Experimental demonstration of tunable scattering spectra at microwave frequencies in composite media containing CoFeCrSiB glass-coated amorphous ferromagnetic wires and comparison with theory

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We demonstrate composite media with ferromagnetic wires that exhibit a frequency region at the microwave regime with scattering spectra strongly dependent on an external magnetic field or stress. These tunable composite materials have recently been proposed theoretically; however, no direct experimental verification has been reported. We used composite materials with predominantly oriented CoFeCrSiB glass-coated amorphous wires having large magnetoimpedance at GHz frequencies. The free-space measurements of reflection and transmission coefficients were conducted in the frequency range 1–8 GHz in the presence of an external static magnetic field or stress applied to the whole sample. In general, the transmission spectra show greater changes in the range of 15 dB for a relatively small magnetic field of 5–10 Oe or stress of 0.1 MPa. The obtained results are quantitatively consistent with the theoretical spectra predicted by the effective medium arguments for permittivity in conjunction with the wire electric polarization dependent on the ac surface impedance. A number of applications of proposed materials are discussed, including the field tunable microwave surfaces and the self-sensing media for the remote nondestructive evaluation of structural materials.

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I. INTRODUCTION

Composite structural materials containing periodic or random arrays of conducting scattering elements have received much attention because they make it possible to engineer a selective spectral response to the electromagnetic radiation. It has been demonstrated that composites with conducting wires possess a number of unique properties at microwave and optical frequencies, such as anomalous large photonic band gaps,[1] appearance of bulk plasmon modes,[2] and resonancelike anomalous dispersion of the effective dielectric function also associated with negative values.[3–6] A recent trend is to achieve the adjustability of these materials. This would be in a great demand in numerous applications, in particular, wireless communication, telemetry, and remote nondestructive testing. Several methods were proposed based on biased ferroelectric or ferrite substrates[7] and reconfigurable resonant elements.[8–10] It has also been proposed recently that in the composites filled with magnetic wires exhibiting a large magnetoimpedance (MI) effect it should be possible to tune the scattering spectra at GHz frequencies by applying a weak magnetic field or a stress.[11–15] However, this tunability concept has not been experimentally verified. In this paper, using the free-space measuring technique in the frequency band of 1–8 GHz, it has been demonstrated that the transmission of the electromagnetic wave through a thin sheet of the composite containing CoFe-based amorphous glass-coated microwires can be changed over 15 dB by a weak dc magnetic field in the range of 5–10 Oe or a tensile stress, applied to the whole sample. The experimental results agree well with the theory[11] based on the effective medium concept and the wire electric polarization depending on its magnetic structure.

The main physical principles which determine the behavior of tunable microwave composites filled with ferromagnetic wires have already been established. Wire-based composite materials can be treated as continuous media at least in the radiative near or far field region and can be then characterized by the effective permittivity ε_{eff}. If the skin effect in wires is strong, the permittivity ε_{eff} is determined by the wire geometry, concentration (or lattice period), and the permittivity of the host material. In a general case of a moderate skin effect when the wire radius is in the range of the skin penetration depth $a \approx \delta$, the dispersion characteristics of ε_{eff} will also depend on the internal electric and magnetic properties of the wires. The influence of the skin effect on the effective dielectric response in diluted wire-based materials has been first established theoretically[3,16] and experimentally[4,6] considering nonmagnetic wires with different electric conductivity. It has been then demonstrated theoretically[11,12] that the controlling quantity is related to the surface impedance of the wires, which involves the wire permeability μ coupled with the dc magnetic structure. Therefore, in composite containing ferromagnetic wires exhibiting MI effect at microwave frequencies[17,18] the effective permittivity may depend on a static magnetic field via the corresponding dependence of the surface impedance. The surface impedance can also be changed by applying a stress which modifies the magnetic anisotropy and domain structure in wires. Hence, the effective permittivity ε_{eff} may also depend on the external stress or strain.

The first experimental attempt to demonstrate the effect of magnetic properties of wires on the permittivity ε_{eff} was made by measuring reflection/transmission spectra from lattices of long parallel ferromagnetic wires with different magnetic structures whereas the sample topology (the wire length, diameter, spacing in the lattice) was nearly identical.[12] The reported results did demonstrate different permittivity spectra; however, it was not easy to attribute those changes to the wire magnetic structure alone.

A natural approach to this problem would be to investigate the possible variations in the scattering response of the same magnetic composite sample under the effect of applied stimuli, such as magnetic field or stress, which could change the magnetization in the wires. However, this direct experiment has not been realized previously because of anticipated problems of applying a magnetic field to a large sample (at least 30×30 cm^2) within a free-space installation. We are therefore motivated to experimentally verify the effect of large adjustability of the electromagnetic response of the composites with magnetic wires. The amorphous microwires are soft magnetic materials and their magnetization change can be made with a small magnetic field. For example, in Co-based microwires with a negative magnetostriction and a circular domain structure, the magnetization is rotated towards the axis by a field of a few Oe, which is accompanied by a large change in impedance in the range of 100% at the GHz frequencies.[18] In our experiments an essentially uniform field up to 12 Oe in the area of 50×50 cm^2 was produced with a specially designed planar coil mounted on the sample. The coil is invisible for the irradiating wave when the current leads are perpendicular to the electrical field vector. The other method to externally control the electromagnetic response from magnetic wire composites is applying a stress or strain to change the dc magnetization in wires. Using a matrix of a high-elasticity module, a relatively small stress applied to the whole composite in the range of 0.1 MPa will produce sufficiently large stress acting on the wire via strain transfer.

In this work, we have demonstrated that the composites containing short CoFeCrSiB wires demonstrate a sufficiently narrow band gap associated with the antenna resonance in wires and negative values of ε_{eff} beyond the resonance. For circular magnetic configuration in the wire, its magnetic properties play no effect on the propagation of the electromagnetic waves through the composite since the relevant ac permeability is unity. Applying a magnetic field and establishing the magnetization along the axis corresponds to the case of a high permeability and high surface impedance. This broadens the antenna resonance and opens bandpass: from nearly -27 dB to -12 dB. In the case of composite having wires with induced helical anisotropy, the effect of the tensile stress would be reverse: the application of the stress creates the band gap.

The proposed technology to control the microwave propagation with magnetic wire structured composite materials is cost effective and is suitable for large scale applications, in particular, as tunable frequency selective surfaces and coatings. The other large area of application includes stress-sensitive materials for microwave nondestructive testing and control in civil engineering. The fiber reinforced materials have become an important class of constructing materials.[19,20] Their performance depends on the bond between the fibers and the matrix, which is not easy to measure or model. Utilizing magnetic wires along with other polymer fibers and measuring microwave response can provide insight on the adhesion quality and stress transfer in the composite.

This paper is structured as follows. In Sec. II the dispersion characteristics of two types of wire composites are considered, demonstrating that the surface impedance of wires can control the scattering spectra. In Sec. III we review the results on MI in wires relevant to tunable microwave composites. Section IV describes the experimental samples, techniques, and results. In Sec. V we compare the obtained transmission spectra with the theoretical ones and propose future investigations based on a tunable phase of the reflected signal.

II. DISPERSION OF THE EFFECTIVE PERMITTIVITY

In electrodynamics of composites, conductive wire inclusions may induce unusual polarization properties in response to radiation in various frequency ranges over the electromagnetic spectrum. It has been proposed that these structured materials should be treated as continuous media, at least in the radiative field region with a characteristic effective permittivity ε_{eff}.[3,4,6] The ability of these artificial dielectrics to manipulate the electromagnetic radiation can be then viewed as a consequence of a specific dispersion of ε_{eff}. In this work, the main emphasis is placed on composites filled with short conductive wires.[3–6] These materials demonstrate the resonancelike dispersion of the effective permittivity ε_{eff} with the characteristic frequencies determined by the antenna resonance $f_{res,n}=c(2n-1)/(2l\sqrt{\varepsilon})$, where c is the velocity of light, l is the wire length, ε is the matrix permittivity (usually, a real constant), and $n\geqslant 1$ is an integer number (discrete spectrum). In the vicinity of the resonance the real part of ε_{eff} decreases with increasing the frequency (anomalous dispersion), whereas the imaginary part has a local maximum. For the wire volume concentrations p below the percolation threshold ($p<p_c\sim 2a/l$, where a is the wire radius),[3] the dispersion properties of these materials can be described in the frame of the Lorentz model for oscillating electrical dipole moments with a damping. The general form of the permittivity is

$$\varepsilon_{eff}(\omega)=\varepsilon+4\pi p\sum_{n=1}^{\infty}\frac{A_n}{(\omega_{res,n}^2-\omega^2)-i\omega\Gamma_n}, \qquad (1)$$

where the summation is carried out over all the antenna resonance frequencies $\omega_{res,n}=2\pi f_{res,n}$, $i=\sqrt{-1}$ is the complex unit, and the strength of oscillators and relaxation are described by the phenomenological parameters A_n and Γ_n, respectively.[21,22] Each phenomenological parameter Γ_n is contributed by resistive and magnetic losses, and also the wave scattering.[11] As has been demonstrated experimentally,[4,6] ε_{eff} recovered from S-parameter measurements is well approximated by Eq. (1), where only the first term is important. The resonance contribution to ε_{eff} can be very large even for small concentrations $p<0.01\%$. Then, ε_{eff} becomes negative in some frequency band past the resonance, and the wave propagation is forbidden in this frequency range. If the relaxation in the system is large, the dispersion of ε_{eff} broadens and its real part remains positive. In this case, the band gap will be essentially eliminated. The magnetic contribution to Γ_n occurs because of the following. When the composite is irradiated by the electrical field e_0 parallel to the wires, this field also creates a circular magnetic field h_φ within the wire (the wire axis is denoted as x axis). At the wire surface, the x component of the total electric field e_t and h_φ are related via the longitudinal component ς_{xx} of the wire impedance tensor,[23]

$$e_t = \varsigma_{xx} h_\varphi. \tag{2}$$

Equation (2) should be taken as a boundary condition when solving the scattering problem from a conducting wire. In a magnetic wire, the parameter ς_{xx} depends on the circular permeability associated with h_φ. The dependence of the surface impedance on the magnetic properties of the wire will be considered in the following section. If the interaction between the short wire inclusions is neglected, the average polarization η of a wire and the effective permittivity ε_{eff} of the whole composite take simple analytical forms,[14]

$$\eta = \frac{1}{2\pi \ln(l/a)(\tilde{k}a)^2}\left(\frac{2}{\tilde{k}l}\tan(\tilde{k}l/2) - 1\right),$$

$$\varepsilon_{eff} = \varepsilon + 4\pi p \eta. \tag{3}$$

Here a is the wire radius and $\tilde{k}$ is the renormalized wave number depending on the surface impedance in the following way:

$$\tilde{k} = k\left(1 + \frac{ic\varsigma_{xx}}{\omega a \ln(l/a)}\right)^{1/2}, \tag{4}$$

where $k=\omega\sqrt{\varepsilon}/c$ is the wave number of the dielectric matrix. In Eq. (3), the polarization η was derived assuming that the radiation losses can be neglected in comparison with the magnetic and resistive losses, which is reasonable in the case of a moderate skin effect. The renormalization of the wave number in Eq. (4) is also essential in the same approximation.

For comparison, we now consider the system of long (continuous) wires and will demonstrate that its effective permittivity also depends on the wire surface impedance. In this case, there are no charge distribution along the wires and associated dipole resonances, so the system electromagnetic response will be different from that described by the Lorentz model above. Pendry *et al.*[2] have demonstrated that the response of such an array is similar to that of a low-density plasma of very heavy charged particles, which is a consequence of confining the electrons within thin wires. As a result, the effective permittivity for waves with the electric field polarization along the wires has a characteristic plasma dispersion behavior $\varepsilon_{eff}=\varepsilon-(\omega_p/\omega)^2$ but with strongly reduced plasma frequency ω_p,[2]

$$\omega_p = \frac{2\pi c^2}{L^2 \ln(L/a)}, \tag{5}$$

where L is the cell parameter (the average distance between the wires). The value of the plasma frequency would be in the GHz range for the radius a of few microns and the cell parameter L of few millimeters.

Within this approach, the plasma frequency (5) and the effective permittivity of continuous wire arrays are independent of the internal conductive and magnetic properties of the wires. In Ref. 16, ε_{eff} of such composites with nonmagnetic wires was deduced by solving the Maxwell equations within and outside the wires and employing a homogenization procedure,

$$\varepsilon_{eff} = \varepsilon - p\frac{2\varepsilon_c F_1(k_c a)}{(ak_c)^2 F_1(k_c a)\ln(L/a) - 1},$$

$$F_1 = J_1(x)/xJ_0(x), \tag{6}$$

where $p=\pi a^2/L^2$ is the wire volume concentration, $\varepsilon_c=4\pi i\sigma/\omega$ is the dielectric permittivity of the conductor, σ is the wire conductivity, $k_c^2=4\pi i\omega\sigma/c^2$ is the wave number in the wire, and $J_{0,1}$ are the Bessel functions. In the case of a strong skin effect ($ak_c \sim a/\delta \gg 1$), Eq. (6) becomes independent of the wire conductivity and reduces to that of plasma dispersion with the plasma frequency given by Eq. (5). We have extended the described approach to the case of magnetic wires. It is possible to write the solution of the electric field in the system which depends on the wire conducting and magnetic properties through the surface impedance. Then, Eq. (6) can be represented as follows:

$$\varepsilon_{eff} = \varepsilon - p\frac{\omega_p^2}{\omega^2\left(1 + i\dfrac{c\varsigma_{xx}}{\omega a \ln(L/a)}\right)}. \tag{7}$$

Examining Eq. (7), we can conclude that the plasma frequency undergoes renormalization and becomes dependent on the wire surface impedance in a similar way as the wave number for a system of short wires in Eq. (4). Therefore, we have demonstrated that in both types of wire-based composites, the dispersion of the effective permittivity can be controlled by changing the surface impedance in the wires. In Refs. 11 and 12 it was proposed to use the field-dependent impedance in the ferromagnetic wire with a specific magnetic anisotropy to change the microwave response.

III. MAGNETOIMPEDANCE (MI) IN AMORPHOUS MAGNETOSTRICTIVE WIRES

We have demonstrated that the microwave dielectric response of structured composites may depend on magnetic properties of constituent elements via the surface impedance, which can be used as the boundary condition (2) to find the electromagnetic field distribution outside the filling inclusions. In the case of a strong skin effect in the inclusions and a scalar permeability μ, the surface impedance is determined by the Leontovich condition,

$$\varsigma_{xx} = (1-i)\sqrt{\frac{\omega\mu}{8\pi\sigma}}. \tag{8}$$

In fact, Eq. (8) has an illustrative character. First of all, the condition of a strong skin effect may not be valid or may not represent the case of physical interest. In our case, when the skin effect becomes strong, the effective permittivity will mainly depend on the structural parameters of the system and not on the internal properties of the inclusions. The other difficulty is related to the permeability calculations. This parameter depends on the dynamic magnetization processes, which could be quite complicated in real systems, resulting in a particular frequency and spatial dispersion of μ. Furthermore, the permeability and hence impedance have a tensor form even in uniform materials because of the magnetization precession. The detailed analysis of the surface impedance in magnetic wires with uniform magnetization (in a single domain state) valid for any frequency and type of magnetic anisotropy has been made in Refs. 23 and 24. The calculation of the impedance tensor $\hat{\mathbf{s}}$ was based on the solution of the Maxwell equations for the ac electric **e** and magnetic **h** fields inside the wire, together with the equation of motion for the magnetization vector **M**. Considering a linear approximation with respect to the time-variable parameters **e**, **h**, $\mathbf{m}=\mathbf{M}-\mathbf{M}_0$, where $\mathbf{M}_0$ is the static magnetization, and neglecting the exchange effects, the problem was simplified to finding the solutions of the Maxwell equations with a given ac permeability tensor $\hat{\boldsymbol{\mu}}=1+4\pi\hat{\boldsymbol{\chi}}$, $\mathbf{m}=\hat{\chi}\mathbf{h}$. In a single domain wire, the linear magnetization dynamics is described by a uniform precession of the total magnetization vector **M** around $\mathbf{M}_0$. The susceptibility tensor is obtained from the linearized Landau-Lifshitz equation considering the magnetic potential energy of the form

$$U_m = -K_{eff}(\mathbf{n}_K\cdot\mathbf{M})^2/M_0^2 - (\mathbf{H}\cdot\mathbf{M}),$$

$$\mathbf{H} = \mathbf{H}_{ex} + \mathbf{h}, \tag{9}$$

where $(\mathbf{n}_K\cdot\mathbf{M})$ and $(\mathbf{H}\cdot\mathbf{M})$ are the scalar products. In Eq. (9), a uniaxial magnetic anisotropy is considered with the effective anisotropy constant K_{eff} and easy axis $\mathbf{n}_K$ having some angle α with the wire x axis. The external magnetic field H includes the dc field $\mathbf{H}_{ex}$ (which will be used as an external factor to change the impedance and microwave response) and the high-frequency field **h**. For moderate values of $\mathbf{H}_{ex}$, only its projection on the wire axis will influence the magnetic configuration because of the large shape anisotropy in the normal direction (demagnetizing effects). In amorphous materials the magnetic anisotropy excluding shape anisotropy is mainly determined by the magnetoelastic interaction and will depend on the external stress or strain that also can be used as a tuning parameter. In the coordinate system with the axis x' directed along the static magnetization, the susceptibility tensor has the following form with respect to the unit vector (n_z, n_y, n_x):

$$\hat{\boldsymbol{\chi}} = \begin{pmatrix} \chi_1 & -i\chi_a & 0 \\ i\chi_a & \chi_2 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{10}$$

The susceptibility components χ_1, χ_2, and χ_a depend on the frequency ω and the magnetic configuration as following:

$$\chi_1 = \omega_M(\omega_1 - i\tau\omega)/\Delta,$$

$$\chi_2 = \omega_M(\omega_2 - i\tau\omega)/\Delta,$$

$$\chi_a = \omega\omega_M/\Delta,$$

$$\Delta = (\omega_2 - i\tau\omega)(\omega_1 - i\tau\omega) - \omega^2,$$

$$\omega_1 = \gamma\{H_{ex}\cos(\theta) + H_K\cos[2(\alpha-\theta)]\},$$

$$\omega_2 = \gamma[H_{ex}\cos(\theta) + H_K\cos^2(\alpha-\theta)],$$

$$\omega_M = \gamma M_0, \quad H_K = 2K_{eff}/M_0. \tag{11}$$

Here, γ is the gyromagnetic constant, τ is the spin-relaxation parameter, H_K is the anisotropy field, H_{ex} is the external magnetic field along the wire axis, and θ is the angle between the dc magnetization $\mathbf{M}_0$ and the wire axis. Solving the Maxwell equations with the permeability tensor (10) allows the surface impedance to be expressed as the asymptotic series with respect to the parameter a/δ, where $\delta=c/\sqrt{2\pi\sigma\omega}$ is the nonmagnetic penetration depth. In the case of a strong skin effect $(a/\delta \gg 1)$, the longitudinal component of the impedance takes the following form:

$$\mathrm{s}_{xx} = (1-i)\sqrt{\frac{\omega}{8\pi\sigma}}\left(\sqrt{\tilde{\mu}}\cos^2(\theta) + \sin^2(\theta) + \frac{\delta(1+i)}{4a}\right), \tag{12}$$

$$\tilde{\chi} = \chi_2 - \frac{4\pi\chi_a^2}{1+4\pi\chi_1}, \quad \tilde{\mu} = 1 + 4\pi\tilde{\chi}, \tag{13}$$

where $\tilde{\chi}$ and $\tilde{\mu}$ are the effective susceptibility and permeability, respectively, defined as a response to circular excitation field h'_φ in the prime coordinate system $(x' \parallel \mathbf{M}_0)$. The first two terms of Eq. (12) correspond to Eq. (8) where $\sqrt{\mu}$ should be replaced with $\sqrt{\tilde{\mu}}\cos^2\theta+\sin^2\theta$. Therefore, the high-frequency impedance in a magnetic wire depends on both the ac permeability and static magnetization angle. However, for low-magnetic fields, with increasing frequency (beyond the ferromagnetic resonance frequency), the permeability tends to be unity and the dependence on θ vanishes. The last term in Eq. (12) gives evidence that the actual expansion parameter is δ_m/a with a magnetic skin depth determined as $\delta_m = \delta/\sqrt{\tilde{\mu}}$. In the case of amorphous wires $(\sigma \sim 10^{16}\ \mathrm{s}^{-1})$ δ_m calculated with the value of $\tilde{\mu}$ defined from Eq. (13) with parameters typical of amorphous wires is 1.2 μm. Since the permeability value decreases with frequency, the value of δ_m changes little in the frequency range 1–10 GHz. This estimation shows that the high-frequency asymptote may be not satisfactory even in this frequency range for very thin wires with the radius of few microns. In this case, the asymptotic solution for the opposite limit $(a/\delta \ll 1)$ should be used,

$$\varsigma_{xx} = \frac{k_m c}{4\pi\sigma}\frac{J_0(k_m a)}{J_1(k_m a)} + \frac{1}{54}\left(\frac{a}{\delta}\right)^4 \frac{c\mu_3^2}{\pi\sigma a}. \quad (14)$$

Here, for reader convenience, the designations of Ref. 23 are used: $k_m^2 = \mu_1(4\pi i\omega\sigma/c^2)$, $\mu_1 = 1 + 4\pi\cos^2(\theta)\tilde{\chi}$, and $\mu_3 = -4\pi\sin(\theta)\cos(\theta)\tilde{\chi}$.

In Refs. 11 and 18 it was shown that in the microwave range a weak external magnetic field $H_{ex} < H_K \leqslant 10$ Oe or a moderate stress of less than 1 GPa does not result in noticeable changes in the permeability $\tilde{\mu}$. Therefore, the change in high-frequency impedance can be realized by controlling the angle θ of the static magnetization $\mathbf{M}_0$ with respect to the wire x axis. Because of this unusual property, high-frequency magnetoimpedance was referred to as a "valve impedance" $\varsigma_{xx} \sim \cos^2(\theta)$. In the MHz range, this dependence becomes more complicated because $\tilde{\chi}$ also depends on the external magnetic field or stress.

Amorphous magnetic wires of Co-rich compositions having a small negative magnetostriction $\lambda \approx -10^{-7}$ exhibit valve impedance in the range of 100% at 1–2 GHz and they would be most suitable to realize magnetically tunable microwave composites. For a general analysis of the dc magnetization process under the effect of the external field H_{ex} and/or a tensile stress σ_{ex}, we assume that the effective anisotropy is contributed by some axial anisotropy K, an axial tensile stress $\sigma_a = \sigma_{res} + \sigma_{ex}$ which includes the residual σ_{res} and external σ_{ex} components, and also the torsion σ_t. All theses contributions to the anisotropy are considered to be uniform inside the wire. This is certainly a simplified model; nevertheless, it describes quantitatively well the experimental results on hysteresis loops and impedance. The residual stress and torsion are typically introduced by the wire drawing process with $\sigma_{res} \gg \sigma_t$. The latter, however, should not be neglected. Because of torsion, the magnetization deviates from the ultimate states, axial or circumferential, and the hysteresis process is described by a smooth function. The torsion can be partitioned into two perpendicular stresses $\pm\sigma_t$, each acting at 45° relative to the wire axis, and Eq. (9) for the magnetostatic energy U_m can be written in the following form with the equivalent uniaxial anisotropy:[15]

$$U_m = -|\tilde{K}|\cos^2(\alpha - \theta) - M_0 H_{ex}\cos\theta.$$

$$\tilde{K} = \frac{K + (3/2)\lambda\sigma_a}{\cos(2\tilde{\alpha})},$$

$$\tilde{\alpha} = \frac{1}{2}\tan^{-1}\frac{3|\lambda\sigma_t|}{|K + (3/2)\lambda\sigma_a|}. \quad (15)$$

Here, the anisotropy angle α is chosen as follows: (a) if $K + (3/2)\lambda\sigma_a > 0$, then $\alpha = \tilde{\alpha}$; (b) if $K + (3/2)\lambda\sigma_a = 0$, then $\alpha = 45°$ and $\tilde{K} = 3\lambda\sigma_t$; and (c) if $K + (3/2)\lambda\sigma_a < 0$, then $\alpha = 90° - \tilde{\alpha}$.

For a negative magnetostriction ($\lambda < 0$), small values of K and σ_t ($K < (3/2)|\lambda|\sigma_{res}, \sigma_t \ll \sigma_{res}$) the anisotropy is nearly circumferential. Applying an axial magnetic field larger than the anisotropy field H_K (which is of the order of few Oe in some CoFe-based amorphous wires) establishes the axial magnetization, thus enabling a change in the wire microwave impedance.[17,18] For such anisotropy, however, the application of a tensile stress (when $H_{ex}=0$) does not change the magnetization and hence its effect on the impedance is small. In the opposite limit when the uniaxial anisotropy constant K is large, the situation is reversed: the overall anisotropy is nearly axial anisotropy, the effect of H_{ex} (when $\sigma_{ex}=0$) is small, and the tuning parameter becomes σ_{ex}. The application of the tensile stress will rotate the anisotropy axis and the magnetization towards the circumference, which will result in the dependence of Z on σ_{ex} in zero field.[15] In the intermediate case of a helical anisotropy, both H_{ex} and σ_{ex} would cause a change in the static magnetization but the sensitivity of the impedance change would be smaller.

IV. EXPERIMENTAL

Composite materials demonstrating tunable microwave response were fabricated using CoFe-based glass-coated amorphous wires, the magnetic anisotropy of which can be modified by certain stress annealing treatments to get a preferable magnetic structure. As-prepared wires of negative magnetostrictive alloys typically have a nearly circumferential anisotropy due to a predominant axial residual stress coupled with a negative magnetostriction [$K \approx 0$, $\sigma_t \approx 0$ in Eq. (15)]. These wires will be suitable to realize field-tunable composites. To demonstrate the effect of stress on the microwave response, the wire anisotropy should be ideally established along its axis. It has recently been reported[18,25] that such anisotropy may be induced during stress annealing at a certain temperature regime and cooling under stress. It was assumed that during this process the resulting residual stress becomes compressive although annealing was performed under tension. The existence of a dominant axial anisotropy in as-prepared negative magnetostrictive CoFeSiB wires having relatively thick glass coating of 10 μm, which is nearly equal to the metal core radius, has also been reported recently.[26] It should be noted that the exact mechanism of forming a "reverse" anisotropy is not clear and could be attributed to both a creep-induced anisotropy and/or compressive stress.

A. Magnetic and magnetoimpedance characteristics of wires

We have used amorphous wires of the composition $Co_{68}Fe_4Cr_3B_{14}Si_{11}$ having a small negative magnetostriction of about -10^{-7}. For field-tunable composites, the wires having a metal core diameter of 22 μm and a total diameter of 35 μm were heated at a temperature of 160–250° C for about 10–30 min under a tension of 20–30 kg/cm^2, which results in a better established anisotropy with smaller deviation of the easy axis from circumference due to stress relaxation. For stress-sensitive composite materials, it is important to have a substantial deviation of the anisotropy from the circumferential direction (in the ideal case axial anisotropy would be preferred). The type of the anisotropy was modified by heating the wires under a larger applied tensile stress with the following slow cooling also under tension. For this case, the wires of the same composition as above were used but

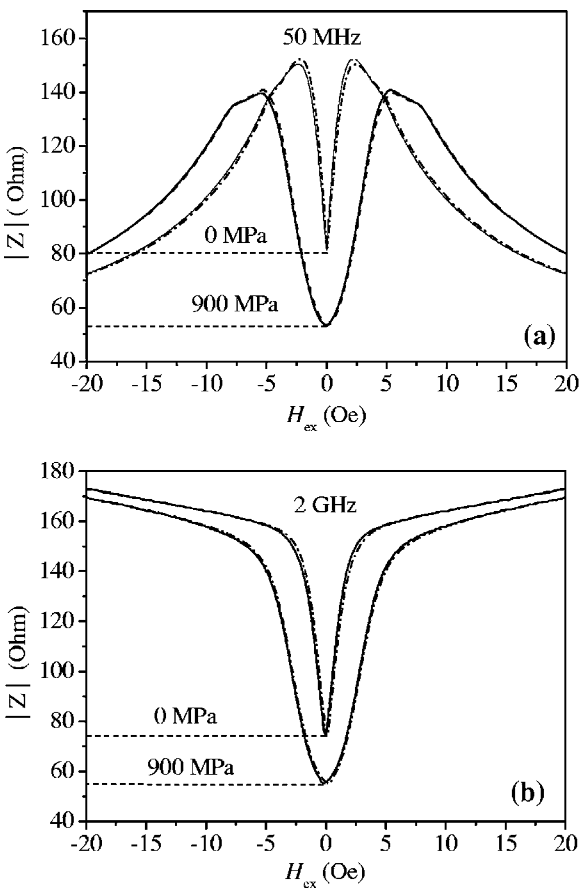

FIG. 1. Impedance plots vs external field measured in CoFeCrSiB wires with a helical anisotropy and external tensile stress as a parameter ($\sigma_{ex}=0$ and $\sigma_{ex}=900$ MPa). Frequency is 50 MHz in (a) and 2 GHz in (b). As the frequency is increased the impedance peaks seen in (a) tend to broaden and at GHz frequencies the impedance shows very gentle sloping for high fields $H_{ex}>H_K$. Applying tensile stress establishes a circumferential anisotropy and the impedance value drops at $H_{ex}=0$.

with a smaller total diameter of 20.3 μm and the metal core diameter of 14.5 μm. The wires were heated at a temperature of 250° C for 30 min under a tension of 50–70 kg/cm^2. This thermomechanical treatment creates a helical anisotropy.

The impedance $Z=\varsigma_{xx}(2l/ca)$ of a short wire sample of 6 mm in length was obtained from the S_{11} parameter (reflection coefficient) measured by means of a HP8720ES vector network analyzer with a coaxial sample holder in which the wire is placed as a central conductor. The measurements were made for frequencies from 10 MHz to 6 GHz, under different tensile stress and external field in the range ±50 Oe supplied by a solenoid parallel to the wire axis. One end of the wire is soldered to a SMA connector and the other to a non-magnetic movable bar with a micrometer screw to apply a tensile stress (up to 900 MPa). To calculate the applied stress, the Young modulus of the composite structure (amorphous metal core coated with glass) was measured in an Instron 4400 tensile/compression test system. For the wire geometry used, the Young modulus was found to be 170 GPa.

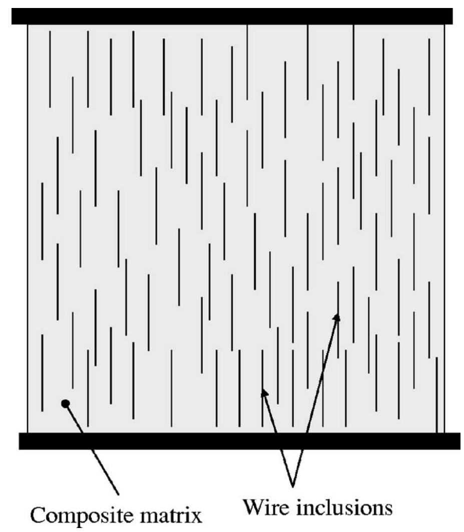

FIG. 2. Sample of composite materials with predominantly aligned wire inclusions.

The impedance plots versus H_{ex} are shown in Figs. 1(a) and 1(b) for the two frequencies of 50 MHz and 2 GHz, respectively. The impedance data are given for the wires annealed under a stress of 50 kg/cm^2 to establish a helical anisotropy, which was confirmed by hysteresis measurements. In both cases of a relatively low frequency of 50 MHz and high frequency of 2 GHz, the application of the stress of 900 MPa substantially lowers (by 45% and 36%, respectively) the value of the impedance at zero field. It also can be seen that the stress sensitivity of the impedance increases in the presence of a small magnetic field. At lower frequency of 50 MHz, the field behavior of the impedance is characterized by a minimum at zero field and two symmetrical peaks seen at the field nearly equal to the anisotropy field of 2.4 Oe for $\sigma_{ex}=0$ and 5 Oe for $\sigma_{ex}=900$ MPa. When the frequency is increased towards GHz frequency range, the impedance behavior for fields smaller than the anisotropy field changes little, but the peaks flatten and for a frequency of 2 GHz there is a very slow increase in impedance for higher fields. The impedance plots versus field seem to be typical of wires with a circumferential anisotropy or a helical anisotropy, as discussed above in Sec. III. The decrease in impedance under the effect of the applied tension can be related with decrease in the value of cos θ [see Eq. (12)], meaning that the angle between the static magnetization and the wire axis increases; thus, this indicates that a helical anisotropy exists in wires annealed under a larger stress, which transforms to a circumferential one in the presence of σ_{ex}. In the case of thicker wires annealed with a small stress, the minimum in impedance at zero field is much deeper and the applied stress has no effect on its value. In this case, the annealing treatment helps to refine the circumferential anisotropy, and the relative change in impedance is as much as 1000% per 5 Oe at a frequency of 50 MHz (data not shown).

B. Composite manufacture

The wires described above were used to fabricate two types of composite materials to demonstrate the effect of external stress and/or magnetic field applied to a whole material on microwave response. The wire pieces of 5 cm in length were situated between two sheets of elastic fabric with in-plane size of 34×38 cm^2. One layer of fabric was soaked in silicon glue, while the wire inclusions were distributed on the second dry layer. The wire pieces were predominantly aligned in one direction as shown in Fig. 2. The number of wire centers (a point at half length) per cm^2 is convenient to use as a characteristic planar concentration $\bar{p}$ of the inclusions. In the tested composite, this concentration was kept very small, not exceeding 0.5 centers per square centimeter. The wires were covered with the soaked layer to form a sandwiched structure that was then placed under a gentle press until all the glue was cured. After polymerization the structure preserves its flexibility. The total thickness of the composite sample was about 2 mm. The sample was attached to two strips of wood, which allows the uniform stress application with a load P.

It seems that the application of the external magnetic field to such a large sample would be impractical and this is why in Ref. 12 the magnetic tunability was demonstrated indirectly by using wires with different dc magnetic structure. In this work, a magnetic field as large as 12 Oe was applied to the whole composite using a specially designed coil. We have used a planar coil of size 50×50 cm^2 with the wire diameter of 0.8 mm and 50 turns mount with a relatively large step d of 1 cm. To provide the maximum uniformity of the generated field, this distance should be equal to that between the coil layers (as for the Helmholtz coil). Yet, the field alteration along the coil length remains very large. To avoid this, the coil turns in the two layers have to be shifted with respect to each other at $d/2$. If the wire inclusions are ordered in one direction, the coil turns have to be perpendicular to the wires to generate the field along them. For the wire inclusions with randomly distributed angles, the "valve-like" behavior of the magnetoimpedance $\varsigma_{xx}\sim\cos^2(\theta)$ will guarantee that for a sufficiently large magnetic field $H_{ex}>H_K$, most of the wires will be switched to the high-impedance state. In addition, the coil turns have to be perpendicular to the electric field of the incident radiating wave to ensure the wave transmission through. It should be noticed that such a coil can be easily integrated with the composite sample to constitute a single structural element. For a coil wire diameter of 1 mm and a current of 10 A, the 50×50 cm^2 coil consumes 106 W and induces a sufficiently uniform magnetic field of 12 Oe. The other way to apply the field to a large planar sample is with the help of a current bus. However, since the field would be generated in large volume (not concentrated in the surface layer), its value is small for realistic configurations and power consumption, being in the range of 1 Oe, which is less than the anisotropy field in the wires.

C. Free-space measurement technique and transmission spectra

The measurements of the effective microwave properties of composites were conducted in free space using standard technique of the through-reflection-line (TRL) calibration,

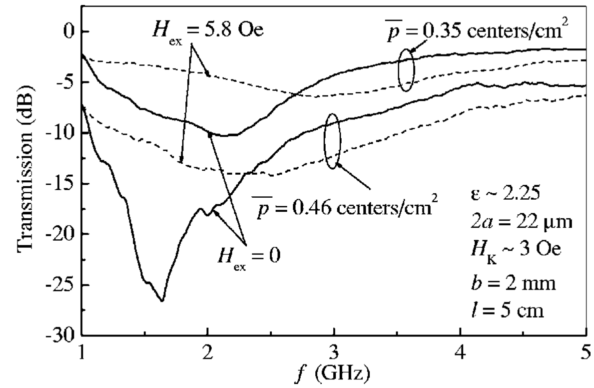

FIG. 3. Transmission spectra measured in the composite containing CoFeCrSiB amorphous wires with a circumferential anisotropy for two values of the in-plane concentration $\bar{p}$=0.35 and $\bar{p}$=0.46 centers/cm^2. The data are given for H_{ex}=0 and H_{ex}=5.8 Oe. A transmission minimum seen at H_{ex}=0 has a tendency to tail up in the presence of a sufficiently large field $H_{ex}>H_K$.

which has been successfully explored for a long time in testing dielectric materials.[27–31] The measuring system includes a pair of broadband horn antennas connected to the analyzer's ports and a mini anechoic chamber covered inside with a microwave absorber. In general, it is possible to avoid the parasitic scattering in the measuring track without use of an anechoic chamber by employing a time-domain option,[27] which enables the separation of the useful signal from background scattering. The broadband horn antennas have the following characteristics: length 887 mm, aperture 351×265 mm^2, a frequency range of 0.85–17.44 GHz, standing-wave radio (SWR)<1.7, and the effective area more than 150 mm^2 in the range of 0.85–15 GHz. The front walls of the chamber, on which the antennas were fastened, were made mobile to enable the distance between the antennas to be variable. This is necessary for the preliminary TRL calibration of the measuring track. A thin planar composite sample is fastened on fiberglass ropes in the middle of chamber at a distance of 40 cm from each horn antenna. This space guarantees the measurements in the radiative near field for the frequencies higher than 1 GHz. In our experiments we do not use focusing lenses to obtain the averaged response from many wire inclusions.

For the study of tunable properties, two types of external stimuli were applied to the composite sample: (i) dc magnetic field H_{ex} induced by a planar coil, and (ii) tensile stress σ_{com} applied to the sample frame by means of a load hung outside the chamber. The maximal value of H_{ex} was 12 Oe and the maximal value of σ_{com} was 0.1 MPa. Since silicone materials have a relatively low stiffness of 50–100 MPa the stress transferred through the matrix strain to the wires was very large, in the range of GPa, which would be sufficient to saturate the magnetization in wires with a helical anisotropy in the circumferential direction, as discussed above (see Fig. 1). A precise calculation of the stress imposed on wires would require special investigation to take into account the effect of wires on the matrix stiffness and on the adhesion between the wires and matrix. The transmission/reflection spectra are then measured and plotted for different values of

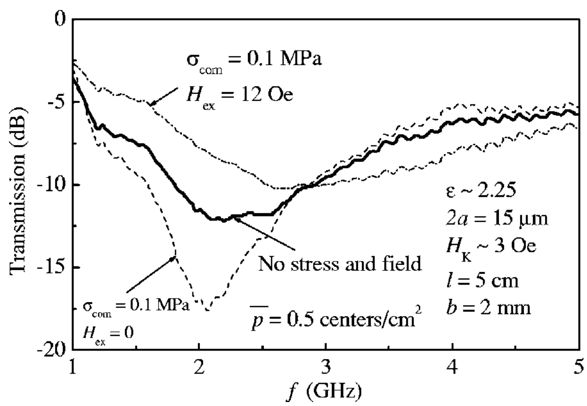

FIG. 4. Transmission spectra measured in the composite material containing CoFeCrSiB amorphous wires having a helical anisotropy with the external stress and field as parameters. The plot when no external stimuli are applied is between those with σ_{ex} and (σ_{ex}, H_{ex}) as the dc magnetization in wires changes from helical to circumferential and axial, respectively.

H_{ex} and σ_{com}. In this work, we were not in a position to deduce the effective permittivity parameter from these spectra. However, in the next section we will make comparison of the experimental results with the theoretical ones obtained on the basis of the effective permittivity model.

The spectra of the transmission coefficient (S_{21} or S_{12} parameters) through the composite sample containing wires with a circumferential anisotropy are shown in Fig. 3 for two values of the planar concentration $\bar{p}=0.35$ and $\bar{p}=0.46$ centers/cm^2 without the field and for $H_{ex}=5.8$ Oe. This field is higher than the circumferential anisotropy field in wires ($H_K \approx 3$ Oe). The frequency dispersion has a resonance character that becomes more pronounced with increasing $\bar{p}$ and when no field is applied. The minimum of $S_{21}(f)$ corresponds to the first antenna resonance in wires. The resonance frequency is seen to depend slightly on the values of the concentration and field, indicating that the resonance condition for an individual wire is held in composites as well. This is in agreement with the model of the scale-dependent effective medium,[3] and for normal incidence can be proven by a rigorous scattering theory, taking into account the interaction between wires.[32] Using the value for resonance, the permittivity of the matrix can be estimated: $\varepsilon = [c/(2lf_{res,1})]^2$, which gives the value of about 2.25 for $l=5$ cm and $f_{res,1} \approx 2$ GHz. The effect of the field on the transmission coefficient is very large near the resonance: for $\bar{p}=0.35$ centers/cm^2, the transmission increases from -10 dB up to -3.5 dB at $f=2.1$ GHz; and for $\bar{p}=0.46$ centers/cm^2, it increases from -27 dB up to -12 dB at $f=1.7$ GHz. A very large sensitivity of $S_{21}(f)$ near the antenna resonance agrees with the theoretical predictions. At the resonance, the current distribution in wires strongly depends on their surface impedance. In general, the scattering properties of wires as embedded antennas are greater if the surface impedance is low.[33] In magnetic wires, the surface impedance has a minimum when the magnetization is along the circumference which is realized for a circumferential anisotropy and $H_{ex}=0$. Thus, a deep minimum in transmission is seen when no field is applied. The surface impedance in the wires increases in the presence of the field as $\sqrt{\tilde{\mu}}$ since the magnetization rotates towards the wire axis. In this case the transmission spectrum is very broad with the resonance minimum shifted to higher frequencies. This will be explained in detail in the next section on the basis of the effective permittivity concept, providing quantitative comparison of the experimental and theoretical results.

Next, we investigated the stress effect on the transmission spectra of wire composites. In this case we have to use as inclusions wires with helical anisotropy that show stress-sensitive magnetoimpedance (see Fig. 1). The transmission spectra of such composite are shown in Fig. 4. Since the wires had also the length of 5 cm, the transmission minimum related to the antenna resonance in wires is seen at a similar frequency of 2 GHz. After the sample was loaded and the stress was applied to the whole composite, the transmission minimum deepens, changing from -12 dB down to -17.5 dB. The application of tensile stress via the matrix strain to wires with a negative magnetostriction establishes the circumferential anisotropy, and therefore lowers the wire surface impedance. As a result, the scattering at the resonance increases and the rate of transmission is low. While the sample was under stress, a relatively large external field is applied to overcome the effect of applied stress and saturate the magnetization along the axis. Estimating the value of stress imposed on wires to be in the range of GPa, this field will be sufficient to saturate the wire magnetization along the axis, thus realizing the magnetic state with the maximum surface impedance. In this case the transmission coefficient becomes about -7 dB. Therefore, we have demonstrated that rotating the magnetization in the wires from a circumference to the axis, which increases the surface impedance in approximately $\sqrt{\tilde{\mu}}$ times, and changes substantially the transmission spectra: from that having a narrow and deep resonance minimum to those with broader dispersion and increased transmission.

It should be noticed that the transmission at minimum seen in Fig. 4 is greater than that in Fig. 3; however, the planar concentration is larger. In this case we used wires with smaller diameter so the volume concentration is smaller. Therefore, there are in general a number of critical parameters to control the microwave response, such as the wire geometry including its length and diameter, the wire concentration in the composite, and the matrix thickness and its permittivity. We believe that by optimizing the system it would be possible to realize tunable microwave selective surfaces.

V. THEORETICAL SCATTERING SPECTRA AND COMPARISON WITH EXPERIMENT

The transmission experiments will now be analyzed quantitatively on the basis of the effective permittivity concept as described in Sec. II. We will demonstrate that this simple approach works well. In general, it is not possible to assign the effective parameters to a noncontinuous material unless its structural scale is much smaller than the wavelength. In the considered composites, the wire length is of the order of the wavelength to realize the condition of the antenna reso-

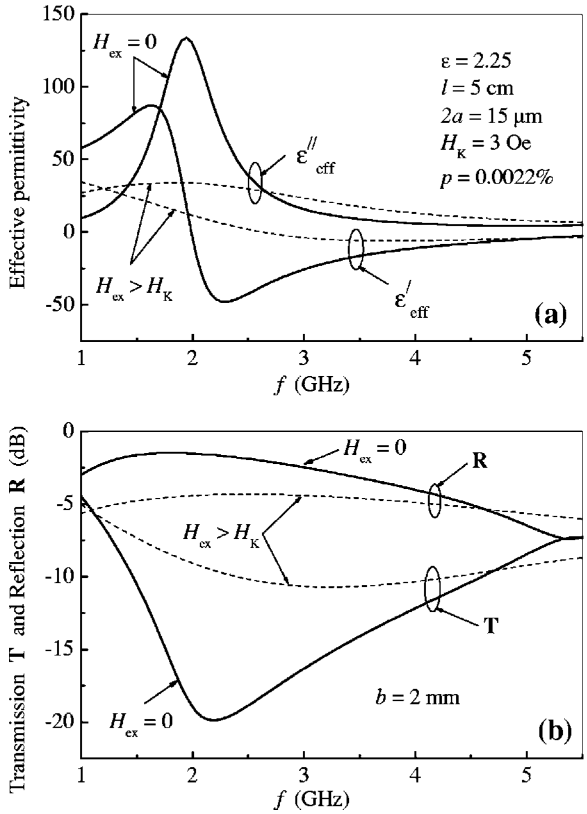

FIG. 5. Theoretical dispersion of the effective permittivity in (a) and coefficients of transmission and reflection in (b) for a composite containing wires with a circumferential anisotropy. The plots are shown for two limiting cases: $H_{ex}=0$ (circumferential magnetization and low impedance) and $H_{ex}>H_K$ (axial magnetization and high impedance). Notice that there exists a frequency band with a negative real part of ε_{eff} for $H_{ex}=0$ where transmission is strongly suppressed. Calculation parameters are given in the figure inset.

nance. Yet, the microwave response from such composites can be treated as from continuous media. This was validated by numerical simulations and experiments,[4,6,32,34] and could be proven by a rigorous analytical method.[32] Furthermore, we will demonstrate that the experimental transmission spectra are consistent with the frequency-dependent form of the permittivity ε_{eff}, comprising the averaged wire polarization induced by the incident wave. We will also demonstrate that the dependency of ε_{eff} on the magnetic properties of the wires predicted by theory quantitatively describes the change in transmission seen experimentally under the effect of the external magnetic field or stress.

The concept of the effective permittivity of wire-based composite, which is sensitive to magnetic properties of the inclusions, was developed in Ref. 11. Here we calculate ε_{eff} using simplified Eq. (3) with structural and magnetic parameters relevant to the experiment. It should be noted that for the experimental configuration (predominantly aligned wires), ε_{eff} will be of a diagonal tensor form with only one component different from the matrix permittivity. The magnetic anisotropy in wires is taken as circumferential and the controlling parameter is an axial magnetic field which will saturate the magnetization along the wire axis, thus ensuring the necessary magnetization change for low and high surface impedances. The permittivity spectra are shown in Fig. 5(a). It is seen that the plots for $H_{ex}=0$ (circumferential magnetization) have a resonance behavior with a resonance frequency of 2 GHz for $l=5$ cm, a large absorption peak, and a negative real part of the permittivity in a wide frequency band past the resonance. In the presence of the magnetic field larger than the anisotropy field which saturates the magnetization along the axis and increases the wire surface impedance [see Eqs. (12)–(14)], the dispersion of ε_{eff} becomes a relaxation type with a gradual decrease in the real part, which is always positive and with very broad losses. At the resonance frequency, the dielectric loss parameter decreases more than six times in the presence of the field.

We now compute the transmission T and reflection R coefficients for waves incident normally to a slab of thickness $b=2$ mm of continuous material with the permittivity function shown in Fig. 5(a). Their frequency plots for $H_{ex}=0$ and $H_{ex}>H_K$ are given in Fig. 5(b). The comparison of the theoretical transmission spectra and the experimental ones seen in Figs. 3 and 4 shows a very good agreement for both magnetic states in the wire. In the case of the circumferential magnetization which exists in wires with a circumferential anisotropy ($H_{ex}=0$) or in wires with a helical anisotropy under tensile stress, the transmission is very low at the level of −20 dB for certain wire concentration in the vicinity of the resonance. Both high losses and negative values of the real part of the permittivity strongly oppose the wave propagation. Establishing the magnetization along the wire axis by applying a sufficiently high magnetic field changes the effective permittivity and recovers the wave propagation. The transmission coefficient becomes in the range of −8 dB. For fixed values of the wire length and the permittivity of matrix, precise values of T will depend on the wire concentration and its diameter $2a$. The theoretical results are given for $2a=15$ μm and $p=0.0022\%$, which correspond to the experimental data of Fig. 3. The agreement in this case is surprisingly very good considering the complexity of the material and many parameters involved.

It should be noticed that the reflection spectra does not show very pronounced resonance behavior and they are much less sensitive to the magnetic structure of the wire. The reason is the existence of absorption. A similar tendency was observed in the experiment. It seems to be a great deficiency for a number of practical applications, including stress monitoring of large civil structures for which reflection parameter would be measured. However, changing the structural parameters of the composites such as the matrix permittivity ε, the wire diameter $2a$, and its length l, it is possible to realize a more pronounced dispersion of the reflection coefficient R with a high sensitivity to the magnetization direction in wires. This will require larger values of ε and $2a$, and also smaller l to keep resonance at similar frequency. An example of such behavior is shown in Fig. 6.

The considered composites will also exhibit the reversal of phase in the reflected wave from π to $-\pi$ at a frequency near the peak of the imaginary part of the effective permittivity. Since the position of this peak shifts towards higher frequencies when the magnetization in wires is rotated along its axis (by applying a magnetic field higher than the anisotropy field), the phase reversal also will occur at higher fre-

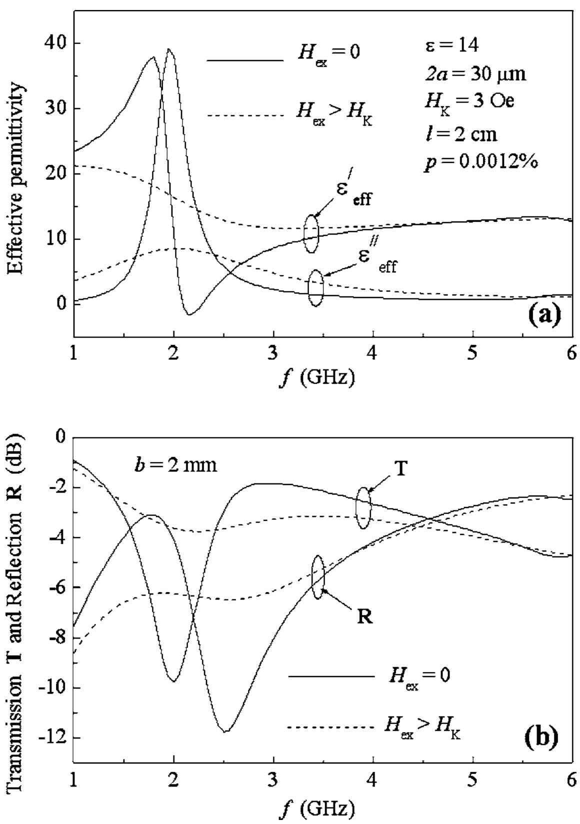

FIG. 6. Theoretical dispersion of the effective permittivity in (a) and coefficients of transmission and reflection in (b) for a composite with high-permittivity matrix $\varepsilon=14$ containing thicker wires with a metal core diameter of 30 μm. The plots are shown for two limiting cases of $H_{ex}=0$ and $H_{ex}>H_K$. Both reflection and transmission parameters show a resonance dispersion and high sensitivity to the magnetic state of the wires.

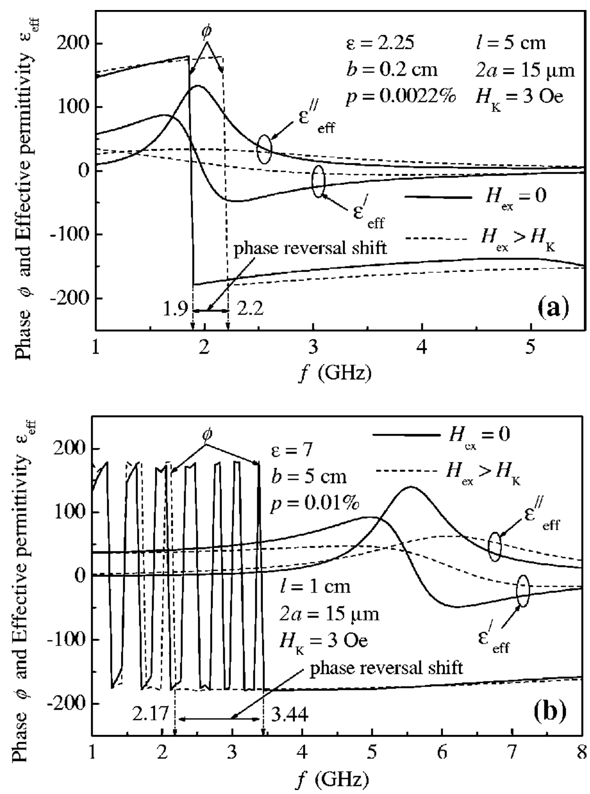

FIG. 7. Theoretical dispersions of the phase of the reflection coefficient for thin (a) and thick (b) composites, respectively, with the external field as a parameter. For comparison, the permittivity dispersion is given as well.

quencies, as shown in Fig. 7(a). This frequency shift could be conveniently used in realistic applications such as stress monitoring, where the transformation of the dispersion curves will be similar but due to the tensile stress. A similar behavior of the reflective coefficient phase was recently reported in the high-impedance surfaces with mechanically reconfigurable resonance elements.[35] The phase effect is much greater in thicker composite samples allowing multiple reflection. Figure 7(b) demonstrates the dispersion of the reflective coefficient phase for a composite sample 5 cm thick and having $\varepsilon=7$, which is typical of a dry concrete. The volume concentration of the wire inclusions is taken larger ($p=0.01\%$) since in this case a high absorption would not be a problem. The phase has frequency oscillations because of multiple reflections that become not possible at the onset of the dispersion in the effective permittivity characterized by an increase in the dielectric loss (10 dB attenuation criterion). In the presence of a magnetic field large enough to saturate the wire magnetization along the axis, the dispersion of ε_{eff} broadens and the frequency oscillations stop at a much lower frequency. For the considered parameters, the frequency shift is more than 1 GHz.

We have demonstrated the magnetic tunability of the phase parameter on the example of a composite sample having magnetic wires with a circumferential anisotropy. Consequently, a controlling parameter was a magnetic field. In composites having wires with a helical anisotropy, tuning could be realized by applying a tensile stress. Therefore, this method could be used for sensitive remote stress measurements analyzing the frequency trace of the reflected wave phase.

VI. CONCLUSION

In this work, we have demonstrated tunable microwave composites containing short CoFeCrSiB glass-coated wires through the direct free-space measurements and effective medium analysis. The transmission spectrum of the proposed wire medium may have a sufficiently narrow band gap associated with the antenna resonance in wires. A striking feature of this behavior is that changing the magnetic structure in wires by applying a weak magnetic field or stress influence the antenna resonance and opens bandpass. The achieved tunability typically exceeds 10 dB for external fields of few Oe. The reflection spectrum also alters, depending on the magnetization in wires. A particular interest for practical application seems to be a considerable shift in the frequency of the phase reversal of the reflected wave. We have briefly discussed a number of applications predicting that the magnetic wire composites are likely to have a large impact on the technology of remote sensing and control. Certainly, further intensive investigations are needed to fully characterize the potential of the proposed microwave materials.

We also have demonstrated that it is appropriate to treat the wire composite medium as a homogeneous material with frequency-dispersive effective permittivity. This reduction in complexity is essential to further understanding of microwave properties of magnetic wire composites including prediction of new effects. One example is the case of composites with wires having a nonzero off-diagonal component of the surface impedance. It should be possible to excite the wires with a magnetic field of the incident wave, thus realizing the "optical" activity typical of chiral structures.[36]

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