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Comparison of nonlinear optical responses of periodic and quasiperiodic superlattices

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For one-dimensional superlattices consisting of unit cells with two films, one of which exhibits nonlinear response to the electromagnetic field, the transmissivity as a function of incident intensity and frequency is calculated for periodic and quasiperiodic structures. It is found that gap-soliton-mediated resonances for periodic structures arise from the Fabry-Pérot oscillations in the transmission band. Solitons in the stop gap are not found in the quasiperiodic case.

I. INTRODUCTION

The study of transport in and transmission through one-dimensional superlattices with quasiperiodic structure has aroused significant interest in the last several years,[1-3] predating the discovery of quasicrystals. Merlin *et al.* synthesized the first one-dimensional semiconductor quasiperiodic structure.[3] The theoretical work of Kohmoto *et al.*[4] and Ostlund *et al.*[5] laid the ground work for experimental studies of quasiperiodic potentials involving phonon dispersion,[2] classical wave transmission,[6] and conductance fluctuations.[7] Among a variety of topics, these studies center on the transitions from extended to critical to localized states as the structure varies from periodic to quasiperiodic to random. To represent the quasiperiodic system, it is customary to use a Fibonacci-sequence superlattice with a "Bravais" lattice constant having two values with a ratio equal to the golden mean, each component distinguished by a different potential. On the theoretical side, a plethora of techniques are brought to bear on this problem with the central goal to determine a band structure for the system. One arrives at a Cantor set of allowed energy levels and eigenfunctions having self-similar representations. All of these studies explore wave-propagation characteristics, within a linear theory.

Another topic of interest in the study of transmission in superlattices has been to include the effect of nonlinear material layers whose response characteristics depend on the amplitude of the incoming beam.[8-11] In this case the structure is periodic and the transmission now varies with the intensity of the beam. In a specific case, alternating layers of dielectric (linear) and antiferromagnetic (nonlinear) materials have been modeled to study transmission phenomena.[8,10,11] Among the interesting results, one finds multistable solutions, the breakdown of reciprocity (i.e., transmission from left to right is different from right to left), gap-soliton-mediated resonances, and regions of chaotic transmission, when viewed as a function of the beam intensity.

The purpose of this paper is to describe the combined effect of nonlinearity and quasiperiodicity on the transmission properties of superlattices. Our understanding is that quasiperiodic superlattices can be fabricated with little difficulty. A more serious concern is in generating systems which are sufficiently lossless for some of the effects described here to be observed. The paper is structured as follows. In Sec. II a brief description of the computational technique used for nonlinear systems will be given. Section III will describe the quasiperiodic superlattice and make contact with previous work. Section IV will compare the features of the quasiperiodic system with the periodic system. And in Sec. V a discussion of the distinctions between the two layers will be given.

II. COMPUTATIONAL TECHNIQUE

In the present paper we will limit ourselves to the discussion of a plane-polarized electromagnet wave propagating normal to the interfaces of the superlattice; the extension to other transport phenomena follows similar construction, with the main conclusions being unaltered. The nonlinear material is modeled by an antiferromagnet, assumed to be an easy-magnetization axis, two-sublattice material with an easy axis normal to the interfaces. A multilayer dielectric structure is easily treated within the same framework. If no external field is present, then the plane-polarized wave propagates through the material with no rotation of the plane of polarization.[10] As was shown in Refs. 9 and 11 when the energy per unit time flowing down the structure is independent of z, the direction parallel to the normal to the interface, then the discretization of the resulting nonlinear Schrödinger equation prescribes an iterated map given by

$$x'=y+F^2x+2\left[xy-\frac{\epsilon_i^2}{\epsilon^2}\sin^2(K)\right]^{1/2}F\ ,$$
$$y=x,\quad x=x'\ ,\qquad (2.1)$$

with boundary conditions at interfaces given by

$$\frac{1}{\epsilon_-}(x'-x)=\frac{1}{\epsilon_+}(x-y)\ . \qquad (2.2)$$

The following definitions have been employed:

$$x=[h(z_n)/h_i|T|]^2\ , \qquad (2.3)$$

where h_i is the incident magnetic field and $h(z_n)$ the field at position z_n. Here x' and y correspond to the equivalent formula for $h(z_{n-1})$ and $h(z_{n+1})$, respectively. The quantity F is given by

$$F=E-\alpha_0-\alpha_1|h_i|^2|T|^2x\ , \qquad (2.4)$$

where

$$E=-2+K^2 \qquad (2.5)$$

and $K=k_0\xi$ with ξ the width of the interval in the basic spatial grid and k_0 the wave vector in the vacuum. Finally,

$$\alpha_0=K^2[1-(1+4\pi\chi)(\epsilon_\perp/\epsilon)] \qquad (2.6)$$

and

$$\alpha_1=K^2 4\pi\chi\epsilon_\perp\lambda/\epsilon\ , \qquad (2.7)$$

where $\epsilon_\perp$ is the dielectric function of a layer in the plane of the interfaces which is perpendicular to the easy axis of the antiferromagnet, ϵ is the dielectric function of the vacuum, χ is the frequency-dependent susceptibility of the antiferromagnet, and λ describes the lowest order of nonlinearity. Both of these parameters are functions of frequency and are determined by the antiferromagnetic material which is used.[11]

Each bilayer in our model superlattice consists of one dielectric film ZnF_2, a nonmagnetic dielectric, and one antiferromagnetic film of FeF_2. The parameters representative of FeF_2 are $\epsilon_\perp=4$, the anisotropy field is 200 kG, and the exchange field is 540 kG. We model ZnF_2 as the dielectric material for which $\epsilon_\perp=8$.[12] As a result of the nearly equal lattice constants of these materials, bilayers show little deformation at the interfaces.[13]

The transmission (as well as the magnetic field as a function of position) is found by starting at the output end with initial conditions $x=y=1$ and iterating to the incident side (in the linear region more efficient matrix techniques can be used to replace the iterating scheme). The resultant value of the incident intensity is then determined by continuing the procedure into the vacuum and extracting the reflected intensity. The ratio of the transmitted intensity (which is equal to 1 from the initial conditions) divided by the incident intensity is the transmission coefficient $|T|^2$. Note, however, that the nonlinearity arising from F is essentially a product of the $|h_i|^2$ and $|T|^2$; therefore, to obtain $|T|^2$ versus the $|h_i|$ one first obtains $|T|^2$ versus $h_0=h_i|T|$, which now serves as an input parameter. This relationship has a single valued nature. Knowing T and h_0 allows the calculation of $h_i=h_0/|T|$ with the rescaling to the variable h_i left to the reader (see Ref. 11).

For the sake of comparison with our results for the quasiperiodic case, a 3D plot of transmission versus h_0 and $d\omega$ (measured from the antiferromagnetic resonance frequency, in G), the periodic case is presented in Fig. 1. Along the $h_0=0$ axis, the usual stop gaps and transmission bands are seen. As h_0 increases, the location of the bands moves to higher frequencies. Therefore, for constant frequency, a scan of h_0 will traverse transmission bands and stop gaps. This feature accounts for the peaks and valleys seen in those constant $d\omega$ curves. The variations become increasingly dense as h_0 gets large. Note also the relative flatness of the structure (for constant ω) for small h_0; rescaling the h_0 axis by $|T|$ in this region will not lead to multistability. A threshold exists before the nonlinear effects become significant. Constant $d\omega=150$ G curves are given in Ref. 11 and represent cross sections of Fig. 1 for the appropriate number of layers.

III. QUASIPERIODIC LATTICE

The quasiperiodic lattice with which we will deal is one constructed from a "Fibonacci sequence" of layers. The $(l+1)$th-generation Fibonacci structure is formed by appending the $(l-1)$th-generation Fibonacci structure to the lth. This concatenating operation can be summarized through the recursive rule $S(l+1)=S(l)|S(l-1)$ for $l\geq 2$, with initial conditions $S(1)=A$ and $S(2)=AB$, the next few Fibonacci structures are $S(3)=ABA$, $S(4)=ABAAB$, and $S(5)=ABAABABA$. In the present case, $A=$ a dielectric layer with a thickness of 169.9 μm and $B=$ an antiferromagnetic layer with a thickness of 105 μm [the ratio of the thickness being approximately the golden mean, $\frac{1}{2}(1+\sqrt{5})$]. The transmission side is the origin of the sequence. In Fig. 2 a comparison is made among several superlattices in the linear regime ($h_0\ll 1$): 2(a) a periodic alternating superlattice ($ABABAB\cdots$) with equal layer thicknesses; 2(b) a periodic alternating superlattice with alternating layers having thicknesses of approximately the golden mean; and 2(c) a Fibonacci superlattice. The effect of unequal thicknesses tends to narrow the transmission bands and shift them to slightly lower frequencies. For the Fibonacci superlattice, the lower end of each of the transmission bands is split off from the rest of the band, creating some spike resonances in what was previously a stop-gap region. The oscillations in the transmission regime arise from the finite size of the superlattice, in this case 21 layers. As evident from Fig. 2(c), incremental changes in frequency can cause strong fluctuations in transmission, as previously noted in Ref. 7. In the linear case, substantial effort has been expended to understand the transmission structure of the quasiperiodic superlattice. The reader is referred to Refs. 1, 7, and 14 for a review of some of the common techniques for calculating wave transmission in linear materials.

As the intensity is increased, the nonlinear effects become more prominent. The distinction between transmission bands and stop gaps becomes blurred at lower frequencies. Resonances appear in the gaps and the transmission bands narrow. As the intensity increases still further, very little is left of the band structure, and a series of spikes as a function of frequency remains. This information is displayed in Fig. 3. In Fig. 4, a 3D plot of periodic structure is compared to that of a Fibonacci superlattice.

IV. GAP-SOLITON-MEDIATED RESONANCES

One of the remarkable features of transmission through the superlattice is the existence of soliton-mediated transmission in regions near band edges. This nonlinear effect has been described by Mills and Trullinger[15] and seen in model calculations in Refs. 8 and 11. In this section, we describe how solitons arise from the transmission band and examine their existence in a quasiperiodic superlattice.

When one examines transmission versus frequency of the incoming radiation, in the linear regime (very low intensity) for a finite superlattice, oscillations are seen in the transmission band. We find that with increasing intensity, the positions of the oscillation peaks veer toward higher frequencies. When these frequencies are at, or just beyond, the transmission-gap boundary (the boundary being determined by linear conditions) the oscillations evolve into the soliton resonances previously described. This relation between Fabry-Pérot oscillations and gap solitons is a new and unexpected result. The soliton resonances are characterized by perfect transmission, a field versus position profile with an approximately sech^2 envelope, and transmission peaks at positions corresponding to an integral number of these envelopes. Soliton-mediated transmission in the gap refers to this transmis-

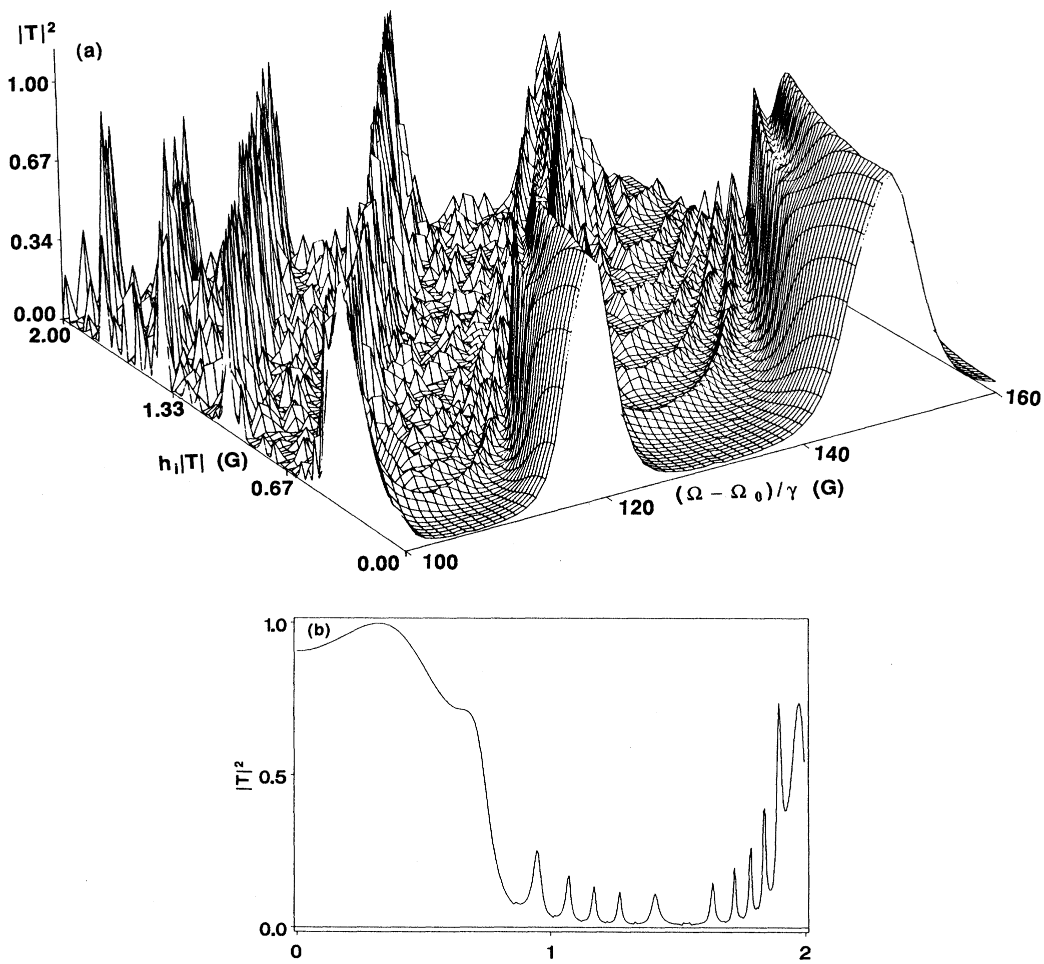

FIG. 1. (a) A plot of $|T|^2$ as a function of $h_i|T|$ and $d\omega$ for two alternating bilayers of antiferromagnetic (FeF_2) and dielectric (ZnF_2) materials with $d_1 = d_2 = \lambda_0/2 = 105$ μm. (b) A cross section of (a) for $d\omega = 150$ G [see Fig. 3(a) in Ref. 11]. In these figures, the antiferromagnetic material is exposed to the incident wave.

sion resonance exhibited at parameter values appropriate to soliton resonances even in regions which correspond to the gap in equivalent linear systems. In Fig. 4(a) the features of the transmission versus frequency and intensity are displayed for a ten-bilayer superlattice. Note the onset of the gap region at about $d\omega = 128$ G as indicated by the near-zero transmission at low intensities.

What we have observed in our model calculations is the similarity of the field versus position plots for all parameter values along one of the transmission peaks. That is, ridges in Fig. 4(a) represent nearly identical magnetic fields throughout the superlattice. In Fig. 5 a plot of h^2 versus position is made for frequencies and intensities corresponding to points along a ridge. The envelopes have nearly identical shapes, with only a mild increase in amplitude as a function of frequency. The number of envelopes as well as the number of fast oscillations within each envelope are unchanged. This is in contradiction with previous studies since the "length scale of the soliton" does not diverge at the gap edge.[15] It appears that the solitons evolve from the interference patterns seen in the transmission bands. Analytic calculations describing the envelope function for the wave pattern depend on

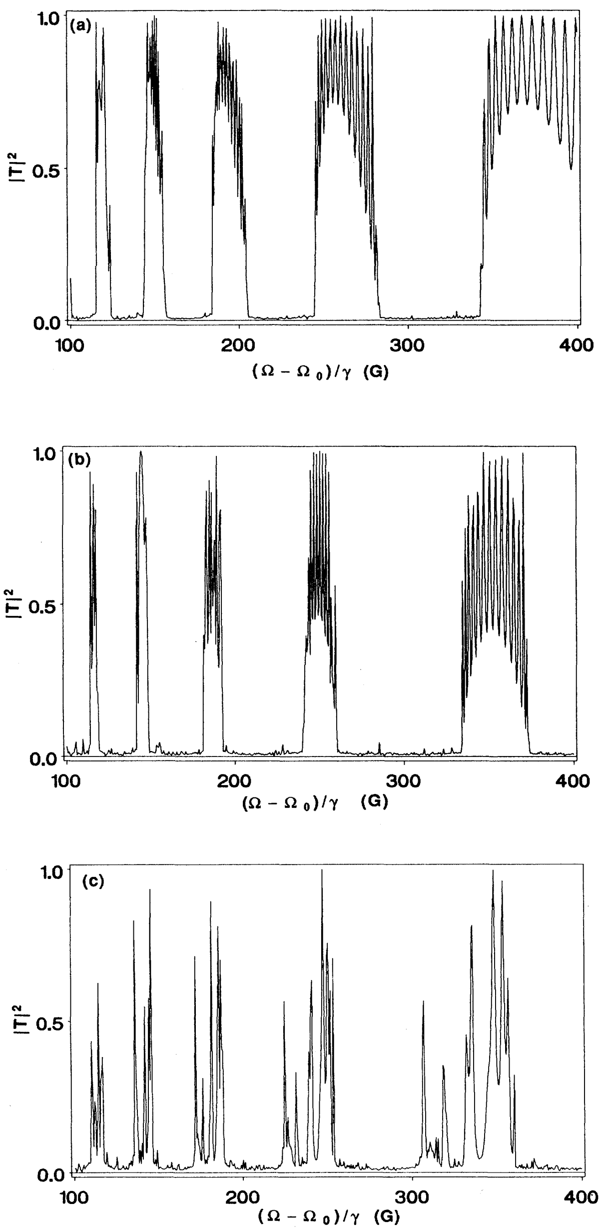

FIG. 2. (a) A plot of $|T|^2$ is given as a function of $d\omega$ in the linear regime, $h_i|T| = 0$ for 34 layers of superlattice structure as described in Fig. 1. (b) Same as (a) except that $d(ZnF_2) = 1.618d(FeF_2)$ $[d(FeF_2) = \lambda_0/2]$. (c) Same as (b) except that the layers are ordered according to the Fibonacci construction.

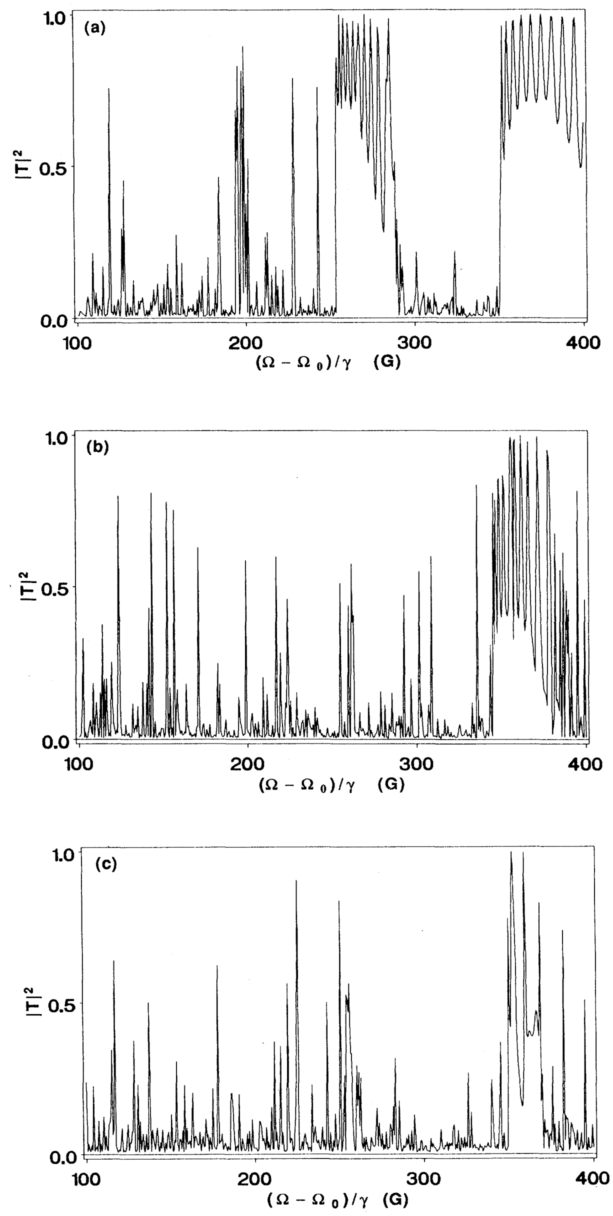

FIG. 3. Same as Fig. 2 with $h_i|T| = 1.0$.

small oscillations of the potential or low-incident intensities.[15,16] Neither of these approximations are appropriate in the present case, and therefore, it is not surprising that the conclusions vary.

In the case of a Fibonacci sequenced superlattice, no evidence of solitons was found. The envelopes were irregular and the fast oscillations reflected the Fibonacci sequence in that in each dielectric layer a node in the magnetic field occurred. Since these spacings are not periodic, neither are the fast oscillations. It is also important to note that the resonances which arise in the gap region are much narrower when viewed as a function of intensity at constant frequency. These resonances appear to be of different character from the soliton resonances.

V. DISCUSSION

The main focus of this paper has been to compare the nonlinear effects of optical transmission in periodic and

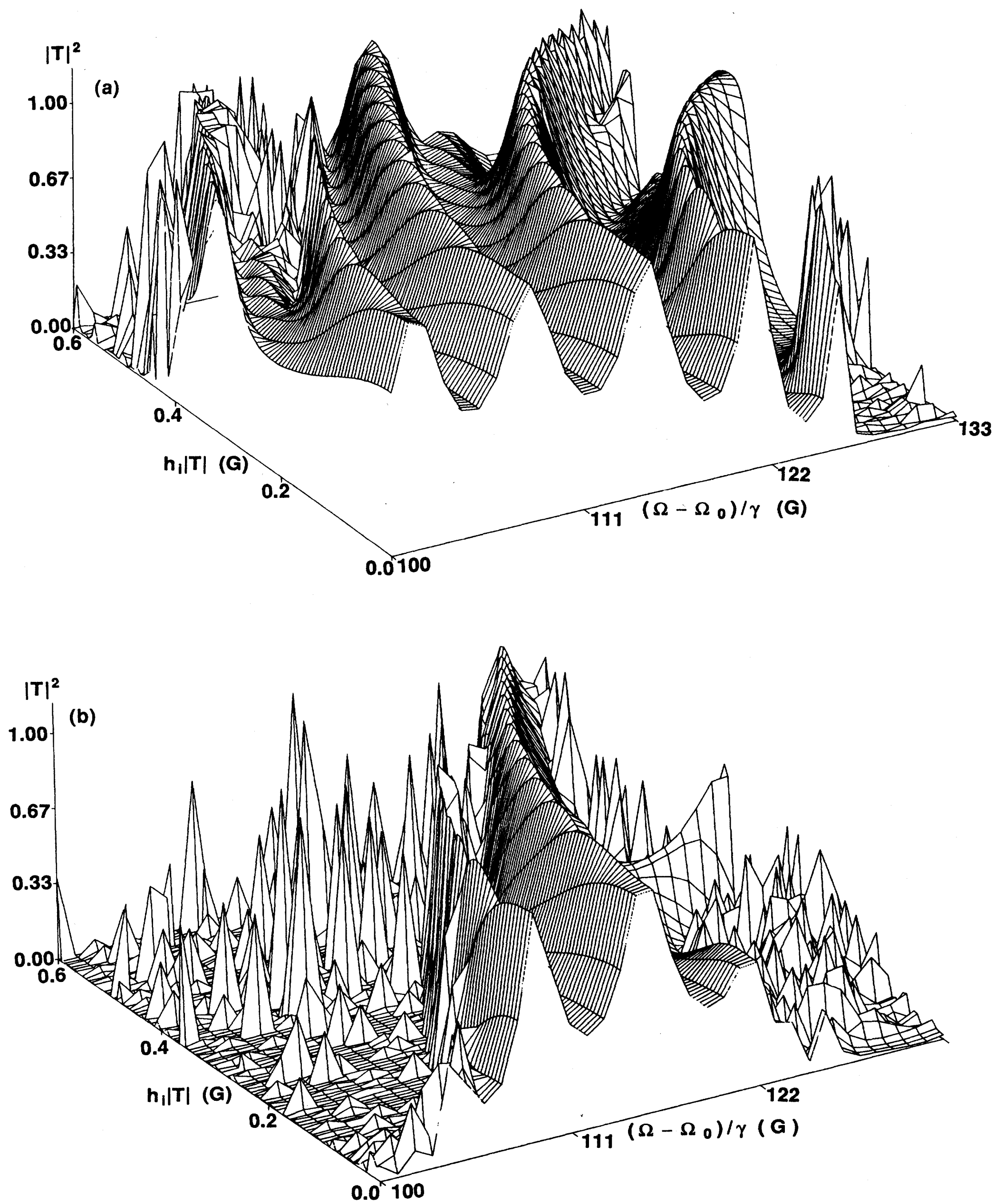

FIG. 4. A plot of $|T|^2$ as a function of $h_i|T|$ and $d\omega$ for a superlattice with layer thickness. (a) $d(ZnF_2)=d(FeF_2)=\lambda_0/10=21$ μm arranged in a ten-bilayer alternating periodic structure, and (b) $d(ZnF_2)=1.618d(FeF_2)$ [$d(FeF_2)=21$ μm] for 21 layers ordered according to the Fibonacci construction.

quasiperiodic superlattices. In general, the lattices share a number of features in the linear regimes which have been well documented. Both structures exhibit optical reciprocity, a band structure with transmission bands and stop gaps, finite-size effects (oscillations in transmission within the band resulting from the finite size of the lattice), nodes in the magnetic field intensity appearing in the dielectric layer (allowing ready prediction of the number of fast oscillations within each structure), and unique output for a given input.

In the nonlinear regime, both types of structures violate optical reciprocity and have multistable solutions

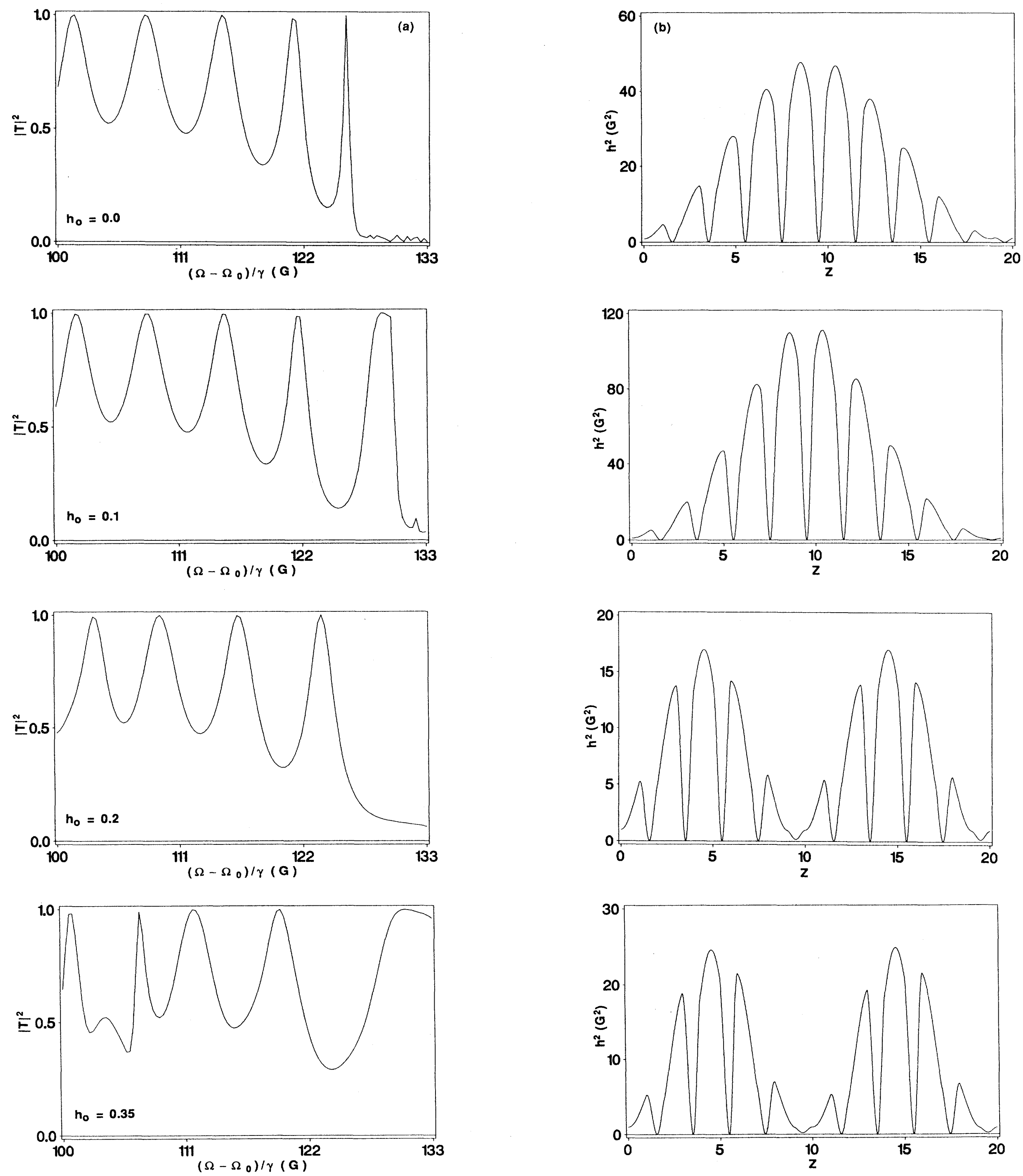

FIG. 5. (a) Cross sections of Fig. 4(a) for $h_0=0$, $h_0=0.1$, $h_0=0.2$, and $h_0=0.35$. (b) Magnetic field intensity h^2 as a function of position at the maximum frequency in each frame of (a) which yielded a peak (note change in scales).

but in general have much less in common. As we have seen in the periodic structure, gap-soliton-mediated resonances evolve from Fabry-Pérot resonances in the transmission band. In the quasiperiodic structure, very sharp resonances arise in the gap but do not appear connected to the transmission bands. The resonances appear as islands in frequency-incident field space. These resonances are very sharp and do not, in general, yield the near-perfect transmission seen in the periodic case. The magnetic field intensity in the periodic case shows a well-defined envelope of fast periodic spatial oscillations. No such envelope is seen in the quasiperiodic case. While it is possible to make some analytic progress in understanding the soliton structure in the periodic case, at present no aids such as Bloch waves (the wave functions are more localized critical states) give insight to the quasiperiodic nonlinear system. The origin of gap resonances in the nonlinear quasiperiodic case remains an open question.

As mentioned in Ref. 11, our model of the nonlinear antiferromagnetic material is only approximate. We have assumed that the magnetization generated by the electromagnetic field may be written in the form

$$m=\chi_0^{(1)}+\chi_0^{(3)}|h|^2 h \tag{5.1}$$

with higher-order terms ignored.

[1] M. Kohmoto, S. Sutherland, and K. Iguchi, Phys. Rev. Lett. **58**, 2436 (1987).
[2] M. Nakayama, H. Kato, and S. Nakashima, Phys. Rev. B **36**, 3472 (1987).
[3] R. Merlin, K. Bajema, R. Clarke, F. Y. Juang, and P. K. Bhattacharya, Phys. Rev. Lett. **55**, 1768 (1985).
[4] M. Kohmoto, L. P. Kadanoff, and C. Tang, Phys. Rev. Lett. **50**, 1870 (1983).
[5] S. Ostlund, R. Pandit, D. Rand, H. J. Schellnhuber, and E. D. Siggia, Phys. Rev. Lett. **50**, 1873 (1983).
[6] D. T. Smith, C. P. Lorenson, R. B. Hallock, K. R. McCall, and R. A. Guyer, Phys. Rev. Lett. **61**, 1286 (1988); Jpn. J. Phys. **26**, 285 (1987).
[7] S. D. Sarma and X. C. Xie, Phys. Rev. B **37**, 1097 (1988).
[8] Wei Chen and D. L. Mills, Phys. Rev. Lett. **58**, 160 (1987); Phys. Rev. B **35**, 524 (1987); **36**, 6269 (1987).
[9] F. Delyon, Y. E. Levy, and B. Souillard, Phys. Rev. Lett. **57**, 2010 (1987).
[10] N. S. Almeida and D. L. Mills, Phys. Rev. B **36**, 2015 (1987).
[11] L. Kahn, N. S. Almeida, and D. L. Mills, Phys. Rev. B **37**, 8072 (1988).
[12] J. K. Vassiliou, J. Appl. Phys. **59**, 1125 (1986).
[13] V. Jaccarino (private communication).
[14] E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1970).
[15] D. L. Mills and S. E. Trullinger, Phys. Rev. B **36**, 947 (1987).
[16] H. G. Winful, J. H. Marburger, and E. Garmine, Appl. Phys. Lett. **35**, 379 (1979).