Vector frequency-comb Fourier-transform spectroscopy for characterizing metamaterials

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Abstract. We determine infrared transmission amplitude and phase spectra of metamaterial samples at well-defined incidence and polarization with a vector (‘asymmetric’) frequency-comb Fourier-transform spectrometer (c-FTS) that uses no moving elements. The metamaterials are free-standing metallic hole arrays; we study their resonances in the 7–13 $\mu$m and 100–1000 $\mu$m wavelength regions due both to interaction with bulk waves (Wood anomaly) and with leaky surface plasmon polaritons (near-unity transmittance, coupling features and dispersion). Such complex-valued transmission and reflection spectra could be used to compute a metamaterial’s complex dielectric function directly, as well as its magnetic and magneto-optical permeability functions.

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1. Introduction

Metamaterials are artificially structured materials designed for specific electromagnetic properties; they were known as ‘artificial dielectrics’ in early microwave electronics. Metamaterials with a negative refractive index \( n \), suggested by Mandelshtam (1950) and Veselago (1968), have in the past been experimentally demonstrated for microwaves (Smith et al 2000) up to THz frequencies (Yen et al 2004). Much activity is presently devoted to their realization for infrared and visible radiation (Dolling et al 2006).

A negative \( n \) requires both electric permittivity \( \varepsilon < 0 \) and magnetic permeability \( \mu < 0 \). The basic design of a negative-index metamaterial is to incorporate subwavelength-sized elements exhibiting either electric or magnetic resonance, and to arrange both types in a regular lattice with a subwavelength-size period. A microscopic optical characterization of such elements is possible by scattering-type scanning near-field microscopy (s-SNOM); this method has already enabled mapping of the electric near-field distribution surrounding resonant structures at sub-micrometer resolution (Hillenbrand and Keilmann 2001, Hillenbrand et al 2003, Jia et al 2008, Valk and Planken 2002, Yu et al 2007, Zentgraf et al 2008).

The optics application of a metamaterial depends, however, crucially on its far-field effects which relate to its spatially averaged optical properties, its electric permittivity and its magnetic permeability. Far-field optical methods can usually determine two observables simultaneously, for example transmittance and reflectance, or amplitude and phase in interferometry, or psi and delta in ellipsometry. The measurement of these two quantities then enables determination of two optical constants, usually \( \varepsilon_1 = \text{Re}(\varepsilon) \) and \( \varepsilon_2 = \text{Im}(\varepsilon) \), at each wavelength. Since metamaterials have at least four independent optical constants (the real and the imaginary parts of both \( \varepsilon \) and \( \mu \)) their complete characterization requires more measurements, for example, at two different angles of incidence \( \alpha \) which, of course, require a well-defined collimated beam. Higher complexity arises with metamaterials containing low-symmetry elements such as split rings which induce bianisotropy, requiring more measurements for the determination of their additional, magneto-optical permittivity properties (Padilla et al 2006).

In the near-infrared, broadband interferometric techniques have been used to measure negative-index metamaterials (Dolling et al 2006, Zhang et al 2005). An ideal instrument for far-infrared characterization is a pulsed-laser-based broadband THz spectrometer, allowing vector spectra with a collimated beam (Adelberger and Cheung 1985, Mittleman 2003). It uses a sampling detector for tracing out the electric field oscillation of the THz pulse with a mechanical delay stage, and has been extended to even mid-infrared frequencies.
Figure 1. Optical layout of vector THz spectrometer using the ASOPS technique. To record complex THz amplitude and phase spectra, a trigger pulse is derived to mark the temporal overlap of the pulse trains from both lasers, generated here by cross correlating (CC) two sample beams split off the laser beams in a BBO crystal. (Kübler et al 2005) yet metamaterials have been studied with this technique only in the far infrared (Azad et al 2008, Gomez Rivas et al 2003, Winnewisser et al 1999).

We suggest here metamaterial characterization by two recently introduced, coherent mid-infrared and THz spectrometers which contain no moving parts. Their laser-like beam can be well collimated, they measure vector information, and the absence of mechanical motion allows high-speed acquisition. The spectrometers’ common, basic principle is the use of two pulsed laser beams with slightly different repetition frequencies, \( f_r \) and \( f_r + \Delta \) (van der Weide and Keilmann 1998).

2. Far-infrared experiment

In a first configuration for THz vector spectrometry, one laser generates a THz beam (magenta in figure 1), whereas the other is used for asynchronous electro-optic sampling of the THz pulse shape (ASOPS) (Bartels et al 2006, 2007, Yasui et al 2005, Yasui et al 2006). Such an ‘asymmetric’ THz spectrometer has recently illuminated a scattering-type scanning THz near-field microscope (von Ribbeck et al 2008). Here, we show that it can characterize metamaterials in a short time, which could be useful in studies of dynamic metamaterials (Chen et al 2008, Driscoll et al 2008). Our setup uses two 5W-laser-pumped Ti:S oscillators (FemtoSource Compact Pro, Femtolasers) emitting 10 fs pulses at 800 ± 50 nm wavelength, and \( f_r = 125.11 \) MHz. The first beam is focused with a 100 cm focal length lens onto a GaAs emitter (Tera-SED, Gigaoptics) biased at 10 V, to produce a 0.3–3 THz beam. This is collimated by a paraboloidal mirror with 25 mm effective focal length and, fully reflected by an ITO-coated glass slide (TS-GSHR, Bioscience Tools) which transmits the second Ti:S laser beam. Both are focused with a paraboloidal mirror with 25 mm effective focal length onto a 1mm thick ZnTe crystal. The THz beam travels 25 cm in total. The sampling beam is split by a polarizing cube, and the signal of a differential detector (2107, New Focus) is recorded on a scope (WaveSurfer 422, LeCroy). In order to retrieve the phase information, ASOPS transients are recorded together.
Figure 2. THz spectroscopy at normal incidence of a $t = 1$ mm thick brass plate with a nominally square array of $d = 204 \mu$m dia. holes (inset: electron micrograph); (a) transients (averaged over 50 repeats, acquired within 1.4 s) with (red) and without (black) sample; (b) amplitude and phase spectra without sample, obtained by Fourier transforming the black transient in (a), showing signatures due to atmospheric absorption in the 25 cm long THz beam path (magenta in figure 1); (c) complex transmission spectrum of holey brass plate showing superluminal phase velocity and strong attenuation in the cutoff region ($< 0.86$ THz), as well as high transmittance in the resonance region (0.9–1.1 THz); diffraction at the sample sets in at $c/g = 1.22$ THz; (d) polar plot of the complex transmission spectrum (c), with the frequency as parameter (amplitude in linear scale, from 0 to 0.8).

with trigger pulses. These are obtained as before (Brehm et al 2006, Schliesser et al 2005) by a BBO cross-correlator (CC in figure 1), which in this experiment is driven by two beams split off from the periphery of each laser beam by mirrors with sharp edges.

A piezoelectric transducer in one of the Ti : S lasers allows us to precisely set the offset frequency $\Delta$, as before (Keilmann et al 2005, Schliesser et al 2005). We use $\Delta = 375.3$ Hz in order that the time dilatation factor becomes $f_t/\Delta = 333333$, such that a 1 THz oscillation appears down-sampled as a 1 THz/333 333 = 3 MHz oscillation. Transient pulses accordingly repeat at $\Delta = 375.3$ Hz rate, but much higher repetition rates could be obtained by setting a higher $\Delta$, or by manipulating one of the lasers (Keilmann et al 2005, Schliesser et al 2005). We record the transients for 20 $\mu$s to obtain spectra at (333 333/20 $\mu$s) = 0.017 THz resolution, and perform on-line averaging of 35 transients s$^{-1}$, as limited by our oscilloscope, before Fourier transformation. The THz spectra (figure 2(b)) exhibit absorption lines from

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the laboratory atmosphere. The phase is seen to decrease when an absorption resonance is approached from the low-frequency side, due to the increased refractive index, in accord with the sign convention of $\exp(2\pi(f t - x/\lambda))$ for a wave propagating in the $x$-direction. The molecular lines cancel out completely when transmission spectra are calculated as ratios of spectra with and without sample (figure 2(c)), illustrating the excellent repeatability and resolution of the spectrometer.

3. Far-infrared results

We study a metallic metamaterial consisting of a square array of cylindrical holes drilled at a nominal period of $g = 246 \, \mu m$ into a $t = 1 \, mm$ thick brass plate. The sample exhibits a broadband of high transmission (enhanced by $\approx 20\%$ over the areal fraction of the holes) near 1 THz, and this is accompanied by a monotonic phase change (figure 2(c)) which leads to an approximately circular trajectory in the complex transmission plane (figure 2(d)). Note this band is in the sub-diffractive frequency region $f < c/g$, well below the Wood anomaly given by the Rayleigh condition (Rayleigh 1907) $f_{\text{wood}} = c/g = 1.22 \, \text{THz}$ that marks the onset of scattering into free-space diffraction orders. We assign this band to the leaky-wave resonance of a periodically corrugated or perforated metallic slab, here at frequency $f_{\text{res}} \approx 1 \, \text{THz} = 0.82 \, c/g$. This resonance has first been analyzed and explained in the remarkable, pioneering work (Ulrich 1974) to arise from the interaction of the beam with modified Zenneck waves (Zenneck 1907) propagating in the plane of the guiding slab. Ulrich demonstrated in (Ulrich 1974), both in theory and experiment, that the periodic corrugations generate photonic Bloch waves and also a photonic band structure, and thereby he initiated no less than the physics of photonic crystals. In particular, he derived that on one hand there exist truly trapped or guided modes he named $s_1$ which are completely confined by the slab without any coupling to external radiation (like the ‘spoof plasmons’ on a corrugated metal surface of a later derivation (Pendry et al 2004) that, unfortunately, did not refer to Ulrich’s work). Further he demonstrated that the slab also supports leaky modes—named ‘guided resonances’ in a related study (Fan and Joannopoulos 2002) and in a review (Garcia de Abajo 2007) of ‘enhanced’ optical transmission of hole arrays that unfortunately, also do not reference to (Ulrich 1974). These are directly connected to external radiation by the grating-coupling mechanism since the in-plane wavevector needs only to be conserved up to a reciprocal lattice vector. A leaky surface wave thus consists of two coupled parts, an evanescent surface wave and a free-space beam, as has been discussed in context with thermal emission from a metamaterial (Chan et al 2006, Laroche et al 2006). In our experiment the external beam is at normal incidence, and therefore, it has no electric field component normal to the slab: it cannot excite the lowest leaky mode (a$_1$), which is asymmetric in respect to the slab plane (Ulrich 1974). Efficient coupling occurs, however, to the next higher leaky mode (s$_2$), which is symmetric, and this coupling is so strong that a rather broad resonance results at $f_{\text{res}} < c/g$; its peak transmittance reaches 100% for the case of a perfect conductor (Ulrich 1974). A detailed experimental test of Ulrich’s analytical model of leaky-wave coupling and of other theoretical developments reviewed in (Garcia de Abajo 2007) could be achieved in future by quantitative vector spectra (figures 2(c) and (d)). For this purpose higher-quality periodic structures are needed since the one used has evident irregularities.

The complex transmission spectrum (figure 2) gives further insight into wave propagation in and along the holes themselves because these are longer than a typical free-space wavelength, and have a large aspect ratio of depth $t$ to diameter, of $t/d \approx 5$. Hollow metal waveguide
Transport can thus be separated from the free-space-to-hole and hole-to-free-space couplings via surface waves at both surfaces (Keilmann 1981). A prominent property of hollow metal waveguides is their cutoff effect. It occurs below the cutoff frequency which for a perfectly conducting metal is $f_{\text{cut}} = 0.586 \frac{c}{d}$. In fact, we observe (figures 2(c) and (d)) that the transmission amplitude reduces sharply below $f_{\text{cut}} = 0.86$ THz derived from $d = 204 \ \mu m$ in our sample. Interestingly, the phase of transmission stays advanced throughout the cutoff region $f \leq f_{\text{cut}}$ at about 9 rad compared to free space. While an advance of the order of 1 rad can be expected from the combined input and output couplings, as known from thin metal mesh where the holes do not contribute a waveguide propagation (see the results of figures 4 and 5, and of Winnewisser et al (2000)), the main effect of about 8 rad advancement can be attributed to superluminal propagation along the holes. Thus we derive the phase velocity in the waveguides to be $v_{\text{ph}} = c \left(1 + \frac{8c}{2\pi f t}\right) = 14c$ in the cutoff frequency region. Theoretically the phase velocity in perfectly conducting waveguides is infinite at and below $f_{\text{cut}}$, but this presents no contradiction; rather, a reduced velocity is to be expected from absorption and roughness scattering at the guide walls (Keilmann 1981) and could well serve to determine these hard-to-access quantities with high sensitivity. Note that the vector THz spectrometer (figure 1) could easily be adapted to measure also complex reflection spectra.

4. Mid-infrared experiment

In a second configuration of frequency-comb Fourier-transform spectrometer (c-FTS) for infrared vector spectrometry, we generate two infrared beams which we superimpose. To understand the basic principle, consider the frequency domain. Both infrared beams have harmonic frequency-comb spectra, regular sequences of modes with frequencies $nf_r$ and $n(f_r + \Delta)$, respectively, caused by the regular pulse repetition and the process of frequency difference generation (Schliesser et al 2005). The offset $\Delta$ is chosen small enough that a low-frequency beating occurs only between the elements of both combs that have identical harmonic number $n$, at beat frequencies $n\Delta$. Then the beat spectrum uniquely replicates the infrared spectrum at the time-dilatation factor $f_r/\Delta$ (Keilmann et al 2004, Schliesser et al 2005, van der Weide and Keilmann 1998) chosen here with $\Delta = 25.02$ Hz to be 500 000, such that a 30 THz mid-infrared oscillation appears down-scaled as a 30 THz/500 000 = 6 MHz oscillation. Here we demonstrate the capability of mid-infrared c-FTS to acquire amplitude and phase transmission spectra. Our setup (figure 3) is a simpler version of our earlier arrangement for demonstrating the acquisition of amplitude and phase spectra of back-scattering from the tip of a scattering-type, ‘apertureless’ near-field microscope (s-SNOM) (Brehm et al 2006).

The Ti:S lasers and the trigger arrangement are identical to the THz experiment of figure 1. The infrared beams are generated by focusing the laser beams onto 200 $\mu m$ thick GaSe crystals oriented between 50° and 65°, for the difference-frequency generation of 9 THz wide spectra in the 20–40 THz range, equivalent to 8–15 $\mu m$ wavelength. In contrast with former setups, (Brehm et al 2006, Keilmann et al 2004, Schliesser et al 2005) the beams are not superimposed on a dielectric beam combiner. Instead, we use wavefront combination by a mirror with a sharp edge (Ganz et al 2008) to avoid a dielectric plate and the associated problems of multiple reflections and dielectric dispersion. This choice should be especially useful for extending the instrument to super-decade-wide infrared spectra (Kübler et al 2005). The infrared beams are refocused at 95 cm, by Au-coated paraboloidal mirrors with 25 mm effective focal length. The resulting 1–2 mm diameter spot allows us to measure small-size

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Figure 3. Optical layout of vector mid-IR spectrometer with no moving part. Two mid-infrared frequency-comb beams are generated by difference frequency generation in GaSe and adjusted for parallel propagation. Interference occurs on an HgCdTe infrared detector and results in self-scanned, repetitive interferograms. With the sample in one of the beams, transmission spectra are recorded in amplitude and phase simultaneously.

samples at well-defined incidence angles, as the beam convergence angle is only about 0.8° full width at half maximum (FWHM). For initial alignment we temporarily place a 2 mm diameter HgCdTe detector just after the combining mirror, to maximize the signal of each beam. The beams are adjusted for parallel propagation at about 2 mm center-to-center distance, by translating and orienting the mirror which has a sharp edge, and they are focused after 105 cm by an $f = 25$ mm Ge lens onto a 200 µm diameter HgCdTe detector (KMPV11, Kolmar). The signal is preamplified at 30 MHz bandwidth and recorded on an oscilloscope (WaveSurfer 422, LeCroy). Transient interferograms appear at a rate of 25.02 Hz and are recorded for 6.1 µs, giving a spectral resolution of $(f_r/\Delta)/6.1 \mu s = 0.8$ THz equivalent to $2.7$ cm$^{-1}$. For averaging over typically 1000 transients in 40 s we stabilize $\Delta$ by sending the cross-correlator trigger signal to a lock-in amplifier (Stanford Research 510) which is externally referenced at 25.02 Hz, and by applying the X output at 100 ms time constant to the piezoelectric transducer. This simple arrangement allows a hold range of $\pm 0.7$ V equivalent to $\pm 1$ Hz, sufficient to stabilize against thermal drifts over several minutes. To understand the infrared comb-FT spectrometer’s vector capability, especially for measuring phase spectra, consider the field amplitude

$$E(t) = \sum_{n=M}^{N} E_n \cos (2\pi n f_r t + \phi_n),$$

of one beam, and

$$E'(t) = \sum_{n=M}^{N} E'_n \cos (2\pi n (f_r + \Delta) t + \phi'_n)$$

of the other, where a choice of $M \approx 160 000$ and $N \approx 320 000$ would span the range from $M f_r = 20$ THz to $N f_r = 40$ THz, equivalent to $667$ cm$^{-1}$ to $1334$ cm$^{-1}$. The detector signal $U(t) \propto (E(t) + E'(t))^2$ contains, apart from two dc terms, several series of interference terms. Choosing $\Delta < f_r/2N \approx 195$ Hz assures that only one series,

$$\sum_{n=M}^{N} E_n E'_n \cos (2\pi n \Delta t + \phi'_n - \phi_n),$$

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lies in the frequency range \(<f_r/2\) and can be selected using a low-pass filter at \(<f_r/2 \approx 63\text{ MHz}.\) This signal’s radio-wave spectrum is the direct replica of the mid-IR spectrum. In particular, its phase spectrum is identical to the infrared phase spectrum.

5. Mid-infrared results

Metamaterials of periodically perforated metal sheets are chosen to demonstrate the capabilities of c-FTS vector spectroscopy in the mid infrared. Near unity transmittance is observed with the metamaterial structure in figure 4, due to the \(s_2\) leaky-wave resonance mentioned above, (Ulrich 1974) here at \(f_{\text{res}} \approx 975\text{ cm}^{-1} = 0.78\c/g\) which is again considerably lower than the onset of diffraction at \(f_{\text{Wood}} = c/g = 1250\text{ cm}^{-1}\). Note that at resonance the transmission is sixfold enhanced over the area fraction occupied by the holes of only about 17%. This means that the average infrared intensity in the holes is more than sixfold enhanced over the incident (and likewise the transmitted) far-field intensity. The phase advances continuously from about zero as the resonance is traversed from the high-frequency side. Since the holes have a rather small depth \(t/\lambda \approx 0.2,\) we can expect that waveguiding effects in the holes are of minor importance to the transmission; both the amplitude and the phase spectra are dominated by the coupling effects of free-space waves into and out of the hole array. Here, we note that a special kind of a transmittance resonance has been theoretically described (Garcia-Vidal et al 2005) and measured (Lee et al 2006, 2007, Ruan and Qiu 2006) for a single hole shaped and oriented similarly as in figure 4; it occurs at \(f_s \approx c/2L,\) where \(L\) is the width of the hole; thus for a single hole with \(L \approx 5\mu m\) as in our sample one would expect \(f_s \approx 1000\text{ cm}^{-1}.\)

In figure 5, we show results of measuring a more open metamaterial with square symmetry. The spectrometer’s range was set to cover both the resonant-transmission and Wood anomaly regions. At normal incidence, the onset of diffraction leaves a marked signature at \(f_{\text{Wood}} = c/g = 1250\text{ cm}^{-1}\) (dashed vertical line). It is well known that the Wood anomaly is expressed...
Figure 5. Complex transmission spectra (a) of a $t = 1 \, \mu \text{m}$ thick Au foil with square holes of 3.5 mm width arranged at period $g = 8 \, \mu \text{m}$, taken at varied incidence angles $\alpha$ (p-polarization); diffraction at the sample sets in at $c/g = 1250 \, \text{cm}^{-1}$; (b) complex polar plot of amplitude and phase data from (a) for three angles, with the marked points separated by 0.5 THz (amplitude in linear scale, from 0 to 1).

in a stepwise decrease of the (zeroth order) transmittance when power is carried away into four first-order diffracted beams emanating at grazing angles. What our spectrometer reveals is that this onset of diffraction also induces a characteristic dip structure in the transmission phase spectrum. At non-normal incidence, both these amplitude and phase signatures tune pairwise according to the expected relation $f_{\text{Wood}} = (1 \pm \sin \alpha)^{-1} c/g$.

The transmission resonance due to the $s_2$ leaky-wave interaction (figure 5(a)) reaches near-unity amplitude at $f_{\text{res}} \approx 1050 \, \text{cm}^{-1} = 0.84 \, c/g$. In addition, but only at non-normal incidence, the transmission spectrum shows the onset of a marked amplitude dip at about $1040 \, \text{cm}^{-1}$ and a marked phase signature. Already at $4^\circ$ the dip reaches down to 50% transmission amplitude, equivalent to a transmitted power of only 25%. This sharp resonance with a $Q$-value of $f/\Delta f$(FWHM) $\approx 50$ originates from the excitation of an $a_1$ leaky wave which has its field oriented normally (Ulrich 1974) hence its excitation must vanish at normal incidence. Since this $a_1$ resonance (high-$Q$) occurs within the broad band of the $s_2$ resonance (low-$Q$), the resulting transmission line shape expresses the interference of two interactions (‘direct and indirect pathways’ in (Fan and Joannopoulos 2002)), and therefore it is in this respect analogous to Fano’s analysis (Fano 1961) of a sharp transition interfering with a continuum (Ulrich 1974). Our measurement is capable of supplying the complete information on this coupling resonance including the phase signature. For an illustration we plot three of the spectra of figure 5(a) in a polar diagram where the transmission amplitude and phase are the coordinates (figure 5(b)). The spectra in the coupling resonance region describe approximately circular trajectories which are traversed clockwise with rising frequency. Relative to such $a_1$ coupling features, the Wood anomaly appears as a smaller effect that describes an arc also traversed clockwise.

In the coupling dip, the missing power is converted via the $a_1$ surface wave (whose wavelength $\lambda_{\text{SPP}}$ matches the condition $1/\lambda_{\text{SPP}} = 1/g - (\sin \alpha) / \lambda$) both into metal absorption...
Figure 6. Dispersion of the $a_1$ leaky wave (black dots) determined from the positions of the coupling dip minima in figure 5(a); open triangles mark the Wood anomaly; the corresponding angle of incidence can be read from the inserted scale. Also shown is the light line (full), and its back-folded continuation (dashed) beyond the Brillouin zone edge. Also shown is the dispersion (red squares) of the leaky wave determined for the sample of figure 7.

The $a_1$ leaky surface wave on the sample in figure 5 asymptotically approaches, at $0^\circ$, a frequency of about $1040 \text{ cm}^{-1}$ and a phase velocity of $1040/1250 = 0.83 c$; the coupling to external radiation vanishes. Caution is advised in interpreting the observed $Q \approx 50$ to be solely governed by nonradiative damping of the $a_1$ evanescent wave part, by absorption in the metal (Ulrich 1974). Rather, the resonance can be inhomogeneously broadened by residual wrinkle of the stretched foil; an additional mechanism is contributed by the mere width of our probing beam: when we assume 1 mm width, the transit time of the surface wave is $T = 1 \text{ mm}/0.83 c$ which induces a homogeneous broadening of $1/T = 8.3 \text{ cm}^{-1}$.

The sample in figure 7 has the same periodicity and exhibits a similar light transmission resonance due to the $s_2$ leaky-wave interaction, at $f_{res} \approx 1000 \text{ cm}^{-1} = 0.8 c/g$. Yet in contrast, the asymptotic frequency and phase velocity of its $a_1$ mode are both considerably reduced to about $930 \text{ cm}^{-1}$ and $930/1250 = 0.76 c$, respectively. It takes higher incidence angles $\alpha$ to observe a red-shifting of the dip which merges with the Wood feature above about $15^\circ$. In the polar plot in figure 7(b) the Wood and $a_1$ coupling features can be distinguished in the $10^\circ$ and $14^\circ$ trajectories, the latter as rather weak loops, but no longer at $22^\circ$ where they form a strong combination signature. The overall transmission is retarded in this sample by about 1 rad, and
Figure 7. Complex transmission spectra (a) of a $t = 8\mu m$ thick Au foil with 4 $\mu m$ wide and 500 $\mu m$ long slits arranged at period $g = 8\mu m$, taken at varied incidence angles $\alpha$ (p-polarization); (b) complex polar plot of amplitude and phase data from (a) for four angles, with the marked points separated by 0.5 THz (amplitude in linear scale, from 0 to 1).

even more at oblique incidence. This might be caused by a Fabry–Perot resonance in the deep slits, but not by a reduced phase velocity which should equal $c$ for a TEM mode in a parallel-plate waveguide (Jackson 1975, Mendis and Grischkowsky 2001).

6. Outlook for metamaterials

The metamaterials investigated here do not contain deeply subwavelength structure elements that possess localized electric and magnetic resonances as needed to induce a negative refractive index. Yet we expect that it will be with such samples that vector c-FTS can serve with its quantitative measurement capabilities, given the ease demonstrated here of taking amplitude and phase spectra at well-controlled directions of both beam propagation and electric field. To retrieve effective-medium dielectric and magnetic functions from vectorial spectra, appropriate Fresnel-type analytic expressions have been given (Chen et al 2004, Smith et al 2002). A sensitive quantity is the slab thickness applicable to the case of single-layer metamaterials such as holey metal films. In practice, this thickness has been assumed equal to the in-plane unit-cell period (Driscoll et al 2007, Padilla et al 2007). Two-layer structures have allowed a better-defined determination, in two cases with an out-of-plane period which was about an order of magnitude smaller than the in-plane period (Dolling et al 2006, Zhang et al 2005). Experimentally retrieved optical constants of 1–4 layer metamaterials have recently been reported (Azad et al 2008). Quite generally the probing wavelength should greatly exceed the period of the metamaterial elements (this requirement, however, might have to

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be relaxed to obtain a strong magnetic response (Rockstuhl et al. 2007)). Otherwise optical probing does not deal with the effective dielectric and magnetic responses alone. Rather, a rich set of surface and bulk excitations, such as the \(a_1\) leaky-wave and Wood interactions in the above examples, contributes characteristic signatures. These depend on incidence angle and beam width, and need to be understood and corrected for to avoid artefacts in the electric permittivity and magnetic permeability functions. Note these quantities might even be intrinsically non-local because inter-element coupling causes them to depend on momentum, not just frequency (Garcia de Abajo and Saenz 2005, Koschny et al. 2005).

In conclusion, we have demonstrated amplitude and phase spectra by a vector comb-based spectrometer. It has enabled us to present complex-valued transmission spectra of metal hole arrays and identify phenomena such as leaky-wave interaction and cutoff-waveguide propagation. The method can characterize the optical properties of metamaterials more completely than previously possible, and may help to resolve issues such as the origin of negative refraction or the non-locality of dielectric and magnetic responses.

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