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# Four-wave mixing approach to *in-situ* detection of nanoparticles

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We report on the development and experimental validation of a laser-based technique which uses coherent Rayleigh-Brillouin scattering (CRBS) to detect nanoparticles with characteristic sizes ranging from the atomic scale to tens of nanometers. This technique is aimed (non-exclusively) for the detection of nanoparticles produced by volumetric nanoparticle synthesis methods. Using CRBS, carbon nanoparticles of dimensions less than 10 nm and concentrations of  $10^{10} \text{ cm}^{-3}$  are detected *in-situ* in a carbon arc discharge with graphite electrodes. This four wave mixing approach should enable advances in understanding of nanoparticle growth that can potentially lead to improved modeling of the growth mechanisms, and thus, to improve synthesis selectivity of nanoparticles and yield.

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## I. INTRODUCTION

Volumetric nanoparticle synthesis methods, such as flames[1, 2], arc discharges[3–5] and laser ablation[6] have shown great potential for production of industrial-scale quantities of various types of nanoparticles[7, 8] with different structures, such as: fullerenes[6], carbon[9] and boron nitride nanotubes[7, 10], nanowires[11], etc. Optimization of these techniques requires a better understanding and control of the nanoparticle synthesis process to increase nanoparticle selectivity and produce nanostructures with desired structural characteristics and properties. For typical volumetric synthesis conditions, there are no conventional *in-situ* optical diagnostic methods capable of resolving sub-nanometer and nanometer scale nanoparticles.

Due to the harsh environment in which nanoparticle nucleation and growth usually occur (e.g. high temperature), detection is usually done via optical techniques, which are less perturbing than other techniques such as mechanical extractors[12]. A number of diagnostic tools are employed to map the nanoparticle formation process from the precursor atoms and molecules to the end product. Species identification and measurements of temperatures and relative atomic and molecular densities can be obtained from optical emission spectroscopy (OES) in which collisionally excited light emitted from the nanoparticle synthesis region is spectroscopically resolved[13, 14]. Laser induced fluorescence provides quantitative measurements of atomic and molecular densities[15]. The sizes of nanoparticles of dimensions larger than 20 nm can be measured via laser induced incandescence (LII) in which particles are heated by a laser beam and the decay time of the resulting incan-

descence is measured and modeled[16]. The intermediate regime between initial nucleation and nanoparticle growth particles with sizes of 10-20 nm is particularly challenging for nanoparticle detection. The only optical technique previously applied in this regime is Raman spectroscopy[17]. However, Raman spectroscopy only probes nanoparticle structure and does not provide nanoparticle dimensional information. Nanoparticles in this regime have been detected using x-ray radiation from synchrotron sources to perform small-angle x-ray scattering (SAXS)[18–20]. Although successful, this technique requires access to a collimated synchrotron light source, which can be costly and infrequent. Thus, nanomaterials research would benefit from a nanoparticle detection technique based on a table top laser system. In this paper, we demonstrate the first use of Coherent Rayleigh-Brillouin Scattering (CRBS), a demonstrated laser-based gas measurement technique[21, 22], for *in-situ* volumetric detection of nanoparticles in an arc discharge.

## II. COHERENT RAYLEIGH-BRILLOUIN SCATTERING: AN OVERVIEW

CRBS is a non-resonant four-wave mixing process, which relies on inducing an electrostrictive grating through the interference of two intense laser beams, termed the *pump* beams, within a medium. The pump beams have a wavelength of  $\lambda_{pump}$  and identical polarization (Figure 1). By varying the frequency difference  $\Delta f$  of the pump beams, an optical lattice moving at a phase velocity given by  $v_g = (\Delta f \cdot \lambda_{pump}) / (2 \cdot \sin(\phi/2))$  is created. Individual particles that have a translational velocity equal or close to  $v_g$  feel an attraction towards the high intensity regions of the interference pattern due to the optical dipole force[23]. Thus, the density modulation induced in the medium effectively probes the velocity distribution function of the medium, as  $\Delta f$  is scanned.

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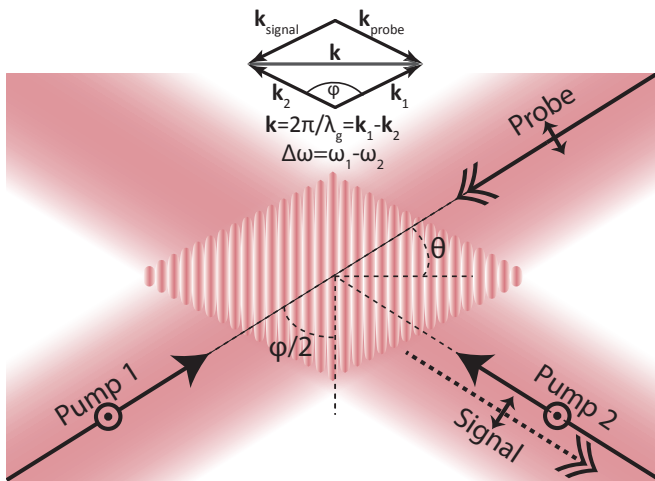


FIG. 1. Typical planar CRBS geometry. Two nearly counter-propagating pump beams of the same polarization interfere within a medium to create an optical lattice. A probe beam with polarization normal to that of the pumps, is incident on this lattice at the Bragg angle to give the fourth beam, the signal beam

The wavelength  $\lambda_g$  of the resulting optical lattice is determined by the crossing half-angle  $\phi/2$  of the pump beams through the relation  $\lambda_g = \lambda_{pump}/(2 \cdot \sin(\phi/2))$  (Figure 1).

A third beam, called the probe, with wavelength  $\lambda_{probe}$  and polarization normal to that of the pumps, is incident upon the optical lattice at an angle  $\theta$  that fulfills the first order Bragg condition  $\lambda_{probe} = \lambda_g \cdot \sin(\theta)$ [24]. A fourth beam, termed the *signal*, is generated through the interaction of the probe beam with the optical lattice. A CRBS spectrum is obtained by measuring the intensity of the signal beam versus the optical lattice velocity. The CRBS signal intensity  $I_S$  is proportional to the square of the induced refractive index modulation:  $I_S \propto L^2(\Delta N)^2 I_1 I_2 I_{pr} \propto \Delta n^2$ , where  $I_1$  and  $I_2$  are the pump intensities,  $L$  is the length of the interaction region,  $I_{pr}$  is the probe intensity and  $\Delta n = \frac{\alpha_{eff} \Delta N}{2\epsilon_0}$  where  $\alpha_{eff}$  is the effective polarizability of the particles (atoms or molecules) and  $\Delta N$  is the induced periodic density modulation[22, 25]. In comparison to spontaneous Rayleigh scattering in which scattering occurs into a  $4\pi$  solid angle, the CRBS signal is a laser beam with the characteristics of the probe beam. CRBS has proven to be a powerful technique for the determination of the temperature, speed of sound, pressure, polarizability, shear, and bulk viscosity of a gas or gas mixture[25–29].

The size of the particles that can be detected by CRBS is determined by the Rayleigh criterion, which defines the maximum particle dimension that can be observed as  $\lambda/10$ , where  $\lambda$  is the detection wavelength. CRBS has never before been applied to gas-nanoparticle mixtures. Nanoparticle detection with CRBS was initially suggested in reference 30 which examined the feasibility of detecting  $C_{60}$  in an atmospheric pressure Argon background. The resulting CRBS line shape is dominated

by the Rayleigh peak since phonons cannot be launched in nanoparticle ensembles at such low densities. This detection technique preferentially measures the smallest nanoparticles in the measurement region, since the Rayleigh peaks of heavier particles would be masked by the Rayleigh peaks of lighter particles. For an ensemble of nanoparticles with known or estimated temperature, the mass of the detected nanoparticles can be derived from the width of the Rayleigh peak. By calibrating the CRBS signal intensities in known conditions (e.g. atmospheric pressure air), reliable estimates of the nanoparticle densities can also be obtained from the Rayleigh peak amplitude[25].

### III. CRBS EXPERIMENTAL SETUP

CRBS has been applied to in-situ detection of carbon nanoparticles produced by an arc discharge. The DC arc operates in the reactor chamber between two 6 mm diameter graphite electrodes with an inter-electrode gap of  $\sim 4$  mm in a helium atmosphere of 500 Torr. This setup is similar to the one used for the initial demonstration of production of carbon fullerenes[31] and nanotubes[32]. The arc current ranges from 40 to 60 A; the arc voltage is maintained at  $\sim 25$  V. The graphite anode ablates during arc operation and evaporated carbon products deposit on the cathode and chamber walls[4].

The CRBS system used in this study is described in detail elsewhere[25, 33]. The two pump laser beams and the probe beam have a wavelength of  $\lambda_{pump} = \lambda_{probe} = 1064$  nm, a pulse duration of  $\sim 150$  ns and can achieve energies of up to  $\sim 250$  mJ/pulse/beam. The system utilizes the single-shot chirped lattice approach to obtain the CRBS spectrum, with chirp rates as high as  $\Delta f = 1.5$  GHz over the pulse duration[34]. In this approach, all of the velocity components required to obtain a CRBS spectrum are present during a single laser pulse. This is important since nanoparticles are expected to have a residence time of  $\sim 1$  ms[35] in the CRBS interaction region which typically has a spatial extent of  $\sim 150$   $\mu$ m. The arc typically operates for  $\sim 2$  minutes (limited by vacuum chamber heating). Thus, fast spectral acquisition times are required, which non-chirped CRBS configurations with acquisition times on the order of 5-10 minutes, cannot achieve. The CRBS beams cross at a distance of  $\sim 13$  mm from the arc core where nanoparticles are expected to form (Figure 2)[12, 35]. Helium is not expected to contribute to the CRBS signal due to its very small polarizability[25]. The typical planar CRBS geometry (Figure 1) is not optimal for nanoparticle detection in an arc discharge because the pump beam counter-propagating the signal beam is Rayleigh and Mie scattered from nanoparticles in its path[21, 22, 25, 27, 29, 34]. The pump beam is also scattered and depolarized, contributing a large background signal to the measurements. To overcome this, a folded geometry with the probe incident on the lattice from a plane normal to the plane of

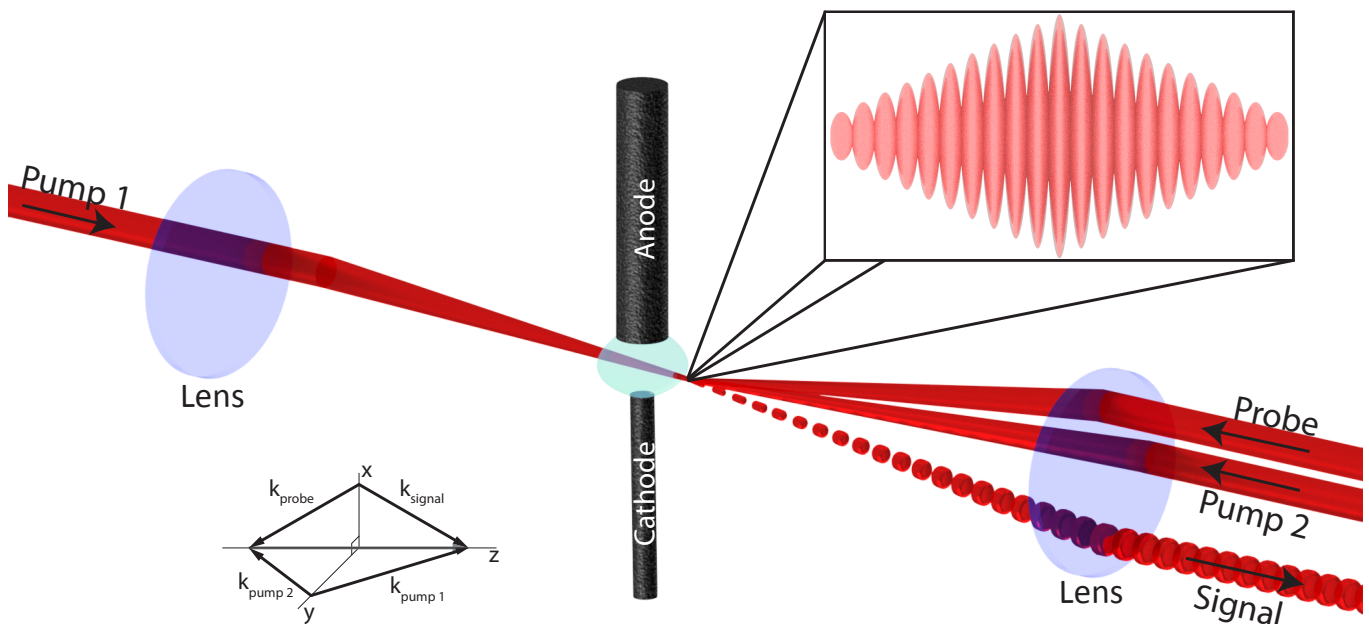


FIG. 2. The folded CRBS geometry used in the experiment described in this paper. Two counter-propagating, co-planar and equipolarized pump beams are focused by two  $f = 500$  mm lenses to interfere at a distance of 13 mm from the arc core, formed between two graphite electrodes when a current of 50-60 A runs through them. For a CRBS signal beam to be produced, a probe beam, with its polarization normal to that of the pumps, must be incident on the induced optical lattice at the Bragg angle. In order for the background noise from the pumps to be minimized in the line of sight of the resulting CRBS signal beam, the probe is incident on the optical lattice from the plane perpendicular to that of the pumps. It is also important for the interaction region to be formed on the side from which the probe enters, so that the signal does not pass through the arc core and weaken due to scattering and absorption.

propagation of the pumps was used, as shown in Figure 2. This ensures that there is minimal unwanted scattering in the line of sight of the signal beam. Another way to overcome this would be to use a probe of different wavelength and to use spectral filtering to eliminate unwanted scattering of the pumps.

#### IV. IN-SITU DETECTION OF NANOPARTICLES WITH CRBS

In gases, a single-shot CRBS spectrum can be obtained using an optical lattice with the zero velocity component in the temporal centre of the pulse with chirping to non-zero velocities toward the beginning and end of the pulse[25, 34]. In the case of carbon nanoparticle measurements, the laser intensities required to obtain a CRBS signal ( $10^{14}$  Wm $^{-2}$  per beam) are sufficient to destroy the nanoparticles, since carbon nanoparticles have an absorption maximum close to the wavelength used[36]. For other nanoparticles, such as boron nitride nanoparticles, this effect would be less important since the absorption maximum is in the ultraviolet region. This effect is clearly demonstrated in Figure 3, which shows the CRBS signal (red lines) for six different applied optical lattice velocity ranges (black lines). If the nanoparticles were not destroyed, the resulting CRBS spectrum would be reproduced regardless of the temporal location of the zero velocity component during the pulse. In our measure-

ments only the initial part of the pulse produces a useful CRBS signal since the nanoparticles have been destroyed due to laser evaporation by times later in the pulse. This effect has also been observed in studies in which x-rays have been used to probe similar structures[37, 38] and is known as *diffraction-before-destruction*. For CRBS we refer to this phenomenon as *scattering-before-destruction*.

Although the folded geometry employed in our experiments minimizes the background signal, some background signal is observed, as seen in Figure 3. The CRBS signal sits on top of this background, which is due to Rayleigh and Mie scattering of the pumps from nanoparticles in the vicinity of the arc. Since there is no useful information in the signal at end of the pulse, we measure the ratio of the signal in the initial part of the pulse, which is the time interval before the nanoparticles are destroyed and constitutes the scattering-before-destruction CRBS signal, to the signal in the final part of the pulse, which is simply the background level. This is done for a series of scanned velocity ranges, such as those shown in Figure 3, to reconstruct the nanoparticle Rayleigh peak profile. This approach assumes a uniform particle size distribution and a uniform density per shot for the 10 Hz laser pulse repetition rate.

The ratio of CRBS signal to background signal plotted versus the lattice phase velocity at the beginning of the laser pulse is shown in Figure 4 (black dots). This plot represents the CRBS spectrum, which has a maximum value at zero lattice phase velocity. The measured

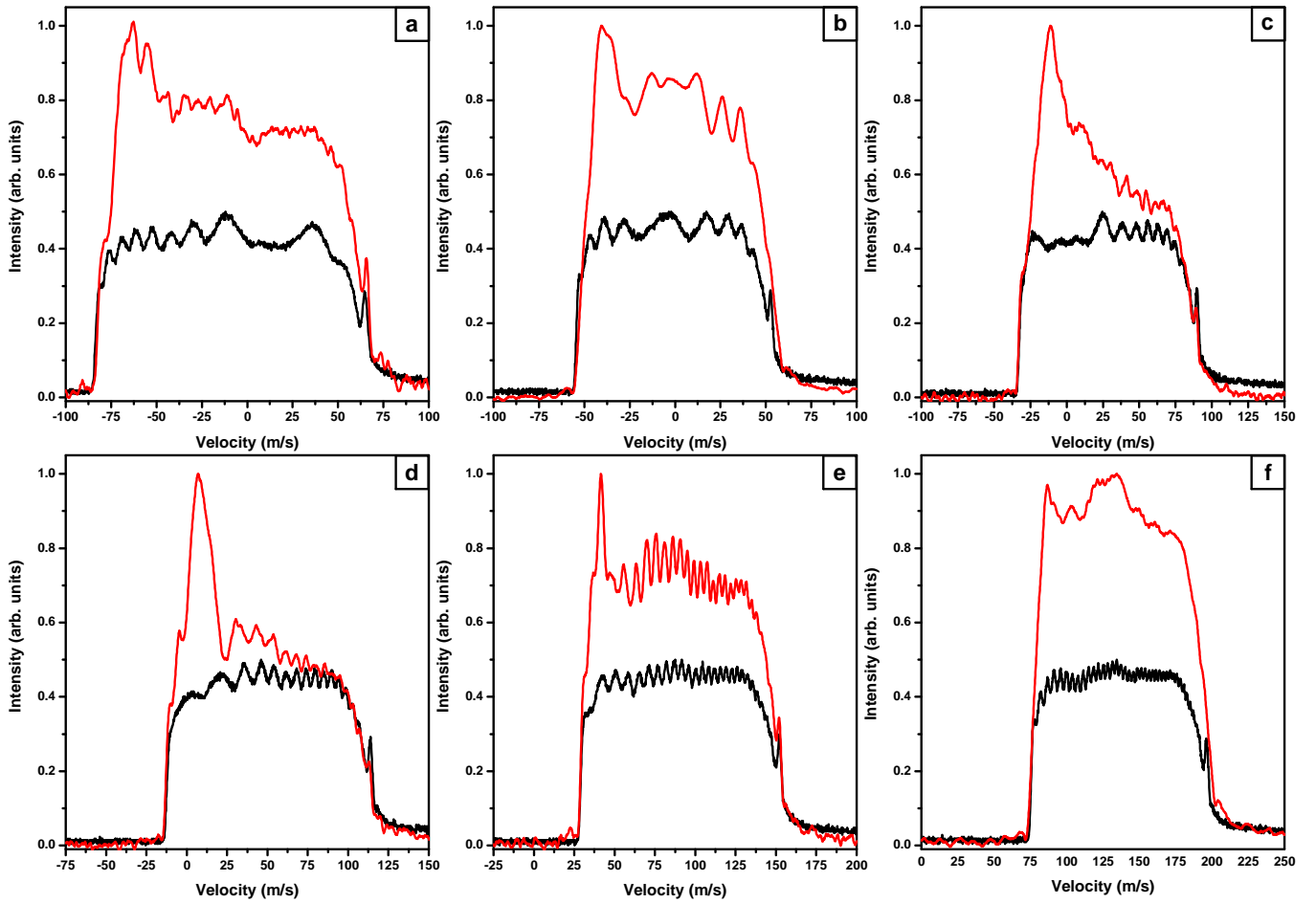


FIG. 3. CRBS signals from the arc (red lines) for different chirp rates, recorded by beating the two pump beams on a fast photodiode (black lines)

data points were fitted to reconstruct the Rayleigh peak, given the assumptions mentioned earlier (dashed line). For an estimated temperature  $T$  of 1500 K at the measurement location [35, 39], the mass  $m$  of the nanoparticles is calculated from  $e^{-\frac{mv^2}{2kT}}$ , where  $v$  is the velocity and  $k$  is Boltzmann's constant (red line). From this fit, the nanoparticle mass is estimated to be  $\sim 500,000$  atomic mass units (a.m.u.). For comparison the Rayleigh peak was also calculated for  $C_{60}$ , which has a mass of 720 amu and a diameter of  $7 \text{ \AA}$  (orange line). Assuming that the density of the nanoparticles is the same as for  $C_{60}$  ( $1.72 \text{ g/cm}^3$ ) and have a mass that is  $\sim 700$  times larger, then the measured nanoparticles have dimensions of  $\sim 6$  nm. This result assumes hollow nanoparticles, such as nanotubes or fullerenes. If the density equal to that of solid graphite (e.g. soot) is assumed ( $2.266 \text{ g/cm}^3$ ), the particles have dimensions of  $\sim 5$  nm.

It is important to note that this result represents a lower size detection limit since heavier, larger particles could also contribute to the observed CRBS signal. Since their Rayleigh peak width is smaller, their presence is masked by the Rayleigh peak of the lighter particles. It is

also important to note that since the laser fluences used are  $\sim 10^{14} \text{ Wm}^{-2}$ , *scattering-before-destruction* would only give a CRBS signal for the initial size of the nanoparticles, as these would immediately be destroyed into their constituents, and not just modified to smaller sized particles. This is also suggested by the simulation presented in Fig. 5. Also, if size modification or alteration would indeed take place, this would be manifested with a much wider Rayleigh peak, which is only mass dependent, for a given temperature.

This result is consistent within an order of magnitude with measurements performed in similar conditions with other methods [12, 16]. This result assumes spherical particles being detected. In order for particle elongation or asymmetry to be detected one would need to monitor the scattered polarization ratio since spherical particles would scatter the same polarization as that of the probe, while elongated particles would scatter only the polarization projection to their elongated axis. Although elongated nanoparticles, such as nanotubes, are randomly oriented in volumetric synthesis environments, through polarization monitoring we should also be able to identify any potential alignment of these nanoparti-

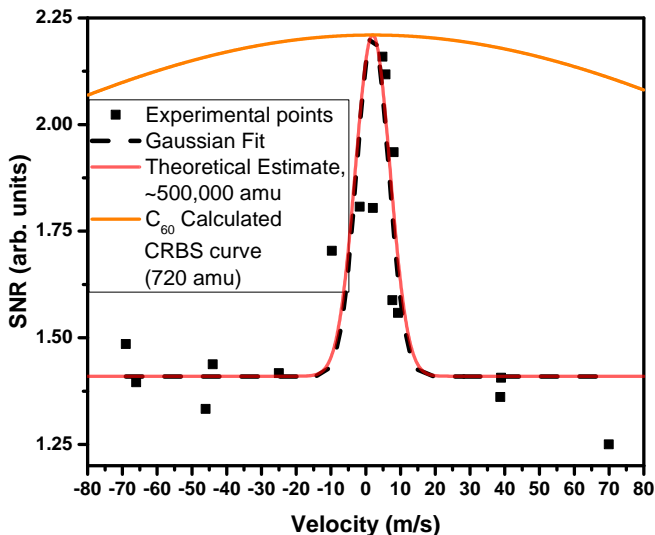


FIG. 4. Reconstructed Rayleigh profile for nanoparticles measured  $\sim 1.3$  cm from the arc core (red line). This estimate is derived from the ratio of the scattering-before-destruction CRBS signal at the beginning of the laser pulse to the background level (black dots). Each dot represents the ratio for the CRBS signal over a single laser shot. Also plotted for comparison is the simulated profile for  $C_{60}$  (orange line).

cles to external fields. This is something we plan to do in the future. Additionally, since clustering is a time-dependent dynamic process or a process occurring in a stream in different spatial areas then, by observing the change in the shape of the CRBS signal, it would also be possible to detect nanoparticle clustering, in combination with the aforementioned simultaneous polarization monitoring. The peak signal level corresponds to that from air at a pressure of 30 Torr which yields a measured nanoparticle density of  $\sim 10^{10}$   $\text{cm}^3$  using our detection system calibration[25]. It has to be pointed out that CRBS in the counter-propagating geometry used here, has got a theoretical detection limit of  $10^8$   $\text{cm}^{-3}$  which arises from the necessity of having at least one occupant per lattice site in order to obtain a coherent signal. Finally, a strong correlation between the observable CRBS signal and arc current is observed: as the arc current decreases, the CRBS signal also decreases while as the arc current increases, the CRBS signal increases. The same effect is also observed as the measurement location changes: closer to the arc center the CRBS signal disappears, as there is no nanoparticle production there.

## V. SCATTERING-BEFORE-DESTRUCTION: THEORETICAL ESTIMATES

To understand the effect of high laser intensities on spherical graphitic nanoparticles, we numerically calculated the time-resolved nanoparticle diameter profiles during and after the laser pulses by solving three coupled ordinary differential equations for the nanoparticles

energy, charge, and mass balance. The appropriate energy balance equation is:

$$\dot{U}_{Int} = Q_{Arc} + Q_{Las} - Q_{Sub} - Q_{Con} - Q_{Rad} - Q_{The}, \quad (1)$$

where  $\dot{U}_{Int}$  is the rate of change of the nanoparticle internal energy [40], and the terms denoted by  $Q$  describe the following heat transfer processes: 1)  $Q_{Arc}$  describes radiative heating from the arc discharge plasma channel [41], 2)  $Q_{Las}$  describes absorption of energy from the laser pulse using the Rayleigh approximation [42, 43] (assuming a complex index of refraction of  $1.57 - 0.56i$  [44]), 3)  $Q_{Sub}$  describes thermal conduction from the evaporative cooling due to laser-induced sublimation [45, 46], 4)  $Q_{Con}$  describes thermal conduction from the nanoparticle surface to the ambient gas in the free-molecular flow regime [40], 5)  $Q_{Rad}$  describes thermal radiation, and 6)  $Q_{The}$  describes thermionic emission from laser-heated nanoparticles [41, 47]. The charge-balance equation considers a volumetric buildup of positive charge in the nanoparticle due to thermionic emission of electrons [47]. The mass balance equation considers mass loss from laser-induced sublimation; for spherical nanoparticles, nanoparticle diameter is related to mass by  $\rho_S \frac{\pi D^3}{6}$ , where  $\rho_S \sim 2$   $\text{g/cm}^3$  is the density of graphite and  $D$  is nanoparticle diameter. The formula used for modeling nanoparticle mass loss is,

$$\frac{dM}{dt} = -\pi D(t)^2 \frac{W(T(t))p_V(T(t))}{R_P T(t)} \sqrt{\frac{R_M T(t)}{2\pi W(T(t))}}, \quad (2)$$

where  $T(t)$  is the nanoparticle's time-resolved temperature profile,  $R_M$  is the ideal gas constant in effective mass units ( $8.3145 \times 10^7$   $\text{g cm}^2/\text{mol K s}^2$ ),  $R_P$  is the ideal gas constant in effective pressure units ( $83.145$   $\text{bar cm}^3/\text{mol K}$ ), and  $W(T(t))$  and  $p_V(T(t))$  are the mean temperature-dependent values of the molecular weight and vapor pressure of the sublimed carbon clusters, respectively. The formulas used for  $W(T(t))$  and  $p_V(T(t))$  [45], were calculated from fits to data published in Leider *et al.* [46].

Numerically solving the energy, charge, and mass balance equations yields the time evolutions of the nanoparticle diameters shown in Figure 5, which illustrate the trend of increased nanoparticle evaporation with increased laser intensity. The data presented in Fig. 5, which are derived from Eqs. 1-2, are meant to qualitatively show that higher laser intensities will destroy carbon nanoparticles with diameters ranging from 5–50 nm. There are certainly uncertainties in the terms used in Eqs. 1-2, but using any reasonable parameters, from e.g. ref. 45, would produce a similar result: laser intensities in excess of  $10^{13}$   $\text{Wm}^{-2}$  are high enough to completely and almost instantaneously destroy carbon nanoparticles.

Figure 5b) shows that laser-induced sublimation would result in a  $\sim 10 - 30$  % reduction in nanoparticle diameters for nanoparticles with different initial diameters

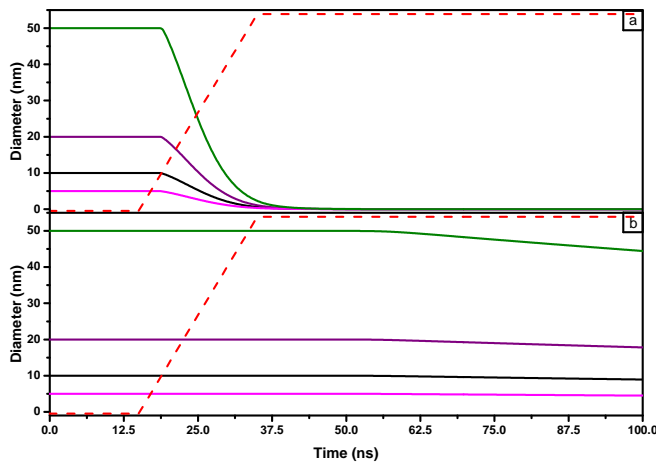


FIG. 5. Simulations of time-resolved nanoparticle diameter profiles (solid lines) following interaction with a laser pulse (dashed line) of a)  $10^{12} \text{ Wm}^{-2}$  and b)  $10^{10} \text{ Wm}^{-2}$ .

in the range 5 – 50 nm assuming a laser intensity of  $10^{10} \text{ Wm}^{-2}$ . However, a higher laser intensity of  $10^{12} \text{ Wm}^{-2}$  results in stronger laser-induced nanoparticle sublimation, and the complete destruction of laser-heated nanoparticles, as shown in Figure 5a). This simplified calculation illustrates the worst case scenario. In reality the laser intensity seen by the nanoparticles can be lower due to scattering and absorption in a process of the passage through a mixture of gas, nanoparticles and soot particles adjacent to the observation area. In addition, as the nanoparticles evaporate, they become surrounded by dense, strongly absorbing cloud, which shields them from further absorption of the incident laser energy. This mechanism would considerably slow down complete nanoparticle evaporation due to absorption.

## VI. CONCLUSIONS

In summary, we report the first in-situ detection of nanoparticles of 5 – 6 nm with concentrations of  $10^{10} \text{ cm}^{-3}$  synthesized in a carbon arc. The results presented here could allow for the in-situ detection and monitoring of volumetric nanoparticle formation and growth using a table top laser system. CRBS could also be applied to detection of nanoparticles in other gas phase synthesis methods (e.g. laser, flame or plasma torch), as long as the density of nanoparticles is above the CRBS detection limit of CRBS for nanoparticles ( $\sim 10^8 \text{ cm}^{-3}$ ). For an estimated temperature at the point of measurement of 1500 K, the average mass of the measured nanoparticles was estimated to be 500,00 a.m.u. Thus, for a known shape of the measured nanoparticles (e.g. from ex-situ evaluation), the average size of nanoparticles can accurately be deduced if the temperature is known from an independent measurement. We plan to do this in the future. We are working on increasing the detection sensitivity of our system to allow detection of nanoparticles without destroying them. This would also allow us to directly obtain a CRBS spectrum in a single laser shot (as is the case in gases), in comparison to the tens of shots that are required with the current setup. The digital data for this paper can be found at <http://arks.princeton.edu/ark:/88435/dsp01x920g025r>.

## VII. ACKNOWLEDGMENTS

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