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X-ray reflectivity of Fibonacci multilayers

F. Domínguez-Adame, E. Maciá [1]

Departamento de Física de Materiales, Facultad de Físicas, Universidad Complutense, E-28040 Madrid, Spain

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Abstract

We have numerically computed the reflectivity of X-rays incident normally onto Fibonacci multilayers, and compared the results with those obtained in periodic approximant multilayers. The constituent layers are of low and high refractive indices with the same thickness. Whereas the reflectivity of periodic approximant multilayers changes only slightly with increasing the number of layers, Fibonacci multilayers present a completely different behaviour. In particular, we have found a highly fragmented and self-similar reflectivity pattern in Fibonacci systems. The behaviour of the fragmentation pattern on increasing the number of layers is quantitatively described using multifractal techniques. We end with a brief discussion on possible practical applications of our results in the design of new X-ray devices.

1. Introduction

The realization of well-controlled quasiperiodic superlattices [1,2] has led to a widespread theoretical interest in the study of one-dimensional quasiperiodic systems [3–5]. From the very beginning, most researchers have considered the Fibonacci sequence as the archetype of a quasiperiodic system [6,7]. This point of view is firmly established by X-ray diffraction analyses, which clearly reveal the quasiperiodic nature of Fibonacci superlattices, even if moderately large growth fluctuations during sequential deposition are present [1,3]. It is by now well known that Fibonacci lattices exhibit highly-fragmented energy spectra with a hierarchy of splitting subbands displaying self-similar patterns [8–10]. These novel features are directly related to the peculiar topological order displayed by the underlying structure, namely its quasiperiodic order [11]. But it is most important to stress that new and striking phenomena are not only found in the case of electron dynamics. In fact, harmonic vibrations in quasiperiodic lattices also present highly-fragmented and self-similar frequency spectra [12]. In addition, theoretical studies on incoherent [14] and Frenkel [15] excitons in Fibonacci systems have revealed a very different dynamics compared with that shown in both random and periodic lattices.

In this Letter we consider the X-ray reflectivity of a multilayered system where the refractive indices of the layers are arranged according to the Fibonacci sequence. The aim of this paper is twofold. In the first place, we carry out a theoretical investigation of Fibonaccian systems from a different perspective to shed more light onto the role played by quasiperiodic order in their physical properties. In the second place, our present study suggests new practical applications of Fibonacci systems. In particular, we shall demonstrate that Fibonacci multilayers can be used as selec-

[1] Also at Instituto de Estudios Interdisciplinares, El Guijo, Z4 Galapagar, E-28260 Madrid, Spain.

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tive filters for soft X-rays, allowing for a fine tuning of different narrow lines. This may be compared with recent results reported on multilayers with randomly varying thicknesses, which have been proposed as a broad bandwidth X-ray mirror [16].

The system we study in this work is a Fibonacci multilayer (FM) consisting of two different kinds of layers of the same thickness d. Layers A (B) have low (high) refractive indices n_A (n_B). For the sake of simplicity we neglect the absorption of X-ray incident normally onto the sample surface, so that the refractive indices are real parameters. Finally, we assume that the whole system is placed in vacuum. In general, a Fibonacci system of order l is generated from two basic units A and B by successive applications of the inflation rule A → AB and B → A. This sequence comprises F_{l-1} elements A and F_{l-2} elements B, F_l being the lth Fibonacci number given by the recurrence law $F_l = F_{l-1} + F_{l-2}$ with the initial values $F_0 = F_1 = 1$. As l increases the ratio F_{l-2}/F_{l-1} converges toward the so-called inverse golden mean $\tau = \frac{1}{2}(\sqrt{5} - 1) \sim 0.618\ldots$. The reflectivity of the FM structure is then easily computed numerically using Rouard's method (see, e.g., Ref. [16]). To facilitate direct comparison with previous studies of Yoo and Cue in random multilayers [16], we have taken the same physical parameters, namely $d = 50$ Å, $n_A = 0.9200$ and $n_B = 0.9995$. In this way we can separate those features of the reflectivity spectrum stemming from the underlying quasiperiodic order in a straightforward manner.

In periodic multilayers arranged according to the refractive indices sequence $n_A n_B n_A n_B \ldots$, the reflectivity shows a pronounced peak centered at 184 Å, with a bandwidth of about 11 Å [16]. Note that in this case the ratio c between the number of high and low refractive index layers is $c = \frac{1}{2}$. This value is not close to τ, so that it is difficult to carry out a direct comparison with the reflectivity obtained in FMs. Instead, it becomes more appropriate to consider periodic approximants to the quasiperiodic FM. To this end we have constructed periodic structures by a repetition of blocks of the form ABAAB ($c = 0.667$), or ABAABABA ($c = 0.600$), or ABAABABAABAAB ($c = 0.625$), namely the fourth, fifth and sixth order approximants to the FM. The number of blocks is repeated in any realization until the total number of layers N roughly equals the desired Fibonacci number.

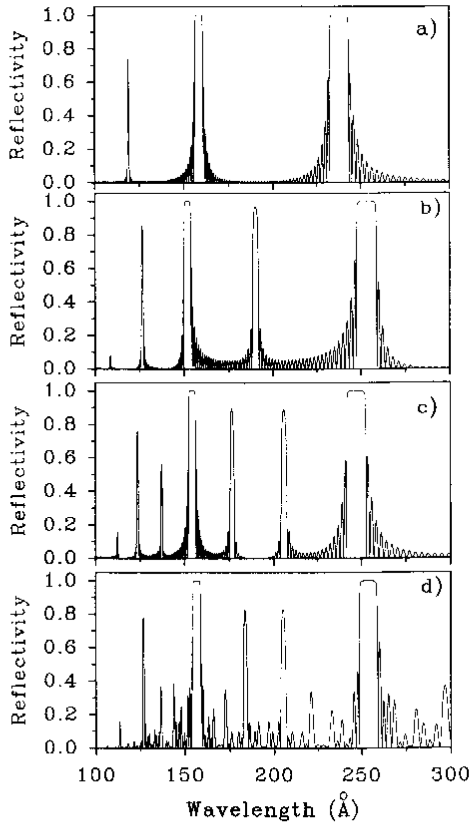

Fig. 1. X-ray reflectivity of N-multilayer structures with two basic layers of refractive indices $n_A = 0.9200$ and $n_B = 0.9995$, each layer thickness being $d = 50$ Å. Results for periodic approximants of (a) fourth order with $N = 235$, (b) fifth order with $N = 232$, (c) sixth order with $N = 236$, and (d) quasiperiodic FM with $N = F_{12} = 233$ are shown.

Fig. 1 shows the reflectivity $R(\lambda)$ corresponding to these periodic approximants (1a, 1b and 1c) and to the quasiperiodic $N = F_{12} = 233$ FM (1d). In all cases, two major reflection peaks are clearly observed at about $\lambda = 150$ Å and $\lambda = 250$ Å, whose bandwidths are 4 Å and 11 Å respectively, and a less pronounced peak close to $\lambda = 125$ Å. The position at which these major peaks are centered varies slightly depending on the order of the approximant considered. In addition to those major peaks, a set of *subsidiary* peaks displaying high reflectivity arises in periodic approximants, the number of them being increasingly large as the order of the approximant increases and, consequently, the envelope of $R(\lambda)$ is less uniform.

The origin of these subsidiary peaks can be eas-

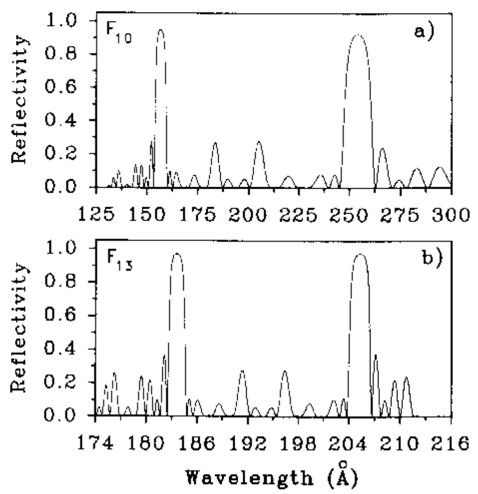

Fig. 2. Self-similarity of X-ray reflectivity of quasiperiodic FM with the same physical parameters n_A, n_B and d as in Fig. 1. The number of layers is (a) $N = F_{10} = 89$ and (b) $N = F_{13} = 377$.

ily visualized from a closer inspection if Fig. 1. Let us start by noting that no subsidiary peaks appear between the two major peaks in the reflectivity pattern corresponding to the fourth order approximant (1a). As soon as we increase by one step the order of the approximant considered, *one* prominent subsidiary peak arises between them (1b). By increasing the order of the approximant two steps, *two* subsidiary peaks appear; instead meanwhile *one* minor subsidiary peak develops between the major peaks centered at about $\lambda = 150$ Å and $\lambda = 250$ Å (1c). By further increasing the order of the approximant, an increasing number of subsidiary peaks progressively appear (this number is $F_{n-3} - 1$, $n \geqslant 3$ being the order of the considered approximant), until we obtain the reflectivity pattern of the quasiperiodic FM (1d). The resulting pattern forms a dense set of sharp and narrow peaks, some of them presenting a reflectivity larger than 50%.

Self-similarity and multifractal properties are both characteristic features of quasiperiodic orderings. Then it follows in a natural way that one looks for such properties in the reflectivity of FMs. In Fig. 2 we compare the reflectivity pattern of a FM containing $F_{10} = 89$ layers with the central portion of the reflectivity pattern of a FM containing $F_{13} = 377$ layers. This figure clearly shows the self-similar characteristics of the reflectivity peaks, that is to say, a given interval of wavelengths of a short FM is mapped onto a small interval of a larger FM. The rescaling procedure relates FMs with F_l and F_{l+3} layers, in an analogous fashion to self-similar electronic spectra [17].

In view of this result, we believe that it is important to get a quantitative estimate of the fragmentation as N increases. To be specific, it is clear that reflectivity of periodic approximants of any order changes very little on increasing the number of layers, whereas a hierarchical fragmentation process occurs in FMs. This is similar, although not identical, to what is found in the case of electronic properties (energy spectra and wave functions) in Fibonacci lattices. Following this analogy, we make use of multifractal analysis to get insight into the fragmentation of the reflectivity pattern. In particular, the participation ratio as defined, for instance, in Ref. [18], has been successfully used to describe the spatial nature (extended or localized) of electron wave functions. This method is readily generalized to the case of any positive measure defined in the system. Particularly, we can apply the same concepts to $R(\lambda)$ since it is a positive-defined quantity. Thus we introduce the participation ratio for the reflectivity as $P(N) = [\int R(\lambda)\,d\lambda]^2 / \int R^2(\lambda)\,d\lambda$. The value of $P(N)$ gives an estimate of the overall reflectivity of the sample as a function of the number of layers: The higher its value the higher the whole reflectivity. It is worth mentioning that performing the numerical integration requires very tiny integration steps since $R(\lambda)$ presents a more and more detailed structure on increasing N due to the hierarchical fragmentation scheme previously discussed. Thus one must repeat numerical integration with smaller and smaller integration steps until convergence is reached. Typically, 1.6×10^4 grid points are needed for a maximum number of layers of $F_{18} = 4181$. Fig. 3 shows the results obtained for both the fourth order approximant and the quasiperiodic FM as a function of the number of layers. Notice that periodic multilayers present an almost constant value of P for different numbers of layers, in agreement with the fact that the envelope of $R(\lambda)$ remains almost unchanged on increasing N in those systems. On the contrary, the value of P in FMs increases monotonically with N and, in addition, it is always larger than the corresponding value for periodic approximants of the same size. Therefore, we can conclude that the characteristic fragmentation process observed in quasiperiodic multilayers leads to an overall increase of their X-ray reflectivity. This in-

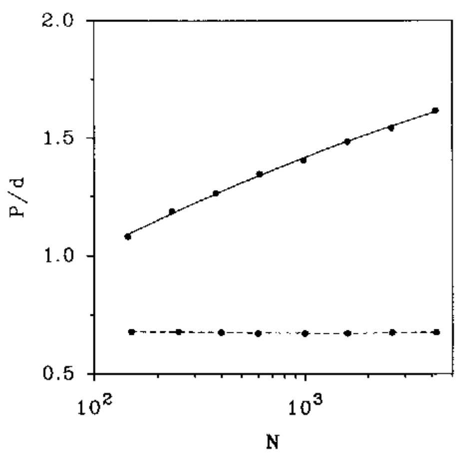

Fig. 3. Participation ratio P in units of d as a function of the number of layers N in periodic approximants (dashed line) and quasiperiodic FMs (solid line).

teresting property can be directly related to the self-similarity displayed by the reflectivity pattern. In fact, both the total number and the average height of subsidiary peaks progressively increase and new peaks appear after inflating the multilayer structure.

To summarize, we have numerically evaluated X-ray reflectivity of Fibonacci multilayers and compared it with that corresponding to periodic approximants. The main result is that both systems present a very different X-ray reflectivity incident normally onto the surface. Whereas reflectivity of periodic multilayers changes only slightly on increasing the number of layers, quasiperiodic multilayers present a completely different trend. We have observed a highly fragmented reflectivity pattern as a function of the incident wavelength. Whenever the order l of the FM increases (i.e., the inflation rule is applied), the subsidiary peaks increase their heights and new peaks arise. These new peaks also increase reflectivity on further increasing the system size. These conclusions have been established quantitatively by means of multifractal analysis and, in particular, using the participation ratio P. The value of P in periodic approximants remains constant on increasing N whereas in quasiperiodic systems it increases monotonically. Moreover, P is always higher in the later systems. Hence the overall reflectivity is larger in this case. Finally, some comments regarding multilayers with random thicknesses are in order. Yoo and Cue have recently demonstrated that random multilayers present a broad reflectivity peak, whose bandwidth broadens as fluctuations are stronger [16]. Hence, random multilayers increase the whole reflectivity on increasing fluctuations, and the system acts as a broad bandwidth mirror. We have found that Fibonacci multilayers also behave as a mirror but, unlike random systems, they should be regarded as X-ray selective filters instead. Notice that the position of subsidiary peaks can be changed by varying the refractive indices of the two constituent layers and/or the corresponding thicknesses. Therefore, Fibonacci multilayers open new possibilities in the *engineering* of soft X-ray devices.

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