

A Simple Doublet Lens Design for mid-IR Imaging

CLAIRE E. NELMARK AND ARNALDO L. SERRANO*

Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, IN 46556, USA

**arnaldo.serrano@nd.edu*

Abstract: Wide-field mid-IR hyperspectral imaging offers a promising approach for studying heterogeneous chemical systems due to its ability to independently characterize the molecular properties of different regions of a sample. However, applications of wide-field mid-IR microscopy are limited to spatial resolutions no better than $\sim 1\ \mu\text{m}$. While methods exist to overcome the classical diffraction limit of $\sim \lambda/2$, chromatic aberration from transmissive imaging reduces the achievable resolution. Here we describe the design and implementation of a simple mid-IR achromatic lens combination that we believe will aid in the development of resolution enhanced wide-field mid-IR hyperspectral optical and chemical absorption imaging. We also examine the use of this doublet lens to image through polystyrene microspheres, an emerging and simple means for enhancing spatial resolution.

1. Introduction

Infrared (IR) spectroscopy, and its ultrafast nonlinear variants, is a powerful technique useful for studying a wide range of chemical systems, materials [1–3], and phase separating systems [4,5] over a variety of timescales, [6] and has potential for biological diagnostics. [7,8] It provides extensive molecular information with a minimal amount of sample, and can be implemented in a nondestructive manner with the potential to be done in a label-free fashion. By coupling chemically sensitive information with microscopic imaging, mid-IR hyperspectral imaging collects molecular information with spatial resolution, providing an image with chemical contrast. [9,10] This is especially important in spatially heterogeneous samples, where a sample can have a diverse chemical or molecular make up and it can be impossible to see how this varies using other conventional imaging modalities. [11] However, a major drawback of IR spectroscopy when studying micro-heterogeneous samples is its relatively low spatial resolution. [12]

While mid-IR imaging has been developed and used by numerous labs over decades, [13–25] due to the relatively long wavelengths of mid-IR light ($\sim 3\text{--}8\ \mu\text{m}$ or $\sim 1250\text{--}3300\ \text{cm}^{-1}$) used in ultrafast mid-IR imaging, the classical diffraction limit makes it impossible to resolve smaller features such as organelles in a cell, [26] or microscopic liquid droplets. [27–29] Additionally, when a broadband light source is used for spectroscopy, chromatic optical aberrations may further limit the achievable spatial resolution. [30] Since chromatic aberration leads to reduced spatial resolution, it needs to be corrected in order to achieve diffraction-limited resolutions. Chromatic aberration is an optical aberration specific to systems utilizing a polychromatic light source, such as an ultrashort pulse of light in nonlinear optics, that arises from the wavelength dependence of the refractive index of transmissive optics. This results in the focal length of the lens being wavelength dependent, making it impossible to bring all the light from a source into focus simultaneously. [31]

Chromatic aberration needs to be addressed in even the most sophisticated transmissive optical imaging systems, regardless of the wavelength range. However, it is particularly problematic for coherent imaging in the mid-IR wavelength regime, where

commercial solutions to this problem are not widely available. With so many potentially transformative applications for mid-IR imaging, particularly in the biomedical field, there is significant interest in the development of mid-IR optical systems that can image with sub-diffraction limited resolution. [32,33] However, imaging techniques requiring a broadband light source, which is necessary for many spectroscopic applications, will suffer from reduced spatial resolution until the chromatic aberration is mitigated. While techniques to reduce chromatic aberration have been developed for visible light and include a variety of lens designs such as superchromats [34–36], hybrid lenses [37,38], metasurfaces [39,40], adaptive optics [41,42], as well as post-processing algorithms [43–46], many of these methods are expensive, time consuming, or under-developed for the mid-IR.

ATR-FTIR is a common technique touted for its ease of use and is technically capable of sub-diffraction limited resolution, however, this is only true for the sample nearest the ATR crystal. This technique is best for probing surface properties of samples, and can typically reach no farther than 5 μm into the sample, thus limiting the range of samples to be studied with it. [47,48] Reflective objectives, such as the Schwarzschild objective, are another common commercially available solution because they are intrinsically achromatic. However, these objectives have been shown previously to limit resolution capabilities for coherent imaging applications, especially near the resolution limit, due to the central obscuration of the entrance pupil caused by their secondary mirror. [13,49,50] Similarly, parabolic mirrors are inherently achromatic, and have been shown previously to also have the benefit of avoiding the central obscuration that is innate to the reflective objectives when used off-axis. [51] However, they introduce more aberration to a system when using it off-axis. Because of this, they are not favored in laser-scanning applications. [36] With an end-goal of sub-diffraction limited ultrafast imaging in mind, which requires a coherent light source, we chose to focus on the possible refractive lens options.

Besides the reflective options mentioned above, refractive solutions have been demonstrated as well. Kazarian and coworkers utilized FTIR to exhibit the ability of semi-hemispherical lenses placed on the sample to diminish chromatic aberrations. [25,52,53] Hanninen and Potma produced a promising broadband commercially constructed achromatic doublet lens that spans the visible range up to $\sim 4.5 \mu\text{m}$. [36] While these are encouraging options for mid-IR techniques, they can be cost- and time-prohibitive as they are custom optics, and may require a custom optical mount. [54] Here we take a similar approach and present a simple doublet lens optimized for the 4-6 μm ($\sim 1665\text{--}2500 \text{ cm}^{-1}$) wavelength region for future use in ultrafast mid-IR imaging, assembled in-lab with off-the-shelf components. This wavelength range was chosen with future ultrafast hyperspectral imaging applications studies in mind, which primarily consist of collecting information within the amide-I and nitrile region of the IR spectrum. While our design only partially corrects for the chromatic aberration present in our system, it is achievable at a fraction of the cost and time required of a fully custom solution.

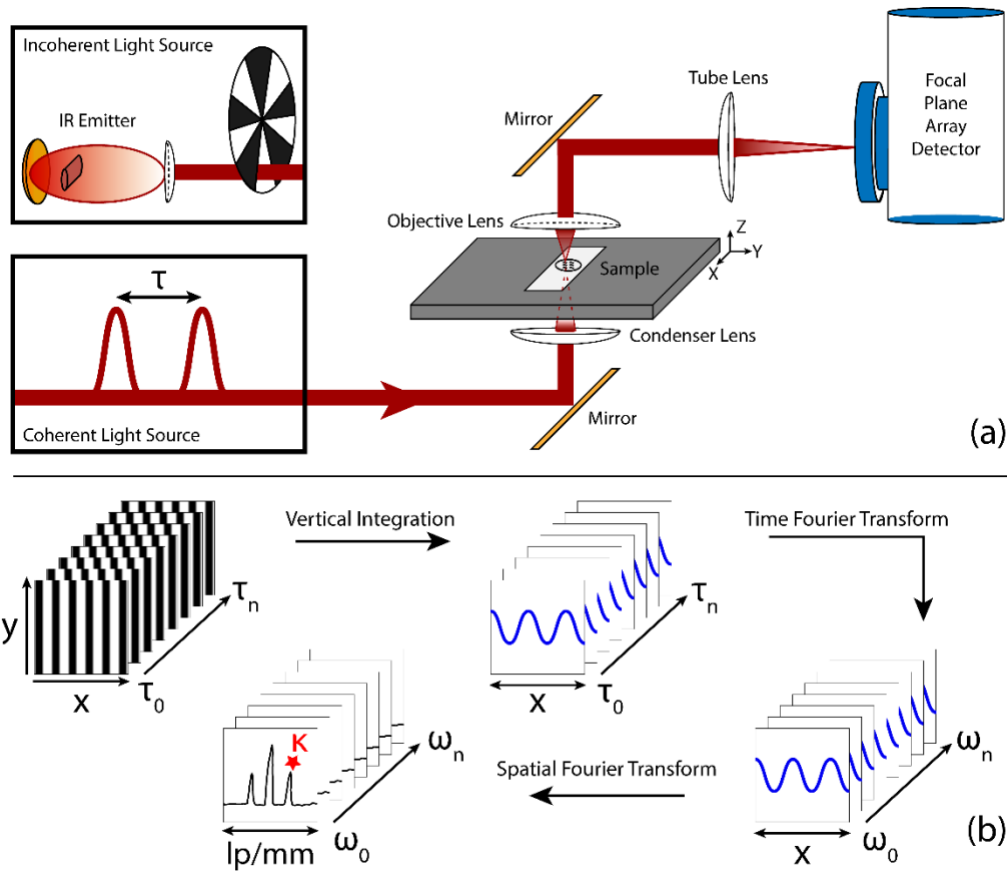
Additionally, we show early work combining this doublet lens with another relatively simple technique for resolution enhancement that has been demonstrated previously. Imaging through high-index dielectric microspheres placed near to or in direct contact with the object of interest in transmission microscopy has been shown to achieve sub-diffraction resolution with visible light, [55–58], resolution enhancement when done with mid-IR light, [59] as well as effectively increase the observed magnification of the image in both cases. It is commonly believed that the reason for this resolution enhancement is related to the ability of the microspheres to focus light into photonic nanojets that have intense and narrow, sub-diffraction beam waists owing to near field effects [60,61]. Thus, coupling both the dielectric microsphere with an achromatic doublet lens has the potential to result in even greater resolving power. In

94 this manuscript, we have tested this approach by quantifying the resolution of the doublet lens,
95 as well as the combination of the doublet and microsphere, by measuring their modulation
96 transfer functions (MTFs) using a set of uniform resolution targets. Comparing amplitudes of
97 the MTFs for different imaging systems offers an approach for identifying any resulting
98 enhancement.

100 2. Methods

101 An air-spaced achromatic doublet consisting of a variable CaF_2 lens and an uncoated, 1”
102 diameter, 12.7 mm focal length ZnSe aspheric lens (Edmund Optics) was designed in
103 OpticStudio by ZEMAX. The optimal lens geometry was found with the built-in contrast
104 optimization function that works by maximizing the intensity of the MTF at a given spatial
105 frequency – the spatial frequency used for the optimization was 30 cycles per mm. The
106 combination was first optimized allowing the thickness of the lens, the air spacing between the
107 two lenses, the air spacing from the second lens to the image plane, and the radius of curvature
108 of both sides of the CaF_2 lens, to vary.

109 From here, the optimized CaF_2 lens was compared to off-the-shelf models from
110 various manufacturers, and a meniscus lens from Thorlabs, Inc. very nearly matched the
111 optimized parameters. The specifications for this meniscus lens were swapped out for the
112 optimized CaF_2 lens in OpticStudio and the doublet was reoptimized allowing the space
113 between lens and the space after the lens to the image plane to both vary. The CaF_2 meniscus
114 lens was placed in a slotted lens tube with the ZnSe aspheric lens at approximately the
115 optimized distance. Fine adjustments were then made with the retaining rings while optimizing
116 the apparent contrast in the live digital microscope images. A diagram of the microscope is
117 shown in **Fig. 1(a)**.



118

119 Fig. 1. (a) Diagram of the microscope setup. For coherent experiments, the light source in the
 120 black outline is a double pulse sequence generated by our AOM-based pulse shaper using
 121 broadband pulsed laser light from our femtosecond (fs) mid-IR laser system. For incoherent
 122 experiments, the setup in the top black outline was used. The objective lens was also switched
 123 between the asphere and the doublet lens, depending on the experiment. (b) Depiction of the
 124 FTIR image processing scheme. Images without NAP spectra were processed in a similar
 125 manner, but without the time Fourier transformation step.

126 A homebuilt wide-field mid-IR microscope was configured using the newly designed
 127 doublet lens as the objective lens, and a similar ZnSe aspheric lens (Edmund Optics) with a
 128 numerical aperture (NA) of 0.71 as the condenser lens. A CaF_2 lens with a 50 mm focal length
 129 was used as the tube lens to allow for $\sim 50\times$ magnification of the images. Images were collected
 130 on a 128×128 -pixel mercury cadmium telluride (MCT) focal plane array detector (Catalina,
 131 Teledyne) at the imaging plane.

132 A custom chrome patterned CaF_2 resolution target (II-IV Optical Systems) with
 133 Ronchi grating patterns from 50 to 500 line pairs per millimeter (lp/mm), in increments of 50,
 134 was used for imaging with both incoherent and coherent light sources. The broadband coherent
 135 light source, centered at $6 \mu\text{m}$ with a full-width at half-maximum band width of $\sim 150 \text{ cm}^{-1}$,
 136 originated from an AOM-based ultrafast mid-IR laser system described previously. [59] A 9W
 137 IR light emitter (Newport) placed in front of a 75 mm focal length gold spherical mirror and
 138 collimated with a 150 mm focal length CaF_2 lens was used as an incoherent light source. To

image through microspheres, 100 μm polystyrene (PS) microspheres (Spherotech) with a mid-IR refractive index ~ 1.5 [62] suspended in spectroscopic grade methanol were placed dropwise onto the set of gratings until each grating had at least one lone microsphere for imaging. Images were collected at each grating frequency, with and without a PS microsphere, using both coherent and incoherent light sources. Blank images were also collected through unobscured regions of the CaF_2 target at the same focal settings as each collected grating image, for later background subtraction.

After images were processed (i.e., background subtracted and dead pixels were replaced with the averaged value of nearest neighbors), horizontal cuts of the intensity were taken from the center of each image and vertically averaged over a number of rows, with the averaged cut being Fourier transformed to generate a spatial frequency spectrum. With a lp/mm axis the amplitude of the peak nearest the expected spatial frequency was collected. This was typically the second largest peak in the spatial frequency spectrum, shown in **Fig. 1(b)** and denoted as κ , since the center peak, corresponding to 0 lp/mm, had the largest amplitude. The intensity of the chosen peak was plotted against the actual spatial frequency of the grating to generate the experimental MTFs. The MTFs for the gratings with PS microspheres were each normalized to a 0 lp/mm point. The 0 lp/mm point was taken by collecting an image through a portion of solid chrome on the grating slide, a cut was taken through this in the same manner as the other cuts, and subtracted from the cut for 50 lp/mm. This new subtracted cut was divided by two to account for half of every line pair being covered with chrome, and Fourier transformed to collect the spatial frequency in the same way as the others.

Error bars were determined by choosing wing regions of the Fourier transform that did not include the expected peak, or any intense peaks from higher harmonics of the given spatial frequency. The root-mean-square (RMS) of the wing region was then calculated to give the error in MTF amplitude for each data point. Error was also calculated to look at the noise in the data collection process. For this, 100 shots were collected and averaged into a file. Ten of these files were collected, and individually processed following the same procedure described above. The intensity of the peaks was collected for each of these ten files, and the standard error of the mean was calculated for this data set. As this error was much smaller than that calculated from the wing region in our processing (on the order of $\sim 1.5\%$), we decided this error was negligible and thus presented the larger error from our processing.

The final set of data collected contained FTIR images taken of N-acetyl proline (NAP) on the gratings to determine the spatial resolution of chemical absorption imaging. For this, 200 mM D_2O -exchanged NAP solution was placed dropwise over the gratings. This was covered with a 50 μm greased Teflon spacer and a 0.35 mm CaF_2 window. Images were collected in a purged environment at the focus of the gratings through the NAP solution and averaged for ~ 10 minutes. As the blank, a sample of D_2O was prepared in the same way on a blank CaF_2 window and a spectrum was collected at the same focus as the NAP sample was collected. Data processing was done following the depiction in **Fig. 1(b)**. During image collection, our femtosecond IR pulses were shaped into double pulse pairs with varying phase and time delays, τ . A series of images, shown first, were collected and processed with a two-frame phase cycling scheme described in [13] to produce time-domain interferograms for each pixel in the images. The data was then vertically integrated, and time Fourier transformed to generate single beam spectra for the stack of images, as shown by the next two arrows and stacks of images. The blank sample described above was processed in the same way to generate blank spectra that were used in calculating the absorbance spectra for the image stack. Finally, as illustrated by the last arrow and stack of images in **Fig. 1(b)**, the data underwent spatial Fourier transformation to extract the spatial modulation at each grating frequency. Within these averaged FTIR spectra, the 1700 cm^{-1} peak characteristic of NAP [63], shown in **Fig. 7(a)**, was

baseline-corrected for each set of gratings using a linear fit, and the peak intensity was plotted against the corresponding spatial frequency of the grating. The error bars were generated in a similar fashion to the optical MTF described above.

3. Results

3.1 Designing the Lens

A doublet was chosen for its simplicity and relatively small size. For a doublet to correct for chromatic aberration between two different wavelengths, there must be a dispersion difference between these materials. Thus, the materials for a doublet are typically selected based on their location in an Abbe diagram, [64] which plots the Abbe number V vs. the refractive index n of a set of materials over a specified wavelength range. Since the Abbe number is a measure of the dispersion of a material, this number should be significantly different for the two materials. The Abbe number can be calculated with Eq. 1, where n_{center} is refractive index of the material at the center wavelength of the chosen range, n_{short} is at the shortest wavelength of the range, and n_{long} is at the longest wavelength in the range. [65] An Abbe diagram consisting of common mid-IR optical materials was calculated with refractive index data from Zemax OpticStudio [66] from 4-6 μm and is shown in Fig. 2.

$$V = \frac{n_{center} - 1}{n_{short} - n_{long}} \quad (1)$$

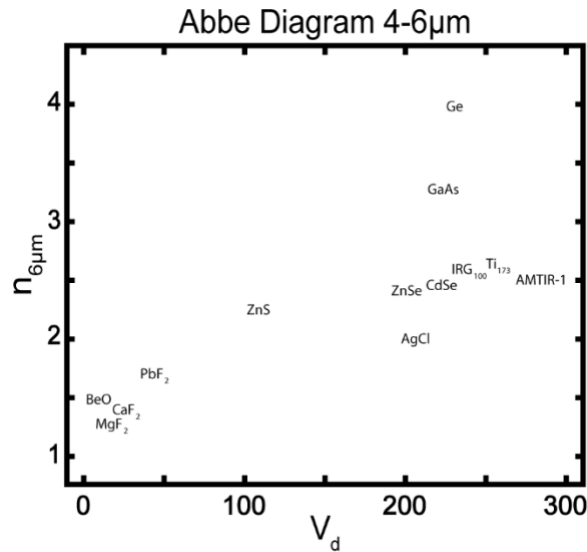


Fig. 2. Abbe Diagram from 4-6 μm ($\sim 1665\text{-}2500\text{ cm}^{-1}$) for mid-IR transmissive materials. Data for refractive index and Abbe number from Zemax OpticStudio [66], and diagram adapted from Salv, et. al. [67]

Numerous combinations of materials on opposite ends of the Abbe Diagram were used to model the doublet lens (data not shown), but ultimately, we chose to use a ZnSe asphere as the second lens and vary the geometric parameters of the first lens. In the end, CaF₂ was chosen because it is a very common mid-IR material and had a wide separation from ZnSe in the Abbe diagram.

215 Optimizations were run to design a manufacturable lens that would fit within the space
 216 constraints of our microscope, allowing every aspect of the CaF₂ lens to vary except for its
 217 aperture size. Due to these manufacturing and space constraints, thickness constraints were
 218 placed on the optic to keep it between 2-15 mm, and on any air spacings (between the two
 219 lenses, and from the asphere to the imaging plane) to be between 2-50 mm, with all edge
 220 thicknesses at a minimum of 2 mm. Contrast optimization produced a CaF₂ lens with a
 221 favorable combination of MTF, spot size, and PSF with similar dimensions to a CaF₂ meniscus
 222 lens that could be purchased off the shelf from Thorlabs. **Table 1** compares the parameters of
 223 the two lenses.

224 **Table 1. A Comparison of the two CaF₂ lenses: the theoretical lens produced by contrast**
 225 **optimization and the real meniscus lens from Thorlabs.**

	Optimal Lens	Real Lens
Radius of Curvature – Front Side (mm)	42.007	42.0
Radius of Curvature – Back Side (mm)	21.151	20.2
Center Thickness (mm)	7.367	4.0
Edge Thickness (mm)	11.662	6.5

226

227 The parameters for the optimized CaF₂ lens were replaced with those from the
 228 commercially available meniscus lens, and the optimization process was redone, this time only
 229 allowing the distances between lenses and from the second lens to the imaging plane to vary.
 230 The resulting metrics did not change dramatically from the optimized case and were deemed
 231 suitable for our purposes.

232 Comparison of the final doublet design with the commercial lens versus the asphere
 233 is shown in **Fig. 3**. These results are for contrast optimization at 30 cy/mm for 4-6 μ m in
 234 OpticStudio. The spot size of the doublet was narrowed to an RMS radius of 12.404 μ m, which
 235 is a huge improvement over that of the asphere at 49.323 μ m. Finally, and most importantly for
 236 experimental comparisons, the normalized MTFs show that the doublet has increased amplitude
 237 at higher spatial frequencies when compared to that of the asphere. This correlates to improved
 238 image resolution.

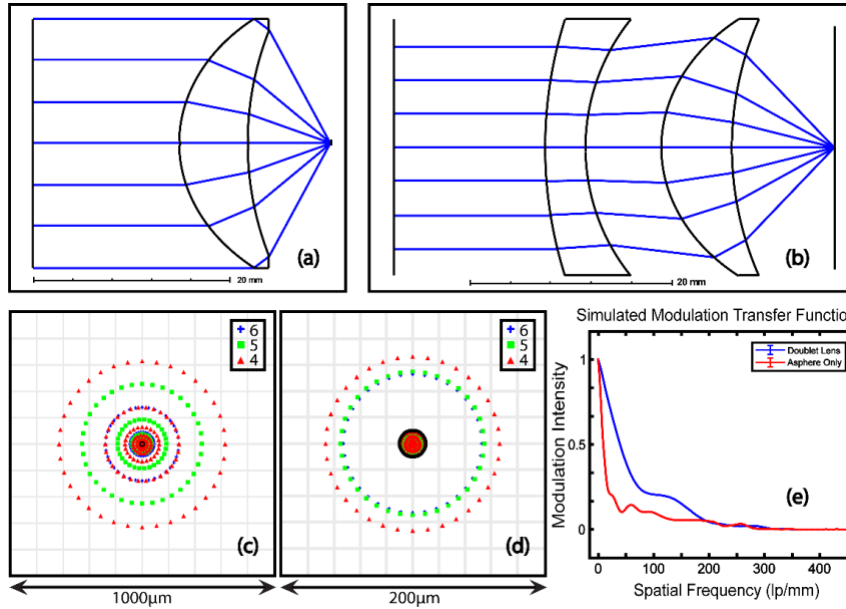


Fig. 3. Comparison of asphere and doublet evaluation methods in OpticStudio. (a) Asphere layout. (b) Doublet layout. (c) Spot size diagram for 4, 5 and 6 μm light through the asphere. (d) Spot size diagram for 4, 5 and 6 μm light through the doublet. The black ring in the middle is the Airy disk size. The black rings in the middle of the spots mark the Airy radii for the given clear aperture of the imaging system. (e) Normalized MTFs for the asphere (red) and doublet (blue). The doublet shows a much higher intensity at lower spatial frequencies, which would result in clearer images.

3.2 Evaluation of the Lens

Both lenses – the CaF_2 meniscus lens (**Table 1**) and the ZnSe asphere – were housed in a slotted lens tube and held in place with adjustable retaining rings. Adjustments were made to the lens spacing, and the microscope was reconfigured with the new doublet prior to collecting and processing images as described in the Methods section. Following this, horizontal cuts of the intensity were taken through the center of the image to avoid consistent imaging artifacts from the edges of the detector array, which can be exemplified by noise in the top right corner of each image. These cuts were vertically averaged over the region shown by the white box in each image. Selected images and their cuts are shown in **Fig. 4**.

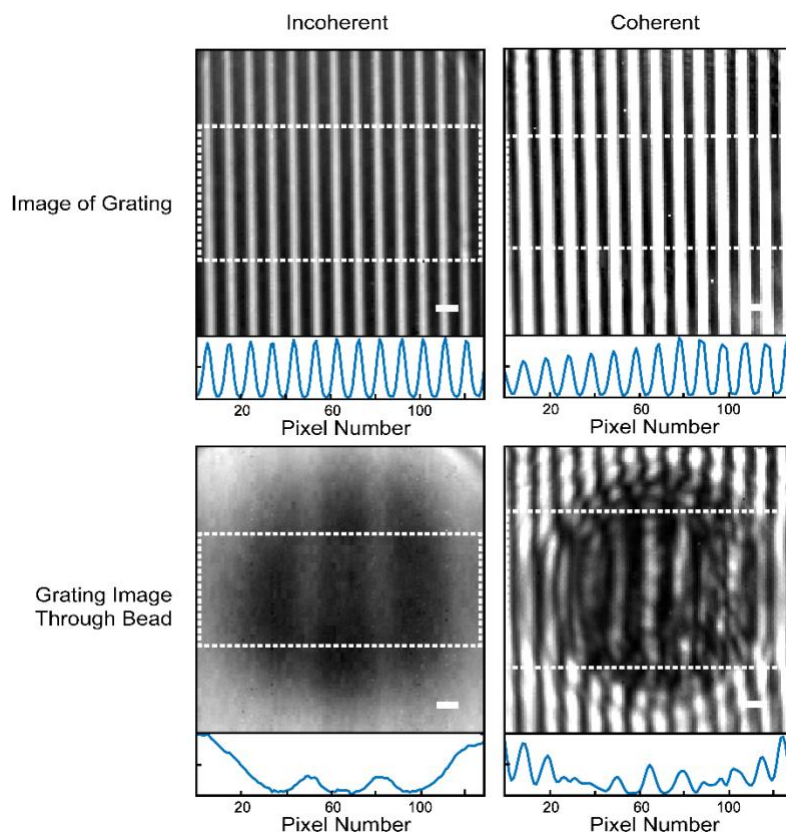


Fig. 4. Comparison of incoherent and coherent microscope images for the 100 line-pairs per millimeter (lp/mm) grating. The top row shows images of just the grating, while the bottom row of images is through 100 μm PS microspheres. The blue trace below each image is the averaged cut taken from the box within the dotted white line on each image. The white scalebar in each image represents 12 μm .

The averaged cuts were each Fourier transformed to pick out the amplitude of the peak nearest the expected spatial frequencies. When analyzing images taken through microspheres, averaged cuts to be Fourier transformed were only taken from the section of the image containing the microsphere to look for possible resolution enhancement, rather than including sections of plain grating for analysis. The intensity of the chosen peak was plotted against the actual spatial frequency of the grating to generate the experimental MTFs. Calculation of the error bars is described previously in Methods. **Fig. 5** shows a comparison of the experimental MTFs produced by the doublet versus by the asphere. MTFs resulting from the coherent light case are shown in **(a)** and the MTFs from the incoherent light are shown in **(b)**. All MTFs were normalized to the 50 line-pairs per millimeter (lp/mm) point.

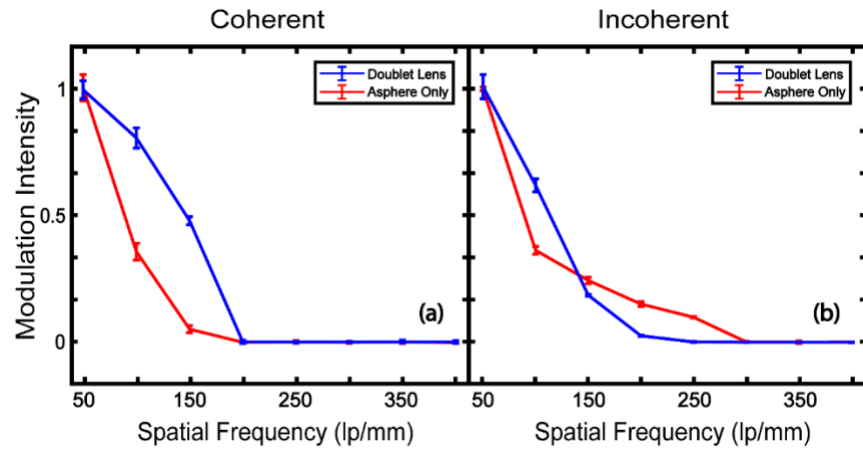


Fig. 5. Normalized MTFs for asphere (red) and doublet (blue) collected with (a) coherent light centered at 6 μm and (b) incoherent mid-IR light. All MTFs were normalized to the 50 lp/mm point.

For the coherent light source, the doublet shows a clear increase in modulation intensity over the asphere at low spatial frequencies, corresponding to resolution enhancement. However, both the asphere and doublet cutoff at the same spatial frequency of 200 lp/mm, which corresponds to the coherent cutoff for 6 μm at ~ 166 lp/mm. This means that when used with a coherent mid-IR light source, the doublet produces images with more distinguishable features than the asphere but is not able to show any features smaller than the asphere is capable. The incoherent light source yields different results. The doublet initially shows some improvement in modulation intensity when imaging at lower spatial frequencies but performs worse than the asphere at spatial frequencies starting at 100 lp/mm and higher. The asphere was capable of imaging smaller features overall than the doublet, as well.

Next, images were taken through PS microspheres placed on the gratings. As discussed earlier, the addition of a PS microsphere has been shown to increase resolution. Thus, the MTFs presented in **Fig. 6** were calculated for the images through a 100 μm PS microsphere on each grating. However, these MTFs appear to not follow the expected behavior. The lack of spatial resolution enhancement with the addition of the PS beads is discussed later.

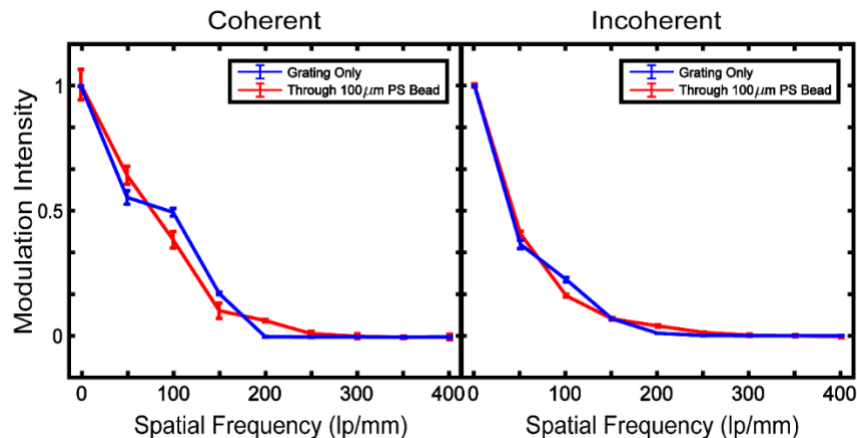


Fig. 6. MTFs taken with the doublet through 100 μm PS microspheres compared to images collected of just the grating for (a) coherent light centered at 6 μm and (b) incoherent mid-IR light. In both graphs, the blue curve is the grating only, and the red curve represents imaging through the PS microspheres.

Finally, FTIR microscope images were collected in order to assess the spatial resolution in FTIR absorption imaging. For this set of data, images were collected of the gratings through NAP solution superimposed on the gratings. Since the gratings were more difficult to see through the NAP than on the clean resolution targets, a smaller selection of grating spacings was collected. A normalized spectrum of the NAP peak used to generate the MTF is shown in **Fig. 7(a)**. The resulting MTF can be seen in **Fig. 7** (red), with the optical MTF collected from the gratings earlier (blue) for comparison.

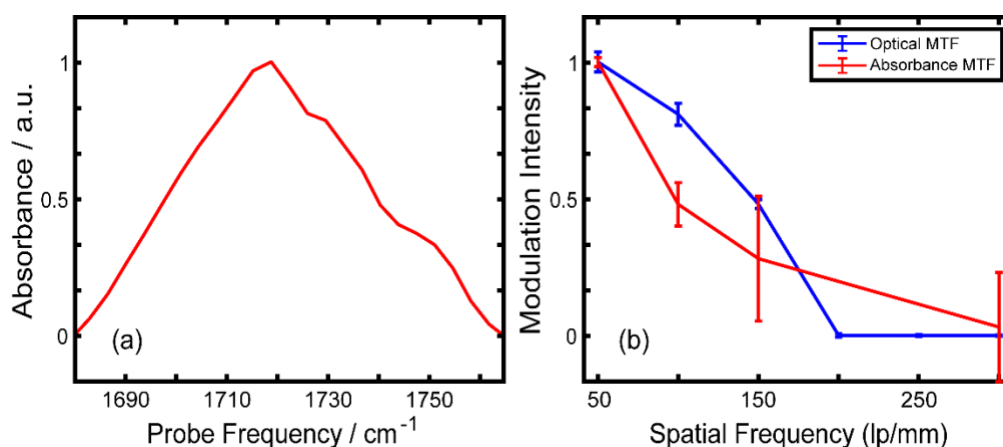


Fig. 7. (a) Normalized FTIR spectra of NAP resulting from the absorbance processing described above. The absorbance peak was collected and baselined for each of the selected grating spacings with the intensity used as the modulation intensities in the absorbance MTFs in (b). (b) Normalized MTFs collected with the doublet lens to compare the spatial resolution through optical intensity (blue) and absorbance measurements (red). The optical MTF (blue) is the same as the blue MTF plotted in **Fig. 6(a)** collected through the plain grating. The absorbance MTF (red) was collected with the absorbance spectrum of NAP on each of the grating spacings.

4. Discussion

As shown in **Fig. 5**, the resolution is improved when imaging with the doublet with coherent light when compared to previous results from the asphere. This is due, in part, to a reduction in coherent ringing artifacts that has previously been reported in coherent mid-IR imaging. [59,68,69] Additionally, when normalizing the grating MTF with coherent light to 0 lp/mm, **Fig. 6**, the experimental doublet produced an MTF that follows a similar shape as the simulated doublet MTF produced in OpticStudio shown in **Fig. 3**. Imaging done with incoherent light, however, did not produce the same results. Instead, the doublet only appears to slightly increase the resolution at 50 and 100 lp/mm, before being outperformed by the asphere. One reason for the incoherent light not performing as well as the coherent light in terms of the MTF might be the fact that the doublet was optimized for 4-6 μm ($\sim 1665\text{-}2500\text{ cm}^{-1}$).

¹) light in OpticStudio. The incoherent light source spans from approximately 1-25 μm ($\sim 400\text{-}10,000\text{ cm}^{-1}$), which is much broader light than the doublet is optimized for, making lower resolution likely. However, in the future, this can be mitigated with a bandpass filter that only allows through a similar wavelength range as that of the coherent light source.

The addition of the 100 μm PS microspheres shows almost no resolution enhancement as compared to images of the bare grating. This is unexpected given our previous work, in which the microspheres enabled us to resolve gratings at much higher spatial frequencies than calculated frequency cutoffs would predict. [59] One possibility for this could be due to an effect caused by the addition of the CaF_2 lens. We hypothesize that because PS has a mid-IR refractive index similar to CaF_2 , [53] the ZnSe asphere and PS previously acted to offset each other, similar to a crude doublet lens. The addition of the CaF_2 may have disrupted this offset, causing the benefits of the microspheres to all but disappear. This effect seems to be worse in the incoherent case, in support of the idea that the incoherent light source is too broad for the doublet as currently designed and arranged. In the MTF generated with FTIR absorption imaging of NAP, we see somewhat similar spatial resolution capabilities as with the intensity MTF, though it does drop off more gradually and ends around the same grating spacing.

While the doublet lens designed here is meant to correct for chromatic aberration in IR imaging systems using broadband sources, an alternative approach would be to utilize a tunable source such as a quantum cascade laser, as is used in discrete frequency IR imaging systems. In such cases, chromatic aberrations could be mitigated by simply adjusting the focal plane at every frequency, without the need of the achromat.

5. Conclusion

Here we show the results of a simple doublet lens that was assembled using all off-the-shelf optics that improves the resolution of our mid-IR microscope by reducing the chromatic aberration present in the system. While our design does not address as wide a wavelength range nor diminish the chromatic aberration as much as some alternative solutions, it is intended to work as a cost- and time- effective solution for ultrafast mid-IR hyperspectral imaging in the 4-6 μm ($\sim 1665\text{-}2500\text{ cm}^{-1}$) range, for both optical (intensity) and chemical absorption imaging. We also hope to further explore the use of the doublet lens with microspheres, and test different materials to compare how they interact with the combination of materials present in the doublet lens.

Funding. This work was funded under NSF 2108727

Acknowledgments. We would like to thank Dean Edun for valuable discussions.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document.

References

[Click or tap here to enter text.](#)

367 1. S. Veerasingam, M. Ranjani, R. Venkatachalapathy, A. Bagaev, V. Mukhanov, D. Litvinyuk, M. Mugilarasan,
368 K. Gurumoorthi, L. Gunganathan, V. M. Aboobacker, and P. Vethamony, "Contributions of Fourier
369 transform infrared spectroscopy in microplastic pollution research: A review," *Crit Rev Environ Sci Technol*
370 **51**, 2681–2743 (2021).

371 2. Y. Bai, D. Zhang, L. Lan, Y. Huang, K. Maize, A. Shakouri, and J. X. Cheng, "Ultrafast chemical imaging by
372 widefield photothermal sensing of infrared absorption," *Sci Adv* **5**, (2019).

373 3. F. Rosi, C. Miliani, R. Braun, R. Harig, D. Sali, B. G. Brunetti, and A. Sgamellotti, "Noninvasive Analysis of
374 Paintings by Mid-infrared Hyperspectral Imaging," *Angewandte Chemie International Edition* **52**, 5258–
375 5261 (2013).

376 4. B. Das, S. Roychowdhury, P. Mohanty, A. Rizuan, J. Chakraborty, J. Mittal, and K. Chattopadhyay, "A Zn-
377 dependent structural transition of SOD1 modulates its ability to undergo phase separation," *EMBO J* **42**,
378 e111185 (2023).

379 5. D. N. Edun, M. R. Flanagan, and A. L. Serrano, "Does liquid-liquid phase separation drive peptide folding?,"
380 *Chem Sci* **12**, 2474–2479 (2021).

381 6. A. Ghosh, J. S. Ostrander, and M. T. Zanni, "Watching Proteins Wiggle: Mapping Structures with Two-
382 Dimensional Infrared Spectroscopy," *Chem Rev* **117**, 10726–10759 (2017).

383 7. A. Ebner, P. Gatteringer, I. Zorin, L. Krainer, C. Rankl, and M. Brandstetter, "Diffraction-limited hyperspectral
384 mid-infrared single-pixel microscopy," *Sci Rep* **13**, 281 (2023).

385 8. M. Jackson, M. G. Sowa, and H. H. Mantsch, "Infrared spectroscopy: A new frontier in medicine," in
386 *Biophysical Chemistry* (Elsevier B.V., 1997), Vol. 68, pp. 109–125.

387 9. P. Lasch, M. Stämmeler, M. Zhang, M. Baranska, A. Bosch, and K. Majzner, "FT-IR Hyperspectral Imaging
388 and Artificial Neural Network Analysis for Identification of Pathogenic Bacteria," *Anal Chem* **90**, 8896–
389 8904 (2018).

390 10. M. Hermes, R. B. Morrish, L. Huot, L. Meng, S. Junaid, J. Tomko, G. R. Lloyd, W. T. Masselink, P. Tidemand-
391 Lichtenberg, C. Pedersen, F. Palombo, and N. Stone, "Mid-IR hyperspectral imaging for label-free
392 histopathology and cytology," *Journal of Optics (United Kingdom)* **20**, 023002 (2018).

393 11. R. A. Schultz, T. Nielsen, J. R. Zavaleta, R. Ruch, R. Wyatt, and H. R. Garner, "Hyperspectral imaging: A
394 novel approach for microscopic analysis," *Cytometry* **43**, 239–247 (2001).

395 12. K. Ataka, S. T. Stripp, and J. Heberle, "Surface-enhanced infrared absorption spectroscopy (SEIRAS) to
396 probe monolayers of membrane proteins," *Biochim Biophys Acta Biomembr* **1828**, 2283–2293 (2013).

397 13. A. L. Serrano, A. Ghosh, J. S. Ostrander, and M. T. Zanni, "Wide-field FTIR microscopy using mid-IR pulse
398 shaping," *Opt Express* **23**, 17815 (2015).

399 14. C. R. Baiz, D. Schach, and A. Tokmakoff, "Ultrafast 2D IR microscopy," *Opt Express* **22**, 18724 (2014).

400 15. S. Junaid, J. Tomko, M. P. Semtsiv, J. Kischkat, W. T. Masselink, C. Pedersen, and P. Tidemand-Lichtenberg,
401 "Mid-infrared upconversion based hyperspectral imaging," *Opt Express* **26**, 2203 (2018).

402 16. C. A. Tibbetts, A. B. Wyatt, B. M. Luther, and A. T. Krummel, "Practical Aspects of 2D IR Microscopy," in
403 *Emerging Trends in Chemical Applications of Lasers* (American Chemical Society, 2021), Vol. 1398, pp.
404 109–134.

- 405 17. J. S. Ostrander, A. L. Serrano, A. Ghosh, and M. T. Zanni, "Spatially Resolved Two-Dimensional Infrared
406 Spectroscopy via Wide-Field Microscopy," *ACS Photonics* **3**, 1315–1323 (2016).
- 407 18. A. Dorsa, Q. Xie, M. Wagner, and X. G. Xu, "Lock-in amplifier based peak force infrared microscopy,"
408 *Analyst* **148**, 227–232 (2023).
- 409 19. G. Srinivasan and R. Bhargava, "Fourier transform-infrared spectroscopic imaging: The emerging evolution
410 from a microscopy tool to a cancer imaging modality," *Spectroscopy (Santa Monica)* **22**, 30–43 (2007).
- 411 20. B. Holcombe, A. Foes, S. Banerjee, K. Yeh, S.-H. J. Wang, R. Bhargava, and A. Ghosh, "Intermediate
412 antiparallel beta structure in amyloid plaques revealed by infrared spectroscopic imaging," *bioRxiv*
413 2023.04.18.537414 (2023).
- 414 21. B. J. Davis, P. S. Carney, and R. Bhargava, "Theory of mid-infrared absorption microspectroscopy: II.
415 Heterogeneous samples," *Anal Chem* **82**, 3487–3499 (2010).
- 416 22. N. Kröger, A. Egl, M. Engel, N. Gretz, K. Haase, I. Herpich, B. Kränzlin, S. Neudecker, A. Pucci, A. Schönhals,
417 J. Vogt, and W. Petrich, "Quantum cascade laser–based hyperspectral imaging of biological tissue," *J.*
418 *Biomed. Opt* **19**, 111607 (2014).
- 419 23. E. N. Lewis, I. W. Levin, P. J. Treado, R. C. Reeder, G. M. Story, A. E. Dowrey, and C. Marcott, "Fourier
420 Transform Spectroscopic Imaging Using an Infrared Focal-Plane Array Detector," *Anal Chem* **67**, 3377–
421 3381 (1995).
- 422 24. F. S. Ruggeri, J. Habchi, S. Chia, R. I. Horne, M. Vendruscolo, and T. P. J. Knowles, "Infrared
423 nanospectroscopy reveals the molecular interaction fingerprint of an aggregation inhibitor with single
424 A β 42 oligomers," *Nat Commun* **12**, (2021).
- 425 25. K. L. A. Chan and S. G. Kazarian, "Correcting the effect of refraction and dispersion of light in FT-IR
426 spectroscopic imaging in transmission through thick infrared windows," *Anal Chem* **85**, 1029–1036 (2013).
- 427 26. G. E. Greaves, D. Kiryushko, H. W. Auner, A. E. Porter, and C. C. Phillips, "Label-free nanoscale mapping of
428 intracellular organelle chemistry," *Commun. Biol* **6**, 583 (2023).
- 429 27. D. Bracha, M. T. Walls, and C. P. Brangwynne, "Probing and engineering liquid-phase organelles," *Nat*
430 *Biotechnol* **37**, 1435–1445 (2019).
- 431 28. H. Zhang, S. Shao, Y. Sun, H. Zhang, S. Shao, and Y. Sun, "Characterization of liquid–liquid phase
432 separation using super-resolution and single-molecule imaging," *Biophys Rep* **8**(1), 2–13 (2022).
- 433 29. X. Zhang, H. Li, Y. Ma, D. Zhong, and S. Hou, "Study liquid-liquid phase separation with optical microscopy:
434 A methodology review," *APL Bioeng* **7**, 21502 (2023).
- 435 30. K. Tachi, T. Hirasawa, S. Okawa, A. Horiguchi, K. Ito, and M. Ishihara, "Chromatic-aberration-free
436 multispectral optical-resolution photoacoustic microscopy using reflective optics and a supercontinuum
437 light source," *Appl Opt* **60**, 9651 (2021).
- 438 31. P. Wang, N. Mohammad, and R. Menon, "Chromatic-aberration-corrected diffractive lenses for ultra-
439 broadband focusing," *Sci Rep* **6**, 1–7 (2016).
- 440 32. B. Guo, Y. Wang, C. Peng, H. L. Zhang, G. P. Luo, H. Q. Le, C. Gmachl, D. L. Sivco, M. L. Peabody, and A. Y.
441 Cho, "Laser-based mid-infrared reflectance imaging of biological tissues," *Opt Express* **12**, (2004).
- 442 33. J. Kilgus, G. Langer, K. Duswald, R. Zimmerleiter, I. Zorin, T. Berer, and M. Brandstetter, "Diffraction
443 limited mid-infrared reflectance microspectroscopy with a supercontinuum laser," *Opt Express* **26**, (2018).

- 444 34. J. Sasian, "Method to design apochromat and superachromat objectives," *Optical Engineering* **56**, 1
445 (2017).
- 446 35. Q. Lu, Y. Ding, W. Wang, S. Liu, and M. Xu, "VIS-NIR superachromatic triplet design with five-color
447 correction for a broadband interferometer," *Appl Opt* **61**, 8880 (2022).
- 448 36. A. M. Hanninen and E. O. Potma, "Nonlinear optical microscopy with achromatic lenses extending from
449 the visible to the mid-infrared," *APL Photonics* **4**, 080801 (2019).
- 450 37. A. P. Wood, "Design of infrared hybrid refractive–diffractive lenses," *Appl Opt* **31**, 2253 (1992).
- 451 38. Y. Qiu, J. Li, T. Li, X. Ma, and X. Jiang, "Photo-Curing Vis-IR Hybrid Fresnel Lenses with High Refractive
452 Index," *Macromol Chem Phys* **222**, 2100311 (2021).
- 453 39. Y. Yuan, Z. Yan, P. Zhang, Z. Chang, F. Peng, R. Chen, Z. Yang, S. Chen, Q. Zhao, and X. Huang, "A
454 Broadband Achromatic Dielectric Planar Metalens in Mid-IR Range," *Photonic Sensors* **13**, 1–9 (2023).
- 455 40. F. Aieta, M. A. Kats, P. Genevet, and F. Capasso, "Multiwavelength achromatic metasurfaces by dispersive
456 phase compensation," *Science* (1979) **347**, 1342–1345 (2015).
- 457 41. N. Ji, "Adaptive optical fluorescence microscopy," *Nat Methods* **14**, 374–380 (2017).
- 458 42. P. S. Salter and M. J. Booth, "Adaptive optics in laser processing," *Light Sci Appl* **8**, 1–16 (2019).
- 459 43. Y. Liu, Z.-H. Yang, Y.-J. Yu, L.-A. Wu, M.-Y. Song, and Z.-H. Zhao, "Chromatic-Aberration-Corrected
460 Hyperspectral Single-Pixel Imaging," *Photonics* **10**, 7 (2022).
- 461 44. J. Wu, D. Li, A. Cui, J. Gao, K. Zhou, and B. Liu, "Digital infrared chromatic aberration correction algorithm
462 for membrane diffractive lens based on coherent imaging," *Appl Opt* **61**(34), 10080–10085 (2022).
- 463 45. J. T. Korneliussen and K. Hirakawa, "Camera processing with chromatic aberration," *IEEE Transactions on*
464 *Image Processing* **23**, 4539–4552 (2014).
- 465 46. M. Kozubek and P. Matula, "An efficient algorithm for measurement and correction of chromatic
466 aberrations in fluorescence microscopy," *J Microsc* **200**, 206–217 (2000).
- 467 47. M. M. Eid, "Characterization of Nanoparticles by FTIR and FTIR-Microscopy," *Handbook of Consumer*
468 *Nanoproducs* 1–30 (2021).
- 469 48. G. L. Liu and S. G. Kazarian, "Recent advances and applications to cultural heritage using ATR-FTIR
470 spectroscopy and ATR-FTIR spectroscopic imaging," *Analyst* **147**, 1777–1797 (2022).
- 471 49. K. Seidl, J. Knobbe, and H. Gröger, "Design of an all-reflective unobscured optical-power zoom objective,"
472 *Appl. Opt.* **48**(21), 4097-4107 (2009).
- 473 50. K. Yeh, I. Sharma, K. Falahkheirkhah, M. P. Confer, A. C. Orr, Y. T. Liu, Y. Phal, R. J. Ho, M. Mehta, A.
474 Bhargava, W. Mei, G. Cheng, J. C. Cheville, and R. Bhargava, "Infrared spectroscopic laser scanning
475 confocal microscopy for whole-slide chemical imaging," *Nature Communications* 2023 14:1 **14**, 1–12
476 (2023).
- 477 51. E. S. Lee, S.-W. Lee, J. Hsu, E. O. Potma, H. 3 R. C. Mendelsohn, M. E. Chen, and D. J. Rerek, "Vibrationally
478 resonant sum-frequency generation microscopy with a solid immersion lens," *Biomed. Opt. Express* **5**(7),
479 2125-2134 (2014).

480 52. K. L. A. Chan and S. G. Kazarian, "Aberration-free FTIR spectroscopic imaging of live cells in microfluidic
481 devices," *Analyst* **138**, 4040–4047 (2013).

482 53. K. L. A. Chan, A. Altharawi, P. Fale, C. L. Song, S. G. Kazarian, G. Cinque, V. Untereiner, and G. D.
483 Sockalingum, "Transmission Fourier Transform Infrared Spectroscopic Imaging, Mapping, and Synchrotron
484 Scanning Microscopy with Zinc Sulfide Hemispheres on Living Mammalian Cells at Sub-Cellular
485 Resolution," *Appl Spectrosc* **74**, 544–552 (2020).

486 54. J. A. Kimber, L. Foreman, B. Turner, P. Rich, and S. G. Kazarian, "FTIR spectroscopic imaging and mapping
487 with correcting lenses for studies of biological cells and tissues," *Faraday Discuss* **187**, 69–85 (2016).

488 55. X. Hao, C. Kuang, X. Liu, H. Zhang, and Y. Li, "Microsphere based microscope with optical super-resolution
489 capability," *Appl Phys Lett* **99**, 203102 (2011).

490 56. K. W. Allen, N. Farahi, Y. Li, N. I. Limberopoulos, D. E. Walker, A. M. Urbas, V. Liberman, and V. N.
491 Astratov, "Super-resolution microscopy by movable thin-films with embedded microspheres: Resolution
492 analysis," *Ann Phys* **527**, 513–522 (2015).

493 57. H. Yang, N. Moullan, J. Auwerx, and M. A. M. Gijs, "Super-Resolution Biological Microscopy Using Virtual
494 Imaging by a Microsphere Nanoscope," *Small* **10**, 1712–1718 (2014).

495 58. V. N. Astratov, A. V. Maslov, K. W. Allen, N. Farahi, Y. Li, A. Brettin, N. I. Limberopoulos, D. E. Walker, A. M.
496 Urbas, V. Liberman, and M. Rothschild, "Fundamental limits of super-resolution microscopy by dielectric
497 microspheres and microfibers," in *Nanoscale Imaging, Sensing, and Actuation for Biomedical Applications*
498 *XIII* (SPIE) (2016), paper 97210K.

499 59. D. N. Edun, C. E. Nelmark, and A. L. Serrano, "Resolution Enhancement in Wide-Field IR Imaging and Time-
500 Domain Spectroscopy Using Dielectric Microspheres," *Journal of Physical Chemistry A* **124**, 5534–5541
501 (2020).

502 60. B. S. Luk'yanchuk, R. Paniagua-Domínguez, I. Minin, O. Minin, and Z. Wang, "Refractive index less than
503 two: photonic nanojets yesterday, today and tomorrow [Invited]," *Opt Mater Express* **7**, 1820 (2017).

504 61. H. Yang, R. Trouillon, G. Huszka, and M. A. M. Gijs, "Super-Resolution Imaging of a Dielectric Microsphere
505 Is Governed by the Waist of Its Photonic Nanojet," *Nano Lett* **16**, 4862–4870 (2016).

506 62. J. Liang, M. Tu, W. Liu, Z. Qiao, G. Cui, al Jianfeng Liang, and W. Peng, "Optical constant determination of
507 cross-linked polystyrene in the infrared," **10623**, 63–68 (2018).

508 63. B. Boeckx, R. Ramaekers, and G. Maes, "The influence of the peptide bond on the conformation of amino
509 acids: A theoretical and FT-IR matrix-isolation study of N-acetylproline," *Biophys Chem* **159**, 247–256
510 (2011).

511 64. Z. Zhang and R. Lu, "Initial structure of dispersion objective for chromatic confocal sensor based on
512 doublet lens," *Opt Lasers Eng* **139**, 106424 (2021).

513 65. B. Gleason, "Designing Optical Properties in Infrared Glass," Ph.D. dissertation (Clemson University, 2015).

514 66. Zemax (An Ansys Company); OpticStudio.

515 67. A. R. Salvà, "OUFTI-NEXT: STUDY OF REFRACTIVE LENSES FOR AN INFRARED CAMERA," Master's Thesis
516 (The Technical University of Catalonia, 2019).

517 68. K. Yeh, S. Kenkel, J. N. Liu, and R. Bhargava, "Fast infrared chemical imaging with a quantum cascade
518 laser," *Anal Chem* **87**, 485–493 (2015). 68. S. Ran, S. Berisha, R. Mankar, W.-C. Shih, and D. Mayerich,

519 "Mitigating fringing in discrete frequency infrared imaging using time-delayed integration," Biomed Opt
520 Express **9**, 832 (2018).

521 69. S. Ran, S. Berisha, R. Mankar, W.-C. Shih, and D. Mayerich, "Mitigating fringing in discrete frequency
522 infrared imaging using time-delayed integration," Biomed Opt Express **9**, 832 (2018).

523

524

525

526