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ABSTRACT

Despite the recent achievements in the stretchability of kirigami metallic glasses, relevant research is still mainly relying on the single-level kirigami structure. In our work, three different hierarchical levels of patterns were developed for kirigami metallic glass structures for both strip and square units. The degree of freedom that reflects the availability for morphing is shown to be a key factor affecting the mechanical response of the hierarchical metallic glasses. It is found that a high hierarchical order with a large degree of freedom leads to the high stretchability of kirigami metallic glasses. Kirigami metallic glasses, designed with square units, with a large degree of freedom show high stretchability compared to those designed with strip units, with a small degree of freedom, at the same hierarchical level. Our study, combining the degree of freedom in the hierarchical construction, highlights the potential for a multilevel architected structure as a programmable block for stretchable mechanical metamaterials.

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I. INTRODUCTION

Metallic glasses, being strong and stiff materials with the short-medium range order structures,^{1–3} have attracted attention for fundamental research and engineering applications. However, their macroscale brittleness remains an issue for engineering applications.^{4,5} Two approaches are commonly adopted to enhance the mechanical properties of metallic glasses: the design of the intrinsic structures and the external structures. Various intrinsic structures have been developed, such as nanoglass (metallic glasses with nanoscale grain structures)^{6–8} and rejuvenescent metallic glass (more disordered metallic glass structure).^{9,10} However, intrinsic structure designs are complex and difficult to control. On the other hand, an architected external structure, which is engineered to overcome the strain–stress trade-offs and unleashes new property space in materials, has been applied in various materials, such as fabrics,¹¹ kaptons,¹² graphene,¹³ hydrogels,¹⁴ and metallic glass to enhance their mechanical properties. Chan and co-workers introduced kirigami patterns to metallic glass,¹⁵ and their findings showed that metallic glasses designed with kirigami patterns extended the intrinsic mechanical performance of metallic glasses.

The kirigami metallic glass showed a large elastic strain, more than 198%, and a small strain energy loss of less than 3%.¹⁵ In addition, Chan and co-workers found the elastic strain and stretchable performance of kirigami metallic glass depended on the kirigami pattern, such as curved elements, cut distances, and circle-shaped node-cuts.¹⁶ Furthermore, they illustrated the deformation mechanism underlying the morphable responses of kirigami patterns by introducing beam deflection theory and a stress concentration factor, predicting the experimental critical force and explaining the origin of the kirigami-inspired deformation.¹⁷ However, the design space of the morphology and mechanical response of the kirigami patterns of metallic glasses still remains to be broadened. Cho *et al.* introduced hierarchical structures into the kirigami patterns with square units and found that hierarchical structures increased the tunability of the engineered materials.¹⁸ Grima *et al.* showed that the mechanical response of hierarchical structures is influenced by the connectivity of the architected structure.¹⁹ Dudek showed that the deformation behavior of hierarchical structures can be tailored by changing the resistance to the rotational motion of the architected structure.²⁰ Chen and co-workers further showed the connectivity and rotation of the architected structures are related to the degree of

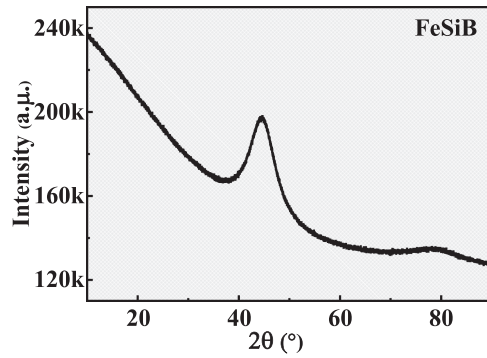


FIG. 1. XRD pattern of the $\text{Fe}_{78}\text{Si}_9\text{B}_{13}$ metallic glass.

freedom (DOF).²¹ Considering the movements of the rigid kirigami primitives inspire the deformation response of the architected metallic glasses, DOF, which reflects the availability of a rigid object morphing through 3D space, is a key parameter in understanding the deformation information of kirigami structures. However, the direct relationship between DOF and deformation behavior of hierarchical structures remains to be determined.

In this study, new hierarchical kirigami patterns are introduced in metallic glasses. The hierarchical kirigami patterns enable the realization of the programmable stretchability of metallic glasses. Furthermore, the kirigami-inspired deformation behavior of metallic glasses is investigated, shedding light on the underlying relationship between DOF and the deformation response of hierarchical kirigami metallic glasses. The study also shows that the spatial

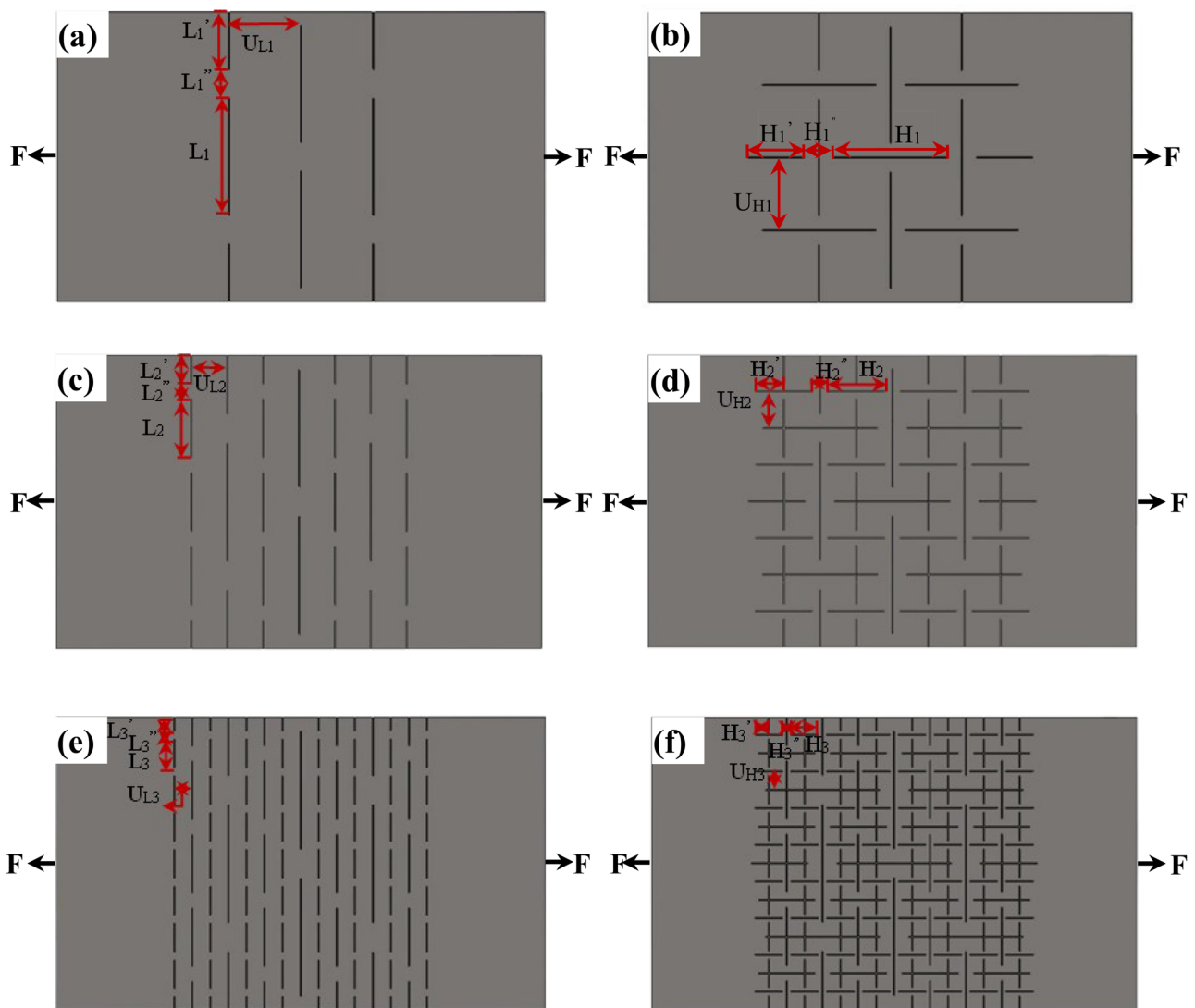


FIG. 2. Schematic illustration of hierarchical kirigami patterns. H represents the horizontal length of the pattern units, and L represents the vertical length of the pattern units. (a) Strip order 1 pattern. (b) Square order 1 pattern. (c) Strip order 2 pattern. (d) Square order 2 pattern. (e) Strip order 3 pattern. (f) Square order 3 pattern.

TABLE I. The parameters of the kirigami patterns, $U_0 = 280 \mu\text{m}$.

Strip order 1	$L_1 = 32U_0$	$L_1' = 16U_0$	$L_1'' = 8U_0$	$U_{L1} = 20U_0$
Strip order 2	$L_2 = 16U_0$	$L_2' = 8U_0$	$L_2'' = 4U_0$	$U_{L2} = 10U_0$
Strip order 3	$L_3 = 8U_0$	$L_3' = 4U_0$	$L_3'' = 2U_0$	$U_{L3} = 5U_0$
Square order 1	$H_1 = 32U_0$	$H_1' = 16U_0$	$H_1'' = 8U_0$	$U_{H1} = 20U_0$
Square order 2	$H_2 = 16U_0$	$H_2' = 8U_0$	$H_2'' = 4U_0$	$U_{H2} = 10U_0$
Square order 3	$H_3 = 8U_0$	$H_3' = 4U_0$	$H_3'' = 2U_0$	$U_{H3} = 5U_0$

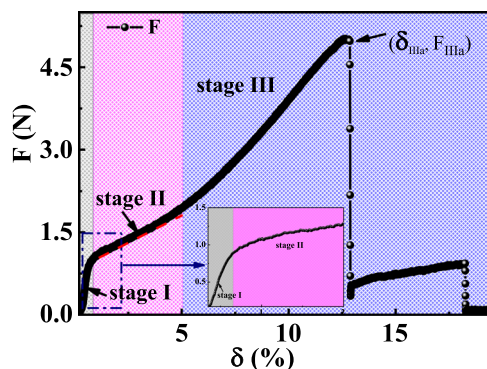
direction makes a difference in the DOF effect on the stretchability of kirigami metallic glasses.

II. EXPERIMENTS

$\text{Fe}_{78}\text{Si}_9\text{B}_{13}$ (at.%) metallic glass was chosen as the model material. Photochemical machining (PCM) was applied to a $\text{Fe}_{78}\text{Si}_9\text{B}_{13}$ metallic glass ribbon with a thickness of $40 \mu\text{m}$ to obtain kirigami patterns. The length and width of the sample ribbons are 38.6 and 22.4 mm, respectively. The amorphous structure of the sample was confirmed by XRD, as shown in Fig. 1. The kirigami samples were designed with three hierarchical pattern levels for the strip units and the square units, respectively. Details of the patterns are shown in Fig. 2, with the size of the unit primitives of level 1 twice that of level 2, while the size of the units of level 2 is twice that of level 3. Further information on the patterns is shown in Table I. Tensile tests were conducted using an MTS CMT5205, with a strain rate of $5 \times 10^{-3} \text{ s}^{-1}$.

III. RESULTS AND DISCUSSIONS

The force-strain responses of the kirigami samples that involve three stages, caused by the deformation of the kirigami patterns, are displayed in different colors in Fig. 3. During stage I, which is very short, the sample displays in-plane deformation with a linear elastic response. During this stage, the force increases sharply with strain. During stage II, the sample displays elastic out-of-plane deformation, which is first caused by the buckling of the first-order patterns, then buckling of the second-order kirigami pattern occurs,

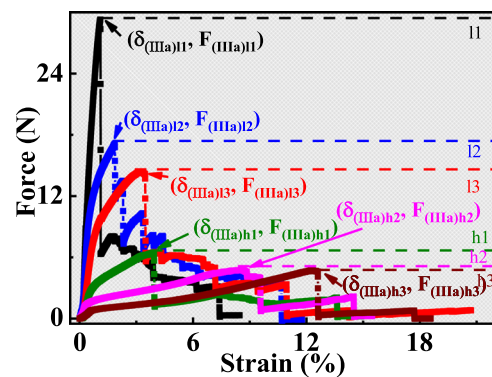
**FIG. 3.** The strain-force curve of kirigami metallic glass. The inset shows the enlarged curve corresponding to the region covered by the dashed line.

and following buckling of the third-order kirigami pattern. In this stage, the deformation changes from 2D to 3D for the kirigami patterns. The sample displays out-of-plane softening deformation in this stage. During stage III, the sample displays out-of-plane stiffening deformation in the initial phase. Following the stiffening deformation, the first-order kirigami patterns are broken due to the fracture of the connection of the architected structure, resulting in serrations in the force-strain curve. The kirigami patterns with the first order but not the kirigami patterns with other orders lead to the failure of the kirigami samples. The strain of the first serration in stage III is defined as δ_{IIIa} , which is the starting strain of the failure, as shown in Fig. 3. The morphologies of the kirigami patterns with the corresponding deformation stages have been presented in our other paper; interested readers are referred to Ref. 11. The strain-force curves of kirigami metallic glasses with different hierarchical levels are presented in Fig. 4. δ_{IIIa} decreases with decreasing hierarchical levels, and δ_{IIIa} for strip units is smaller than that of the square units, as shown in Fig. 4.

Considering that the planar kirigami metallic glass is a thin sheet system with joint-cuttings, the kirigami structure can be taken as quads with links.²¹ As shown in Fig. 5, the quads are separated and the neighboring nodes of the quads are connected by hinges. With this setup, the cutting problem is converted to a linkage problem in kirigami metallic glass. The degree of freedom and the set of independent dimensions of the translation and rotation of the rigid matrix^{22,23} play a crucial role in the deformation behavior of the link patterns. The minimum degree of freedom (DOF) for the kirigami patterns is related to the set of positions for the links; $(DOF)_{\min}$ for the kirigami metallic glass with blocks of $m \times n$ quads is taken as²¹

$$(DOF)_{\min} = 3mn - 2\delta(mn), \quad (1)$$

where m is the number of quads in each line, n is the number of quads in each column, $3mn$ is the total number of DOF for the $m \times n$ kirigami patterns with no connected links, and $\delta(mn)$ is the number of links to rigidify the kirigami samples. Since the reduction of the maximum number of DOF is two, after adding one link, the reduction of the maximum number of DOF is $2\delta(mn)$ after adding $\delta(mn)$ links, and hence, the minimum number of DOF can be

**FIG. 4.** Tensile curves of kirigami metallic glasses.

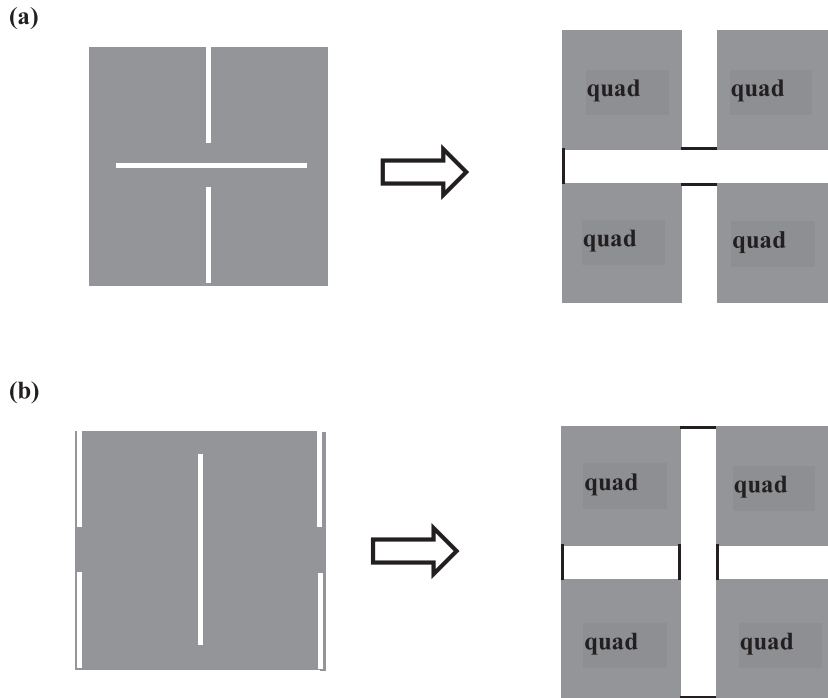


FIG. 5. Planar kirigami with the corresponding segment of quads with hinges. (a) Kirigami structure with strip units. (b) Kirigami structure with square units.

derived as Eq. (1). Based on Eq. (1), $(DOF)_{\min}$ for the hierarchical kirigami metallic glass in this study is calculated and presented in Table II. The trend of δ_{IIIa} with $(DOF)_{\min}$ is shown in Fig. 6. For the kirigami samples with strip pattern (or square pattern), the higher

TABLE II. The minimum degree of freedom for kirigami metallic glasses with different architected patterns. L1 is the strip pattern with hierarchical order 1, L2 is the strip pattern with hierarchical order 2, and L3 is the strip pattern with hierarchical order 3; H1 is the square pattern with hierarchical order 1, H2 is the square pattern with hierarchical order 2, and H3 is the square pattern with hierarchical order 3. $(DOF)_{\min}$ is the minimum degree of freedom for the certain kirigami sample.

Kirigami pattern	L1	L2	L3	H1	H2	H3
$(DOF)_{\min}$	4	30	110	8	40	136

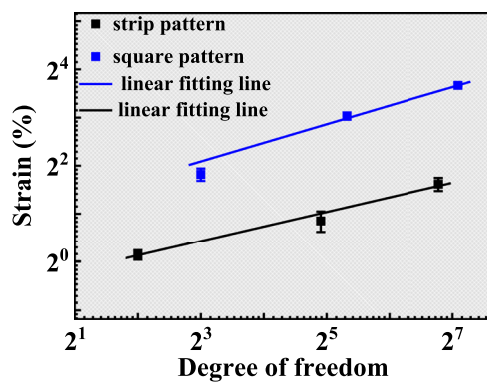


FIG. 6. The change of δ_{IIIa} with the minimum degree of freedom.

the hierarchical order, the more $(DOF)_{\min}$, and the more possible positions for kirigami sample stretching. The square kirigami samples with $(DOF)_{\min}$ show higher stretchability than those strip kirigami samples, at the same order of hierarchical structure. However, the changing trend of δ_{IIIa} with the minimum degree of freedom for kirigami samples with different kinds of architected pattern is not presented in a single curve, i.e., the relationship between δ_{IIIa} and the minimum degree of freedom shows an obvious linear trend only in the kirigami samples with the same type of kirigami patterns (strip pattern or square pattern). It indicates not only the number but also the spatial direction of the links that affect the role of DOF in the kirigami-inspired deformation response, which would be further studied in our following work. As the hierarchical level increases, the stretchability is enhanced linearly for both the square kirigami samples and the strip kirigami samples. By introducing the hierarchical pattern, the stress-strain response is more regulable and programmable. The mechanical responses of kirigami metallic glasses can be controlled by the degree of freedom of the kirigami patterns.

IV. CONCLUSION

Hierarchical structures enrich the regulatory and programmable nature of kirigami metallic glass with space-saving and shape transformation, contributing to tailoring the mechanical response and designing the ultra-soft materials. A high hierarchical order with a large degree of freedom leads to the high stretchability of kirigami metallic glasses. In addition, mainly affected by the degree of freedom, square kirigami samples with a high degree of freedom show higher stretchability than strip kirigami samples with a low degree of freedom at the same hierarchical level.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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