Nonlinear optics of periodic and quasiperiodic structures

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Periodic and quasiperiodic structures are known to possess some very special properties. Recent technological breakthrough in poling techniques in nonlinear materials has rendered nonlinear variants of such structures realizable. We review some of the latest developments in nonlinear optics of these systems. We show that the use of such structures leads to several major advantages like lowering of threshold, increase in efficiency of nonlinear mixing, fabrication of media with better characteristics, etc.

RECENT years have witnessed an intense development of nonlinear optics of periodic and quasiperiodic media[1]. Even the linear counterpart of such structures are interesting because of their very special properties[2,3]. The simplest possible example of such systems can be a periodic or quasiperiodic arrangement of dielectric slabs. Optical properties of such systems can be quite distinct from those of the bulk constituents because of 'structural' dispersion. In fact, the transmission through such structures becomes frequency selective though the constituent layers do not have material dispersion. The most distinct property of periodic structures is the existence of stop gaps[2], while quasiperiodic systems are known to exhibit exotic properties like self-similarity, scaling and multifractal spectra[3]. The other important feature is the ability of such structures to support modes with very high quality factors and large local field enhancement. For example, for finite periodic structures, such modes are located near the edge of the stop gap. These high Q resonances play a very important role in lowering the threshold of various nonlinear optical processes. In fact, diode lasers with sub-watt power levels can now be used to observe most of the nonlinear optical effects.

In the context of layered media (both periodic and quasiperiodic) one needs to distinguish the case of normal incidence from that of oblique incidence[1]. In the former, the system is equivalent to coupled Fabry–Perot cavities, while in the latter, one needs to take into account the possible surface and guided modes of the structure. The case of oblique incidence is more complicated because of the necessary distinction of s- and p-polarizations. It should be noted here that guided mode structures are of tremendous technological value since they offer large power densities over longer propagation distances compared to what can be achieved in bulk samples. Besides, they offer the possibility of integrating various functionalities on the same optical 'chip'.

Till recent times most of the development in nonlinear optics of periodic and quasiperiodic media has been in the theory front, largely due to the lack of proper and comparatively cheap fabrication techniques. A real technological breakthrough has been the domain reversal technique[4] in ferroelectric materials like $LiNbO_3$, $LiTaO_3$, KTP, etc. The most important application of this technique has been in quasi phase matched[5] nonlinear mixing in second order materials[4,6].

Periodicity/quasiperiodicity henceforth needs to be understood in a much broader perspective. For example, the system can be composed of periodic (quasiperiodic) arrangement of dielectric slabs with given optical properties and widths or can be a medium with the corresponding variation of, perhaps, refractive index. It can also be a layered medium with one or more interfaces with a periodic (quasiperiodic) profile. Irrespective of the details, these structures possess interesting spectral features. In what follows we treat the cases of periodic and quasiperiodic structures separately to bring out the manifestations of various nonlinear effects.

Nonlinear periodic structures

Linear optical properties of periodic media are now well understood[2]. As mentioned earlier, irrespective of the details, these structures possess forbidden frequency gaps or stop gaps as a direct consequence of Floquet–Bloch theory. These gaps are located around Bragg frequencies, which are determined by the modulation period and the average refractive index. Waves with frequencies in the stop gap of an infinite structure are reflected and the gaps are perfect. For finite structures sharp transmission resonances occur at the gap edges. The location of the gap and such resonances has critical dependence on the refractive index. It is in this context

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that Kerr nonlinearity (leading to intensity-dependent refractive index) plays a crucial role. A periodic structure made of alternating Kerr nonlinear slabs exhibits transmission, which has very strong power dependence. In fact, one can tailor both the transmission resonances and the gaps by changing input power levels. This suggests tremendous and diverse potentials for all-optical devices. Some potential applications of nonlinear periodic structures were earlier reviewed by Winful and Stegeman[7]. These include optical bistability, nonlinear coupling and tuning of surface and guided modes, tunable filters, pulse compression, etc. Other important applications such as power limiting[8], all-optical logic gating[9], VLSI compatible switching devices[10] and diode action have also been demonstrated.

The exact analytical treatment of nonlinear periodic structures even with the simplest possible nonlinearity (viz. Kerr nonlinearity) can be quite complicated. As of now exact solutions for a single nonlinear slab are known only for normal incidence, which were later generalized to the case of a lossless nonlinear periodic medium[11]. For oblique incidence, the case of *s*-polarized light was dealt with by Leung[12]. However, the solutions for *p*-polarization even for a single nonlinear layer are yet to be worked out. In view of the complexity of the exact solutions, and sometimes their unavailability, various numerical and approximate schemes were developed. A particularly simple and general method has been the nonlinear characteristic matrix method[13], which has been applied to various layered media, including periodic[14] and quasiperiodic media[15–17]. The method can incorporate the effects of losses though it is limited only to the case of weakly nonlinear systems. There have been other approximate and exact matrix methods, which rely heavily on computation[18]. In the context of nonlinear periodic media the coupled mode theory[19] as well as the envelope function approach[20] have been quite popular.

The major outcome of the studies on Kerr nonlinear periodic media has been the prediction of solitary wave profiles with the frequency in the stop gap of the structure. The static (immobile) profiles, termed as 'gap solitons' were first discovered in numerical experiments carried out by Chen and Mills[11]. The exact method was applied to calculate the transmission from a finite superlattice with alternating linear and nonlinear slabs. The superlattice was illuminated with normally incident plane waves with frequency in the stop gap but close to the edge. Multivalued output in transmission was noted. The intensity distribution corresponding to the total transmission state exhibited *sech* profile. Thus, it was interpreted that the total transmission through the nonlinear structure is mediated by these gap solitons. Existence of analogous solitons in other contexts were reported by several authors[21].

There have been several experiments on periodic structures with intensity-dependent refractive index. One of the preferred realizations of such structures is the corrugated waveguide. Sankey *et al.*[22] studied a silicon on insulator waveguide with the top surface of the Si layer corrugated. Two surface gratings were used to couple in and out the radiation from the waveguide. A similar structure (Figure 1) was studied to demonstrate the possibility of VLSI compatible switching[10]. The major experimental result is given in Figure 2, where the typical incident and reflected pulse shapes are shown. At the leading edge of the pulse the reflected signal mimics the incident signal. With an increase in the incident energy, the shift of the stop gap can be sufficient for the incident field to tune itself out of the stop gap, which leads to a sudden drop of the reflection and corresponding decrease in the peak amplitude of the reflected signal. As the incident intensity decreases, a sudden rise occurs in the reflected amplitude, after which there is a steady decrease following the incident pulse. Along with other applications[23] power limiting has been a major goal. Power limiting using periodic structures has been demonstrated by several groups[8,24]. The origin of power limiting in periodic structures is simple. The intensity-dependent refractive index has two major effects on the stop gap. For a defocusing nonlinearity the gap shifts to the lower wavelengths because of decrease in the average refractive index, and at the same time a broadening of the gap occurs due to an increase in the refractive index contrast (assuming the nonlinear layers having lower index). For the red edge there is a competition between these two effects. On the other hand, for the blue edge a decrease in transmission will occur leading to power limiting.

An important class of problems in nonlinear optics has been wave-mixing phenomena. The high *Q* modes of periodic structures can play a very important role in en-

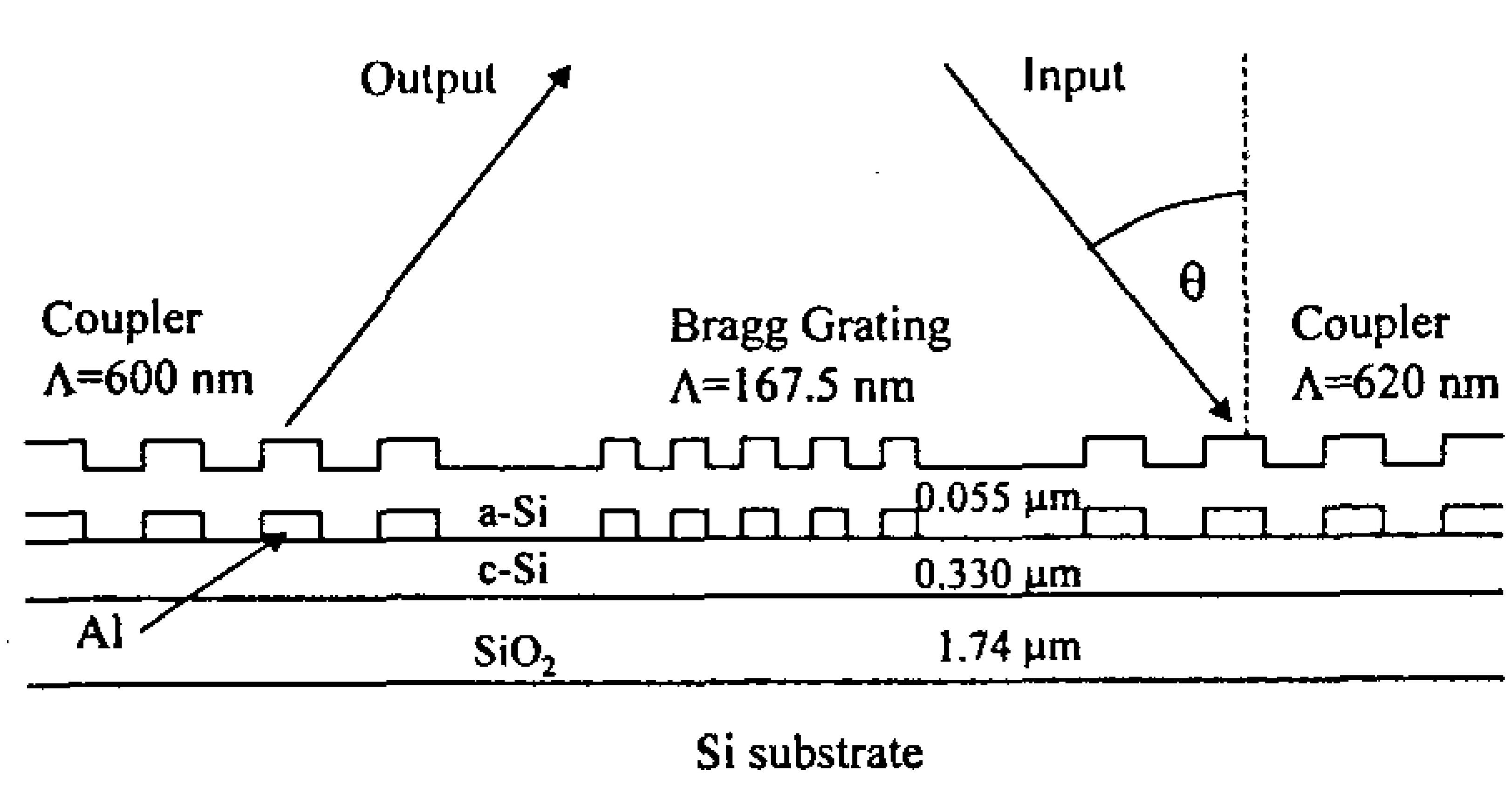

Figure 1. Schematic view of the experimental set-up of Bieber *et al.*[10]. The waveguide with 55 nm thick a-Si layer has end couplers for input and output radiation. The separation between the couplers and the Bragg grating is 0.5 nm. Second order backward coupling is used to excite the TM_0 guided mode.

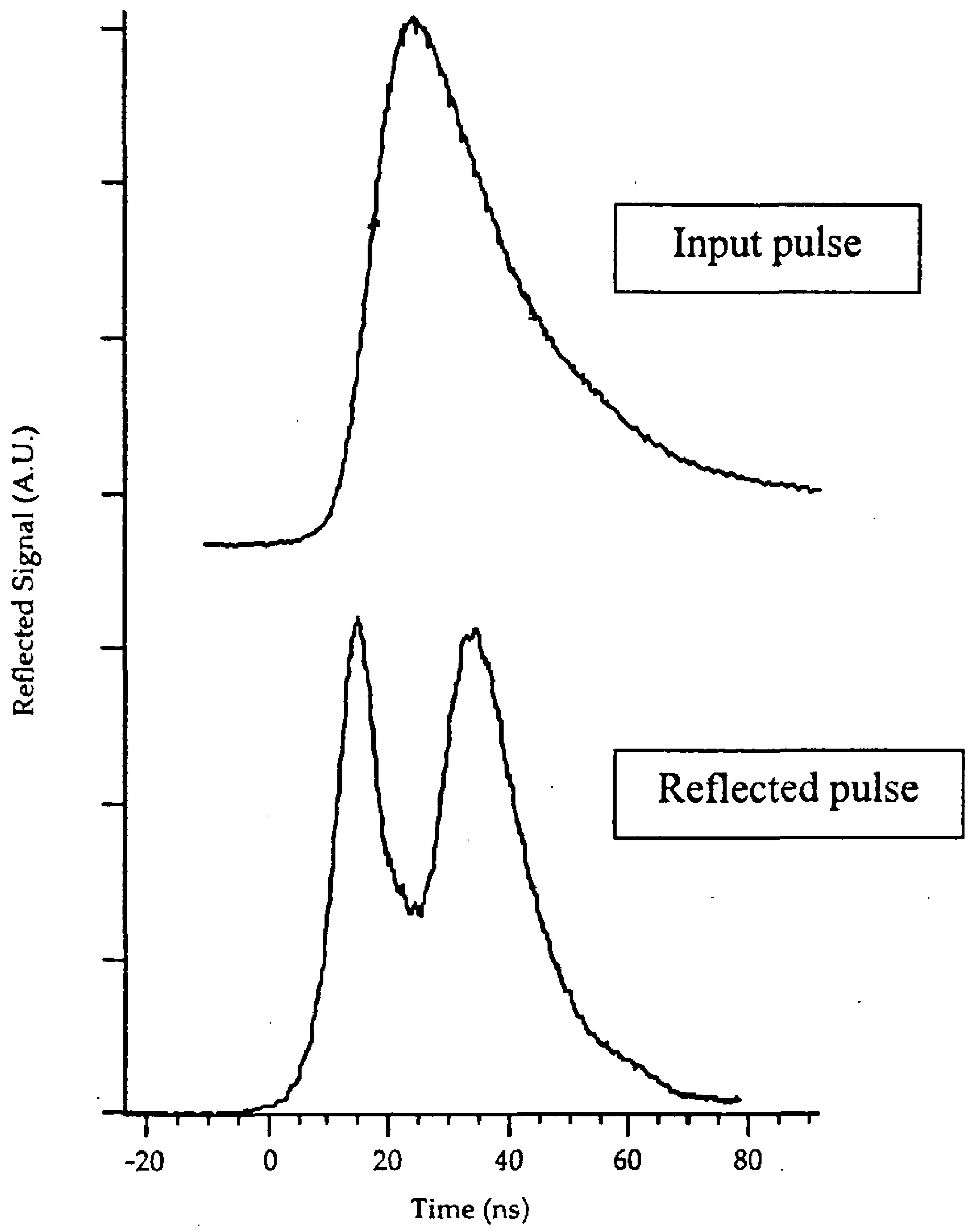

Figure 2. Incident and reflected pulse profiles for the structure of Figure 1 (ref. 10).

hancing the mixing efficiency. In the context of harmonic generation the advantages of such resonances can be easily understood. Fundamental distribution in the layered sample will be enhanced if its frequency coincides with one of the mode frequencies of the periodic structure. Since the fundamental distribution along the periodic structure acts as the source for the generated harmonic, there will be an increase in the output of the harmonic wave[25]. The general theory of wave-mixing in layered media, in particular, in periodic media, albeit under undepleted pump approximation, has been developed by Bethune[26] and by Hashizume *et al.*[27]. Bethune's approach is a straightforward extension of the earlier work of Bloembergen for a single slab, adding further the elegance of transfer matrices, while the other group uses Green's function technique to arrive at the harmonic output and, in our opinion, gives a better understanding of the underlying physics. The work of Hashizume *et al.* contains also the experimental results, which show excellent agreement with theoretical predictions.

Perhaps the most important application of periodic structures in the context of harmonic generation and three-wave-mixing has been in quasi phase matching (QPM) in waveguide geometries. The major advantage of wave-mixing in guided wave structures stems from the fact that in addition to conventional phase matching techniques (using birefringence) one has the option of QPM which can make use of the largest d-coefficient of the nonlinear crystals. The principle of QPM has been known for quite sometime since the classic work by Armstrong *et al.*[5]. However, the actual realization has been achieved only recently using the periodic domain reversal techniques in ferroelectric crystals leading to a layered media with opposite signs of nonlinear coefficient in alternate layers[4,6]. In principle, in order to achieve QPM, any relevant waveguide parameter (e.g. refractive index or the nonlinear coefficient) can be spatially modulated with modulation vector $K(= 2\pi/\Lambda$, Λ is the period). The second harmonic (SH) effective index $n^{2\omega}_{\rm eff}$ can then matched with a Bragg scattered fundamental effective index $n^{\omega}_{\rm eff}$ as follows

$$n^{\omega}_{\rm eff} \pm (mK)\frac{c}{2\omega} = n^{2\omega}_{\rm eff}, \qquad (1)$$

where m is an integer.

Note that in guided wave structures both the fundamental and the SH need to be the modes of the waveguide with distinctive transverse field distributions. A large overlap of the transverse profiles is crucial for efficient harmonic output. Note also that we used effective indices in eq. (1) to characterize the modes of the waveguide. Though there is an option to use refractive index modulation for achieving QPM, one never exploits the possibility in reality. Rather one patterns the nonlinear susceptibility by means of suitable masks. The method of patterning in ferroelectric materials depends on the specifics of the crystal, especially their Curie temperature[4]. The most feasible method has been the patterned dopant technique involving patterned in- or out-diffusion of certain dopants leading to a corresponding distribution of reversed domains. For example, in lithium niobate, periodic domain reversal can be achieved by in-diffusion of a Ti film patterned into a grating of a desirable period by standard photolithographic technique at temperatures close to the Curie temperature.

Blue light generation by up-converting diode laser output is in demand for purposes of optical storage and xerography because of the compact nature of such devices. QPM has played an important role in such devices. QPM has been used in $LiTaO_3$, $LiNbO_3$ and KTP to obtain such blue sources. A domain inverted $LiTaO_3$ channel wave-guide was shown to lead to a 2.4 mW of SH at 424 nm wavelength[28]. Gain switching and locking of the oscillation wavelength by grating feedback were shown to lead to a better efficiency. A 4.5 mW of average blue light power with 13% conversion efficiency was achieved later. Other efficient designs using a continuous wave Nd-YAG laser and exploiting an array of channels and a fan-patterned domain inverted grating were tested resulting in an efficiency of 17% (ref. 29). Another interesting geometry leading to surface emitting SH corresponds to the case of counter-propagating fundamental waves that belong to the same waveguide mode[30]. Since the resulting nonlinear polarization does not have any surface component, the generated SH is radiated normal to the surface. GaAs based waveguides, where some form of QPM in multilayered geometry could be used, show a lot of promise. Novel proposals have been put forth for phase matching in periodically corrugated waveguides for surface emitted SH. One such proposal makes use of higher order spatial harmonics for both the fundamental and the SH in the guiding layer. Recently the proposal has been tested using a corrugated polymer waveguide on a glass substrate[31]. There have been other interesting suggestions for combining counter-propagating geometry with the QPM. Results based on coupled mode theory for such structures were compared with the case of co-propagating fundamental waves. In most situations the counter-propagating geometry turns out to be more advantageous.

Cascaded second order nonlinearity has been at the focus of some of the recent studies. A major goal of these studies was to achieve nonlinear phase shift, much like in third order materials. The principle behind the intensity-dependent phase shift in cascaded second order nonlinear processes is as follows: a fundamental is up-converted to, say, the SH, which is then down-converted back to the fundamental. The resulting fundamental wave can have a phase, which is distinct from that of the original wave. Since second order nonlinear coefficients are much larger than the third order counterparts, the effective 'third order' effect due to cascading can be orders of magnitude larger than conventional Kerr phase shifts. The first experimental demonstration of cascading in guided wave geometry was given by Sundheimer *et al.*[32], who used a QPM KTP waveguide to measure the self-phase modulation, where the phase information was inferred from spectral data. A direct interferometric measurement was carried out only recently by the same group. A proposal for a novel type of frequency shifter was presented by Gorbunova *et al.*[33]. The proposed structure involves SH generation in a waveguide by counter-propagating fundamentals at frequency ω, followed by difference frequency generation in a second waveguide on top with an input wave at ω_s. Recall that the counter-propagating geometry leads to SH emitted normal to the surface of the waveguide. This SH can then interact with the probe at ω_s leading to an idler at frequency $\omega_i = 2\omega - \omega_s$, which propagates in the opposite direction of the signal. A multilayered QPM structure between the waveguides with additional mirrors was shown to lead to conversion efficiency much larger than that of typical waveguide shifters made of GaAs/AlGaAs multilayers.

A very interesting consequence of self-phase modulation due to cascaded processes in periodic structures has been the emergence of solitary wave profiles in such systems. Clausen *et al.*[34] considered the propagation of a cw beam at both fundamental and its SH frequencies in a QPM second order nonlinear slab waveguide, where only the nonlinear susceptibility was modulated. Existence of rapidly oscillating solitary waves was demonstrated. Analogous problems have also been considered by Torner and Stegeman[35] who investigated numerically the formation and evolution of spatial solitons with allowance for the fluctuations of the domain lengths to bring out the effects of the randomness.

A major area of activity in nonlinear optics has been the search for new nonlinear materials with better characteristics. Synthesis of new materials with higher nonlinear coefficients, faster response times and higher damage thresholds is one such direction. An alternate scheme to enhance the nonlinear susceptibility with existing materials was proposed by Sipe and Boyd[36], exploiting the local field effects in composite materials. A composite consists of two or more constituents as 'grains'. Grain size or the linear dimension of each constituent must be large enough so that they may be represented by their bulk properties. At the same time, they should be small enough (much less than the wavelength) so that an effective medium description holds. In order to understand the scope of such materials let us consider a composite consisting of spherical inclusions with dielectric constant ε_i embedded in a host with dielectric constant ε_h. A single spherical inclusion in the composite experiences the local field $\bar{\mathbf{E}}_{\rm loc}$ (and not the applied field $\bar{\mathbf{E}}$) which is given by:

$$\bar{\mathbf{E}}_{\rm loc} = \bar{\mathbf{E}} + \frac{4\pi}{3\varepsilon_h}\bar{\mathbf{P}}. \qquad (2)$$

The field inside the spherical inclusion $\bar{\mathbf{E}}_{\rm loc,i}$ is given by:

$$\bar{\mathbf{E}}_{\rm loc,i} = \frac{3\varepsilon_h}{\varepsilon_i + 2\varepsilon_h}\bar{\mathbf{E}}_{\rm loc}. \qquad (3)$$

If, for example, the inclusion is made of a metal with $\mathrm{Re}(\varepsilon_i) < 0$, one may choose the host and inclusion such that $\mathrm{Re}(\varepsilon_i + 2\varepsilon_h) = 0$. This corresponds to the plasmon resonance and there is the possibility of resonant enhancement of the field, which can be exploited for low threshold nonlinear phenomena. For several such inclusions Maxwell–Garnett calculations along with other approaches exist to arrive at the effective index. Similar approaches can be extended to nonlinear composites in order to derive the higher order effective nonlinear susceptibilities[37]. While considering a general problem of nonlinear inclusion in a nonlinear host, Sipe and Boyd[36] demonstrated that a careful choice of the linear properties of the constituents and their volume fraction can lead to an effective medium with nonlinear susceptibilities larger than those of the constituents. The enhanced effective nonlinear susceptibility of the composite results as a consequence of the local field corrections and does not necessarily need some of the constituents to be metals. Despite the elegance of the theoretical predictions involving spherical inclusions, the realization of such composites turned out to be a formidable job because of stringent and unachievable conditions. Again periodic layered media with layer widths much less than the wavelength came to the rescue. The theory of layered composites was presented by Boyd and Sipe[38], who calculated the effective nonlinear susceptibilities for various nonlinear processes. For example, for SH generation and for electric field polarized perpendicular to the layers, the effective second order nonlinearity is given by:

$$\chi^{(2)}_{\mathrm{eff}}(2\omega=\omega+\omega)=f_1\chi_1^{(2)}\frac{\varepsilon_\perp^2(\omega)\varepsilon_\perp(2\omega)}{\varepsilon_1^2(\omega)\varepsilon_1(2\omega)}+ f_2\chi_2^{(2)}\frac{\varepsilon_\perp^2(\omega)\varepsilon_\perp(2\omega)}{\varepsilon_2^2(\omega)\varepsilon_2(2\omega)}, \tag{4}$$

with

$$\frac{1}{\varepsilon_\perp}=\frac{f_1}{\varepsilon_1}+\frac{f_2}{\varepsilon_2}, \tag{5}$$

where f_i, $\chi_i^{(2)}$ and ε_i are the volume fraction, second order susceptibility and the linear dielectric constant of the ith constituent, respectively. The results for nonlinearity only in one component ($\chi_1^{(2)}\neq 0$, $\chi_2^{(2)}=0$) are shown in Figure 3. It is clear from the figure that an enhancement of three can be achieved for an optimal choice of parameters. Though we cited the results for SH generation, the experiments on enhancement of the effective nonlinear susceptibility exploiting local field corrections were carried out for the third order process leading to nonlinear phase shifts[39]. A z-scan technique was used to measure the nonlinear phase shift in the composite made of alternating linear (titanium dioxide) and nonlinear (PBZT) layers with layer thickness of about 500 Å. It was shown that the predictions of the effective medium theory match the experimental results well.

Nonlinear quasiperiodic structures

Quasiperiodicity implies the presence of two or more incommensurate periods. Thus quasiperiodic structures are intermediate between periodic and random media.

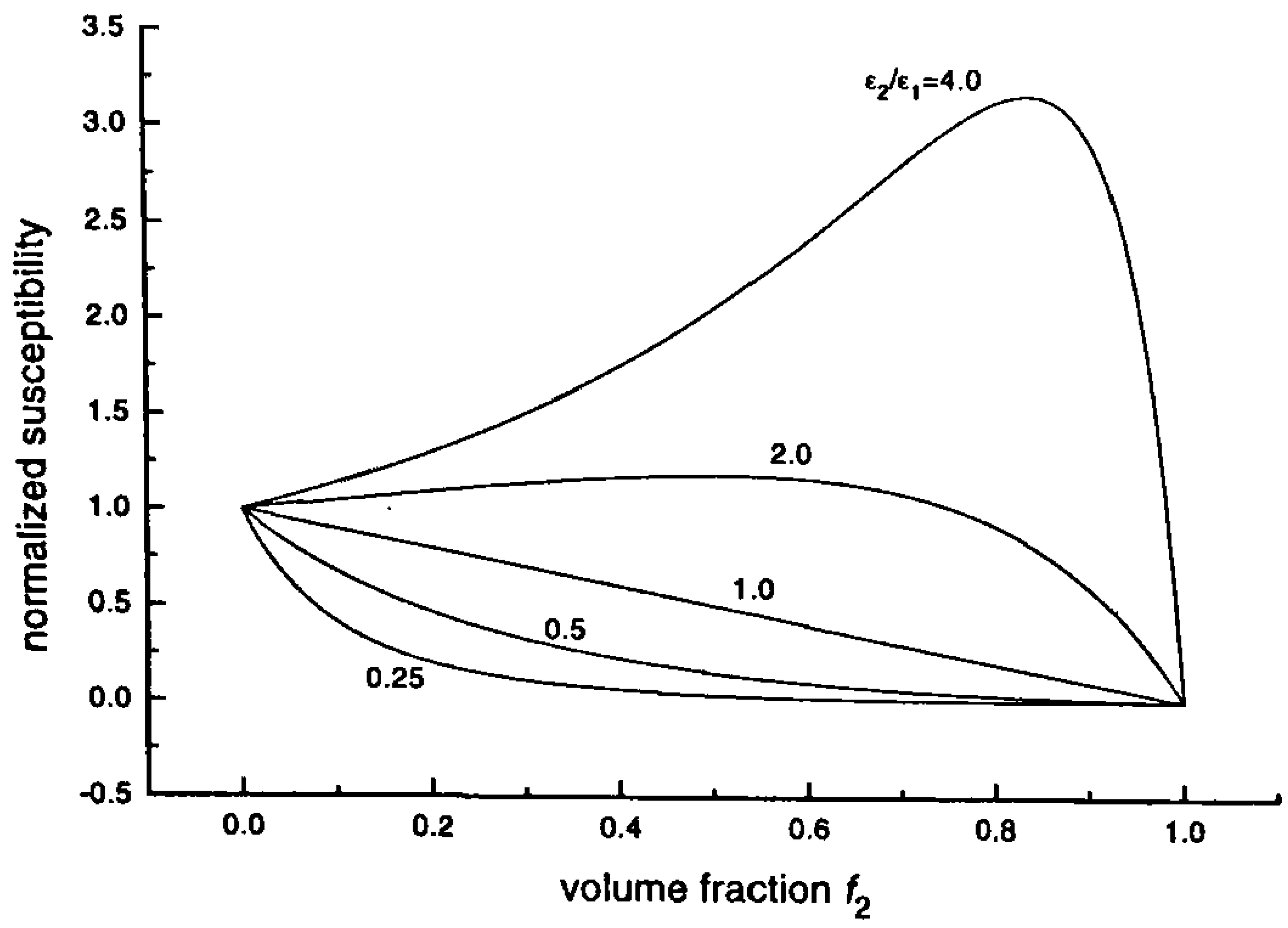

Figure 3. Effective second order nonlinear susceptibility $\chi_{\mathrm{eff}}^{(2)}$ normalized to $\chi_1^{(2)}$ as a function of the volume fraction of the second component f_2 for a layered composite[35]. Different curves are labelled by the corresponding values of the ratio of the linear dielectric constants $\varepsilon_2/\varepsilon_1$.

The interest in quasiperiodic structures stems from the realization that such structures can be ideal candidates for weak localization. In fact, localization in solid state systems has been a very 'hot' field ever since the discovery of Anderson localization. Weak localization in quasiperiodic solid state systems has been studied by several groups[3]. The theoretical studies received an impetus with the experimental realization of quasiperiodic superlattice by Merlin *et al.*[40]. The first ever proposal for weak localization in optical systems was presented by Kohmoto *et al.*[41]. The optical systems in general have several advantages over their solid state counterparts. Optical experiments are purer since the photons are noninteracting. On the other hand, in solid state systems electron–electron and electron–phonon interactions are unavoidable and can pose a real threat for direct observation of theoretical predictions. Besides, the polarization of light adds a new dimension to the problem of localization. Kohmoto *et al.*[41] considered the transmission of a plane wave through a Fibonacci optical multilayer. A Fibonacci multilayer can be constructed recursively, for example, by dielectric slabs A and B (with distinct refractive indices) as

$$S_{j+1}=S_{j-1}S_j,\ \text{with } S_0=(B) \text{ and } S_1=(A). \tag{6}$$

Thus second generation S_2 is given by (BA), S_3 by (ABA), S_4 by ($BAABA$), etc. Assuming the same optical width for both slabs, it was shown that the system can be described by a (2 × 2) map, which can be further reduced to a trace map. The most interesting property of such structures is the self-similarity of the transmission coefficient as a function of optical width for various generations. Another interesting feature is the fact that allowed states (characterized by total transmission) form a Cantor set with Lesbegue measure zero[41]. Sendler and Steel[42] presented a detailed treatment of the system and a comparison with a periodic structure. The Landauer resistivity defined by $R_L = R/(1-R)$, where R is the reflectivity of the total structure, was studied. It is well-known from solid state results that R_L as a function of system length becomes exponential or power law bounded for localized and critical states, respectively. An extended state is characterized by a constant or a bounded function of system length. A major finding was the following: for a very large number of layers, all the observable states are exponentially localized surface states. For a smaller number of layers some states appear to be critical, but cross over to exponentially localized states as the system size (number of layers) increases. The above conclusions were reached at by looking directly at the field distribution along the length of the system and noting the dependence of R_L on the number of layers j or $\log(j)$. Experimental demonstration of the scaling behaviour and the self-similarity was achieved much later using a SiO_2 and TiO_2 multilayer[43].

Perhaps the first ever study of nonlinear quasiperiodic structure was carried out by Dutta Gupta and Ray, who considered a Fibonacci multilayer with one or both the constituent layers with Kerr type non-linearity[15–17]. The nonlinear characteristic matrix approach was applied to calculate the transmission through such structures. It was shown that nonlinearity coupled with the richness of Fibonacci spectra can lead to a wide variety of bistability and multistability[15]. A very important observation was the existence of bulk localized states (described by a *sech* distribution)[16]. In fact, the total transmission of the structure was shown to be mediated by these bulk localized states. The situation is quite analogous to the gap solitons of nonlinear periodic structures. The symmetric (with respect to the center of the structure) distribution in the quasiperiodic system (lacking any such symmetry) results from a delicate interplay between dispersion and nonlinearity. A detailed numerical study of the effects of nonlinearity on localized, extended and critical states was carried out by the same authors[17]. It was shown that strong surface localized states in linear theory spread over the whole structure as the power is increased. Thus nonlinearity leads to delocalization. However, the extended states corresponding to $R \sim 0$ in the linear regime do not change their character under the influence of nonlinearity. There was also evidence of critical-like states. As mentioned earlier, the most important result was the emergence of bulk localized states without any linear counterpart, which persisted even for large system sizes. In fact, these states exhibited a direct scaling with the system size.

A significant achievement in the field of nonlinear optics of quasiperiodic media was the report of SH generation in QPM quasiperiodic structures[44]. QPM in a quasiperiodic structure has its own specifics due to the presence of incommensurate periods. The relevant phase matching condition, for example, for two incommensurate periods can be written as follows:

$$\Delta k = k_{2\omega} - k_{\omega} - K_{m,n} = 0, \qquad (7)$$

where m, n are integers, $K_{m,n}$ is the reciprocal vector. The reciprocal vector in eq. (7) needs two integers for labelling because of the two incommensurate periods of the structure. Note that for periodic structures the reciprocal vector $K_m(= mK)$ is indexed with only one integer. In view of the much broader options for $K_{m,n}$, QPM in quasiperiodic structures can cover a large number of harmonic frequencies. The nonlinear quasiperiodic structure in the experiment of Zhu *et al.*[44] was fabricated using the pulse field poling technique in a $LiTaO_3$ wafer at room temperature. Each of the building blocks A and B of the structure had one positive and one negative ferroelectric domain and they were arranged in a Fibonacci sequence with the starting generations given by A and AB. The tunable output of a parametric oscillator was used to record an efficiency of 5 to 20% as the input wavelength was tuned to 0.9726, 1.0846, 1.2834, 1.3650 and 1.5699 μm. Self-similarity feature was shown to be destroyed because of dispersion in constituent layers.

There have been studies of quasiperiodic structures other than that given by a Fibonacci sequence. A self-similar Fabry–Perot resonator consisting of dielectric slabs of two different refractive indices, with the high index slabs forming a Cantor set was studied. Along with the waveguide realization of the above structure these authors studied the nonlinear Cantor corrugated waveguide to show bistability with the narrow transmission resonances[45].

In conclusion, we presented some of the new directions in research involving nonlinear periodic and quasiperiodic structures. Avoiding mathematical derivations (interested readers may find them in ref. 1) we tried to reveal the immense scope of such structures for device applications. It is perhaps because of the theoretical achievements and technological progress in such fields that Bloembergen[46], father of nonlinear optics, commented that nonlinear optics has entered a technology phase. The problem now is not in having high power lasers, but in fabricating nonlinear materials with sufficiently high damage thresholds. Hence, some of the very interesting predictions of theory are yet to be tested. The search for such materials is on and success in this regard will lead to ever more exciting results in this field.

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ACKNOWLEDGEMENTS. I thank the Department of Science and Technology and National Laser Programme, Government of India for supporting this work.