

Spectral gaps and mode localization in Fibonacci chains of metal nanoparticles

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Abstract: In this paper we study the spectral, localization and dispersion properties of dipolar modes in quasi-periodically modulated nanoparticle chains based on the Fibonacci sequence. By developing a transfer matrix approach for the calculation of resonant frequencies, oscillation eigenvectors and integrated density of states (IDS) of spatially-modulated dipole chains, we demonstrate the presence of large spectral gaps and calculate the pseudo-dispersion diagram of Fibonacci plasmonic chains. The presence of plasmonic band-gaps and localized states in metal nanoparticle chains based on quasi-periodic order can have a large impact in the design and fabrication of novel nanophotonics devices.

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References and links

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1. Introduction

Quasi-periodic photonic structures are defined by simple mathematical rules which generate a non-periodic modulation of optical constants, encoding a fascinating complexity.

The Fibonacci sequence is the chief example of long-range order without periodicity, and can be easily generated according to the following inflation rule [1]: $A \rightarrow AB, B \rightarrow A$. The sequence F_n starts with an arbitrary seed element, $F_0=A$ for instance, and the inflation rule is repeatedly applied to obtain: $F_1=AB, F_2=ABA, F_3=ABAAB$, etc, which displays the well known Fibonacci symmetry: $F_{n+1}=\{F_n, F_{n-1}\}$ for $j \geq 1$.

Materials structures based on the Fibonacci sequence show distinctive diffraction patterns which cannot be found in either periodic or disordered systems [2]. Unlike periodic and random media, the Fourier spectra of Fibonacci structures consist of isolated Bragg peaks with incommensurate periods, which densely fill the reciprocal space [3]. Remarkably, the Bragg peaks of these characteristic types of spectra, known as singular continuous Fourier spectra, can only be indexed using two integers [3]. These spectral features, including the appearance of more Miller indices than the dimensionality of the physical system, induce fascinating physical properties in Fibonacci structures, which have been widely investigated in different physical contexts [3,4]. In particular, It has been shown that in Fibonacci structures the transport of wave-like excitations (such as electronic [5], optical [6], mechanical [7], spin waves [8]) as well as "mixed-waves" (polariton waves [8]) exhibits unique properties described by multi-fractal energy spectra [9], critical wavefunctions with power-law scaling [10] and anomalous diffusion [11].

In the context of optics, linear and non-linear Fibonacci dielectric multilayer structures have been widely investigated, both theoretically and experimentally, as a model of quasi-periodicity [6]. Light localization and spectral gaps formation in dielectric Fibonacci structures were theoretically investigated by Kohmoto et al [6]. Shortly after, pioneering experiments were performed by Gellermann et al. [12] who observed self-similarity in the transmission spectrum of Fibonacci dielectric multilayers and by Hattori et al. [13] who measured the Fibonacci dispersion curves. The dynamical transport properties of dielectric Fibonacci multilayers have been recently investigated by Dal Negro et al. [14], showing sizable field enhancement effects and group velocity reduction at the critically-localized band-edge states.

On the other hand, the present need to control and manipulate sub-wavelength local fields over nanodevices platforms has recently led to a "re-birth" of fundamental studies on surface plasmon propagation and electromagnetic coupling in metal nanoparticle arrays [15, 16]. In particular, periodic metal-nanoparticle chains [17] and bi-dimensional arrays [18] have been intensively investigated in the last five years, both theoretically and experimentally, in relation to the possibility of guiding/controlling sub-wavelength resonant fields via engineered near-field electromagnetic coupling [15-17,19]. However, the impact of quasi-periodicity on the plasmonic transport and localization properties of Fibonacci-based nano-devices has not been directly addressed so far.

In this paper we study the spectral, localization and dispersion characteristics of dipolar modes in quasi-periodically modulated nanoparticle chains based on the Fibonacci sequence. Based on a simple transfer matrix approach valid within the dipole approximation, we

calculate the resonant frequencies and dipole oscillation modes of a Fibonacci nanoparticles chain. In addition, we calculate the integrated density of states (IDS) and pseudo-dispersion diagrams of Fibonacci chains, and we demonstrate the presence of large spectral gaps in quasi-periodic plasmonic systems.

2. Computational method

In this section, we will introduce a simple and efficient transfer matrix approach for solving the set of coupled-dipole equations describing metal nanoparticle chains with arbitrary non-periodic modulation of the inter-particle spacing.

In particular, we will apply this model to the numerical calculation of the collective oscillations of finite-size, nanoparticle chains with Fibonacci modulation, and contrast our results against the simpler behavior of periodic chains of finite-length. As in Refs. [17, 19], we will consider only nearest-neighbors interactions within the dipole approximation, and we will assume Ag nanoparticles with minimum spacing of 25 nm. These assumptions are generally valid as long as higher order multipolar interaction terms do not significantly influence the quasi-static approximation. Even though more general models including all multiple orders have now been successfully developed for periodic chains [20, 21], it has been shown experimentally that the dipole approximation is sufficiently accurate for the description of electromagnetic transport in chains of Ag nanoparticles with up to 50 nm diameter and 25 nm inter-particle separation [17]. Additionally, since here we are only interested in comparing the transport properties originating from the geometrical modulation of the particle positions, we will limit our discussion to dipole chains, which have been largely investigated in the context of periodic metal-nanoparticle arrays [15-21].

Differently from the periodic case, a Fibonacci chain can be simply obtained by a modulation in the particle positions which results from the identification: $d_A \equiv A$ and $d_B \equiv B$ in the symbolic Fibonacci sequence of any given order F_n [see Fig. 1(a)].

In this paper, without loss of generality, we will discuss the properties of Fibonacci chains with 144 particles (corresponding to F_{11}), which is a good approximation of a “bulk” Fibonacci structure. In addition, we will choose $d_A=25$ nm and $d_B=30$ nm which, as explained above, ensures the validity of our dipole approach [17,19,20].

It is well-known that for a linear chain of Hertzian dipoles, the dipole equations of motion are given by [19]:

$$\ddot{p}_{i,n} = -\omega_0^2 p_{i,n} - \Gamma_e \dot{p}_{i,n} + \frac{\Gamma_R}{\omega_0^2} \ddot{p}_{i,n} - \gamma_i \alpha_n^2 (p_{i,n-1} + p_{i,n+1}) \quad (1)$$

where: $p_{i,n} = qx_{i,n}$ is the individual dipole, $x_{i,n}$ is the distance from equilibrium in the i polarization direction at point n along the chain, ω_0 is the metal plasma frequency, Γ_e is the electronic relaxation frequency, Γ_R is the relaxation frequency due to radiation into the far-field, γ_i is a polarization-dependent term ($\gamma=1$ for transverse modes and $\gamma=-2$ for longitudinal) and $\alpha_n^2 \propto 1/d_n^3$ is the near-field coupling term [19]. All the materials parameters used here are as in Ref. [19], with the important exception that for non-periodic chains the dipole-dipole coupling term α_n is not constant, because nearest neighbors of the n th dipole do not repeat regularly, but they are now modulated according to the quasi-periodic Fibonacci sequence. Therefore, the positional disorder of the Fibonacci chain induces a non-uniform coupling along the chain. As a result, an analytical solution cannot be found for the eigenmodes of the Fibonacci dipole chain. However, simple transfer matrix approaches exist for the numerical solution of harmonic chains with spatially-modulated coupling [22, 23]. In the past, transfer matrix techniques have been extensively applied to the calculation of phonon modes in quasicrystals [3]. Here, based on a similar approach, we have developed a simple technique for the numerical solution of modulated dipole chains, and we have specifically

investigated the Fibonacci case. We will first transform Eq. (1) in the frequency domain, and rearrange the terms to obtain:

$$u_{n+1} = \left[\frac{\omega^2 - \omega_0^2 - i(\Gamma_e \omega - \Gamma_R \omega^3 / \omega_0^2)}{\gamma_i \alpha_n^2} \right] u_n - u_{n-1} - u_{n+1} \quad (2)$$

where u_n is the Fourier transform of the oscillation amplitude of dipole n . Equation (2) can now be reduced to a simpler transfer matrix form as:

$$\begin{pmatrix} u_{n+1} \\ u_n \end{pmatrix} = T_n \begin{pmatrix} u_n \\ u_{n-1} \end{pmatrix} \quad (3)$$

where we have defined the unimodular transfer matrices T_n , relating the u_n amplitudes to each other, as:

$$T_n = \begin{pmatrix} \frac{\omega^2 - \omega_0^2 - i(\Gamma_e \omega - \Gamma_R \omega^3 / \omega_0^2)}{\gamma_i \alpha_n^2} & -1 \\ 1 & 0 \end{pmatrix} \quad (4)$$

It is important to notice that the position-dependent coupling terms α_n , which depend purely on the geometrical arrangement of the particles, are known “in advance” by the application of the Fibonacci rule. Once all the α_n have been found, the displacement of the N th dipole with frequency ω , $u_N(\omega)$, can be easily calculated by cascading the individual transfer matrices:

$$\begin{pmatrix} u_{N+1} \\ u_N \end{pmatrix} = T_N T_{N-1} T_{N-1} \dots T_2 T_1 \begin{pmatrix} u_1 \\ u_0 \end{pmatrix} = Q \begin{pmatrix} u_1 \\ u_0 \end{pmatrix} \quad (5)$$

Imposing the fixed boundary condition $u_0 = u_{N+1} = 0$, which describes an N -particle chain, we see that the vibration frequencies of the plasmonic chain must satisfy the eigenvalue equation:

$$Q_{11}(\omega) = 0 \quad (6)$$

which can now be solved numerically. The eigenvectors of Eq. (6) yields the oscillation amplitudes of the dipole chain, while the eigenvalues yield the oscillation frequencies. Here, following Ref. [19] we will assume only negligible plasmon damping, and therefore will only deal with the real part of Q_{11} , restricting the root finding procedure to real numbers. This choice will allow us to directly focus on the plasmonic transport properties uniquely induced by the quasi-periodic geometry of the Fibonacci chain, regardless of the specific materials parameters. However, it remains clear that the described approach is also suited for arbitrary dispersion parameters, as long as complex-plane root finding is allowed. Once the oscillation properties of the plasmonic chain have been obtained, the integrated density of states (IDS) of the chain can be simply obtained as [3]:

$$g(\omega) = \sum_k \delta(\omega - \omega_k) \quad (7)$$

The character of the eigenstates is described by the participation ratio [3]:

$$P(\omega_k) = \frac{\sum_n |e_n(\omega_k)|^2}{N \sum_n |e_n(\omega_k)|^4} \quad (8)$$

where $e_n(\omega_k)$ is the eigenvector defining the polarization of the atomic displacement for the mode of frequency ω_k on the atom at position n . When the orthogonality relation is imposed on the eigenvectors:

$$\sum_n |e_n(\omega_k)|^2 = 1 \quad (9)$$

the value of the participation ratio yields a quantitative measure of the localization degree of the eigenstates [3]. For extended modes $P(\omega_k) \approx 1$, while for the most localized modes (where $|e_n(\omega_k)|^2 = 0$ for all n except one) $P(\omega_k) \approx 1/N$, which is almost zero in the limit of long chains. The participation ratio of an arbitrary mode can therefore take a value between $1/N$ and 1, depending on the degree of localization of the modes.

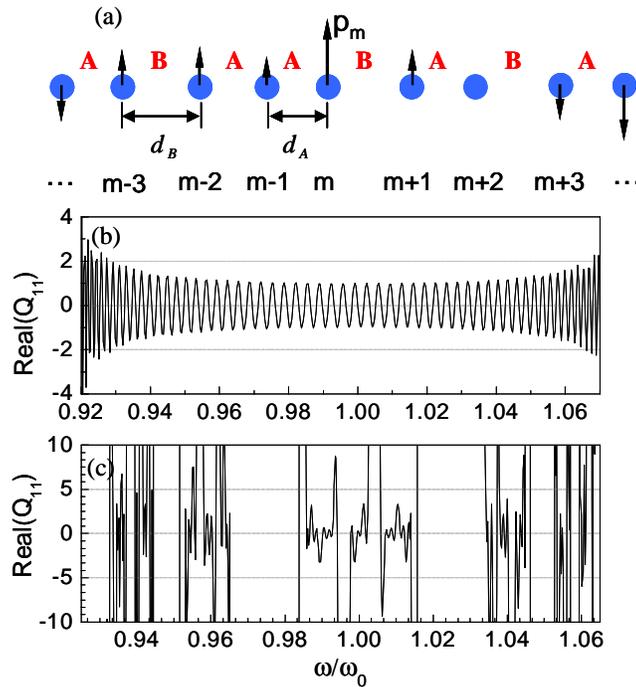


Fig. 1. (a). Schematics of the aperiodic nanoparticle chain, and real parts of the transfer matrix element Q_{11} for (b) periodic and (c) aperiodic Fibonacci chains.

3. Results and discussion

In Figs. 1(b) and 1(c) we plot the real part of Q_{11} for a periodic and a Fibonacci quasi-periodic chain, respectively. As expected for a general N -particles chain, in both the cases the number of oscillation frequencies (the zeros of Eq. 6) equals the number of particles in the chain. However, in the case of the Fibonacci chain, the distribution of zeros shows non-periodic frequency gaps separating groups of allowed modes. In order to clearly relate this observation with the plasmon transport along the Fibonacci chain, we calculated the Integrated Density of States (IDS) of periodic and Fibonacci plasmon chains, as described in the previous section.

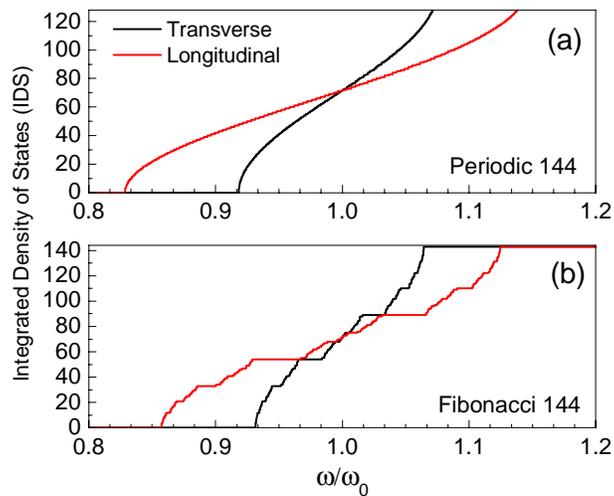


Fig. 2. Integrated density of states (IDS) for the (a) periodic and (b) quasi-periodic Fibonacci chains.

In Fig. 2 we show the longitudinal and transverse IDS branches calculated for a periodic (a) and a Fibonacci chain (b) with the same number of particles. We notice that, contrary to the case of a periodic chain, the Fibonacci quasi-periodicity induces a non-uniform distribution of IDS spectral gaps for both polarizations [Fig. 1(b)]. As seen from Fig. 2(b), the longitudinal IDS gaps are significantly wider than the transverse ones due to the stronger electromagnetic coupling of longitudinal modes. In addition, it is important to notice here that the two central Fibonacci gaps shown in Fig. 2(b) already appear even for shorter Fibonacci chains down to only 21 particles. However, as the particle number is increased, smaller and smaller satellite plasmonic gaps, nested in a self-similar hierarchy emerge across the frequency range. This is the typical behavior of the Fibonacci energy spectra, which in the limit of many particles converge to Cantor set fractals. Interestingly, the Fibonacci IDS gaps shown in Fig. 2(b) appears to follow a non-uniform distribution similar to the multi-fractal Devil's staircase distribution [24], which describes intermittent chaos and quasi-periodicity in many different areas of physics [25, 26].

In Fig. 3 we show the spatial profiles of the most localized dipole modes (in order of decreasing participation ratio) found in periodic [Fig. 3(a)] and Fibonacci chains [Figs. 3(b), 3(c)]. The mode shown in Fig. 3(a) corresponds to the well known transverse band-edge mode found in periodic chains [3]. On the other hand, the Fibonacci modes shown in panels (b) and (c) display the characteristic features of critical and gap-edge modes, respectively [3]. Our findings show that, in the case of negligible plasmon damping, localized states with very low participation ratio exist in coupled-dipole chains based on quasi-periodic Fibonacci order.

However, the Fibonacci frequency gaps (band-gaps) can only be completely understood by studying the band-dispersion diagram of the structures, as shown in Fig. 4. In the case of periodic plasmon chains [Fig. 4(a)], our numerical model accurately reproduces the analytical solutions calculated, in the limit of low damping, in Ref. 19. As discussed in Ref. 19, we notice that for periodic chains the group velocity vanishes at the borders of the Brillouin zone, and that the bandwidth of the longitudinal branch is larger than that of the degenerate transverse branch. Interestingly, the transverse branch shows a negative slope across most of the Brillouin zone, compatible with a negative index of refraction.

On the other hand, both the definition and the calculation of the dispersion diagrams for a Fibonacci quasi-periodic chain [Figs. 4(b) and 4(c)] certainly deserve more attention. In fact, the lack of periodicity in a Fibonacci chain prevents proper definition of a reciprocal space, and modes cannot be considered as simple plane waves with well-defined wavevectors [3].

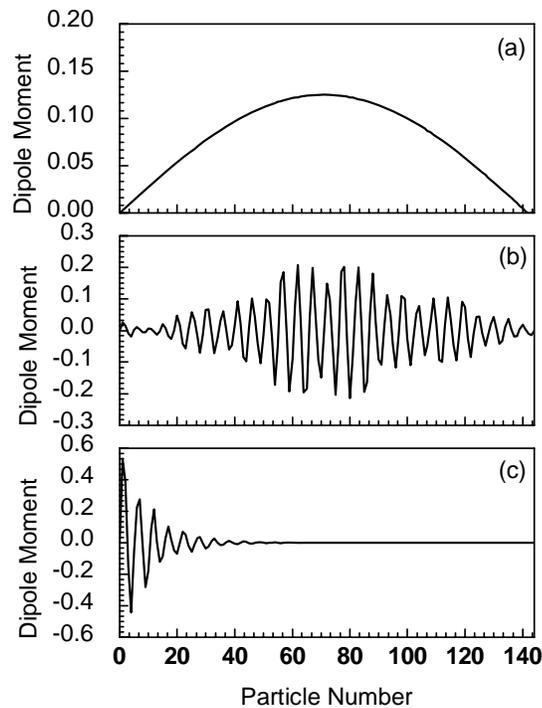


Fig. 3. Dipole moments of the (a) extended state of a periodic chain, (b) critical state ($\omega/\omega_0 = 0.986$) and (c) the most localized states ($\omega/\omega_0 = 0.984$) of a quasi-periodic Fibonacci chain. The participation ratios for cases (b) and (c) are 0.201 and 0.044.

Furthermore, the description of excitations in terms of a dispersion law $\omega = f(k)$ becomes inappropriate in non-periodic chains. In the case of a quasi-periodic chain, the spatial disorder (Fibonacci quasi-periodicity) induces spatially localized modes which can be described by a superposition of numerous Fourier components with different relative intensities. This situation is reflected in the Fibonacci mode profiles shown in Fig. 3. However, by determining the spatial frequencies associated with Fibonacci modes of different angular frequencies, it is possible to define dispersion diagrams, known as pseudo-Brillouin zones, even in the case of non-periodic structures [3].

The pseudo-dispersion curves of the Fibonacci plasmon chain, for the two different polarizations, are shown in Figs. 4(b) and 4(c). The intensity of the Fourier components of each mode is suggested by the dot sizes and colors. We notice that Fibonacci pseudo-dispersion diagrams of coupled-dipole chains show a remarkable similarity with pseudo-Brillouin zones widely investigated in the context of quasicrystals phonons [3, 4, 7].

Indeed, the pseudo-dispersion diagrams shown in Figs. 4(b) and 4(c) are characteristic of monatomic chains with a modulation period incommensurate with the lattice constant [3] and, as for any general linear chain system, *mass modulation* and *gap openings* in the energy spectrum are closely related phenomena.

Our results clearly demonstrate that the Fibonacci modulation of the electromagnetic coupling in plasmonic chains is responsible for the gap formation in the pseudo-Brillouin zone, for both longitudinal and transverse modes.

The presence of plasmonic band-gaps and localized states in metal nanoparticle chains based on quasi-periodic order can have a large impact in the design and fabrication of novel nanophotonics devices.

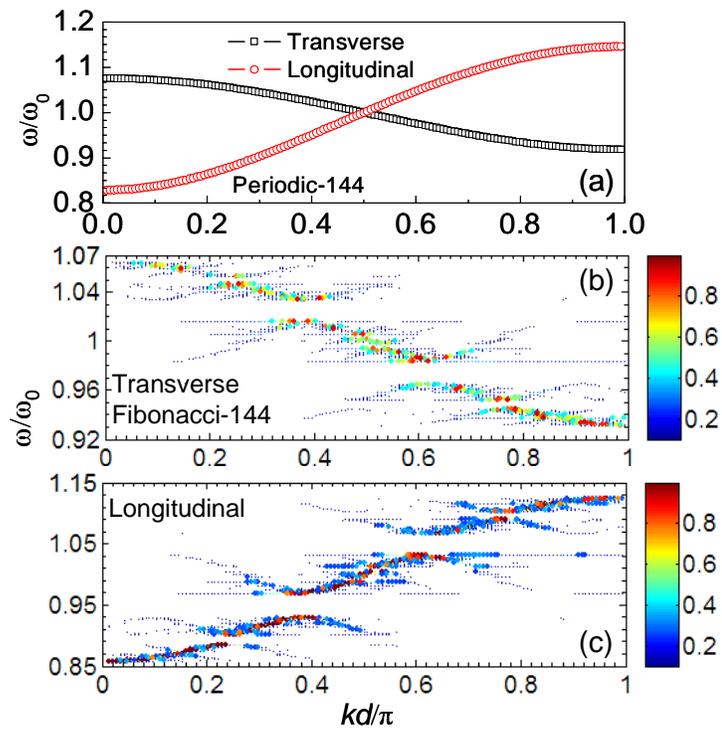


Fig. 4. Dispersion diagrams for (a) periodic chain: (b) quasi-periodic Fibonacci chain, transverse polarization; (c) quasi-periodic Fibonacci chain, longitudinal polarization.

4. Summary

In this paper we have investigated the spectral, localization and dispersion properties of dipolar modes in non-periodically modulated nanoparticle chains based on the Fibonacci sequence. We have developed a simple transfer matrix approach for the calculation of resonant frequencies, oscillation eigenvectors and integrated density of states (IDS) of arbitrarily modulated dipole chains, and we have demonstrated the presence of plasmonic band-gaps in the pseudo-dispersion diagram of Fibonacci chains.

Acknowledgments

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