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Magnetic dipole and electric dipole resonances in TiO_2 microspheres at terahertz frequencies

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ABSTRACT

In a non-magnetic dielectric sphere of high-permittivity (ε >20), effective magnetic response occurs as a result of the 1st Mie mode, known as the magnetic dipole resonance. This resonance produces a similar effect as split ring resonators, making it possible to use dielectric spheres as metamaterial components. In the terahertz (THz) part of the spectrum, where dielectrics with ε ~100 can be found, all-dielectric metamaterials can potentially reduce absorption and provide isotropic and polarization-independent properties. In this contribution, we discuss TiO_2 micro-spheres, ~1/10 of the wavelength in diameter. Such spheres are expected to support the magnetic and electric dipole resonances. To detect these resonances in a single TiO_2 microsphere we use THz near-field microscopy with the sub-wavelength size aperture probe. This method allows detection of Mie resonances in single sub-wavelength spheres. Fano-type line-shape is observed in the near-field amplitude and phase spectra. The narrow line-width of the magnetic resonance and the sub-wavelength size of the TiO_2 microspheres make them excellent candidates for realizing low-loss THz metamaterials.

Keywords: Terahertz, metamaterials, dielectric resonator, near-field imaging, Mie mode, TiO_2 micro-sphere, Fano line-shape, THz spectroscopy

1. INTRODUCTION

The key element for metamaterials is a sub-wavelength size resonator with an effective magnetic response. High-permittivity dielectric resonators [1] have attracted much interest recently as an alternative to split ring resonators, which exhibit undesirable anisotropy, polarization dependent response and non-negligible metallic Ohmic losses. In a high-permittivity dielectric sphere, an effective magnetic response occurs as a result of the 1st Mie mode, known as the magnetic dipole (MD) resonance [1].

TiO_2 has a large potential as a material for THz sub-wavelength size resonators. Microspheres of TiO_2 can be fabricated by assembling and annealing clusters of TiO_2 nano-crystals. Their size, density and crystallinity can be controlled through varying the fabrication process conditions. Poly-crystalline TiO_2 microspheres have the average permittivity of ~100, large enough for a microsphere of only 1/10 the wavelength (20-30 μm) in diameter to have the 1st Mie resonance in the region of 1-2 THz [2].

In order to determine the intrinsic line-widths of the MD and ED modes and to avoid inhomogeneous broadening, the microspheres should be probed individually. Characterization of the Mie resonances in a sub-wavelength size object however is difficult with the standard THz spectroscopy methods due to the reduction of the absolute scattering cross-section at the MD resonance. As the permittivity increases, the effective scattering cross-section of the sphere increases approximately as $\varepsilon^{1/2}$. Its physical cross-section however decreases approximately as $1/\varepsilon$. We estimate that a single TiO_2 sphere of diameter $d \approx \lambda/n = \lambda/10$ (for $\varepsilon = n^2 = 100$) placed in the focus of a typical THz beam produces only small attenuation (~1%) of the beam intensity at the frequency of MD resonance, i.e. 0.5% attenuation for the field amplitude. This small value of attenuation is comparable to the noise level in a typical THz time domain spectroscopy system. As a result, the far-field THz characterization of individual TiO_2 microspheres is highly challenging.

In the vicinity of a high-permittivity resonator, on the other hand, the electromagnetic energy is present as an evanescent field. Near-field microscopy methods therefore may provide an alternative method for detecting the resonator modes. Here we investigate near-field probing of TiO_2 microspheres and demonstrate that the MD as well as the electric dipole (ED) resonances can be characterized using the THz near-field microscopy approach with a sub-wavelength (~λ/50) size aperture probe [3]. The Mie resonance signatures appear in the amplitude and phase spectra. This technique allows us to determine the intrinsic width of MD and ED modes as well as to map the electric field distribution near the resonator [4]. The observed narrow line-width of the MD resonance confirms the potential of the TiO_2 microspheres for realizing low-loss THz metamaterials.

2. TRANSMISSION THROUGH A SUB-WAVELENGTH APERTURE

To probe the properties of TiO_2 microspheres, we utilize the effect of enhanced transmission through a sub-wavelength aperture when a high-permittivity micro-sphere is placed in front of the aperture [4,5]. The effect can be understood as the localization of the resonant field near the sphere due to the reduced wavelength inside the high-permittivity material and due to energy storing inside the resonator. The confined field couples through the sub-wavelength aperture more efficiently than the incident THz beam, resulting in enhanced transmission at the frequencies of Mie resonances. Figure 1 illustrates this effect in the waveforms of the THz pulse transmitted through a 5 μm aperture, with and without a 27 μm diameter TiO_2 sphere (see Fig. 1, inset) placed in front of the aperture.

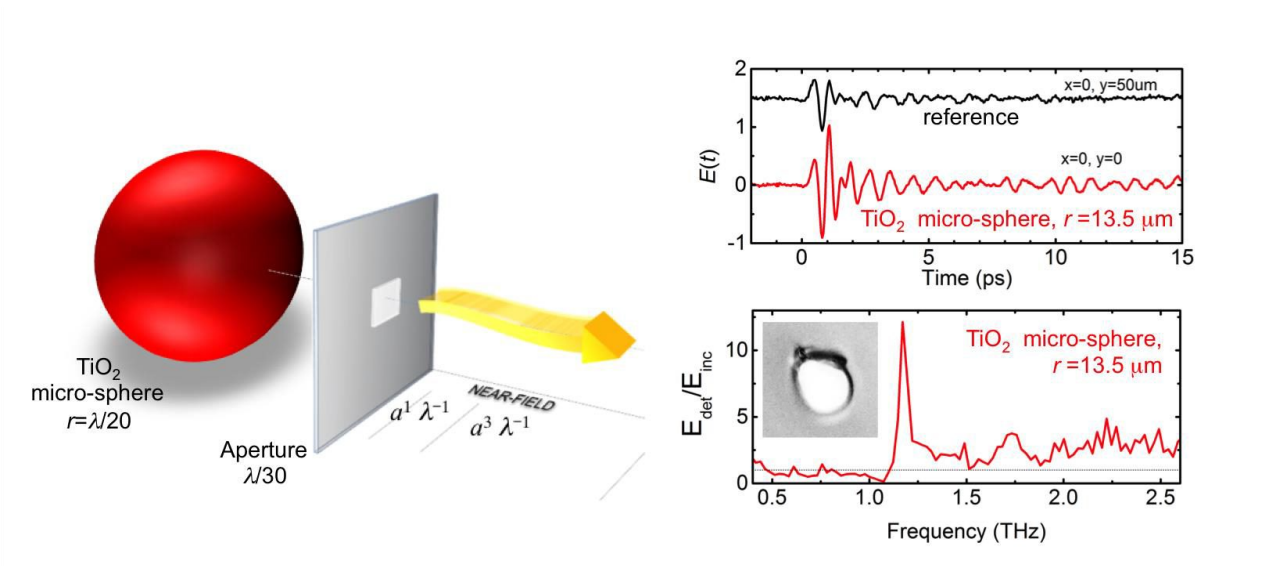

FIG. 1: Schematic diagram of the experimental setup: a 13.5 μm radius TiO_2 sphere is positioned at a distance z=1 μm from a 5 μm aperture in a gold screen. The THz pulse waveform measured after transmission through the aperture with the sphere placed in front of the aperture (red trace) is compared with the waveform measured without the sphere (black trace). The amplitude transmission spectrum calculated using Fourier transmforms of the waveforms; *inset*: an optical microscope image (60 μm × 60 μm) of the sample.

The waveforms show that the amplitude of the pulse transmitted through the aperture is higher for the case of the sphere. The corresponding waveform [Fig. 1, red trace] also shows coherent regular oscillations lasting over several cycles. These oscillations are absent in the waveform of the incident pulse [Fig. 1, black trace]. Fourier transforms of the waveforms reveal that the enhanced transmission through the aperture occurs only at a selected frequency, which corresponds to the frequency of the observed oscillations. The oscillations indicate that a resonance is formed inside the sphere.

The spectrum in Fig. 1 suggests that the electric field amplitude in the near-field region of the sphere is enhanced at the resonance frequency by up to 10 times. For non-resonant frequencies, the detected field amplitude however remains practically unaffected. On the low frequency side of the peak, the amplitude drops to zero showing the distinctive line shape of a Fano-type resonance.

After testing a number of spheres ranging from 20 to 30 μm in diameter and using the 5 μm as well as 10 μm aperture probes [3] we find that the relative transmission spectra show similar line shapes and similar values of enhancements at the resonance frequency. The frequency of the resonance itself varies with the size of the sphere [4].

The clear signature of the observed resonance shows the advantage of near-field probing over far-field characterization of sub-wavelength size high-permittivity resonators. The signal-to-noise ratio in the measured spectra exceeds the signal-to-noise estimated for the far-field transmission characterization, whereas The Fano line shape suggests that an analytical model can be applied for extracting the resonator parameters.

3. MAGNETIC AND ELECTRIC DIPOLE RESONACES

The spectrum in Fig. 1 also shows a smaller and broader peak at ~1.7 THz. The set of two resonances matches the lowest order Mie modes, the MD and ED modes, expected for a spherical particle 25 μm diameter with $\varepsilon \approx 100$. The finite-difference time-domain method (FDTD) is used to simulate the field distributions for these modes. Figure 2 shows the E_x component of the field in the section through the sphere. The first resonance occurs at 1.213 THz. The electric field lines of this mode form closed loops within the sphere. Such a field distribution corresponds to an induced magnetic moment M_y within the sphere and therefore it is called the magnetic dipole mode. The second resonance is at 1.66 THz. The field distribution of this mode is similar to that of the electric dipole.

The set of resonances observed in the experiment confirms that the enhanced coupling effect is due to the excitation of normal modes in the microsphere (TiO_2 has no material-related modes in this spectral range). The resonance frequencies decrease as the resonator size increases, whereas their ratio $\omega_{MD}/\omega_{ED} \approx 0.7$ remain practically the same. It varies slightly from sample to sample due to the non-ideal shape of the spheres.

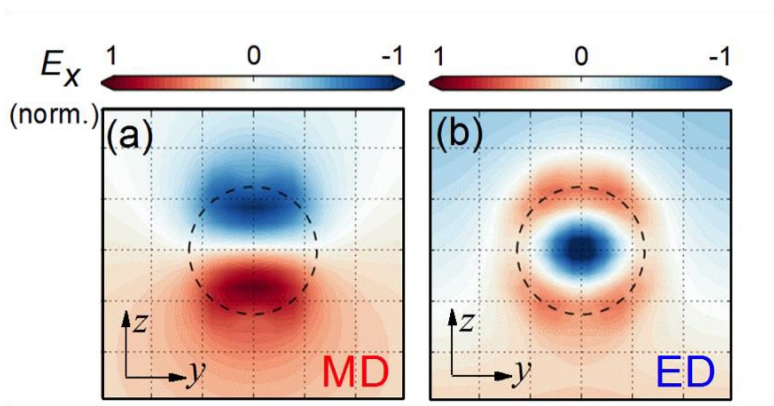

FIG. 2: Electric field distribution in the central section of a TiO_2 sphere ($\varepsilon = 100$, radius $r = 12.5$ μm) for the two lowest order modes. The magnetic dipole (MD) mode is at 1.213 THz (a) and the electric dipole mode (ED) is at 1.66 THz. The modes are excited by an x-polarized incident EM wave propagates along the z-axis. Color shading represents the electric field (E_x).

Both modes are observed in the near-field measurements for the spheres excited by an unfocused THz beam. The ED resonance exhibits a significantly weaker spectral feature in our measurements. The signature strength in general depends on the coupling of the incident wave to the resonance mode and on the coupling to the near-field probe. As shown by the simulations in Fig. 2, the E_x component of the field at the location of the aperture exhibits similar spatial distributions for both modes. Therefore we don't expect substantially different sensitivities of the near-field probe to the ED and MD modes. We attribute the difference therefore to the difference in the coupling of the incident wave to the two modes.

4. FANO LINE SHAPE

The MD resonance signature in the near-field spectrum normalized to the spectrum of the incident field follows the Fano line shape. Figure 3 displays the experimental spectrum for a TiO_2 sphere of r = 11 μm (red), the numerically simulated spectrum, and a Fano resonance signature fitted to the data. The analytical description uses only three parameters: the resonance frequency, the line-width, and the Fano parameter q. The agreement of the experimental results and the model is nevertheless close.

The three parameters in the Fano formula have the following physical interpretation: the Fano parameter q corresponds to the aperture transmission enhancement defined by the amplitude of the resonance peak in the spectrum; the Fano line-width parameter and the resonance frequency determine the width and position of the spectral peak.

If the enhancement factor becomes zero, the Fano shape changes to a Lorentzian shape with the Lorentzian line-width parameter. Therefore the MD resonance line-width can be estimated directly from the line-shape model fit to the data. The full width at half maximum is found to be $\Gamma/(2\pi)$=20±10 GHz for the sample in Fig. 1 and 50 GHz for the sample in Fig. 3. We note that the fitted line-width is similar to the measurement resolution line-width of 40 GHz, which we estimate form the duration of the recorded waveform (25 ps). Therefore we can only conclude that the observed line-width for the sample in Fig. 1 is less than 40 GHz. Details of this analysis can be found in Ref. 4.

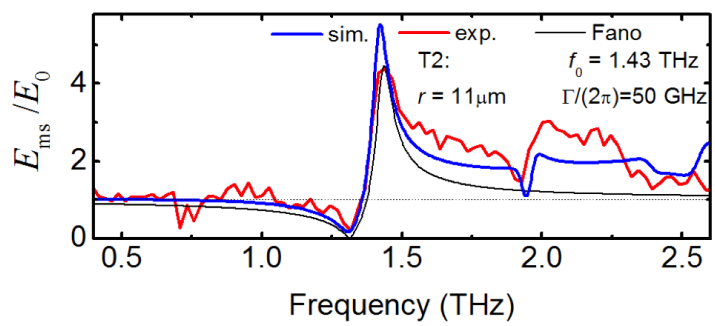

FIG. 3: The amplitude spectrum for the closest distance z<1 μm measured by a 5 μm aperture probe (red line), the fitted Fano resonance line shape (black line) and the simulated spectrum of a TiO_2 sphere.

The spectral signatures in Fig. 1 and 3 correspond to the spheres positioned directly in front of the aperture and the MD moment oriented parallel to the plane of the aperture. If the MD moment is oriented perpendicular to the aperture plane, the phase of the MD resonance depends on the location of aperture with respect to the sphere and the corresponding spectral signature can be reversed, with the zero amplitude point on the high frequency side of the resonance. This effect is presented Fig. 4. Panels (a) and (b) show two instantaneous THz images of another TiO_2 particle, one immediately after the THz pulse excitation and the other ~5 ps after the excitation.

The later image shows a noticeably different pattern, which is anti-symmetric with respect to the y-axis. THz spectra measured at the locations of the field minimum and maximum (marked by the red and blue arrows in the image) show resonances in the frequency region of the MD mode (~1.4 THz), however the two line-shapes are reversed. The change of the line shape correlates with the phase spectra shown in Fig. 4(d). The images and the spectra therefore

indicate the excitation of a MD mode with the magnetic moment oriented along perpendicular to the image plane. Detailed analysis of these observations can be found in Ref. 4.

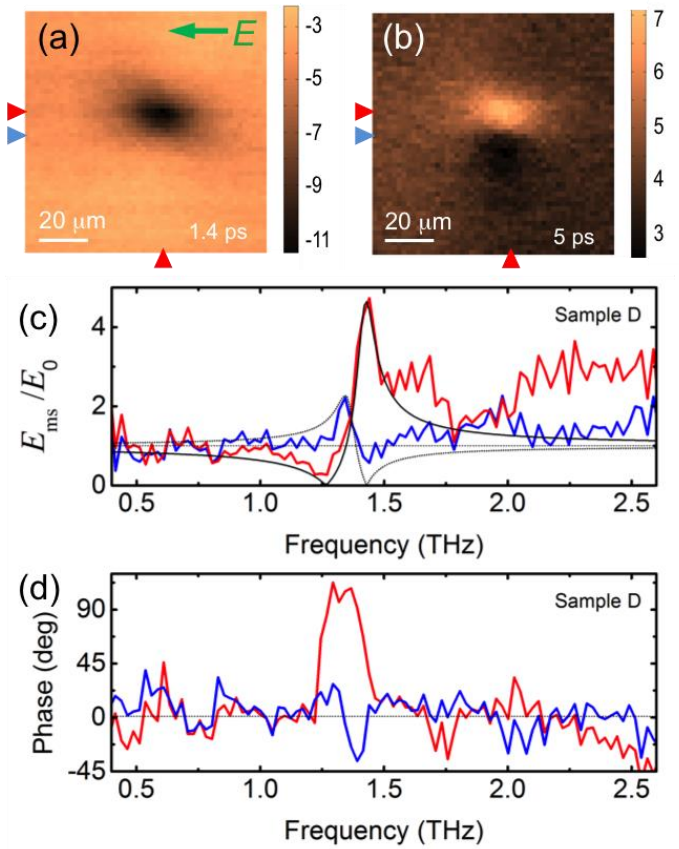

FIG. 4: Electric field maps $E_x(x,y)$ at z=~1 μm from the TiO_2 sphere immediately after the excitation of the sphere by the THz pulse, t=1.4 ps, and at t=5 ps (image area: 100 μm × 100 μm). The spectral amplitude ratio (c) and phase (d) measured at the sphere center (x_0, y_0) and at (x_0, y_0-10 μm); these location are indicated by the red and blue arrows in (a) and (b).

5. CONCLUSIONS

Poly-crystalline TiO_2 microspheres 20-30 μm in diameter exhibit magnetic dipole and electric dipole resonances in the THz frequency range. The magnetic dipole resonance line-width for a single resonator is less than 40 GHz, substantially narrower than the inhomogeneously broadened line-width observed for an ensemble of spheres [2]. The narrow line-width and the sub-wavelength size ($2r \approx \lambda/10$) make the TiO_2 microspheres excellent candidates for realizing the effective magnetic response for low-loss THz metamaterials. In comparison with SRR, the TiO_2 resonators can provide a narrower line-width at THz frequencies and eliminate the anisotropy and polarization dependence of the effective magnetic permeability μ.

The near-field probe with a sub-wavelength aperture allows probing Mie resonances in individual TiO_2 resonators. The MD spectral signature follows the Fano line shape in the amplitude and phase spectra. The analytical description of the spectrum allows estimating the line-width of the resonance despite the asymmetry of the line shape.

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