New Twists of 3D Chiral Metamaterials

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Rationally designed artificial materials, called metamaterials, allow for tailoring effective material properties beyond (“meta”) the properties of their bulk ingredient materials. This statement is especially true for chiral metamaterials, as unlocking certain degrees of freedom necessarily requires broken centrosymmetry. While the field of chiral electromagnetic/optical metamaterials has become rather mature, the field of elastic/mechanical metamaterials is just emerging and wide open. This research news reviews recent theoretical and experimental progress concerning 3D chiral mechanical and optical metamaterials, with special emphasis on work performed at KIT.

1. Introduction

A macroscopic crystal such as, e.g., silicon is a highly complex quantum mechanical many-body system composed of about $10^{24}$ protons and electrons. By describing silicon via effective material parameters referring to a simplified fictitious continuum, e.g., via its electric conductivity, its magnetic permeability, or its Young’s modulus, we can reduce this complexity. Thereby, we enable the design of advanced devices and systems such as transistors and computer chips. Likewise, it would generally be hopeless to design optical circuitry or complex mechanical systems while considering the material at the level of atomic unit cells.

Metamaterials can be seen as artificial crystals based on rationally designed unit cells. Each unit cell is composed of a large number of atoms. Unit cell sizes range from the sub-micrometer scale to the centimeter scale. Based on this simple idea, composite materials become possible, the properties of which can go beyond (“meta”) those of the constituent materials—qualitatively and quantitatively. In some cases, the resulting properties are even unprecedented, not found in nature, or were previously deemed impossible. Again, to reduce complexity, it is highly desirable to assign effective parameters to metamaterials, such as an effective conductivity, an effective magnetic permeability, or an effective Young’s modulus.

The idea of metamaterials is not entirely new. For example, in 1920, Lindman realized arrangements of randomly oriented copper helices, leading to 3D chiral metamaterials exhibiting very large isotropic optical activity at microwave frequencies.[1] In 1987, Lakes discussed foams with negative Poisson’s ratio from positive constituents.[2] However, the idea of rationally designed artificial crystals got a tremendous boost when magnetic composites based on split-ring resonators were discussed theoretically in 1999 and realized experimentally in 2000.[3,4] Magnetism at elevated and even optical frequencies then enabled negative refractive indices, which created considerable excitement.[5] Thereafter, the field of metamaterials has strongly benefitted from advances in nanotechnology and 3D additive manufacturing of complex chiral architectures that would be hard or impossible to make by other means.[6,7]

Today, the field of metamaterials has become very broad, encompassing nearly all aspects of solids, including optical, electrical, magnetic, thermal, and mechanical properties—linear and nonlinear as well as static and dynamic.[8–10] Metamaterials,
Unlike metasurfaces, their 2D cousins, are inherently 3D. In this research news, to avoid repetition, we build on extensive previous review articles by us and others on chiral optical metamaterials\cite{7,11,12} as well as on reviews on mechanical metamaterials in general.\cite{13,14} In this research news, we rather focus on selected aspects of recent interest concerning 3D chiral metamaterials and on corresponding challenges researchers at KIT are working at. We start with an introduction pointing at analogies between the effective-medium descriptions of chiral materials in electromagnetism and mechanics. Next, we continue with mechanical metamaterials, for which the number of future challenges is by far larger than the number of past accomplishments. Finally, we discuss the idea of extreme chirality that has recently emerged in electromagnetism but has not yet been addressed in mechanics.

2. Effective-Medium Descriptions

Equations (1) and (2) are descriptions of effective electromagnetic and mechanical (or elastic) material properties, respectively.\cite{15,16} While no continuum description is unique or perfect, Equations (1) and (2) are at a fairly advanced level already. We assume linear response, passivity, reciprocity and include general anisotropies. For the sake of compactness, we use the Einstein summation convention. In Equation (1), the electric rank-two permittivity tensor with components \( \varepsilon_{ij} \) with \( i, j = 1, 2, 3 \) describes the excitation of electric dipoles in the material by the electric component of the electromagnetic field and the thereby changed relation between the components of the dielectric displacement vector field \( \mathbf{D} \) and those of the electric field vector, \( \mathbf{E} \). Likewise, the magnetic permeability tensor with components \( \mu_{ij} \) describes the excitation of magnetic dipoles in the material by the magnetic part of the electromagnetic light and the thereby changed relation between the magnetic induction vector components \( \mathbf{B} \) and those of the magnetic field vector, \( \mathbf{H} \). Here, \( \varepsilon_0 = 8.854 \times 10^{-12} \, \text{A s V}^{-1} \text{m}^{-1} \) and \( \mu_0 = 4\pi \times 10^{-7} \, \text{V s A}^{-1} \text{m}^{-1} \) are the vacuum permittivity and permeability, respectively; \( c_0 = 1/\sqrt{\varepsilon_0 \mu_0} \) is the vacuum speed of light and \( i \) is the imaginary unit. As pointed out above, a sizable magnetic response by metamaterials created considerable excitement some years ago

\[
\begin{align*}
D_i &= \varepsilon_0 \varepsilon_r \varepsilon_i E_i + i\varepsilon_r^{\varepsilon} \zeta_{ij} \vec{\phi}_j + \mathcal{D}_i, \\
B_i &= \varepsilon_r^{\varepsilon} \zeta_{ij} \vec{\phi}_j + \mu_r \mu_i H_i + \mathcal{B}_i
\end{align*}
\]

with \( \zeta_{ij} = -\zeta_{ji} \) due to reciprocity (note the swapping of the indices \( i \) and \( j \)).\cite{15} In the presence of centrosymmetry, all elements \( \zeta_{ij} \) of the chirality tensor are strictly zero. This is easy to see: performing a space inversion, \( \mathbf{r} \rightarrow -\mathbf{r} \), generally turns a right-handed structure into a left-handed structure and vice versa. Furthermore, the electric field is a polar vector and the magnetic field is an axial vector, i.e., the components transform according to \( E_i \rightarrow -E_i \), \( D_i \rightarrow -D_i \), \( B_i \rightarrow B_i \), and \( H_i \rightarrow +H_i \). Any even-rank tensor (not pseudotensor) is invariant with respect to space inversion. If the material is centrosymmetric, its properties must not change when performing a space inversion. Looking at Equation (1), this can only be true if \( \zeta_{ij} = 0 \). In the presence of chirality, which requires the absence of centrosymmetry, this means that electric dipoles can also be excited by the magnetic component of light. Vice versa, magnetic dipoles can be excited by the electric component of light. The assumption of reciprocity directly connects these two “off-diagonal” tensors.\cite{15} This cross-coupling generally leads to biaxiality, the best known special case of which is optical activity. In the case of isotropy, all material tensors turn into scalars.

Let us compare Equation (1) with its mechanical counterpart Equation (2). There, all material tensors are of rank four rather than rank two; they have four indices \( i, j, k, l \) and \( l = 1, 2, 3 \). Intuitively, this aspect is connected to the fact that elastic waves (or phonons) can generally have two orthogonal transverse polarizations and one longitudinal polarization. In electromagnetism, longitudinal waves for zero permittivity are a rare exception. The counterpart of the rank-two electric permittivity tensor is the rank-four elasticity tensor with components \( C_{ijkl} \). It connects the stress tensor and the generalized strain tensor, with components \( \sigma_{ij} \) and \( \varepsilon_{ij} \) respectively. It is quite unfortunate at this point that the symbol \( \varepsilon \) is used for both, the electric permittivity in electromagnetism and for the strain in elasticity. Nevertheless, we stick to this widespread nomenclature

\[
\begin{align*}
\sigma_{ij} &= C_{ijkl} \varepsilon_{kl} + D_{ijkl} \phi_k \\
\varepsilon_{ij} &= B_{ijkl} \sigma_{kl} + A_{ijkl} \phi_k
\end{align*}
\]

with \( D_{ijkl} = B_{ijlk} \) due to reciprocity (note that the first and second pair of indices are interchanged and that, in addition, \( l \) and \( k \) are swapped).\cite{16} The tensor with components \( A_{ijkl} \) connects a torque field with components \( \mathcal{M}_j \) and a rotational field with components \( \phi_{ij} \), both of which form pseudotensors. By comparing Equation (2) with Equation (1), we already see that the electrical degrees of freedom in electromagnetism are analogous to translational degrees of freedom in mechanics. Likewise, the magnetic degrees of freedom are analogous to rotational degrees of freedom.

The tensor components \( B_{ijkl} \) and \( D_{ijkl} \) describe the coupling of translational to rotational degrees of freedom and vice versa. In the presence of centrosymmetry, in close analogy to our above reasoning in electromagnetism for \( \zeta_{ij} = 0 \), we find that \( B_{ijkl} = D_{ijkl} = 0 \). In other words, to obtain a coupling between translational and rotational degrees of freedom, the breaking of centrosymmetry is mandatory.

Equation (2) is well established as Eringen micropolar continuum mechanics. Textbook Cauchy elasticity is only a special case, namely the case of “point mechanics,” for which \( A_{ijkl} = B_{ijkl} = D_{ijkl} = 0 \) holds true. It is sometimes believed that Cauchy elasticity, with 21 independent parameters in the triclinic case, is all there is in continuum mechanics. Likewise, it was long believed that everything in optics can be described by just the electric permittivity, perhaps with minute corrections. Fortunately, both beliefs are incorrect, opening a plethora of opportunities for metamaterials with unprecedented properties in both electromagnetism and mechanics.

We have emphasized the analogies of Equations (1) and (2). However, there are also conceptual differences. The permittivity, permeability, and the chirality parameter in Equation (1) are unavoidably dispersive, i.e., frequency dependent. We have therefore also formulated them in the frequency domain.
In the static case, all components $\xi_{ij}$ are always zero, even if the material is chiral. Equation (2) can be interpreted either in the time domain or in the frequency domain. The components $B_{ijkl}$ can be nonzero for a chiral medium, even in the static case. This aspect is an important difference between optics and mechanics. There are no static chiral effects in optics, whereas static chiral effects have been observed experimentally in mechanics for finite-size samples (see below). However, for samples much larger than the size of one unit cell, the effects of chirality also disappear in static mechanics. This includes the limit of elastic-wave propagation for elastic wavelengths $\lambda \to \infty$.

Clearly, to arrive at a closed set of equations, the above constitutive material equations need to be coupled to equations of motion: the Maxwell equations for the case of electromagnetism and an adequate adaptation of Newton’s law for the case of continuum mechanics.[16,17] In electromagnetism, no further material parameters enter. In contrast, in continuum mechanics, the mass density or the mass density tensor additionally enters via the equation of motion. Together with its rank-four tensors discussed above, continuum mechanics is thus substantially more complex than electromagnetism with its rank-two tensors.[16]

3. Chiral Mechanical Metamaterials

**Static Regime:** While the theory of Eringen chiral micropolar continuum mechanics (and Cosserat mechanics) has been worked out many years ago,[16] detailed blueprints for experiments on corresponding chiral 3D metamaterials have only been published quite recently.[18–23] Figure 1 shows a selection of schemes of corresponding metamaterial unit cells.

For chiral effects to be significant, the sample size or the wavelength of an elastic wave or both need to become comparable to the metamaterial lattice constant. In the static regime, 3D metamaterial samples based on some hundreds of unit cells like the one shown in Figure 1b have shown a large twist effect of about $2^\circ/\%$ (degrees twist angle per axial strain) when pushing onto a metamaterial beam composed of the unit cells as shown in Figure 1b.[19] In addition, the Young’s modulus showed a characteristic dependence on the number of unit cells, whereas it is expected to be independent on the number of unit cells in Cauchy elasticity. Such loss of scale invariance has recently also been found for achiral micropolar metamaterials in 2D and 3D.[24,25] This loss of scalability is intimately connected to the presence of a finite characteristic length scale.[25] For sample sizes $L$ much larger than this scale, the surface-to-volume ratio of the sample decreases according to $1/L$ and the twist effect decreases accordingly $\propto 1/L$ (see above). Therefore, Cauchy behavior with zero twist is asymptotically obtained in the limit $L \to \infty$. Metamaterials with bistable or buckling unit cells also show such scale-dependent behavior. It allows for “programming” the effective properties of the metamaterial over a relatively wide range.[10,26]

Other recent work on artificial chiral architectures in 3D has pointed at the importance of chirality in shearing auxetics (see Figure 1d), in that mechanically rigid and compliant structures can be created by combining elements with different handedness.[20]
As emphasized in the introduction, the mapping of any of these metamaterial microstructures onto effective-medium parameters is a crucial aspect for the metamaterial concept. Some recent work has not performed this mapping at all, but not in a unique manner. Ref. [21] is a notable exception. For the model structure composed of slender beams shown in Figure 1e, the authors have employed the approximation of Euler–Bernoulli beams and have uniquely determined the parameters of the Eringen elasticity tensors in Equation (2). The structure in Figure 1e is cubic, just like the ones in Figure 1a,b. While this structure is ideal concerning its mathematical analysis, it is not really favorable in regard to ease of manufacturing.

Ultimately, one would not only like to map a given metamaterial microstructure onto effective-medium parameters, but one would also like to be able to start from a desired set of effective-medium parameters and systematically construct a corresponding microstructure. This problem is sometimes referred to as the inverse problem. It has not been solved for chiral mechanical metamaterials. To be fair, however, it should be mentioned that the inverse problem has not been solved for chiral optical metamaterials either, despite the fact that this field is much more mature.

A more humble and reachable goal toward the inverse problem is the rational design of metamaterials with large characteristic lengths. The characteristic lengths can be connected to combinations of effective-medium parameters. In 2018, this goal has been achieved for certain achiral micropolar mechanical metamaterials, in that, microstructures were suggested leading to conceptually infinitely large characteristic lengths.[23] For chiral mechanical metamaterials, we are working toward a solution.

The idea underlying our respective work at KIT (see Figure 1g) is to not connect identical chiral unit cells directly, but rather connect them via intermediate achiral cells. If one connects them directly, the displacement vectors at touching interfaces between unit cells point in opposite directions and cancel each other. Therefore, only the unit cell interfaces at the sample surface contribute to the twist, leading to a scaling of the twist effect with the sample’s surface-to-volume ratio, i.e., according to \( \propto 1/L \), with the sample side length \( L \), or according to \( \propto 1/V^{1/3} \) for a cube with volume \( V \). In the opposite limit, if one does not connect the unit cells at all, the individual unit cells do twist around their fixed centers of mass, but these individual microrotations do not translate into a macrorotation of the sample unless they are effectively coupled through the boundary conditions, e.g., by attaching them to a stamp. For an overall chiral response, one therefore has to connect the individual unit cells inside of the metamaterial in some way. A successful example is an alternation of chiral unit cells and achiral intermediate cells. This approach corresponds to building unit cells of multiple meta-atoms just like alloys or diatomic crystals. Such chiral–achiral heterostructures can lead to very large characteristic length scales if the intermediate cells are easily deformable. Preliminary calculations based on the approximation of slender Timoshenko beams for the architecture shown in Figure 1g have shown twist effects as large as about 1°/% for 3D metamaterials containing more than 20 000 of such supercells.[23]

Chiral mechanical metamaterials explore the fundamental limits of accessible material degrees of freedom. In addition, chiral mechanical metamaterials open interesting new perspectives for applications. For example, the mentioned push-to-twist conversion effects can be used together with linear piezoelectric actuators to generate twist motions on the micrometer scale in the quasistatic regime. Further potential applications arise in the dynamic regime.

**Dynamic Regime:** Experiments on 3D chiral mechanical metamaterials in the dynamic regime are elusive to date. Calculations of metamaterial phonon band structures for 3D crystals based on the unit cell shown in Figure 1b have been published by us (see the Supporting Online Material of ref. [19]). Within micropolar continuum mechanics, the modes of a metamaterial beam can be separated into two orthogonal subspaces—the twist and (longitudinal) compression modes on the one hand and the two (transverse) shear modes on the other hand. In the presence of chirality, the twist and compressional modes are coupled. This coupling leads to mixed modes, forming the direct dynamic counterpart of the static push-to-twist conversion effects discussed above. The twist modes can only occur for finite sample size. Furthermore, chirality couples the two otherwise independent and orthogonal transverse shear modes toward two effective chiral modes. For these modes, the displacement vector at a fixed sample position circles around its rest position in perfect analogy to the behavior of the electric field vector for circular polarization of light. Interestingly, chiral phonons have only very recently been observed in 2D natural materials.[27] In optics, such eigen states lead to the well-known phenomenon of optical activity, which allows converting an incident transverse linear polarization into the orthogonal transverse linear polarization after some propagation distance. The corresponding phenomenon of “mechanical activity” has not been observed so far, neither have chiral phonons in 3D (meta)materials been generated directly. We work toward both at KIT. Furthermore, mechanics offers opportunities beyond optics because longitudinal waves are generally allowed in elasticity too. Therefore, chiral modes composed of mixed transverse and longitudinal character, “trochoidal phonons”, seem possible.

For all of these wave propagation effects, it is highly desirable to work with constituent materials exhibiting small elastic losses at high frequencies. It is presently not quite clear, inasmuch this goal is accessible with polymer-based metamaterial samples, which can directly be manufactured by 3D laser printing. However, recent work on the pyrolysis of polymer structures has shown promising results.[14,28] Both, the associated shrinkage by about a factor of five along all three spatial directions, as well as the stiffening of the constituent material by about an order of magnitude toward Young’s moduli in the range of \( E = 20 \text{ – } 30 \text{ GPa} \), are favorable for high-frequency operation. In addition, the higher material strength at smaller length scales reaching 2–3 GPa in the sub-micrometer regime compared to 200 MPa for macroscopic dimensions would also be beneficial to the structural engineering applications of chiral metamaterials.[29]
Chirality necessarily requires broken space-inversion symmetry. If, in addition, time-inversion symmetry is broken, additional degrees of freedom arise. Mathematically, the tensors B and D in mechanics (see Equation (2)), and the tensors $\xi$ and $\zeta$ in electromagnetism (see Equation (1)) are not fully determined by each other anymore. Breaking reciprocity can be achieved by time symmetry breaking elements like external magnetic fields. The Faraday effect in electromagnetism is one well known consequence. In mechanics, spinning elements can play the role of magnetic moments or static magnetic fields in electromagnetism. \cite{30,31}

While inspiring model experiments have been performed in two dimensions, \cite{31} 3D chiral mechanical metamaterials of that sort are elusive to date.

4. Chiral Optical Metamaterials

In recent years, the activities at KIT in the field of chiral optical metamaterials have focused on the question of how to design motifs from which metamaterials can be made whose chiral response is as large as fundamentally possible. \cite{6,32} In this endeavor, one is led to the question: “How chiral is a given chiral object?” It turns out that providing an answer is not trivial.

Geometrical chirality is well defined. If an object lacks centro-symmetry, all mirror symmetries, and all rotation–reflection symmetries, it is chiral (see example in Figure 2a). Otherwise, it is achiral. However, this binary criterion does not allow a graded quantification of chirality, which is important to sorting and comparing objects according to it as well as to having a clear target toward which one can optimize metamaterials.

Over the past decades, it has become clear that there is no consistent measure of geometric chirality. \cite{33} The first step toward a solution is to change the point of view from geometry to interaction. The electromagnetic chirality (em-chirality) of an object is a measure of how differently general fields of opposite polarization handedness do interact with it. \cite{14} General fields of pure polarization handedness (helicity) are the sum of an arbitrary number of propagating and/or evanescent plane waves that are all either left- or right-handed circularly polarized.

Their natural representation are the Riemann–Silberstein combinations \cite{35}

$$\sqrt{2} G_{\alpha} = E \pm i Z H$$

where $Z = \frac{\mu_0}{\varepsilon_0}$ is the impedance of the medium surrounding (index “S”) the system of interest, characterized by permittivity $\varepsilon_s$ and permeability $\mu_s$. For a field with pure helicity of $+1(-1)$, $G_+ (G_-)$ is zero at all space-time points.

The quantification of em-chirality is accomplished using the interaction operator of the object, a.k.a. the T-matrix. Such operator contains all information about the interaction of the object with the electromagnetic field. First, the operator is decomposed into two parts that correspond to the two helicities of the incident fields. Next, singular-value decomposition is used to compute a distance between the two suboperators. This distance is the em-chirality of the object. \cite{34} A crucial property of em-chirality is that it is upper-bounded, contrary to the traditional geometrical definition of chirality. The maximum value that em-chirality can achieve is equal to the total interaction cross section of the object. One can therefore speak about maximally emchiral objects. For reciprocal objects, reaching the bound is equivalent to being transparent to all the fields of one helicity.

At KIT, the motivation to focus on extreme em-chirality partly comes from its potential applications. For bulk devices, helicity filtering and angle independent glasses for use in stereoscopic projection systems would become possible. \cite{14} At smaller scales, helicity dependent photon processing in optical chips, and the enhancement of the chiral response from molecules would become possible. Hence, a key question is: How can we design actual systems that approach maximal em-chirality? The first step is to apply the transparency requirement to obtain restrictions in the typical models that describe light–matter interaction, which will then guide the design. For small particles in the dipolar approximation, whose interaction is described by a polarizability tensor

$$\begin{pmatrix} d \\ m \end{pmatrix} \begin{pmatrix} \alpha_{dE} & \alpha_{dH} \\ \alpha_{mE} & \alpha_{mH} \end{pmatrix} \begin{pmatrix} E \\ H \end{pmatrix}$$

the transparency condition imposes

$$\alpha_{dE} = \alpha_{dH} = E_s, \alpha_{mH} = \mp i \frac{\alpha_{mE}}{Z_s}, \alpha_{mE} = \mp i \mu_s \frac{\alpha_{mH}}{Z_s}$$

where we have also assumed that the particle is reciprocal. Furthermore, with the reciprocity assumption, transparency at the level of the constitutive relations imposes

$$Ze_0 E_0 = \mu_0 \mu_s |Z| = \mp Z_s$$

It is clear that a 3D metamaterial made of small maximally emchiral inclusions would feature effective constitutive relations meeting these restrictions.

A truly maximal emchiral inclusion is invisible to all fields of a given helicity at all frequencies. We can also consider the less challenging case of designing an object that achieves the transparency condition at a single frequency of light, or in a narrow frequency band. An example for such a structure is a circular helix with particular geometrical parameters made of
an ideal conductor. It is shown in Figure 2b.[34] Efforts are currently put in place at KIT to identify objects with comparable properties at much shorter wavelengths, which constitutes a bigger challenge.

There is one notable consequence of the addition of reciprocity to the requirement of transparency. It forces the system to preserve the helicity of light, or, in other words, to not couple the G and G fields upon interaction. This requirement defines a system with electromagnetic duality symmetry.[36] The duality condition in the dipolar approximation reads

$$\alpha_{\text{el}} = \varepsilon \alpha_{\text{mH}} \quad \text{and} \quad \alpha_{\text{mE}} = -\alpha_{\text{el}} / \mu.$$

and, at the level of the constitutive equations

$$Z \varepsilon_0 \varepsilon_\parallel \mu_0 / Z \varepsilon_\parallel = \xi_\parallel \xi_0,$$

Along our path to work toward maximal emchiral 3D metamaterials, we also investigate scatterers that approach duality and study metamaterials thereof. This is an important research endeavor. Not only is duality a prerequisite to reach maximal emchiral materials, but, additionally, dual systems are a key ingredient for technologically relevant effects like backscattering suppression and artificial optical activity, and a requisite for transformation media. Duality is also one of the ingredients in some systems featuring topologically protected photonic states.[37]

Frequently, dielectric scatterers can only be made dual in dipolar approximation. This happens for spheres made from high-permittivity materials at a specific wavelength for a given radius.[38] However, if such dipolar spheres are closely packed together to form a metamaterial, higher-order multipole moments gain importance in the interaction. The nonvanishing excitation of higher-order multipole moments causes a metamaterial consisting of such spheres to be no longer dual. By using optimized dielectric core–shell particles, the situation can be improved. Higher-order multipole moments can also be driven into the duality regime.[39] Similarly, initial results indicate that core–shell spheres made of materials with intrinsic chirality can be optimized to achieve large values of em-chirality (see Figure 2c). At KIT, we investigate the possibility of providing the necessary intrinsic chirality by using chiral carbon nanotubes. These are promising steps toward designing 3D metamaterials of very high duality and very large electromagnetic chirality.

5. Conclusion

In conclusion, while the idea of 3D chiral metamaterials is at least one century old, it still poses a wealth of interesting scientific questions—both in optics and mechanics. Due to a close mathematical analogy on the level of effective-medium descriptions, both fields can benefit from each other. Broadly speaking, a long-standing dream of materials science is to rationally design materials, to avoid tedious trial-and-error experimentation. However, this goal has been realized in only a few exceptions to date. Metamaterials are an entire class of such exceptions.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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