

Microspectroscopy on perovskite-based superlenses

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Abstract: Superlenses create sub-diffraction-limit images by reconstructing the evanescent fields arising from an object. We study the lateral, vertical, and spectral field distribution of three different perovskite-based superlenses by means of scattering-type near-field microscopy. Sub-diffraction-limit resolution is observed for all samples with an image contrast depending on losses such as scattering and absorption. For the three lenses superlensing is observed at slightly different frequencies resulting in an overall broad frequency range of 3.6 THz around 20 THz.

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References and links

1. V. Veselago, "The electrodynamics of substances with simultaneously negative values of ϵ and μ ," *Sov. Phys. Usp.* **10**, 509–514 (1968).
2. D. R. Smith, W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, "Composite medium with simultaneously negative permeability and permittivity," *Phys. Rev. Lett.* **84**, 4184–4187 (2000).
3. H. J. Lezec, J. A. Dionne, and H. A. Atwater, "Negative refraction at visible frequencies," *Science* **316**, 430–432 (2007).
4. J. Yao, Z. Liu, Y. Liu, Y. Wang, C. Sun, G. Bartal, A. M. Stacy, and X. Zhang, "Optical negative refraction in bulk metamaterials of nanowires," *Science* **321**, 930 (2008).
5. U. Leonhardt and T. Philbin, "General relativity in electrical engineering," *New J. Phys.* **8**, 247–1–18 (2006).
6. U. Leonhardt, "Optical conformal mapping," *Science* **312**, 1777–1780 (2006).
7. J. B. Pendry, D. Schurig, and D. R. Smith, "Controlling electromagnetic fields," *Science* **312**, 1780–1782 (2006).
8. D. Schurig, J. Mock, B. Justice, S. Cummer, J. Pendry, A. Starr, and D. Smith, "Metamaterial electromagnetic cloak at microwave frequencies," *Science* **314**, 977–980 (2006).
9. J. Valentine, J. Li, T. Zentgraf, G. Bartal, and X. Zhang, "An optical cloak made of dielectrics," *Nature Mater.* **8**, 568–571 (2009).
10. J. B. Pendry, "Negative refraction makes a perfect lens," *Phys. Rev. Lett.* **85**, 3966–3969 (2000).

11. N. Fang, H. Lee, C. Sun, and X. Zhang, "Sub-diffraction-limited optical imaging with a silver superlens," *Science* **22**, 534–537 (2005).
12. T. Taubner, D. Korobkin, Y. Urzhumov, G. Shvets, and R. Hillenbrand, "Near-field microscopy through a SiC superlens," *Science* **313**, 1595 (2006).
13. X. Zhang and Z. Liu, "Superlenses to overcome the diffraction limit," *Nature mater.* **7**, 435–441 (2008).
14. M. C. K. Wiltshire, J. Pendy, I. Young, D. Larkman, D. Gilderdale, and J. Hajnal, "Microstructured magnetic materials for RF flux guides in magnetic resonance imaging," *Science* **291**, 849–851 (2001).
15. T. J. Yen, W. Padilla, N. Fang, D. Vier, D. Smith, J. Pendry, D. Basov, and X. Zhang, "Terahertz magnetic response from artificial materials," *Science* **303**, 1494–1496 (2004).
16. C. Enkrich, M. Wegener, S. Linden, S. Burger, L. Zschiedrich, F. Schmidt, J. Zhou, T. Koschny, and C. Soukoulis, "Magnetic metamaterials at telecommunication and visible frequencies," *Phys. Rev. Lett.* **95**, 203901 (2005).
17. N. Setter, D. Damjanovic, L. Eng, G. Fox, S. Gevorgian, S. Hong, A. Kingon, H. Kohlstedt, N. Y. Park, G. B. Stephenson, I. Stolitchnov, A. K. Taganste, D. V. Taylor, T. Yamada, and S. Streiffer, "Ferroelectric thin films: Review of materials properties, and applications," *J. Appl. Phys.* **100**, 051606 (2006).
18. S. Jin, T. Tiefel, M. McCormack, R. Fastnacht, R. Ramesh, and L. Chen, "Thousandfold change in resistivity in magnetoresistive La-Ca-Mn-O films," *Science* **264**, 413–415 (1994).
19. M. K. Wu, J. Ashburn, C. Torng, P. Hor, R. Meng, L. Gao, Z. Huang, Y. Wang, and C. Chu, "Superconductivity at 93 K in a new mixed-phase Y-Ba-Cu-O compound system at ambient pressure," *Phys. Rev. Lett.* **58**, 908–910 (1987).
20. J. Scott, *Ferroelectric Memories* (Springer Series in Advanced Microelectronics, Vol. 3, Heidelberg, New York, 2000).
21. R. Waser, *Nanoelectronics and Information Technology* (Wiley-CH, Weinheim, 2003).
22. S. C. Kehr, Y. Liu, L. Martin, P. Yu, M. Gajek, S.-Y. Yang, C.-H. Yang, M. Wenzel, R. Jacob, H.-G. von Ribbeck, M. Helm, X. Zhang, L. Eng, and R. Ramesh, "Near-field examination of perovskite-based superlenses and superlens-enhanced probe-object coupling," *Nat. Commun.* **2**, 249, DOI:10.1038/ncomms1249 (2011).
23. R. Shelby, D. Smith, and S. Schultz, "Experimental verification of a negative index of refraction," *Science* **292**, 77–79 (2001).
24. W. Spitzer, R. C. Miller, D. Kleinman, and L. Howarth, "Far infrared dielectric dispersion in BaTiO₃, SrTiO₃, and TiO₂," *Phys. Rev.* **126**, 1710–1721 (1962).
25. S. Kamba, D. Nuzhnyy, M. Savinov, J. Šebek, J. Petzelt, J. Prokleška, R. Haumont, and J. Kreisel, "Infrared and terahertz studies of polar phonons and magnetodielectric effect in multiferroic BiFeO₃ ceramics," *Phys. Rev. B* **75**, 024403 (2007).
26. T. D. Kang, G. S. Lee, H. S. Lee, H. Lee, Y. S. Kang, S.-J. Cho, B. Xiao, H. Morkoç, and P. G. Snyder, "Infrared ellipsometric study on PZT thin films," *J. Korean Phys. Soc.* **49**, 1604–1610 (2006).
27. F. Zenhausern, M. O'Boyle, and H. Wickramasinghe, "Apertureless near-field optical microscope," *Appl. Phys. Lett.* **65**, 1623–1625 (1994).
28. F. Zenhausern, Y. Martin, and H. Wickramasinghe, "Scanning interferometric apertureless microscopy: Optical imaging at 10 Angstrom resolution," *Science* **269**, 1083–1085 (1995).
29. E. Betzig, J. K. Trautman, T. D. Harris, J. S. Weiner, and R. L. Kostelak, "Breaking the diffraction barrier: Optical microscopy on a nanometric scale," *Science* **251**, 1468–1470 (1991).
30. S. C. Schneider, J. Seidel, S. Grafström, L. Eng, S. Winnerl, D. Stehr, and M. Helm, "Impact of optical in-plane anisotropy on near-field phonon polariton spectroscopy," *Appl. Phys. Lett.* **90**, 143101 (2007).
31. S. C. Kehr, M. Cebula, O. Mieth, T. Härtling, J. Seidel, S. Grafström, L. Eng, S. Winnerl, D. Stehr, and M. Helm, "Anisotropy contrast in phonon-enhanced apertureless near-field microscopy using a free-electron laser," *Phys. Rev. Lett.* **100**, 256403 (2008).
32. G. Wurtz, R. Bachelot, and P. Royer, "Imaging a GaAlAs laser diode in operation using apertureless scanning near-field optical microscopy," *Eur. Phys. J. Appl. Phys.* **5**, 269–275 (1999).
33. B. Knoll and F. Keilmann, "Enhanced dielectric contrast in scattering-type scanning near-field optical microscopy," *Opt. Commun.* **182**, 321–328 (2000).
34. R. Hillenbrand and F. Keilmann, "Complex Optical Constants on a Subwavelength Scale," *Phys. Rev. Lett.* **85**, 3029–3032 (2000).
35. R. Hillenbrand, T. Taubner, and F. Keilmann, "Phonon-enhanced light-matter interaction at the nanometre scale," *Nature* **418**, 159–162 (2002).
36. T. Taubner, F. Keilmann, and R. Hillenbrand, "Nanomechanical Resonance Tuning and Phase Effects in Optical Near-Field Interaction," *Nano Lett.* **4**, 1669–1672 (2004).
37. P. Mühlischlegel, H.-J. Eisler, O. J. F. Martin, B. Hecht, and D. W. Pohl, "Resonant optical antennas," *Science* **308**, 1607–1608 (2005).
38. I. Fedorov, J. Petzelt, V. Zelezny, G. A. Komandin, A. A. Volkov, K. Brooks, Y. Huang, and N. Setter, "Far-infrared dielectric response of PbTiO₃ and PbZr_{1-x}Ti_xO₃ thin ferroelectric films," *J. Phys.: Condens. Matter* **7**, 4313–4323 (1995).

1. Introduction

Metamaterials show many fascinating optical properties [1] such as negative refraction [2–4], optical cloaking [5–9] and perfect imaging with planar superlenses [10–13]. In the last decade, such materials have been designed for different frequencies ranging from Gigahertz [14] via Terahertz (THz) [15] up to optical frequencies [3, 16]. On the other hand, perovskites exhibit many intriguing properties such as piezoelectricity and ferroelectricity [17], colossal magnetoresistance [18], and superconductivity [19], that lead to numerous applications [17, 20, 21]. In the mid-infrared range, pairs of certain perovskites show permittivities that are suitable for a superlens for electric fields [22]. In this paper, we discuss the performance of three different perovskite-based superlenses. These lenses together show a resonant response with a bandwidth of 3.7 THz at around 20 THz, which we study by means of near-field infrared microscopy in combination with a free-electron laser.

A planar slab of a material with negative refractive index acts as a superlens [10]: propagating and evanescent waves arising from an object on one side of the lens are focused in the image plane on the opposite side, resulting in a resolution beyond the classical diffraction limit of $\lambda/2$. In order to achieve negative refractive index, a material needs to show negative permittivity and permeability at the same frequency [1]. Such properties are not observed in any natural material, but they can be created in periodical arrays of artificial structures such as split-ring resonators in combination with metallic wires [2, 23]. However, a planar slab of a material with negative permittivity only (see Fig. 1a), which can naturally be found in many materials at certain frequencies, does not focus propagating waves, but it still reconstructs the evanescent electric fields that carry the intriguing sub-wavelength information about the object [10–13].

Pairs of different perovskites are in particular suitable as superlenses for several reasons [22]: firstly, perovskite oxides show phonon-resonances in the range of 1 to 25 THz, which result in negative permittivities on the high-frequency sides of some of these resonances [24–26]. As the crystal structures of different perovskites are similar (Fig. 1b), they show phonon modes at similar frequencies (Fig. 1c). Consequently, at certain frequencies we find matching pairs of materials A and B with small dielectric constants of opposite signs, which fulfill the superlensing condition of $\epsilon_A = -\epsilon_B$ (Fig. 1d). Secondly, at the frequencies of interest the corresponding imaginary parts are small resulting in small absorption of the materials. Moreover, the similarity in crystal structure allows for the growth of epitaxial heterostructures resulting in low scattering at highly crystalline interfaces. Together, small absorption and low scattering lead to low losses, which is in general a limiting factor of metamaterial structures.

In this paper, we compare three different superlenses made out of different pairs of perovskite oxides such as strontium titanate (STO), bismuth ferrite (BFO), and lead zirconate titanate (PZT) concerning resolution, image contrast, and frequency range.

2. Experimental methods

We study vertically layered lenses, which consist of (1) structured objects made out of the metallic perovskite strontium ruthenate (SRO) on STO substrates, (2) a spacer layer A of BFO or STO with $\epsilon_A \cong +1$ and (3) a superlens layer B of STO or PZT with $\epsilon_B \cong -1$ at the frequencies of interest (see Fig. 1a). All films are grown by means of pulsed laser deposition (PLD) resulting in highly crystalline films with atomically flat interfaces (see [22]). We study three different lenses with the two layers A-B being BFO-STO, PZT-STO, and BFO-PZT, which fulfill the superlensing condition $\epsilon_A = -\epsilon_B$ at different frequencies in the range from 19 to 23 THz (Fig. 1d).

In order to excite and study the evanescent waves in the image plane of the lenses, we combine a scattering-type near-field infrared microscope (s-NSIM) [27–29] with the free-electron laser (FEL) light source at Forschungszentrum Dresden Rossendorf, Germany, which is pre-

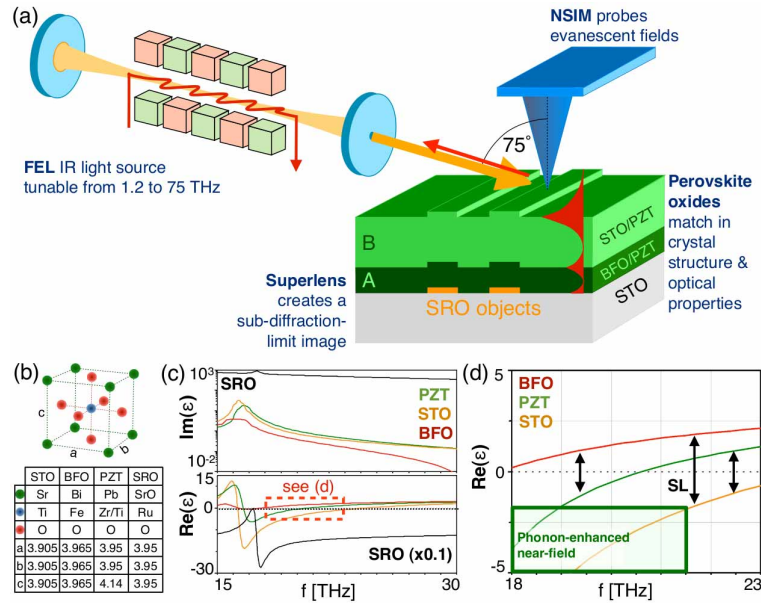


Fig. 1. s-NSIM setup and perovskite properties. (a) Sketch of the experimental setup including the superlens structure, the geometry at the near-field probe, and the free-electron laser light source [22]. The superlenses consist of two layers A and B of thicknesses d and $2d$ ($d=200$ nm), respectively, with A-B being BFO-STO, PZT-STO, or BFO-PZT. As for objects, we study structured SRO on a STO substrate. (b) Perovskite structure of the materials used with lattice constants in Å determined by X-ray diffraction; (c) imaginary and real parts of the dielectric constants ϵ of all constituents taken either from literature (for BFO, STO, and PZT [24–26]) or determined by Fourier transform infrared (FTIR) spectroscopy (for SRO, see [22]); (d) real parts of the dielectric constants at the high-frequency side of their phonon resonances depicted in c. The arrows indicate the frequencies at which superlensing is expected for superlens systems with A-B being BFO-PZT, BFO-STO, and PZT-STO (from small to large frequencies), whereas the green box highlights the area of a phonon-enhanced near-field signal in the top-most layers STO and PZT. (Figure adapted from [22])

cisely tunable from 1.2 to 75 THz (Fig. 1a) [30, 31]. This combination allows us to probe the evanescent waves in the image plane of the lens and to study their spectral behavior [22]. Moreover, as the basis of the microscope is an atomic force microscope, we are able to probe the vertical and lateral distribution of the optical signal as well of the topography of the sample.

In general, with NSIM one measures localized electric fields such as evanescent waves by placing a near-field probe close to a sample surface [27–29]. We use the method of higher-harmonic demodulation [32–34] in order to separate the near-field (NF) from the much larger background signal: the probe-sample distance is modulated resulting in an enhanced sensitivity to strongly varying fields such as evanescent waves. These fields can be enhanced resonantly by nonlocalized polariton modes in the sample or due to the superlensing effect.

For the perovskite-based lenses we observe polariton-enhanced near-field signals on the high-frequency side of the phonon-resonances, where $\Re(\epsilon) = -5$ to -1 (see box in Fig. 1d) [31, 35]. The specific spectral position of these resonances depends on the probe-sample distance h with decreasing resonance frequency for smaller distances [33, 36] and show maxima of different phases in different higher-harmonic demodulation frequencies. As the origin of this resonantly

enhanced near-field is a nonlocalized polariton mode, it is present on all areas of the samples. Particularly, it does not carry any information about the SRO objects on the opposite side of the lens. However, on areas with SRO objects, we observe an additional contribution to the near-field signal due to the superlensing effect, which is located at slightly higher frequencies than the propagating polariton mode, namely, when $\epsilon_B = -\epsilon_A$ (see arrows in Fig. 1d). Here, on both interfaces of layer B coupled polariton modes are excited which result in the localized evanescent fields that form the superlens image [10–12, 22].

3. Near-field imaging of perovskite-based superlenses

In the following we compare the near-field signals of three different perovskite superlenses as well as of SRO objects without a superlens (Fig. 2). For all superlenses we observe a clear object-related contrast with sub-diffraction-limit resolution when $\epsilon_A = -\epsilon_B$. In addition, we show the topography images on the examined areas (Fig. 2, column 1) and the distance dependence of the near-field signals (Fig. 2, column 2). Here, we focus on the comparison of different perovskite superlenses. For a more detailed discussion in particular of the near-field examination of superlenses please see [22].

For metallic SRO no polariton-enhanced signal is expected as $\text{Re}(\epsilon_{\text{SRO}}) \cong -150 \ll -5$ in the measured frequency range. The third-harmonic near-field signal $\text{NF}_{3\Omega}$ is the same over a wide frequencies range and decreases to zero within a distance of 300 nm reflecting its evanescent character (Fig. 2a, second column). The images shown are measured with a CO₂ laser with a frequency of 28.3 THz. However, we expect a similar response at shorter frequencies in general, except for some geometrical resonances due to antenna effects as known from plasmonics [22, 37]. We compare the second and third harmonic signals, $\text{NF}_{2\Omega}$ and $\text{NF}_{3\Omega}$, on a given structure. In both harmonics, the near-field signal is enhanced on the SRO objects. The signal on 50 nm thick objects is rather weak and, hence, the signal-to-noise ratio is low. Consequently, in $\text{NF}_{3\Omega}$ the structure is hardly visible.

Compared to the plain SRO objects the topography of all superlenses is rough (see Fig. 2b–d, first column) with grains, which depend on the condition of growth in PLD. In particular the PLD-growth condition for the superlens consisting of PZT and STO are not ideal: the growth temperature for STO is so high, that the PZT layer would be damaged. Consequently, the STO layer needs to be grown at a lower temperature resulting in a rough surface as can be seen in Fig. 2c. Please note that these grains are only present on the surface of layer B, but not at the A-B interface, as we know from in-situ reflection high-energy electron diffraction (RHEED). In general, these problems can be overcome in using other growth techniques such as chemical vapour deposition or sol-gel growth, which are both in addition suitable for growth of thick films and large samples but result in a lower crystalline quality of the films. However for the BFO-PZT and BFO-STO superlenses, the temperature problem does not occur and the surface appears comparably smooth.

Both effects described above, polariton- and superlens-enhanced signals, result in a resonant response of the superlenses. The latter appears on the SRO objects only and at somewhat larger frequencies. We compare the distance dependence of the third-harmonic signals $\text{NF}_{3\Omega}$ on both areas shown in the second column of Fig. 2. Even though we probe the objects at a distance of 600 nm, $\text{NF}_{3\Omega}$ on all superlens structures is much higher than the corresponding signal on the SRO objects only. For BFO-STO we observe the strongest enhancement of about 10 times the SRO response. Please note that this number can only be a qualitative figure as the samples were measured with different probes. At frequencies much shorter than the superlensing frequency, $\text{NF}_{3\Omega}$ is similar on areas with and without SRO object. At larger frequencies the signals show a different distance dependence, which is present on the SRO objects only.

Even though the propagating polariton enhances the near-field signal it is not localized and

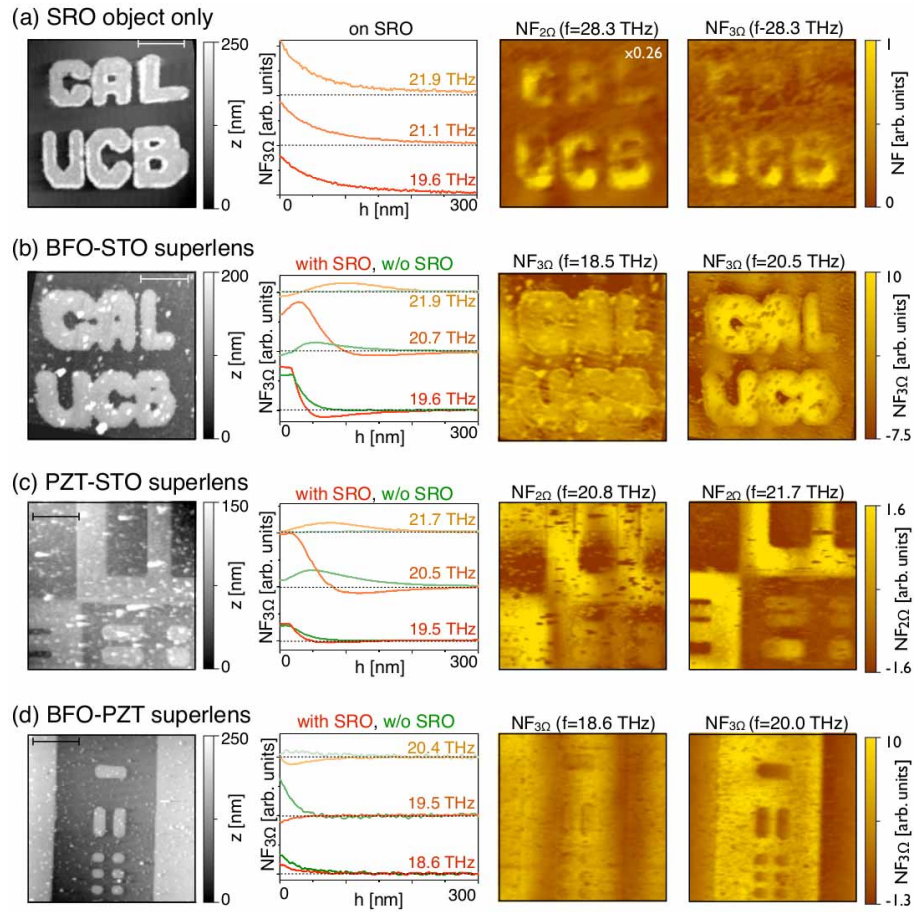


Fig. 2. Near-field images of different superlenses. For different sample types, we show (from left to right): topography images (scalebar is $10\ \mu\text{m}$), third-harmonic near-field signal $NF_{3\Omega}$ as a function of the distance h (vertical offset added for better visibility, dashed lines show $NF_{3\Omega} = 0$), and near-field images showing $NF_{2\Omega}$ or $NF_{3\Omega}$ at two selected frequencies (same color range) at which either the phonon-enhanced signal (left) or the superlens-enhanced signal (right) dominates. (a) SRO objects only, distance dependence measured with FEL, near-field images with CO_2 laser. (b)-(d) Different types of superlenses. On areas with and without SRO objects on the opposite site of the lens (see Fig. 1a), we observe different distance dependences: at low frequencies, the phonon-enhanced signal is present on all areas of the sample, whereas at high frequencies localized fields are present only on SRO objects due to the superlensing effect. At these frequencies, we observe a clear contrast with sub-wavelength resolution (images on the far-right). Please note that for the BFO-PZT superlens (d) the phase of the superlens contribution is opposite to the phase of the phonon-enhanced signal resulting in an inverted contrast.

will not create an image correlated to the SRO objects. Hence, we expect an enhanced contrast with sub-diffraction-limit resolution only when superlensing occurs. The near-field images on the right hand side of Fig. 2 show the signal when either exciting the propagating polariton mode (left) or the localized polariton modes resulting in superlensing (right). For the first case, the structures appear blurred with a slightly larger signal on areas with objects due to scattering. However, at larger frequencies, a clear contrast can be observed, which reproduces the shape of the objects.

In order to determine the resolution of the superlenses, we compare the smallest structures resolved in the near-field image with their lateral size in the topography. These are at least as small as $0.75\ \mu\text{m}$ for BFO-STO and $1\ \mu\text{m}$ for PZT-STO and BFO-PZT corresponding to a resolution of $\lambda/20$, $\lambda/14$ and $\lambda/15$, respectively. Please note that the resolution of the superlens might be much higher, but scattering at the topography edges makes it impossible to define a resolution by means of the signal change at a steep edge, which is commonly used in scanning probe microscopy.

4. Spectral response of perovskite-based superlenses

4.1. Numerical simulations: transfer functions

The performance of a superlens can be described by its transfer function, that is the transmittance $|T|^2$ through the lens as a function of the wavenumber k . For superlensing, the evanescent waves are enhanced over a large range of k vectors, that allow for the formation of a sub-diffraction-limit image. Here, the isothermal contour of the transmittance show an extended tail towards large k .

In Figure 3, we compare the transfer functions for the three different perovskite-based superlenses. We find the highest k -vector ranges at 21.7 THz, 22.5 THz, and 20.1 THz for the BFO-STO, PZT-STO, and BFO-PZT superlenses, respectively. These findings match well the prediction by the simple SL-condition of $\epsilon_A = -\epsilon_B$ as indicated by the arrows in Fig. 1d. Moreover, we find that the BFO-STO superlens shows the largest bandwidth in terms of the largest frequency range with supported high k .

4.2. Near-field spectroscopy on perovskite-based superlenses

We experimentally compare the spectral response of the superlenses based on different perovskites for different distances h between probe and sample surface (Fig. 4). For each lens type, third-harmonic signals $\text{NF}_{3\Omega}$ on areas with (red) and without (green) SRO object are shown as well as the dielectric constants of its constituents for comparison. For all lenses, we observe a modified signal on the SRO objects at frequencies where the dielectric constants of both constituents have different signs. This response characteristically shifts towards larger frequencies with increasing h , which is discussed in detail in [22] and which is related to the frequency shift of polariton-enhanced near-field coupling as discussed in [36]. Due to different values of $\text{Re}(\epsilon_A)$ and $\text{Re}(\epsilon_B)$ the different samples show superlensing at slightly different frequency ranges reaching from 19.2 to 22.2 THz for BFO-STO (Fig. 4a), 19.5 to 22.2 THz for PZT-STO (Fig. 4b), and 18.6 to 21 THz for BFO-PZT (Fig. 4c).

As described above, we observe two contributions to the near-field on the perovskite-based superlenses, namely polariton- and superlens-enhanced signals. As both signals are resonant at slightly different frequencies and with different phases, we observe constructive and destructive superposition of both (see [22] for details). For BFO-STO and PZT-STO, a decreased signal is observed e.g. at $h = 50\ \text{nm}$ for $f < 20.3\ \text{THz}$ and $f < 20.5\ \text{THz}$, respectively, whereas for larger frequencies, the near-field is increased (see Figs. 4a,b). For the BFO-PZT superlens (Fig. 4c) both resonances are spectrally located closer to each other compared to the other superlenses,

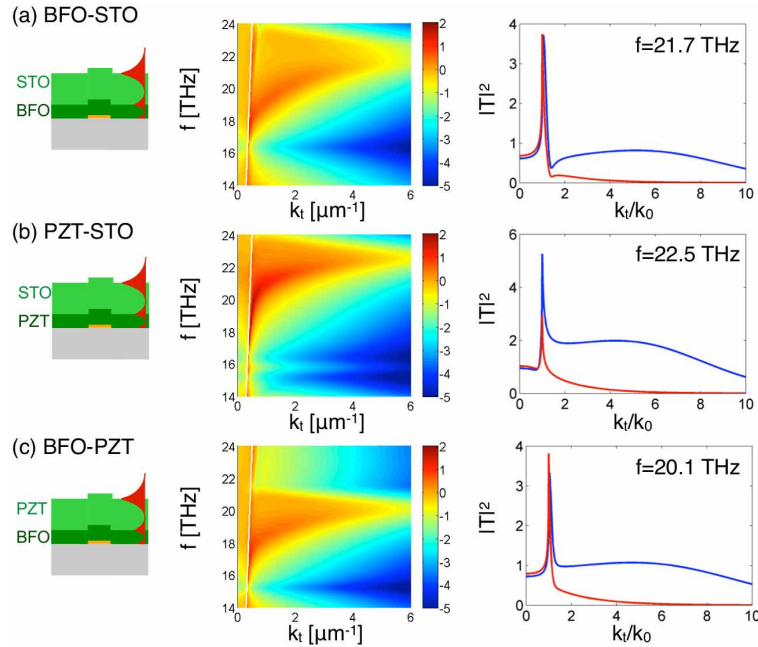


Fig. 3. Transfer functions for the three different perovskite-based superlenses, namely (a) BFO-STO, (b) PZT-STO, and (c) BFO-PZT. The transmittance $|T|^2$ is shown as a function of frequency f and wavenumber k using materials properties from literature [24, 25, 38]. For each superlens, we show (from left to right): 1. sketch of the superlens, 2. isothermal contour of the transfer function (the white line is the light line), and 3. the transmittance as a function of the tangential wave vector up to $10k_0$ for the corresponding superlensing frequencies (peaks at $k_t = k_0$ correspond to total internal reflection). For the latter, we compare the response of the superlens (blue) with the response of a reference sample (red) for which layer B consists of material A.

resulting in a reduced signal on the SRO objects for most distances and frequencies. Only for $h = 0$ nm, we observe a small increase at frequencies between 20.4 and 21.4 THz.

It is absorption and interface roughness that determine the strength of the near-field signals and the image contrast of the lenses. Highest absorption is observed for the BFO-PZT superlens with $\Im m(\epsilon_{PZT})$ reaching 2, whereas the lenses with an active layer of STO show a highest absorption of 0.7 in the range of interest. On the other hand for the PZT-STO superlens the interface roughness is higher due to the growth conditions as discussed above, which results in higher loss due to interface scattering. The comparison of the signal strength on the different types of superlenses is shown for $h = 0$ nm in Fig. 5. The BFO-STO superlens shows the highest signal as well as the broadest bandwidth ($f = 19.2$ to 21.45 THz). The signal strength for the PZT-STO and the BFO-PZT superlenses are decreased by a factor of about 2.5 and 5 with their frequency range located at $f = 20.1$ to 21.75 THz and $f = 18.6$ to 20.4 THz, respectively. Please note that, compared to absorption losses, the high roughness and the corresponding scattering losses of the PZT-STO superlens have a rather small effect on the signal strength and image contrast. Hence, it seems to be material absorption rather than surface quality that is the key parameter for designing an efficient superlens.

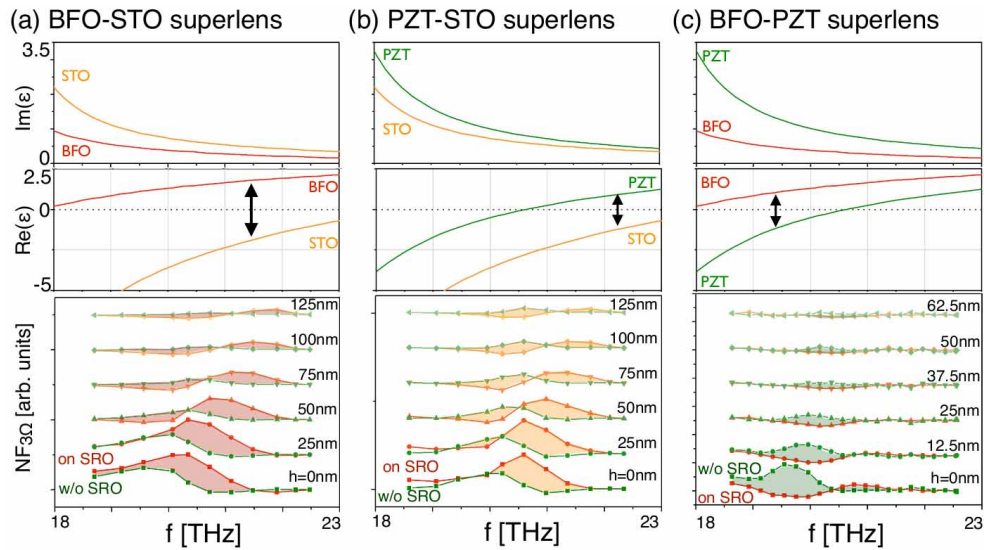


Fig. 4. Near-field spectroscopy on three different samples. We compare the spectral near-field response on areas with (red) and without (green) SRO objects for three different superlenses. The highlighted areas mark the additional fields on the SRO objects due to the superlensing effect, which is present when the real parts of the permittivities of the layers A and B have opposite signs (theoretical position marked with arrows in the $\text{Re}(\epsilon)$ diagrams). For different types of superlenses, this response is located at slightly different frequencies with different bandwidth. It characteristically shifts to larger frequencies with increasing distance h between probe and sample surface [22]. (a) BFO-STO superlens, (b) PZT-STO superlens, and (c) BFO-PZT superlens.

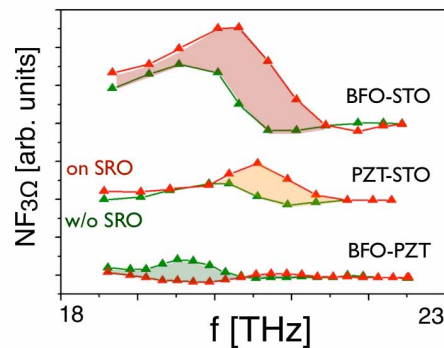


Fig. 5. Comparison of the near-field spectra on all lenses for $h = 0$ nm with the same $\text{NF}_{3\Omega}$ -scale. The BFO-STO superlens shows the highest signal, whereas it is decreased for PZT-STO and BFO-PZT due to interface roughness and higher material absorption, respectively.

5. Conclusion

We examined the near-field of three different perovskite-based superlenses. At the superlensing frequencies, we observed an image of structures on the opposite side of the lens with a resolution beyond the classical diffraction-limit. Different pairs of perovskites show slightly different spectral positions and bandwidths of the superlensing frequencies.

With the three different lenses studied in this paper, a frequency range from 18.6 to 22.2 THz is covered corresponding to an overall bandwidth of about 3.6 THz. Other pairs of perovskite oxides or similar materials might lead to an extension of this range. A combination of different perovskite superlenses might be usable as combined bandpass filters for near-field signals, with possible application for spectroscopical examination of e.g. biological samples as objects.

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