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Nonlinear optical microscopy with achromatic lenses extending from the visible to the mid-infrared

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ABSTRACT
With the advent of near-infrared broadband sources stretching into the mid-infrared (MIR) region, there is a growing demand for optical components with utility over an increasingly broad spectral range. For refractive lenses, color correction over such broad bandwidths can be a challenge. In this work, we discuss and demonstrate a two-element lens design with achromaticity spanning the visible to the mid-infrared. The air-spaced doublet designed from commercially available materials shows a significant reduction in spot size and chromatic shift compared to single lens alternatives. We have tested these new broad bandwidth achromats for the purpose of laser-scanning sum-frequency generation microscopy, confirming their improved performance for nonlinear optical imaging applications. The super broadband achromatic lenses represent an attractive alternative to reflective components in ultrabroadband applications, as they enable compact transmission-based optical designs and good focusing performance at off-axis field angles.

I. INTRODUCTION
The ability to generate high-brilliance, coherent mid-infrared (MIR) radiation has improved significantly over the past decade. Continuing developments in quantum cascade lasers, parametric oscillators, and super continuum light sources have made a wavelength range that was once accessible only with synchrotron light sources now available for benchtop applications. With the growing availability of coherent radiation over a wider spectral range, broadband applications that utilize light anywhere in the visible-MIR range are becoming ever more practical.

A case in point is nonlinear optical (NLO) microscopy based on MIR light to excite selective molecular vibrational modes and visible/near-infrared (NIR) light for probing the excited vibrations. Both photothermal MIR microscopy and sum-frequency generation (SFG) microscopy are based on this principle, which makes it possible to probe MIR-driven spectroscopic transitions with conventional visible photo-detectors. Such applications also underline the need for optical components that show high performance over an exceptionally wide spectral range. Since conventional optical microscopy systems are based on refractive focusing elements, expanding their use into the MIR range poses several challenges. Refractive lenses exhibit chromatic aberrations, and reducing such aberrations becomes particularly relevant for imaging applications that rely simultaneously on visible/NIR and MIR laser beams. Therefore, for MIR-based NLO microscopy and other broadband imaging applications, development of super broadband achromatic lens systems is highly desirable.

An achromatic lens consists of at least two lens components with different refractive properties that together effectively cancel chromatic aberrations. Such achromatic lenses are relatively standard in the visible range of the spectrum and a myriad of visible achromats are commercially available. However, fewer solutions exist in the near-infrared (NIR) because suitable materials that display both good transmission and dispersion properties are scarcer and only a handful of options are available for the mid-infrared (MIR). Many of the available NIR and MIR achromats are made out of materials that exhibit limited optical transmission in the
visible range. Conversely, achromats designed for the visible range generally display unfavorable transmission properties for NIR/MIR light.

Conventional lens materials include amorphous glasses based on silicon-dioxide (SiO₂), or silica, with a small percentage of varying dopants. Of the 200 commonly used glasses, none show favorable transmission characteristics in the MIR due to strong IR absorption beyond 2.5 μm attributed to the hydroxyl bonds in silica. This limitation has severely hampered the advancement of red-shifted achromats, and although they can be designed for color correction out to their transmission limit, they cannot operate beyond the 2.5 μm cliff. Mid-infrared achromats made of crystallized silicon and germanium components are commonly used to achieve achromacity between 3 and 5 μm; however, they exhibit transparency limitations in the visible and NIR. An alternate solution for color correction is a hybrid lens which uses both a curved refracting surface and a diffractive surface, a solution that has yet to demonstrate its utility for ultrabroadband applications. Multiple high-performance imaging systems have been developed and commercialized for hyperspectral and multispectral applications, spanning the visible to the MIR using refractive systems, zoom lenses, and folded geometry reflective systems.11 Broadband imaging systems are attractive because they can capture a single field of view (FOV) onto focal plane arrays (FPA) while covering multiple spectral windows.11 This has many advantages including less weight, a requisite for field deployment, aeronautic applications, and airborne reconnaissance.12

The conventional solution for focusing and conditioning broadband radiation is the use of reflective optics such as parabolic mirrors or three mirror anastigmats, or to use a lens singlet with broad transmission such as fluorites, e.g., calcium-fluoride. Simple light focusing and light collection achieved with parabolic mirrors elegantly combine two great attributes: they eliminate spherical aberration and are inherently achromatic. These single element curved mirrors perform well on-axis, but off-axis focusing is significantly hindered by deleterious aberrations rendering these components less attractive for laser-scanning systems.13 Singlet lenses do not offer diffraction-limited focusing on-axis due to spherical aberration. However, their off-axis performance is only marginally worse compared to their on-axis performance, which makes them still attractive for beam scanning applications. Achromatic doublets are the preferred implementation as they embody the best attributes of both low-performance alternatives, like the color correction, and diffraction limited performance on-axis of the reflector and the minimal aberrations off-axis of the singlet lens.

In this work, we present a new doublet lens system with achromatic focusing capabilities in the range spanning from the visible to ~4.5 μm. We discuss the focusing performance of the super broadband achromatic lens and demonstrate its utility for the imaging system of a NIR/MIR laser-scanning SFG microscope.

II. ATTRIBUTES OF BROADBAND ACHROMATIC LENSES

A. Material selection and lens design

For a two-element lens system that eliminates primary color at a given lens power (ϕ), the linear dispersion must be zero and satisfy the following equations:

\[ \phi_{\text{total}} = \sum_{n=1}^{3} \phi_n = \phi_1 + \phi_2, \]

\[ \sum_{n=1}^{2} \frac{\phi_n}{V_n} = \frac{\phi_1}{V_1} + \frac{\phi_2}{V_2} = 0, \]

where \( V_n \) is the modified IR-Abbe number defined over the wavelength range of interest,

\[ V_n = \frac{n(\lambda_2) - 1}{n(\lambda_1) - n(\lambda_2)}. \]

Here, \( n \) is the index of refraction and \( \lambda_1 = 1 \mu m, \lambda_2 = 3 \mu m, \) and \( \lambda_3 = 5 \mu m \). These wavelengths were chosen based on the targeted spectral range, although somewhat arbitrarily, as there is no standard definition for the Abbe number outside the visible spectrum. Rearranging Eq. (2) shows that \( \phi_n = -\phi_2 V_1/V_2 \). Expanding Eqs. (1) and (2) to a third term would enable an apochromat triplet, albeit at the expense of simplicity. In the preferred embodiment of a doublet, the Fraunhofer design, a positive-power low index crown element is followed by a negative-power high index flint element with a large difference in Abbe numbers to reduce the optical power of each component. In principle, lens doublets can be either cemented together for easier alignment and fabrication, or separated by an air-spacer for improved design performance. However, lenses designed for the MIR cannot be cemented and must be air-spaced due to transmission loss. Optical cements are commonly variations of PDMS (polydimethylsiloxane), which suffer from the same parasitic optical absorption at wavelengths above 2.5 μm as glass.

For our design, we have selected calcium-fluoride (CaF₂) and sapphire as the crown and flint elements, respectively. These substrates are suitable for practical infrared refracting optical systems based on their optical transmission, complimentary dispersion properties, mechanical strength, and availability.16,17 Other substrates such as the soluble salts, e.g., sodium chloride and potassium bromide, have been excluded due to their restriction to applications in carefully controlled environments. KRS-5 and arsenic trisulfide were excluded because of toxicity, limited transmission in the visible, and excessive softness which limit manufacturability. ZnSe and ZnS are commonly used for thermal imaging applications; however, their disparate partial dispersion values and large optical nonlinearities preclude compatibility for color corrected doublets with high-power and pulsed lasers. In addition, currently available chalcogenide glasses are not transparent in the visible. Of the fluorite crystals, calcium fluoride has the best mechanical properties and is commonly used. CaF₂, however, has a low refraction index which requires high surface curvatures, resulting in increased spherical aberrations. CaF₂ also has an anomalous partial dispersion that helps to reduce the degree of the secondary spectrum. Sapphire is commonly used and is reasonably priced compared to other MIR materials. Although it cannot be conveniently diamond-turned due to its hardness, special suppliers are able to produce components at a high yield. Sapphire is birefringent, yet when the uniaxial crystal axis is aligned with the optical axis, known as C-cut, birefringence can be suppressed with little additional cost, enabling polarization.
resolved applications. We have designed the elements with spherical surfaces for cost efficiency. While possible to use precision molding of chalcogenide glasses to form IR optical components with aspherical and freeform features, aside from the large setup costs for tooling, the components are often used as single elements without color correction. Some relevant optical properties of CaF$_2$ and sapphire are summarized in Table I.

B. Ray tracing analysis

Figure 1 highlights the performance differences between the three different approaches to focusing: (a) reflective geometry, (b) a refractive singlet, and (c) a refractive doublet. Here, a 1/2-in. aperture is centered on a 1.0-in. optic with a 2.0-in. focal length. Two field angles are shown, on-axis (black) and a 2° field angle (red). The corresponding spot diagrams, computed in Zemax, provide a measure of the quality of the focal spot. The black circle represents the Airy disk, and diffraction limited performance is obtained when all spots fall within this circle. Simulations are shown for two wavelengths, a 1030 nm NIR beam (blue) and a 3.6 μm MIR beam (green), for each of the focusing geometries. Figure 1(a) shows that the on-axis performance of the reflective focusing element is excellent, a quality that is maintained for both target wavelengths as the reflective optic is inherently achromatic. However, the off-axis performance is compromised. The spot has expanded by more than an order of magnitude, indicated by the many spots that fall well outside of the black circle; the latter can be found in the middle of the diagram with a much smaller relative size.

It is clear from Fig. 1(b) that the focusing performance of the singlet CaF$_2$ lens on axis is worse than that of the reflective element. For both wavelengths, spots can be found beyond the black circle, mostly due to spherical aberrations. Although the spot diagram for the off-axis focusing geometry also shows a performance affected by aberrations, the relative difference between the on-axis and off-axis relative spot sizes is within 10% for both target wavelengths. The similarity in the spot size at both field angles comes from a balance of aberrations including (chromatic) defocus, spherical, coma, astigmatism, and distortion.

The refractive doublet shown in Fig. 1(c), on the other hand, exhibits diffraction limited performance on axis for the 1030 nm beam and near diffraction limited performance for the 3.6 μm beam. Off axis, aberrations affect the quality of the focal spot; however, the relative rms spot size is about one order of magnitude smaller than the off-axis focal rms spot size obtained in the reflective geometry.

| TABLE I. Properties of optical substrates used for super broadband color correction. |
|-----------------------------------|-----------------|-----------------|
|                                   | CaF$_2$         | Sapphire        |
| $n(\lambda_1)$, $n(\lambda_2)$, $n(\lambda_3)$ | 1.4289, 1.4179, 1.7557, 1.7122, 1.6239 | |
| IR-Abbe number ($V_n$)            | 13.98           | 5.41            |
| Transmission                      | 180 nm–8.0 μm   | 150 nm–5.0 μm   |
| IR-partial dispersion             | 0.3666          | 0.3300          |
| Knoop hardness                    | 158             | 1370            |

Importantly, both the NIR and MIR beams are affected to a similar degree, retaining a good overlap of the energy contained in the focal spots at the two different wavelengths. This discussion underlines that the refractive doublet offers reduced aberrations for the target wavelengths at off-axis focusing angles, which is directly relevant for laser-scanning optical imaging techniques such as NLO microscopy.

Figure 2 shows the axial chromatic focal shift of a CaF$_2$ singlet vs the doublet, computed for a 50 mm focal length. Over the broad spectral range from 800 nm to 4.5 μm, the achromat, shown in red, shows a narrow degree of the secondary color with a maximum focal shift of 0.375 mm and perfect color correction at 1030 nm and 3.60 μm. The relative axial chromatic shift when tuning the MIR from 3.0 μm to 4.0 μm, a wavelength regime relevant for some of the experiments reported here, is 0.163 mm. The singlet lens, indicated by the black line, on the other hand, has 3.16 mm of continuous chromatic shift in the axial dimension in the 800 nm–4.5
μm range. Note that the target wavelengths are chosen to coincide with the output of a Yb\textsuperscript{3+}-fiber laser and a MIR frequency that is in the vicinity of the fundamental CH-stretching vibrational modes. However, the optical design can be readily modified to instead accommodate visible or even UV radiation. By adjusting the design wavelengths in the simulation, achromaticity can be easily achieved over the entire transparency window of these components as specified in Table I, spanning both vibrational and electronic resonance regimes.

C. Lens fabrication

We have fabricated two broadband achromatic lenses using CaF\textsubscript{2} as the crown and C-cut sapphire as the flint material. The first lens is a 25.4 mm diameter optic with a focal length of 50 mm. This lens is designed to function as a scan lens in a laser-scanning microscope. The second lens is a 32 mm diameter optic with a focal length of 180 mm. This lens is designed to function as a tube lens and its dimensions mimic the dimensions of a standard tube lens in an Olympus IX71 frame. Both lenses are designed for complete achromatization at the target wavelengths of 1030 nm and 3.6 μm. Using only spherical surfaces, both lenses are engineered to suppress aberrations. At a 90% aperture, on-axis performance shows a Strehl ratio greater than 0.93 in both systems. This value could have approached 1 but is marginally compromised, although still above the diffraction limit criterion, in favor of off-axis performance. Optimizing the lenses for incorporation into a beam-scanning microscope, we have judiciously balanced imaging performance at multiple field angles. Another consideration for nonzero field angles is lateral chromatic aberration, as it is equally important to ensure the beam overlap at the image plane, as is illustrated in Fig. 1(c) by the overlap of the blue and green dots in the off-axis spot diagram.

We have performed Monte Carlo simulations in Zemax using standard precision tolerances to determine manufacturability. The worst offenders in the analysis are surface and element decenteration of the sapphire element, and the air-spacer thickness due to the large ray angles after the first element. The scan lens is a fast f/6 optic, which benefits from looser tolerances with similar performance. The lenses are manufactured to specifications by Supply Chain Optics (Irvine, CA) with a commercially available broadband antireflection (BBAR) coating from 1 to 5 μm applied to the sapphire element, while the CaF\textsubscript{2} element was left uncoated due to its already low index of refraction and consequently <3% Fresnel reflection loss per surface interface. The assembled lenses are shown in Fig. 3.

III. EXPERIMENTAL SETUP

The two super achromatic lenses considered in this work are designed to function as a scan and tube lens of a laser-scanning microscope. To evaluate the performance of the lenses, we have characterized the linear and nonlinear imaging properties of the microscope with the achromats incorporated and compared the results with the imaging properties attained with CaF\textsubscript{2} singlet scan and tube lenses. The light source used in this study is a picosecond Yb\textsuperscript{3+}-fiber laser (Aeropulse, NKT Photonics), producing 5 ps pulses at 1030 nm with a repetition rate of 76 MHz. This laser functions as a pump source for a synchronously pumped optical parametric oscillator (Levante OPO IR, A.P.E., Berlin), delivering a signal beam and an idler beam. Both the signal and idler beams can be spectrally tuned. For the studies discussed here, the idler is tuned to 3.401 μm, which coincides with the 2940 cm\textsuperscript{-1} CH-stretching vibration of collagen fibers.

The idler beam is spatially filtered and collinearly recombined on a dichroic mirror with the 1030 nm pump beam after passing a delay stage. Both beams are steered toward a laser-scanning microscope (Fluoview 300, Olympus) interfaced with an inverted microscope frame (IX71, Olympus). The aluminum-coated galvanometric mirrors support both the NIR and MIR beams. The scan lens of the Fluoview system has been replaced with either a CaF\textsubscript{2} singlet (f = 50 mm) or a CaF\textsubscript{2}/sapphire scan lens. Similarly, the tube lens in the microscope frame has been substituted with a CaF\textsubscript{2} singlet (f = 150 mm) or with a CaF\textsubscript{2}/sapphire tube lens. The objective lens is a reflective Schwarzschild-Cassegrain (5007, Beck) with a numerical aperture (NA) of 0.65. Since this lens is reflective, no chromatic aberrations are introduced by the microscope objective, allowing a
direct comparison of chromatic effects introduced by the scan/tube lens system.

For the linear imaging experiments, the NIR and MIR beams are detected in transmission mode using a InAs-photovoltaic detector (P10090-01, Hamamatsu), which exhibits good photosensitivity in the 1.0–3.6 μm range. The imaging targets are a 1951 USAF resolution test chart for evaluating the imaging resolution and contrast and a Ronchi ruling for evaluating lateral field distortions within the field of view (FOV). For the nonlinear imaging experiments, we used the SFG signal to characterize the quality of the beam overlap between the MIR (ω₁) and NIR (ω₂) beams, and to study the chromatic focal shift across the field of view. In SFG, the NIR and MIR beams combine to generate a signal at ω₃ = ω₁ + ω₂; see Fig. 4(a). In the experiments reported here, the SFG signal is generated at λ₃ = 792 nm. Average intensities at the sample are ~5 mW for the MIR and up to 30 mW for the NIR. The signal is captured in the forward propagation direction by a refractive condenser lens (NA = 1.40 oil), filtered by using a 780 ± 20 nm bandpass filter, and directed to a photomultiplier tube (7422-50, Hamamatsu). See Fig. 4(b) for a schematic of the optical setup. The detected signal is electronically amplified and sent to a discriminator for single photon counting.

IV. EXPERIMENTAL RESULTS

A. Linear imaging performance

Before studying the imaging performance of the lenses for nonlinear SFG microscopy, we have characterized the linear imaging properties of the combined achromatic scan and tube lenses. Figures 5(a) and 5(c) show a 1951 USAF resolution chart visualized with standard CaF₂ singlet lenses for illumination at 1030 nm and 3.60 μm, respectively. In the image taken at 1030 nm, all features are resolved well. In contrast, the MIR image appears blurry. The line structures labeled 1 have a width of 3.91 μm and are not completely resolved, as shown by the red line in the cross-cut depicted in Fig. 5(e). Although the MIR wavelength is longer, the diffraction-limited resolution at this wavelength is ~2.8 μm, and we thus expect these features to be completely resolved. The main reason for the blurry appearance is the axial chromatic shift introduced by the refractive part of the imaging system, giving rise to a defocus of the MIR beam relative to the focus of the NIR beam. The axial chromatic shift is suppressed when using the achromatic scan and tube lenses, as shown in panels 5(b) and 5(d). Here, the focal plane of the MIR beam is within the depth of focus of the NIR beam, resulting in a much sharper image. By suppressing the defocus, the 3.91 μm wide line structures are now completely resolved with the 3.60 μm beam, as shown by the black line in Fig. 5(e).

We also expect the lateral chromatic shift across the field of view to be improved when using an achromatic imaging system. A measure of the lateral chromatic shift can be obtained from a Ronchi ruling. Figure 6(a) shows an image of the Ronchi ruling (5 μm/line pair) taken at 1030 nm using the singlet scan/tube lens system. By comparing the images obtained at 1030 nm and 3.60 μm, a measure of the lateral deviation due to chromatic aberrations can be obtained. Figure 6(b) depicts the relative lateral...
chromatic shift between 1030 nm and 3.60 μm. It can be seen that the lateral deviation for the singlet scan/tube system, shown in red, is rather significant. The total chromatic shift is more than 9 μm across the 175 μm FOV. For NLO imaging, the lateral shift needs to be smaller than the diffraction limited focal spot size of the MIR beam (~2.8 μm) to enable imaging across the full FOV. Clearly, this cannot be achieved with the singlet scan/tube lens system. Much better performance is observed with the achromatic doublet system, shown by the black line in Fig. 6(b). The total chromatic shift across the FOV is 2.2 μm, which is within the diffraction-limited spot size of the MIR beam. Consequently, we expect good quality NLO signals over the full FOV when the achromatic doublet scan/lens system is used.

B. Nonlinear imaging performance

The broadband achromat has been specifically designed to improve the imaging properties of the SFG modality of the NLO microscope. In NLO microscopy, two or more photons are focused to a common focal volume to induce a nonlinear polarization, which produces radiation that can be captured with a far-field photodetector. Paired with beam scanning mirrors and an optical relay, a NLO microscope allows for rapid generation of images by raster-scanning collinearly focused laser beams across the sample to achieve submicron resolutions. Since in SFG the ω₁ and ω₂ photons are spectrally distant, achieving a good overlap of the two tightly focused beams for various beam angles is a challenge. Common refractive optics of laser-scanning microscopes are incompatible to handle both NIR and MIR input beams. The achromatically corrected scan tube lens presented here enables laser-scanned SFG microscopy with much improved imaging performance.

An example is shown in Fig. 7, where clusters of 0.1 μm barium titanate (BaTiO₃) nanoparticles on a coverslip are visualized. BaTiO₃ exhibits a high nonresonant χ(2) and is a good test sample to examine the SFG imaging performance. In panel 7(a), the sample is imaged with CaF₂ singlets for the scan/tube lens relay. Whereas the SFG signal in the center of the image is optimized for maximum overlap of the NIR and MIR beams, the resolution and magnitude of the SFG signal decrease toward the margins of the image. This observation can be largely attributed to the lateral chromatic shift in the focal plane, which affects the spatial overlap of the NIR and MIR foci. For larger beam angles, the chromatic shift is more pronounced, thus lowering the signal and spatial resolution away from the center of the image. The lateral chromatic shift is dramatically reduced when using the achromatic scan/tube lens system, as shown in panel 7(b). The SFG signal is significantly stronger across the image, especially toward the margins, effectively increasing the FOV. This corroborates the findings shown in Fig. 6(b). In addition, the imaging resolution remains high even near the image margins, resolving features that are not resolved in the image of panel 7(a). By reducing chromatic aberrations, several individual nanoparticles are seen with the achromatic scan/tube system that remain invisible with the CaF₂ singlet-based relay system.

These new optical components enable high-contrast and fast imaging using the SFG modality. The improved image quality across
a larger FOV makes the use of the achromatic doublets more practical for imaging larger areas of the sample. This capability is particularly important for imaging of biological tissues, which display microscopic features at the mesoscale. Figure 8 shows an SFG mosaic of a rat tail, which is rich in fibers of type I collagen. Strong SFG signals are seen when tuning $\omega_1$ to the 2950 cm$^{-1}$ vibrational resonances of collagen.22,23 This image is the result of four frames stitched together. Because of the improved FOV, intensity variations near the seam lines between the image frames are virtually absent. Each individual image is captured at 1 frame/s and averaged over 6 frames. This figure highlights the readiness of tabletop SFG microscopy for biological applications.

V. DISCUSSION

Expanding the spectrally accessible range in optical microscopy implies expanding spectroscopic capabilities. The NIR range includes important spectroscopic transitions, mainly vibrational overtones and combination bands that carry chemical information.24 Extending into the MIR allows targeting the much stronger fundamental vibrational modes of molecules. Opening up the accessible window to 4.8 $\mu$m, which can be reached with 1030 nm-pumped OPOs, enables spectroscopic imaging based on the N–H, O–H, C–H, and C≡N stretching vibrations. Given that these chemical moieties are the building blocks of organic structures, the expansion from the VIS/NIR into the MIR opens up a whole new set of molecular fingerprints that can be used in sensing/imaging applications. The achromatic doublets make it possible to collect images without chromatic blur, as demonstrated here for SFG microscopy.

The utility of broadband achromats operating over the NIR/MIR range goes beyond SFG microscopy. The innovation reported herein has potential benefits for fields which require broadband light sources including high harmonic XUV generation, MIR diagnostics, hyperspectral sensing based on vibrational overtones, combination bands, and now fundamentals in the MIR. In addition, other types of microscopy such as two- and three-photon fluorescence with long wavelength NIR light sources could also benefit from achromatic imaging optics in the 0.8–3 $\mu$m range. IR-based photothermal microscopy, which uses a MIR excitation beam and a visible probe beam, is another technology for which achromatic imaging optics can be used to improve imaging performance.

To better facilitate improved microscopy techniques, it would be advantageous to have a high NA objective lens designed to operate over a broad spectral range including the MIR. An achromatic, refractive high numerical aperture lens designed for an ultrabroad spectral range currently does not exist. When microscopic focusing of light in the NIR to MIR range is required, reflective objective lenses are the only commercial solution. The highest NA lenses are the Schwarzschild-Cassegrain reflector objectives which at best boast a modest 0.5–0.7 NA. Although free of chromatic aberrations, the Cassegrain objective produces quasi-Bessel type beams with a reduced resolution along the optical axis due to the obscuration of the center portion of the beam. In this work, we have selected a reflective objective with a relatively small obscuration area, yet the axial resolution is still reduced by ~15% relative to a refractive objective of the same NA. This makes the use of reflective objectives less attractive for depth-resolved nonlinear optical imaging. The availability of an ultrabroadband refractive, catadioptric, or folded geometry reflective objective would be a breakthrough for the use of MIR excitation in NLO microscopy as it would enable signal generation with an IR vibrational molecular contrast and a high 3D resolution over a larger field of view.

VI. CONCLUSION

In this work, we have presented a cost-effective solution to ultrabroadband focusing of light based on CaF$_2$:sapphire achromatic lenses. We have used these broadband achromatic lenses as an optical relay for a beam-scanning, vibrationally sensitive SFG microscope, optimized for color correction extending from the NIR to the MIR. With minimal changes to the optical design, achromatic lenses of this type could be readily manufactured with color correction extending into the visible and even the UV. The optical components are compatible with standard microscopes and optical systems for simple integration. The optical tools described herein thus enable new applications including selective MIR-heating for
fabrication purposes with a visible source as a guide and targeting the IR-active vibrational modes of biologically relevant molecules. Higher-order lens designs, i.e., apochromatic triplets, would further increase the image quality. Future work may consider other optical substrate combinations to extend the operational wavelengths further into the MIR, targeting the fingerprint region between 5 and 10 μm.

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REFERENCES

1. M. S. Vitiello, G. Scalari, B. Williams, and P. D. Natale, Opt. Express 23, 5167 (2015).
2. T. Steinle, F. Neubrech, A. Steinmann, X. Yin, and H. Giessen, Opt. Express 23, 011105 (2015).
3. Y. Yu, X. Gai, P. Ma, D.-Y. Choi, Z. Yang, R. Wang, S. Debbarma, S. J. Madden, and B. Luther-Davies, Laser Photonics Rev. 8, 792 (2014).
4. D. Zhang, C. Li, C. Zhang, M. N. Slipchenko, G. Eakins, and J.-X. Cheng, Sci. Adv. 2, e1600521 (2016).
5. S. Bai, D. Zhang, C. Li, C. Liu, and J.-X. Cheng, J. Phys. Chem. B 121, 010249 (2017).
6. V. Raghunathan, Y. Han, O. Korth, N.-H. Ge, and E. O. Potma, Opt. Lett. 36, 3891 (2011).
7. A. Hanninen, M. Wat Shu, and E. O. Potma, Biomed. Opt. Express 8, 4230 (2017).
8. A. P. Wood, Appl. Opt. 31, 2253 (1992).
9. D. Jeong, J. H. Lee, H. Jeong, C. Min Ok, and H.-W. Park, Curr. Opt. Photonics 2(3), 241–249 (2018).
10. A. Mann and Society of Photo-Optical Instrumentation Engineers, Infrared Optics and Zoom Lenses (SPIE, 2009), p. 164.
11. T. Yang, J. Zhu, W. Hou, and G. Jin, Opt. Express 22, 9193 (2014).
12. T. A. Palmer, C. C. Alexay, and S. Vogel, Proc. SPIE 8012, 801223 (2011).
13. Y.-S. Jung, Y.-S. Ryu, Y.-J. Kim, J. Kim, S.-K. Youn, K.-W. Park, and H. B. Lee, J. Opt. Soc. Korea 15, 174 (2011).
14. P. N. Robb, Appl. Opt. 17, 2677 (1978).
15. E. S. Lee, S.-W. Lee, J. Hsu, and E. O. Potma, Biomed. Opt. Express 5, 2125 (2014).
16. R. L. Sinclair, M. High, P. Panchhi, and J. A. Dobrowolski, Proc. SPIE 3061, 376 (1997).
17. D. Ren and J. R. Allington-Smith, Opt. Eng. 38, 537 (1999).
18. D. Gurganus, J. D. Owen, M. A. Davies, B. S. Dutterer, S. Novak, and A. Symmons, in Optical Manufacturing and Testing XII, edited by R. Rascher, R. Williamson, and D. W. Kim (SPIE, 2018), p. 28.
19. R. Mittal, M. Balu, P. Wilder-Smith, and E. O. Potma, Biomed. Opt. Express 4, 2196 (2013).
20. A. M. Hanninen, R. C. Prince, R. Ramos, M. V. Plikus, and E. O. Potma, Biomed. Opt. Express 9, 4807 (2018).
21. A. Hanninen, R. Prince, and E. Potma, IEEE J. Sel. Top. Quantum Electron. 25, 6800411 (2019).
22. I. Rocha-Mendoza, D. R. Yankelevich, M. Wang, K. M. Reiser, C. W. Frank, and A. Knoserson, Biophys. J. 93, 4433 (2007).
23. Y. Han, J. Hsu, N.-H. Ge, and E. O. Potma, J. Phys. Chem. B 119, 3356 (2015).
24. C. Pasquini, I. Braz. Chem. Soc. 14, 198 (2003).
25. M.-C. Chen, P. Arpin, T. Popmintchev, M. Gerrity, B. Zhang, M. E. Seaberg, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. 105, 173901 (2010).
26. A. B. Seddon, Phys. Status Solidi B 250, 1020 (2013).
27. R. M. Balabin and S. V. Smirnov, Talanta 85, 562 (2011).
28. N. G. Horton, K. Wang, D. Kobat, C. G. Clark, F. W. Wise, C. B. Schaffer, and C. Xu, Nat. Photonics 7, 205 (2013).