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Microwires enabled metacomposites towards microwave applications

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Abstract:

The work describes the microwave behavior of polymer composites containing parallel Febased and continuous/short-cut Co-based microwire arrays. A magnetic field-tunable metacomposite feature has been identified in the hybrid microwires composite containing 3 mm spaced Co-based wires and confirmed by the presence of transmission windows in the frequency band of 1 to 3.5 GHz. The magnetically tuned redshift-blueshift in the transmission window is due to the competing dynamic interactions between the different wires and the ferromagnetic resonance of the Fe-based microwires. When the Co-based inter-wire spacing is increased to 10 mm, dual-band transmission windows in the 1.5-3.5 GHz and 9-17 GHz bandwidths were observed. These transmission windows are likely induced by the ferromagnetic resonance of Fe-based wires and the long range dipolar resonance arising between Fe-Co wire couples. The hybridization of parallel Fe-based and short Co-based wires in the composites leads to a significant enhancement of the transmission window in the 1 to 6 GHz band due to the band-gap nature of the Co-based wires. The hybrid metacomposites containing microwires seem attractive in radio frequency identification application.

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1. Introduction

Left-handed metamaterials have stimulated tremendous fundamental and practical interests in recent years owing to their peculiar electromagnetic (EM) properties and potential engineering applications in a wide range of operating frequencies. Because of their artificial double negative (DNG) properties (i.e., negative permittivity and permeability), several metamaterial concepts and potential applications have been developed including invisibility cloaking,¹ perfect metamaterial absorbers,² perfect lenses,³ mechanical metamaterials^{4,5} All these applications have originated from the first experimental demonstration of the performance of split ring resonators (SRRs) and metallic wires.⁶ To date some significant efforts have been devoted to design low loss, boardband^{7,8} and tunable metamaterials⁹, and the integration of mechanical and EM metamaterials¹⁰ in order to maximize or optimize their EM performance. However, the conventional design of left-handed metamaterials involves the use of the overall dielectric and magnetic response of materials' meso-structures with less focus on the intrinsic properties of the metamaterials' building blocks. Moreover, the increasing complexity of the topologies and designs associated to modern metamaterials with broadband capability leads to the increasing use of nano-materials synthesis technologies, with added higher capital costs involved.¹¹

Luo et al. have recently developed a metacomposite with the DNG characteristics, which is made from continuous Fe-based amorphous ferromagnetic microwire arrays and aerospace-grade polymer-based composites.¹² This design provides a much simpler left-handed structure and shows that DNG features can be embedded in a 'real' material from an engineering standpoint, rather than a merely functionalized structure. On the other hand, Co-based microwires feature a distinct giant magnetostriction,¹³ this makes the EM performance

of Co-based microwires to be easily modulated by external magnetic fields or mechanical stresses.^{14,15} It is plausible to incorporate the Co-based microwire arrays into the Fe-based wire/polymer metacomposite system with the aim to obtain a metacomposite with significant external stimuli-controlled behavior. Therefore, it is worth investigating the underlying physics behind the proposed hybrid microwire composites when they interact with incident microwaves. In a recent work, Luo et al. have introduced the concept of critical wire spacing in composites containing Fe-based wire arrays, and have explicitly demonstrated that the wire spacing plays an import role in the metacomposite features.¹² It is therefore of much interest to further explore how the Co-based wire spacing would influence the microwave performance of the hybrid wire composites and to understand how the interactive magnetic resonance could be realized between the Co- and Fe-based wire arrays by taking into account their different wire alignment, unique EM properties and dimensions. All these information would be invaluable to expand the DNG operating frequencies or to enhance the microwave absorption.

In the present work, we hybridize short-cut and continuous Co-based wire arrays respectively into the Fe-based microwire metacomposite and investigate the microwave properties. Key findings of this work are summarized as follows. (i) For the continuous wires composites with high concentration of Co-based wires, the left-handed microwave behavior is enhanced by the emerging transmission windows when the external field is larger than 300 A/m. The transmission window peak shifts towards the red with field increasing up to 600 A/m due to the strong long range dipolar resonance (LRDR) between wires, and subsequently shifts to the blue with fields larger than 3000 A/m, suggesting a field-tuned metacomposite behavior; (ii) a dual-band metacomposite characteristic (without external fields) is revealed in the continuous hybrid composites in absence of external bias when the spacing of Co-based array is reduced to 10 mm; (iii) a transmission-window/band-gap feature is identified in the

hybridized composite system that contains short-cut Co-based wires and continuous Fe-based wire arrays.

2. Material and methods

2.1 Material processing

Amorphous glass-coated ferromagnetic microwires Fe77Si10B10C3 (total diameter of 20 µm, glass coat thickness of 1.7 µm) and Fe₄Co_{68.7}Ni₁B₁₃Si₁₁Mo_{2.3} (total diameter of 20.77 μm, glass coat thickness of 2.4 μm) were manufactured using the modified Taylor-Ulitovskiy technique¹⁶ and supplied by TAMAG, Spain. To investigate the influence of wire topological arrangement on the microwave behavior of the composites, the Co-based and Fe-based microwires were hybridized into aerospace-grade 913 E-glass prepregs in two topological arrangements, i.e., parallel Co-based and parallel Fe-based wire arrays (Fig. 1(a)) and shortcut Co-based and continuous orthogonal Fe-based wire array (Fig. 1(b)). To consider the effect of wire spacing on the EM properties of the composites in the continuous hybrid structure the spacing of the Co-based wire array is arranged as 10 mm and 3 mm, respectively. A standard polymer curing procedure was then followed after embedding the wires. The prepregs were heated at a rate of 2.4 °C/min to 125 °C and the temperature was kept for 120 min before naturally cooling down to room temperature. The pressure was elevated to 0.62 MPa at a rate of 0.07 MPa/min and remained for 270 min before decreasing at 0.07 MPa/min per minute.¹⁷ All the resultant composite samples have an in-plane size of 500×500 mm² and thickness of 1 mm. It should be noted that the Fe-based and Co-based microwire arrays were arranged in separate prepreg layers to avoid the large reflection loss induced by the physical microwire contact.¹² Besides, the in-plane spacing between the Co-based and Fe-based microwire arrays was intentionally mismatched by approximately 1 mm to minimize the undesirably high reflection caused by the wire superposition. In comparison, polymer composites containing continuous Co- or Fe-based microwire array, orthogonal Fe-based

wire array and short-cut Co-based wire array with same wire spacing as in the hybrid wire composites in Fig. 1 were also fabricated by the same experimental protocol.

2.2 Microwave characterization

The microwave behavior of the microwire composites was examined using free-space measurements in the frequency band of 0.9 to 17 GHz with the electrical component E_k parallel to the glass fiber direction (Fig. 1). To track the magnetic field tunable properties, an additional external dc magnetic bias up to 3000 A/m was applied along the glass fiber direction. A brief description is presented herein in terms of how the external field was applied. The wire-composite specimens were firmly fixed in a wood frame to ensure a uniform magnetic field distribution on the specimen surface. A planar coil was winded on the frame to make the specimen be conveniently tuned by a high magnetic field up to 3000 A/m. The frame included with specimen was installed in the center of an anechoic chamber made of plywood walls and covered inside by a microwave absorber to minimize the signal loss and neutralize the effect of noises. Detailed information of the microwave measuring rig can be found elsewhere.^{18,19} The effective permittivity was extracted via the obtained *S*-parameters by an implanted computer program: Reflection/Transmission Epsilon Fast Model. Further details of this experimental setup can be found in Ref [20].



FIG. 1 (Color online) Schematic illustration of the hybridization of (a) continuous parallel Febased microwire array plus continuous Co-based microwire array and (b) orthogonal Fe-based microwire array plus short-cut Co-based microwire array.

3. Results and discussion

3.1 Continuous hybrid composites containing 3 mm spaced Co-based wires

The subfigures (a) to (i) of Figure 2 describe the transmission, reflection and absorption coefficients of the continuous (10 mm Fe-based, 3 mm Co-based) composites in the frequency band of 1 to 6 GHz. It is clear that the EM performance of the composite with the Fe-based microwires can be tuned with the presence of significant external fields larger

than 1000 A/m (Figs. 2(b), (e), (h)). A transmission dip (Fig. 2(b)) associated with an absorption peak (Fig. 2(h)) is identified approximately at 2.5 GHz, which can be attributed to the ferromagnetic resonance (FMR),¹² as evidenced by the blueshift of resonance peaks according to Kittel's equation.²¹ The absorption intensity reduces in the vicinity of the FMR frequency with increasing levels of magnetic field. For the Fe-based microwires there is a trade-off between the effective permeability (limited values but yet not unity in this case) and the ferromagnetic resonance frequency as derived from Snoek's law:

$$(\mu - 1)f_{\rm FMR} = \frac{2}{3}\gamma 4\pi M_{\rm s},\tag{1}$$

Where γ , M_s , f_{FMR} and μ denote the gyromagnetic factor, saturation magnetization, the FMR frequency and the permeability, respectively.²² The observed blueshift frequency results in the reduction of the effective permeability and therefore of the microwave absorption. In contrast, the field-tunability of the composite containing the Co-based wires is saturated at rather high fields up to 600 A/m (Figs. 2(c), (f), (i)), indicating a degradation of the magnetic softness of the wires. The degradation may be explained by the damage of the circumferential anisotropy of the microwires induced by the high temperature and pressure during the composite curing process, in view of their very significant heat and stress sensitivity.^{23,24} In particular, the absorption of the composite containing the Co-based wires increases more significantly with increasing external field compared to Fe-based wires (Fig. 2(i)). This is due to the interaction effect of the long-range dipolar resonance (LRDR) between the Co-based wires because the 3 mm wire spacing is below the critical spacing.^{12,25} Interestingly, the LRDR peaks in the absorption spectrum shift to the red with increasing fields (Fig. 2(i)). With increasing magnetic fields, the skin depth of the Co-based wires at the LRDR frequency is significantly decreased due to the increased μ according to Eq. 2:

$$\delta_{\rm res} = \sqrt{\frac{\rho}{\pi f_{LR}\mu}},\tag{2}$$

Where ρ and f_{LR} are electrical resistivity and the long range dipolar resonance frequency, respectively.²⁶ The decrease of the skin depth leads to high eddy current losses and the resonance frequency is then reduced to compensate this loss. It should be noted that the 10 mm spacing in the Fe-based microwire composite is too large to induce this type of resonance and the shift towards the blue is dictated by their FMR.¹²



FIG. 2 (Color online) Frequency plots of the transmission coefficients, S₂₁, of the composite samples containing (a) hybrid wire arrays with 3 mm spaced Co-based wires, (b) pure Fe-based wires and (c) pure Co-based wires; the reflection coefficients, S₁₁, of composite samples containing (d) hybrid wire arrays with 3 mm spaced Co-based wires, (e) pure Fe-based wires and (f) pure Co-based wires; the absorption coefficients of composites containing (g) hybrid wire arrays with 3 mm spaced Co-based wires, (h) pure Fe-based wires and (i) pure Co-based wires.

It should be also noted that the composites containing the single Fe-based wire array exhibit no DNG characteristic due to the fairly low plasma frequency of 1.4 GHz¹² evident from Fig. 3(a), which is caused by the longitudinal domain structure of the Fe-based wires.²⁷ Although the plasma frequency can be enhanced in the Co-based wires contained composites spaced by 3 mm, the overall high reflection loss suppresses the DNG features (Fig. 2(f)). We expect a solution to this problem by introducing the Co-based microwires into the Fe-based wire composite. A lower reflection loss is indeed shown by the hybrid metacomposites due to the improved impedance match (Fig. 2(d)). Quite remarkably, a transmission window can be observed in the frequency band between 1 GHz and 3.5 GHz for the hybridized wire array composite when the fields are larger than 300 A/m. This suggests that an abnormal transmission dispersion is established in the continuous hybridized Fe-/Co-based wire composite system, which is quite distinct from the previously reported natural transmission windows independent of magnetic field and appears to be controlled by a critical spacing in the single Fe-based wires composites.¹¹ It is worth emphasizing that the existence of this abnormal transmission dispersion does not guarantee that our hybrid microwire-composite is a DNG media, as it may also possess a double positive media behavior. This point has been validated from experimental microwave tests featured in some recent studies.^{15, 28} Further, with the increase of the external field, the transmission window peak experiences a redshiftblueshift change (Fig. 2(a)) because the long range dipolar resonance, due to the dynamic interactions between wire couples,^{12,29} dominates at low magnetic fields (below 600 A/m), while the FMR of the Fe-based wires prevails at higher fields than 600 A/m.

Figure 3 shows the effective permittivity of the pure Fe-based wire composite and the continuous hybridized composites with and without external magnetic fields (up to 3000 A/m). Previous studies have shown that the permittivity of composites containing microwires

can be modulated under different magnetic fields, and the undelaying physics has been thoroughly investigated.^{30,31} In this work we argue that for specific wire topology, composites containing wire arrays show a DNG metamaterial property and their permittivity is tunable with negative values. In the presence of external fields, the variation of the permittivity of the composite containing the single Fe-based wire array spaced by 10 mm is rather limited due to the formation of nanocrystallites on the microwires after the curing process, which degrades the field-tunable properties to a large extent.^{12,23,32} (Fig. 3(a)). Moreover, the f_p of the Febased wires is heavily reduced in contrast to the theoretical value.³³ The dielectric response of the continuous microwire composite system can be described as the dilute plasmonic behavior and interpreted via the following expression:

$$f_p = \sqrt{\frac{c^2}{2\pi b^2 ln\left(\frac{b}{a}\right)}},\tag{3}$$

Where $c=3\times10^8$ m/s, b and a denote the vacuum light velocity, wire spacing and diameter, respectively. To provide an explanation for the observed degraded value of f_p ,¹² we have introduced a term related to the effective diameter a_{eff} to consider the circumferential domain volume of the microwires, which gives the actual contribution to their dielectric performance.³³ For the Fe-based wires a_{eff} is much smaller than their actual diameter because their circular domain only occupies a tiny portion of the whole domain volume. Hence f_p is heavily reduced (Fig. 3(a)) and Eq. 3 should be modified as:

$$f_p^2 = \frac{c^2}{2\pi b^2 ln\left(\frac{b}{a_{eff}}\right)}.$$
(4)



FIG. 3 (Color online) Frequency plots of the real part of effective permittivity, ε', of the composites containing (a) continuous Fe-based wire array spaced by 10 mm and (b) hybrid arrays with 3 mm spaced Co-based wires in the presence of magnetic fields up to 3000 A/m.

For the hybridized microwire array composite, the permittivity magnitude shows a decrease-increase trend with the increase of magnetic fields (Fig. 3(b)). As the external fields are less than 1000 A/m the absolute value of the permittivity decreases because of the improved impedance match (Z), which subsequently compromises the effective ε :

$$Z = \sqrt{\frac{\mu}{\varepsilon}}.$$
(5)

By further increasing the magnetic bias to 3000 A/m the dispersion of the permittivity becomes stronger due to the increased μ arising from the FMR of the Fe-based microwires, therefore creating circular fields that greatly enhance the dielectric response of microwires. To this end the value of f_p for the hybridized wire composites can also be tuned by external fields due to the additional magnetically enhanced circular field and therefore can be significantly increased up to approximately 4.5 GHz (Fig. 3(b)). Another influential factor impacting on the dielectric performance is the wire-wire interaction. It is believed that the inter-wire interactions due to LRDR with the presence of external fields among the Co-Co and Co-Fe wire couples after the hybridization have provided an essential offset to the limited a_{eff} in the continuous hybrid composite system, thus enhancing the value of f_p to some degree. This feature can be justified when taking into account the fact that the circumferential magnetic response excited by the external fields of a single microwire is beneficial to expanding a_{eff} of its neighbor wires.¹²

It should be also noted that the measurement of the magnetic permeability is problematic when using a conventional Nicolson-Ross-Weir (NRW) method in a free-space measurement, which is normally adopted for traditional highly lossy system SRRs metamaterials.³⁴ Our microwire composite, although it generates lossy microwave signatures, possesses a rather limited magnetic permeability that makes difficult to extract useful experimental values. Here we focus our attention to another physical term that defines the double negative features, i.e., the phase velocity, which is a prerequisite to obtain double negative metamaterials.²⁸ In a normal wave-matter interaction, according to classic solid state physics, waves are considered to propagate in a double positive medium (ε and μ are both positive) and their propagation phase velocity is regarded as positive values, i.e., waves are moving forward. However, when ε and μ are both negative, phase velocity is expected to enter into negative zone, rendering backward waves. We present in Fig. 4 the frequency plots

of the transmission phase of the hybrid composite with the presence of an external magnetic bias. Clearly, it can be seen that the derivative of the phase function (phase velocity) reverses its sign when magnetic fields exceed 300 A/m, therefore implying the presence of double negative EM parameters.³⁵ This field-controlled behavior is also consistent with the above discussed results about the redshift-blueshift properties of the transmission coefficients (Fig. 2(a)) and the decrease-increase trend of the effective permittivity (Fig. 3(b)). Moreover, the identified negative refractive index region (2 to 4 GHz as displayed in Fig. 4) overlaps with the above-discussed transmission window frequencies for the negative permittivity dispersion (Fig. 3(b)). In some recent studies model predictions also support the fact that in the GHz range the magnetic permeability of the microwires arranged in a parallel array possesses negative values, albeit with a very small magnitude.^{36,37} It is sufficient to conclude at this stage that under the tuning effect of external fields the negative permeability can also be obtained. It is worth mentioning that this negative phase velocity is also identified in our previous parallel¹² and orthogonal³⁰ microwire composites as well as other hybrid metacomposites that will be introduced later. However for brevity we will not repeat the discussion on the phase analysis for the latter metacomposite systems. All these results give further indication of the existence of a double negative media in the microwire-composite system. This magnetic bias-tunable metacomposite behavior can be potentially used from an operational perspective to obtain a microwave invisibility cloaking that can be activated or deactivated by conveniently using an additional magnetic field.



FIG. 4 (Color online) Frequency dependence of transmission phase variation of the dense continuous hybrid microwire composite in presence of different external magnetic fields

3.2 Continuous hybrid composites containing 10 mm spaced Co-based wires

It has been mentioned above that this field-tuned metacomposite features cannot be present at high frequencies in dense hybrid composites due to the rather high reflection in place. It is natural therefore to imagine that a high frequency transmission window could be obtained in a continuous hybrid composite system by increasing the Co-based wire spacing. This transmission window would be generated by the possible magnetic resonance between the couples of wires, and it is actually featured by the composites containing 10 mm spaced Co- and Fe-based wire arrays and their hybridized arrays in the frequency band of 0.9 to 7 GHz (Fig. 5). First, the Co-based wire composite displays weak transmission and field-tunable properties that are consistent with the overall high reflection loss (Fig. 5(b)). This feature suggests that the 10 mm spacing in the single Co-based wire array would still be too narrow to maintain double-negative features in the bandwidth considered in this work. It is however remarkable that a transmission window is identified with the 10 mm spaced Co-

based wire array added to the 10 mm spaced Fe-based microwire composite between 1.5 GHz to 5.5 GHz without dc fields, which experimentally proves existence of the DNG characteristic. This behavior resembles to the one exhibited by Fe-based microwire composites with the natural DNG feature.¹² By introducing the additional Co-based wire array into the composite the magnitude of f_p is enhanced due to the increase of the continuous wire medium complexity therein³³ as also evidenced in Fig. 7, which results in the negative permittivity dispersion in the lower frequency band. Meanwhile, the negative permeability should be maintained at frequencies between the NFMR (2.5 GHz according to above analysis) and ferromagnetic anti-resonance ones (FMAR) of the Fe-based wires. The FMAR frequencies are however larger than maximum measuring bandwidth. A slightly different window position where the metacomposite features emerge is however noted, i.e., the transmission window initiates from 1.5 GHz rather than 2.5 GHz. This small deviation can be attributed to the non-uniform distribution of frozen-in stresses during the fabrication of the microwires,^{32,38} as well as the residual stress distribution at the microwire/polymer interface.²⁹ Co-based microwires have a much smaller NFMR frequency compared to Febased ones, but the position of the resonance peak can be effectively tuned by the external magnetic bias.^{13,15} However, the fact that the position of transmission window is not affected by the presence of external fields implies that the left-hand behavior is independent of the intrinsic properties of Co-based microwires at the low frequencies regime (Fig. 5(a)). This is due to the weakened dynamic coupling existing between the EM waves and the single Cobased microwire array that arises from the large dielectric loss (Fig. 5(b)). One might expect to extend the transmission window to even higher frequencies by decreasing the spacing of adjacent Fe-based wires, in the hope of increasing the value of f_p and obtaining a stronger magnetic excitation from the composite with higher wire content. However, further investigations on the hybridization of Fe-based wire arrays with spacing of 7 or 3 mm plus 10 mm Co-based wire array show that the transmission window is cancelled out (*not shown here*). This is because both the heavily loaded Co- and Fe-based wires show such a significant reflectance that forbids the occurrence of the transmission window.



FIG. 5 (Color online) (a) Transmission, (b) reflection and (c) absorption coefficients of composites containing 10 mm spaced Fe-based wire array, the 10 mm spaced Co-based wire array and their hybridized wire arrays in the frequency band of 0.9 to 7 GHz.

Figure 6 displays the transmission, reflection and absorption coefficients of the continuous microwire composites in the frequency band between 7 GHz and 17 GHz. Overall, the strong transmission and reflection of the Co-based wire array composite are still present because of the drastically diminished skin depth at higher frequencies and consequently large

magnetic and dielectric losses.³⁹ A feature worth noting is that transmission enhancement together with reflection and absorption dips are now present at frequencies between 9 to 17 GHz for the Fe-/wide spaced Co-based hybrid wire array composite, indicating a natural DNG feature at such high frequencies. An absorption peak at 8.5 GHz is also noted, which implies that magnetic resonance occurred and is responsible for the identified high frequency EM wave-induced transparency. We have explained in a previous study that a decrease of the wire spacing to 3 mm would induces strong dynamic wire-wire interactions with microwave and gives rise to LRDR.¹² Hence, by further reducing the wire spacing to 1 mm (the mismatched spacing between the Fe-based and Co-based microwire arrays), we can generate a similar effect and produce noticeable absorption by interaction. Moreover, the circumferential fields created by the coupling between the E_k and the longitudinal anisotropy of Fe-based wires⁴⁰ can also overlap and interact with the circumferential anisotropic field of Co-based microwire array, thus enhancing the magnetic excitation. At this point, this interwire resonance indicates the presence of a negative permeability dispersion above the resonance frequency. The observed high-frequency transparency also suggests a negative permittivity related to the enhanced value of f_p . Overall, this transmission increase achieved by wire misalignment/offset and the dynamic excitation from the propagating EM waves broadens the metacomposite operating frequencies in success. According to Kittel's relations,²¹ the above observed two magnetic resonances and transmission peaks should have shifted towards the blue with the presence of an increasing external field. However, the experimental results confirm that these peaks are quite independent of dc fields, and this can be explained by the degraded magnetic performance of the microwires during the curing cycle.^{23,24} A remedial strategy for this issue is the application of a proper pretreatment to the microwires such as magnetic field⁴¹ or current annealing⁴² to obtain better static and dynamic EM properties.



FIG. 6 (Color online) (a) Transmission, (b) reflection and (c) absorption coefficients of the same composites as shown in Fig. 4 measured in the frequency band of 7 to 17 GHz.

To understand the dielectric behavior of the identified dual-band metacomposite characteristics one can observe Fig. 7, which shows the effective permittivity of the dilute composites containing Fe-based, Co-based and their hybrid wire arrays in the presence of an external field up to 3000 A/m. By substituting b=10mm, $a_{Co}=15.97$ µm and $a_{Fe}=16.6$ µm (inner core diameter) into Eq. 3 we obtain the values of $f_{p,Co}=4.9$ GHz and $f_{p,Fe}=4.8$ GHz. Compared with the measured value for the monolithic Fe- or Co-based microwire array

composite (Fig. 7), the value of f_p is overestimated by the equation. By employing the modified Eq. 4,¹² the value f_p for the Co-based wire array composite is believed to be slightly higher than that of Fe-based ones, as confirmed in Fig. 7, although it is still much lower than the theoretical value. This is because Co-based wires have a larger circumferential domain volume and therefore a larger a_{eff} . The overestimated f_p value however justifies the physical reason why the left-handed features are not accessible in the monolithic Fe- or Co-based microwire array composites. A prominent increase of f_p is attained with the hybridization of Fe- and Co-based microwire arrays in the composite, i.e., by obtaining negative permittivity dispersion in nearly the whole frequency measure bandwidth (Fig. 7(a)). As per Eq. 4, the effective diameter is closely associated to the intrinsic domain structure of the microwires. In the present case, when taking into account the small spacing (~ one prepreg thick, i.e., 0.25 mm) existing between the Co- and Fe-based wire layers, the two neighboring Fe- and Cobased wires can be regarded as a wire pair/couple that interacts with microwaves (Fig. 1). The hybridized composite effectively consists of 50 wire couples with approximately 10 mm spacing. As such, the long range dipolar resonance arising from the dynamic wire interactions can greatly enhance the dielectric excitation of the wire pair at higher frequencies, therefore improving the a_{eff} and consequently f_p . In polymer metacomposites, engineering the value of f_p at high frequencies is always a critical task for the metamaterial design. From Eq. 3 it appears that feasible solutions to tune f_p at high frequencies would involve the reduction of the wire spacing or to size up the metamaterial building blocks, but neither of these would effectively suppress the excessive reflection loss or the complexity in the fabrication process due to the large amount of necessary functional units. These particular aspects make the use of the established dilute medium model inapplicable.^{18,33} As a brief summary, the Co-based wire spacing has significant effects on the microwave behavior of hybrid composites. The magnetic field-tunable properties could only be preserved by decreasing the spacing of Cobased wire array to 3 mm. Moreover, increasing the Co-based wire spacing can mitigate the presence of high reflection loss at high frequencies and induce the LRDR between the Fe-Co wire couples, therefore producing a high frequency transmission window.



FIG. 7 (Color online) Frequency plots of (a) real part ε ' and (b) imaginary part ε '' of effective permittivity of composites containing10 mm spaced Fe-based wire array, the 10 mm spaced Co-based wire array and their hybrid wire arrays, respectively.

3.3 Short-cut hybrid composites

At this point, we have discussed the influence of continuous Co-based wire arrays on the microwave response in the microwire composite system. What would happen if we introduce the short-cut Co-based wire arrays in the system? Figure 8 shows the EM parameters of the polymer composites containing the short-cut Co-based wire array, the continuous orthogonal Fe-based wires and their hybridized wire arrays, respectively. As discussed in previous work, the orthogonal microwire array can generate DNG features in the microwire-polymer system due to the significantly improved f_p arisen from the additional dielectric contribution provided by the vertical wires.²⁹ However, it remains an issue to tune the identified transmission windows in such orthogonal structure. With reference to Fig. 8(b), the transmission spectrum of the composite containing single short-cut Co-based wires reveals a typical band-stop dispersion indicated by a sharp transmission dip and a reflection peak at approximately 6 GHz. Note that short wires behave as dipoles when they collectively interact with incident waves and the dipole resonance frequency is expressed as Eq. 6: ^{20,43}

$$f_{dr} = \frac{c}{2l\sqrt{\varepsilon_m}},\tag{6}$$

Where ε_m and *l* denote the permittivity of matrix materials and wire length, respectively. Substituting ε_m =3 and *l*=15 mm in the present study, we obtain f_{dr} =5.8 GHz, which coincides with the identified resonance peak present in Fig. 8(c). The microwave opaqueness in the short Co-based wires composite appears to be induced by the wire configuration. Of particular note is that the band-gap characteristic is maintained in the hybridized composite and enhances the transmission window in the 1 GHz to 6 GHz of the orthogonal Fe-based wire array composite. The reason for preservation of transmission window is due to the relatively low absorption of the Co-based wires in 1-6 GHz, as also verified by similar absorption coefficients (Fig. 8(c)). Hence the short-cut Co-based wire array has synergistically enhanced the double negative features of the composite. As opposed to conventional left-handed metamaterials, this enhanced transmission coefficients in the wire metacomposites are associated with the careful selection of magnetic/dielectric properties of its building blocks. From the perspective of the absorption spectra (Fig. 8(c)), absorption peaks are identified at 2 GHz and 6.5 GHz, which are likely originated from the FMR of Febased wires and the dynamic wire-wire interaction in the orthogonal Fe-based structure, respectively.¹² An additional peak at 8.5 GHz is also noted in the hybridized wire composite which is likely due to the dynamic wire-wire interaction between the short Co-based and continuous Fe-based wires, indicating that the Co-based wires can enhance the absorption of the hybrid metacomposite.



FIG. 8 (Color online) (a) Transmission, S_{21} (b) reflection, S_{11} and (c) absorption coefficients of composites containing orthogonal Fe-based wire array, short-cut Co-based wire array and their hybridized wire arrays in the 0.9 to 17 GHz frequency range.

The effective permittivity of the composites containing the short-cut Co-based wires, the orthogonal Fe-based wires and their hybrid wire array is displayed in Fig. 9. One observes that the hybridized composite has stronger permittivity dispersion than the pure Fe- and Cobased wire composites due to the large dielectric response induced by the inserted short Cobased wire array. Meanwhile, the composite containing the short-cut Co-based wire array reveals a non-plasmonic behavior verified by all-positive values of ε' in the whole measuring band because of the discontinuous alignment of microwire arrays. The ε'' peak at 6 GHz also coincides with the transmission dip due to the dipole resonance that results in the metacomposite/stop-band features, as displayed in Fig. 8.



FIG. 9 (Color online) Frequency plots of the (a) real part, ε ', and (b) imaginary part, ε '', of effective permittivity dispersion of composites containing orthogonal Fe-based wire array, short-cut Co-based wire array and their hybridized wire arrays.

As discussed above, several effects, i.e., FMR in Fe-based wire, Fe-Fe wire, Co-Co wire and Fe-Co wire interactions are all involved and the dominating mechanism varies at different frequencies. A combination of coarse and fine control of metacomposite behavior is therefore available via the manipulation of the wire spacing and arrangement in this composite system. In view of these merits, the present hybrid metacomposites may be potentially used in microwave applications related to radio frequency identification (RFID)

where structural polymer composites are widely used. The RFID is a contactless data acquisition technique that uses RF waves to automatically identify objects. The everincreasing applications of RFID for commercial inventory control in warehouses, supermarkets, hospitals as well as in military friend-or-foe (FoF) identification has resulted in considerable interest on researching about low-costs and long range sensors design. The recent unfortunate MH370 loss calls for an efficient RFID technology to identify civil airplanes by their distinguished microwave response. However, existing conventional tags involve structures with complicated shapes and therefore lead to high manufacturing costs.44,45 Furthermore, the undesirable parasitic coupling effect of these structures when interacting with EM waves makes the accurate analysis of their EM performance rather difficult. The present study proposes a type of versatile composite containing microwire arrays with a simple structure featured with DNG characteristics and exceptional mechanical properties. They may be potentially oriented for RFID application which identifies practical and societal significance to distinguish civil from military planes, or to identify general airborne vehicles. cloaking invisibility at this stage, and importantly, with more societal significance

4. Conclusions

A systematic study on the microwave properties of hybrid polymer composites containing Fe-based and/or Co-based with different mesostructure architectures has been conducted. The particular influences of the continuous/short-cut Co-based microwire arrays on the microwave response of hybrid composites can be summarized in the following three aspects: (i) in the continuous hybrid composite containing 3 mm spaced Co-based wires, the wire array can induce magnetic field-tunable metacomposite characteristics when the fields are higher than 300 A/m. The transmission windows display a redshift-blueshift trend with the increase of external magnetic fields due to the competition between the wire-wire

interactions between the Fe- and Co-based wires and the FMR of Fe-based wires; (ii) in the composite containing 10 mm spaced Co-based wire array, a dual-band DNG feature is observed at 1.5-5.5 GHz and 9-17 GHz. The transmission window displayed at high frequencies likely originates from the long range dipolar resonance between the Fe-Co wire couples. (iii) The composite containing short Co-based wires and orthogonal Fe-based wires manifests a metacomposite/stop-band feature. This arises from the microwave opaqueness of the Co-based wire arrays in the mid-frequencies of the measuring range, which provides a synergistic effect on the identified transmission windows. All these findings showcase unique advantages created by the hybridization of the Co-based and Fe-based microwires, which make the proposed metacomposites a potential candidate for microwave radio frequency identification applications.

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References:

¹J. B. Pendry, D. Schurig, and D. R. Smith, Science **312**, 1780 (2006).

²N. I. Landy, S. Sajuyigbe, J. J. Mock, D. R. Smith, and W. J. Padilla, Phys. Rev. Lett. **100**, 207402 (2008).

³J. B. Pendry, Phys. Rev. Lett. **85**, 3966 (2000).

⁴Z. G. Nicolaou and A. E. Motter, Nat. Mater. **11**, 608 (2012).

⁵J. H. Lee, J. P. Singer, and E. L. Thomas, Adv. Mater. **24**, 4782 (2012).

⁶R. A. Shelby, D. R. Smith, and S. Schultz, Science **292**, 77 (2001).

⁷I. J. McCrindle, J. Grant, T. D. Drysdale, and D. R. Cumming, Adv. Opt. Mater. **2**,149(2013).

⁸H. Li, L. H. Yuan, B.Zhou, X. P. Shen, Q. Cheng, and T. J. Cui, J. Appl. Phys. **110**, 014909 (2011).

⁹C. L. Chang, W. C. Wang, H. R. Lin, F. J. Hsieh, Y. B. Pun, and C. H. Chan, Appl. Phys. Lett. **102**, 151903 (2013).

¹⁰D. Shin, Y. Urzhumov, D. Lim, K. Kim, and D. R. Smith, Nature 4, 4084 (2014).

¹¹ Shalaev VM. Optical negative-index metamaterials. Nat. Photon. **1**, 41 (2007).

¹²Y. Luo, H. X. Peng, F. X. Qin, M. Ipatov, V. Zhukova, A. Zhukov, and J. Gonzalez, Appl. Phys. Lett. **103**, 251902 (2013).

¹³M. H. Phan and H. X. Peng, Prog. Mater. Sci. 53, 323 (2008).

¹⁴J. Carbonell, H. García-Miquel, and J. Sánchez-Dehesa, Phys. Rev. B 81, 024401 (2010).

¹⁵H. García-Miquel, J. Carbonell, and J. Sánchez-Dehesa, Appl. Phys. Lett. **97**, 094102 (2010).

¹⁶V. S. Larin, A. V. Torcunov, A. Zhukov, J. Gonzalez, M. Vazquez, and L. Panina, J. Magn. Magn. Mater. **249**, 39 (2002).

¹⁷H. X. Peng, F. X. Qin, M. H. Phan, J. Tang, L. V. Panina, M. Ipatov, V. Zhukova, A. Zhukov, and J. Gonzalez, J. Non-Cryst. Solids **355**, 1380 (2009).

¹⁸F. X. Qin and H. X. Peng, Prog. Mater. Sci. **58**, 183 (2013).

¹⁹D. Makhnovskiy, A. Zhukov, V. Zhukova, and J. Gonzalez, Adv. Sci. Technol. (Faenza, Italy) **54**, 201 (2008).

²⁰D. P. Makhnovskiy, L. V. Panina, C. Garcia, A. P. Zhukov, and J. Gonzalez, Phys. Rev. B **74**, 064205 (2006).

²¹C. Kittel, Phys. Rev. **73**, 155 (1948).

²²J. L. Snoek, Physica **14**, 207 (1948).

²³H. Wang, F. X. Qin, D. Xing, F. Cao, X. D. Wang, H. X. Peng, and J. Sun, Acta Mater. **60**, 5425 (2012).

²⁴H. Wang, D. W. Xing, H. X. Peng, F. X. Qin, F. Y. Cao, G. Q. Wang, and J. F. Sun, Scr. Mater. **66**, 1041 (2012).

²⁵Y. J. Di, J. J. Jiang, G. Du, B. Tian, S. W. Bie, and H. H. He, Trans. Nonferrous Met. Soc. China **17**, 1352 (2007).

²⁶F. X. Qin, H. X. Peng, N. Pankratov, M. H. Phan, L. V. Panina, M. Ipatov, V. Zhukova, A. Zhukov, and J. Gonzalez, J. Appl. Phys. **108**, 044510 (2010). also J. Appl. Phys. **108**, 07A310, 2011.

²⁷M. Vázquez and A. Zhukov, J. Magn. Magn. Mater. 160, 223 (1996).

²⁸H. García-Miquel, J. Carbonell, V. E. Boria, and J. Sánchez-Dehesa, Appl.Phys. Lett. **94**, 054103 (2009).

²⁹Y. Luo, H. X. Peng, F. X. Qin, M. Ipatov, V. Zhukova, A. Zhukov, and J. Gonzalez, J. Appl. Phys. **115**, 173909 (2014).

³⁰O. Reynet, A-L. Adenot, S. Deprot, O. Acher, and M. Latrach, Phys. Rev. B **66**, 094412 (2002).

³¹L. Liu, K. N. Rozanov, and M. Abshinova, Appl. Phys. A **110**, 275 (2013).

³²F. X. Qin, Y. Quéré, C. Brosseau, H. Wang, J. S. Liu, J. F. Sun, and H. X. Peng, Appl. Phys. Lett. **102**, 122903 (2013).

³³J. B. Pendry, A. J. Holden, W. J. Stewart, and I. Youngs, Phys. Rev. lett. **76**, 4773 (1996).

³⁴D. R. Smith, S. Schultz, P. Markoš, and C. M. Soukoulis, Phys. Rev. B 65, 195104 (2002).

³⁵O. F. Siddiqui, M. Mojahedi, and G. V. Eleftheriades, IEEE Trans. Antennas Propag. **51**, 2619 (2003).

³⁶C. S. Olariu, G. Ababei, N. Lupu, and H. Chiriac, Physica B (2015) In Press.

³⁷L.V. Panina, D. P. Makhnovskiy, A. T. Morchenko, and V. G. Kostishin, J. Magn. Magn. Mater. **383**, 120 (2015).

³⁸H. Wang, D. W. Xing, X. D. Wang, and J. F. Sun, Metal. Mater. Trans. A **42**, 1103 (2011).

³⁹F. X. Qin, H. X. Peng, and M. H. Phan, Mater. Sci. Eng. B **167**, 129 (2010).

⁴⁰L. Kraus, Z. Frait, G. Ababei, and H. Chiriac, J. Appl. Phys. 113, 183907 (2013).

⁴¹J. S. Liu, D. Y. Zhang, F. Y, Cao, D. X. Xing, D. M. Chen, X. Xue, and J. F. Sun, Phys. Status Solidi A **209**, 984 (2012).

⁴²J. S. Liu, F. X. Qin, D. M. Chen, H. X. Shen, H. Wang, D. W. Xing, M. H. Phan, and J. F. Sun, J. Appl. Phys. **115**, 17A326 (2014).

⁴³F. X. Qin, C. Brosseau, H. X. Peng, H. Wang, and J. Sun, Appl. Phys. Lett. **100**, 192903 (2012).

⁴⁴J. McVay, A. Hoorfar, and N. Engheta, IEEE Radio Wireless Symp. San Diego, CA, Jan. 17–19, 2006, pp.199–202.

⁴⁵I. Jalaly and I. D. Robertson, Eur. Microw. Conf. Paris, France, Oct. 4–6, 2005, vol. 2, pp. 4–7