

X-RAY PROPAGATION THROUGH A QUASI- ORDERED MULTILAYERED STRUCTURE

by

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ABSTRACT

We investigate the propagation of short wavelength transverse electric x-rays through a quasi-ordered (Fibonacci) atomically commensurate multilayered structure using a transfer matrix model which treats each atomic plane as a diffraction unit. The reflectance spectrum has a rich structure being dominated by peaks associated with certain critical points of the system. Peaks around these special points exhibit self-similarity with a scaling factor approximately equal to the cube of the golden mean. At a critical point itself, the electromagnetic energy distribution is neither localised nor extended. The Landauer resistance at the reflection peaks exhibits a power law behaviour at low Fibonacci generation numbers but eventually increases exponentially; in the presence of absorption, the Landauer resistance reaches a plateau after its initial power law behaviour. The persistence, in the presence of absorption, of some reflectivity peaks at small angles of incidence is in line with current interest in the use of quasi-ordered multilayered structures as short wavelength near-normal incidence x-ray mirrors.

Keywords : X-rays, Fibonacci multilayered structures

INTRODUCTION

It has, for some time now, become technically possible to fabricate layered systems with film thicknesses as low as two or three atomic planes with a good degree of crystallinity (Schuller, 1980). These advances have resulted in the design and construction of novel artifacts such as (a) quantum-well heterostructures and solid state superlattices (Esaki, 1986), and (b) high reflectance x-ray mirrors (Underwood & Barbee, 1981; Lee, 1982; Evans *et al.* 1994). In the former case, because, often, the properties to be theoretically predicted are such that they are determined by those electron wavefunctions whose coherence are determined by characteristic length scales comparable with the layer dimensions, novel methods of theoretical analysis have had to be developed (Bastard, 1982). In the latter case, quantum size effects are not as important, since the interaction of x-rays with crystalline matter principally involves the low-lying energy bands. Here, the ratio of the wavelength to the film-thickness is an important parameter. With typical atomic plane spacings of about 5 Å and comparable x-ray wavelengths of 1-10 Å, a dielectric continuum model of crystal x-ray diffraction would be unreliable (Barbee, 1986). In fact, the scattering (Ashcroft & Mermin, 1988) approach recognises and accounts for the discreteness of the interaction through the form factor

$$\sum_{\mathbf{r}_n} \exp[i(\mathbf{k} - \mathbf{k}_0) \cdot \mathbf{r}_n]$$

where the lattice sites are denoted by \mathbf{r}_n , and where \mathbf{k} and \mathbf{k}_0 are respectively the diffracted and incident wave-vectors. As it is well-known, this sum is zero unless the scattering vector coincides with a reciprocal lattice vector, thereby expressing the Bragg diffraction condition. However, as secondary scattering is excluded, the anticipated interference with reflected radiation from the next and subsequent atomic planes cannot be considered. The method is thus insufficient for accurate reflectance calculations, unless the computationally demanding higher order scattering terms are included. The transfer matrix (Born & Wolf, 1989) method then becomes an attractive alternative.

Harper & Ramchurn (1987) have introduced a transfer matrix model (plane iterative model) where, within each plane, induced polarisation is averaged over atomic positions and varies spatially depending only on the phase of the incident and reflected radiation. Transverse to the plane, the polarisation is finite only within its atomic dimension. Effectively, the free-space radiative field changes discontinuously through the atomic plane. The atomic planes for this purpose are regarded as identical, any relative displacement as in the hexagonal close packed structure be-

ing disregarded. In this model, for the transverse electric case, the amplitudes E_i^2 and E_r^2 of the incident and reflected electric fields at an atomic plane are related to the corresponding amplitudes, E_i^1 and at the previous atomic plane by a transfer matrix, $T(\theta)$, constructed from a free-space E_r^1 , propagation matrix and a trans-plane matrix:

$$T(\theta) = \begin{bmatrix} \exp(i\psi) & 0 \\ 0 & \exp(-i\psi) \end{bmatrix} \begin{bmatrix} 1 + \frac{ik_0 a \chi}{2\cos(\theta)} & \frac{ik_0 a \chi}{2\cos(\theta)} \\ -\frac{ik_0 a \chi}{2\cos(\theta)} & 1 - \frac{ik_0 a \chi}{2\cos(\theta)} \end{bmatrix} \quad (1)$$

where, for an angle of incidence θ , $\psi = k_0 a \cos(\theta)$, a is the interplanar separation, and χ is the electric susceptibility. For an incident x-ray wavelength of λ , the free-space wavenumber k_0 is $k_0 = \frac{2\pi}{\lambda}$. Multiple scattering is included through the continuity conditions on the electric and magnetic field components at each boundary.

It has already been shown that, in the limit of small a , the model reduces to standard dielectric continuum models, and that multilayer x-ray reflection can be treated within this framework (Harper & Ramchurn, 1987). Using (1), the dynamical Darwin-Prins, modified Bragg, and kinematical relationships (James, 1948) for x-ray reflection can be derived. It is our purpose here to investigate the propagation of short wavelength transverse electric x-rays through an atomically commensurate quasi-ordered (Fibonacci) multilayer structure using this matrix model.

FIBONACCIMULTILAYEREDARTIFACTS

A generalised Fibonacci sequence is generated using the inflation rule

$$L \rightarrow L^m M, M \rightarrow L$$

L and M are two fundamental building blocks which, if they themselves are constituted of other repetitive blocks, lead to a superlattice (Merlin *et al.* 1985) structure. Being neither ordered nor disordered, these structures are termed quasi-ordered. With $m=1$, the popular Fibonacci (golden mean) lattice results. Thus, if S_n denotes the n^{th} sequence, then, with $S_0 = M$ and $S_1 = L$, we have $S_2 = LM$, $S_3 = LML$, and

$S_n = LMMLM$, etc. In any sequence S_n ($n \geq 2$), there are F_{n-1} blocks of type L and F_{n-2} blocks of type M, where F_n is a Fibonacci number with $F_n = F_{n-1} + F_{n-2}$, and $F_0 = 1$ and $F_1 = 1$. $n \rightarrow \infty$ corresponds to the golden mean quasiperiodic limit with

$$\frac{F_{n-1}}{F_{n-2}} \rightarrow \frac{1 + \sqrt{5}}{2} (= \tau)$$

Fibonacci multilayered artifacts are constructed by identifying the blocks L and M as layered artifacts. The study of these structures has become a subject of intense interest (e.g. Hu *et al.* 1986; Karkut *et al.* 1986; Kohmoto *et al.* 1987a; Chow & Guenther, 1993; Gellermann *et al.* 1994) following the extension of the original work of Kohmoto *et al.* (1983) and of Ostlund *et al.* (1983) on quantum quasiperiodic systems to the problem of localisation in optics (John, 1991). There has also been the realisation that the investigations of such systems may shed light on the properties of quasicrystals (Shechtman *et al.* 1984) and that novel physical properties may be associated with them. Kohmoto *et al.* (1983, 1984) studied the spectral properties of the quasiperiodic Schroedinger equation

$$\Psi_{n-1} + V_n \Psi_n + \Psi_{n+1} = E \Psi_n \quad (2)$$

where Ψ_n is the electronic probability amplitude at lattice site n and the on-site potential V_n takes on two values arranged in a Fibonacci way. The energy spectrum is a Cantor set with multifractal (Halsey *et al.* 1986) scaling. The allowed wavefunctions are neither localised nor extended but are either self-similar or chaotic and are termed critical (Kohmoto *et al.* 1987a; Kohmoto *et al.* 1987b). A one-dimensional model of a quasicrystal can be constructed by arranging bonds of two types of length in a Fibonacci sequence. A simple tight-binding model of the electronic states of this quasicrystal is given by

$$t_n \Psi_{n-1} + t_{n+1} \Psi_{n+1} = E \Psi_n \quad (3)$$

This is the off-diagonal version of (2). The coupling constant t_n represents the strength of hopping of an electron between sites n and $(n + 1)$ and, like V_n , takes on two values, t_A and t_B arranged in a Fibonacci sequence. The spectral properties of (3) are the same as those of (2) (Kohmoto *et al.* 1987b). The energy band structure is again self-similar (multifractal) with scaling factors, α, β , depending on the potential strength $\lambda = |t_A - t_B|$. At the centre of the spectrum, $\alpha \rightarrow \tau^3$ as $\lambda \rightarrow 0$

It is known that a renormalisation group approach can be used to describe the evolution of these systems if a scaling approach to the quasiperiodic limit is used. The band-structure problem can then be reduced to the study of a trace map with an energy independent constant of motion in the quantum case (Kohmoto *et al.* 1983). In the case of light propagating through a Fibonacci multilayered structure, a phase dependent constant of motion is defined (Kohmoto *et al.* 1987a). The effect of the quasiperiodicity is very strong when the optical phase d across each of the layers is an odd integral multiple of $\frac{\pi}{2}$. Prominent self-similarity and scaling is then observed in the reflectance spectra.

Another important quantity to investigate is the electromagnetic energy density distribution across the multilayered structure. For incident and reflected field amplitudes E_i and E_r at a layer-layer interface, the energy density there is given by

$$W = \epsilon \left[|E_i|^2 + |E_r|^2 + \left\{ \text{Re} \left(E_i E_r^* \exp(i2\delta) \right) \right\} (1 - \cos(2\theta)) \right]$$

where d is the optical phase, θ is the angle of incidence at the interface, and ϵ is the dielectric constant. We have computed the energy density distributions for light propagating normally through two dielectric (continua) media, L and M, of refractive indices $n_L = 2$ and $n_M = 3$ layered in Fibonacci sequences and bounded by free-space. Fig. 1 shows an S_2 structure. The thicknesses, d_L and d_M , of the layers and the wavelength of the incident radiation are chosen such that the optical phases $\delta_L = \delta_M = \frac{3\pi}{2}$. The critical energy distributions (Fig. 2) manifest the imbedded horse-shoe dynamics (Kohmoto & Ono, 1984) underlying the evolution of the trace map under successive renormalisations and illustrates the quasi-localisation of light in this structure. These distributions, as we have recently shown (Ramchurn & Baijnath, 1998), have multifractal properties. The spectrum of singularities (Halsey *et al.* 1986) are smooth and extend only over a finite range of scaling indices as is required of multifractals. The extension of some of the above ideas to the soft x-ray and extreme ultraviolet regimes has been performed by Feng *et al.* (1989) and Feng *et al.* (1990) through the studies of Fibonacci superlattices with the blocks L and M each consisting of two layers. The reflectance spectra here again display self-similarity and scaling.

Our objective, now, is to investigate whether these features of self-similarity, scaling, and critical electromagnetic energy distribution are preserved at atomically commensurate x-ray wavelengths, dimensions at which we expect the dielectric continuum model used in the above calculations to break down and the atomic planes to create additional superlattice effects. Our numerical investigations build upon those

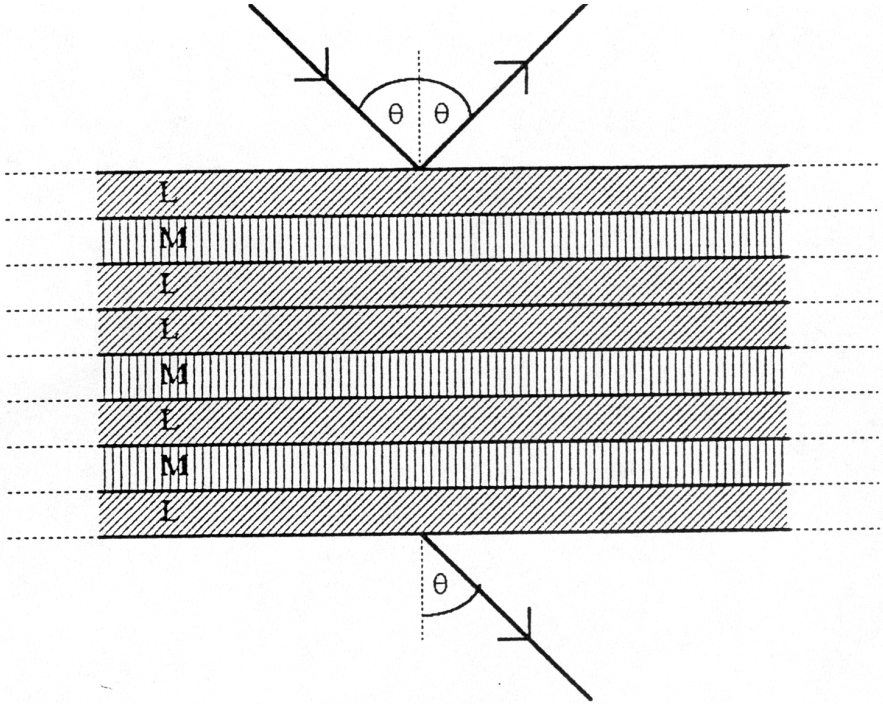


Fig. 1 Example of a Fibonacci multilayered structure. An \mathbb{S}_2 structure, bounded by free-space, is shown with radiation incident on it at an angle of incidence θ .

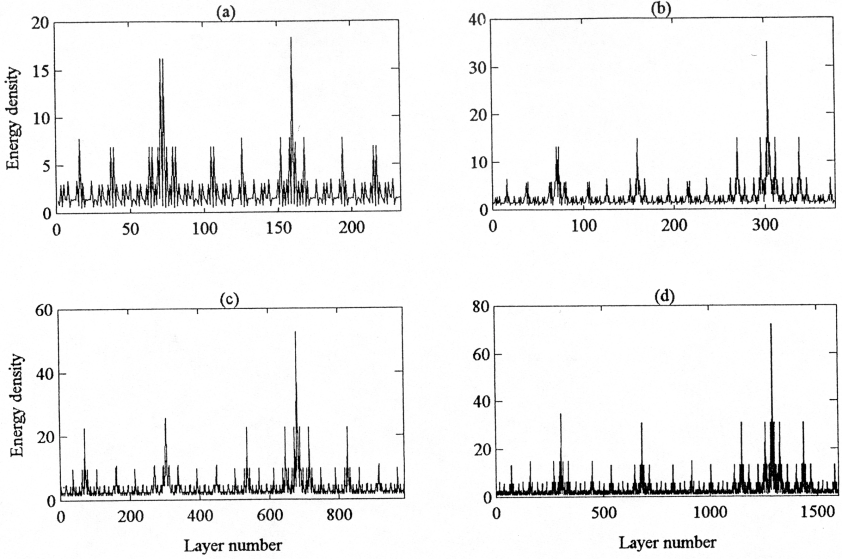


Fig. 2 Variation of the electromagnetic energy density (in arbitrary units) as light propagates normally through multilayered structures with layers of refractive indices 2 and 3 arranged in the following Fibonacci sequences: (a) S_{12} (b) S_{13} (c) S_{15} , and (d) S_{16} . The incident medium and the substrate each have a refractive index of unity. The wavelength of the incident radiation and the thicknesses of the layers are chosen such that the optical phase across each layer is $\frac{3\pi}{2}$.

we have already carried out (Harper & Ramchurn, 1987) for an atomically commensurate multilayer system with translational symmetry. In our previous work, we considered the reflection of transverse electric x-rays of wavelength 8.34 \AA ($AIK\alpha$) incident on a periodic double layer multilayered structure, with each period consisting of three atomic planes of platinum (corresponding to a thickness of 11.75 \AA) and ten atomic planes of carbon (corresponding to a thickness of 25 \AA), deposited onto a substrate of crystalline silicon. Here, we assume a Fibonacci multilayered structure with the building block L consisting of 4 atomic planes of carbon and 3 atomic planes of platinum while M consists of 10 atomic planes of carbon and 3 atomic planes of platinum such that $\frac{d_M}{d_L} = 1.69 = \tau$. We consider the reflection of transverse electric x-rays of wavelength 8.34 \AA incident from free-space on this structure. The substrate is again taken as silicon. The electric susceptibilities used are $\chi(\text{platinum}) = -2.1 \times 10^{-3} + i6.1 \times 10^{-4}$, $\chi(\text{carbon}) = -3.9 \times 10^{-4} + i1.9 \times 10^{-5}$, $\chi(\text{silicon}) = -3.8 \times 10^{-4} + i1.7 \times 10^{-5}$, and are derived from the atomic scattering factors tabled by Henke *et al.* (1981). The complex refractive index η is $\eta = \sqrt{1 + \chi}$. The real and imaginary parts of $\chi = \frac{1}{2} \chi = \delta - i\kappa$, are related to the atomic scattering factors $f_1 + i f_2$ according to $\delta + i\kappa = \frac{\hbar^2 \lambda^2}{2\pi} N_a (f_1 + i f_2)$, where $\hbar = 2.82 \times 10^{-12} \text{ m}$, and N_a is the number of platinum (or carbon) atoms per unit volume in the pure bulk material (Batterman & Cole, 1964).

Fig. 3 shows the results of our reflectance computations using the plane iterative model for real χ 's for the S_3 , S_{12} , and S_{15} Fibonacci sequences. Our analyses reveal scaling and self-similarity at various parts of the spectrum. We have, in particular, investigated such behaviour around the near-normal angle of incidence of $\theta = 15.8^\circ$. For purely real χ 's, we find a self-similar structure about this point, the scale change between either the S_3 and S_{12} structures, or the S_{12} and S_{15} structures being approximately equal to τ^2 . This value corresponds to the scaling factor at the centre of the spectrum of the corresponding quantum Fibonacci system in the limit of the strength of the potential tending to zero. For comparison, Fig. 3 also includes the reflectance spectrum for an S_{15} structure using the dielectric continuum model. It is obvious that the additional superlattice effects created by the atomic planes in the plane by

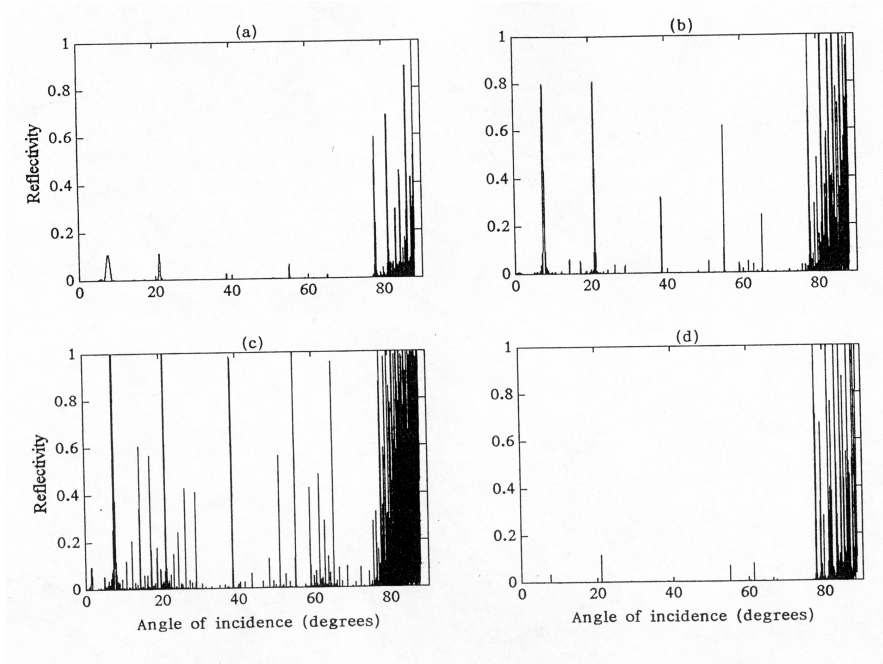


Fig. 3 Reflectivities, as a function of the angle of incidence, computed using the plane iterative model for the (a) S_9 , (b) S_{12} , and (c) S_{15} platinum-carbon Fibonacci superlattices described in the text. The incident x-ray radiation has a wavelength of 8.34 Å and each atomic plane is treated as a diffraction unit. (d) corresponds to an S_{15} computation using a dielectric continuum model. The electric susceptibilities used are $\chi(\text{Pt}) = -2.1 \times 10^{-3}$, $\chi(\text{C}) = -3.9 \times 10^{-4}$, $\chi(\text{Si}) = -3.8 \times 10^{-4}$, the substrate being silicon.

plane approach enhance reflection particularly at small angles of incidence. The electromagnetic energy distribution (neither localised nor extended) at $\theta = 15.8^\circ$ (Fig. 4), together with the scaling properties, emphasize this angle as a critical point of the system.

We have also calculated the Landauer (Landauer, 1970) resistance $\mathcal{R} = \frac{\mathcal{R}}{1 - \mathcal{R}}$ of the reflectivity peaks, of amplitude \mathcal{R} , with increasing Fibonacci generation number n . In general, the initial power law dependence of r on F_n changes to an exponential

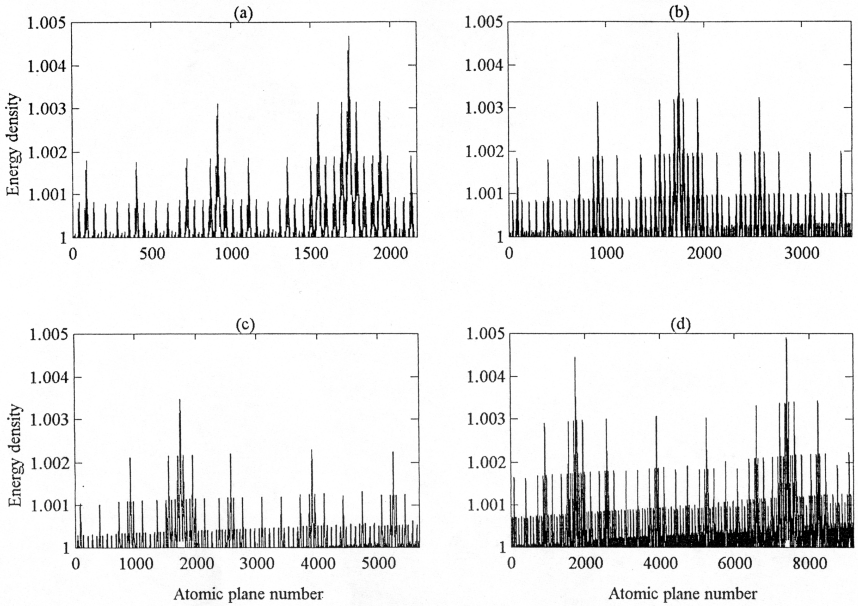


Fig. 4 Variation of the electromagnetic energy density (in arbitrary units) across the platinum-carbon Fibonacci superlattices described in the text for the S_{12} to S_{15} sequences at a critical point of the system. The wavelength of the incident x-ray radiation is 8.34 \AA and the electric susceptibilities used are as for Fig. 3.

one for large n . Fig. 5 shows this behaviour for the peak at $\theta \approx 7.6^\circ$. This is explained by the evolution of the trace, Γ , of the transfer matrix characterising the structure: $|\Gamma| < 2$ gives a power law growth while $|\Gamma| > 2$ leads to an exponential growth; $|\Gamma| = 2$ is a transition point.

More realistic reflectance computations with the use of complex ϵ' 's are shown in Fig. 6. The reflectivities are sensibly reduced, more so at the larger angle of incidence. Also, as Fig. 7 shows, the resistance reaches a plateau for $n \approx 13$. Spiller

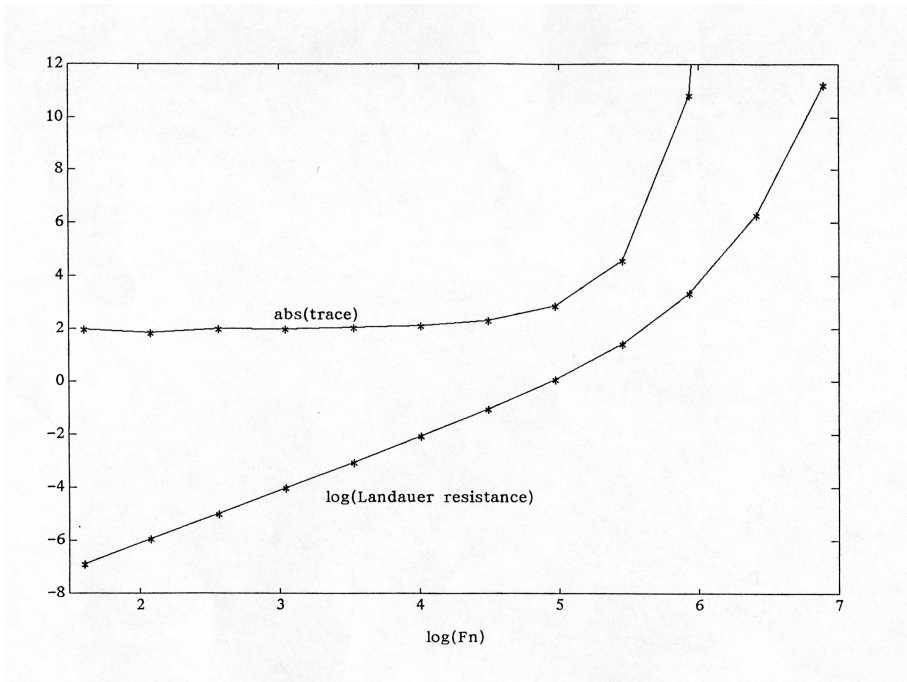


Fig. 5 Variation of the Landauer resistance with the Fibonacci number for the Fibonacci superlattices described in the text for the peak at an angle of incidence of approximately 7.6° . The wavelength of the incident x-ray radiation is 8.34 \AA and the electric susceptibilities used are as for Fig. 3. The variation of the trace of the matrix characterising the structure is also shown.

(1981) has estimated the number of layers, N_{max} , beyond which the reflectivity will not be increased in the absorbing case as $N_{\text{max}} = \frac{\cos^2(\theta)}{2\pi\kappa_{\text{sp}}}$ where κ_{sp} is the imaginary part of a refractive index. Here, for the peak at $\theta \approx 7.6^\circ$ and with $\kappa_{\text{sp}} \approx 10^{-4}$ to 10^{-5} , this corresponds to $N \approx 11$ to 16 . The effect of the number of layers contributing to reflection decreases with increasing angle of incidence and competes with the reflecting power of each atomic plane which increases with θ being given by $r = \frac{\chi}{\cos^2(\theta)}$ for $|\chi| \ll \cos^2(\theta)$. Obviously, for the case we are considering, the former is more important, being enhanced by the quasi-ordered arrangement of the layers. This contrasts with the periodic case whose Bragg reflection peaks increase in intensity as θ increases. Our work thus rejoins current efforts (Peng *et al.* 1991) to use Fibonacci multilayered structures as short wavelength near normal incidence x-ray mirrors. For a first order, and therefore high-intensity, peak at $\theta \approx 7.6^\circ$ at a wavelength of 8.34 \AA , a periodic x-ray multilayer structure requires a period of approximately 4.21 \AA ; this may be difficult to deposit at a wavelength of 8.34 \AA , a periodic x-ray multilayer structure requires a period of approximately 4.21 \AA ; this may be difficult to deposit.

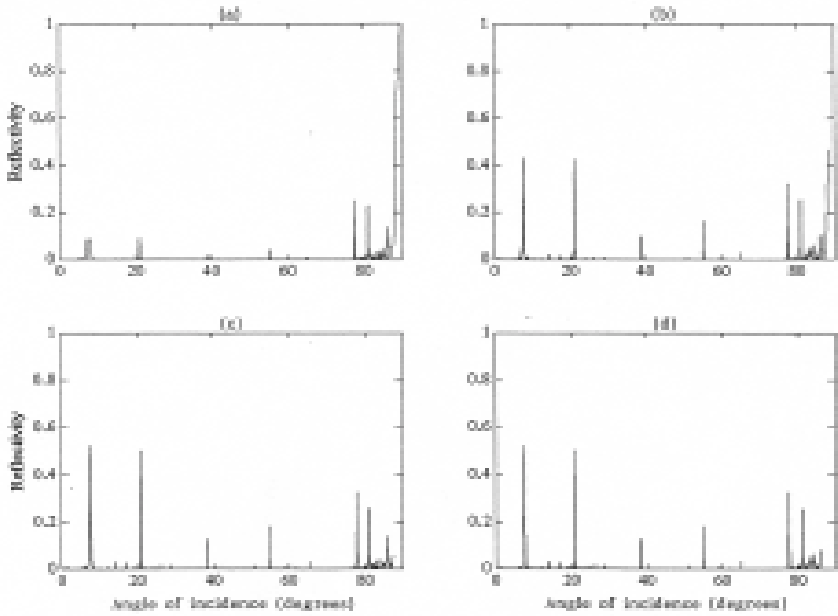


Fig. 6 Same as for Fig. 3 for the (a) S_9 , (b) S_{12} , (c) S_{15} , and (d) S_{16} platinum-carbon Fibonacci superlattices described in the text. However, the electric susceptibilities are now complex $\chi(\text{Pt}) = -2.1 \times 10^{-3} + i6.1 \times 10^{-4}$, $\chi(\text{C}) = -3.9 \times 10^{-4} + i1.9 \times 10^{-5}$, and $\chi(\text{Si}) = -3.8 \times 10^{-4} + i1.7 \times 10^{-5}$

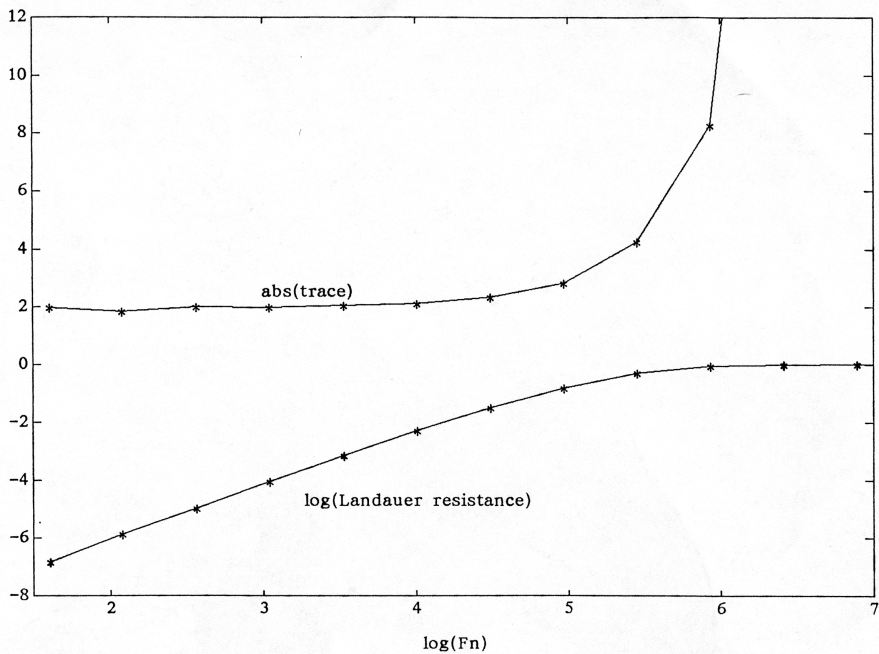


Fig. 7 Same as Fig. 5, with the use of complex electric susceptibilities:

$$\chi(\text{Pt}) = -2.1 \times 10^{-3} + i6.1 \times 10^{-4},$$

$$\chi(\text{C}) = -3.9 \times 10^{-4} + i1.9 \times 10^{-5}, \quad \text{and}$$

$$\chi(\text{Si}) = -3.8 \times 10^{-4} + i1.7 \times 10^{-5}$$

CONCLUSION

As concluding remarks, the following seem important: (a) the additional superlattice effects created by the atomic planes enhance reflection particularly at small angles of incidence, (b) peaks in the reflectance spectra are associated with certain critical points of the system, (c) in the presence of absorption, as shown by Landauer resistance computations, only a certain number of planes contribute to reflection with the persistence of some high reflectance peaks at some small angles of incidence, and (d) in the non-absorbing case, the computed electromagnetic energy at a critical point is neither localised nor extended. Reflectance peaks around the critical points exhibit self-similarity, a prominent scaling factor being approximately equal to the cube of the golden mean suggesting some three-cycle property about the critical points.

Further computational investigations reveal that significant departures from the orderly arrangement of the atomic planes do not affect the reflectance characteristics significantly. This robustness of reflectance characteristics to disorder - a special feature of quasiperiodic ordering - has also been observed experimentally (Todd *et al.* 1986; Gellermann *et al.* 1994) thereby emphasizing quasi-ordered superlattices as good candidates for high reflectance short wavelength x-ray mirrors even at angles near normal incidence and in spite of their absorbing nature. However, the controlled deposition of the atomic planes over many layers remains an experimental challenge (Chow & Guenther, 1993; Gellermann *et al.* 1994).

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