Bioinspired, mechanical, deterministic fractal model for hierarchical suture joints

The MIT Faculty has made this article openly available. Please share how this access benefits you. Your story matters.

<table>
<thead>
<tr>
<th>Citation</th>
<th>Li, Yaning, Christine Ortiz, and Mary C. Boyce. “Bioinspired, Mechanical, Deterministic Fractal Model for Hierarchical Suture Joints.” Physical Review E 85.3 (2012). ©2012 American Physical Society</th>
</tr>
</thead>
<tbody>
<tr>
<td>As Published</td>
<td><a href="http://dx.doi.org/10.1103/PhysRevE.85.031901">http://dx.doi.org/10.1103/PhysRevE.85.031901</a></td>
</tr>
<tr>
<td>Publisher</td>
<td>American Physical Society</td>
</tr>
<tr>
<td>Version</td>
<td>Final published version</td>
</tr>
<tr>
<td>Citable Link</td>
<td><a href="http://hdl.handle.net/1721.1/71526">http://hdl.handle.net/1721.1/71526</a></td>
</tr>
<tr>
<td>Terms of Use</td>
<td>Article is made available in accordance with the publisher's policy and may be subject to US copyright law. Please refer to the publisher's site for terms of use.</td>
</tr>
<tr>
<td>Detailed Terms</td>
<td></td>
</tr>
</tbody>
</table>
Many biological systems possess hierarchical and fractal-like interfaces and joint structures that bear and transmit loads, absorb energy, and accommodate growth, respiration, and/or locomotion. In this paper, an elastic deterministic fractal composite mechanical model was formulated to quantitatively investigate the role of structural hierarchy on the stiffness, strength, and failure of suture joints. From this model, it was revealed that the number of hierarchies (N) can be used to tailor and to amplify mechanical properties nonlinearly and with high sensitivity over a wide range of values (orders of magnitude) for a given volume and weight. Additionally, increasing hierarchy was found to result in mechanical interlocking of higher-order teeth, which creates additional load resistance capability, thereby preventing catastrophic failure in major teeth and providing flaw tolerance. Hence, this paper shows that the diversity of hierarchical and fractal-like interfaces and joints found in nature have definitive functional consequences and is an effective geometric-structural strategy to achieve different properties with limited material options in nature when other structural geometries and parameters are biologically challenging or inaccessible. This paper also indicates the use of hierarchy as a design strategy to increase design space and provides predictive capabilities to guide the mechanical design of synthetic flaw-tolerant bioinspired interfaces and joints.

DOI: 10.1103/PhysRevE.85.031901 PACS number(s): 87.10.Pq, 87.10.Kn, 68.35.Ct, 62.20.mm
suture joints is evaluated; in Sec. VI, the failure mechanisms, strength, and fracture toughness of hierarchical suture joints are studied. Finally, the benefits and potential applications of the hierarchical design are discussed, providing guidelines for the development of potential bioinspired synthetic systems.

II. GEOMETRY OF A BIOINSPIRED DETERMINISTIC FRACTAL MODEL FOR HIERARCHICAL SUTURE JOINTS

While the fractal model developed here is generic and can be applied broadly to hierarchical suture joints, it is particularly relevant to the ammonite shell septal suture, which forms the junction between the septa of buoyancy chambers and the external wall of the phragmocone [22–25]. Ammonites, which are extinct marine invertebrates in the subclass Ammonoidea of the class Cephalopoda, have hierarchical sutures, which have long been recognized as one of the most complex and rapidly evolving structures in the fossil record, showing consistently increased suture complexity during evolution [25]. Representative hierarchical suture seams, best observed by polishing the outer layers of the shell, are shown in Fig. 1(a).

In this paper, the hierarchical suture seam is modeled in a deterministic manner by iteratively superimposing a self-similar wave on each wave form of the former profile as shown in Fig. 1(b). By defining the center line of a straight interface layer as the zeroth-order suture line \((L_0)\), an \(N\)th-order suture seam can be generated via successive superposition of \(N\) wave forms. Thus, \(N\) is also the total number of hierarchies or order of the suture joint. Here, we focus on suture seams characterized by triangular-wave forms, given their presence in many natural systems [32]. Schematics of typical RVEs of hierarchical suture joints of different order \(N\) are shown in Fig. 1(c).

To illustrate the geometric features of the RVEs of the hierarchical suture joints, the hierarchy map of the RVE of a third-order suture joint is shown in Fig. 2(a) as an example and then is generalized to that of an \(N\)th-order joint in Fig. 2(b). The nomenclature for a general \(N\)th-order suture takes hierarchy level \(m = 1\) to correspond to the longest wavelength \(\lambda_1\) and largest amplitude \(A_1\) of the hierarchy [corresponding to \(L_1\) of Fig. 1(b)] with each successive level possessing a wavelength and an amplitude that decreases in a self-similar manner (as mathematically specified later). Figure 2(a) shows that the effective interface layer of the third-order RVE at hierarchical level \([m = 1]\) is a second-order suture joint at a smaller scale, and the effective interface layer of this second suture joint is a first-order suture joint. Generalizing, the hierarchy map of an \(N\)th-order suture joint is abstracted in Fig. 2(b). It can be seen that the geometry of an \(N\)th-order suture joint is determined by the total number of hierarchies \(N\), the wave amplitudes \(A_m\) \((m = 1 \ldots N)\), wavelengths \(\lambda_m\), and the width of the effective interface layer\(^1\) \(g_m\) at each level of hierarchy \(m\).

\(^1\)Note \(g_m\) is taken along the propagation direction of the wave line at the \(m\)th hierarchy. \(g_m\) equals the thickness of the effective interface layer at the \(m\)th hierarchy divided by \(\cos \theta\).
Due to our imposition of self-similarity, \( i_m \) and \( \alpha_m \) are constants. By taking \( i_m = i \) and \( \alpha_m = \alpha \), three independent nondimensional parameters \( i \), \( \alpha \), and \( N \) determine the geometry of the suture seam. The suture complexity index is derived from Eq. (1) as \( I_N = i^N \). For a triangular-wave suture seam, \( i \) is related to the tip angle of the triangular tooth \( \theta \) via \( i = 1/\sin \theta \). From Eqs. (1) and (2), using the fractal box-counting method [28] and taking the wavelength of an \( m \)th level suture line as a unit measure \( e_m \), \( e_m = \lambda_m = \lambda_i a^{m-1} \), the fractal dimension of a suture seam is derived as

\[
D = \lim_{m \to \infty} \frac{\log(J_m)}{\log(e_m)} = 1 - \frac{\log(i)}{\log(\alpha)} = 1 + \frac{\log(\sin \theta)}{\log(\alpha)}, \tag{3}
\]

where \( J_m \) is the number of \( e_m \) units over the entire length of the suture line \( J_m = \frac{\text{length of suture}}{e_m} \), where \( L_m = L_0 i^m \).

The log-log plot of \( e_m \) vs \( J_m \), Fig. 3(a) (left), shows that the self-similar triangular hierarchical suture seam has a constant fractal dimension \( D \) independent of \( N \) and, therefore, is a deterministic fractal. \( D \) is a function of \( \theta \) and \( \alpha \) with a value between 1 and 2. \( D \) is larger when \( \theta \) is smaller or \( \alpha \) is larger as shown in Eq. (3) and its corresponding plot, Fig. 3(a) (right). A schematic of the box-counting method is shown in Fig. 3(b).

The values of \( D \) of a large variety of ammonite suture seams have been shown to lie in the range of \( \sim 1.2-1.8 \) [33]. For some physical systems, a deterministic fractal model has been shown to be a good approximation for the random fractal system with the same fractal dimension [34].

Due to similarity, the wave number \( n_w \) for each straight segment is a constant at each hierarchy, related to \( \theta \) and \( \alpha \) by \( n_w = (2\alpha \sin \theta)^{-1} \). By using Eq. (3), we obtain \( n_w = 0.5\alpha^{-D} = 0.5(\sin \theta)^{-D/(D-1)} \). Examples of hierarchical suture seams with the same \( n_w \) = 2.5 and different \( \theta \) and, therefore, different \( D \) are shown in Fig. 3(c).

In order to quantify the effective mechanical properties of hierarchical suture joints, the volume fraction of the components in the composite interface region must be taken into account. When the total volume fraction of the interface is the same for suture joints with different orders \( N \), the length of the interface increases, hence, the width of the interface layer, at each level of the hierarchy \( g_m \), decreases and follows a power law relationship: \( g_m = g_i i^{m-1} \) [Fig. 2(b)].

As shown in Fig. 4(a), fundamental building blocks (i.e., the repeating geometry unit) of a self-similar hierarchical suture joint are RVEs of a first-order suture joint. These building blocks are self-similar and are assembled through \( N \) hierarchies to construct an \( N \)th-order hierarchical suture joint. By defining the tooth volume fraction of a single building block as \( f \) (\( N = 1 \)), the total tooth volume fraction of an \( N \)th-order hierarchical suture joint \( f_N \) (including minor teeth at all levels)
(c) examples of self-similar suture seams with the same wave number at each level of hierarchy

Thus, (a) hierarchical suture seam; (b) schematics of the box-counting method; (c) parametric space of the geometric constraint (the maximum tip angle $\theta^*$ changing with $f$) of this model (region 1: no hierarchical suture joint exists, region 2: geometric constraint, and region 3: no constraint).

### III. THE RIGID TOOTH COMPOSITE MODEL

Tensile loading within the suture plane and normal to the suture axis is a prevalent physiologic loading condition $[13,18,35]$ [Fig. 1(b)]. In this section, we determine the tensile stiffness of the self-similar hierarchical suture joint model for the case of hierarchical sutures where the teeth are assumed to be rigid and only the interfacial material is deformable, which provides an upper bound for the suture stiffness. Under this tensile load, tensile stress is transmitted from the major teeth (i.e., the largest teeth or the first-order teeth) to minor teeth (i.e., smaller teeth in all other levels of hierarchy or the higher-order teeth) and interface through tension and shear. Therefore, in this section, first, the tensile and shear stiffness of the first-order suture joint is determined; and then, the load transmission mechanism is analyzed; finally, the tensile stiffness of an $N$th-order hierarchical triangular suture joint model, (a) schematics of the configuration of a self-similar $N$th-order hierarchical triangular suture joint model, (b) the total volume fraction $f_v$ of self-similar hierarchical suture joints as a function of the volume fraction of a building unit $f$ and the number of hierarchies $N$, and (c) parametric space of the geometric constraint (the maximum tip angle $2\theta^*$ changing with $f$) of this model (region 1: no hierarchical suture joint exists, region 2: geometric constraint, and region 3: no constraint).

#### A. First-order suture joint model under tension and shear

**Tension.** For a first-order suture joint under tension perpendicular to the suture axis [Fig. 1(b)], the tension $\tau_T$ is balanced by interfacial shear stress $\tau_T$ and normal stress $\sigma_T$ as shown in the free body diagram of Fig. 5(a).
FORCE EQUILIBRIUM, IN THE X AND Y DIRECTIONS, ON THE FREE BODY DIAGRAMS OF A TOOTH [FIG. 5(a)] YIELDS [32]

$$\sigma_T = \tau_T \tan \theta = \sigma \sin^2 \theta, \quad \tau_T = \sigma \sin \theta \cos \theta. \quad (6)$$

The strain energy $U_{\text{RTM}}^T$ of a RVE of the system due to the applied tension can be expressed as

$$U_{\text{RTM}}^T = \frac{f^2}{2} \left( \frac{1}{E_{\text{RTM}}^T} V + \frac{1}{2} \left( \frac{\sigma_T^2}{E_0^T} + \frac{\tau_T^2}{G_0} \right) V_0 \right). \quad (7)$$

where $E_{\text{RTM}}^T$ is the effective tensile modulus of the first-order suture joint; $E_0^T$ and $G_0$ are the plane strain modulus$^2$ and shear modulus of the interface material, respectively; $V$ is the total volume of the RVE; $V_0$ is the volume of the interface material in the RVE, and $V_0 = \frac{1}{f}$ (for first-order suture joints, $f = f_0$). By substituting Eq. (6) into Eq. (7), the effective tensile stiffness of the first-order suture joint is obtained

$$E_{\text{RTM}}^T = \frac{f^2}{1 - f} \left( \frac{1}{G_0} \sin^2 \theta \cos^2 \theta + \frac{1}{2} \frac{\sigma_T^2}{E_0^T} \sin^4 \theta \right)^{-1}. \quad (8)$$

From Eq. (8), we can see that, when $2\theta < 90^\circ$, $G_0$ dominates, whereas, for $2\theta > 90^\circ$, $E_0^T \tan^2 \theta$ dominates.

Shear. When a first-order RTM suture joint is subjected to simple shear, the applied shear stress at the tooth edges $\tau$ is balanced by a uniform interfacial shear stress $\tau_S$ and tensile and compressive normal stress $\sigma_S$ as shown in Fig. 5(b). The teeth on the top and bottom rows experience a relatively rigid body displacement $\delta$ as shown in Fig. 5(b). For small elastic deformation, equilibrium together with the kinematic analysis of the interfacial segments yield,$^3$

$$\sigma_S = \pm E_0^T \varepsilon_S = \pm E_0^T \frac{\delta}{g}, \quad \tau_S = G_0 \gamma_S = G_0 \frac{\delta}{g} \tan \theta, \quad (9)$$

where $\varepsilon_S$ is the normal strain of the interface, $\gamma_S$ is the shear strain of the interface, and $g$ is the width of the interface as shown in Fig. 5(b). The total strain energy $U_{\text{RTM}}^S$ of an RVE due to the applied shear can be expressed as

$$U_{\text{RTM}}^S = \frac{1}{2} G_{\text{RTM}} \left( \frac{\delta}{A} \right)^2 V = \frac{1}{2} \left( \frac{\sigma_S^2}{E_0^T} + \frac{\tau_S^2}{G_0} \right) V_0, \quad (10)$$

where $G_{\text{RTM}}$ is the effective shear modulus of the first-order suture joint and $A$ is the amplitude of the interface. By substituting Eq. (9) into Eq. (10), the effective shear modulus is derived as

$$G_{\text{RTM}} = \frac{f^2}{1 - f} \left( \frac{E_0^T \sin^2 \theta \cos^2 \theta + \frac{1}{2} \frac{G_0}{E_0^T} \sin^4 \theta}{\tan^2 \theta} + G_0 \right). \quad (11)$$

Equation (11) shows that the effective shear modulus $G_{\text{RTM}}$ of the first-order suture joint increases when the tip angle $\theta$ decreases. Also, as expected, larger tooth volume fraction $f$ and larger stiffness of interface material correspond to larger $G_{\text{RTM}}$.

**B. Tensile stiffness of the hierarchical RTM**

The load transmission mechanism between two successive hierarchies is shown in Fig. 6. In a top-down view of the hierarchy, the tensile load applied at the top hierarchy level ($m = 1$) is transmitted to the interface layer down through all hierarchies. The load transmitted to the teeth of all other hierarchies ($m > 1$) is the superposition of tension and shear. Following the construction process of a hierarchical suture joint, the effective interface of the $N$-th order hierarchical suture joint is an $(N - 1)$-th order hierarchical suture joint as shown in Fig. 2(b).

Based on this load transmission mechanism and Eqs. (8) and (11), the effective tensile modulus and shear modulus of the second-order hierarchical suture joint can be determined by the corresponding properties of the first-order hierarchical suture joint,

$$E_{\text{RTM}} = \frac{f^2}{1 - f} \left( \frac{1}{G_{\text{RTM}}} \cos^2 \theta \sin^2 \theta + \frac{1}{2} \frac{G_0}{E_0^T} \sin^4 \theta \right)^{-1},$$

$$G_{\text{RTM}} = \frac{f^2}{1 - f} \left( \frac{E_{\text{RTM}}^T}{\tan^2 \theta} + G_{\text{RTM}} \right). \quad (12a)$$

where $E_{\text{RTM}}$ and $G_{\text{RTM}}$ can be calculated from Eqs. (8) and (11). The effective tensile modulus and shear modulus of an $N$th-order hierarchical suture joint can be determined by the
corresponding properties of the \((N-1)\)th-order hierarchical suture joint following explicit iterative process whereby properties of the \(N\)th-order joint are determined using properties of the \((N-1)\)th-order joint as shown in Eqs. (12b),

\[
E_N = \frac{f^2}{1-f} \left( \frac{1}{G_{N-1}} \sin^2 \theta \cos^2 \theta + \frac{1}{G_{N-1}} \sin^4 \theta \right)^{-1},
\]

\[
G_N = \frac{f^2}{1-f} \left( \frac{E_N - G_{N-1}}{\tan^2 \theta + G_{N-1}} \right).
\]  

Using the iterative Eqs. (12), the tensile stiffness and shear modulus of the RTM hierarchical suture joints with an arbitrary order \(N\) (>1) is obtained. Interestingly, when \(f > 0.618\), which corresponds to the golden ratio \(\frac{f^2}{1-f} > 1\), then \(E_N > G_{N-1}\) is guaranteed. The tensile behavior of hierarchical hard biological tissues with arrays of bonded rigid rectangular components were modeled with a similar iterative relation [4] where load transfer and deformation were more simply due to interface shear [36].

To verify this load transmission mechanism and Eqs. (12a), finite element (FE) models of a first-order and a second-order self-similar suture joint model are constructed with \(f = 0.667\), and \(2\theta = 60^\circ\) as shown in Fig. 7. It can be seen that Eqs. (12a) are very accurate, and remarkably, the addition of one hierarchy in the structure increases both the tensile and the shear modulus by approximately 1 order of magnitude. Also, the piecewise spatially nonuniform strain (including regions of both compression and tension) in the hierarchical interfaces significantly indicates a potential benefit of local noncatastrophic failure (an energy dissipation mechanism) for the hierarchical suture geometry where higher-order teeth can act sacrificially protecting lower-order teeth. The stress in the major teeth and interfacial material will be uniform due to the triangular geometry [32], indicating the maximum strength.

The iteration system of Eqs. (12a) and (12b) is plotted in Fig. 8, showing how the hierarchy order influences the stiffness of RTMs constructed by building blocks with different geometries. Figures 8(a) and 8(b) show that the nondimensional stiffnesses for all combinations of geometric parameters are exponentially dependent on the hierarchy number \(N\). The exponent \(k(f, \theta)\) is plotted in Fig. 8(c) and is a function of the two independent geometric parameters of the building block \(f\) and \(\theta\),

\[
k(f, \theta) = \frac{d \log E_N}{d N} = \frac{d \log f^2}{d N} = \frac{d \log G_N}{d N}.
\]

Therefore, the effective stiffness of hierarchical RTMs can be expressed as

\[
E_N^\text{RTM} = E_0 \times 10^{N k(f, \theta)}.
\]

Equation (14) shows that, for the same building block, when the number of hierarchy \(N\) increases, the effective stiffness increases exponentially. Because the influence of \(N\) is amplified by \(k(f, \theta)\) and \(k\) only is determined by the geometry of the building block, we refer to \(k\) as a geometry amplification factor.

FIG. 6. (Color online) Load transmission mechanisms of the RTM between two successive hierarchies.

FIG. 7. (Color online) FE verification of Eqs. (12a) for first- and second-order RTM suture joints, under the loading cases of (a) shear and (b) tension. \(\tilde{E}\) and \(\tilde{G}\) refer to FE results (symbols), and the effective shear and \(E_N^\text{RTM}\) and \(G_N^\text{RTM}\) calculated from Eqs. (12a), respectively, refer to the analytical results (solid and dashed lines).

FIG. 8. (Color online) Load transmission mechanisms of the RTM between two successive hierarchies.
k(f, θ) is obtained numerically using Eqs. (12a) and (12b) and is plotted in Fig. 8(c). Figures 8(a) and 8(c) show that k increases rapidly when θ decreases. For RTMs with relatively large tip angles (θ > 90°), θ has much less influence on k than those with relatively small tip angles (θ < 90°). Also, for all tip angles, k increases when f increases.

IV. THE DEFORMABLE TOOTH COMPOSITE MODEL

A. First-order suture joint model under tension and shear

The RTM captures the main effects of increasing levels of hierarchy on enhancing the stiffness and strength of suture joints. However, when the tooth tip angle is small, deformation of the tooth must be taken into account as shown in this section. Similar to the RTM, the strategy to develop the mechanical hierarchical deformable tooth model (DTM) is to derive relationships for the shear and tension of a first-order DTM first and then to develop models for the higher-order sutures.

For a first-order DTM under tension, the total strain energy of its RVE can be written as

$$U_{T}^{DTM} = \frac{1}{2} \frac{\sigma^2}{E_{T}^{DTM}} V = \frac{1}{2} \left( \frac{\sigma^2}{E_0} + \frac{\tau^2}{G_0} \right) V_0 + \frac{1}{2} \frac{\sigma^2}{E_1} V_1, \quad (15)$$

where V_0 is the volume of the skeleton teeth V_1 = V − V_0. Compared to Eqs. (8) and (15) now includes tooth strain energy.

Similarly, by substituting Eq. (6) into Eq. (15), the effective tensile stiffness of a first-order DTM is derived as

$$E_{T}^{DTM} = \frac{f^2 E_1}{(1 - f) \left( \frac{E_T}{E_0} \sin^2 \theta \cos^2 \theta + \frac{E_T}{E_0} \sin^4 \theta \right) + f}. \quad (16)$$

This same relationship was derived in Ref. [32] using a kinematic analysis.

The shear behavior of the first-order DTM is more complicated than that of the first-order RTM due to the influence of tooth deformation. FE models of first-order suture joints with a wide range of tip angles (θ = 2.9°, 5.7°, 11.3°, 21.8°, 38.7°, and 58.0°) and stiffness ratios (R_S ≈ E_T/E_0 = 10, 100, 1000) were constructed and were subjected to simple shear loading to gain insight into the deformation mechanisms and then to compare to analytical models. These results are then compared with the results of the RTM in Fig. 9(a).

Figure 9 demonstrates that the inclusion of tooth deformability leads to a fundamentally different mechanical behavior relative to the rigid tooth system with R_S and 2θ. In contrast to the RTM in which the shear modulus goes to infinity when the tip angle decreases, the shear modulus of the DTM achieves a maximum at a tip angle 2θ* that is dependent on R_S and 2θ [Fig. 9(a)]. The tip angle 2θ* corresponds to competition of two dominant deformation mechanisms: a moderate tooth coupled bending and shear occurring simultaneously with deformation of the compliant interface material in shear and tension or compression [Fig. 5(b)]. Generally, when R_S and 2θ decrease, tooth deformation becomes more dominant, whereas, when R_S and 2θ increase, the deformation of interfacial layers becomes more dominant, and the teeth tend to behave more like rigid bodies.

This trend is shown clearly by the contours of the maximum principal strain of various deformed FE suture joint models under the same overall shear strain as shown in Fig. 9(b). For example, when R_S = 1000 and 2θ = 43.6°, teeth behave rigidly, and interface layers experience a quasistatic stress state; when R_S = 10 and 2θ = 5.8°, shear of teeth is the dominant deformation, and the inverse rule of mixtures is an accurate evaluation for mechanical properties; when R_S = 1000 and 2θ = 5.8°, bending is the dominant deformation of teeth; for all other intermediate cases, the deformation of teeth is a combination of shear and bending. For all cases, the maximum principal strains are located in the middle of the interface layers, which could be a precursor to interface failure.

Extending the derivation of the RTM, the effect of deformable teeth shown above can be captured quantitatively by an analytical model developed as follows. Two variables due to coupling between shear and bending are introduced in Eq. (9): the average deflection δ_b and the rotation γ_b of deformable teeth. Thus, comparison of Fig. 9(a) (top) and Fig. 5(b) (left) yields

$$\tau_x = G_0 \left( \frac{\delta - \delta_b}{g} \tan \theta - \gamma_b \right), \quad \sigma_x = E_0 \frac{\delta - \delta_b}{g}. \quad (17)$$
To evaluate $\delta_b$ and $\gamma_b$, the RVE of a first-order DTM is modeled as a cantilevered beam. An effective shear load is applied at the end of this cantilevered beam as shown in Fig. 9(a) (top). The equations governing the deformation of the beam under the coupled shear and bending are

$$G_s A_s v'_b = E_1 I v''_b, \quad E_1 I v''_b = P,$$

where $E_1$ is the Young’s modulus of the tooth material; $I$ is the moment of inertia of the cantilever beam such that $I \approx \frac{1}{3} \ell^2$; $G_s$ is the local shear modulus, which is calculated by the Voigt law $G_s = G_0(1 - f) + G_1 f$; $A_s$ is the effective shear area of the cross section where, for a rectangular cross section, $A_s = \frac{1}{2} (\lambda - 2g)$; $P$ is the effective concentrated force at the end of the beam due to a shear stress $\tau$; where $P = \tau (\lambda - 2g)$. The general solution of Eq. (18) is

$$v(x) = v_s(x) + v_b(x),$$

where $v_s$ is the deflection due to shear and $v_b$ is the deflection due to bending. The boundary conditions imply that $v(0) = 0$, $v'_b(0) = 0$, $v''_s(A) = 0$, and $v'_b(0) = -\frac{P}{E_1 I}$. Therefore, the total deflection can be solved as

$$v(x) = \frac{P x^3}{6 E_1 I} + \frac{P A x^2}{2 E_1 I} + \frac{P x}{G_s A_s}.$$

The average influence of tooth deformation on the kinematics of the interface is evaluated in the middle of the RVE as

$$\delta_b = v \left( \frac{A}{2} \right) = \frac{5 P A^3}{48 E_1 I} + \frac{P A}{2 G_s A_s},$$

$$\gamma_b = v' \left( \frac{A}{2} \right) = \frac{3 P A^2}{8 E_1 I} + \frac{P}{G_s A_s}.$$

Thus, the effective shear modulus of a first-order DTM is derived as

$$\bar{G}_s^{\text{DTM}} = f \frac{\tau A}{\delta} = \frac{f^2 C E_0 E_1}{f C C^b E_0 + (1 - f)(1 + C') E_1},$$

where

$$C = \frac{G_0}{E_0} + \frac{E_0^{PS}}{E_0} (\tan \theta)^{-2},$$

$$C^b = \frac{3 E_1}{4 G_s} + \frac{5}{16} (\tan \theta)^{-2},$$

and

$$C' = \frac{3 G_0}{2 G_s} + \frac{9}{8} \frac{G_0}{E_1} (\tan \theta)^{-2}.$$

When $\frac{E_1}{E_0} \rightarrow \infty$, Eq. (23a) degenerates into Eq. (11) as a connection between DTM and RTM solutions. The parameters $C$, $C^b$, and $C'$ in Eq. (23a) are nondimensional and depend on $\theta$ and the tension and/or shear stiffness ratios of the two phases, as shown in Eqs. (23b)–(23d).

**B. Tensile stiffness of the hierarchical DTM**

The tensile stiffness of a hierarchically suture joint with deformable teeth can be calculated using a similar iterative approach as that presented earlier for the hierarchical DTM. Therefore, for a second-order DTM,

$$\bar{E}_2^{\text{DTM}} = \frac{f^2 E_1}{(1 - f)[\frac{E_0}{E_1} \sin^2 \theta \cos^2 \theta + \frac{E_0}{E_1} \sin^4 \theta] + f},$$

$$\bar{G}_2^{\text{DTM}} = \frac{f^2 C_2 E_1^{\text{DTM}}}{f C_2 E_1^{\text{DTM}} + (1 - f)(1 + C'_2) E_1},$$

where $C_2$ and $C'_2$ are the coupling effects among the stiffness ratios and the tooth angles (lower) and the free body diagram of DTM accounting for the coupling effects among shear, bending of a tooth, and the rigid tooth movement (upper) and (b) FE results of the maximum principal strain distribution as a function of the tip angle and the stiffness ratio (in order to show the deformation clearly, a magnification factor of 5 is used).
where $\tilde{E}_{\text{DTM}}$ and $\tilde{G}_{\text{DTM}}$ are the effective interfacial properties of the second-order suture joint and are obtained from Eqs. (16) and (23a)–(23d), respectively. The expressions of the nondimensional parameters $C_2, C'_2$, and $C''_2$ in Eqs. (24a) are as follows:

$$C_2 = \frac{\tilde{G}_{\text{DTM}}}{\tilde{E}_{\text{DTM}}} + (\tan \theta)^{-2}.$$  \hspace{1cm} (24b)

$$C'_2 = \frac{3E_1}{4G_2^N} + \frac{5}{16} (\tan \theta)^{-2},$$ \hspace{1cm} (24c)

$$C''_2 = \frac{3\tilde{G}_{\text{DTM}}}{2G_2^N} + \frac{9}{8} \frac{\tilde{E}_{\text{DTM}}}{E_1} (\tan \theta)^{-2},$$ \hspace{1cm} (24d)

where the effective local shear modulus $G_2^N$ of the second-order RVE follows the Voigt law,

$$G_2^N = \tilde{G}_{\text{DTM}} (1 - f) + G_1 f.$$ \hspace{1cm} (24e)

For an $N$th-order DTM, the iterative system of equations is as follows:

$$\tilde{E}_{\text{DTM}}^N = \frac{f^2 E_1}{(1 - f)(1 + C_N^N) E_1} + f,$$

$$\tilde{G}_{\text{DTM}}^N = \frac{f^2 C_N^N \tilde{E}_{\text{DTM}}^N}{f C_N^N + (1 - f)(1 + C_N^N) E_1},$$ \hspace{1cm} (25a)

where

$$C_N^N = \frac{\tilde{G}_{\text{DTM}}^N}{\tilde{E}_{\text{DTM}}^N} (1 - f) + (\tan \theta)^{-2},$$ \hspace{1cm} (25b)

$$C'_N^N = \frac{3E_1}{4G_N^N} + \frac{5}{16} (\tan \theta)^{-2},$$ \hspace{1cm} (25c)

$$C''_N^N = \frac{3\tilde{G}_{\text{DTM}}^N}{2G_N^N} + \frac{9}{8} \frac{\tilde{E}_{\text{DTM}}^N}{E_1} (\tan \theta)^{-2},$$ \hspace{1cm} (25d)

and

$$G_N^N = \tilde{G}_{\text{DTM}}^N (1 - f) + G_1 f.$$ \hspace{1cm} (25e)

FIG. 10. (Color online) Tailorable nondimensional stiffness-to-density ratio of self-similar hierarchical suture joints ($\tilde{E}$ in the y axes refers to either $\tilde{E}_{\text{RTM}}$ [Eqs. (12a) and (12b)] or $\tilde{E}_{\text{DTM}}$ [Eqs. (24a)–(24l) and (25)]). (a) Schematics of self-similar hierarchical suture joints in comparison, constructed through the method shown in Fig. 4(a) and the tailorability of the nondimensional stiffness-to-density ratio predicted by RTM and DTM, depending on: (b) number of hierarchy $N$ (by taking $f = 0.8$, $2\theta = 60^\circ$, and $R_s = 10$, 100, 1000, and 10 000 as examples), (c) stiffness ratio (by taking $f = 0.8$, $2\theta = 60^\circ$, and $N = 1–3$ as examples, the shaded pink region indicates the region of stiffness ratios for many biological systems and nanocomposites [38,39]), (d) tip angle $2\theta$ (by taking $f = 0.8$, $R_s = 1000$, and $N = 1–4$ as examples), (e) the volume fraction of a building block $f$ (by taking $2\theta = 60^\circ$, $R_s = 1000$, and $N = 1–4$ as examples), and (f) tailorability of tensile stiffness when the total volume fraction of teeth is fixed at $f_v = 0.98$. 

031901-9
V. RESULTS OF THE RTM AND DTM MODELS: TENSILE STIFFNESS OF HIERARCHICAL SUTURE JOINTS

The numerical results of Eqs. (24a)–(24l) and (25) are shown in Fig. 10. In order to evaluate the effect of $N$ for the same composite composition, we define and plot the stiffness-to-density ratio \[ 37 \], i.e., the effective stiffness per unit volume skeletal tooth material\[ 4 \] stiffness-to-density ratio \[ 37 \], as a function of $N$ as shown in Fig. 10(b).

As shown in Fig. 10(b), the RTM provides an upper bound to the DTM. As predicted by the DTM, the tensile stiffness-to-density ratio of a hierarchical suture joint increases with the total number of hierarchies $N$, but this trend is limited by the stiffness ratio. When $N$ increases, the nondimensional stiffness-to-density ratio $\bar{E}/f_0, E_0$ has an asymptotic upper limit determined by the stiffness ratio, which is approximately $10^{6}$. When $N > N_c (R_S, \theta, f)$, any further increase in $N$ causes little increase in the effective stiffness-to-density ratio, and $N_c$ varies approximately with $\log(R_S)$, depending on the values of $R_S, \theta$, and $f$. $N_c$ increases when $R_S$ or $\theta$ increases and when $f$ decreases.

More importantly, when the stiffness ratio of the two phases is in the range of $10–10^4$ as shown in Fig. 10(c) (shaded interfacial region), it is very efficient to increase the stiffness of a hierarchical suture joint by increasing the number of hierarchies $N$. Many biological systems or nanocomposites lie in this stiffness ratio range \[ 38,39 \]. For a very small stiffness ratio, the stiffness-to-density ratio cannot be increased very much (and can even be reduced slightly if $R_S$ is small) by increasing $N$. Moreover, when $N$ increases, the stiffness significantly increases for all $\theta$ and $f$, but the stiffness becomes less sensitive to $\theta$ and $f$. When $N > 1$, the sensitivity of the effective stiffness to the geometric parameters $\theta$ and $f$ is reduced significantly at small $\theta$ and large $f$ [Figs. 10(d) and 10(e)].

When the total volume fraction is fixed, the influence of $N$ on the effective tensile stiffness can be evaluated via Eqs. (25a)–(25e) and (4). As shown in Fig. 10(f), which indicates that, when the same composite composition is used to construct a hierarchical suture joint, the effective stiffness of the RTM significantly depends on its geometry, described by the number of levels of hierarchy $N$ and the tip angle $\theta$.

For the same tip angle $\theta$, when $N$ increases while holding the total volume fraction $f$, constant, the volume fraction $f$ of the building block needs to decrease as shown in Fig. 4(b). Thus, when $N$ increases, two mechanisms compete with each other: (1) $N$ is increased, which is the factor that increases the stiffness, however, (2) $f$ is decreased, to keep the same $f_0$, which is the factor that decreases the stiffness. Therefore, a peak stiffness is achieved for a certain $N = N^\ast$: when $N < N^\ast$, the stiffness increases with $N$; after $N$ exceeds $N^\ast$, instead, the stiffness decreases with $N$. Generally, $N^\ast$ increases when $R_S$ or $\theta$ increases and when $f$ decreases. As shown in Fig. 10(f), for very small tip angles (such as $2^\circ$), increasing $N$ from 1 to 2 does not increase the stiffness; this is because the influence of the competing mechanism, Eq. (2), exceeds that of Eq. (1), which is because the shear stiffness is very small when $\theta$ is small; whereas, for modest to relatively large tip angles ($2\theta > 20^\circ$), the stiffness increases sharply when $N$ increases, indicating increasing $N$ to 2 is a very efficient way to increase the stiffness of hierarchical suture joints with modest to relatively large tip angles.

VI. FAILURE MECHANISMS, STRENGTH, AND TOUGHNESS OF THE SELF-SIMILAR HIERARCHICAL SUTURE JOINT MODEL

Here, we formulate a theoretical foundation for the failure of first-order and hierarchical suture joints and explore the role of geometry in tailoring failure mechanisms, strength, and toughness. Each material component (tooth and compliant interface) is assumed to behave elastically (as previously) and possesses a critical failure stress above which catastrophic failure takes place (this theory can be extended easily to nonlinear elastic-plastic materials). The tooth material is assumed to be bonded perfectly to the compliant interface material. For hierarchical suture joints ($N > 1$), three potential failure mechanisms are identified: (I) catastrophic failure of the major (the first-order) teeth, (II) catastrophic failure of the second-order teeth; and (III) relative sliding or shear between the first- and the second-order teeth [Fig. 11(a)]. Interfacial failure is not considered for hierarchical suture joints because mechanical interlocking between higher-order teeth prevents the interfacial fracture and/or flaws that develop from propagating further, thereby preventing catastrophic failure of the joint and providing flaw tolerance. Mechanism III is more likely to happen when the tooth material enters into the plastic regime. Here, we focus on failure mechanisms I and II.

Since failure mechanisms of first-order suture joints ($N = 1$) are different than those of hierarchical suture joints ($N > 1$), the failure analysis of each is derived separately as follows.

A. Failure mechanisms and strength of first-order suture joints ($N = 1$)

For first-order suture joints, potential failure mechanisms that are considered include: (I) failure of the major teeth leading to catastrophic failure of the joint and (II) failure of the compliant interfacial material also leading to catastrophic failure of the joint. The normal stress $\sigma_T$ and shear stress $\tau_T$ in the interface layer are related to the tensile stress \[ 30 \] in the teeth via Eqs. (6). Thus, the stress state of the interfacial material is $[\sigma_T, \nu_0\sigma_T, \tau_T]$, and the stress state of the tooth material is $[\sigma_f, 0, 0]$. A failure criterion of maximum principal stress is used for both materials. We define the tensile strength of tooth material to be $\sigma_f$, and the tensile strength of interfacial material to be $\sigma_T^0$. When $\sigma = \sigma_f$, tooth failure occurs; when the maximum principal stress $\sigma_T^0 = (1+\nu_0)^{1/2} + [\nu_0/(1+\nu_0)]^{1/2} + (\tau_T)^2$ of the interfacial material satisfies $\sigma_T^0 = \sigma_T^0$, the first-order suture joints fail by interface failure. Hence, the effective strength of the first-order
suture joints ($N = 1$) according to each mechanism is as follows:

$$S_{I}[1] = f \sigma_f \quad \text{(tooth failure),}$$

$$S_{II}[1] = \frac{\sigma_f}{c H_1(\theta)} \quad \text{(interface failure),}$$

where $S_{I}[1]$ is the effective strength of the first-order suture joints when tooth failure occurs, $S_{II}[1]$ is the effective strength of the first-order suture joints when interface failure occurs and

$$H_1(\theta) = \sin^2 \theta \left[ \frac{1 + v_0}{2} + \sqrt{\left(1 - \frac{v_0}{2}\right)^2 + \tan^{-2} \theta} \right],$$

and $c$ is the strength ratio of the two materials where

$$c = \frac{\sigma_f}{\sigma_0}.$$  

The function $H_1(\theta)$ provides the failure mechanism indicator of the first-order suture joints and is plotted in Fig. 11(b). If $H_1(\theta) < \frac{1}{c}$, tooth failure occurs, otherwise, interface failure occurs. If $H_1(\theta^\circ) = \frac{1}{c}$, the critical tip angle $\theta^\circ$ corresponds to the transition between the two failure mechanisms. When $\theta < \theta^\circ$, tooth failure occurs, and when $\theta > \theta^\circ$, interface failure occurs. The tensile strength $S_{[1]}$ of the first-order suture joints is determined by the strength of the two materials $\sigma_f$ and $\sigma_0$ and the volume fraction of teeth $f$ and tip angle $\theta$. Based on Eq. (26), $S_{[1]}$ is the smaller value of $S_{I}[1]$ and $S_{II}[1]$ expressed as

$$S_{[1]}(\sigma_f, \sigma_0, f, \theta) = \min[S_{I}[1], S_{II}[1]].$$

**B. Failure mechanisms and strength of the hierarchical suture joints ($N > 1$)**

Failure in the higher-order sutures begins by assessing the stress state in the higher-order teeth. The relationships between
the tensile stress in the first-order teeth $\sigma_{[1]}$, the tensile stress of the second-order teeth $\sigma_{[2]}$, and the shear stress between the first-order and the second-order teeth $\tau_{[2]}$ are as follows:

$$\tau_{[2]} = \sigma_{[1]} \frac{6 A_2}{f \lambda_2} = \frac{f \tau_{[2]} 3}{\tan \theta}, \quad (31)$$

where $\sigma_b$ is the local maximum normal stress in the second-order teeth, generated by tooth bending [similar to what is shown Fig. 8(a)] due to the shear stress $\tau_{[2]}$. $\sigma_b$ is expressed as

$$\sigma_b = f \frac{6 A_2}{f \lambda_2} = \frac{6 f \tau_{[2]} 3}{\tan \theta}, \quad (30)$$

where $A_2$ and $\lambda_2$ are the amplitude and wavelength of the second-order teeth. It is worth noting that $\sigma_b$ only exists for failure mechanisms I and II of hierarchical suture joints. For the first-order suture joints, the term $\sigma_b$ does not exist. Also, when the tooth material fails by relative sliding in the plastic region between major and minor teeth, the term $\sigma_b$ disappears in Eq. (30), and therefore, a higher strength is expected for this mechanism.

$$H(f, \theta) = 0.5 \left( \frac{1}{f} + 3 \cos^2 \theta \right) + \sqrt{0.25 \left( \frac{\sin^2 \theta - \cos^2 \theta}{f} + 3 \cos^2 \theta \right)^2 + \left( \frac{\sin \theta \cos \theta}{f} \right)^2}. \quad (33)$$

If we define that the strength of the hierarchical suture joints corresponds to catastrophic failure, then the strength $S_{[N]}$ of the hierarchical suture joints ($N > 1$) is determined by the tooth material strength $\sigma_f$, and the geometric parameters of the building block (i.e., $f$ and $\theta$) and is expressed as

$$S_{[N]}(\sigma_f, f, \theta) = \min \left[ S^I_{[N]}, S^{II}_{[N]} \right] \quad (N > 1). \quad (34)$$

The function $H(f, \theta)$ is the failure mechanism indicator of hierarchical suture joints. If $H(f, \theta) > 1$, failure mechanism II occurs, otherwise, failure mechanism I occurs. However, $H(f, \theta)$ is always larger than 1 for all values of $f$ and $\theta$ as shown in Fig. 11(b), which means that, under the assumption of material elasticity for the hierarchical suture joints, catastrophic failure will never occur in mechanism I. By fixing the total volume fraction $f_v$ of the tooth material, the effective strength $S_{[N]}$ of hierarchical suture joints ($N > 1$) and that of the first-order suture joints $S_{[1]}$ are nondimensionalized by $f_v \sigma_f$ and are plotted and are compared in Fig. 11(c).

C. Fracture toughness of the first-order and hierarchical suture joints

We define the nondimensional tensile fracture toughness $\Gamma^*$ as the effective tensile toughness of the suture joints normalized by the tensile toughness of tooth material, which is expressed as

$$\Gamma^* = \frac{(S)^2}{(S_{[1]})^2} \frac{E_1}{E_N} \quad (35)$$

where $S$ is the effective strength of the suture joint, referring to $S_{[1]}$ in Eq. (29) (when $N = 1$) and $S_{[N]}$ in Eq. (34) (when $N > 1$). The influences of hierarchy $N$ and geometry on tensile fracture toughness of the first-order and hierarchical suture joint model are plotted and are compared in Fig. 11(d) when the total volume fraction $f_v$ is fixed at 0.98 and the stiffness ratio is fixed at 100.

Figures 11(c) and 11(d) show that the first-order suture joints may fail for either of the two failure mechanisms (I for small $\theta$ and II for large $\theta$), whereas, hierarchical suture joints only fail for mechanism II. Also, the tensile strength and fracture toughness of the first-order suture joints depend on the strength ratio $c$, decreasing with increasing values of $c$, while the tensile strength and fracture toughness of hierarchical suture joints ($N > 1$) do not depend on $c$. If the same composite composition (the same $f_v$) is used to construct either a first-order suture joint or a self-similar hierarchical suture joint, when the tip angle $\theta$ is small, the tensile strength and fracture toughness of the first-order suture joints are larger than that of hierarchical suture joints, and the strength and tensile fracture toughness of different order hierarchical suture joints are quite close to each other. However, for relatively large tip angles, the fracture strength and fracture toughness are increased significantly by increasing $N$ from 1 to a value larger than 1. For hierarchical suture joints ($N > 1$), when $N$ increases, the fracture toughness increases, but the strength decreases.

VII. CONCLUSIONS

In this paper, we construct an elastic deterministic fractal composite model for distributing hierarchically structured interfaces and suture joints to quantitatively investigate the role of structural hierarchy on the mechanical stiffness, strength, and failure of suture joints. Suture joints in nature
allow for segmenting monolithic shells into more compliant structures to accommodate biological functions, such as growth, respiration, locomotion, and/or predatory protection [32,40]. The elastic regime studied here is expected to be biologically relevant for typical physiological loading conditions.

The use of hierarchy in natural suture joints is an effective geometric-structural strategy to achieve different properties with limited material options in nature. The hierarchical structure is employed to increase the design space when other structural geometries and parameters (such as the tooth tip angle) are biologically challenging or inaccessible. This paper also provides predictive capabilities to guide the mechanical design of synthetic flaw-tolerant bioinspired interfaces and joints.

A. Mechanical tailorability and amplification

The key finding of this paper is the capability of the hierarchy in a linear elastic system to tailor and to amplify mechanical behavior (e.g., stiffness, strength, and fracture toughness) nonlinearly and with high sensitivity over a wide range of values (orders of magnitude) for a given volume and weight.

**Stiffness**. It is found that, if the teeth of a hierarchical suture joint are rigid sufficiently relative to the more compliant interlayer, the effective tensile stiffness increases exponentially with $N$ and is enhanced further by a geometry amplification factor $k(f, \theta)$. This is because the mechanical properties of the effective interface are amplified via the hierarchical geometry. However, if tooth deformation is considered, the increase in stiffness with the increase in hierarchy is limited by the stiffness ratio between the skeletal teeth and the interfacial material $R_S$. When $N \gg \log R_S$, no significant improvement can be achieved by further increasing $N$ [Fig. 10(a)]. Remarkably, as shown in Figs. 10(c)–10(e), when the stiffness ratio of the two phases is in the range of $10^{-1}$–$10^4$ (many biological systems lie in this stiffness ratio range [38,39]), the stiffness of the hierarchical suture joint is very sensitive to the number of hierarchies. Therefore, for the biological systems (or synthetic systems with stiffness ratios in the same range), it is very efficient to increase the stiffness of the suture joint by increasing the number of hierarchies $N$. However, when the total volume fraction of skeletal teeth is fixed, there is an optimal $N^\ast$ (depending on $R_S$ and $\theta$), beyond which, no further stiffening can be achieved by increasing $N$ as shown in Fig. 10(f).

**Strength and fracture toughness**. Generally, for the same composite composition, there are two efficient ways to increase the tensile strength and fracture toughness: using the first-order suture joints ($N = 1$) and decreasing $\theta$, promoting tooth failure; for relatively large $\theta$, using the higher-order hierarchical suture joints ($N > 1$) to increase the effective strength of the effective interface, promoting failure in the minor teeth [Fig. 2(b)]. For relatively large $\theta$, when $N$ increases, the fracture toughness increases, but the strength-to-density ratio decreases because of the local normal stress, arisen by the coupled bending effect of minor teeth under shear. In biological systems, plasticity will also play a role in toughness, and including postyield deformation and dissipation would further enhance the effects on strength and on toughness presented here. Hence, for hierarchical suture joints, the postyielding strength and fracture toughnesses are expected to be improved significantly when $N > 1$.

B. Interlocking of higher-order teeth and flaw tolerance

The continuity of the teeth at different hierarchies along the effective interface provides a substantial advantage in reducing interfacial failure. Increasing hierarchy was found to result in mechanical interlocking of higher-order teeth, which creates additional load resistance capability and local nonuniform interfacial stress distributions with regions of compression or tension (Fig. 7), thereby preventing catastrophic failure in lower-order teeth (a sacrificial mechanism) and providing flaw tolerance, providing more recovery time for the self-healing process of either biological or engineering systems [41,42].

C. Relevance to natural systems

Hence, the results shown in this paper demonstrate the great sensitivity of mechanical behavior to hierarchy indicating that the diversity of hierarchical and fractal-like interfaces and joints found in nature have definitive functional consequences. Hierarchy is one geometric strategy that nature used to address the limited material option. The use of hierarchy as a design parameter in biological systems likely is employed when other structural geometries and parameters (for example, $\theta$) are biologically challenging or inaccessible. This model may also facilitate a better understanding of the evolution of biological sutures.

(a) *The pelvic suture of Gasterosteus aculeatus* (the threespined stickleback [17,20]) exhibits a first-order suture $N = 1$ with a spatially graded range of $\theta$ (∼$2^\circ$–$60^\circ$). As previously shown [32], *G. aculeatus* utilizes small $\theta$ to provide increased stiffening near the trochar joint.

(b) *The turtle carapace suture* [40] exhibits a complex three-dimensional suture joint with two morphologies in two cross sections [40]: a first-order suture in the sagittal plane and a fractal one in the transversal plane within the suture region.

(c) *Vertebrate cranial sutures* [12,13,18,19] possess morphological evolution in different stages of growth. For example, the cranial suture of humans develops from a simple relatively straight line to very complex first-order and even fractal-like patterns as one matures [43]; the cranial suture of mammals with horns, such as the white tailed deer [21] exhibit more complex patterns than that of humans to improve the mechanical properties to resist occasional impact loading transmitted via the horns.

(d) *Ammonite septal sutures* [22–25], which are hierarchical ($N = 1$–3) or fractal, with low $\theta$ [compared with (a)] are observed to increase complexity continuously through several mass extinctions [25], implying that the ammonites have adapted and have evolved toward a functional design.

**ACKNOWLEDGMENTS**

We gratefully acknowledge support of the US Army through the MIT Institute for Soldier Nanotechnologies (Contract No. DAAD-19-02-D0002), the Institute for Collaborative Biotechnologies through Grant No. W911NF-09-0001 from the US Army Research Office, and the National Security Science and Engineering Faculty Fellowship Program (Grant No. N00244-09-1-0064).