Enhanced temperature uniformity by tetrahedral laser heating

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(Received 25 February 2004; accepted 18 August 2004; published 29 October 2004)

Temperature profile on a spherical sample that is heated by laser beams in various geometries while processed in vacuum is analyzed. Sample heating by one or four laser beams was considered. An analytical expression was derived for directional sample heating cases. It suggests an enhanced temperature uniformity over the samples when heated with four diffuse laser beams arranged in a tetrahedral geometry. This was experimentally verified by heating a spherical stainless steel sample by laser beams. Both the calculated and experimentally determined temperature variations over the sample suggest that use of diffuse four beams arranged in tetrahedral geometry would be effective in reducing temperature variation to within 1 K. The enhancement in the temperature uniformity for four diffuse beams arranged in a tetrahedral geometry by a factor of 50 over a single focused beam is promising to accurately measure of thermophysical properties. This drastic improvement in temperature uniformity might even enable atomic diffusion measurements in the undercooled liquid states of the bulk glass forming alloys since Marangoni and gravity driven convection will be substantially reduced. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1804351]

I. INTRODUCTION

Usefulness of the high temperature electrostatic levitator and acoustic levitator for measurements of thermophysical properties of molten materials and for studies of solidification processes is being widely recognized in the materials science community.[1–9] Although many of these experiments are temperature sensitive, they are usually performed assuming the sample temperature is represented by a single value, overlooking the temperature variance over the sample. A temperature gradient across a liquid sample tends to create density gradient as well as surface tension gradient that are responsible for inducing convective flows in the sample. Such convective flows will affect measurement results of physical properties, particularly those transport properties such as atomic diffusion, thermal conduction, and viscosity. Convective mixing can completely obliterate the diffusion profile making quantitative analysis difficult if not impossible. In designing experiments to determine transport properties one ought to be especially careful about temperature gradients if the sample is going to be heated by directional heating sources.

In this article we analyze the temperature profile on a spherical sample when it is heated by one or more laser beams in search of the conditions that minimize the temperature variations over the sample. The Laplace equation, subjected to boundary conditions appropriate for a sphere that is heated by laser while it is radiating heat through the surface to the surrounding vacuum, was solved. The analytical results suggests a substantial decrease in temperature variations over the sample when heating with four diffuse laser beams arranged in a tetrahedral geometry. The calculations were verified on a stainless steel ball using various laser beam heating geometries that were set up in a high vacuum chamber.

II. THEORETICAL CONSIDERATION

The magnitude of temperature variations in the spherical droplets under conditions of heating with one or more laser beams (or any other source) can be assessed by solving the steady state (time-independent) Fourier heat flow equation. This equation is subjected to boundary conditions appropriate for a sphere heated by absorption of laser power at the surface and radiating energy from the surface by Stephan–Boltzmann radiation to a surrounding vacuum. We assume that the sample is suspended in a high vacuum so that any heat conduction through gaseous medium or any mechanical contact is ruled out. The radiated power per unit area of surface is given by

$$P_{\mathrm{rad}} = \varepsilon \sigma_{\mathrm{SB}} T_s^4, \tag{1}$$

where T_s is the local surface temperature, ε is the hemispherical total emissivity of the surface, and σ_{SB} the Stefan–Boltzmann constant. The power input is determined by the spectral emissivity of the surface (equal to the spectral absorption coefficient) at the laser wavelength and the incident

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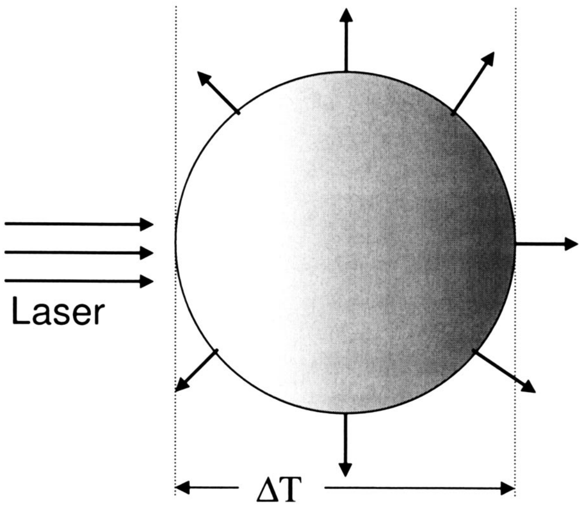

FIG. 1. Schematic illustration of single beam heating by a laser beam and radiation loss by the sphere producing a temperature gradient.

power per unit area of the laser beam projected on the sphere surface. The situation is illustrated in Fig. 1. To obtain the exact steady state temperature distribution in the sphere, one solves the steady state Fourier equation (essentially Laplace's equation) for the boundary condition

$$J_r(R,\theta,\varphi) = -\kappa[1/r(r\partial T/\partial r)]_{r=R} = \varepsilon\sigma_{\mathrm{SB}}T_s^4 - P_{\mathrm{laser}}(\theta,\varphi), \tag{2}$$

where $J_r(R,\theta,\varphi)$ is the net radial component of the heat flux out of the sphere surface, κ is the thermal conductivity of the sample, R is the sample radius, and p_{laser} is the laser power input at the sphere surface, which depends on location (θ,φ). For small temperature gradients compared to the average temperature, $\Delta T/T_{\mathrm{av}} \ll 1$, where T_{av} is the average steady state temperature of the sphere, one can neglect the higher order effect of surface temperature variations on the local radiated power loss and assume that power is radiated isotropically.

The steady state heat flow equation in a sphere is a solution to the Laplace equation and can be solved by expanding temperature T in spherical harmonics Y_{lm}:

$$T(r,\theta,\varphi) = \sum_{l=0}^{\infty}\sum_{m=-l}^{+l} A_{lm}\frac{r^l}{R^l}Y_{lm}(\theta,\varphi) \Rightarrow \nabla^2 T = 0. \tag{3}$$

The coefficients A_{lm} in the expansion are to be determined from the boundary condition that the heat flow at the surface equals the net radiated power density p:

$$\kappa\nabla T\cdot\mathbf{n}|_{\mathrm{surface}} = \left.\kappa\sum_{l=0}^{\infty}\sum_{m=-l}^{+l} A_{lm}\frac{lr^{l-1}}{R^l}Y_{lm}(\theta,\varphi)\right|_{r=R} = p \Rightarrow A_{lm} = \frac{1}{\kappa}\frac{R}{l}\oiint_{4\pi} Y_{lm}^{*}p\,d\Omega. \tag{4}$$

This net radiated power density includes laser heating and radiative cooling. If the sample is nearly uniform in temperature (temperature variations are small compared to the average temperature) the Stefan–Boltzmann equation can be linearized around the average temperature and substituted back into the expression for the coefficients

$$p \approx p_{\mathrm{laser}} + \varepsilon\sigma_{\mathrm{SB}}T_0^4 + 4\varepsilon\sigma_{\mathrm{SB}}T_0^3(T-T_0), \tag{5}$$

$$A_{lm} = \frac{1}{\kappa}\frac{R}{l}\left(\oiint_{4\pi} Y_{lm}^{*}p_{\mathrm{laser}}d\Omega + 4\varepsilon\sigma_{\mathrm{SB}}T_0^3A_{lm}\right) \Rightarrow A_{lm} = \frac{1}{\kappa}\frac{R}{l}\left(1-\frac{1}{\kappa}\frac{R}{l}4\varepsilon\sigma_{\mathrm{SB}}T_0^3\right)^{-1}\oiint_{4\pi} Y_{lm}^{*}p_{\mathrm{laser}}d\Omega. \tag{6}$$

The correction term for the nonuniformity in temperature is smaller than 1% in the temperature region of interest, so the radiative cooling term is unimportant in all but the zero order spherical harmonic, which corresponds to the average temperature. In other words, we can, to a very good approximation, ignore the third term in Eq. (5).

For one tightly focused beam hitting the sample at the pole, the coefficients are given by

$$A_{lm} = \frac{1}{\kappa}\frac{R}{l}Y_{lm}^{*}(0,0)\frac{P}{R^2} \text{ with } P = 4\pi R^2\varepsilon\sigma_{\mathrm{SB}}T_0^4, \tag{7}$$

which results in the same functional dependence on the sample material (ε,κ), size (R), and temperature (T_0, or total laser power P) for each of the coefficients

$$A_{lm} \propto \frac{P}{\kappa R} \propto \frac{\varepsilon}{\kappa}RT_0^4. \tag{8}$$

This functional form is also maintained for other distributions of the incident power over the sphere surface; only the numerical value of the proportionality factor changes with changes in geometry. The case of a single laser beam focused to a point is the worst possible case for temperature uniformity. Uniformity can be improved by using a (preferably symmetric) arrangement of multiple heating laser beams and/or spreading the beam. In a symmetric arrangement any spherical harmonics that do not have that symmetry (this typically includes the lowest-order spherical harmonics) are eliminated, while spreading the beam reduces the amplitude of higher order spherical harmonics. For example, for a single spread-out beam hitting the sample normal to the pole the coefficients are

$$A_{l0} = \frac{R}{l\kappa}\int_0^{2\pi}\int_0^1 Y_{lm}^{*}(\vartheta,\varphi)\cos(\vartheta)d\cos(\vartheta)d\varphi\frac{P}{\pi R^2}. \tag{9}$$

Note that the integral here runs only over a half sphere, this is not the interval over which spherical harmonics are orthogonal. In practice, the beam may not be spread out completely uniformly and the absorption will depend on the angle of incidence, but the principle for calculating the coefficients is the same.

The coefficients of the leading term in Eq. (7) were computed for single beam (polar) and tetrahedral four-beam case, with the beam(s) focused to a point or uniformly spread over an entire hemisphere, for sample sizes of $R=1$ mm and $R=6.35$ mm. A T302 stainless steel ball with a thermal conductivity of $\kappa=16.7$ W/m K (at 1000 K) and a hemispherical total emissivity of $\varepsilon=0.3$ was considered.[10] For comparison, the bulk metallic glass forming alloy

TABLE I. Calculated coefficients of the leading term in the spherical harmonics expansion reflecting the maximum temperature variations over the sample for $Zr_{58}Nb_3Cu_{16}Ni_{13}Al_{10}$ samples of 1 mm in radius and a stainless steel ball with a radius of 6.35 mm heated with different heating geometries.

Sample	Radius of sphere (10^{-3} m)	Average temperature (K)	1 beam focused (K)	1 beam spread out (K)	4 beams focused (K)	4 beams spread out (K)
Vit106a	1	960	3.1	2.1	1.3	0.055
		900	2.4	1.6	1	0.043
		740	1.1	0.7	0.47	0.020
SST302	6.35	960	33.7	22.5	14.1	0.59
		900	26	17.3	10.9	0.46
		740	11.9	7.9	4.9	0.21

$Zr_{58}Nb_3Cu_{16}Ni_{13}Al_{10}$ (Vit106a) was chosen,[11] with $\varepsilon=0.26$ and $\kappa=25$ W/m K (at 1000 K).[12] Three different temperatures were considered, representative of the entire undercooled liquid region: 960 K, above the nose of the TTT diagram, 900 K, at the nose, and 740 K, below the nose. The results are summarized in Table I. For the case of a single point-focused beam, the leading term is the dipole term with coefficient $A_{l0}=\sqrt{(3/4\pi)}(P/\kappa R)=\sqrt{12\pi}(\varepsilon\sigma_{SB}/\kappa)RT_0^4$ [Eq. (7)]. At 960 K this evaluates to 3.1 K for a Vit106 sphere of 1 mm radius. Spreading the beam uniformly results in 2/3 of that value, or 2.1 K. By using four heating laser beams in a tetrahedral arrangement, the dipole and quadrupole terms are eliminated by symmetry. In addition, spreading the beam reduces the higher-order moments of the temperature distribution much more effectively than the dipole and quadrupole moments. By chance, the octupole term is reduced to zero when the beam is spread uniformly. The leading term the multipole expansion is then the 16-pole ($l=4$) term, with an amplitude of 0.055 K for a 1 mm radius sphere at 960 K. Clearly, modifications to the geometry of the heating laser beam arrangement should show remarkable effect in lowering the magnitude of the temperature variation on the sphere. These values are calculated for a steel sample and will be lowered if sample rotation is considered. Sample rotation is present in most levitation experiments and it smears out the temperature distribution and thereby lowers temperature gradients.

III. EXPERIMENTAL SETUP

The calculations suggest a dramatic enhancement in temperature uniformity if the sample is heated by four diffuse laser beams arranged in a tetrahedral geometry. In order to verify the predicted reduction of temperature variations over the sample, a T302 stainless steel ball, 6.35 mm in radius, with thermal conductivity of 16.7 W/m K was used. The relatively large sample size enhanced temperature variations and facilitated attaching thin thermocouples. The sample was suspended by four thin wires. A yttritium–aluminum–garnet laser with a Gaussian beam profile was used to heat the sample. Four beams of roughly equal intensity were produced by splitting the main laser beam as shown in Fig. 2. This setup consists of three 50/50 beam splitters and three mirrors. The use of beam splitters to divide the single laser beam into four beams eliminates the uncorrelated fluctuations in power, which would arise if separate lasers were used. With the present setup the laser power drift will only affect the average temperature but will not induce variations in temperature gradients (such as dipole, quadrupole, etc.).

In our setup this optical system was mounted on the top flange of a vacuum chamber in such a way that three of the four vertical beams went through the three windows that are located at the apexes of a equilateral triangle and the fourth beam went through an window that was located at the center of the triangle. The electrode assembly was so positioned in the chamber that the central beam irradiated the top of the sample while the three side beams were directed to the sample through mirrors. All the optical components are arranged in such a way that the four beams irradiate the sample satisfying a tetrahedral geometry.

The power of the roughly divided four beams were made equal by adjusting three attenuators that were inserted in the three stronger beam paths so that after the adjustment they had the same intensity (within experimental error) as the weakest fourth beam. This power equalizing procedure was done as follows: First, a reference temperature was established by measuring steady state sample temperature that was achieved when the sample was heated by the weakest laser beam alone. The sample temperature was measured by a fine thermocouple that was welded on the diametrically opposite point of the laser heating spot. The similar procedure was repeated one after another for three other beams,

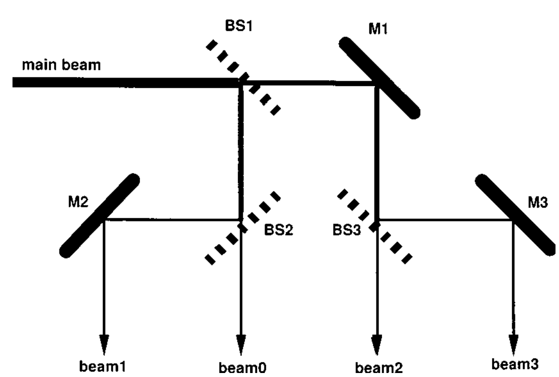

FIG. 2. Schematic illustration of the equalization of four laser beams originated from one laser. Three beam splitters (50/50) and three mirrors were used to divide the main laser beam into four beams of roughly equal power.

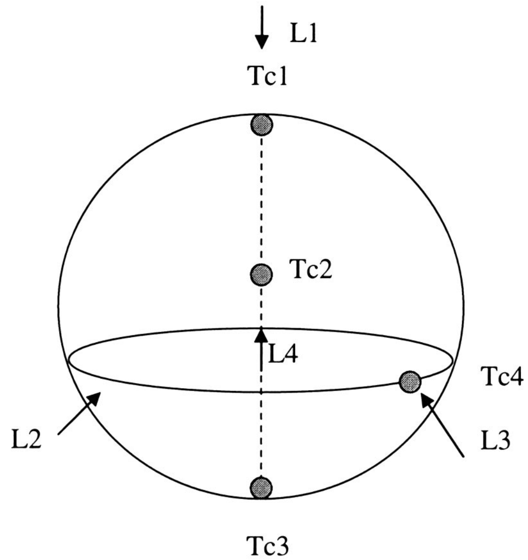

FIG. 3. Schematic illustration of the positions where the thermocouples were attached (shown by circle) and the spots where the laser beams impinged on the sample (→). For the one beam case, $L1$ was used and the maximum temperature variations for this case were measured by Tc3 and Tc1.

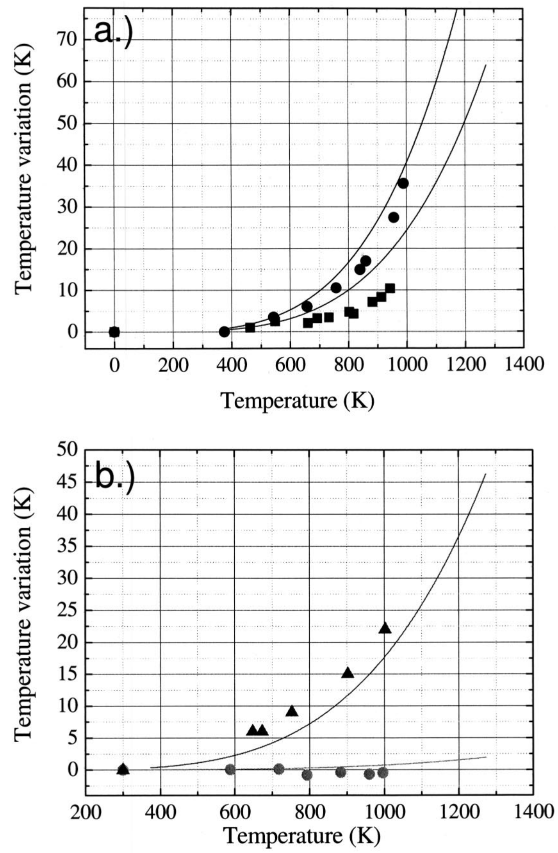

FIG. 4. Temperature variations within the sample for 1 beam heating geometry (a) [focused beam (solid circle) diffusive beam (solid square)] and 4 beam heating geometry (b) [focused (solid triangle) and diffuse (solid circle)]. The solid curves resulted from the calculations according to Eq. (7).

this time the sample temperature achieved by each beam was lowered by adjusting the attenuator until the reference temperature was achieved.

Four thermocouples were connected to the sample's surface as shown in Fig. 3. They were positioned in such a way that they detected the maximum temperature variations in the sample. Thermocouples Tc1 and Tc3 were utilized to measure the maximum temperature variations for the one beam case and Tc2 and Tc4 for the maximal temperature variation in the four beam case. Tc2 is located in the middle of the triangle formed by the laser spot 1, 2, and 3. K-type thermocouples of 76 μm in diameter were used and similar wires were used to support the sample. The temperature gradients were measured after the sample was allowed to equilibrate at each temperature. For calibration purpose, the characteristics of different thermocouples were tested in a uniform 1100 K temperature field established by a resistant furnace. Their temperature readings varied less than 0.7 K.

IV. EXPERIMENTAL RESULTS

Figure 4(a) shows the measured maximum temperature variations over the sample for a sample that was heated by a single laser beam. When the beam was focused on the sample to a spot of about 1.6 mm in diameter, the temperature variation on the sample was as much as 35 K for an average sample temperature of 980 K. If the beam was spread out to about 10 mm in diameter (diffuse beam) the temperature variation went down to 10.4 K at the average temperature 943 K. When the sample was heated by four focused laser beams in a tetrahedral geometry as shown in Fig. 4(b), the temperature variation still was as high as 22 K at an average temperature of 1003 K. However, as soon as the four beams in the tetrahedral geometry were spread out, the temperature variation was dramatically decreased to less than 0.5 K at an average sample temperature of 1003 K. This temperature variation was in the range of the experimental error of 0.7 K. Also shown in Fig. 4 is the amplitude of the temperature variations calculated according to Eq. (7) in which only the leading term was considered. An emissivity of 0.3 was used for the calculations.[10] Since the maximum temperature variations scales linearly with the samples radius, Fig. 4 can be used to extrapolate the temperature variations for different sample sizes.

The measurements of temperature uniformity confirm the main features of the calculations. The temperature variations scale with temperature to the fourth power, and spreading the beam as well as going to a tetrahedral four beam arrangement drastically reduce the temperature gradients. The calculated temperature variations show reasonable agreement with the experimental results. A source of quantitative discrepancy between the calculations and the experimental results may be explained by the position of the thermocouple which might not be exactly positioned at the largest temperature variations. General trends and scaling relations, however, would not be affected by such systematic errors. Further possible sources of error include imperfect alignment of the laser beams with the sample and/or with each other, and the unavoidable disturbance of the laser beams and the temperature profile by the thermocouple leads and support wires. According to a numerical integration of the heat flow through thin wire radiating heat from its surface the heat loss through the thermocouple and support wires is less than 3×10^{-3} W for temperatures below 1100 K. This is 0.25% of the total power input to the sphere at that temperature, resulting in a temperature variation 0.25% of that for the one beam focused case, i.e., less than 0.5 K. In addition, the temperature variations from thermocouple and support wires on opposite sides of the sphere largely cancel one another, so even with four thermocouples and four support wires, the temperature variation caused by heat loss through the wires is less than 1 K and therefore have a minor influence on the experimental error.

ACKNOWLEDGMENTS

This work was supported by the National Aeronautics and Space Administration (Grant No. NAG8-1744). The authors express their gratitude to Sundeep Mukherjee for technical assistance.

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