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In situ microwave characterization of microwire composites with external magnetic field

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Rubber composites containing Co68.15Fe4.35Si12.25B15.25 glass-covered amorphous microwires were fabricated. For samples containing 5 mm long wires and randomly dispersed in the polymer matrix, our results demonstrate that the current induces resonance of circumferential permeability (magnetoimpedance resonance) resulting in a significant change of the effective permittivity of the wire-filled composites with application of a high magnetic field. For samples containing longer wires (70 mm) periodically arranged in the polymer matrix, the permittivity spectrum shows two resonance peaks due to, respectively, dipole and magnetoimpedance resonance. The induced magnetoimpedance resonance has a narrower linewidth than the dipole one as magnetic field is increased. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4712126]

Despite being the subject of intense research for almost two decades, optimized materials for microwave engineering are just beginning to be understood. Previous research considered materials with specific permittivity via the application of an electric field, magnetic permeability via a magnetic field excitation, and indirect manipulation of the permittivity (respectively, the magnetic permeability) via a magnetic field (respectively, an electric field) excitation, e.g., multiferroics.

In this report, we study the microwave properties of composite structures containing ferromagnetic microwires. The results reveal that a magnetic field can induce significant variation of the impedance via the skin effect at high frequencies. The current distribution along the wire induces dipolar polarization. This, in turn, permits to manipulate the effective permittivity by an applied magnetic field. There have been few previous studies on this subject in the literature. Specifically, Liberal and co-workers considered an analytical formulation of the electromagnetic response of arrays of ferromagnetic microwires. They further showed that the magnetic response of the microwires allows controlling of the effective permittivity of the composite material. That is, the resistance of the microwires can be controlled by the magnetic losses existing at the ferromagnetic resonance frequency, while the reactive load is inductive or capacitive, as determined by the sign of the real part of the magnetic permeability of the microwires.

Panina and co-workers and Acher and co-workers used microwire arrays to realize left-handed materials which offer cross-tuning possibility-tuning of the permittivity by magnetic field. Starostenko and Rozanov gave an in-depth discussion of the magnetic bias effect of the effective permittivity of composites containing random wires and demonstrated the suitability of this kind of composites for controlling the attenuation of electromagnetic waves. Di and co-workers, Marin and co-workers, and Zhang and co-workers also reported on the microwave absorption of microwire composites. Likewise, our previous studies showed that such composites have multifunctional properties and may serve for sensing and structural health monitoring components. We use our own purpose-designed and built facility for in situ microwave characterization of the composites over a broad range of frequencies and concomitant application of magnetic field excitation.

The samples used in this study consisted of soft magnetic microwires, with nominal composition Co68.15Fe4.35Si12.25B15.25 and average diameter of about 40 µm, embedded in a rubber matrix. Details of a sample preparation can be found elsewhere. The hysteresis loop of the wires was measured by using a conventional induction method. The wires possess low coercivity and large magnetostriction coefficient which make them suitable for...

(a)

(b)

FIG. 1. (a) The M-H hysteresis loop of the melt-extracted Co68.15Fe4.35Si12.25B15.25 microwire. (b) Stress-strain curve of a microwire studied in this study.

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sensing applications. An INSTRON machine with a load cell of 1 kN was used to obtain the tensile stress-strain curve of single wires (Fig. 1(b)).

For the present study, two kinds of composites were prepared with different wire lengths. In sample A, 50 mg of 5 mm long wires was randomly dispersed in a silicon rubber by mechanical mixing. The mixture was subsequently mould casted to obtain samples with dimensions $70 \times 10 \times 1.8$ mm$^3$ and cured at ambient temperature for 24 h (Fig. 2(a)). The result sample has a microwire content of 3 wt. %. In sample B, we used 70 mm long microwires and aligned them in a periodical manner (Fig. 2(b)) with fixed wire spacing of 0.77 mm into silicon rubber matrix sheets which were bonded together using silicone resin. The dimensions of sample B are approximately similar to those of sample A.

The effective complex (relative) permittivity $\varepsilon = \varepsilon' - i\varepsilon''$ spectra were measured at room temperature with and without applied magnetic bias over the frequency range 300 MHz–6 GHz. Full details on the design and specification of the microwave spectrometer are available in the previous article, and only a brief summary will be provided here. Briefly, our experiments consist of measuring the transmission and reflection coefficients of an asymmetric microstrip transmission line containing the sample in the presence of a magnetic bias. The electromagnetic measurement was carried out with the wave configuration as shown in Fig. 2. The measurement of the scattering parameters (S parameters) was achieved using an Agilent H8753ES network analyzer with short-open-load-thru (SOLT) calibration. A utility program extracts the data and generates $\varepsilon$ (and also magnetic permeability) spectra as individual files in txt format. As outlined in Ref. 17, the quasi-transverse electric and magnetic mode which is the only mode that propagates in the structure makes analysis of the complex transmission and reflection coefficients created by the continuity between the line and the sample relatively uncomplicated. Using the Nicolson-Ross procedure for the transformation of the load impedance by a transmission line, $\varepsilon$ is determined by the transmission $S_{21}$ and reflection $S_{11}$ parameters. An error analysis indicates modest uncertainties in $\varepsilon'$ ($<5\%$) and $\varepsilon''$ ($<1\%$) for the data. One further feature of the measurement system is worth commenting on. To obtain accurate measurements of $\varepsilon$, it is particularly important to account for the residual air-gap between the sample and the line walls, i.e., the gap is determined by the roughness of the surfaces of the measured samples.

Representative spectra of the real, $\varepsilon'$, and imaginary, $\varepsilon''$, parts of the complex permittivity at five different magnetic bias values for sample A are shown in Fig. 3. At zero magnetic field, the permittivity shows a relaxation behavior. When a magnetic field of 100 Oe is applied, $\varepsilon'$ is found to increase and the relaxation remains visible. With further increase of magnetic field up to 500 Oe, an absorption peak is seen near 4.1 GHz (Fig. 3(b)). This maximum shifts to higher frequency of, respectively, 4.6 and 4.7 GHz as the magnetic field is increased to 1 and 1.5 kOe, with increased height and width. The peak observed in Fig. 3(a) is associated with anomalous dispersion of $\varepsilon'$. This dispersion is related to negative values of $\varepsilon'$ for frequencies higher than 4.7 GHz when the magnetic field is larger than 1 kOe.

Fig. 4 presents the magnetic bias dependence of the real and imaginary parts of the effective complex permittivity for sample A at 2.5 and 4.5 GHz. At 2.5 GHz (off resonance), the variations in $\varepsilon'$ and $\varepsilon''$ are small. This contrasts with the rather pronounced changes of $\varepsilon'$ and $\varepsilon''$ observed at 4.5 GHz (resonance). It can also be seen that, at 4.5 GHz, the application of a magnetic field excitation gives rise to a $\varepsilon''$ larger than $\varepsilon'$, which is not seen at 2.5 GHz.

In order to examine the influence of the magnetic bias on the permittivity of sample B, we turn now to analyzing the microwave absorptive behavior probed by our electromagnetic measurements (Fig. 5). Two well separated absorption lines are found in the ranges 1–2 GHz and 4–5 GHz.
Since the applied magnetic field strongly influences the current distribution in the microwires through the skin effect, this gives rise to a significant change of the dielectric response in the GHz range of frequencies. Absorption via the skin effect predominates over dipolar absorption since the magnetoimpedance resonance frequency is smaller than the dielectric resonance frequency.\textsuperscript{26} From the inset of Fig. 3(b), we note that our measurements of the blueshift of the magnetoimpedance resonance frequency with the applied field $H_{dc}$ are consistent with the field effect on the impedance profiles calculated from the $S$ parameters (Fig. 3(c)). Our comprehensive data set also follows the trend of the theoretical calculations of Liberal et al.,\textsuperscript{9} obtained for physically reasonable parameter choices. The interest in the microwave behavior of these composite samples lies also partly in its anomalous dispersion.\textsuperscript{23} Since application of a magnetic bias can lead to large eddy current losses, we suggest that wire-filled composites can be exploited for designing microwave absorbers having large absorption bandwidth. In addition, the anomalous dispersion is often associated with a negative value of $\varepsilon'$.\textsuperscript{7} Thus, a variety of unusual properties can be engineered to meet the requirements of a high-performance frequency selective surface. It should be noted that while the magnetic bias has a strong influence on the permittivity, it has negligible effect on the magnetic permeability (not shown here). Since the wire concentration is low, the magnetic permeability is close to unity in the GHz range of frequencies. For sample B, the wires which are perpendicular to the microwave magnetic field do not contribute whereas those which are parallel have no response to the field in the GHz range of frequencies. Hence, we predict that the sensitivity of permittivity to frequency and magnetic field will result in useful applications of this kind of material samples.

The second point deals specifically with sample B. Comparing with sample A, it is notable that it has a much lower dipole resonance frequency at 1.1 GHz according to $f_{res} = c/(2\ell \sqrt{\varepsilon_m})$ which is in good agreement with the lower frequency peak shown in Fig. 4. The difference observed with the experimental value of the resonance frequency can be attributed to interfacial defects in the composite, e.g., imperfect bonding.

Now, we come to the third point of our discussion. Very recently, experimental studies, e.g., Ref. 27, have studied the magnetoimpedance effect and field sensitivity in Co$\textsubscript{84.55}$Fe$\textsubscript{4.45}$Zr$\textsubscript{7}$B$\textsubscript{4}$ amorphous ribbons with 50 nm thick Co coating layer on the free ribbon surface. The magnetoimpedance ratio, $\Delta Z/Z = 100\%[Z(H) - Z(H_{max})]/Z(H_{max})$, where $Z(H)$ and $Z(H_{max})$ represent, respectively, the impedance in a magnetic field $H$ and in the maximum field ($H_{max} = 120$ Oe), reaches the largest value of 24% at 2 MHz. Remarkably, a similar maximum value ($\approx 20\%$) of the surface impedance ratio can be observed in the spectra shown in Fig. 3(c) in the GHz range of frequencies. This effect is clearly seen at high fields. These results show that our hybrid materials are of practical importance for designing highly sensitive magnetic sensors in the microwave range of frequencies.

To summarize, we have performed a detailed study of the microwave response of composite samples containing glass-covered amorphous microwires embedded in a rubber matrix with different wire length and topological arrangement. Our

![Graph](image1.png)

FIG. 4. Magnetic field dependence of the real and imaginary parts of the effective complex permittivity for sample A at 2.5 and 4.5 GHz.

respectively. One notes that the bias dependence of permittivity can be clearly observed for each resonance region. As the magnetic field is increased, the absorption line at 4–5 GHz grows in magnitude and becomes narrower. The linewidths of the dielectric and magnetoimpedance resonances present opposite variations as the field is increased.

Three aspects of the above results should be noted. The first is that microwires respond to the electromagnetic wave like electric dipoles for sample A. The dipole resonance can be expressed as $f_{res} = c/(2\ell \sqrt{\varepsilon_m})$, where $c$ is the electromagnetic wave velocity in vacuum, $\ell$ is the wire length, and $\varepsilon_m$ is the permittivity of the host matrix. When $\ell$ is chosen at 5 mm, the resonance frequency is close to 15 GHz which is out of the measurement range of the present work. This explains the absence of the absorption line in the spectrum when no magnetic field is applied, or when the magnitude of the magnetic field is low. With the application of high magnetic field, our results demonstrate that the current induces resonance of circumferential permeability, i.e., the magnetoimpedance resonance leads to a significant change of the effective permittivity of the wire-filled composites.\textsuperscript{24–26}

![Graph](image2.png)

FIG. 5. Frequency dependence of the real and imaginary parts of the effective complex permittivity for sample B at zero magnetic field and 1.5 kOe.
results show that there is a strong dependence of the permittivity spectra on the external magnetic bias. For wire composites containing randomly dispersed wires of 5 mm length, the real part of the permittivity increases as the magnetic bias is increased until it attains a maximum which corresponds to the maximum absorption induced by the magnetoimpedance resonance. The absorption resonance frequency and linewidth can be conveniently tuned by the magnetic bias. Composites containing periodically arranged longer wires (70 mm) are characterized by a double-peak permittivity spectrum. The higher frequency resonance is associated with magnetoimpedance resonance. It has a narrower linewidth than that of the lower frequency (dipole) resonance as the magnetic field is increased.

The present study points to interesting physics in our understanding of the electromagnetic properties of composite materials with magnetically tunable microwave properties. Further, microwave measurements under applied stress will be carried out to provide a better understanding of the electromagnetic behavior for stress sensing and structural health monitoring applications.28

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