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介电体超晶格

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朱永元 王振林 陈延峰 陆延青 祝世宁



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第五章 离子型声子晶体与超构材料

陆延青

以准位相匹配理论的拓展作为物理基础,以具有有序调制畴结构的铁电晶体作为材料基础,以激光与非线性光学效应作为应用牵引,形成了在介电体超晶格光学方向研究探索的一条清晰思路。然而,从介电体超晶格这一名词中可以看出,我们是在介电体中建构一种与天然晶体结构物理对应的人工超结构,而不是仅仅寻求非线性光学效应的增强。正如描绘晶体的物性取决于其组元(即原子)和结构,介电体超晶格通过人工微结构的设计来调控物性,获得不同于其基质材料的崭新性能。从这一点看,介电体超晶格的理论和实验体系,打开了一扇通过微结构获得天然材料所不具备的超常特性的的大门,这和现在十分热门的超构材料(或称之为超材料)殊途同归。

在介电体超晶格的理论体系中,离子型声子晶体概念的提出与验证、拓展,不但呈现出从介电体超晶格到超构材料的异曲同工的演进思路,也体现了我们在超构材料领域的研究特色。离子型声子晶体实际上是与离子晶体对应的一种人工电磁材料。当电磁波入射于离子晶体中,电磁场与晶格振动相互作用,形成与晶体横振动光学声子耦合的模量子,即声子极化激元。为纪念其奠基者黄昆先生,它又被称为“黄子”。这是最先得到研究的极化激元。随后激子极化激元、磁振子极化激元等也相继被提出。

1999年,我们提出了“离子型声子晶体”的概念。详见本章第一篇代表性论文。在5.1中,我们将声子极化激元概念推广到介电体超晶格。与普通声子晶体不同,我们设计制备的离子型声子晶体是由自发极化方向首尾相接的具有周期畴结构的铌酸锂超晶格构成。由于相邻带电畴界可看作是异号的电荷中心,于是一维离子型声子晶体就可以完美对应一维异性离子链。不过,实际上,离子型声子晶体中超晶格振动与电磁场的耦合来源于压电效应。基于压电方程,我们得到了离子型声子晶体中超晶格和电磁波的耦合运动方程组及其极化激元的色散关系。其形式与离子晶体中反映晶格振动与电磁波耦合的黄昆方程完全一致。正是由于电磁波与超晶格的耦合导致的极化激元模,离子型声子晶体中出现了其基质材料不具备的诸多新颖物性,如微波波段的介电反常、电磁吸收,乃至类拉曼散射、类布里渊散射等。这就把若干原本存在于红外波段的经典物理概念推广到微波领域,进一步深化了人们对这些物理过程物理本质的认识。

以介电反常为例,正如5.1文中指出的,在微波波段,“出现了一个介电常数为负的带隙。其对应的折射率为虚数,从而相应波段的电磁波被反射。然而,它与光子晶体中的带隙不一样,却是源于声子和光子相互作用”。因此,基于局域的压电共振以及各单元之间的同相增强,可通过结构设计,获得所需波段的负介电常数。这与通过亚波长局域共振结构的设计,用有效介质方法,得到等效的负磁导率并结合金属的本征负介电常数实现负折射的超构材料在物理思路出现了有趣的一致。只不过,我们是通过压电共振获得负介电常数,而常

规超构材料是通过亚波长电磁共振获得负磁导率。如果我们不拘泥于负折射这一较狭义的超构材料定义,而是着眼于通过微结构设计获得与基质材料截然不同的奇异物性参数,我们就实现了从离子型声子晶体到超构材料的概念的自然过渡。基于离子型声子晶体带来的超晶格声子极化激元这一新思路,我们开展了系列相关的研究工作。在本章中,除 5.1 之外,我们还选取了另外 10 篇代表性论文,从中我们可以看出相关物理思想演进的自然脉络。

离子型声子晶体的概念着眼于压电体超晶格与离子晶体的物理对应。在此物理现象得到证实后,我们即围绕电磁场与超晶格的耦合过程,以微结构对极化激元的调控为主线,开始了进一步深入研究和拓展。5.2 是其中的一个重要工作。在离子晶体中,声子极化激元只能由横向光学声子与光子耦合形成,光子不可能与声学声子发生相互作用。我们发现,由于离子型声子晶体中的光、声耦合实际上源于压电效应,这就使得压电体超晶格中的声子极化激元具有不同的形成条件。除了色散关系曲线的一些典型不同外,在压电体超晶格中,不但横向超晶格声子能够与光子耦合形成声子极化激元,而且还存在纵向超晶格声子与光子耦合形成的声子极化激元,从而扩展了传统的极化激元的概念。其物理原因是,由于压电效应的各向异性,超晶格横振动和纵振动都有可能伴随着超晶格的横向极化,该极化与电磁波耦合,从而导致了新型声子极化激元的产生。

进一步的研究(5.3)表明,由于材料的压电效应,偏振方向相互垂直的电磁波在超晶格中传播时能够发生强烈耦合,这种耦合导致了压电体超晶格中两种新型极化激元的产生。耦合使其中一个模存在禁带,而另一个模则可以通过。这种奇特的现象在传统的晶格中并不存在,说明通过超晶格的设计不但可以在新波段拓展某些物理效应,而且通过合适的微结构和应用场景设计还可以实现更加新颖的物理过程。

当然,既然是超晶格,其结构的人工设计不但带来了特征尺度的可控,也提供了灵活设计超越普通周期性结构的可能。5.4 则是我们将离子型声子晶体和压电体超晶格的概念拓展到准周期结构的一个代表性工作。我们曾在准周期钽酸锂超晶格中成功实现了多波长二次谐波和高效的直接三倍频,将准周期超晶格结构引入压电体,同样出现了一个有趣的物理现象。我们从理论与实验两个方面,研究了具有推广的一维二组元准周期结构的压电体超晶格中的声子极化激元。结果显示,声子极化激元的频率位置由推广的一维二组元准周期结构的倒格矢的位置决定;而带隙大小由材料阻尼系数、机电耦合系数以及傅立叶展开系数的大小决定。小的阻尼系数,以及大的机电耦合系数以及傅立叶展开系数有利于带隙的展宽。结果还显示,具有推广的一维二组元准周期结构比一维周期,以及一维二组元的 Fibonacci 结构在应用上有着更大的灵活性,因此也拓宽了压电体超晶格的声子极化激元的潜在应用范围。我们结合具有推广的一维二组元准周期结构的压电体超晶格中声子极化激元的物理特征,阐述了其在光学、声学以及相关应用领域的可能应用,如制作偏光器、光通信中的多波复用器件,以及环境保护材料等。

同样电磁波与声子的耦合也可发生在压磁材料和压电/压磁复合材料中。在 5.5 一文中,我们与当时在 UCLA 的张翔教授合作,研究了压磁系数周期调制的超晶格中电磁波的传播特性。由于压磁效应,导致了电磁波与晶格振动的耦合,形成一种不存在普通磁性材料中的磁极化激元。令人兴奋的是,正如在离子型声子晶体中出现负介电常数一样,在这种压磁超晶格中,磁导率的异常色散也导致了在某些共振频率出现的负磁导率。于是,通过超晶格的设计,我们实现了负介电常数和负磁导率的人工调控,这何尝不是另一种实现“超构材

料”的途径呢?

很自然地,5.6一文从理论和实验两方面对压电/压磁超晶格开展了讨论,该超晶格材料由压电和压磁薄膜交替生长而成,可以看作压电和压磁的复式超晶格。当电磁场与这种超晶格相互作用时,由于电磁场包括电场和磁场矢量,因此显示了新的特点。由于电场和磁场同时激发了超晶格振动,因而产生了两种宏观激化激元,这两种激化激元通过超晶格振动发生耦合,完全改变了原来各自的特征。实验表明,同一个超晶格可以实现两个谐振频率声波的激发,分别对应于电场激发和磁场激发的振动。这些有趣的结果昭示着压电/压磁超晶格不但是研究电、磁、声间复杂耦合作用的平台,也有着重要的应用前景。这方面的探索研究,特别是与其他超构材料的联系值得深入研究。

当然,光学材料的表面效应的研究吸引了众多关注,表面等离激元的研究可谓如火如荼,离子型声子晶体等材料的表面效应也是十分值得关注的问题。5.7一文是我们在此方向的一项基础性的工作。由于电磁波与压电超晶格的耦合能带来负介电常数,而正是负介电常数才使得金属表面等离激元(SPP)得以存在,那在离子型声子晶体中必然也存在着一种对应的表面模式。基于这一思想,我们预言压电超晶格表面声子极化激元的存在,并导出了其相应的色散关系。由于表面等离极化激元(SPP)在诸多超构材料、微纳光子器件中扮演了重要的角色,或许,我们预言的表面声子极化激元也可在微波、太赫兹(THz)波等重要频段的新型电磁材料和器件的设计上发挥出重要的作用。

在以上的这些代表性工作中,声子起到了十分关键的作用。组成超晶格的各单元可以看作一系列的振子,具有自身的谐振频率。它们之间又在物理上链接在一起,这样,光子通过压电或压磁效应与超晶格的集体振动能量量子即声子,发生相互作用,产生丰富的物性。然而,由于声学谐振系统的尺寸往往较大,对应的频率一般在微波波段,这就极大地限制了其应用范围。

为提高工作频率,要减小各微结构单元的尺寸和单元之间的间隔,这就必须考虑各共振单元之间的耦合。借助于超晶格研究中关注耦合效应的研究思路,研究中我们并不仅仅考虑磁共振单元共振性质的平均效应,而是更加关注单元之间的耦合效应所带来的新奇性质。5.8一文介绍了由两个完全相同的金属开口环组成的磁共振“超构分子”,两个磁共振“原子”之间会形成很强的耦合作用,导致原来单独磁原子的共振模劈裂为磁共振分子的杂化模:成键模与反键模,这种模式劈裂的大小决定于单元之间的距离。

进一步研究发现,两个开口环之间的耦合强度不但与两个单元之间距离有关,还与两个单元相互旋转形成的不同空间立体构型有关,从而可以形成立体超构分子。通过与德国斯图加特大学合作,利用高精密的电子束光刻对准技术,制备出了样品并进行了测量,实验与理论符合的较好。相关工作发表在 Nature Photonics 上,即论文 5.9。

一维原子链是固体物理中分析原子之间耦合作用形成晶格振动波的经典理论模型。由“超构分子”过渡到类似于—维原子链的“超晶格”是十分自然的拓展。我们利用开口环作为结构单元组成—维超晶格。由于单元之间距离很小,我们发现单元之间磁共振耦合也会形成一种类似格波的传播模式,由于这种波是很多磁共振单元耦合形成的集体振荡,我们把它称为磁等离激元波。相关的工作参见论文 5.10。

这样,在介电体超晶格和考虑单元耦合效应的驱动下,我们的超构材料研究由“超构分子”,向—维超构原子链、二维超构晶体演进。显然,向三维系统的过渡是十分自然的趋势。

由于金属纳米颗粒具有独特的光学响应,我们设想将金属纳米棒粒子镶嵌于介电材料中,形成三维立方晶格(见 5.11)。由于电磁波能够激发纳米棒中自由电子的运动,且电子运动在纳米棒两端形成电荷聚集,故产生电偶极矩。后者能够辐射电磁波并与入射波进行干涉。因而在一定的条件下,入射电磁波与金属粒子的等离激元波(极化波)能够发生强烈的耦合。我们称这种独特的三维“超构晶体”为等离激元晶体。通过将等离激元晶体与离子晶体类比,我们发现适用于离子晶体的黄昆方程也能够被推广到等离激元晶体。原先建立于离子晶体的一系列长波光学性质,如极化激元激发、极化激元带隙、介电异常、红外吸收等,都能够在等离激元晶体中产生。

回顾 1999 年,我们在离子型声子晶体中利用黄昆方程来研究入射电磁波与超晶格的相互作用并导出极化激元、介电异常等新颖性质。等离激元晶体与离子型声子晶体呈现出惊人的一致。虽然材料体系、单元共振基质、工作频率发生了巨大的变化,但在这一步步变迁中蕴含的物理脉络却是如此的清晰。我们从离子型声子晶体到超构材料走出了一条特色的发展之路,并且还在不断的发展中。我们相信这类的物理体系中还蕴含着太多的新机制、新现象、新效应,值得我们不断研究探索。这证实了“介电体超晶格”这一材料体系中包含着丰富的物理思想和强大的生命力!

Chapter 5 Ionic-Phononic Crystals and Metamaterials

Yanqing Lu

Dielectric superlattice(DSL) has been a well-recognized research achievement. Although it covers a wide range from optics to acoustics, from classical phenomena to quantum world, the laser and nonlinear optical applications still dominate its progresses in the past years. However, DSL is not just a quasi-phase matched(QPM) nonlinear material. As can be seen from the unique term, “dielectric superlattices”, we are actually building a correspondence between natural crystal structures and artificial superlattices, rather than merely seeking some enhanced nonlinear optical effects. The DSL actually supplies a platform to manipulate various physical properties through the intentional design of microstructures. It opened a door to achieve extraordinary properties that do not exist in natural materials, which is quite similar to a very hot topic, metamaterial.

In the entire theoretical matrix of DSL, the proposal, demonstration and expansion of “ionic type photonic crystal”(ITPC), not only reflect the concept evolution from DSL to metamaterial, but also represent our unique research style in the field of metamaterials.

The so-called ITPC, a counterpart of ionic crystal in the mesoscopic scale, is an artificial electromagnetic(EM) material. As long as the EM wave propagates in an ionic crystals, the EM field would interact with transverse lattice vibration then forms photon polaritons. This phenomenon and related mechanism were firstly revealed by a famous Chinese physicist, Prof. Huang Kun, in 1951. Afterwards, other types of polaritons such as exciton polaritons, magnon polaritons, etc. have also been proposed.

In 1999, we proposed the concept of ITPC that is composed of two piezoelectric media aligned periodically in a superlattice structure.(5.1) Just like a real ionic crystal, there exists the coupling between vibrations of the superlattice and the electromagnetic(EM) field when the EM wave propagates in the ITPC, which results in the long wavelength optical properties. After proposing a simple method for calculating the dispersion relation of a 1-dimentional(1D) phononic crystal, we studied the coupling effects in ITPC and obtained the fundamental equations that describe the coupling between the superlattice vibration and the EM wave. These equations have the same type as the Huang's equations that describe the coupling between the lattice vibration and EM wave in ionic crystals.

From the fundamental coupling equations in ITPC, some long-wavelength optical properties that originally exist in ionic crystal were predicted and experimentally demonstrated based on a LiNbO_3 superlattice. Among them, there are the LST relation, the EM wave absorption, the dielectric abnormality and the polariton mode. Even the Raman scattering and Brillouin scattering in the microwave band might also be expected in an ITPC. The only difference is different response frequencies.

Taking the dielectric abnormality as an example, as we pointed out in the paper, "There is a band gap in which the dielectric constant is negative. The corresponding refractive index becomes imaginary. The incident radiation with these frequencies will be reflected. However, this gap does not originate from the interference of EM waves due to the periodic structure but, rather, originates from the interaction of the photon and the transverse optics phonon." As a consequence, based on the local piezoelectric resonance and in-phase enhancement between different units, we may design suitable domain structure to obtain negative dielectric constant in corresponding wavelength bands. As we know, the key steps toward negative index are negative permeability and negative permittivity. Metallic materials normally have to be used, while our results give the possibility to achieve exotic negative parameter in a dielectric medium. In this case, if we do not stick to strict definition of negative index metamaterial, but rather focus on obtaining extraordinary material parameters through microstructure design, well, we thus realize a natural transition from ITPC to metamaterials.

Afterwards, starting from the induced polariton in ITPC, we further carried out a serial of related research work. In this chapter, we still picked other 10 representative papers in addition to paper 5.1. From these work, we may see a clear trend emerge from ITPC to material.

Paper 5.2 is an important work after the ITPC and its basic properties were demonstrated. In an ionic crystal, the phonon-polariton can only be accompanied with the transverse optical phonons, whereas in the piezoelectric superlattice(PSL), the coupling could occur between the photons and longitudinal phonons as well as transverse phonons. Physically, it is the polarization of dipoles induced by lattice vibrations that interferes with the EM waves in the ionic crystal, whereas it is the polarization of bound charges induced by superlattice vibrations that interferes with the EM waves in the PSL. Due to the anisotropic nature of the piezoelectricity, the polarization of bound charges can be induced by transverse superlattice vibrations, or longitudinal superlattice vibrations as well. In this case, even a transverse polarization can be induced by a longitudinal wave which couples strongly to the EM wave in some particular frequency regions, resulting in the creation of a new type of polariton that does not exist in ionic crystals. The forbidden band associated with the polariton is not due to the Bragg reflection, but rather to the coupling.

Further study(5.3) revealed that the superlattice vibration not only can be excited by and coupled strongly to the incident EM wave, but they can also induce coupling between

two orthogonally polarized EM waves. This coupling results in two types of polariton modes, one is supported in the band gap and the other prohibited. We have noticed that in the literature two interesting cases occurred in the stop band. In one case, the coupled-thin-film structure can support guided-wave plasmon polaritons; and in the other, the propagation of a probe light beam is allowed within the polariton stop band via exciton-biexciton coupling. Our case presents another example, where the phonon polariton is involved. This unusual coupling effect is not present in real lattices.

On the other hand, as we know, the discovery of quasicrystals has fired up a new field of condensed-matter physics and given rise to many practical applications since 1984. For example, the multiwavelength second-harmonic generation and the direct third-harmonic generation have been realized by Zhu et al. in a Fibonacci superlattice. Furthermore, compared with a one-dimensional (1D) two-component Fibonacci superlattice, a 1D two-component generalized quasiperiodic superlattice (GQPSL) possesses more freedom for applications. As a consequence, we further investigate the influence of structural variation of the piezoelectric superlattice upon the phonon polaritons on the basis of a 1D two-component GQPSL. (5.4) We measured the dielectric function of the GQPSL at the microwave region, thereby obtaining the physical information required to deduce the properties of polaritons. The possible applications are discussed, which include WDM devices in optical communications, and for EM wave(or sound wave)-proof materials in environmental protection.

Furthermore, our vision could go beyond piezoelectric materials. In paper 5.5, we collaborated with Prof. Xiang Zhang who was with UCLA at that time to study the propagation of an electromagnetic (EM) wave in a piezomagnetic superlattice with piezomagnetic coefficient being modulated. Because of the piezomagnetic effect, the coupling between the EM wave and vibration of superlattice also can be established, resulting in the creation of a new type of magnetic polariton that does not exist in ordinary magnetic material. At some resonance frequencies, the abnormality of dispersion of permeability introduces negative value and piezomagnetic superlattice can make a kind of negative permeability material. Therefore both negative permittivity and permeability could be realized through the design of suitable microstructures, which might lead to a possible way toward new electro-magnetic materials.

As a straightforward extension, in paper 5.6 we investigated the propagation of EM waves in a piezoelectric-piezomagnetic superlattice (PPS). In a PPS, the electric and magnetic vectors of EM waves could simultaneously couple with the identical superlattice vibration, respectively, due to piezoelectric effect and piezomagnetic effect, which results in magnetoelectric effect. Consequently, two orthogonally polarized EM waves could simultaneously couple with the identical vibration, which would give birth to coupled phonon polaritons. Attributed to this mechanism, in a PPS, the propagation of EM waves varies drastically. EM waves perpendicular to the PPS vector can propagate, while the

propagation is inhibited along the PPS vector in the original band gap of either the piezoelectric superlattice or the piezomagnetic superlattice. The origin of the differences in propagation is analyzed and some potential applications are discussed. These phenomena and applications reveal that the dielectric superlattice has become a versatile platform to study the complex couplings among electronic, magnetic and mechanic processes.

However, just like the surface plasmon polariton mode resulted from the negative permittivity, we also predicted a surface phonon polariton (SPhP) based on ITPCs in paper 5.7. Due to the scale of the superlattice structure, this SPhP mode normally exists in megahertz-gigahertz band. This might be a quite fundamental work since many surface EM mode related phenomena thus could be artificially designed.

Among all these representative works above, the phonons play very critical roles. The superlattice units could be viewed as a serial of oscillators with corresponding resonance frequencies. Due to the physically tight link between adjacent units, the photons thus could interact with the superlattice's phonons through piezoelectric or piezomagnetic effects. Many interesting phenomena were observed. However, due to the large scale of mechanical/ultrasonic oscillation systems, the corresponding frequencies normally exist in microwave band, which limits their high frequency applications.

To raise the operation frequency then further induce coupling between different units, metallic split ring resonators are ordinarily used choices. Due to the background of optical superlattice, not only the magnetic plasmon oscillations of single units may be considered, but also a kind of magnetic dimer (MD) consists of two single split-ring resonators (SSRR) were reported in paper 5.8. The fundamental MD resonances are viewed as bonding and antibonding combinations of individual SSRRs eigenmodes. The so-called hybridization of the magnetic plasmon modes were demonstrated at near-infrared frequencies. A metamaterial made of a large number of coupled magnetic dimers could be utilized as a tunable optically active medium with possible applications in optical elements and devices.

Based on the ideas above, we propose a new concept in nanophotonics, namely stereometamaterials, which refer to metamaterials with the same constituents but different spatial arrangements. In collaboration with Universität Stuttgart in Germany, we theoretically and experimentally study a typical stereometamaterial, i.e., meta-dimers, which consist of a stack of two identical SSRRs with various twist angles. (5.9) We found that the twisting of stereometamaterials offers a way to engineer complex plasmonic nanostructures with a tailored electromagnetic response.

If more coupled units are taken into account, a linear SSRR chain is formed that is just similar to a 1D superlattice. (5.10) These SSRRs interact mainly through the exchange of conduction current, resulting in stronger coupling as compared to the corresponding magneto-inductive interaction. This configuration could be used to support propagation of long range MP polaritons with some new applications in subwavelength transmission lines for a wide range of integrated optical devices.

Eventually, our superlattice inspired metamaterial researches move to 3D cases as reported in representative paper 5.11. We studied the long-wavelength optical properties of a plasmonic crystal composed of 3D nanorod matrix. Just like the original ITPC, we emphasized the concept of the polariton, which is due to the coupling between the photons and the long transverse plasmon wave. The polariton stop band, associated with the coupling effect rather than the Bragg reflection, has been suggested. The results also show that the long-wavelength method developed for an ionic crystal can be applied to a plasmonic crystal and that the artificial and classic lattices may share a common physics. In fact, to study the effect, the Huang's equations were also used.

As we know, the Huang's equations were established originally in ionic crystals where exist a strong coupling between the photons and lattice vibrations. Look back to 1999, we adopted these equations to study the coupling between incident EM wave and the superlattice vibration in an ITPC. Since the similar equations are used, the long-wavelength optical properties suggested in the former, such as the dielectric abnormality, polariton excitation, etc., can also be found in the plasmonic lattice. Although the material systems, resonance units, and related frequencies are quite different, the evolution of the related concepts, mechanisms, and properties are so clear. We developed a unique way from ITPC to metamaterial under the framework of dielectric superlattice. We believe there are still many new physics and phenomena that deserve further and continuous investigation. These are just the powers and charms of dielectric superlattices.