LFE device is also found suitable for operating in liquid phase. The results are important for designing high performance liquid LFE sensors on langasite substrate.

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