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cknowledgement.....

本文提出了微米超晶格(声学超晶格和光学超晶格)的概念,系统研究了微米超晶格中经典波(声波和光波)的激发和传播,发展了一些新的概念和理论,发现了若干新现象,它们包括;

- 1、通过对声学超晶格中弹性波的激发和传播的理论研究, 首次发现了二类共振现象(1,2,3):
- A、主共振, 决定于声学超晶格周期的大小而不是决定于其总厚, 因此有可能用声学超晶格制备工作频率高达几千兆的声学器件。
- B、卫星共振, 决定于声学超晶格的总厚, 可通过调节晶体生长的工艺参量, 使超晶格中的正、负畴厚度相等而消除之。
- 2、首次成功地制备了LiNbO, 声学超晶格,验证了所发展的理论,证实了声学超晶格在发展超高频声学器件方面有着重要的应用前景。
- A、利用生长条纹方法,成功地制备了适用于 200~1000 M Hz 声学 器件的超晶格 LiNbO₃ 晶体。
- B、实现了声学超晶格中的高频共振, 定量地验证了所发展的理论⁽¹⁾。
- C、利用声学超晶格研制成功超高频谐振器和换能器的原型。证实了可以通过调节声学超晶格的参数,使器件的阻抗与测量系统的相匹配,从而使其插入损耗近于零⁽⁴⁾。
- 3、在理论上和实验上获得了准周期(Fibonacci)声学超晶格中的超声激发谱,发现了其谱结构具有自相似性。首次在亚宏观超晶格中证实了"倒空间中谱的自相似性是实空间中结构自相似性的反映"这一普遍规律^(5,6)。
- 4、研究了准周期 (Fibonacci) 光学超晶格中的二次谐波谱, 首次发现了光的色散效应破坏了二次谐波谱的自相似性以及类似于 X-射线 衍射或电子衍射中出现的消光现象,并得到了消光规律⁽⁷⁾。

- 5、首次提出了利用准周期 (Fibonacci) 光学超晶格直接得到高效 三倍频的实验方案,并充分地论证了该方案的可行性⁽⁸⁾。
- 6、对二次谐波的发生作了较严格的数学处理,提出了利用反向传播的二次谐波的实验方案,即通过在光学超晶格中使反向传播的二次谐波实现准位相匹配的方案⁽⁹⁾。
- 7、研究了准周期 (Fibonacci) 光学超晶格中的电光效应,获得了因电光效应而产生的透射谱,再次证实了光的色散效应破坏谱结构的自相似性⁽¹⁰⁾。
 - 8、首次对准周期结构中的参量过程提出了位相匹配的概念(8,18)。
- 9、在亚宏观准周期 (Fibonacci) 超晶格中首次获得了经典波反射率的解析表达式,研究了反射谱的性质,得到如下结论⁽¹¹⁾:
 - A、由于光波的色散效应, 其谱结构是非自相似的;
 - B、由于声波无色散效应, 其谱结构是自相似的。
 - C、二者都存在消光效应。

ABSTRACT

The concept of micron superlattice has been proposed in this thesis. The excitation and propagation of classical waves (acoustic and optical waves) in the micron superlattice has been studied systematically. Some new concepts and theories have been developed and some new phenomena have been discovered which include:

1. After the theoretical study of excitation and propagation of elastic waves in the acoustic superlattice, two kinds of resonances are predicted for the first time which are 1-3:

A. The main resonance is determined by the period of the acoustic superlattice, not by its total thickness which makes it possible to fabricate acoustic devices with high frequency up to several gigahertz.

B. The satellite-like resonance is related to thetotal thickness of the acoustic superlattice and is the counterpart of the satellite in x-ray orelectron diffraction. It can be eliminated by adjusting the technological parameters in crystal growth tomake thethicknesses of positive and negative domains equal.

2. For the first time we have successfully prepared the acoustic superlattice of LiNbO₃ crystals and thus verified the theory developed by us. These show that the acoustic superlattice is very promising in developing the high-frequency acoustic devices.

A. Using the method of growth striations, we have successfully prepared the acoustic superlattice of Linbo.

crystals applicable to the fabrication of acoustic devices operating at frequencies of 200-1000MHz.

B. With acoustic superlattices we have realized high-frequency resonance and verified the theory quantitatively 1.

C.Prototypes of high-frequency resonators and transducers have been fabricated successfully with the acoustic superlattice. We have confirmed that by adjusting the parameters of the acoustic superlattice, the acoustic devices can be made to match the 50Ω measurement system, thus an insertion loss near 0dB can be obtained.

3.We have obtained the ultrasonic spectrum of quasiperiodic acoustic superlattice both theoretically and experimentally which shows self-similarity. This is the first time that the universal law, the self-similarity of spectrum in reciprocal space being the reflection of the self-similarity of the structure in real space, has been observed in submacroscopic superlattices 5-6.

4. We have studied the second harmonic spectrum of the Fibonacci optic superlattice. For the first time we have discovered that the spectrum is non-self-similar due to the dispersive effect of light and that there exists an extinction phenomenon much similar to that in x-ray or electron diffraction. A general extinction rule has been deduced.

5. For the first time we have proposed an experimental scheme for obtaining a third harmonic generation (THG) with high

efficiency by using a Fibonacci optical superlattice and fully demonstrated its feasibility.

6.A more rigorous mathematical treatment for the second harmonic generation has been performed. An experimental scheme for obtaining a backward-going second harmonic through quasi-phase-matching in the Fibonacci optical superlattice has been proposed.

7. We have studied the electrooptic effect in the Fibonacci optical superlattice and obtained the transmission spectrum by electrooptic effect. Once again we have proved that the dispersive effect of light destroys the self-similar structure of the spectrum ¹⁰.

- 8. For the first time we have proposed the phase matching concept for parametric processes in quasiperiodic micron superlattices 8,10.
- 9. For the first time we have derived the analytical expression of reflectivity of acoustic and optic waves propagating in submacroscopic quasiperiodic superlattice and studied the properties of the reflectivity. Following conclusions have been obtained 11:
- A. For optical waves, the reflectivity is non-self-similar due to the dispersion of the refractive index.
- B. For acoustic waves, the reflectivity is self-similar due to the non-dispersion of the sound velocity.
 - C. For both, there exists an extinction phenomenon.

参考文献

(已发表, 待发表以及正在撰写的论文)

- 1. Yong-yuan Zhu(朱永元), Nai-ben Ming(闵乃本), Wen-hua Jiang(姜文华), and Yong-an Shui(水永安), "High-frequency resonance in acoustic superlattice of LiNbO₃ crystals", Appl. Phys. Lett. 53(1988)2278-2280.
- 2. Yong-yuan Zhu(朱永元), and Nai-ben Ming(闵乃本),
 "Ultrasonic excitation and propagation in acoustic
 superlattice",(submitted).
- 3. Nai-ben Ming(闵乃本), Yong-yuan Zhu(朱永元), and Duan Feng(冯端), "Ferroelectric crystals with periodic laminar domains as the micron superlattices for optic and acoustic processes", Ferroelectrics, 106(1990) 935-940.
- 4. Yong-yuan Zhu(朱永元), Nai-ben Ming(闵乃本), Wen-hua Jiang(姜文华), and Yong-an Shui(水永安), "Acoustic superlattice of LiNbO₃ crystals and its applications to bulk-wave transducers for ultrasonic generation and detection up to 800MHz", Appl. Phys. Lett. 53(1988)1381-1383.
- 5. Yong-yuan Zhu(朱永元), Nai-ben Ming(闵乃本), and Wen-hua Jiang(姜 文 华), "Ultrasonic spectrum in Fibonacci acoustic superlattices", Phys. Rev. B40(1989)8536-8540.
- 6. Li-xing Zhang(张立新), Yong-yuan Zhu(朱永元), and Nai-ben Ming(闵乃本)," Indexing the self-similar reciprocal vectors of Fibonacci quasilattice", (submitted).
- 7. Yong-yuan Zhu(朱永元), and Nai-ben Ming(闵乃本), "Second-harmonic generation in a Fibonacci optical superlattice and the dispersive effect of the refractive index", Phys, Rev. B42(1990)3676-3679.

- 8. Jing Feng (冯京), Yong-yuan Zhu(朱永元) and Nai-ben Ming(闵乃本). "Harmonic generations in an optical Fibonacci superlattice", Phys. Rev. B41(1990)5578-5582.
- 9. Yong-yuan Zhu (朱永元), A. L Shuvalov, Nai-ben Ming(闵乃本), "Backward-going second harmonic generation and quasi-phase-matching in optical superlattice", (in preparation).
- 10 Yong-yuan Zhu(朱永元), and Nai-ben Ming(闵乃本), "Electrooptic effect and transmission spectrum in a Fibonacci optical superlattice", (submitted).
- 11 .Yong-yuan Zhu(朱永元), and Nai-ben Ming(闵乃本), "Reflection of optic and acoustic waves by micron superlattice", (in preparation).

INTRODUCTION

Superlattices have opened an area to the development of new materials that do not exist in nature. The flexibility in the choice of superlattice materials allows superlattices to exhibit a wide range of tailorable properties which are of interest for scientific and device purposes.

The concept of superlattice was initiated by Rsaki and Tsu in 1969¹. In subsequent twenty years, the study of superlattice has experienced two stages according to its symmetry. 1970 began the first stage which was devoted to the investigation of periodic superlattice. The second stage, the study of quasiperiodic superlattice was triggered by the discovery of quasicrystals in 1984², however the periodic one still remained a subject. Recently, the quasiperiodic structure is of increasing interest because it is intermediate between periodic and random³.

In the early years, most of work was focused OD semiconductor superlattices fabricated from alternating layers of crystalline III-V materials which show many interesting transport and electrical properties associated with quantum size effects 4,5. At that time, it 788 universally accepted that superlattices could produced if the constituents have the same crystal structure and closely matching lattice parameters. Otherwise the density of defects would be so high that the phenomena associated with quantum size effects would be obscured. However, these stringent requirements can be relaxed, which results in two different types of superlattices.

The first came the lattice-mismatched superlattice, called strained-layer superlattice in 1974^{6,7}. All of the lattice mismatch is accommodated by layer strains without generating misfit dislocations if the layers are kept sufficiently thin. Layer strains provide a new mechanism for tailoring the properties of superlattices.

Next, in 1983, Abeles et al. reported for the first time the amorphous semiconductor superlattice. They found that the range of materials from which superlattices can be fabricated can be extented to hydrogenated amorphous semiconductors. The interfaces are essentially defect free and nearly atomically sharp. The new superlattices provide novel ways of studying fundamental properties of amorphous semiconductors and have potential for technological applications because large area films of them can be deposited cheaply 9.

Since in all these three types of semiconductor superlattices, electron movement plays a decisive role, the characteristic length, i.e., the period of periodic superlattice or the modulated wavelength of quasiperiodic superlattice, should be reduced to less than the electron mean free path. Hence, by their characteristic length, these superlattices may be called the nanometer superlattices.

With the development of investigation, it was found that the constituents of superlattice may not be restricted to semiconductors. Other materials can also be used.

Schuller 10 was the first to prepare in 1980 the metallic superlattice. It shows some novel physical characteristics such as superconductivity, magnetism and

elastic properties etc 11,12. It can be used as optical elements for soft x-rays and vacuum ultraviolet. According to its characteristic length, the metallic superlattice can be classified into the nanometer superlattice.

The superlattice materials can be further extented to elastic and dielectric substances. In these materials, the important wave processes are mostly optic and acoustic. Superlattices with characteristic lengths comparable with the wavelengths of lights and ultrasonic waves may participate in these wave processes to produce significant effects.

The elastic superlattice was appeared in 1980¹³. It is made up of elastic rods imbedded in a matrix with different elastic properties. One dimensional or two dimensional structure can be made. Such materials, by combining two or more component phases, are able to realize a performance superior to any of the individual phases alone and are now finding applications in many fields¹⁴. Owing to its technical restrictions¹³, its characteristic length is about of the order of milimeter. Thus, it may be identified as milimeter superlattice.

All the superlattices mentioned above are essentially heterostructures. In 1980s we have developed a different type of superlattice made of dielectric media 15-19. It consists of a single crystal with 180 laminar ferroelectric domain structures and can be prepared by the Czochralski method directly. In this kind of superlattice, the physical tensors such as those with odd-rank change their signs regularly. With this material, we verified the

quasi-phase-matching theory proposed by Bloembergen et In 1985, Feisst et al. 23 repeated the experiment with the superlattice of less periods. More al. realized the second-harmonic recently. Magel et generation of blue light in periodically poled LiNbOq. Since 1988, our group (including the work of this thesis) has systematically developed the theories for acoustic and optic kind of periodic and quasiperiodic this superlattice which have been demonstrated by experiments, thus opened up a new field of studying new materials and novel devices through the control of microstructures of materials 25-33. It is certain that its importance will attract more and more attention in the future. In the field discussed above, the superlattice with its characteristic length in the range of microns is needed and so it is named the micron superlattice.

More generally, the modulation of physical tensors can be realized not only through the modulation of 180° ferroelectric domain structures, but also through the modulation of non-180° ferroelectric domains, crystallographic orientation of through the modulation of the composition thus there may be other types of micron superlattices, such as non-180° laminar ferroelectric domain superlattice, laminar twin superlattice and compositionally modulated superlattice, etc. Whereas the 180° laminar ferroelectric domain superlattice is only one special example of them.

Because these new types of superlattices possesses novel acoustic and optic effects and can be used to

fabricate special ultrasonic and photoelectronic devices, it is also named the acoustic or optic superlattice.

The importance of the latter two types of superlattices lies not only in their practibility but also in their similarity in some respects to quantum mechanical systems. Maynard pointed out that electrons (considered as "waves") in a quantum mechanical system can be imitated by classical waves (elastic or electromagnetic) owing to the fact that they are all governed by a wave equation. With classical systems, direct measurement of eigenvalues, eigenfunctions and properties of quasicrystals 27,41 was realized. In a random classical system, even the phenomenon of Anderson localization was observed these are difficult if not impossible to obtain in the quantum mechanical systems.

Now there are superlattices of different kinds. The constituents of the superlattice can be semiconductors, metals, elastic or dielectric materials etc. The characteristic length of the superlattice can be of the order of nanometer, micron or milimeter. The symmetry can be periodic or quasiperiodic. Each type of superlattice possesses some unusual properties that others do not.

This thesis only deals with the micron superlattice (acoustic and optic superlattice). The work is primarily original, having as its main aim the understanding of the nature of the micron superlattice.

The thesis contains six chapters.

The first chapter is mainly background material. We discuss the fabrication of the micron superlattice and the

mechanism of its formation. The transformation properties of its physical parameters are presented. Chapters 2 and 3 devoted to the study of excitation and propagation of ultrasonic waves through the piezoelectric effect in the micron superlattice. Chapter 2 deals with the periodic structure while chapter 3 is for the quasiperiodic one. Experiments are performed which are in good agreement with the theory. Chapters 4 and 5 analyse theoretically the transmission of light waves in the micron superlattice. The nonlinear optical effect is tackled in chapter 4 and the electrooptic effect in chapter 5. In chapter 6, the reflection of light and acoustic waves by the micron superlattice is discussed.

References of introduction:

- 1.L.Esaki, and R.Tsu, IBM J.Res.Develop.14, 686 (1970).
- 2.D.Schechtman, I.Blech, D.Gratias, and J.W.Cahu Phys.Rev.Lett.53, 1951 (1984).
- 3.D.Levine, and P.J.Steinhardt, Phys.Rev. B34, 596 (1986).
- 4.L.L.Chang, Synthetic Modulated Structure, edited by B.C.Giessen (Academic Press, New York, 1985).
- 5.K.Ploog, and G.H.Dohler, Advances in Physics 32, 285 (1983).
- 6.J.W.Matthews, and A.E.Blakeslee, J.Cryst.Growth 27, 118 (1974).
- 7.G.C.Osbourn, IEEE J. Quan. Electron. QE-22, 1677 (1986).
- 8.B.Abeles, and T.Tiedge, Phys.Rev.Lett.51, 2003 (1983).
- 9.B.Abeles, Superlattices and Microstructures 5, 473 (1989).

- 10.I.K. Schuller, Phys. Rev. Lett. 44, 1597 (1980).
- 11.I.K. Schuller, and C.M. Falco, Surf. Sci. 113, 443 (1982).
- 12.D.B.McWhan, Structure of Chemically Modulated Films, in Synthetic Modulated Structures, edited by L.L.Chang and B.C.Giessen (Academic Press, New York, 1985).
- 13.B.A.Auld, in Proceedings of International Workshop on Acoustic Nondestructive Evaluation, Nanjing, M1 (1985).
- 14.T.R.Gururaja, W.A.Schulze, R.E.Newnham, B.A.Auld, and Y.Wang, IEEE Trans.on Sonics and Ultrasonics, SU-32, 481 (1985).
- 15.N.B.Ming, J.F.Hong, and D.Feng, J.Mater.Sci.17, 1663 (1982).
- 16.N.B.Ming, J.F.Hong, and D.Feng, Acta Phys.Sin.(in Chinese) 31, 104 (1982).
- 17.N.B.Ming, J.F.Hong, Z.M.Sun, and Y.S.Yang, Acta Phys.Sin. (in Chinese) 30,1672 (1980).
- 18. J. F. Hong, and Y. S. Yang, Acta Opt. Sin. (in Chinese) 4, 821 (1986).
- 19.J.F.Hong, Z.M.Sun, Y.S.Yang, and N.B.Ming, Physics (in Chinese)9, 5 (1980).
- 20.D.Feng, N.B.Ming, J.F.Hong, J.S.Zhu, Z.Yang, and Y.N.Wang, Appl.Phys.Lett.37, 607 (1980).
- 21.Y.H.Xue, N.B.Ming, J.S.Zhu, and D.Feng, Acta Physica Sinica (in Chinese) 32, 1515 (1983); Chinese Phys.4,
- 554 (1984)
- 22.J.A.Armstrong, N.Bloembergen, J.Ducuing, and P.S.Pershan, Phys.Rev. 127, 1918 (1962).
- 23.A.Feisst, and P.Koidl, Appl. Phys. Lett. 47, 1125 (1985).
- 24.G.A.Magel, M.M.Fejer, and R.L.Byer, Appl.Phys.Lett.56,

- 108 (1990).
- 25.Y.Y.Zhu, N.B.Ming, W.H.Jiang, and Y.A.Shui,
 Appl.Phys.Lett.53, 2278 (1988).
- 26.Y.Y.Zhu, N.B.Ming, W.H.Jiang, and Y.A.Shui, Appl.Phys.Lett.53, 1381 (1988).
- 27.Y.Y.Zhu, N.B.Ming, and W.H.Jiang, Phys.Rev.B40, 8536 (1989).
- 28. J. Feng, Y. Y. Zhu, and N. B. Ming, Phys. Rev. B41, 5578 (1990).
- 29. J. Feng, and N.B. Ming, Phys. Rev. A 40,7047 (1989).
- 30.Y.Y.Zhu, and N.B.Ming, Phys.Rev.B42,3676 (1990).
- 31. J. Chen, Q. Zhou, J. F. Hong, W. S. Wang, N. B. Ming, and D. Feng, J. Appl. Phys. 66, 336 (1989).
- 32.D.Feng, N.B.Ming, J.F.Hong, and W.S.Wang, Ferroelectrics 91, 9 (1989).
- 33.N.B.Ming, Y.Y.Zhu, and D.Feng, Ferroelectrics 106,935 (1990).
- 34.B.Hadimioglu, L.J.La Comb, Jr., D.R.Wright, B.T.Khuri-Yakub, and C.F.Quate, Appl.Phys.Lett.50, 1642 (1987).
- 35.H.K.Wong, G.K.Wong, and J.B.Ketterson, J.Appl.Phys.53, 6834 (1982).
- 36.C.F.Dewey, and L.O.Hocker, Appl. Phys. 26, 442 (1975).
- 37.J.D.Maynard, in Physics News in 1988, Physics Today 42, S5 (1989).
- 38.S.He, and J.D.Maynard, Phys.Rev.Lett.62, 1888 (1989).
- 39.B.A.Auld, H.A.Kunkel, Y.A.Shui, and Y.Wang, in 1983 IEEE

 Ultrasonics Symposium proceedings, edited by

 B.R.McAvoy, 554.
- 40.Y. Wang, and B.A. Auld, in 1985 IEEE Ultrasonics symposium

proceedings, edited by B.R.McAvoy, 637.

- 41. J.P. Desideri, L. Macon, and D. Sornette, Phys. Rev. Lett. 63, 390 (1989).
- 42.S.He, and J.D.Maynard, Phys.Rev.Lett.57, 3171 (1986).

CHAPTER 1

The fabrication of micron superlattice, its formation mechanism and the related physical parameters

In this chapter, the fabrication of the micron superlattice (MSL), a single crystal with 180 laminar ferroelectric domains, and the mechanism of its formation are discussed. The relation between the physical parameters, such as tensors of dielectric, elastic, piezoelectric, nonlinear optic and electrooptic etc., in positive and negative ferroelectric domains are deduced. In order to meet the needs of the remainder of the thesis, the fourier transformation of the quasiperiodic function is presented.

§1-1. The preparation of micron superlattice

Czochralski method of crystal growth with doped melts is used to grow single LiNbO₃ crystals. In the process of crystal growth, a temperature fluctuation is introduced into the solid-liquid interface either through an eccentric rotation or through the application of an electric current current current, which results in the growth striations, i.e., the regular variation in dopant concentration of growing crystal. When cooling through the Curie point, crystals with superlattice are fabricated automatically. The periods of the superlattice may be adjusted by choosing suitable pulling rate and rotation frequency or by changing the duration of the electric current pulse. In usual Czochralski growth system, the superlattice with its characteristic length of several microns may be achieved and so is given

the name micron superlattice (MSL). Below we will discuss the formation mechanism of the micron superlattice.

81-2. The mechanism of the formation of micron superlattice

Early in 1966 Nassau, and later in 1976 Peuzin and Tasson found that there is a one-to-one correspondence between the growth striations and the domain forms as-grown LiNbO3 crystals. In 1982, Ming et al measured the yttrium concentration distributions over the rotational growth striations in LiNbO, by means of energy dispersive x-ray analysis in the scanning electron microscope, and between temperature established the correspondence fluctuations, growth rate fluctuations, solute concentration fluctuations and the ferroelectric domain structures, thus reached the conclusions that it is the solute concentration gradient which determines the direction of spontaneous polarization of the domain. They believed that the solutes in a crystal are generally ionized but not completely shielded, especially when the temperature is decreasing and passing through the ferroelectric phase transition. Hence the non-uniform solute distribution is equivalent to non-uniform space-charge distribution in the crystal and a non-uniform local internal electric field is produced in it. Although the field is comparatively small, it can induce the ions of lithium and niobium within the lattice to displace preferentially at a temperature close to the Curie point and thus the crystals with the MSL is formed(see Fig.1-1).

At the same time, Ming pointed out that the amplitude of concentration fluctuation decreases as the frequency of

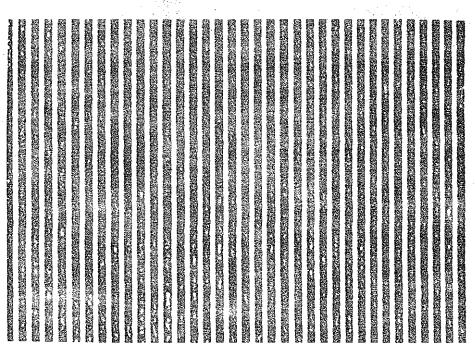


Fig.1-1. Photomicrograph of micron superlattice revealed by etching.

the growth rate fluctuation increases. There exists a cut-off frequency above which the amplitude of concentration fluctuation will approach zero and thus the growth striations will be unclear. He estimated that the cut-off frequency is determined by

$$f_c = D_s / \delta_c^2$$
, (1-1)

where D is the solute diffusion coefficient and δ is the thickness of the solute boundary layer. Applying it to LiNbO crystals, he obtained the minimal space of the growth striations is about one micron, agreement with the experiment.

The problem was further investigated by J.Chen in 1989. He proved that the local internal electric field induced by the solute concentration gradient consists of two parts, one is the space-charge field and the other the elastic equivalent field. He pointed out that the Gibbs free energy is minimized only when the spontaneous polarization \overrightarrow{P} has the same direction as \overrightarrow{E} . Thus a non-uniform local internal electric field can induce a non-uniform distribution of \overrightarrow{P} , i.e., regular laminar ferroelectric domain structures are formed.

As for ferroelectric crystals belonging to 3m point group such as LiNbO $_3$, since the spontaneous polarization \overrightarrow{P}_s is along the z direction, only ferroelectric domains with their polarizations of opposite directions can exist, normally called the 180° domain. Below we will restrict ourselves to this kind of crystals.

§1-3.Relationship of physical parameters between positive and negative ferroelectric domains

According to reference [9] or the crystallographic relationship between positive domain and negative domain², the coordinate systems associated with these two types of domains are as shown in Fig.1—2. These two coordinate systems are interrelated by a 180 rotation about the x-axis which can be expressed by a coordinate transformation matrix

$$A = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{bmatrix} \tag{1-2}$$

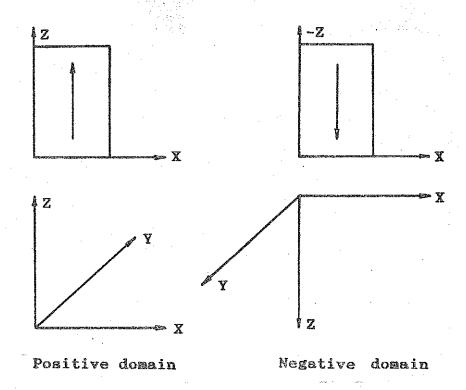


Fig.1-2. The coordinate systems for positive and negative domains

Below we will take the coordinate system for positive domains as the reference.

The physical parameters of LiNbO₃ such as dielectric, elastic, piezoelectric, nonlinear optic and electrooptic coefficients etc. can all be expressed as tensors. In the reference coordinate system, the relations of these parameters between positive and negative domains are as follows.

For dielectric tensor(second-rank)

$$\varepsilon_{ij}^{\prime} = \sum_{i} a_{il} a_{jm} \varepsilon_{lm} . \qquad (1-3)$$

For elastic tensor(forth-rank)

$$C'_{ijkl} = \sum_{im}^{a} a_{im}^{a} a_{im}^{a} c_{im}^{c}$$
mnop
$$(1-4)$$

For third-rank tensors(piezoelectric, nonlinear optic and electrooptic)

$$T'_{ijk} = \sum_{ijk} a_{il} a_{jm} a_{kn} T_{lmn} . \qquad (1-5)$$

Where a_{ij} are elements of matrix A as shown in Eq.(1-2). The quantities with prime are for negative domains, those without prime are for positive domains.

For LiNbO $_3$ crystals, by using Eqs.(1-2)-(1-5), it is easy to show that

$$\varepsilon_{ij}^{\prime}=\varepsilon_{ij}^{\prime}$$
, (1—6)

$$C'_{ijkl} = C_{ijkl}, \qquad (1-7)$$

$$T'_{i,jk} = T_{i,jk} {1...8}$$

That is, the tensors with even-rank remain the same, whereas the tensors with odd-rank change their signs. Clearly, for processes which are associated with the even-rank tensors in a crystal with its symmetry of 3m point group, the crystal is homogeneous and is not a superlattice. Only for those physical effects connected with the odd-rank

tensors, the crystal shows inhomogeneity and therefore is a superlattice.

\$1-4.Description of quasiperiodic superlattice

The initial description of a quasiperiodic superlattice (Fibonacci superlattice) was given by Merlin et al 10. Two different building blocks A and B, each made up of two different materials, are layered according to a well-prescribed rule, ABAABABA... X-ray and Raman scattering measurements have revealed the special features of this non-periodic heterostructure.

Likewise, here we define our Fibonacci MSL. It also consists of two fundamental blocks A and B. The width of these two blocks are different, each composed of one positive and one negative ferroelectric domain. Generally, there are two types of domain configurations as shown in Figs.1—3(a) and 1—4(a), where we choose $I_{A1} = I_{B1} = I$. The quasiperiodicity can be realized by a Fibonacci sequence of blocks A and B fulfilling relations 10

$$S(1) = |A|, S(2) = |AB|, S(3) = |ABA|,$$

$$S(4) = |ABAAB|, S(n) = |S(n-1)S(n-2)|.$$
 (1-10)

In chapters 4 and 5, we have defined

$$I_{A2} = I(1+\eta)$$
,

$$I_{\rm B2} = I(1-t\eta)$$
, (1—11)

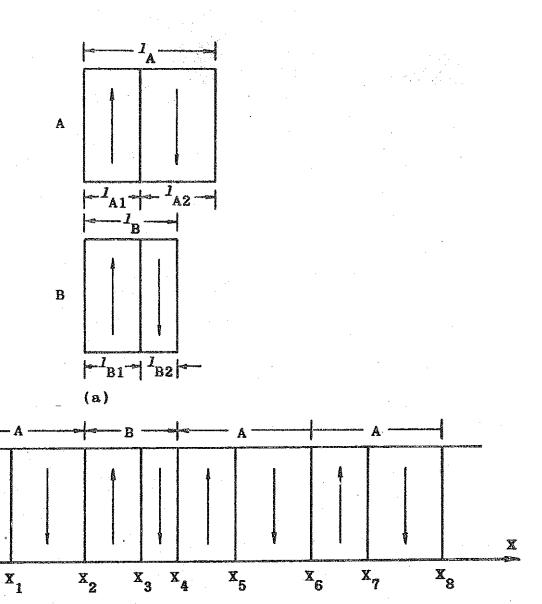


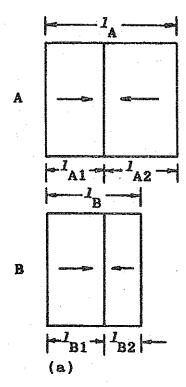
Fig.1—3. The Fibonacci micron superlattice made of a single LiNbO₃ crystal with domains arranging along the x-axis. (The arrows indicate the directions of the spontaneous polarization). (a) The two building blocks, each composed of one positive and one negative ferroelectric domains. (b) Schematic diagram of the superlattice.

where I, t, η are adjustable parameters.

(b)

OX

The Fibonacci superlattice thus obtained is depicted in Figs. 1-3(b) and 1-4(b).



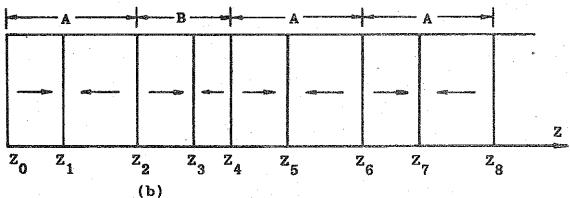


Fig.1-4. The same as Fig.1--3 but for the domains arranging along the z-axis.

Here we should point out that in this thesis the meaning of the term "quasiperiodic superlattice" is the same as the term "Fibonacci superlattice", unless otherwise

specified.

 $\S1-5$. The representation of reciprocal vector of quasiperiodic MSL

For periodic superlattice, a reciprocal vector $|\overrightarrow{G}_m| = 2\pi m/L$ can be used to describe the periodicity. Here L p is the period of the structure and m is an integer. The direction of \overrightarrow{G}_m is along the normal of the domain boundary. The \overrightarrow{G}_m vector participates in many interactions taking place in dielectric media.

The quasiperiodicity of the quasiperiodic superlattice can also be described by reciprocal vectors. They can be obtained through the Fourier transform of a quasiperiodic function which will be encountered frequently in this thesis.

In the previous section, we have proved that the signs of the elements of the third-rank tensor are opposite in positive and negative domains. Hence in this kind of material, the third-rank tensor is no longer a constant, but a quasiperiodic function of the coordinate, and can be expressed as

$$T_{i,jk}(u) = T_{i,jk}f(u),$$
 (1-12)

where u can be the coordinate x or z, T is a constant and

 $f(u) = \begin{cases} +1, & \text{if } u \text{ is in the positive domain,} \\ -1, & \text{if } u \text{ is in the negative domain.} \end{cases}$

Because f(u) is a quasiperiodic function, it must be expanded as a Fourier integral

$$f(u) = \int f(k)e^{-iku}dk , \qquad (1-13)$$

and

$$f(k) = \frac{1}{2\pi} \int f(u) e^{iku} du = \frac{1}{ik\pi} \left\{ \sum_{j} e^{iku} 2j + 1 + e^{i\pi} \sum_{j} e^{iku} 2j \right\}, \quad (1-14)$$

where $\{u_n\}$ are the positions of the ferroelectric domain boundaries (Figs. 1—3 and 1—4).

In Eq.(1-14), the terms inside the curly braces comprise the structure factor, which is divided into two parts with one part lagging behind the other by a phase $e^{i(kl+\pi)}$. Thus Eq.(1-14) can be written as

$$f(k) = \frac{2}{\pi k} e^{i\frac{1}{2}k} \int_{j}^{k} e^{iku} du du$$
 (1-15)

For an infinite array with $l_A/l_B = \tau$, by the use of the direct l_A or the projection method l_A , Eq.(1-15) transforms to

$$f(k) = \frac{2}{\pi} \sum_{m,n} e^{i(\frac{1}{2}kI - X_{m,n})} \frac{\sin \frac{1}{2}kI}{k} \frac{\sin X_{m,n}}{X_{m,n}} \delta(k - G_{m,n}) . \qquad (1-16)$$

Here

$$X_{m,n} = \frac{\pi \tau^2 (m\tau - n)}{1 + \tau^2}$$
, (1-17)

$$G_{m,n} = \frac{2\pi (m+n\tau)}{D} , \qquad (1-18)$$

with

$$D = \tau I_A + I_B$$
 (1—19)

Substituting Eq.(1-16) into Eq.(1-13), we have

$$f(u) = \frac{2}{\pi} \sum_{m,n} e^{\frac{i(\frac{1}{2}G_{m,n}I - X_{m,n})}{G_{m,n}}} \frac{\sin \frac{1}{2}G_{m,n}I \sin X_{m,n}}{G_{m,n}} e^{-iG_{m,n}u} . (1-20)$$

Here G is the reciprocal vector of quasiperiodic superlattice. Analogous to the periodic one, it is reasonable to anticipate that it may play an significant role in interactions proceeding in the quasiperiodic superlattice.

References of chapter 1:

- 1.N.B.Ming, J.F.Hong, and D.Feng, J.Mater.Sci. 17, 1663 (1982).
- 2.A.Rauber, in Current Topics in Materials Science, edited by E.Kaldis and H.J.Scheel (North-Holland, Amsterdam and New York, 1977), vol.1, 481.
- 3. J.F. Hong, and Y.S. Yang, Acta Opt. Sin. (in Chinese) 4, 821 (1986).

- 4.K.Nassau, H.J.Levinston, and G.M.Loiacino, J.Phys.Chem.Solids 27, 983 (1966).
- 5.J.C.Peuzin, and M.Tasson, Phys.Status Solidi (A) 37, 119 (1976).
- 6.N.B.Ming, J.F.Hong, and D.Feng, Acta Phys. Sin. (in Chinese) 31, 104 (1982).
- 7.N.B.Ming, Physical fundamentals of crystal growth (Scientific and technological press, Shanghai, 1982), (in Chinese).
- 8. J. Chen, Q. Zhou, J. F. Hong, W. S. Wang, N. B. Ming, and D. Feng, J. Appl. Phys. 66, 336 (1989).
- 9.Standards on piezoelectric crystals,
 Proc.IRE.37,1378 (1949).
- 10.R.Merlin, K.Bajema, R.Clarke, F.Y.Juang, and P.K.Bhattacharya, Phys.Rev.Lett.55, 1786 (1985).
- 11.D.Levine, and P.J.Steinhardt, Phys. Rev. B34, 596 (1986).
- 12.R.K.P.Zia, and W.J.Dallas, J.Phys.A18, L341 (1985).

CHAPTER 2

Piezoelectric effect of periodic acoustic superlattice

The thicknesses of bulk-wave ultrasonic devices such as resonators and transducers operating at frequencies above 100MHz are too thin to be fabricated by ordinary processing techniques. Here in this chapter we describe a novel method which uses an acoustic superlattice (ASL) to generate detect ultrasonic waves. Some interesting phenomena have been predicted theoretically. First, the resonance frequency is determined by the periodicity of the ASL. Second, the acoustic power emitted by the ASL into the transmission medium is directely proportional to the square of the domain numbers. These two features make the ASL of potential applications in acoustic devices operating at frequencies of several hundred megahertz to several gigahertz. Another is the existence of phenomenon interesting satellite-like resonance frequency, which is related to the total thickness of the ASL and is the counterpart of the satellite in x-ray or electron diffraction. Using the we have fabricated resonators and transducers with operating frequencies in the range of 500-1000 MHz. We have observed for the first time the satellite-like resonances in the ultrasonic spectrum. The transducers with an insertion loss of nearly 0 dB at 555 MHz and a 5.8% 3 dB have been fabricated 2. Experiments confirm the predictions by the theory.

In the periodic ASL, the positive and negative domains arrange in one dimension, say the z axis (Fig. 2-1). We have already proved that in this kind of material, all odd-rank tensors will change signs from one domain the next. Thus the piczoelectric tensor, being a third-rank one, is no longer a constant through the ASL, but a periodic function of the spatial coordinate z(see Fig.2-1(b)). Under the action of an alternating external electric field, the domain walls, where the piezoelectric coefficient shows discontinuity, can be viewed as sound δ -sources as shown in Fig. 2-1(c). The ultrasonic waves excited by these sound &-sources will interfere with each other. Those satisfying the constructive interference will appear as resonant peaks in the ultrasonic spectrum. This is the physics basis for ultrasonic excitation with the ASL.

The above discussed excitation scheme(Fig. 2-1), is determined by the arranging direction of ferroelectric domains of the ASL, will produce a pure longitudinal wave z axis. There are propagating along the many other excitation schemes. For example, Figs. 2-2 show one which will produce one quasi-longitudinal wave and one quasi-shear wave both propagating along the x axis. Without the loss generality, below we will restrict ourselves the discussion of the excitation of the pure longitudinal wave.

Here we assume that the ASL is arranged along the z axis and that the domain walls lie in the xy plane. The thicknesses of the positive and negative domains are a and b, respectively, with the number of periods equal to N, i.e., the periodicity of the ASL is a+b and its total

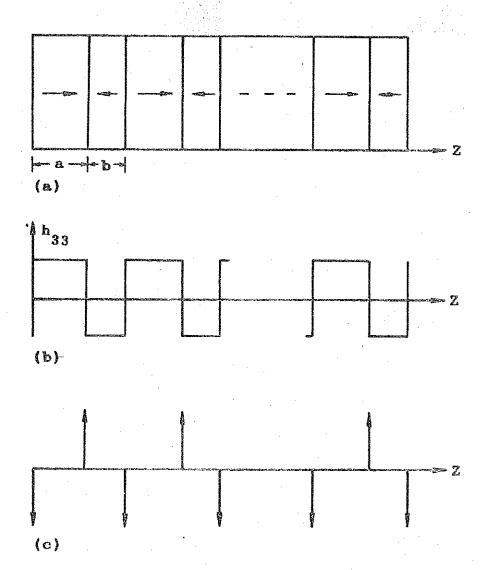


Fig. 2—1.ASL of LiNbO $_3$ for excitation of one pure longitudinal wave. (a)Schematic diagram of ASL (the arrows indicate the directions of the spontaneous polarization). (b)Corresponding piezoelectric coefficient as a periodic function of z. (c)Corresponding sound δ -sources.

thickness is N(a+b), as shown in Fig.2—1(a). Also we assume that the electrode face is parallel to the xy plane and that the transverse dimensions are very large compared with an