Localization of Light Waves in Fibonacci Dielectric Multilayers

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We have measured the optical transmission of quasiperiodic dielectric multilayer stacks of SiO_2 (A) and TiO_2 (B) thin films which are ordered according to a Fibonacci sequence $S_{j+1}=\{S_{j-1},S_j\}$, with $S_0=\{B\}$ and $S_1=\{A\}$ up to the sequence S_9 which consists of 55 layers. We observe a scaling of the transmission coefficient with increasing Fibonacci sequences at quarter-wavelength optical thicknesses. This behavior is in good agreement with theory and can be considered as experimental evidence for the localization of the light waves. The persistence of strong suppression of the transmission (gaps) in the presence of variations in the refractive indices among the layers is surprising.

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Localization of electronic states due to *disorder* (Anderson localization) is one of the most active fields in condensed-matter physics [1]. Recently it was recognized that localization could occur not only in disordered systems but also in the deterministic *quasiperiodic* systems [2]. In a quasiperiodic system two or more incommensurate periods are superimposed, so that it is neither a periodic nor a random system and therefore can be considered as intermediate between the two.

In one dimension, a specific quasiperiodic Schrödinger equation based on the Fibonacci sequence can be analyzed by a renormalization-group type theory [3,4]. In this model a simple binary quasiperiodic Fibonacci sequence is used which is constructed recursively as $S_{j+1}=\{S_{j-1},S_j\}$ for $j\geq 1$; with $S_0=\{B\}$ and $S_1=\{A\}$. In this sequence one has $S_2=\{BA\}$, $S_3=\{ABA\}$, $S_4=\{BAABA\}$, $S_5=\{ABABAABA\}$, and so on. It has been shown that the wave functions for this case (S_∞) are not exponentially localized but rather *quasilocalized* (less than exponential decay at large distance), and that they have a rich structure including scaling. Furthermore, many detailed and exact results have been proved for the spectrum of this problem [5].

While the localization of states was originally regarded as an electronic problem, it was later recognized that the phenomenon is essentially a consequence of the wave nature of the electronic states. Therefore such localization can be expected for any wave phenomenon, such as acoustic waves [6-8] and even optical waves [9], where the latter have relatively high frequencies. There are distinct advantages to studying localization using a classical wave equation as opposed to the quantum mechanical electronic problem. First, in the electronic problem there are other possible interactions, such as electron-electron and spin-orbit effects, that can complicate the situation. In addition, the problem is not strictly one dimensional. From an experimental perspective the fabrication of Fibonacci lattices can be difficult because of the presence of such imperfections as defects at the interfaces that tend to mask the effects of the localization.

In addition to localization, there are other important properties of classical wave phenomena that have analogs with the electronic problem. One such property is the occurrence of a complete "photonic band gap" in certain dielectric microstructures. This phenomenon is the absence of photon propagation modes in any direction for a range of frequencies. Recently this effect has been experimentally verified [10] for the transmission behavior of microwaves in a *periodic* array of air holes in a material with high refractive index.

In *disordered* dielectric materials complete localization of light waves (sometimes called "photon localization") still lacks experimental proof. However, several groups have already been able to show the existence of weak localization, which is a precursor to true localization, demonstrated in *coherent backscattering* of light under certain conditions [11-14]. This effect arises from constructive interference of backward-scattered waves and can be used to obtain values for the optical diffusion coefficient. Complete localization would be indicated by a vanishing diffusion coefficient. Very recently, an unusually small optical diffusion coefficient consistent with an onset of localization has been realized in transmission and scattering experiments with microwaves in *random* mixtures of aluminum and Teflon spheres [15]. Theoretically predicted scaling properties of the transmission with sample thickness were verified.

Concerning *quasiperiodic* systems, there have been attempts to observe the exotic wave phenomena of Fibonacci systems in synchrotron x-ray scattering [16] and Raman scattering spectra [17,18] of semiconductor superlattices [19], and in propagation modes of acoustic waves on corrugated surfaces [20,21]. Generally, however, there are extraneous effects in these systems and it is difficult to study the localization phenomenon by itself.

In this paper we report experiments on *optical Fibonacci dielectric multilayers*. In this system the one-dimensional theory is strictly valid [22]. Compared to semiconductor superlattices the production of thin film dielectric multilayers is easier, and it can be expected that the effects of quasilocalization for the energy band structure become immediately apparent in a simple mea-

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surement of the optical transmission coefficient.

The transmission of a normally incident light wave through interface $B \leftarrow A$ is represented by the transfer matrix

$$T_{BA} = \begin{pmatrix} 1 & 0 \\ 0 & n_A/n_B \end{pmatrix},$$

and the transmission through interface $A \leftarrow B$ by

$$T_{AB} = \begin{pmatrix} 1 & 0 \\ 0 & n_B/n_A \end{pmatrix}.$$

The propagation within a layer of material A (B) is described by

$$T_{A(B)} = \begin{pmatrix} \cos\delta_{A(B)} & -\sin\delta_{A(B)} \\ \sin\delta_{A(B)} & \cos\delta_{A(B)} \end{pmatrix},$$

where the phases δ are given by $\delta_{A(B)} = n_{A(B)} k d_{A(B)}$, where k is the wave vector in vacuum and where $d_{A(B)}$ are the thicknesses of the layers.

For a finite Fibonacci multilayer S_j which is sandwiched between two media of material of type A the corresponding transfer matrix is

$$M_j = M_{j-2} M_{j-1},$$

with the initial conditions $M_1 = T_A$ and $M_2 = T_{AB} T_B \times T_{BA} T_A$. The number of layers in S_j is denoted by F_j, and is given recursively by $F_j = F_{j-1} + F_{j-2}$, with $F_0 = F_1 = 1$. From this expression the transmission coefficient can be calculated as

$$T[S_j] = \frac{4}{|M_j|^2 + 2},$$

where $|M_j|^2$ is the sum of the squares of the four elements of M_j.

The equation for M_j has been considered as a dynamical map and it has been shown that a constant of motion exists in all cases [3]. For the case of normal incidence and $\delta_A = \delta_B = \delta$, a constant of motion is [22]

$$I = \frac{1}{4} \sin^2\delta \sin^2 \left[\delta \left(\frac{n_A}{n_B} - \frac{n_B}{n_A} \right)^2 \right].$$

This constant is always positive and represents the strength of the quasiperiodicity. For the case $\delta = m\pi$ (half-wavelength layer), $I = 0$ and the transmission coefficient is 100%. For the case $\delta = (m + \frac{1}{2})\pi$ (quarter-wavelength layer), I is maximum and the quasiperiodicity is most effective. Also, for the latter case the dynamical map has a period of six, which means that $M_j = M_{j+6}$ for any j at that particular phase. This fact implies that the transmission coefficient $T[S_j]$ exhibits a self-similar behavior about $\delta = (m + \frac{1}{2})\pi$, with $T[S_{j+3}] = T[S_j]$. (The period of the transmission coefficient is three recursions rather than six, due to the explicit forms of the six matrices $M_1, \ldots, M_6$ [22,23].) The scaling behavior of the transmission coefficient is characterized by the scale factor $f = [1 + 4(1+I)^2]^{1/2} + 2(1+I)$. The quantity f gives the scale change of the wave vector between spectra $T[S_j]$ and $T[S_{j+3}]$. Therefore, quasilocalization of the light waves in a Fibonacci dielectric multilayer is demonstrated by the self-similarity of the transmission coefficient under the given boundary conditions.

As thin film materials we chose silicon dioxide (A) and titanium dioxide (B). These are virtually absorption-free above 400 nm. Their indices of refraction at 700 nm are $n_A = 1.45$ and $n_B = 2.30$, respectively. Relative film thicknesses d_A and d_B of materials A and B were chosen such that the phase shift for normally incident light was the same, i.e., $\delta_A = \delta_B = \delta$. In this case $n_A d_A = n_B d_B$. Furthermore, the individual layers were taken as quarter-wave layers, for which the quasiperiodicity is expected to be most effective [21], and the central wavelength was chosen as 700 nm (14285 cm^{-1}). These conditions imply physical thicknesses $d_A = (700\ \mathrm{nm})/4n_A = 121$ nm and $d_B = (700\ \mathrm{nm})/4n_B = 76.4$ nm.

The layer systems were electron-gun evaporated on optically polished substrates of fused silica ($n = 1.46$, 25 mm diameter, 6.5 mm thickness). The base pressure of the evaporation chamber (Balzers BAK 600) before evaporation was better than 10^{-5} mbar. In order to achieve absorption-free coatings, layers were deposited in an oxygen atmosphere of several 10^{-4} mbar. The substrate temperature was 300 °C. Quarter-wave and half-wave optical thicknesses were optically monitored at 700 nm and adjusted to an accuracy of $\pm 2\%$. The coated substrates were contacted to an uncoated substrate of the same thickness using index matching fluid, and the transmission of the resulting sandwiched Fibonacci coating stacks was measured as a function of wave number $\tilde{\lambda} = k/2\pi$ in the 25000 cm^{-1} (400 nm) to 5000 cm^{-1} (2 μm) range (Cary 17 D spectrophotometer).

The experimental results for transmission are shown in Fig. 1 for Fibonacci sequences S_4 to S_9 as curves a, and compared for each case to calculated spectra shown as curves b. [The central wave number $\tilde{\lambda} = k/2\pi$ is at 14285 cm^{-1} (700 nm) and corresponds to a phase shift of 1.5π.] In curves b the wavelength-independent reflectivity at the two air/substrate interfaces (~8%) is not taken into account and therefore has to be subtracted for comparison with curves a, for which the maximum measured transmission reaches 92%. As a general trend it can be seen that with increasing layer number of the sequences, more and more transmission dips develop and some of them approach zero transmission (one-dimensional photonic band gap); therefore the total transmission over the spectral region of interest decreases. Nearly identical transmission behavior as compared to the theory is observed for Fibonacci sequences $S_4 = \{BAABA\}$, $S_5 = \{ABABAABA\}$, $S_6 = \{S_4, S_5\} = \{BAABAABABAABA\}$, $S_7 = \{S_5, S_6\}$, and $S_8 = \{S_6, S_7\}$. Small asymmetries of the transmission peaks occur in S_6 and S_7 near the central wave number,

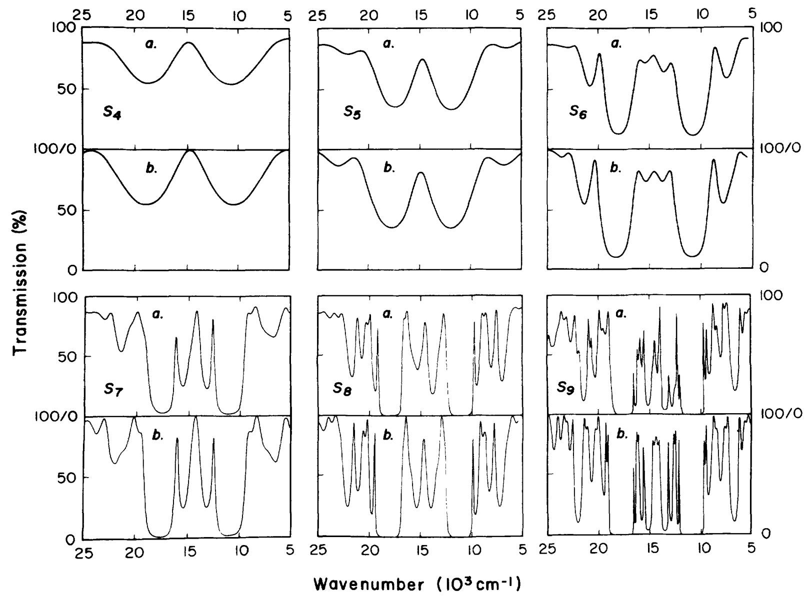

FIG. 1. Optical transmission spectra (transmission T versus wave number $\tilde{\lambda}$) for Fibonacci dielectric coating stacks S_4 to S_9. The dielectric stacks are sandwiched between 6.5 mm thick fused silica substrates. Curves a: experiment; curves b: theory.

and a small extra peak at 16500 cm^{-1} appears in S_8. In S_9, having 55 layers, spectral locations and shapes of the transmission peaks are still in good agreement with calculated spectra in the range 6000–10000 cm^{-1}. At higher energies, the spectral positions of the peaks still agree (with an exception near 20000 cm^{-1}); however, the relative strengths of the peaks are now severely distorted.

In order to check the predicted scaling of the transmission spectra about $\delta = (m+\frac{1}{2})\pi$ (corresponding to the central wavelength 700 nm), we determine the scaling factor f from $f = [1+4(1+I)^2]^{1/2} + 2(1+I)$, and $I = \frac{1}{4}\sin^2\delta \sin^2\delta (n_A/n_B - n_B/n_A)^2$ as $f = 5.11$, and compare in Fig. 2 the transmission spectra S_5 and S_6 with scaled spectra S_8 and S_9, respectively. The experimental curves are shown as solid curves, the calculated ones as dashed curves. Note the scale change for spectra S_8 and S_9, compared to S_5 and S_6. The self-similarity of spectra S_5 and S_8, and S_6 and S_9, respectively, is evident around the central wavelength, while major deviations occur in scaled S_9 near 16000 cm^{-1}. Some of the discrepancies in Fig. 2 outside the main band near 15000 cm^{-1} are to be expected since the scaling factor f must be applied from the center of each "optical band" and, in addition, $f = 5.11$ is only applicable to the central band. We attribute the distortions in the relative strengths of transmission peaks in S_8 and S_9 to experimental difficulties connected with depositing a large number of layers, most importantly perhaps variations in the index of refraction. It is known that with increasing layer number the films become less and less ideal, i.e., the structure of the films changes slowly from an initial homogeneous amorphous phase to an inhomogeneous porous columnlike crystalline phase with empty spaces in between crystallites (pore diameter ~10–100 nm). This change in morphology leads effectively to a change in the index of refraction of the film layers deposited later with respect to the initial ones. In order to simulate this expected effect we calculated the transmission of sequence S_9 assuming a 5% linear modulation of the index of refraction with increasing layer number. (The 5% modulation was chosen empirically as the "best fit" to the data. Experimentally, such a monatomic variation in the index is expected because of systematic changes in the composition of the layers with growth time.) The result is shown in Fig. 3, where the experimentally obtained spectrum is shown again for comparison. In agreement with experiment the simulation shows rather little deviation in the lower energy

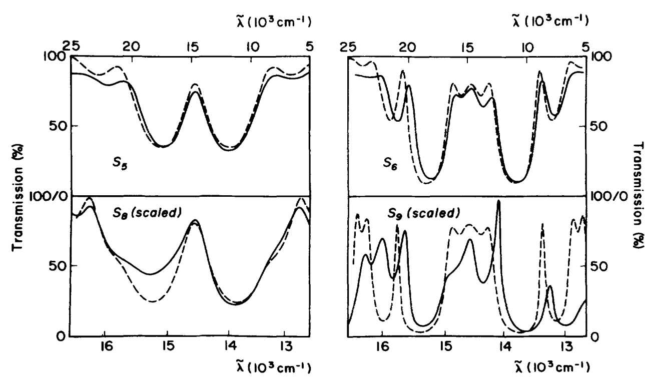

FIG. 2. Comparison of transmission spectra S_5 and S_6 with scaled spectra S_8 and S_9, respectively, showing self-similarity of corresponding transmission spectra. Relative to S_5 and S_6 the $\tilde{\lambda}$ axis of spectra S_8 and S_9 is expanded by the scaling factor $f = 5.11$. Solid curves: experiment; dashed curves: theory.

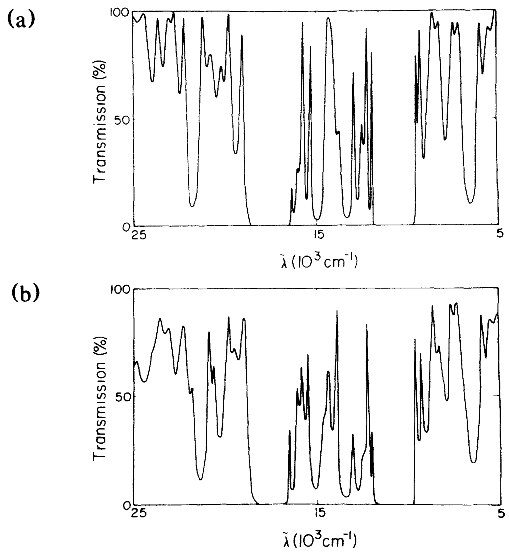

FIG. 3. Transmission spectra as a function of $\tilde{\lambda}$ for Fibonacci dielectric coating stack S_9: (a) calculated spectra for layer system with 5% linear modulation of index of refraction; (b) experiment.

range ($\tilde{\lambda} \leq 10000$ cm^{-1}), but at higher energies large changes in relative peak heights, spectral positions, and resolution occur.

It is interesting to note, particularly in the spectra for S_9 in Fig. 3, that although there are clearly changes in the positions of the peaks in transmission between the calculated and experimental spectra, the suppression of the transmission in the gaps is essentially unaffected by the variation in the indices of refraction. This insensitivity is encouraging for the possible use of these effects in optical devices since layer-to-layer variations are inevitable in real systems.

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Note added.—Since the submission of this manuscript experiments have been published [24] that contain results similar to some of those discussed in this Letter.

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