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Parametric disordering-driven topological transitions in a liquid metacrystal

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Electromagnetic metamaterials are the artificial composites structured on subwavelength level, and their macroscopic characteristics are defined by the design of structural elements (meta-atoms) and architecture of the unit cell. Tunable structures are of particular interest as they can increase functionality and/or broaden the bandwidth of metamaterial devices [1, 2], and reconfigurable *liquid metamaterials* can provide a basis for the design of highly tunable nanophotonic devices [3]. The plasmonic negative-index liquid metamaterials were first suggested in the papers [4, 5]. Later on, the new kind of liquid metamaterial called liquid metacrystal (LMC) was theoretically studied in [6].

It was supposed in [6] that such a material may be realized as an array of *resonant* micro- or nanoparticles (meta-atoms) with *anisotropic* polarizability suspended in viscous medium (e.g. liquid or gel). The external dc electric field applied to LMC aligns meta-atoms along one axis that imparts the anisotropic properties to the metamaterial. The electromagnetic waves propagation in LMC strongly depends on the relative orientation of the anisotropy axis and the wavevector, that makes it possible to change the effective refraction index of LMC simply by changing the orientation of dc electric field. The meta-atoms can also be reoriented by means of high-frequency electromagnetic field that results in strong nonlinearity of LMC. These properties of LMC were predicted in [6, 7] and basically demonstrated in experiment [8].

In this work we report the unusual type of LMC nonlinearity which is caused by the resonant interaction between the individual meta-atoms and linearly polarized amplitude-modulated electromagnetic wave. We consider the resonant interaction of the *mechanical* angular oscillations of elongated meta-atom with the modulated electromagnetic wave when the modulation frequency is close to the doubled mechanical resonance frequency. In dc or ac electric field a meta-atom with anisotropic polarizability orients along the field direction. At the same time, being deviated from the direction of dc external control field, the meta-atom will experience oscillations near this equilibrium state.

It is shown (See Supplemental Material) that the squared eigenfrequency of angular oscillations depends linearly on the squared static field strength and intensity of electromagnetic wave and can be represented as follows:

$$\Omega^2 = F_0 E_0^2 + F_{\sim} \mathrm{Re} G(\omega) |E_{\sim}|^2. \qquad (1)$$

Here E_0 ($E_{\sim}$) is the external dc (ac) electric field, $G(\omega) = \omega_0^2/(\omega_0^2 - \omega^2 + \mathrm{i}\gamma\omega)$ is the function describing a resonance response of the meta-atoms, ω_0 is the corresponding eigenfrequency, γ is the damping coefficient, and F_0 ($F_{\sim}$) is the static (high frequency) form-factor depending on the shape and size of the meta-atom.

The intensity of amplitude-modulated wave changes in time as $I = I_0(1 + \nu_0 \cos \Omega_{\mathrm{M}} t)$ $(0 < \nu_0 < 1)$, and the eigenfrequency of mechanical oscillations of meta-atoms becomes also temporally modulated

$$\Omega^2 = \Omega_0^2 (1 + \nu \cos \Omega_{\mathrm{M}} t), \qquad (2)$$

where $\Omega_0^2 = F_0 E_0^2 + F_{\sim} \mathrm{Re} G(\omega) |E_{\sim}|_0^2$, $|E_{\sim}|_0^2$ is the mean value of $|E_{\sim}|^2$ averaged over the modulation period, and $\nu = \nu_0 F_{\sim} \mathrm{Re} G(\omega) |E_{\sim}|_0^2/(\Omega_0^2)$.

In this case, the dynamics of the angular deviation θ of meta-atom dipole moment from the equilibrium is described by nonlinear oscillator equation with the eigenfrequency explicitely depending on time:

$$\frac{d^2\theta}{dt^2} + \eta \frac{d\theta}{dt} + \frac{1}{2}\Omega_0^2 \left(1 + \nu \cos \Omega_{\mathrm{M}} t\right) \sin 2\theta = 0, \qquad (3)$$

where η is the damping coefficient of mechanical oscillations of the meta-atom. For small deviations when $\sin 2\theta \approx 2\theta$ Eq. (3) is actually Mathieu equation.

It is well-known that Mathieu equation describes the so-called parametric instability: if the modulation frequency is approximately twice the eigen frequency of the oscillator, $\Omega_{\mathrm{M}} = 2\Omega_0(1 + \delta)$ $(\delta \ll 1)$, the oscillator phase-locks to the parametric variation and absorbs energy that leads to the exponential growth of the oscillation amplitude, which saturates only at nonlinear stage.

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The typical temporal dependences of $\theta(t)$ during the instability growth and saturation are shown in Fig. 1a. On the other hand, direct numerical evaluation of Eq. (3) demonstrates that sometimes no stationary state exists and the angular dynamics of the meta-atom becomes chaotic. The transition from dynamical to stochastic regime is clearly seen in Fig. 1b, which shows the dependence of the angular deflection amplitude, $|\theta|_{\max}$ on the modulation parameter ν. The smooth functions that are rather close to the theoretical predictions change into highly irregular dependencies, and the chaotic regime is established at $|\theta|_{\max} \approx 0.64$.

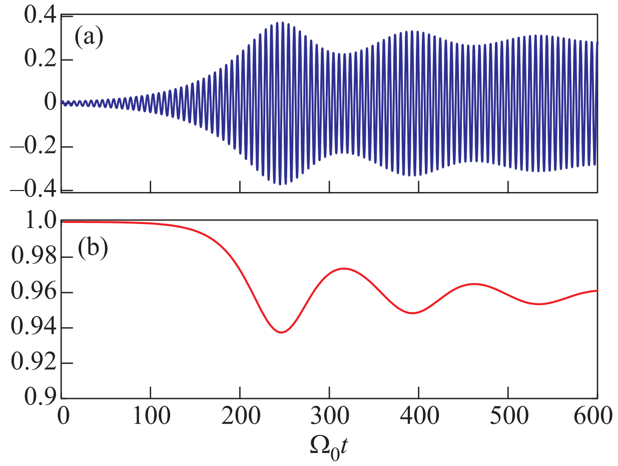

Fig. 1. (Color online) (a) – Time dependence of the angular deviation θ of the meta-atom from its equlibrium. (b) – The amplitude $|\theta|_{\max}$ of the angular meta-atom's deviation from its equlibrium as a function of the modulation amplitude; the corresponding dependencies in stochastic regime are shown with red dashed curves. The numerical results (thick lines) are compared to the theoretical predictions (thin black lines)

The parametric instability changes the mean orientation of the meta-atoms, thus leading to modification of the dielectric tensor of the medium. As in the case of conventional liquid ctystals, this orientational nonlinearity is strong and slow. The nonlinear effects are particularly pronounced when the LMC constitutes a hyperbolic medium in which the longitudinal $\varepsilon_\parallel$ and transverse $\varepsilon_\perp$ components of permittivity tensor have different signs, $\varepsilon_\parallel \varepsilon_\perp < 0$.

An important phenomenon called "topological transition" that manifests itself as the change of topology of the isofrequency surface can appear due to the change of the parameters of anisotropic metamaterial. Parametric oscillations of meta-atoms also can lead to such topological transition. Depending on the oscillations regime, different scenarios may be realized. Namely, hyperbolic isofrequency surface may become elliptic (in stochastic regime) or may oscillate between elliptic and hyperbolic (in nonlinear saturation regime). In the latter case effective dielectric constants become

$$\begin{aligned} \varepsilon_\parallel^{\rm eff} &= \varepsilon_\parallel^{(0)} - (\varepsilon_\parallel^{(0)} - \varepsilon_\perp^{(0)}) \sin^2\theta, \\ \varepsilon_\perp^{\rm eff} &= \varepsilon_\perp^{(0)} + \tfrac{1}{2}(\varepsilon_\parallel^{(0)} - \varepsilon_\perp^{(0)}) \sin^2\theta, \end{aligned} \tag{4}$$

where $\varepsilon_\parallel^{(0)}$ and $\varepsilon_\perp^{(0)}$ are the permittivity components found under the assumption of perfect ordering. Phase-coherent angular motion of the meta-atoms results in oscillation of the anisotropy factor with the frequency $\Omega_{\rm M} \approx 2\Omega_0$, that is slow on the optical time-scale $2\pi/\omega$. For small θ we obtain the time-dependent expressions

$$\begin{aligned} \varepsilon_\parallel^{\rm eff} &\approx \varepsilon_\parallel^{(0)} - 2A_{\rm st}^2(\varepsilon_\parallel^{(0)} - \varepsilon_\perp^{(0)})(1 + \cos\Omega_{\rm M} t), \\ \varepsilon_\perp^{\rm eff} &\approx \varepsilon_\perp^{(0)} + A_{\rm st}^2(\varepsilon_\parallel^{(0)} - \varepsilon_\perp^{(0)})(1 + \cos\Omega_{\rm M} t). \end{aligned} \tag{5}$$

If we have, for example, $\varepsilon_\parallel^{(0)} = -0.5$, $\varepsilon_\perp^{(0)} = 2$, $A_{\rm st} = 0.3$, then approximately half of the time the LMC is a hyperbolic medium and the other half it is elliptic one. When $\varepsilon_\parallel^{\rm eff} > 0$ the medium is transparent, then $\varepsilon_\parallel^{\rm eff}$ becomes negative, and LMC is opaque for the optical wave polarized along the z-axis. In this case the effect of oscillating topology can be realized only in thin (relative to the skin depth) layer of the metamaterial.

Thus, the instability of mechanical oscillations of meta-atoms in LMC in the field of amplitude-modulated electromagnetic wave eventually leads either to regular oscillatory regime that may result in multiple topological transitions or to chaotic regime with the meta-atoms' disordering and effective termalization which also can modify the topology of isofrequency surface.

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