

1 Ultrasonic evaluation of ply and interleaf layer properties of interleaved composite laminates

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3 Yosuke Ishii<sup>a,\*</sup> and Shiro Biwa<sup>a</sup>

4 <sup>a</sup> *Department of Aeronautics and Astronautics, Graduate School of Engineering, Kyoto University, Katsura,*  
5 *Nishikyo-ku, Kyoto 615-8540, Japan*

6  
7 **Abstract**

8 An ultrasonic nondestructive evaluation procedure is presented for the ply and interleaf layer properties of  
9 carbon/epoxy composite laminates having interleaf resin layers at their interlaminar interfaces. It is shown  
10 that the material properties of plies (density, thickness, and transversely isotropic complex elastic moduli)  
11 and elastic interleaf layers (density, thickness, Young's modulus, and Poisson's ratio) can be identified by  
12 best fitting the theoretical energy transmission spectra of longitudinal waves through the laminate immersed  
13 in water to those obtained in ultrasonic measurement. It is also demonstrated that compared to the mass-less  
14 and null-thickness spring-type interlaminar interface model employed in the previous works, the present  
15 finite-thickness interleaf layer model can better reproduce the experimental transmission spectra of a  
16 unidirectional interleaved composite laminate containing ultrasonic bandgaps. Furthermore, the property  
17 characterization in the case where the plies and interleaf layers are assumed to be elastic and viscoelastic,  
18 respectively, is also examined. It is also shown that the ply and interleaf layer properties determined for the  
19 unidirectional laminate can be used to reproduce the transmission spectra measured for a quasi-isotropic  
20 laminate consisting of the same plies and interleaf layers.

21 Keywords: Nondestructive evaluation; Composite laminate; Interleaf layer; Stiffness-matrix method

22 \*Corresponding author: Yosuke Ishii, Department of Aeronautics and Astronautics, Graduate School of  
23 Engineering, Kyoto University, Katsura, Nishikyo-ku, Kyoto 615-8540, Japan.

24 Tel: +81 75 383 3797, E-mail: ishii@kuaero.kyoto-u.ac.jp

## 1. Introduction

Ultrasonic wave propagation in composite laminates has been studied extensively [1–3], leading to the proposal of several nondestructive evaluation methods of mechanical properties. For example, the anisotropic (visco)elastic constants of composite laminates can be identified by measuring the velocity (or time-of-flight) [4–10]/attenuation [10–13] of bulk quasi-longitudinal and quasi-transverse waves, the critical angles at the interface between the laminate and its surrounding fluid [14,15], the amplitude transmission spectrum [16,17], and the dispersion relation of guided waves [18–24]. Besides the average properties of laminates in the layering direction, the elastic constants of a single ply constituting multidirectional composite laminates can also be evaluated by accounting for the wave refraction effects at interlaminar interfaces on the bulk wave velocity [25] and by using the propagation characteristics of Floquet-Bloch waves [26].

Actual polymer-based composite laminates, such as carbon/epoxy and glass/epoxy laminates, have thin resin-rich regions of a few microns thickness between neighboring plies. When the ultrasonic wavelength is sufficiently long, the presence of such interlaminar zones has little influence on the wave propagation, and it is reasonable to assume direct bonding of neighboring plies as in the aforementioned works [4–26]. In contrast, when the wavelength is comparable with the double ply thickness, the quality of interlaminar zones plays a significant role as the wave scattering from interlaminar interfaces can cause constructive interference, resulting in the formation of ultrasonic bandgaps where the wave transmissivity falls to vanishingly low levels. The wave propagation in such a frequency range has been analyzed by the present authors [27–32] by modeling the interlaminar resin-rich zones as mass-less spring-type interfaces [33–42] of equivalent interfacial normal and tangential stiffnesses, and it has been demonstrated that the interfacial stiffnesses, which reflect the elastic properties of interlaminar zones, can be evaluated from the broadband reflection/transmission spectra including the bandgaps.

Recently, a variety of fabrication techniques of carbon-fiber-reinforced composites have been proposed to improve their inherent drawback of low interlaminar strength [43,44], such as the stitching through the layering direction [45–47], Z-pin [48–50], Z-anchor [51,52], and three-dimensional fabric [53,54]. Among them, interleaving heterogeneous resin layers with a typical thickness of 20–30  $\mu\text{m}$  between adjacent plies has widely been adopted in composites for aircraft structures to enhance the interlaminar fracture toughness and to restrict the onset of delaminations [55–59].

In the authors' previous study [60], wider and deeper ultrasonic wave bandgaps have been observed experimentally in the transmission spectra of interleaved composite laminates compared to the conventional non-interleaved ones and reasonably reproduced by the theoretical analysis with the spring-type interlaminar interface model. In principle, the elastic properties of interleaf layers can be estimated in an approximate manner from the equivalent interfacial stiffnesses identified by the spring-type interface model [35]. On the other hand, from a practical perspective, it is an important and intriguing issue to evaluate the interleaf layer properties more directly by modeling them as an elastic layer of finite thickness. Such modeling was employed by Wang and Rokhlin [61] in the analysis of the reflection spectrum of a longitudinal wave at normal incidence to the conventional composite laminates having thin interlaminar resin-rich regions. They also considered the case of oblique incidence, whereas their main concern was not the inverse determination of material properties of interlaminar resin layers but the direct problem of a reflection response of low frequency range where the influence of resin layers was negligible. Okabe and Takeda [62] evaluated the interleaf layer properties (i.e., density, thickness, Young's modulus, and Poisson's ratio) in a destructive manner by the direct reflection measurement from a single interleaf layer that was exposed by the mechanical grinding of interleaved composite laminates. When calculating the theoretical reflection coefficients from the exposed interleaf layer, they neglected the viscoelastic nature of the substrate ply and assumed its elastic properties to be those evaluated beforehand for a non-interleaved composite laminate specimen. To the best of the present authors' knowledge, the characterization of interleaf layer properties in a completely nondestructive manner has not been addressed in the existing literature.

In contrast to the previous studies, the aim of this paper is to evaluate the ply and interleaf layer properties of interleaved composite laminates simultaneously and nondestructively using ultrasonic transmission characteristics. By modeling the fiber-reinforced plies and interleaf resin layers as homogeneous transversely-isotropic viscoelastic and isotropic elastic layers of finite thickness, respectively, the energy transmission spectra of plane longitudinal waves at oblique incidence to the laminate immersed in water are calculated theoretically by the stiffness-matrix method [63,64]. They are then fitted to the measured transmission spectra to identify the material properties of plies and interleaf layers. The evaluation procedure is outlined in Section 2 and applied to experimental transmission spectra measured for a 16-ply interleaved unidirectional composite laminate. The results are compared to the ply stiffnesses evaluated for the same

81 experimental data by the spring-type interlaminar interface model as in the previous studies in Section 3. The  
82 applicability of both models is then discussed in detail for composite laminates with different interlaminar  
83 layer thicknesses. While the present evaluation procedure ascribes the viscoelastic nature to the plies only,  
84 the opposite case of assuming the elastic plies and the viscoelastic interleaf layers is also examined briefly.  
85 Finally, the evaluated ply and interleaf layer properties are used to reproduce the transmission spectra of a  
86 quasi-isotropic laminate. The present study is concluded in Section 4.

## 2. Evaluation of ply and interleaf layer properties

### 2.1. Modeling of the composite laminate

To describe the evaluation procedure for the ply and interleaf layer properties from the ultrasonic measurements, a theoretical model of the composite laminate is first introduced. A unidirectional composite laminate consisting of  $N$  fiber-reinforced plies and  $N-1$  interleaf resin layers which is immersed in water (density  $\rho^w$  and wave velocity  $c^w$ ) is considered as shown in Fig. 1. The plies are modeled as homogeneous transversely-isotropic viscoelastic media and assumed to have the same thickness  $h^p$  and material properties of density  $\rho^p$  and complex elastic moduli  $C_{IJ}^p$  (fiber direction is set as the  $x_1$  axis). The interleaf layers are modeled as homogeneous isotropic elastic layers and assumed to have the same thickness  $h^l$  and material properties of density  $\rho^l$ , Young's modulus  $E^l$ , and Poisson's ratio  $\nu^l$ . The plies and interleaf layers are assumed to be perfectly bonded to each other; namely, the stresses and displacements are continuous across the ply–interleaf layer interfaces. The total thickness and average density of the composite laminate, denoted by  $h^L$  and  $\rho^L$ , respectively, are related to those of plies and interleaf layers as

$$h^L = Nh^p + (N - 1)h^l, \quad (1)$$

$$\rho^L = \frac{N\rho^p h^p + (N - 1)\rho^l h^l}{h^L}. \quad (2)$$

Regarding the above modeling of the composite laminate, it is noted here that there is an inherent difficulty in separating the contributions of the plies and interleaf layers to the viscoelastic loss of ultrasonic waves. For example, Baltazar *et al.* [65] showed that for a three-layered structure with double spring-type interfaces with complex stiffnesses, the dissipative nature of interfaces (i.e., imaginary parts of interfacial stiffnesses) and the attenuation in the intermediate layer had comparable effects on the reflection spectrum. For composite laminates discussed here, both the plies and interleaf layers actually have the viscoelastic nature, but the above nature indicates that it is difficult to quantify the complex-valued stiffnesses of both constituents separately. Hence, in the present study, the interleaf layers are assumed to be elastic, and the wave attenuation is solely ascribed to the plies, which appears justifiable due to the relatively small thickness of the former. This issue is briefly discussed later by examining the results of the opposite case where the interleaf layers are assumed to be solely responsible for the attenuation in the evaluation.

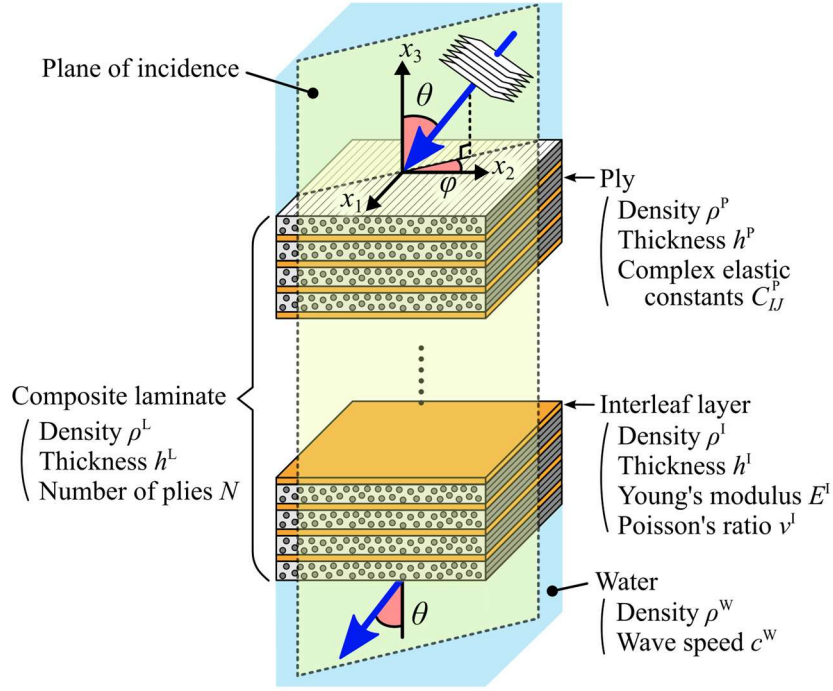


Fig. 1 Unidirectional composite laminate consisting of  $N$  plies and  $N-1$  interleaf layers immersed in water.

## 2.2. Evaluation procedure

In the present study, the density  $\rho^w$  and the wave velocity  $c^w$  of water, the laminate thickness  $h^L$ , the laminate density  $\rho^L$ , and the number of plies  $N$  are assumed to be known in advance. With this assumption, the remaining unknown properties (i.e., the thickness  $h^l$ , the density  $\rho^l$ , Young's modulus  $E^l$ , Poisson's ratio  $\nu^l$  of interleaf layers, the thickness  $h^p$ , the density  $\rho^p$ , and the complex elastic constants  $C_{IJ}^p$  of plies) are evaluated as follows: first, the energy transmission spectra are measured at various incident directions for a composite laminate immersed in water by the through-transmission method [31] to be explained below in Section 2.3. It has been demonstrated in Refs. [31,32,60] that the interlaminar interfacial stiffnesses of composite laminates can be evaluated from the transmission spectra and the bandgap behavior when the spring-type interlaminar interface model is assumed. In this study, similar transmission measurements are performed with a broadband incident longitudinal wave in the frequency range where the wavelength is comparable to the double ply thickness so that the resulting spectra contain several bandgaps. Second, a 14-dimensional real vector  $\mathbf{w}$ , defined with the unknown parameters as  $\mathbf{w} \equiv (h^l, \rho^l, E^l, \nu^l, D_{11}^p, D_{12}^p, D_{22}^p, D_{44}^p,$

128  $D^P_{66}, \eta^P_{11}, \eta^P_{12}, \eta^P_{22}, \eta^P_{44}, \eta^P_{66}$ ) where  $D^P_{IJ} \equiv \text{Re}(C^P_{IJ})$  and  $\eta^P_{IJ} \equiv -\text{Im}(C^P_{IJ})$ , is then optimized in such a manner  
 129 that the following evaluation function  $J(\mathbf{w})$  is minimized:

$$J(\mathbf{w}) = \frac{1}{N_\varphi N_\theta N_f} \sum_{p=1}^{N_\varphi} \sum_{q=1}^{N_\theta} \sum_{r=1}^{N_f} \left| \log_{10} \frac{T^{\text{th}}(\mathbf{w}, f_r, \theta_q, \varphi_p)}{T^{\text{ex}}_{pqr}} \right|, \quad (3)$$

130 where  $T^{\text{ex}}_{pqr}$  are the experimental energy transmission coefficients for the frequency  $f = f_r$  ( $r = 1, 2, \dots, N_f$ ),  
 131 the angle of incidence  $\theta = \theta_q$  ( $q = 1, 2, \dots, N_\theta$ ), and the angle of the plane of incidence  $\varphi = \varphi_p$  ( $p =$   
 132  $1, 2, \dots, N_\varphi$ ). Here,  $N_f$ ,  $N_\theta$ , and  $N_\varphi$  denote the numbers of data points of the frequency, the angle of incidence,  
 133 and the angle of the plane of incidence, respectively. In Eq. (3),  $T^{\text{th}}(\mathbf{w}, f_r, \theta_q, \varphi_p)$  are the theoretical energy  
 134 transmission coefficients of a monochromatic plane longitudinal wave for  $f = f_r$ ,  $\theta = \theta_q$ , and  $\varphi = \varphi_p$ . They  
 135 are calculated by the stiffness-matrix method [63,64] with the optimization variables in  $\mathbf{w}$ , the a-priori known  
 136 properties ( $N, \rho^W, c^W, h^L$ , and  $\rho^L$ ), and  $h^P$  and  $\rho^P$  given from the conditions Eqs. (1) and (2) as

$$T^{\text{th}} = \left| \frac{-2\gamma S^G_{63}}{(\gamma - S^G_{33})(\gamma + S^G_{66}) + S^G_{36}S^G_{63}} \right|^2, \quad (4)$$

137 where  $\gamma \equiv \cos \theta / (2\pi f \rho^W c^W)$  and  $S^G_{mn}$  represent the elements of the  $6 \times 6$  global compliance matrix of the  
 138 composite laminate (see Refs. [63,64] for details on the derivation of Eq. (4)).

139 In the optimization in this study, the complex elastic constants of plies are assumed to be independent of  
 140 frequency. Furthermore, in Eq. (3), the values of  $T^{\text{th}}$  and  $T^{\text{ex}}_{pqr}$  that are less than  $10^{-3}$  are rounded up to  $10^{-3}$ ,  
 141 and the common logarithm is taken for the ratio between  $T^{\text{th}}$  and  $T^{\text{ex}}_{pqr}$  in order to include the transmission  
 142 characteristics not only for small but also for large angles of incidence where the transmissivity is likely to  
 143 be very low. The evaluation function in Eq. (3) is minimized using the MATLAB function “fmincon,” which  
 144 is the constrained nonlinear optimization solver based on the interior point algorithm [66]. From physical  
 145 reasoning, the constraints on the optimization variables are set as  $0 < h^l < h^L/(N-1)$ ,  $0 < \rho^l < h^L \rho^L / [h^l (N-1)]$ ,  
 146  $E^l > 0$ ,  $0 < \nu^l < 0.5$ ,  $D^P_{11} > 0$ ,  $D^P_{22} > 0$ ,  $D^P_{44} > 0$ ,  $D^P_{66} > 0$ ,  $\eta^P_{11} > 0$ ,  $\eta^P_{22} > 0$ ,  $\eta^P_{44} > 0$ , and  $\eta^P_{66} > 0$ , where the  
 147 upper limits of  $h^l$  and  $\rho^l$  are given from Eqs. (1) and (2) with  $h^P > 0$  and  $\rho^P > 0$ .

148 To validate the above evaluation procedure, it has been applied to the artificial transmission coefficients  
 149 computed beforehand by Eq. (4) with material properties representative of interleaved carbon/epoxy  
 150 composite laminates. The procedure has been confirmed to reconstruct the original properties as desired.

### 2.3. Experimental transmission coefficient data

Following the procedure in Refs. [31,32,60], the energy transmission spectra were measured for a 16-ply interleaved unidirectional carbon/epoxy composite laminate immersed in water at various incident directions using the through-transmission method with an ultrasonic polar-scan system (Insight k.k., Tokyo) as schematically shown in Fig. 2. The polar-scan method has been applied to the damage assessment [67,68] as well as the identification of viscoelastic properties of composite laminates [10,15]. The experimental system used in the present study is essentially the same as those in Refs. [10,15,67,68] but applied to the separate evaluation of ply and interleaf layer properties of composite laminates. Two piezoelectric transducers of nominal center frequency 10 MHz were used as the emitter and the receiver. They were held by a computer-controlled rotatable arm in such a manner that their common sound axis intersects a certain point on the specimen surface at various angles  $\theta$  and  $\phi$ , as shown in Fig. 2, with the separation distance being kept at 58 mm. The receiver had a larger diameter to minimize the influence of the shift of the transmitted beam axis due to the refraction at the specimen.

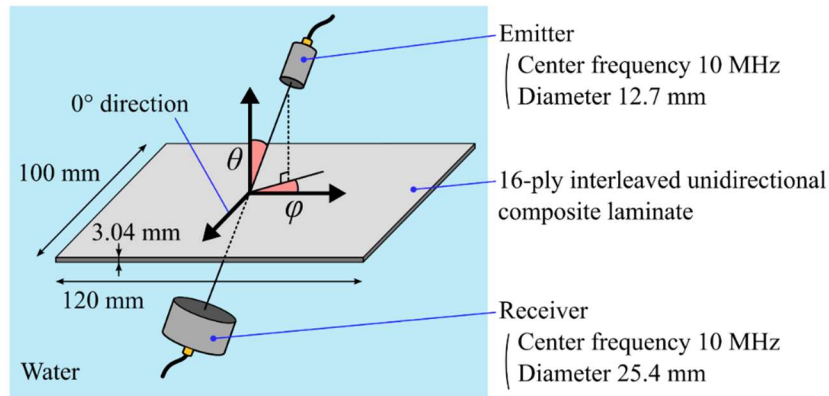


Fig. 2 Schematic of the through-transmission measurement.

The laminate specimen (fabricated from Toray T800S/3900-2B prepregs and having interleaf resin layers) is the same as that used in Ref. [60], and its lateral dimensions are 100 mm  $\times$  120 mm. The temporal transmission waveforms measured at various angles of incidence  $\theta$  are shown in Fig. 3 for three representative angles of the plane of incidence  $\phi = 0^\circ, 45^\circ$ , and  $90^\circ$ . The presence of critical angles is clearly indicated in

these waveforms, i.e., roughly at  $\theta = 30^\circ$  for  $\varphi = 0^\circ$ ,  $\theta = 10^\circ$  and  $36^\circ$  for  $\varphi = 45^\circ$ , and  $\theta = 7^\circ$  for  $\varphi = 90^\circ$ . These waveforms were then fast Fourier transformed with a rectangular window ranging from 37 to 44  $\mu\text{s}$ , whose width was chosen to contain the first arrival waves as well as the subsequent ones caused by multiple reflections inside the specimen, to obtain their power spectra. The resulting power spectra were normalized by the reference spectrum to compute the energy transmission spectra shown in Figs. 4(a)–(c) (shown in Figs. 4(d)–(i) are the theoretically calculated spectra, which will be discussed in the following sections). Note that due to the limited bandwidth of the transducers, the spectra in Figs. 4(a)–(c) are depicted for a finite frequency range of 4 to 12 MHz where the reference signal has at least a quarter of its maximum spectral power (or, equivalently, a half of its maximum spectral amplitude). Several zones with vanishingly low transmissivity seen in the  $f$ – $\theta$  plane in Figs. 4(a)–(c) can be interpreted as ultrasonic wave bandgaps, whose formation mechanism has been examined in detail in the authors’ previous work [60]. Note here that “bandgap” is originally a term defining the frequency ranges where wave propagation in infinitely periodic structures is prohibited, but it will be used below to denote the finite frequency bands where the wave transmission coefficient for finite layered laminates drops to a very low level.

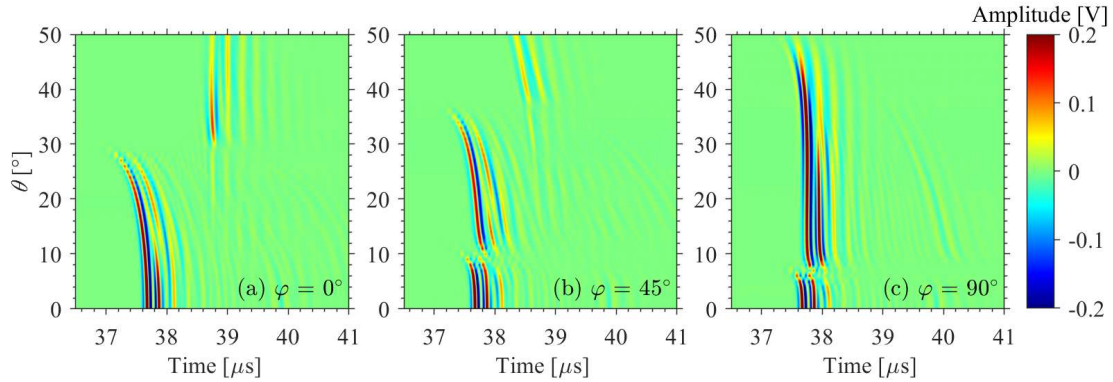


Fig. 3 Variations of the temporal transmission waveforms measured for the 16-ply interleaved unidirectional composite laminate specimen with the angle of incidence  $\theta$  when the angle of the plane of incidence is (a)  $\varphi = 0^\circ$ , (b)  $\varphi = 45^\circ$ , and (c)  $\varphi = 90^\circ$ .

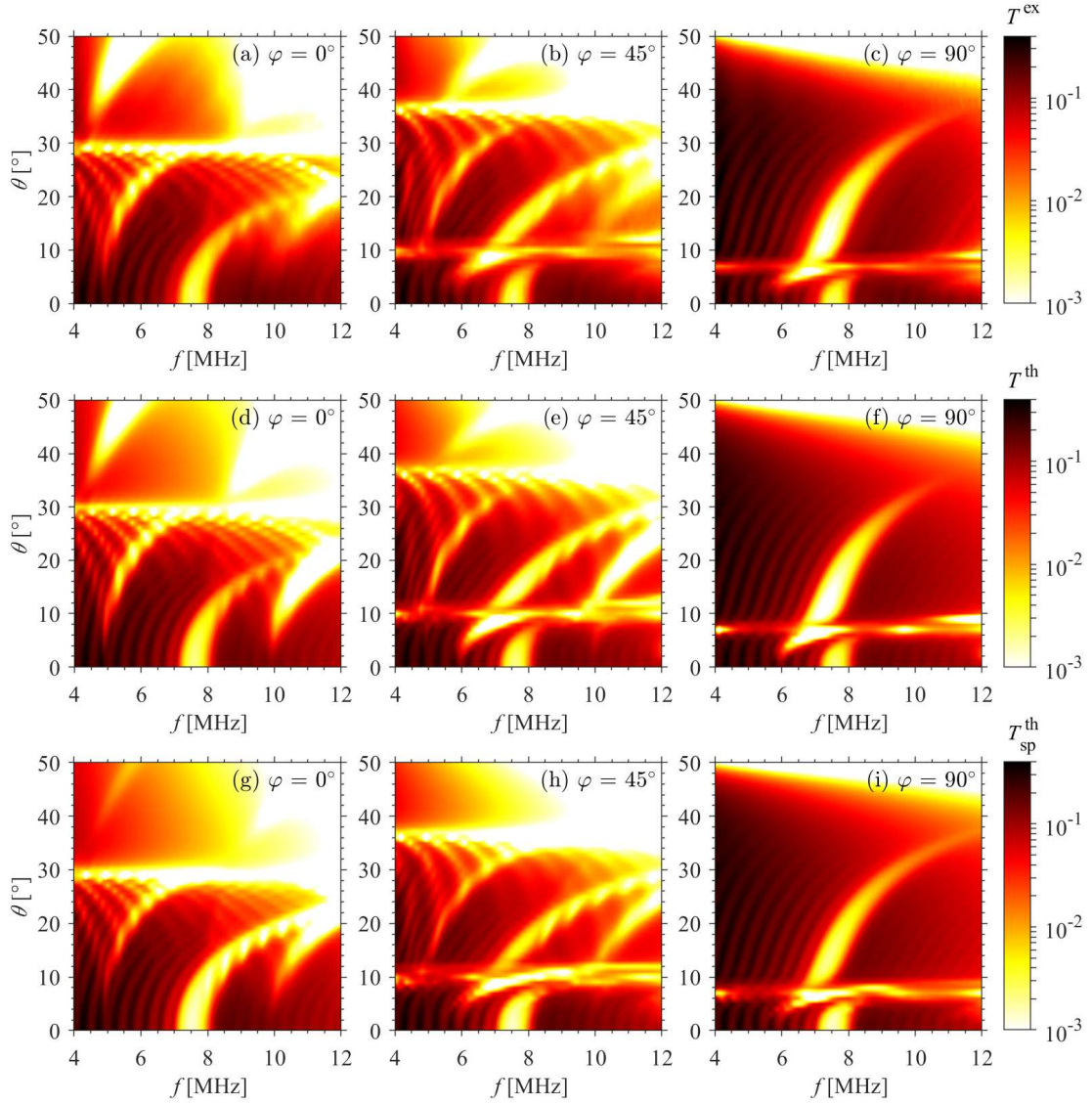


Fig. 4 Distributions of the energy transmission coefficient of the 16-ply unidirectional composite laminate in the plane of the frequency  $f$  and the angle of incidence  $\theta$  when the angle of the plane of incidence is  $\varphi = 0^\circ$ ,  $45^\circ$ , and  $90^\circ$ . (a)–(c) Experiment. (d)–(f) Theoretical analysis with the interleaf layer model. (g)–(i) Theoretical analysis with the spring-type interlaminar interface model.

To increase confidence in the evaluation results, the above polar-scan measurement was performed at 15 different locations on the same specimen, and the evaluation procedure outlined in Section 2.2 is applied to each transmission coefficient data set. The ranges of frequency and incident directions used for the optimization are set as  $4 \leq f \leq 12$  MHz ( $N_f = 106$ , increment  $\Delta f = 0.08$  MHz),  $0^\circ \leq \theta \leq 50^\circ$  ( $N_\theta = 51$ ,  $\Delta\theta = 1^\circ$ ), and  $0^\circ \leq \varphi \leq 90^\circ$  ( $N_\varphi = 7$ ,  $\Delta\varphi = 15^\circ$ ). The known properties of the specimen are listed in Table 1, together with those of water.

203

204 Table 1 A-priori known parameters for the optimization.

|                                                        |      |
|--------------------------------------------------------|------|
| Number of plies $N$                                    | 16   |
| Laminate density $\rho^L [\times 10^3 \text{ kg/m}^3]$ | 1.56 |
| Laminate thickness $h^L [\text{mm}]$                   | 3.04 |
| Density of water $\rho^W [\times 10^3 \text{ kg/m}^3]$ | 1.00 |
| Wave velocity in water $c^W [\text{km/s}]$             | 1.50 |

205

### 3. Results and discussion

#### 3.1. Evaluation results

The average values of the identified parameters for transmission coefficient data measured on 15 different positions on the specimen are summarized in Table 2, together with the minimum and maximum ones in parentheses.

In Table 2, the ply and interleaf layer properties are obtained with relatively small differences between their minimum and maximum values. The initial guess values were chosen arbitrarily such that they were not so far away from representative values of carbon/epoxy composites and epoxy resin layers. The optimization procedure has also been repeated several times with slightly different initial guess values (randomly fluctuated within  $\pm 10\%$  of the ones in Table 2) and confirmed to converge to the same point. The identified complex ply moduli  $C_{IJ}^p$  are representative of the carbon-fiber-reinforced unidirectional composites (e.g., [69]). In Ref. [60], the validity of the equivalent interlaminar interfacial stiffnesses evaluated for the present specimen with the spring-type interlaminar interface model was examined by comparing them with those estimated from the interleaf layer properties evaluated by Okabe and Takeda [62]. Since there is no other literature available for the interleaf layer properties of interleaved composite laminates, Ref. [62] is referred to again here: the present study enables a more direct comparison with the results reported in Ref. [62] since the finite thickness and elastic property of interleaf layers are accounted for. The density, Young's modulus, and Poisson's ratio of the interleaf layer evaluated in Ref. [62] are, in the present notation,  $\rho^l = 1.05\text{--}1.35 \times 10^3 \text{ kg/m}^3$ ,  $E^l = 3.5\text{--}4.8 \text{ GPa}$ , and  $\nu^l = 0.24\text{--}0.42$ , respectively. Compared to them, the interleaf layer properties identified in the present study in Table 2 appear reasonable. Furthermore, a micrographic cross-sectional observation performed for the present specimen provided the interleaf layer thickness of about  $23 \mu\text{m}$  as the average for 150 observation points (with a standard deviation of  $5.1 \mu\text{m}$ ), which is consistent with the identified value of  $h^l = 21.1 \mu\text{m}$  in Table 2. Therefore, it can be concluded that the evaluation procedure outlined in Section 2.2, being of nondestructive nature, can successfully be applied to simultaneously evaluate the ply as well as interleaf layer properties of interleaved composite laminates with reasonable accuracy.

Table 2 Evaluation results for the interleaved composite laminate specimen.

|                                                                | Initial guess | Evaluated value <sup>a</sup> |
|----------------------------------------------------------------|---------------|------------------------------|
| Interleaf layer                                                |               |                              |
| Thickness $h^l$ [ $\mu\text{m}$ ]                              | 15.0          | 21.1 (20.6–21.5)             |
| Density $\rho^l$ [ $\times 10^3 \text{ kg/m}^3$ ]              | 1.20          | 1.38 (1.37–1.39)             |
| Young's modulus $E^l$ [GPa]                                    | 5.0           | 4.4 (4.4–4.5)                |
| Poisson's ratio $\nu^l$                                        | 0.40          | 0.37 (0.36–0.37)             |
| Ply                                                            |               |                              |
| Complex elastic constants                                      |               |                              |
| $C_{IJ}^p = D_{IJ}^p - i \eta_{IJ}^p$ [GPa]                    |               |                              |
| $D_{11}^p$                                                     | 160           | 168 (166–173)                |
| $D_{12}^p$                                                     | 8.0           | 8.0 (7.9–8.1)                |
| $D_{22}^p$                                                     | 14.0          | 15.0 (14.9–15.1)             |
| $D_{44}^p$                                                     | 4.0           | 3.5 (3.5–3.6)                |
| $D_{66}^p$                                                     | 7.0           | 6.9 (6.8–6.9)                |
| $\eta_{11}^p$                                                  | 8.0           | 6.7 (6.4–7.2)                |
| $\eta_{12}^p$                                                  | 0.20          | 0.42 (0.39–0.45)             |
| $\eta_{22}^p$                                                  | 0.40          | 0.43 (0.42–0.45)             |
| $\eta_{44}^p$                                                  | 0.20          | 0.13 (0.13–0.13)             |
| $\eta_{66}^p$                                                  | 0.20          | 0.24 (0.24–0.25)             |
| Thickness $h^p$ [mm] <sup>b</sup>                              | –             | 0.170 (0.170–0.171)          |
| Density $\rho^p$ [ $\times 10^3 \text{ kg/m}^3$ ] <sup>c</sup> | –             | 1.60 (1.59–1.60)             |
| Evaluation function $J$ at convergence point                   | –             | 0.11 (0.11–0.12)             |

<sup>a</sup> The average values over the optimizations for 15 different data sets are shown, together with the minimum and maximum values in parentheses.

<sup>b</sup> The ply thickness is obtained from Eq. (1) with the identified properties.

<sup>c</sup> The ply density is obtained from Eq. (2) with the identified properties.

The theoretical transmission spectra calculated with the properties identified for the experimental data set corresponding to Figs. 4(a)–(c) are shown in Figs. 4(d)–(f). The measured transmission spectra in Figs. 4(a)–(c) are quite well reproduced by the present theoretical model in Figs. 4(d)–(f). The theoretical transmission spectra calculated with the spring-type interlaminar interface model are also shown in Figs. 4(g)–(i) for comparison. Note that in the authors' previous work [60], the ultrasonic wave transmission through the interleaved composite laminates was analyzed using the spring-type interlaminar interface model, and the transmission spectra similar to those in Figs. 4(g)–(i) were reported. In this paper, Figs. 4(g)–(i) are shown for further discussion of the results newly obtained with the interleaf layer model in Figs. 4(d)–(f), as below in Section 3.2.

248

## 249 3.2. Comparison with spring-type interlaminar interface model

### 250 3.2.1. Evaluation results based on spring-type interlaminar interface model

251 The material properties of interleaved composite laminate are now evaluated using the spring-type  
252 interlaminar interface model as in the previous works [27–32,60] to compare with the results of the interleaf  
253 layer model presented above in Section 3.1.

254 For that purpose, the interleaf layers in the theoretical model (Fig. 1) are replaced with mass-less and  
255 null-thickness spring-type interfaces of finite interfacial normal and tangential stiffnesses,  $K_N$  and  $K_T$ ,  
256 respectively. Accordingly, the evaluation procedure presented in Section 2.2 is modified as follows: first, the  
257 14-dimensional optimization vector consisting of unknown parameters  $\mathbf{w}$  is replaced with a 12-dimensional  
258 vector  $\mathbf{w}_{sp} \equiv (K_N, K_T, D_{11}^P, D_{12}^P, D_{22}^P, D_{44}^P, D_{66}^P, \eta_{11}^P, \eta_{12}^P, \eta_{22}^P, \eta_{44}^P, \eta_{66}^P)$ . Second, because of the null  
259 thickness of the interfaces, the conditions Eqs. (1) and (2) are now reduced to  $h^P = h^L/N$  and  $\rho^P = \rho^L$ ,  
260 respectively. The ply thickness and density are, thus, known in advance from the parameters in Table 1 and  
261 fixed throughout the optimization process. Third, instead of the theoretical transmission coefficients  $T^{th}$   
262 given in Eq. (4), those of a composite laminate with spring-type interlaminar interfaces  $T_{sp}^{th}$  are used in Eq.  
263 (3), which are calculated by the stiffness-matrix method again with  $\mathbf{w}_{sp}$  and the a priori known parameters  $N$ ,  
264  $\rho^W$ ,  $c^W$ ,  $h^P$ , and  $\rho^P$  (see Refs. [30,31] for details on the computation of  $T_{sp}^{th}$ ). Finally, the constraints originally  
265 set on  $D_{11}^P$ ,  $D_{22}^P$ ,  $D_{44}^P$ ,  $D_{66}^P$ ,  $\eta_{11}^P$ ,  $\eta_{22}^P$ ,  $\eta_{44}^P$ , and  $\eta_{66}^P$  in Section 2.2 are kept the same but those on  $h^L$ ,  $\rho^L$ ,  $E^L$ ,  
266  $\nu^L$  are replaced with  $K_N > 0$  and  $K_T > 0$  from physical reasoning.

267 This procedure was applied to the same experimental data sets used in Section 3.1, and the identified  
268 properties are summarized in Table 3. The obtained interlaminar interfacial stiffnesses differ slightly from  
269 those reported in Ref. [60] since the present optimization is performed for the newly obtained experimental  
270 data. The ply stiffness components in Table 3 are comparable to their counterparts in Table 2. The minimized  
271 values of the evaluation function in Table 2 are all smaller than their counterparts in Table 3, indicating that  
272 the theoretical transmission coefficients with the interleaf layer model better fit the experimental ones than  
273 those with the spring-type interface model.

274

Table 3 Evaluation results for the interleaved composite laminate specimen based on the spring-type interlaminar interface model.

|                                               | Initial guess | Evaluated value <sup>a</sup> |
|-----------------------------------------------|---------------|------------------------------|
| Interlaminar interfacial stiffnesses [GPa/μm] |               |                              |
| $K_N$                                         | 0.60          | 0.58 (0.56–0.60)             |
| $K_T$                                         | 0.20          | 0.22 (0.21–0.23)             |
| Ply complex elastic constants                 |               |                              |
| $C_{IJ}^P = D_{IJ}^P - i \eta_{IJ}^P$ [GPa]   |               |                              |
| $D_{11}^P$                                    | 160           | 148 (146–152)                |
| $D_{12}^P$                                    | 8.0           | 8.2 (8.2–8.3)                |
| $D_{22}^P$                                    | 14.0          | 15.0 (14.9–15.1)             |
| $D_{44}^P$                                    | 4.0           | 3.4 (3.4–3.5)                |
| $D_{66}^P$                                    | 7.0           | 6.4 (6.3–6.4)                |
| $\eta_{11}^P$                                 | 8.0           | 5.5 (5.2–6.1)                |
| $\eta_{12}^P$                                 | 0.20          | 0.27 (0.25–0.30)             |
| $\eta_{22}^P$                                 | 0.40          | 0.38 (0.37–0.40)             |
| $\eta_{44}^P$                                 | 0.20          | 0.12 (0.12–0.13)             |
| $\eta_{66}^P$                                 | 0.20          | 0.20 (0.20–0.21)             |
| Evaluation function $J$ at convergence point  | –             | 0.15 (0.14–0.15)             |

<sup>a</sup> The average values over the optimizations for 15 different data sets are shown, together with the minimum and maximum values in parentheses.

The theoretical transmission spectra with the spring-type interface model shown in Figs. 4(g)–(i) are calculated with the material properties identified for the experimental data set corresponding to Figs. 4(a)–(c). For relatively small incident angles, the measured transmission characteristics, including the bandgap formation, are well reproduced by both models. On the other hand, the spring-type interface model in Figs. 4(g)–(i) fails to reproduce the bandgaps observed experimentally for larger incident angles. To be more specific, the energy transmission spectra at some representative incident directions (i.e., along some horizontal crosslines of Fig. 4) are compared in Fig. 5.

In Fig. 5, the experimentally observed bandgaps are indicated by arrows. In the case of normal incidence in Fig. 5(e), the wave field in the laminate consists only of the pure longitudinal wave propagating in the layering direction, and it causes the constructive interference to form a bandgap at around 7.5 MHz in the experimental spectrum (solid line). This feature is well reproduced by the interleaf layer (dotted line) and the spring-type interface (dash-dotted line) models. When  $\theta = 8^\circ$  in Fig. 5(d), the quasi-longitudinal and two quasi-transverse waves propagate obliquely in the plies, which cause the interference in combination to form

a bandgap at around 7 MHz. When  $\theta = 15^\circ$  in Fig. 5(c) and  $\theta = 28^\circ$  in Fig. 5(b), the quasi-longitudinal wave becomes evanescent in the plies as the angles of incidence are above the critical angle at the water-ply interface (about  $10^\circ$ ). Hence, the bandgaps appear in these figures due to the interference of quasi-transverse waves. In Figs. 5(b)–(d), slight quantitative differences are seen between the experiment and the theory, but the locations of experimentally observed bandgaps are still favorably reproduced by both models. When the angle of incidence further increases to  $\theta = 45^\circ$  in Fig. 5(a), which exceeds the critical angle of the faster quasi-transverse wave (about  $36^\circ$ ), only the slower quasi-transverse wave can propagate in the plies and cause the interference to form a bandgap at around 7 MHz in the experimental spectrum. For more details on the wave transmission and bandgap characteristics in unidirectional composite laminates, refer to Refs. [30], [60]. Most importantly, the spring-type interface model exhibits monotonic dependence on the frequency without any sign of the bandgap formation in Fig. 5(a). In contrast, the interleaf layer model successfully reproduces the appearance of bandgap at 7 MHz in a reasonable manner. Note here that when computing the evaluation function defined in Eq. (3) in the optimization procedure, the values of energy transmission coefficient less than  $10^{-3}$  have been rounded up to  $10^{-3}$  to suppress the influence of vanishingly small transmission coefficients as mentioned in Section 2.2, which may be responsible for the quantitative difference in bandgap depth between the experiment and the interleaf layer model seen in Fig. 5(a).

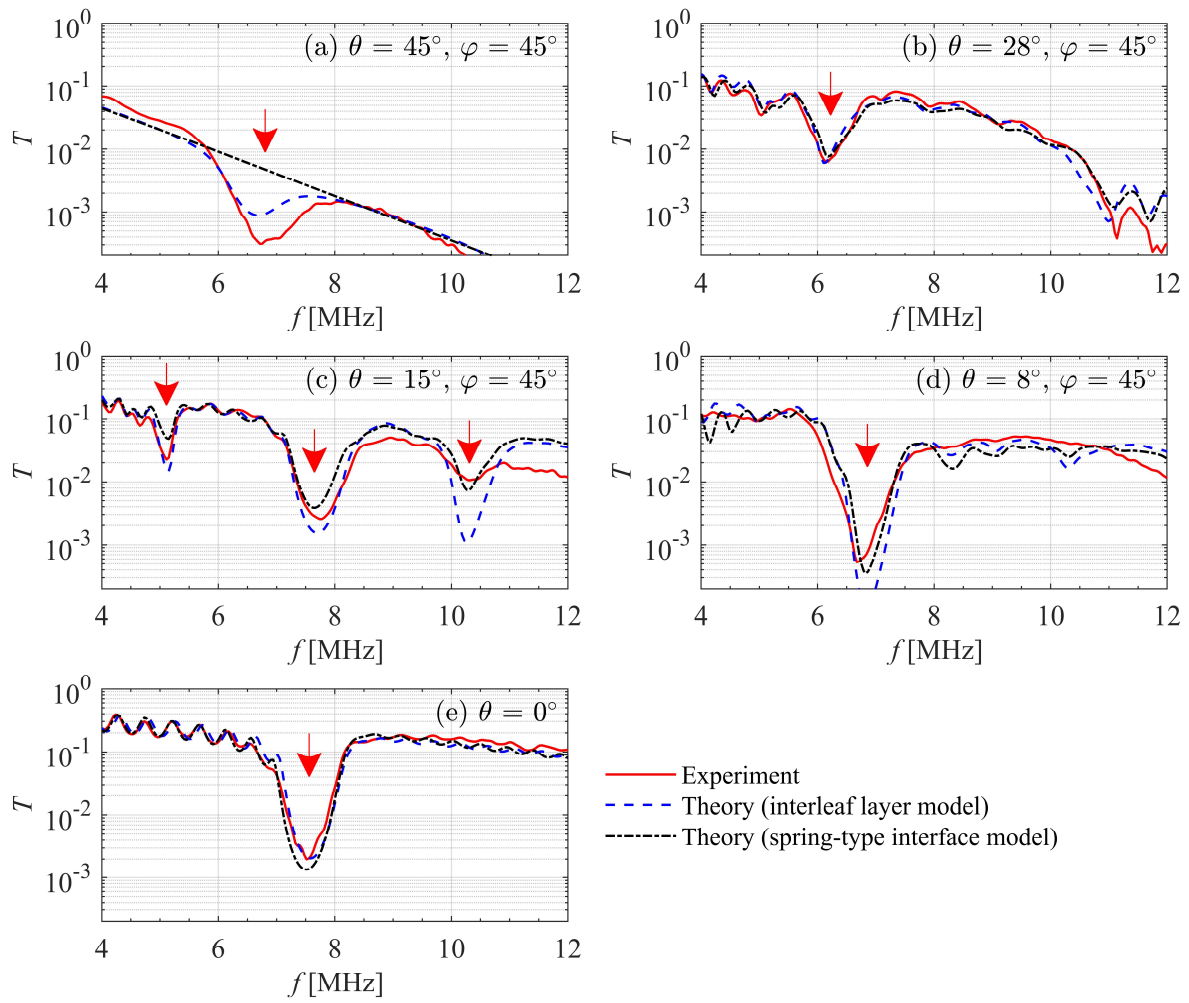


Fig. 5 Comparison of the experimental and theoretical energy transmission spectra (horizontal cross sections of Fig. 4) for some representative incident directions. Arrows indicate the locations of experimentally observed bandgaps.

### 3.2.2. Applicability to non-interleaved composite laminates

In the authors' previous works [27–32], the spring-type interlaminar interface model was applied to evaluate the ply and interfacial stiffnesses of conventional non-interleaved composite laminate specimens. These specimens had interlaminar resin-rich regions whose thicknesses were a few micrometers and considerably small compared to the interleaf layers in the composite laminate investigated in this study. The evaluation procedure in Section 2.2 has also been applied to the experimental transmission spectra measured for a conventional non-interleaved composite laminate specimen, but the optimization has failed to converge. This may be partly due to the complicated matrix-fiber geometry of the interlaminar zones, which makes it

difficult to assign a definite thickness to such zones, but their small thickness may also be a cause of convergence difficulty.

To further examine the above observation, the evaluation function  $J(\mathbf{w})$  in Eq. (3) is computed for artificial transmission coefficients of 16-ply unidirectional composite laminates with different interleaf layer thicknesses. They are calculated beforehand using the interleaf layer model in Eq. (4) with the other material properties fixed to the ones identified for a specific experimental data set in Section 3.1: specifically, they are set as  $E^{l,o} = 4.4$  GPa,  $\rho^{l,o} = 1.38 \times 10^3$  kg/m<sup>3</sup>,  $\nu^{l,o} = 0.37$ ,  $C^{p,o}_{11} = 166 - 6.9i$  GPa,  $C^{p,o}_{12} = 7.9 - 0.43i$  GPa,  $C^{p,o}_{22} = 15.0 - 0.43i$  GPa,  $C^{p,o}_{44} = 3.5 - 0.13i$  GPa,  $C^{p,o}_{66} = 6.9 - 0.24i$  GPa,  $h^{p,o} = 0.170$  mm,  $\rho^{p,o} = 1.60 \times 10^3$  kg/m<sup>3</sup>, where the superscript “o” has been introduced to denote the properties used for the preparation of artificial data sets. The ranges and the numbers of frequencies and incident directions used for the computation of  $J(\mathbf{w})$  are the same as before for the application to experimental data sets (see Section 2.3).

Figure 6 shows the intersections of the resulting 14-dimensional evaluation function  $J(\mathbf{w})$  and the  $E^l$ – $h^l$  plane containing its minimum (i.e., the point given by  $\mathbf{w}^o$  and marked with a cross in Fig. 6) when the original interleaf layer thickness is  $h^{l,o} = 3, 10$ , and  $20$   $\mu\text{m}$ . It is found that the distribution of  $J$  tends to become flatter in the vicinity of its minimum as  $h^{l,o}$  decreases. This implies that the minimization procedure of evaluation function for composite laminates with thinner interleaf layers can be more susceptible to the presence of noise as well as the random fluctuation of interleaf thickness, which may explain the above-mentioned unsuccessful optimization for the conventional non-interleaved composite laminate.

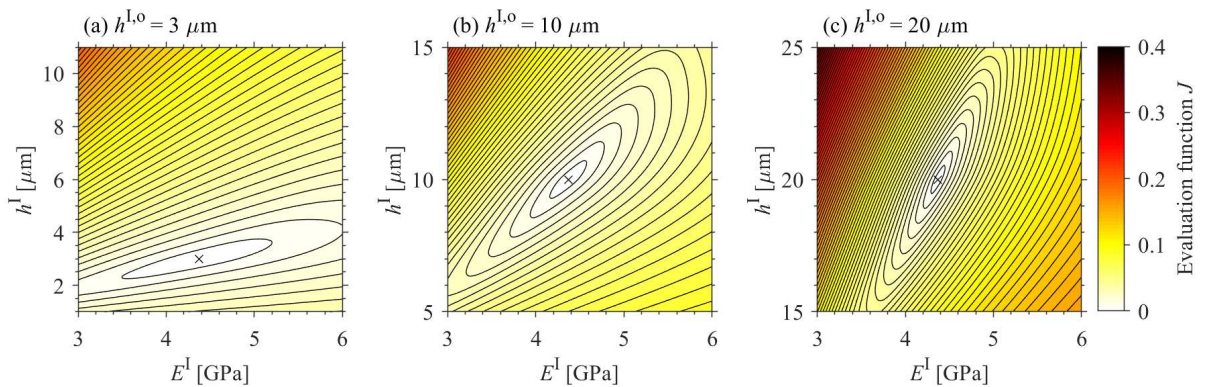


Fig. 6  $E^l$ – $h^l$  cross sections passing through the minimum of the evaluation function computed for artificial transmission coefficient data sets of 16-ply unidirectional composite laminates with interleaf layer thicknesses of (a)  $h^{l,o} = 3 \mu\text{m}$ , (b)  $h^{l,o} = 10 \mu\text{m}$ , and (c)  $h^{l,o} = 20 \mu\text{m}$ . The locations of minima (i.e.,  $h^l = h^{l,o}$  and  $E^l = E^{l,o} = 4.4$  GPa used for generating artificial data sets) are indicated by crosses. The contour lines

correspond to  $J = 0.005m$ , where  $m$  is a non-negative integer.

In Fig. 6, it is commonly observed for the three interleaf layer thicknesses that the contour lines of  $J$  are elongated around its minimum in a certain well-defined direction. This trend is seen in Fig. 6(c) most clearly but can still be noticeable in Fig. 6(a). This implies that the ratio between  $E^I$  and  $h^I$  (which corresponds essentially to the interfacial stiffnesses) can robustly be identified even in the case of thin interleaf layers, suggesting that the use of the spring-type interface model is more suitable for composite laminates with thin interleaf layers. To support this suggestion, the optimization procedure modified in Section 3.2.1 for the spring-type interlaminar interface model is applied to the three artificial data sets used above in Fig. 6. The energy transmission spectra calculated by the spring-type interface model with the so-obtained material properties are then compared in Fig. 7 to their counterparts in the original data sets for some representative incident directions. As already noted in Fig. 5, the spring-type interface model fails to reproduce the transmission characteristics of composite laminates with thicker interleaf layers for large angles of incidence, as can be seen in Fig. 7(c) for  $h^{I,0} = 20 \mu\text{m}$  and  $\theta = 45^\circ$ . This limitation is gradually alleviated with decreasing  $h^{I,0}$ , and the spring-type interface model is in good agreement with the interleaf layer model even in the large angles of incidence when  $h^{I,0} = 3 \mu\text{m}$  in Figs. 7(a), (d), and (g).

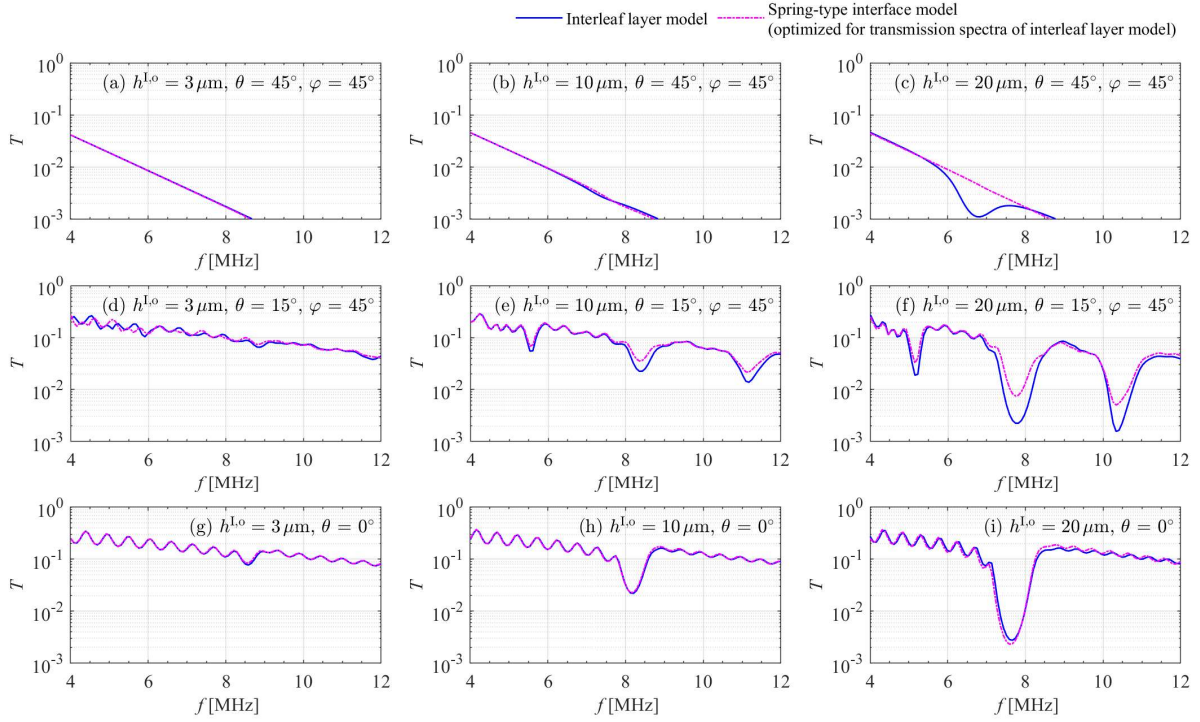


Fig. 7 Comparison of the theoretical energy transmission spectra of the interleaf layer model and the ones calculated by the spring-type interface model with the material properties identified for the transmission coefficient data sets of the interleaf layer model. The left, middle, and right columns correspond to the original interleaf layer thicknesses of  $h^{L_o} = 3, 10$ , and  $20 \mu\text{m}$ , respectively. The bottom, middle, and top rows correspond to the different angles of incidence of  $\theta = 0^\circ, 15^\circ$ , and  $45^\circ$ , respectively.

Therefore, the spring-type interlaminar interface model is more robust for the property characterization of conventional composite laminates with a few micron-thick interlaminar resin-rich zones as in the previous studies [27–32]. On the other hand, the finite-thickness interleaf layer model as employed in the present study is more suitable for modeling interleaved composite laminates with the interleaf layer thickness of 20–30 microns, since the experimental transmission spectra are more accurately reproduced with this model, as discussed above in Figs. 4 and 5.

### 3.3. Note on the viscoelastic property of interleaf layers

The results presented in Section 3.1 have been obtained assuming that the plies are solely responsible for the wave attenuation given the difficulty in distinguishing between the influence of ply and interleaf viscoelastic nature on the transmission spectrum, as mentioned in Section 2.1. On the other hand, it may also

be of interest to see the results of the opposite case where the wave attenuation is solely ascribed to the interleaf layers. In this regard, the evaluation procedure in Section 2 is performed again here, except that the plies are now assumed to be elastic (i.e.,  $\eta^p_{IJ} = 0$ ) while the interleaf layers are assumed to be viscoelastic with complex Young's modulus  $E^l$  and Poisson's ratio  $\nu^l$ . The present evaluation is applied to one of the experimental data sets used in Section 3.1, and the obtained results are shown in Table 4.

The ply and interleaf layer properties in Table 4, except for the imaginary parts of  $E^l$  and  $\nu^l$ , are reasonably compared with those in Table 2. Although there is no literature available for the validation of imaginary parts of  $E^l$  and  $\nu^l$  identified in Table 4, the particularly high value of  $\text{Im}(E^l)$  should overestimate the actual loss property of the interleaf layers considerably due to the neglect of the viscoelastic nature of plies. The quantitative evaluation of the viscoelastic properties of both plies and interleaf layers requires additional information and remains as a subject of future study.

Table 4 Evaluation results for the interleaved composite laminate specimen when the plies and interleaf layers are assumed to be elastic and viscoelastic, respectively.

|                                                                | Initial guess  | Evaluated value |
|----------------------------------------------------------------|----------------|-----------------|
| <b>Interleaf layer</b>                                         |                |                 |
| Thickness $h^l$ [ $\mu\text{m}$ ]                              | 15.0           | 24.6            |
| Density $\rho^l$ [ $\times 10^3 \text{ kg/m}^3$ ]              | 1.20           | 1.37            |
| Complex Young's modulus $E^l$ [GPa]                            | $5.0 - 0.5i$   | $5.3 - 1.7i$    |
| Complex Poisson's ratio $\nu^l$                                | $0.40 - 0.05i$ | $0.36 - 0.03i$  |
| <b>Ply</b>                                                     |                |                 |
| Elastic constants $D^p_{IJ}$ [GPa]                             |                |                 |
| $D^p_{11}$                                                     | 160            | 176             |
| $D^p_{12}$                                                     | 8.0            | 8.5             |
| $D^p_{22}$                                                     | 14.0           | 15.3            |
| $D^p_{44}$                                                     | 4.0            | 3.7             |
| $D^p_{66}$                                                     | 7.0            | 7.0             |
| Thickness $h^p$ [mm] <sup>a</sup>                              | —              | 0.170           |
| Density $\rho^p$ [ $\times 10^3 \text{ kg/m}^3$ ] <sup>b</sup> | —              | 1.60            |
| Evaluation function $J$ at convergence point                   | —              | 0.19            |

<sup>a</sup> The ply thickness is obtained from Eq. (1) with the identified properties.

<sup>b</sup> The ply density is obtained from Eq. (2) with the identified properties.

### 3.4. Transmission spectra of a quasi-isotropic composite laminate

Finally, it is examined if the ply and interleaf layer properties identified above for the unidirectional laminate can reproduce the ultrasonic wave transmission characteristics for other layup configurations of interleaved composite laminates. To this purpose, the energy transmission spectra are calculated for the quasi-isotropic laminate consisting of composite plies and interleaf layers assuming the stacking sequence  $[-45/0/+45/90]_{s2}$  in accordance with the specimen used in the previous study [60] (the stacking sequence of this specimen was noted as  $[-45/0/+45/90]_{2s}$  in Ref. [60] but should be corrected as  $[-45/0/+45/90]_{s2}$ ). This specimen was fabricated using the same prepregs and interleaf resin layers as the unidirectional specimen examined in this study. Therefore, the properties of the plies and interleaf layers evaluated for the unidirectional laminate in Table 2, except for the interleaf layer density, are used to model those for the quasi-isotropic laminate. The ply thickness is assumed to be the same as that identified for the unidirectional laminate (i.e.,  $h^p = 0.170$  mm), and the interleaf layer thickness and density are set as  $h^l = 22.8$   $\mu\text{m}$  and  $\rho^l = 1.32 \times 10^3$  kg/m<sup>3</sup>, respectively, to comply with the measured thickness  $h^l = 3.06$  mm and average density  $\rho^l = 1.56 \times 10^3$  kg/m<sup>3</sup> of the quasi-isotropic laminate specimen. It is noted that these values of interleaf layer thickness and density are within the range obtained in the micrographic observation and the one reported in the literature, respectively (see Section 3.1).

The energy transmission spectra of the ultrasonic longitudinal wave for the quasi-isotropic laminate were measured in the manner described in Section 2.3. The experimental results are shown in Figs. 8(a)–(c) for different angles of the plane of incidence, while the theoretical results based on the finite-thickness interleaf layer model are plotted in Figs. 8(d)–(f). As mentioned in Section 3.1 for the unidirectional laminate, the wave transmission and bandgap formation in the present quasi-isotropic laminate were analyzed in Ref. [60] using the spring-type interlaminar interface model. In this paper, the theoretical transmission spectra thus obtained are also shown in Figs. 8(g)–(i) to compare with the interleaf layer model. They are calculated using the ply and interfacial stiffnesses evaluated for the unidirectional laminate in Table 3 with the ply thickness and density given from the above-mentioned laminate thickness and average density as  $h^p (= h^l/N) = 0.191$  mm and  $\rho^p (= \rho^l) = 1.56 \times 10^3$  kg/m<sup>3</sup>.

It can be seen that the experimental transmission spectra are well reproduced by both models in Fig. 8, thereby again indicating the validity of the ply and interleaf layer properties evaluated in the present study. Compared to the transmission spectra of the unidirectional laminate shown in Fig. 4, those of the quasi-isotropic laminate in Fig. 8 show very low transmissivity for large angles of incidence of about  $\theta > 30^\circ$  due to the significant internal reflection in the multidirectional laminate for obliquely propagating waves. This loss of transmission somewhat obscures the difference between the predictions of the two models highlighted for the unidirectional laminate at large angles of incidence, as mentioned in Section 3.2.1. It is, nevertheless, worth noting that the interleaf layer model still provides a quantitatively better fit to the experimental spectra of the quasi-isotropic laminate than the spring-type interface model, as the resulting evaluation function between the experimental and theoretical energy transmission coefficients is  $J = 0.13$  for the interleaf layer model and  $J = 0.18$  for the spring-type interface model. This feature is consistent with the case of the unidirectional laminate and also demonstrated in the direct comparison of experimental and theoretical transmission spectra shown in Fig. 9 for some representative incident directions.

In the case of normal incidence in Fig. 9(e), the experimental transmission spectrum, including the bandgap formation at around 7.5 MHz indicated by a solid arrow, is very similar to the one in the unidirectional laminate in Fig. 5(e), as the wave field inside the quasi-isotropic laminate is still governed by the pure longitudinal wave propagating perpendicularly to the ply fiber directions. Accordingly, the experimental spectrum is well reproduced by the interleaf layer and the spring-type interface models. For larger angles of incidence, the propagation characteristics (i.e., wave velocity and polarization) of quasi-longitudinal and quasi-transverse waves in the plies are influenced by their fiber directions. As a result, compared to the unidirectional laminate in Figs. 5(b)–(d), quite different spectra containing several bandgaps are obtained in Figs. 9(b)–(d). The more complicated bandgap formation in these figures, still caused by the constructive interference of quasi-longitudinal and quasi-transverse waves, can be analyzed by calculating the energy flux density of partial waves in each ply constituting the quasi-isotropic laminate or using the Floquet–Bloch wave theory; see Refs. [31] and [60] for more details. The general trends of experimental spectra in Figs. 9(b)–(d) are reasonably reproduced by the two models. In particular, the interleaf layer model better fits the experiment, as can be seen around the bandgaps indicated by solid arrows as well as the locations indicated by dotted arrows. When  $\theta = 45^\circ$ , only the slower quasi-transverse wave can propagate in

the plies constituting the quasi-isotropic laminate due to the evanescent nature of quasi-longitudinal and faster quasi-transverse waves. Thus, the experimental transmission spectrum in Fig. 9(a) exhibits a simpler frequency dependence with very low transmissivity, and it is again better reproduced by the interleaf layer model.

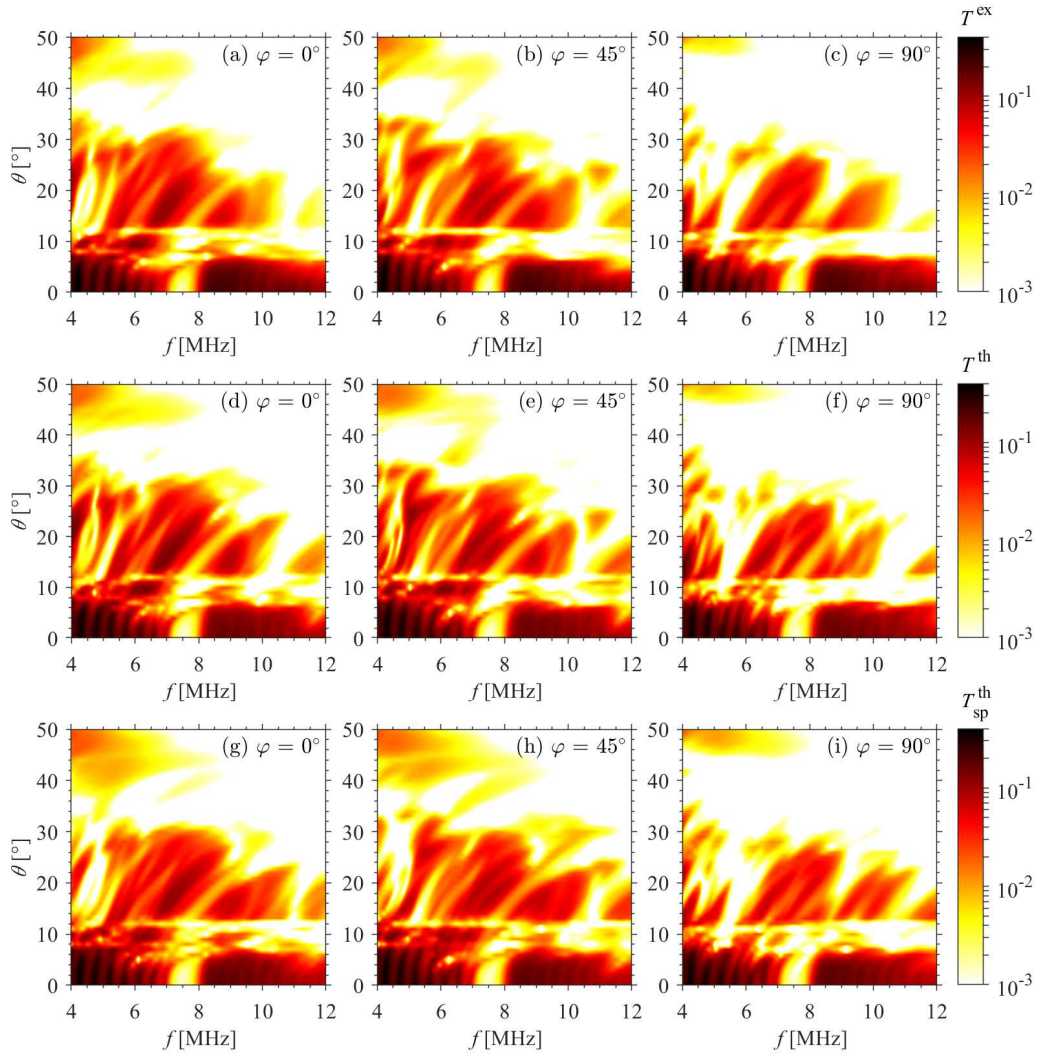


Fig. 8 Distributions of the energy transmission coefficient of the 16-ply quasi-isotropic composite laminate in the plane of the frequency  $f$  and the angle of incidence  $\theta$  when the angle of the plane of incidence is  $\varphi = 0^\circ, 45^\circ$ , and  $90^\circ$ . (a)–(c) Experiment. (d)–(f) Theoretical analysis with the interleaf layer model. (g)–(i) Theoretical analysis with the spring-type interlaminar interface model.

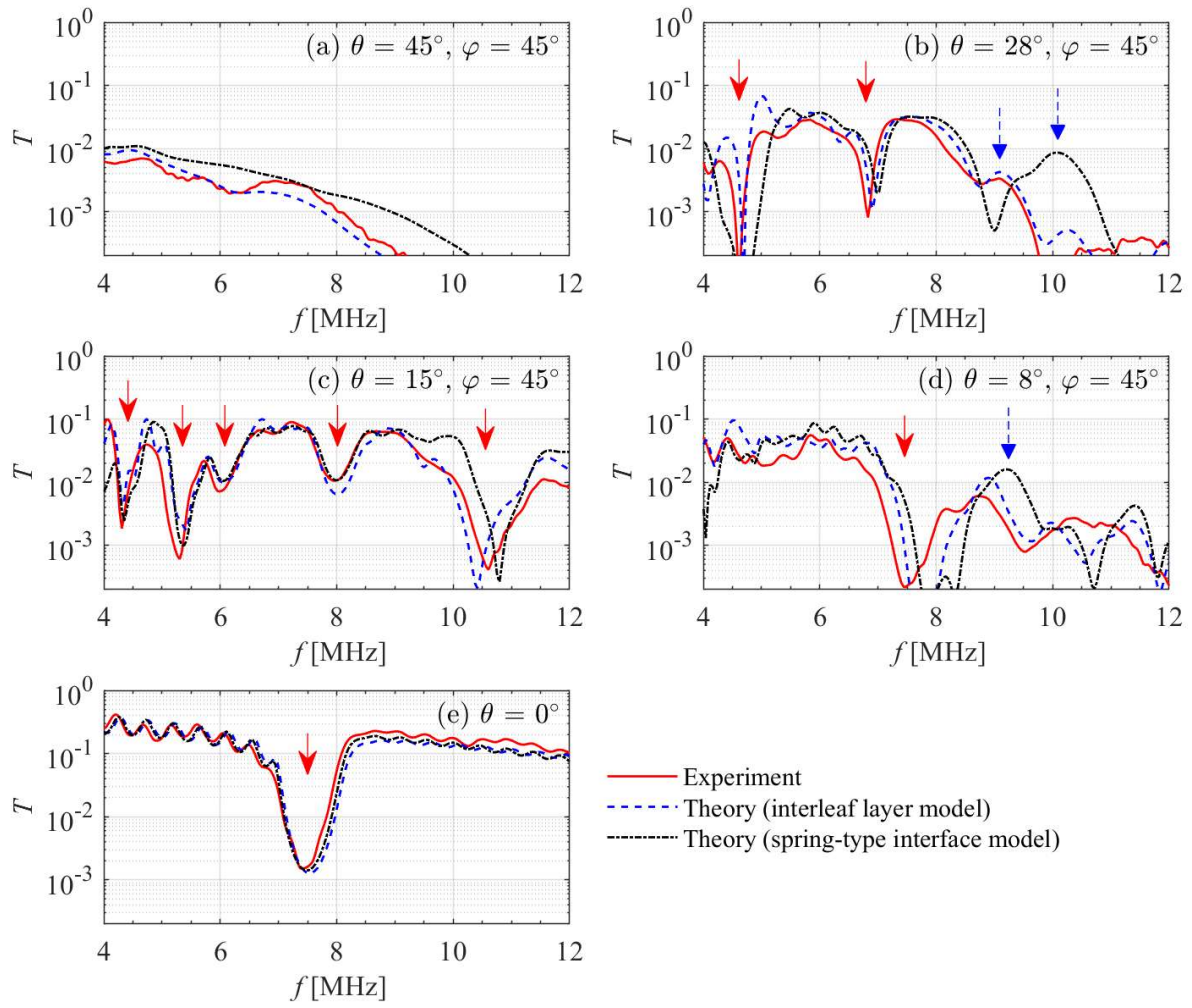


Fig. 9 Comparison of the experimental and theoretical energy transmission spectra for the 16-ply quasi-isotropic composite laminate (horizontal cross sections of Fig. 8) for some representative incident directions. Solid arrows indicate the locations of experimentally observed bandgaps. Dotted arrows highlight the locations where the interleaf layer model better fits the experiment than the spring-type interface model.

#### 4. Conclusions

In this paper, the ply and interleaf layer properties of an interleaved composite laminate have been evaluated nondestructively by best fitting the theoretical energy transmission coefficients of longitudinal waves through the laminate to the experimental ones. In contrast to the existing works, most of which addressed the identification of average properties of composite laminates in the layering direction, the ply and interlayer properties can be identified separately by the present evaluation procedure. Hence, the results obtained in this study can be helpful when the quality of interleaf layers, which is of great importance for the structural integrity of interleaved composite laminates, is to be evaluated nondestructively.

The evaluation results based on the finite-thickness interleaf layer model have been compared with those based on the mass-less and null-thickness spring-type interlaminar interface model adopted in the previous works. By doing so, it has been shown that the interleaf layer model is more suitable for modeling interleaved composite laminates with an interleaf layer thickness of 20–30 microns since it reproduces the experimental transmission spectra more accurately, while the spring-type interlaminar interface model is more robust for the property characterization of conventional composite laminates with a few micron-thick interlaminar resin-rich zones. In particular, it has been demonstrated that the bandgaps observed experimentally for relatively large incident angles of  $\theta > 30^\circ$  can be reproduced in a clearer manner by the present interleaf layer model. The ply and interleaf layer properties determined for the unidirectional laminate have been shown to reproduce the transmission spectra of the quasi-isotropic laminate with good accuracy.

The present study has assumed that either the plies or interleaf layers are solely responsible for the wave attenuation, thus providing the imaginary parts of the stiffness for either one of them. To evaluate the viscoelastic properties of both plies and interleaf layers, the present evaluation procedure should be extended to incorporate additional information to work well without the above assumption, which is left for future study.

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