Laser-heated carbon-nanotube blackbody source for infrared spectroscopy

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ABSTRACT

We report on the demonstration of a laser-heated blackbody source fabricated from vertically aligned carbon nanotubes (VACNTs). This thermal source has potential use for performing micro- and nano- infrared spectroscopies because VACNTs have an extremely high melting point >3000 K, near unity emissivity across the infrared, and are compatible with lithographic microfabrication that can be exploited to maximize etendue of thermal emission.

1.MOTIVATION

Micro-Fourier transform infrared spectroscopy (FTIR) is a fundamental tool in characterizing intrinsic material properties at near-diffraction limited spatial resolution such as metal-insulator transitions[1], heterogeneity[2], or absorption efficiency[3]. As we seek to evaluate smaller specimens, both the spatial resolution and signal to noise ratio (SNR) becomes limited by the light source used. The standard source for mid-infrared spectroscopy for tabletop experiments is the globar, which is a silicon carbide element heated to approximately ~1200 K, which emits as a near-blackbody. The globar is sufficient for spectroscopy on relatively large areas, but as dimensions get smaller, globar radiation becomes impractical because of low irradiance. This is especially true in beyond diffraction-limited techniques such as scattering-type scanning near-field infrared microscopy (S-SNIM)[4], or photothermal induced resonance (PTIR)[5]. Infrared lasers such as CO_2[6], mercury[7], quantum cascade lasers (QCL) [8], difference frequency generation (DFG) [9], or supercontinuum [10] lasers, can cover portions of the infrared with sufficient photon flux. Covering the entire infrared, however, requires multiple laser systems which is impractical from both a cost and setup perspective for most laboratories. There have also been recent developments in ultra-high temperature plasma-based blackbody sources[11–13], although plasma instabilities from convective currents can reduce the short-term output stability, and the use of expensive noble gases makes the operating cost non-negligible.

In this work we describe the development of a blackbody source based on vertically aligned carbon nanotubes (VACNTs). VACNTs have many advantages as a blackbody source, such as near unity emissivity from the visible through far-infrared (100 µm), high melting point (exceeding 3000 K), and various growth platforms[14,15]. This source could potentially fill the temperature gap between the globar (~ 1200 K) and plasma (> 8000 K) blackbodies and could be ideal for small area spectroscopy at a relatively low cost.

2.EXPERIMENTAL SETUP

The VACNT-based microbolometer is an ideal platform for our prototype VACNT blackbody device because it is designed to absorb light efficiently and dissipate heat slowly to the environment by a weak thermal (strong physical) link. [16,17] Electrical heating by means of the bolometer's thermistor could lead to delamination or melting of the thermistor at high temperature. Laser-based heating, however, is practical because VACNTs are a nearly ideal absorber and have extremely high damage threshold. Furthermore, with laser heating we have sufficient optical power to heat a

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small area and our stability depends on the amplitude stability of the laser. We have fiber coupled a 532 nm doubled YAG laser as our heating source as shown in Fig. 1. The output of the fiber passes through a continuously-variable neutral density filter (ND). The ND filter enables control of the laser power incident on our VACNT platform, which can be varied continuously up to approximately 1 W. In operation of our laser-heated source, we have observed that the damage threshold for the VACNTs is beyond 5700 W/cm^2 (1 W of laser power focused down to 150 μm diameter) of heating irradiance. The light then passes through a variable-diameter iris to a pickoff mirror on a flip mount to measure the optical power of the laser. The iris defines the size of the laser spot on the VACNT blackbody area. The laser is then focused through a 50 mm off axis parabolic mirror (OAP) (50.8 mm effective focal length) which has a tapered hole through it. The hole is aligned co-axially with the focus of the mirror. The light passes through a potassium bromide (KBr) window into a vacuum chamber and onto our VACNT blackbody. KBr is transmissive to both the laser and infrared light out to approximately 25 micrometers. We must heat the VACNT blackbody in vacuum otherwise atmospheric interaction would occur at elevated temperatures and degrade the emission performance of the VACNTs. Our VACNT blackbody is heated by the laser and emits radiation which is collected and collimated by the OAP and is then refocused onto a precision pinhole with x-y translation capacity. The pinhole is a spatial filter for the emitted radiation from the VACNT blackbody and any spurious heating laser reflections. The filtering allows us to isolate the area of high irradiance and will have the best reimaging properties. A secondary OAP collimates the light from the pinhole, at which point the radiation can be used by an application. In addition, we have a pilot laser which is aligned to be colinear with the emitted thermal radiation to assist in align further in the beam path. In the future we plan on building an enclosure around the setup to allow for dry-air or nitrogen purging to remove atmospheric absorbance from the emitted radiation.

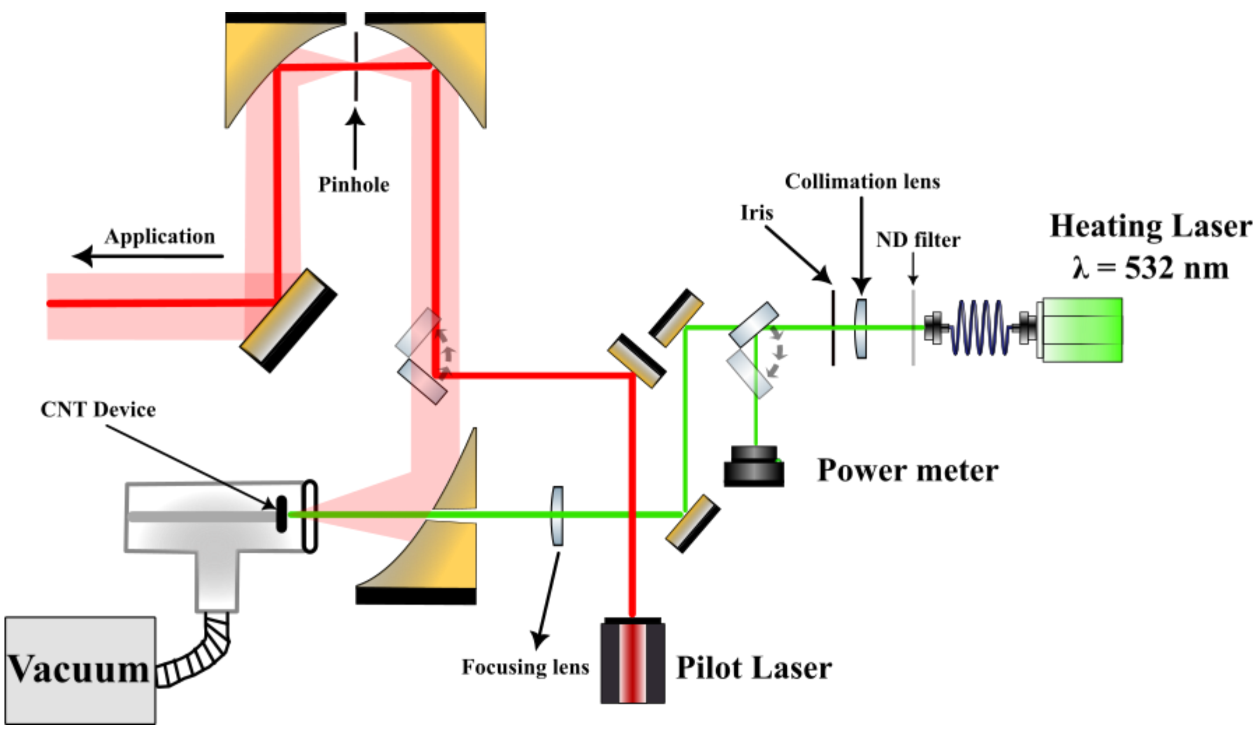

Figure 1: Laser-heated VACNT blackbody setup. Description of beam path for the heating laser, emitted infrared radiation, and pilot laser is detailed in the text.

3.PRELIMINARY RESULTS

We first performed thermal imaging on our blackbody to determine the size of the thermal hotspot from our heating laser. We are defining the hotspot as the focused spot of the heating laser, since this is where the majority of the laser heating is being absorbed. This test was performed at a relatively low laser irradiance (56 W/cm^2), since at higher powers the entire device heats, obscuring the size of the hot spot. An image of the blackbody while off (no laser heating) and on are displayed in figure 2c. From figure 2c we can identify the size of our hotspot to be approximately 150 μm in diameter. The size of this hotspot is important in determining our emission beam divergence and ultimately its reimaging properties. Next we tested our device up to our maximum laser irradiance of 5700 W/cm^2. Our irradiance is calculated by first measuring the incident power and then dividing by the area of our hotspot. This is an approximate irradiance since some of the incident laser power will fall outside of the hotspot. The blackbody under these conditions is displayed in figure 2a and 2b. In figure 2b, the heating laser has been filtered from view via a bandpass filter, and we can clearly see an incandescent hotspot. We used a handheld pyrometer to approximate the temperature of the hotspot under this laser heating condition. We estimate the hotspot temperature to be 3000 K with an uncertainty of 400 K. Our relatively large uncertainty is due to the small area of our thermal emission versus the filament image in the disappearing-filament pyrometer. To quantify the temperature more accurately, we plan to perform NIR grating spectroscopy to assess the peak irradiance wavelength to estimate the blackbody temperature. A clear damage threshold for our VACNT device has not been determined because to do so we need to increase the power of our heating laser beyond its current capacity of approximately 1 W. To increase our available power we will need to upgrade to a higher power fiber. We would like to identify the damage threshold of our VACNT blackbody so that we can pinpoint further improvements to our design and possibly identify the melting point of our VACNTs. Previous studies have suggested a lower bound on the melting point of CNTs at 3400 K[15], and a theoretical maximum at approximately 4000 K[18]. It would be interesting to experimentally determine the melting point of our VACNTs relative to these previous studies. Even at our current estimated temperature this source can provide significantly more flux than refractory sources such as silicon carbide or tungsten. This is because they cannot reach these temperatures without damage, and in the case of tungsten, low emissivity in the mid-infrared[19]. Having established a spot size and approximate temperature we sent our source radiation through an FTIR and onto a mercury cadmium telluride detector (MCT) which can detect between 2 μm – 18 μm. The infrared spectra are plotted in figure 2d for various laser heating irradiances. We did not increase past 2829 W/cm2 of laser heating irradiance since our detector began saturating. At this laser heating level we are far below our peak estimated temperature of ~3000 K, nonetheless these spectra provide qualitative details on our emission performance. One thing to note is the relatively low intensity at the shorter infrared wavelengths (~ 2 μm). We think this is related to a number of factors. For one, the response of our MCT detector has a peak responsivity at 16.5 μm wavelength. In addition, since our emitted radiation is transmitting through multiple windows, the focus position is different for the short and long wavelength regimes. By tuning the position of our detector (closer, or further from the focus position), we can increase the intensity at shorter wavelengths while decreasing the intensity at longer wavelengths and vice versa. Nonetheless it is impressive that our detector can be saturated from predominantly radiation at wavelengths greater then 6 μm considering the peak wavelength for our blackbody is firmly in the near-infrared. Our long wavelength cutoff is coming from the ZnSe window on our MCT detector. The next step is to try re-imaging our source to see the spatial resolution we can achieve and the flux that can be realized. Based on our source size, with reasonably fast focusing optics we expect excellent imaging properties.

There are a number of improvements that could be made to the current device. Since we are only interested in collecting emitted radiation form a small area, we could reduce the size of our VACNT blackbody so that the thermal energy is confined to a smaller area. In our current design, large amounts of optical power (~ 1 W) are required to heat our sample, but with a smaller design we expect less laser power would be required to reach the desired temperatures. As an alternative to reducing the area of our VACNT, we could also potentially use a VACNT microbolometer design with a lower thermal conductance (smaller thermal links or less of them) to retain more heat in the VACNTs and increase the energy efficiency to heat. To boost flux, we could improve our collection optics. Our collection optics could be converted to an on-axis three-mirror anastigmat[20] or Schwarzschild objective[21] that resides inside of the vacuum chamber. This would boost the numerical aperture of collection and increase the photon flux. These higher numerical aperture optics would also reduce the size of our hotspot (since the heating laser would be focused by the collection optics in this setup), which would further improve our ability to reimage the hotpot. To increase our spectral range, we could switch to a diamond window as opposed to KBr. Diamond is transmissive into the far-infrared with approximately

70 % transmission and does not degrade from atmospheric absorption like KBr. We could also explore operating the system in a gas purge as opposed to vacuum to remove the window entirely. Since the VACNT blackbody is small, we could also try to reduce the volume of the vacuum chamber substantially to miniaturize the source.

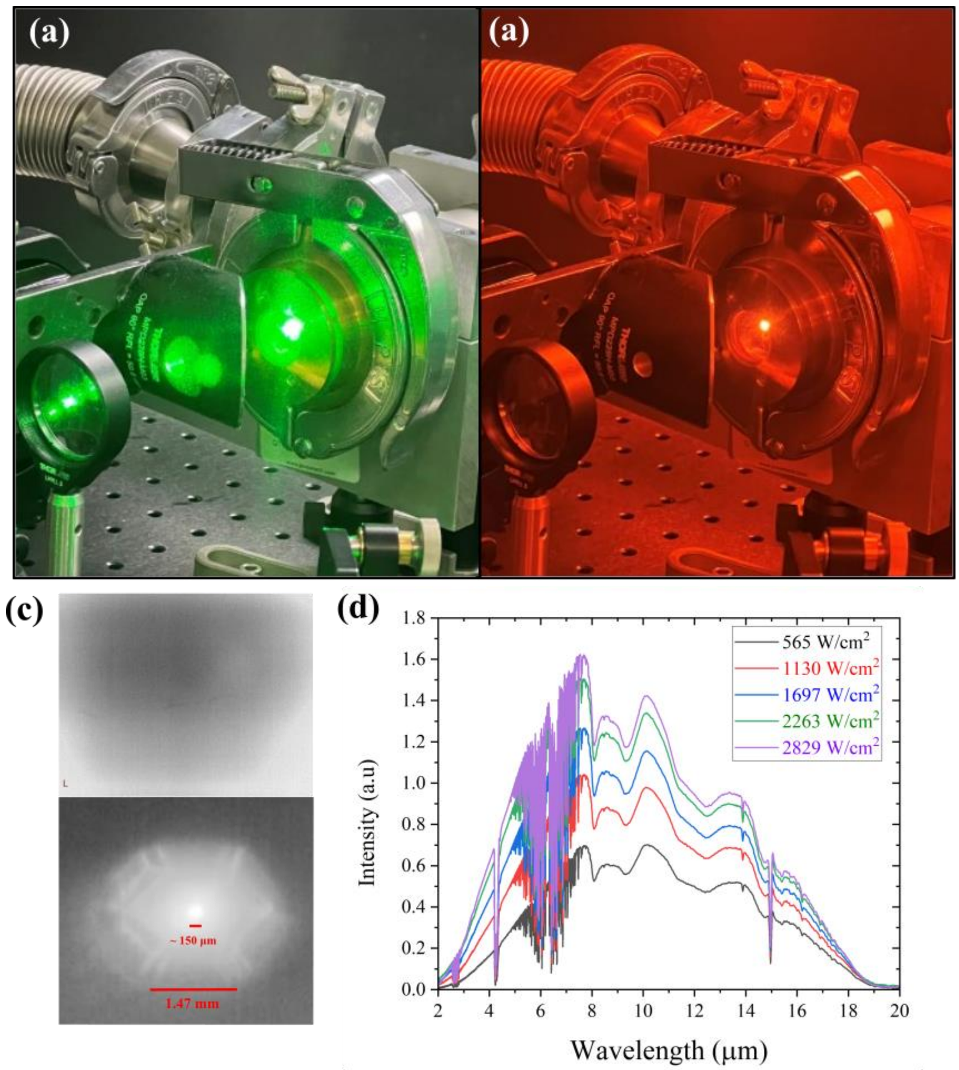

Figure 2: (a) Laser-heated VACNT blackbody in operation. (b) Same as (a), but with heating laser filtered via bandpass filter. The bright spot is incandescent emission from the VACNT area. (c) Thermal images of source while off (top), and on (bottom) heated with 56 W/cm^2 incident irradiance. At low irradiance we can estimate the size of the hotspot to be ~ 150 µm. At elevated laser irradiance, the thermal hotspot is difficult to discern since the entire device heats up and obscures the central hotspot. (d) Infrared spectra taken between 2 µm - 20 µm with varying incident laser irradiance (in steps of 100 mW incident power). Radiation is cut off at ~20 µm is due to the ZnSe window on our mercury cadmium telluride detector and dropping responsivity.

4.CONCLUSION

We have built and demonstrated the use of a laser heated VACNTs as a blackbody source up to a temperature of ~ 3000 K with a source point size of approximately 150 µm diameter. Our source is a VACNT microbolometer detector mounted inside of a vacuum chamber. A high-powered laser is focused onto our VACNT blackbody which emits as it is heated. The near-unity emissivity of VACNTs, high operating temperature and small source point are all ideal properties for infrared spectroscopy of small area materials or devices. Future improvements include redesigning our collection optics, using a diamond window to increase our spectral range and reducing the size of our device to increase the energy efficiency of operation. We hope this device stimulates future work on VACNT type blackbody sources for potential use in infrared micro- and nano- spectroscopy.

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REFERENCES

1. P. McArdle, F.-T. Huang, J. Yang, M.-W. Chu, S.-W. Cheong, and M. M. Qazilbash, "Orbital-selective metallicity in the valence-bond liquid phase of Li 2 RuO 3," Phys. Rev. B **105**(24), 245148 (2022).
2. Q. Wang, P. McArdle, S. L. Wang, R. L. Wilmington, Z. Xing, A. Greenwood, M. L. Cotten, M. M. Qazilbash, and H. C. Schniepp, "Protein secondary structure in spider silk nanofibrils," Nat Commun **13**(1), 4329 (2022).
3. P. McArdle, C. Yung, N. Tomlin, J. Lehman, and M. Stephens, "Micro-DRIFTS for small area hyper-black spectroscopy," Optics Express **31**(26), (2023).
4. X. Chen, D. Hu, R. Mescall, G. You, D. N. Basov, Q. Dai, and M. Liu, "Modern Scattering-Type Scanning Near-Field Optical Microscopy for Advanced MAterial Research," Advanced Materials **2019**(31), 1804774 (2019).
5. A. M. Katzenmeyer, V. Aksyuk, and A. Centrone, "Nanoscale Infrared Spectroscopy: Improving the Spectral Range of the Photothermal Induced Resonance Technique," Anal. Chem. **85**(4), 1972–1979 (2013).
6. N. Sobolev, N. and V. V. Sokovikov, "CO2 Lasers," Soviet Physics Uspekhi **10**(2), 153 (1967).
7. D. R. Smith, R. L. Morgan, and E. V. Loewenstein, "Comparison of the Radiance of Far-Infrared Sources," J. Opt. Soc. Am. **58**(3), 433 (1968).
8. M. A. Belkin and F. Capasso, "New frontiers in quantum cascade lasers: high performance room temperature terahertz sources," Phys. Scr. **90**(11), 118002 (2015).
9. K. Fradkin-Kashi, A. Arie, P. Urenski, and G. Rosenman, "Mid-infrared difference–frequency generation in periodically poled KTiOAsO_4 and application to gas sensing," Opt. Lett. **25**(10), 743 (2000).
10. G. Granger, M. Bailly, H. Delahaye, C. Jimenez, I. Tiliouine, Y. Leventoux, J.-C. Orlianges, V. Couderc, B. Gérard, R. Becheker, S. Idlahcen, T. Godin, A. Hideur, A. Grisard, E. Lallier, and S. Février, "GaAs-chip-based mid-infrared supercontinuum generation," Light Sci Appl **12**(1), 252 (2023).
11. D. J. Lahneman, T. J. Huffman, P. Xu, S. L. Wang, T. Grogan, and M. M. Qazilbash, "Broadband near-field infrared spectroscopy with a high temperature plasma light source," Opt. Express **25**(17), 20421 (2017).
12. M. Wagner, D. S. Jakob, S. Horne, H. Mittel, S. Osechinskiy, C. Phillips, G. C. Walker, C. Su, and X. G. Xu, "Ultrabroadband Nanospectroscopy with a Laser-Driven Plasma Source," ACS Photonics **5**(4), 1467–1475 (2018).
13. D. J. Lahneman and M. M. Qazilbash, "Hyperspectral infrared imaging of surface phonon-polaritons in SrTiO 3," Phys. Rev. B **104**(23), 235433 (2021).
14. J. Lehman, C. Yung, N. Tomlin, D. Conklin, and M. Stephens, "Carbon nanotube-based black coatings," Applied Physics Reviews **5**(1), 011103 (2018).
15. X. Wei, M.-S. Wang, Y. Bando, and D. Golberg, "Thermal stability of carbon nanotubes probed by anchored tungsten nanoparticles," Science and Technology of Advanced Materials **12**(4), 044605 (2011).

16. A. Vaskuri, M. S. Stephens, N. A. Tomlin, C. S. Yung, A. J. Walowitz, C. J. Straatsma, D. M. Harber, and J. H. Lehman, "Microfabricated bolometer based on a vertically aligned carbon nanotube absorber," in *Synthesis and Photonics of Nanoscale Materials XVII*, A. V. Kabashin, J. J. Dubowski, and D. B. Geohegan, eds. (SPIE, 2020), p. 21.
17. C. Yung, N. Tomlin, C. Straatsma, J. Rutkowski, E. Richard, D. Harber, J. Lehman, and M. Stephens, "BABAR: black array of broadband absolute radiometers for far infrared sensing," in *Image Sensing Technologies: Materials, Devices, Systems, and Applications VI*, N. K. Dhar, A. K. Dutta, and S. R. Babu, eds. (SPIE, 2019), p. 14.
18. Y. Miyamoto, S. Berber, M. Yoon, A. Rubio, and D. Tománek, "Onset of nanotube decay under extreme thermal and electronic excitations," Physica B: Condensed Matter **323**(1–4), 78–85 (2002).
19. D. Chester, P. Bermel, John. D. Joannopoulos, M. Soljacic, and I. Celanovic, "Design and Global Optimization of High-efficincy Solar Thermal Systems with Tungsten Cermets," Optics Express **19**(S3), A245 (2011).
20. R. N. Wilson, *Reflecting Telescope Optics I*, 2nd ed., Astronomy and Astrophysics Library (Spring Berlin, Heidelberg, 2004).
21. M. Riedl, *Optical Design Fundamentals for Infrared Systems*, 2nd ed. (SPIE Press Book, 2001).