Nanoscale temperature measurements using non-equilibrium Brownian dynamics of a levitated nanosphere

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Supplementary Information

A Impinging and emerging gas model

Particles that are small in comparison to the laser wavelength (Rayleigh particles) are trapped in three dimensions at the highest intensity, I_0, of the beam due to their dipole moment interacting with the laser's electric field gradient. The trap's restoring force, $\vec{F} = (\vec{p}\cdot\vec{\nabla})\vec{E} = \frac{2\alpha}{c\epsilon_0}\vec{\nabla}I$, depends on the spheres' polarisability, α, and in harmonic approximation leads to trap frequencies $\omega_{x,y,z} = \sqrt{\frac{8\alpha\, I_0}{c\epsilon_0 M}}\, d_{x,y,z}$ for the three spatial directions x, y and z, where M is the sphere mass and $d_{x,y,z}$ are parameters given by the beam geometry in three dimensions[1]. Larger particles behave qualitatively similar but obey the more general Mie theory.

In any particular direction, x, the centre-of-mass motion of the spheres is generally described by the Langevin equation,

$$M\ddot{x}(t) + M\Gamma_{\mathsf{CM}}\,\dot{x}(t) + M\omega_x^2\, x(t) = F^{\mathsf{fluc}}(t), \tag{S.1}$$

where Γ_{CM} is the damping constant of the sphere's motion due to drag exerted by the surrounding gas and $F^{\mathsf{fluc}}(t)$ is the fluctuating force due to collisions with gas particles at times t. Time averages for Gaussian white noise are $\langle F^{\mathsf{fluc}}(t)\rangle = 0$ and $\langle F^{\mathsf{fluc}}(t+\tau)F^{\mathsf{fluc}}(t)\rangle = 2G\,\delta(\tau)$ with G given by the fluctuation-dissipation theorem, $G = k_B T\,\Gamma_{\mathsf{CM}}\, M$ where T is the temperature of the surrounding gas in equilibrium.

Here we consider spheres heated well above the room temperature and up to the material's melting point. Heat from the sphere surface will be transferred to colliding gas particles making them emerge more energetic. Since the sphere has a large accommodation coefficient these collisions are largely inelastic with the particles sticking on the surface and emerging independent in time and energy from the impinging gas particles. This situation is also referred to as diffusive reflection[2]. The distribution of the emerging gas particles is then assumed to be thermal[2,3], for a higher temperature, $T^{\mathsf{em}} > T^{\mathsf{imp}}$. To model the impact of the gas on the sphere's motion we modify the Langevin equation to separately include *two independent baths*, the impinging gas and the emerging gas:

$$M\ddot{x}(t) + M(\Gamma^{\mathsf{imp}} + \Gamma_x^{\mathsf{em}})\,\dot{x}(t) + M\omega_x^2\, x(t) = F^{\mathsf{imp}}(t) + F_x^{\mathsf{em}}(t). \tag{S.2}$$

Here Γ^{imp} and Γ_x^{em} are the damping constants for the sphere associated with each of the baths, and $F^{\mathsf{imp}}(t)$ and $F_x^{\mathsf{em}}(t)$ the corresponding noise terms. While the forces on the sphere due to the impinging gas are homogeneous, the forces due to the emerging gas particles can show spatial asymmetry caused by spatial variations in sphere surface temperature. The spatial dependence of the emerging gas forces is indicated by the subscript x.

As usual, the time-averaged noise terms vanish individually, $\langle F^{\mathsf{imp}}(t)\rangle = 0 = \langle F_x^{\mathsf{em}}(t)\rangle$ while the fluctuation dissipation theorem implies that the individual time-averaged correlations are $\langle F^{\mathsf{imp}}(t+\tau)F^{\mathsf{imp}}(t)\rangle = 2k_B T^{\mathsf{imp}}\,\Gamma^{\mathsf{imp}}\, M\,\delta(\tau)$ and $\langle F_x^{\mathsf{em}}(t+\tau)F_x^{\mathsf{em}}(t)\rangle = 2k_B T_x^{\mathsf{em}}\,\Gamma_x^{\mathsf{em}}\, M\,\delta(\tau)$. Here the subscript x indicates that in the present experiment the gas temperature may differ for different spatial directions. There exists no time correlation between the two baths, i.e. $\langle F^{\mathsf{imp}}(t+\tau)F_x^{\mathsf{em}}(t)\rangle = 0$, since the gas particle collisions with the sphere are strongly inelastic leading to a random delay between the arrival and departure of the gas molecules.

The two fluctuating forces and two damping forces contribute to the sphere's position power spectrum in any spatial direction x in an additive way,

$$P(\omega) = \frac{2k_B}{M}\,\frac{T^{\mathsf{imp}}\Gamma^{\mathsf{imp}} + T_x^{\mathsf{em}}\Gamma_x^{\mathsf{em}}}{(\omega_x^2-\omega^2)^2 + \omega^2\left(\Gamma_x^{\mathsf{em}} + \Gamma^{\mathsf{imp}}\right)^2}. \tag{S.3}$$

Even though the gas is not in an equilibrium state, the sphere's position power spectrum is of canonical form with an effective damping coefficient, $\Gamma_{\mathsf{CM}}^x := \Gamma_x^{\mathsf{em}} + \Gamma^{\mathsf{imp}}$, and an effective temperature, $T_{\mathsf{CM}}^x := \frac{T^{\mathsf{imp}}\Gamma^{\mathsf{imp}} + T_x^{\mathsf{em}}\Gamma_x^{\mathsf{em}}}{\Gamma_x^{\mathsf{em}} + \Gamma^{\mathsf{imp}}}$, defined for the centre-of-mass motion of the sphere in the x direction. We highlight that both, the measured centre-of-mass damping and temperature depend on which spatial dimension is observed due to the non-uniform landscape of the surrounding gas' heating. Finally, the position autocorrelation function (ACF) for the sphere's centre-of-mass motion is given by

$$ACF_{\mathsf{CM}}^x(\tau) = \frac{k_B T_{\mathsf{CM}}^x}{M\omega_x^2}\, e^{\frac{-\Gamma_{\mathsf{CM}}^x\tau}{2}}\,\left(\cos\hat{\omega}_x\tau + \frac{\Gamma_{\mathsf{CM}}^x}{2\hat{\omega}_x}\sin\hat{\omega}_x\tau\right) \tag{S.4}$$

where $\hat{\omega}_x = \sqrt{\omega_x^2 - {\Gamma^x_{\mathsf{CM}}}^2/4}$.

B Derivation of Γ^{em}

Treating the impinging and emerging gases separately enables us to account for the heat flow between sphere and gas. The different directionality of the two gases is taken into consideration when evaluating how each bath damps the motion of the sphere, characterised by the damping constants Γ^{imp} and Γ^{em}. To derive the damping coefficient Γ^{em} due to the emerging gas, we consider the particles emerging from a surface element $\mathrm{d}S$ of the sphere with normal vector defined along the z direction. By assumption their particle flux distribution is thermal at T^{em},

$$n^{S,\mathsf{em}}_{x,y,z} = C\, v_z\, e^{-v^2 h'} \tag{S.5}$$

with $h' = \frac{m}{2k_B T^{\mathsf{em}}}$ with m the mass of the gas particles, particle velocity $v^2 = v_x^2 + v_y^2 + v_z^2$, and a normalisation coefficient C. For the free molecular regime Epstein showed[2] that the impinging particle flux on the surface element is $n^{S,\mathsf{imp}} = \frac{N}{2\sqrt{\pi h}}\,(1 + \sqrt{\pi h}\,\cos\theta\, V)$, where N is the particle density of the gas, $h = \frac{m}{2k_B T^{\mathsf{imp}}}$, V is the speed of the sphere and θ the angle between the direction of the sphere's motion and the normal vector on the surface element $\mathrm{d}S$. The number of emerging particles from any surface element $\mathrm{d}S$ equals the number of impinging particles on that element,

$$n^{S,\mathsf{em}} = \int_0^\infty \int_{\mathbb{R}} \int_{\mathbb{R}} n^{S,\mathsf{em}}_{x,y,z}\, \mathrm{d}v_x \mathrm{d}v_y \mathrm{d}v_z \overset{!}{=} n^{S,\mathsf{imp}}, \tag{S.6}$$

where the integration in z direction considers only emerging particles with positive velocities, $v_z \geq 0$. Executing the integral fixes the coefficient C. We now derive the drag pressure experienced by the sphere due to the emerging gas particles from surface element $\mathrm{d}S$ by integrating over all leaving velocities,

$$p^{S,\mathsf{em}} = -m \int_0^\infty \int_{\mathbb{R}} \int_{\mathbb{R}} v_z\, n^{S,\mathsf{em}}_{x,y,z}\, \mathrm{d}v_x \mathrm{d}v_y \mathrm{d}v_z \tag{S.7}$$

$$= -\frac{mN(1 + \sqrt{\pi h}\,\cos\theta\, V)}{4\sqrt{h}\,\sqrt{h'}}. \tag{S.8}$$

Now considering only the term depending on the sphere velocity V and including the vector projection ratio $\cos\theta$ associated with each surface element and integrating over all surface elements $\mathrm{d}S = R^2\, \sin\theta\, \mathrm{d}\theta \mathrm{d}\phi$ gives the total drag force due to the emerging particles,

$$F^{\mathsf{em}}_{\mathsf{drag}} = -\int_0^\pi \int_0^{2\pi} p^{S,\mathsf{em}}\, \cos\theta\, \mathrm{d}S \tag{S.9}$$

$$= \frac{mNR^2\pi^{3/2}}{3\sqrt{h'}}\, V \overset{!}{=} M\, \Gamma^{\mathsf{em}}\, V. \tag{S.10}$$

This leads to the damping coefficient due to the emerging particles, $\Gamma^{\mathsf{em}} = \frac{\pi}{8}\sqrt{\frac{T^{\mathsf{em}}}{T^{\mathsf{imp}}}}\,\Gamma^{\mathsf{imp}}$, where the damping coefficient due to the impinging particles is given by[2] $\Gamma^{\mathsf{imp}} = \frac{4\pi}{3}\,\frac{mNR^2\bar{v}_{T_{\mathsf{imp}}}}{M}$ with mean thermal velocity $\bar{v}^2_{T^{\mathsf{imp}}} = \frac{8k_B T^{\mathsf{imp}}}{\pi m}$. This expression is valid under the assumption that the particles are diffusively reflected[2] and that the average sphere velocity is much smaller than the mean thermal velocity of the impinging gas particles, $\langle V\rangle << \bar{v}_{T^{\mathsf{imp}}}$. This is fulfilled in the present experiment as the mass of the sphere is very large in comparison to the gas particles. At the millibar pressures used in the experiment corrections due to intermediate Knudsen numbers[3] can be neglected.

References

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3 Beresnev, S.A., Chernyak, V.G., & Fomyagin, G.A. Motion of a spherical particle in a rarefied gas. Part 2. Drag and thermal polarization. *J. Fluid Mech.* **219**, 405-421 (1990).