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Metamaterial near-field sensor for deep-subwavelength thickness measurements and sensitive refractometry in the terahertz frequency range

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We present a metamaterial-based terahertz (THz) sensor for thickness measurements of subwavelength-thin materials and refractometry of liquids and liquid mixtures. The sensor operates in reflection geometry and exploits the frequency shift of a sharp Fano resonance minimum in the presence of dielectric materials. We obtained a minimum thickness resolution of 12.5 nm (1/16 000 times the wavelength of the THz radiation) and a refractive index sensitivity of 0.43 THz per refractive index unit. We support the experimental results by an analytical model that describes the dependence of the resonance frequency on the sample material thickness and the refractive index.

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Terahertz (THz) radiation has attracted considerable attention during the last decade. This spectral range (frequencies between approximately 0.1 and 10 THz) is of special interest for sensing applications because many substances have a specific spectral response in this frequency interval.1,2 The detection of small amounts or very thin layers of a sample material, however, remains a challenge in THz sensing technology. To increase the interaction of the THz field with the sample, several approaches have been proposed recently.3–11 One particularly promising option to increase the sensitivity of THz measurements is the use of metamaterials, artificial materials whose electromagnetic response is determined mainly by the size and shape of subwavelength-sized inclusions. Due to the strong localization of the electromagnetic fields in the vicinity of resonant metallic elements, metamaterials exhibit a strong change of their optical response when a sample material is present. This property may be exploited to construct sensors which promise superior sensitivity. Examples of metamaterial-assisted sensing have been demonstrated in the microwave, THz, and near-infrared frequency ranges in the past years.12–20

Metamaterial-based sensors offer some important advantages over standard terahertz time-domain spectroscopy (THz-TDS). When calculating optical constants from THz-TDS measurements, one has to take great care concerning the influence of signal noise on the calculated values.21 Furthermore, in transmission geometry, the thickness of the sample has to be known exactly. In reflection geometry, an additional uncertainty is the phase of the reference spectrum. These limitations can be overcome by metamaterial-based sensors which allow to derive the refractive index and the thickness of a sample material from frequency measurements rather than from amplitude measurements. As a main advantage over amplitude measurements, frequency measurements are less prone to noise and do not require the knowledge of a reference spectrum, provided that the spectrum of the THz source is smooth in the spectral range of interest.

In this letter, we present a metamaterial-based sensor whose reflection shows a strong frequency shift of a Fano-type resonance minimum in the presence of a dielectric sample. The magnitude of this shift depends on both the refractive index and the thickness of the sample. The metamaterial is designed to operate at frequencies between approximately 1 and 1.7 THz.

The unit cell of the metamaterial consists of four metallic crosses on top of a 10 μm thick dielectric matrix with a relative permittivity εr = 2.67. The unit cell is square with an edge length of 140 μm. Each of the crosses is tilted by an angle of 22.5°. The geometry parameters are shown in detail in Fig. 1(a). Despite the relatively large unit cell, diffraction by the metamaterial occurs only for frequencies greater than approximately 2.1 THz because the first order of diffraction is zero due to the symmetry of the structure. Figures 2(a) and 2(b) show the experimentally measured THz reflection along with corresponding numerical calculations based on the finite integration technique (FIT) provided by CST Microwave Studio®. The spectrum shows a pronounced, narrow reflection minimum. The advantage of operation of the sensor in reflection geometry is that the sample only has to be accessible from one side, which is especially important when measuring liquids. Furthermore, in contrast to transmission measurements, the absorption of the sample does not reduce

FIG. 1. (a) Geometry parameters of the metamaterial unit cell. a = 140 μm, w = 15 μm, g = 4 μm, θ = 22.5°. The metamaterial consists of gold (Au) crosses on top of a BCB film. (b) Distribution of charges (+, −) and currents (arrows) at resonance when excited by a horizontally polarized incident THz wave. (c) Microscope image of a fabricated metamaterial.

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the signal amplitude when measuring reflection. A narrow resonance feature is necessary to accurately determine the magnitude of a frequency shift and thus increases the resolution of the sensor material. In our design, the spectral width \( \Delta \omega \) (full half-width) of the reflection minimum with respect to the resonance frequency \( \omega_0 \) is \( \Delta \omega/\omega_0 \approx 1/26 \).

In the region around the resonance frequency, the reflection spectrum displays a Fano profile.\(^{22} \) It has been shown that this specific lineshape may be caused by a narrowband “trapped mode” that obtains weak coupling to free-space radiation from a symmetry breaking in a metamaterial structure.\(^{23} \) In the metamaterial design presented here, a mode with inversely phased currents along the arms of neighboring crosses can be excited due to the tilting of the crosses. Figure 1(b) shows the distribution of currents and charges of this mode when excited by a horizontally polarized THz wave. The function

\[
\tilde{E}(\omega) = \frac{q \Gamma/2 + \omega - \omega_0}{\omega - \omega_0 + i\Gamma/2} E_0(\omega)
\]

(1)

fits the frequency-dependent complex electric field amplitude of the experimental and numerical data (see Figs. 2(a) and 2(b)), where \( \omega \) is the frequency, \( E_0(\omega) \) is a baseline with linear dependence of the magnitude and the phase, and \( \Gamma \) is the width of the resonance. The complex Fano parameter \( \tilde{q} \) describes the asymmetry of the resonance profile and the amount of dissipation in the system,\(^{24} \) which is directly related to the depth of the resonance minimum.

The shift of the resonance frequency in the presence of a sample material can be explained using an intuitive analytical model which describes the sensor and the sample material as a layered system of homogeneous media (Fig. 3(a)). The normal on the layer surfaces is defined as the \( z \) direction and the direction parallel to the electric field vector of the incident wave as the \( x \) direction. The metamaterial sensor itself can be understood as an inductor-capacitor (LC) circuit. Its resonance frequency \( \omega_0 = (L_{\text{eff}} \times C_{\text{eff}})^{-1/2} \) is determined by an effective inductance \( L_{\text{eff}} \) and an effective capacitance \( C_{\text{eff}} \). Because we only consider nonmagnetic sample materials with a relative permeability \( \mu_r = 1 \), the effective inductance of the system is expected to be independent of the sample material. The effective capacitance, however, is influenced by the relative permittivities of the sensor and the sample material and can be described as

\[
C_{\text{eff}} = \frac{Q_{\text{eff}}}{U_{\text{eff}}} = \int \epsilon_r(z) E(z) \cdot dA = \int \epsilon_r(z) E(z) \cdot \hat{x}dz,
\]

(2)

where \( Q_{\text{eff}} \) is the charge in the considered volume \( V \), \( dA \) is the area element of the boundary of \( V \), \( \epsilon_0 \) is the vacuum permittivity, \( U_{\text{eff}} \) the voltage between the points \( P_1 \) and \( P_2 \) on two opposing cross arms, and \( \hat{x} \) the unit vector in \( x \) direction. Note that \( \epsilon_r(z) \) reflects the spatial dependence of the relative permittivity in the sensor and the sample material. For the \( z \) dependence of the electric field \( E(z) \) we assume that the electric field amplitude decreases exponentially with increasing distance from the metallizations as shown in Fig. 3(a).

\[
k_c(z) = \sqrt{(\epsilon(z)/\epsilon_0)} / \kappa_0,
\]

where \( \kappa_0 \) is the speed of light in vacuum, and \( k_0 = 2\pi / (70\mu m) \) is the transverse wave number of the first non-zero Fourier component of the structure. Note that \( k_c \) depends on the permittivity of the dielectric layers and thus possesses a \( z \) dependence. Reflections at the boundaries between the layers are neglected.

While we cannot reliably quantify the absolute value of the effective capacitance in Eq. (2) without knowing the exact field distribution of the resonant structure, we can calculate a relative effective capacitance for each layered system of materials surrounding the plane of the metallizations.
This enables us to calculate the frequency shift in the presence of a sample material. The resonance frequency $\omega_n$ with a sample material on top of the sensor is then $\omega_n = \omega_{n0} \times \sqrt{C_{\text{eff}}(\omega_n)/C_{\text{eff}}(\omega_0)}$, where $\omega_0$ is the resonance frequency without the sample material. Because $C_{\text{eff}}$ depends on the frequency, we employ an iterative method to determine $\omega_n$.

In order to test the validity of this simple analytical model, we performed two series of full-wave numerical calculations. In the first series, we calculated the reflection spectrum and the resonance frequency of the sensor in the presence of thick layers of dielectric sample materials with a refractive index of 1 and 5 ($\epsilon_r$ between 1 and 25). In the second set of calculations, we investigated the resonance frequencies of the sensor with dielectric layers with constant refractive index $n = 3.42$ and thicknesses between 50 and 1 mm. Figures 3(b) and 3(c) show the dependence of the resonance frequencies on the refractive index and the thickness of the sample material. In both graphs we compared the resonance frequencies obtained from full-wave calculations with the predicted values of the model. Because the model provides no information about the absolute value of the resonance frequency, the resonance frequency without sample material was set to match the value derived from the full-wave calculations. Within the investigated range of refractive indices and layer thicknesses of the sample materials, the resonance frequencies of the model and the numerical calculations agreed very well. The remaining small deviations can be explained by the fact that we neglected more strongly decaying near field components with larger transverse wave numbers.

We fabricated the metamaterial sensors by UV lithography. We used a 10 $\mu$m thick layer of benzocyclobutene (BCB) as a dielectric background material. On top of this layer, we evaporated a 200 nm thick gold structure. We then removed the silicon substrate which we used during the fabrication process. Figure 1(c) shows a microscope image of a fabricated sensor. Details on the fabrication process can be found in an earlier publication.

A requirement on the metamaterial design was robustness against deviations of the structure parameters which may occur during the fabrication process. A slight variation is expected because of the sensitivity of the lithography process with respect to the exposure and development times. We estimated the standard deviation of the resonance frequencies of individual sensors to be smaller than 5 GHz. This means that imperfections and deviations from the nominal design which arise during the fabrication process pose virtually no restrictions on the accuracy of the frequency measurement.

We optically characterized and tested the performance of the sensors by measuring the reflection spectra using THz time-domain spectroscopy. We employed photoconductive switches as emitter and detector. By using a 10 mm thick high-resistivity silicon wafer as a beam splitter, we measured the reflection spectra of the sensors under normal incidence with a spectral resolution of approximately 5 GHz. We took all measurements in a dry air atmosphere (relative humidity <4%) at room temperature (21°C).

In order to experimentally prove the capability of the sensor of measuring the thickness of thin sample materials, we evaporated silicon layers ($n \approx 3.4$) with thicknesses between approximately 50 nm and 1 $\mu$m on top of the metamaterial sensor. Prior to the THz measurements, we measured the thickness of the silicon layers using a surface profilometer. To assist in the fabrication process, we covered the silicon with an additional sub-10 $\mu$m thick layer of BCB. This created an additional frequency shift but did not influence the functionality of the sensor. Figures 4(a) and 4(b) show the variation of the resonance frequency of the sensor for different thicknesses of silicon. We observed frequency shifts in a range from 1.52 THz without silicon to 1.32 THz for a silicon thickness of 1070 nm. The measured frequency shifts agree very well with both the numerical calculations and the predictions of the analytical model. From Fig. 4(b) we read that the sensitivity of the sensor has a maximum value of approximately 0.4 THz/$\mu$m for very small layer thicknesses. Assuming a frequency resolution of 5 GHz, this yields a thickness resolution of 12.5 nm, which corresponds to approximately 1/16 000 of the THz wavelength ($\lambda \approx 200 \mu$m).

In a second series of experiments, we investigated the influence of different liquids and liquid mixtures on the resonance frequency of the metamaterial sensor. We brought the liquids in direct contact with the metallic structure of the sensor. To provide the sensor with higher mechanical stability, we glued it on a 5 mm thick polytetrafluoroethylene (PTFE) substrate. We deliberately chose PTFE as a substrate because of its low refractive index ($n \approx 1.4$) and low absorption at THz frequencies. A substrate with a high index of refraction (such as silicon with $n \approx 3.4$) would have considerably reduced the sensitivity of the sensor.

The liquids
under investigation were isopropanol, glycerin, paraffin, ethanol, butanol, rapeseed oil, cyclohexane, and N-ethyl-2-pyrrolidone (NEP). In addition to the pure liquids, we measured the reflection spectrum of the sensor for different mixtures of isopropanol and glycerin as well as ethanol and water. To calibrate the metamaterial sensor, we determined the refractive indices of the liquids and liquid mixtures in an alternative experimental configuration. For this purpose, we measured the reflection spectra of the liquids through a silicon window. For each liquid or liquid mixture, we obtained the reflection spectra at the air/silicon interface and the silicon/liquid interface. Based on these measurements, we established the relation between the resonance frequency of the sensor and the refractive index of the liquids (Fig. 5). In a refractive index range between 1 and 1.8, the dependence between resonance frequency and refractive index is almost linear with a slope of approximately $-0.43$ THz per refractive index unit. Assuming an error of 5 GHz for the resonance frequency shift of the detector, we could reliably detect refractive index differences as small as 0.01 with the metamaterial sensor. It should be mentioned that such a high resolution can only be achieved for weakly absorbing sample materials. In general, the resolution of the sensor decreases with increasing loss in the sample material. For all investigated sample materials, however, the refractive index resolution was better than 0.09.

In conclusion, we presented a metamaterial-based terahertz sensor for the measurement of thin sample materials with subwavelength thickness and for refractive index sensitive measurements of liquids and liquid mixtures. As a measurement signal we exploited the frequency shift of a sharp Fano resonance in the reflection spectrum of the metamaterial in the presence of a dielectric. The sensor has been devised such that the sample material under investigation faces the metallic structure of the metamaterial while the THz beam is incident from the opposite side. As a major advantage of operation in reflection geometry, the THz beam need not be transmitted through the sample material, thus ensuring low absorption and an increase of the signal-to-noise ratio. For silicon as a sample material we observed a frequency shift of $0.4$ THz/μm, which corresponds to a thickness resolution of 12.5 nm (1/16 000 times the wavelength of the THz radiation). Furthermore, we determined the refractive index of various liquids and liquid mixtures with different mixing ratios. With the sensor, we obtained a refractive index resolution of up to 0.01 by measuring the resonance shift of the detector with a slope of $-0.43$ THz per refractive index unit. We explained the physical behavior of the detector by an analytical model which predicted the correct dependence of the resonance frequency shift on the sample material thickness and the refractive index.

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