Weighing an Optically Trapped Microsphere in Thermal Equilibrium With Air


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(Received 5 July 2020; revised 9 September 2020; accepted 18 September 2020; published 16 October 2020)

We report a weighing metrology experiment of a single silica microsphere optically trapped and immersed in air. Based on fluctuations about thermal equilibrium, three different mass measurements are investigated, each arising from one of two principle methods. The first method is based on spectral analysis and enables simultaneous extraction of various system parameters. Additionally, the spectral method yields a mass measurement with systematic relative uncertainty of 3.0% in 3 s and statistical relative uncertainty of 0.9% across several trapping laser powers. Parameter values learned from the spectral method serve as input, or a calibration step, for the second method based on the equipartition theorem. The equipartition method gives two additional mass measurements with systematic and statistical relative uncertainties slightly larger than those obtained in the spectral method, but over a time interval 10 times shorter. Our mass estimates, which are obtained in a scenario of strong environmental coupling, have uncertainties comparable to ones obtained in force-driven metrology experiments with nanospheres in vacuum. Moreover, knowing the microsphere’s mass accurately and precisely will enable air-based sensing applications.

DOI: 10.1103/PhysRevApplied.14.044027

I. INTRODUCTION

Optical trapping of nano- and microscale objects [1–3] has become a paradigmatic tool in diverse fields, from micromanipulation of biological samples [4–15] to center-of-mass cooling experiments [16–18] aiming to observe macroscopic quantum effects [19–21], to metrology experiments [22–24] with optomechanical sensing applications [25–29]. In such experiments, a tightly focused laser beam, named the optical tweezer [30–32], is used to polarize a dielectric particle and harmonically confine it to the beam’s intensity maximum.

It is often desirable to monitor the trapped particle’s position as a function of time, so a position-sensitive detector must be calibrated. Calibrating the detector usually requires knowledge of the trapped particle’s mass [22]. However, SiO2 nano- and microspheres, often the object of study in levitated optomechanics experiments, do not have a readily known mass. The Stöber process used to manufacture these particles [33] yields very spherical results with a low dispersion of radius (approximately 3%), but a mass density that can vary in excess of 20% [33,34]. Calculated with these values, the uncertainty in mass is about 22%. For this reason, recent work has focused on mass metrology of nano- and microspheres optically trapped in vacuum using methods of electrostatic levitation [34], oscillation [23], and trapping potential nonlinearities [24], and, most recently, a drop-recapture method performed in air [35]. The mass uncertainty achieved in each of these experiments is at the level of one to a few percent. Each has unique advantages like no assumptions on particle geometry, and distinct challenges, e.g., control of the particle’s charge, accurate modeling of local potentials (gravitational, electric, or optical), or vacuum capabilities including feedback cooling.

Here, we report on a mass metrology experiment with uncertainty similar to previous work, but performed on a 1.5 μm radius SiO2 microsphere optically trapped in air [36] at room temperature and pressure. Our experiment employs a dual-beam optical trap [1,37], sketched in Fig. 1(a) and elaborated upon in Ref. [38]. Our system remains in thermal equilibrium at all times making for a simple protocol. Moreover, we explore two distinct methodologies leveraging our detector’s high spatiotemporal resolution. In the first spectral method, we fit an average voltage signal power spectral density (PSD) to simultaneously extract parameters that make no assumptions on the physical conditions of the experiment. We then fix conditions known with high accuracy—the air temperature, air viscosity, and particle radius—to compute the harmonic trap strength k, microsphere mass density ρ, and detector calibration factor β, as well as the uncertainties and correlations of these parameters. The microsphere

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II. POWER SPECTRAL DENSITY PARAMETER ESTIMATION

The dynamics of the trapped microsphere along the x axis may be modeled by the harmonically bound Langevin equation of motion

\[ m\ddot{x} + \gamma \dot{x} + kx = F(t), \]

where \( m = 4\pi \rho R^3 / 3 \) is the mass of the microsphere with radius \( R \) and density \( \rho \), \( \gamma = 6\pi \eta R \) is the Stokes friction coefficient, \( \eta \) is the viscosity of air, and \( k \) is the trap strength. The stochastic thermal force \( F(t) = g\xi(t) \) is assumed to have the form of zero-mean \( \langle \xi(t) \rangle = 0 \), delta-correlated \( \langle \xi(t)\xi(t') \rangle = \delta(t-t') \) white noise with strength \( g = \sqrt{2k_BT\gamma} \) (according to the fluctuation-dissipation theorem), and in which \( T \) is the air temperature, \( k_B \) is Boltzmann’s constant, and \( \langle \cdot \rangle \) denotes ensemble averages over realizations of \( \xi \). Writing Eq. (1) in terms of the Fourier transforms \( \tilde{x}(\omega) \) and \( \tilde{F}(\omega) \) [44] lets one deduce the position PSD \( S_x(\omega) \) such that \( \langle \tilde{x}(\omega)\tilde{x}(\omega') \rangle = S_x(\omega)\delta(\omega - \omega') \) [45], where \( \omega = 2\pi f \) is the angular frequency.

In our experiment, we record a unitless voltage signal \( V(t) = V_-(-t)/V_+(0) \), where \( V_- \) is proportional to the difference in optical power delivered to the two ports of the balanced photodetector at time \( t \) and \( V_+(0) \) is proportional to the total detection power at time \( t = 0 \). Normalizing the signal in this way accounts for small variations in detected power upon changing the trapping laser power. Here \( V(t) \) is proportional to the microsphere’s position along the x axis: \( x(t) = V(t)/\beta \), where \( \beta \) is the calibration factor that we report in units of \( \mu m^{-1} \).

From such considerations, the theoretical (one-sided) PSD of our voltage signal is understood to be

\[ S_V(\omega) = \frac{\beta^2 4k_B T \gamma}{(mo^2 - k^2) + \gamma^2\omega^2}. \]

Multiple trials of experimental power spectra must be averaged together before we attempt to learn relevant physical parameters. We collect ten trials of the voltage signal, each 0.3 s long, at a sampling rate of 50 MHz. In postprocessing, the signal is low-pass filtered by averaging together nonoverlapping blocks of 256 samples for improved spatial resolution. The new effective sampling rate is 195 kHz. Using Bartlett’s method [38,46] with four windows per trial—for a total of 40 averages of length \( T = 84 \) ms—we estimate the experimental voltage PSD, denoted \( \hat{S}_{V,k} = \hat{S}_V(k) \). The index \( k \) labels the discrete frequencies at which the experimental PSD is known. The frequency resolution is \( f_{k+1} - f_k = T^{-1} \).

Once a set of trials is collected, we fit the experimental data \( \hat{S}_V \) to

\[ S_V(f; \theta) = \frac{1}{a + bf^2 + cf^4}, \]

This paper is organized as follows. First, we review the relevant physics and outline our PSD parameter estimation method in Sec. II. In Sec. III we show how the PSD parameters, along with the equipartition theorem, allow us to weigh the microsphere in three different ways. There, we also present the results that we further discuss in Sec. IV. Finally, we conclude with this work’s significance in Sec. V.
in which we have defined the column vector of free parameters \( \boldsymbol{\theta} = (a, b, c)^\top \). In particular, the fit is done using the maximum likelihood estimation method [47–49] that we briefly outline next.

First, note that each data point of an \( n \)-trial-averaged PSD is subject to gamma-distributed noise (the convolution of \( n \) exponential distributions) [48,50], written

\[
\mathcal{P} = \frac{1}{S(\hat{S}_V)} \frac{n^n \Gamma(n)}{\Gamma(n)} \left( \frac{\hat{S}_V}{S(\hat{S}_V)} \right)^{(n-1)} \exp\left( -n \frac{\hat{S}_V}{S(\hat{S}_V)} \right),
\]

where \( \Gamma(n) = (n-1)! \) is the gamma function and \( S(\hat{S}_V) \) is the mean value of the distribution. Then, the likelihood of measuring the entire data set \( \hat{S}_V \) given a model \( S_V(f_k, \theta) \) is the joint distribution

\[
\mathcal{P} = \prod_k \mathcal{P}(\hat{S}_V | \theta).
\]

Maximizing the negative log likelihood

\[
\mathcal{L}(\theta, \hat{S}_V) = n \sum_k \left( \log[S(f_k; \theta)] + \frac{\hat{S}_V(f_k)}{S(f_k; \theta)} \right) + C,
\]

where \( C = \sum_k \log \Gamma(n) - n \log n - (n-1) \log \hat{S}_V(k) \) is a constant with respect to the free parameters and thus inconsequential for the minimization, and \( n = 40 \) is the number of spectra averaged together in the experiment.

Good starting values for the minimization can be calculated analytically and implemented numerically, a convenient feature that is not possible if one attempts to fit directly to Eq. (2) [48]. Maximum likelihood fitting accounts for the gamma-distributed PSD data, unlike more common least-squares fitting algorithms that assume normally distributed noise and thus provide biased PSD parameter estimations [48]. In the end, the minimization gives the best-fit parameters \( \hat{\theta} = (\hat{a}, \hat{b}, \hat{c})^\top \) that maximize the likelihood of the data given the model \( \mathcal{P}(\hat{S}_V | \theta) = \exp(-\mathcal{L}(\theta, \hat{S}_V)) \). In Fig. 2(a) we show experimental PSD and the best-fit curve for two different trapping laser powers and compare to the noise inherent to the detection system. To measure the parameter fitting uncertainty and correlation, and inspired by the profile likelihood method [47,49], we scan \( \theta \) in the vicinity of \( \hat{\theta} \) over a volume of parameter space to build up a three-variate probability distribution \( \mathcal{P} \) [see Figs. 2(b) and 2(c)] that is fit to a three-variate Gaussian distribution

\[
\mathcal{P} \approx \exp \left[ -\frac{1}{2} (\theta - \hat{\theta})^\top \Sigma_\theta^{-1} (\theta - \hat{\theta}) \right],
\]

where \( \Sigma_\theta \) is the variance-covariance matrix of the fitted parameters:

\[
\Sigma_\theta = \begin{pmatrix} \sigma_a^2 & \sigma_{ab} & \sigma_{ac} \\ \sigma_{ab} & \sigma_b^2 & \sigma_{bc} \\ \sigma_{ac} & \sigma_{bc} & \sigma_c^2 \end{pmatrix}.
\]

The absolute residuals \( |P - P_G| \) are bound below the 1% level. The 95th percentile is bound below the 0.1% level [38]. The vector \( \hat{\theta} \) resulting from the fit is taken as the best-fit parameter set. The matrix \( \Sigma_\theta \) resulting from the fit provides the variance-covariance matrix of the fitted parameters.
FIG. 2. (a) Voltage PSD for 6.5 (blue) and 234.0 mW (red) of trapping laser power. Experimental data is depicted with open circles and consists of 40 independent PSD averages and further bin averaged on a logarithmic horizontal scale for visualization purposes. The solid lines are the maximum likelihood best fit to Eq. (3). The vertical dashed lines mark the bounds on the data used in the fit. The lower bound is color coded with the fitting line and the upper bound (black dashed line) is shared. The noise spectrum (solid black) is collected under identical detection conditions as the other two curves but with no microsphere present. We see a technical noise floor at high frequencies and electronic-laser noise, including 60 Hz harmonics and 1/f noise, at low frequencies. The noise peak at around 120 Hz must be omitted when fitting the 234.0 mW spectrum, as indicated by the lower bound (red dashed line). (b) For 6.5 mW of laser power, we plot the isosurfaces of $P = \exp(-L)$ [see Eq. (6)] as a function of fitting parameters $\theta$. The isosurfaces are taken at Gaussian widths of 3 sigma (purple), 2 sigma (blue), and a core from the peak (red) to the 1-sigma width (green). (c) As in (b) but for 234.0 mW of trapping laser power. By fitting the likelihood data clouds shown in (b) and (c) to a three-variate Gaussian distribution, we extract the best-fit parameters $\hat{\theta}$ and the variance-covariance matrix $\Sigma_{\theta}$. (d) Trap strength $k$, (e) microsphere mass density $\rho$, and (f) calibration constant $\beta$, each extracted from PSD fits as a function of laser power $P$. In (d)–(f), error bars reflect systematic uncertainty calculated by error propagation, including correlation among fitted parameters.

We calculate the variance-covariance matrix of the physical parameters in terms of the fitting and constant parameters via the error propagation equation $\Sigma_{\theta} = J_{\theta} \Sigma_{\phi} J_{\theta}^\top$. The Jacobian matrix (evaluated at the optimal fitting parameters) is $(J_{\theta})_{ij} = \left[\partial \Theta_i / \partial \phi_j\right]_{\theta=\hat{\theta}}$. We have verified that the parameters and uncertainties deduced by the procedure described here and conveniently visualized in Figs. 2(b)–2(c) agree quantitatively with the Monte Carlo method that generates and fits many artificial PSDs by sampling the appropriate gamma distribution. Our technique yields directly the probability density, sidestepping the need for binning and fitting or kernel-density estimating the Monte Carlo results.

We now understand how to estimate $k$, $\rho$, and $\beta$, including uncertainty and correlation, from an experimental voltage PSD $\hat{S}_V$. The results are presented in Figs. 2(d)–2(f) for experiments on the same trapped microsphere and that scan the trapping laser power from 6.5 to 257.2 mW. We observe no unexpected dependence of the physical parameters on laser power except for the calibration constant
that exhibits a nonmonotonic curve, first decreasing then increasing with laser power [Fig. 2(d)]. Thus, we conclude that heating of the microsphere due to the laser is inconsequent because of the strong environmental coupling. This is not the case for experiments in vacuum [55]. The trend in $\beta$ is reproducible when the experiment is repeated with different microspheres, suggesting the source is most likely slight beam deviations caused by the HWP-PBS pairs used to control the trapping and detected power.

**III. MASS MEASUREMENT TECHNIQUE**

Upon learning PSD fitting parameters, presented in Sec. II, it is straightforward to estimate the mass using the density and radius of the microsphere. However, the equipartition theorem, $k_B T = m \langle \chi^2 \rangle = k \langle \chi^2 \rangle$, provides two additional possibilities. The three mass measurements written in terms of the augmented independent variables $\phi' = (a, b, c, \langle V^2 \rangle, \langle V' \rangle, R, \eta, T)$ read

$$m_1(\phi') = \frac{4}{3} \pi R^3 \rho,$$

$$m_2(\phi') = \frac{k_B T}{\langle V^2 \rangle} \beta^2,$$

$$m_3(\phi') = \frac{\langle V' \rangle^2}{\langle V^2 \rangle} k.$$

The benefit of $m_2$ and $m_3$ is that, once a PSD fit is used to calibrate the system, further data can be collected to estimate the variances $\langle V^2 \rangle$ and $\langle V' \rangle$, which may be used to update the mass measurement in the case it changes with time. Of course, there is nothing to update if the mass is unchanging. Nonetheless, to make use of methods $m_2$ or $m_3$, we must make an adequate estimate of the required variances. In Figs. 3(a) and 3(b) we show the histograms of position and velocity (proportional to $V$ and $V'$, respectively) for high and low trapping laser powers. The histograms consist of data from a single 0.3 s trial. Overlaid on each histogram are Gaussian fits with variance as the only free parameter. Uncertainty in the variance calculation is taken as the standard deviation of variances calculated across ten trials.

For an uncorrelated voltage trace of length $\tau$, the uncertainty in the variance estimate scales as $\tau^{-1/2}$, which is a thermally limited trend. However, at short times ($\tau < m/\gamma$), the data is correlated due to the microsphere’s dynamics and, at long times, slow drifts in the system tend to affect the signal’s variance. One way to quantify those correlations and to determine the optimal time over which our measurements are thermally limited is performing an Allan-deviation stability analysis [22,28,57,58].

In Fig. 3(c) we show the results of our Allan-deviation experiment performed with 22.8 mW of trapping laser power. Accordingly, our system is stable out to about 30 s, so using 0.3 s of data for estimating the variances allows for 100 independent mass measurements before the slow drifts demand recalibration of the apparatus. It is in this sense that methods $m_2$ and $m_3$ are faster than $m_1$.

In Fig. 4 we show the results of our three mass measurement procedures. We find $\bar{m}_1 = 24.8$ pg, $\bar{m}_2 = 25.1$ pg, and $\bar{m}_3 = 27.4$ pg, where the over bar denotes an average over the 14 experiments at different total trapping laser powers. Error bars reported are calculated [38] by considering covariance of the PSD fitting parameters, and uncertainties in both fixed parameters and variances. We consider such error bars systematic uncertainty, denoted $\sigma_{m_i}^{\text{sys}}$, $i = 1, 2, 3$. The statistical uncertainty (or fluctuation), denoted $\sigma_{m_i}^\text{stat}$, is calculated as the standard deviation across the 14 experiments at different laser powers. Measurement $m_1$, which is based entirely on the PSD analysis, has
the smallest relative error bars \( (\bar{\sigma}_{\text{sys}} / \bar{m}_1 = 3.0\%) \) and the smallest relative statistical uncertainty \( (\sigma_{\text{stat}} / \bar{m}_1 = 0.9\%) \). Measurement \( m_2 \), which supplements the PSD analysis by estimating the voltage-derivative variance, agrees well with \( m_1 \), albeit with relative error bars at 4.1% and relative statistical fluctuations at 1.6%. The benefit of \( m_2 \) is that, once an initial PSD analysis is performed, parameters like mass (or temperature) can be subsequently updated 10 times faster than collecting data for additional PSD analysis. Measurement \( m_3 \) has the largest systematic and statistical relative uncertainties, 6.7% and 3.0%, respectively. Furthermore, method \( m_3 \) displays additional systematic error because it deviates from \( m_1 \) and \( m_2 \) by nearly 10%. We speculate on the source of this uncertainty in the next section.

For comparison, the mass according to the manufacturer values of density \( \rho_{\text{Bangs}} = 2.0 \text{ g cm}^{-3} \) \( (\sigma_{\text{Bangs}} / \rho_{\text{Bangs}} = 20\%) \) and our radius measurement \( R = 1.51 \mu\text{m} \) \( (\sigma_R / R = 2.9\%) \) is \( m_{\text{Bangs}} = 28.84 \text{ pg} \) with an uncertainty of 22% that agrees within the uncertainty tolerance of all our mass measurements, despite the discrepancy of the mean values.

### IV. DISCUSSION

The systematic bias in \( m_3 \) of Fig. 4 is hypothesized to be dominated by the low-frequency electronic noise apparent in Fig. 2(a). By selecting an appropriate lower bound for the fit, the spectral method easily removes the influence of the noise resonances, the most severe of which appears at 120 Hz with a width of about 100 Hz. However, the time domain estimate of \( \langle V^2 \rangle \) includes variance due to that noise. To estimate the effects of such noise, we can model the experimental PSD as the sum of the best-fit PSD and the experimental noise PSD containing the noise peak near 120 Hz, \( S_V(f) = S_{V, \text{est}}^0(f) + S_{V, \text{noise}}^0(f) \). Using Parseval’s theorem, \( \langle q^2 \rangle = \int_0^{\infty} S_q(f) df \), we have

\[
\langle V^2 \rangle \approx \langle V^2 \rangle_{\text{PSD}} + \int_{70 \text{ Hz}}^{170 \text{ Hz}} S_{V, \text{noise}}^0(f) df,
\]

where \( \langle V^2 \rangle \) is the variance of the voltage signal observed in the time domain, \( \langle V^2 \rangle_{\text{PSD}} = k_B T \beta^2 / k \) is the variance estimate provided by the PSD parameters. The excess variance \( \Delta \langle V^2 \rangle \equiv \langle V^2 \rangle - \langle V^2 \rangle_{\text{PSD}} \) is about 10% of \( \langle V^2 \rangle_{\text{PSD}} \), which agrees with the discrepancy between \( m_1 \) and the other measurements. The quantity \( \Delta \langle V^2 \rangle \) can also be estimated by numerically integrating the observed noise spectrum. We find [38] that the integral of the noise PSD in the frequency band 70–170 Hz predicts the observed excess variance and hence also the bias in \( m_3 \).

The effects of low-frequency noise resonances are suppressed when estimating \( \langle V^2 \rangle \). The reason is because, in general, \( S_q(f) = (2\pi f)^2 S_V(f) \), so high-frequency components of a signal have a quadratically larger weighting factor in the variance compared to low-frequency noise. We find, by direct calculation on our data, that \( \Delta \langle V^2 \rangle \sim 2\% \), which agrees with the numeric integration of \( S_{V, \text{noise}}^0(f) \) over all frequencies above 80 kHz [38].

A recent experimental effort [23] measured the mass of 0.143 \( \mu\text{m} \) radius SiO\(_2\) spheres optically trapped in vacuum to be 4.01 fg with 2.8% uncertainty with 40 s of position data. Their oscillating electric field method makes no assumption on particle shape or density, though a density of 2.2 g cm\(^{-3}\) agrees with their measurements. In Ref. [34], a 2.6 \( \mu\text{m} \) radius sphere is optically trapped and levitated with a static electric field as the trapping laser power is reduced, resulting in a mass measurement of 84 pg with 1.8% uncertainty with 42 min of data. The density is also measured to be 1.55 g cm\(^{-3}\) with 5.16% uncertainty. A third strategy used in Ref. [24] stabilizes oscillations of a 0.082 \( \mu\text{m} \) radius sphere in the nonlinear-trapping regime to deduce the detector calibration constant with 1.0% uncertainty and a mass of 3.63 fg with 2.2% uncertainty. Finally, very recent work [35] used a drop-recapture method and camera-based detection with time resolution that could not quite resolve the microsphere’s instantaneous velocity. Fitting position autocorrelation functions, they measured their resin particle’s radius to be 2.3 \( \mu\text{m} \) with 4.3% statistical uncertainty. In the drop-recapture experiments, 90 s worth of trials were used to deduce a mass of 55.8 pg with 1.4% statistical uncertainty and 13% systematic uncertainty. The authors combined the radius and mass measurements to deduce a density of 1.1 g cm\(^{-3}\) with 9.1% statistical uncertainty.

As a comparison, we present a summary of our physical parameter values and uncertainties in Table I. Based entirely on thermal equilibrium analysis, our two most accurate mass estimates have uncertainties of 3% to 4% as compared to the 1% to 2% uncertainty in
FIGURE 4. Mass measurement results on a silica microsphere. A. Sample image showing the microsphere in a non-resonant optical tweezers configuration. B. Results from the spectral measurement method showing the resonance at 45 Hz. C. Results from the equipartition method showing the change in mass from 2 s to 3 s. The error bars represent the uncertainty in the measurement. The work presented here demonstrates the sensitivity of optical tweezers in a scenario of strong environmental coupling, suggesting applications in air-based sensing. For example, single-site ice nucleation could be monitored in real time as a change in mass of the trapped particle. Alternatively, in a system of constant mass, one could first measure the mass using the spectral method and then use the equipartition method to measure changes in temperature within the trapping medium, which could be driven out of equilibrium with a temperature gradient to probe temperature-gradient-induced turbulence at small scales of space and time.

The equipartition theorem may be challenged by nonequilibrium dynamics. However, in the hydrodynamic regime where thermodynamic state variables are relevant in the sense of quasiequilibrium, we believe our method will be quite applicable. The small sensor size means that the dynamics are fast to respond to changes in the environment (on the scale of $m/γ \sim 45 \mu s$ in this work). Even in the complete absence of thermal equilibrium, where the notion of temperature is no longer defined, our position and velocity data may be used to compute more general velocity structure functions when the simple variance appearing in the equipartition theorem is insufficient [59,60]. We consider such nonequilibrium studies a fruitful direction for future optical tweezer experiments.

ACKNOWLEDGMENTS

The authors are grateful to Y. Stratis, I. Bucay, Y. Lu, K. S. Melin, S. Bustabad, L. Gradl for helping with daily lab activities and making the lab a pleasant environment. We also thank T. Li and J. Mo for helpful information and A. Helal for assisting with SEM imaging of the silica microspheres.
APPENDIX: PARAMETER CONVERSIONS

The physical parameters, denoted by the column vector $\Theta = (k, \rho, \beta)^\top$, are functions of the independent variables $\phi = (a, b, c, R, \eta, T)^\top$. First we define

$$
d_1 \equiv b + \sqrt{ac},
$$
(A1)

$$
d_2 \equiv b + 2\sqrt{ac}.
$$
(A2)

Then, for $\Theta(\phi)$, we have

$$
k(\phi) = 12\pi^2 \eta R \sqrt{\frac{a}{d_2}},
$$
(A3)

$$
\rho(\phi) = \frac{9\eta}{4\pi R^2} \sqrt{\frac{c}{d_2}},
$$
(A4)

$$
\beta^2(\phi) = \frac{6\pi^3 \eta R}{k_B T d_2}.
$$
(A5)

The mass measurements $m = (m_1, m_2, m_3)^\top$ are a function of the augmented independent variables, $\phi = (a, b, c, (\phi), (\phi)^2, R, \eta)$ (noting that $\partial m / \partial T = 0$). For explicit formulae, we have

$$
m_1(\phi') = 3\eta R \sqrt{\frac{c}{d_2}},
$$
(A6)

$$
m_2(\phi') = \frac{6\pi^3 \eta R}{d_2} \frac{1}{(\phi^2)},
$$
(A7)

$$
m_3(\phi') = 12\pi^2 \eta R \sqrt{\frac{a}{d_2} \frac{(\phi^2)}{(\phi^2)}}.
$$
(A8)

We next define

$$
u_1 \equiv \frac{3}{16\pi^2 R^2}, \quad u_2 \equiv \frac{1}{2\sqrt{6\pi^3 \eta R k_B T}},
$$
(A9)

$$
u_1 \equiv \frac{1}{4\pi^2}, \quad v_2 \equiv \frac{\pi}{(\phi^2)}, \quad v_3 \equiv \frac{(\phi^2)}{(\phi^2)},
$$
(A10)

and to write the Jacobians

$$
\frac{\partial \Theta}{\partial \phi} = \frac{6\pi^2 \eta R}{\sqrt{ad_2^2}} \begin{pmatrix}
-\frac{d_1}{u_1 c} & -\frac{a}{u_1 \sqrt{a c}} & -\frac{\sqrt{a^2/c}}{u_1 d_1 \sqrt{a/c}} & 2a u_1 d_1 \sqrt{a/c}\sqrt{a/T} & 2a u_1 d_1 \sqrt{a/c}/\eta & 0 \\
-\frac{u_1 \sqrt{a}}{u_2 \sqrt{c}} & -\frac{u_2 \sqrt{a}}{u_2 \sqrt{c}} & -\frac{a c}{u_2 \sqrt{c}} & u_2 a / \sqrt{c} & 0 & 0 \\
-\frac{ad_2 / \eta}{u_2} & -\frac{ad_2 / \eta}{u_2} & \frac{d_2}{u_2} & -\frac{u_2 \sqrt{d_2}}{ \sqrt{a}} & 0 & 0 \\
\end{pmatrix},
$$
(A11)

$$
\frac{\partial m}{\partial \phi} = \frac{6\pi^2 \eta R}{\sqrt{ad_2^2}} \begin{pmatrix}
-\frac{v_1 c}{v_3 a} & -\frac{v_1 \sqrt{ac}}{v_3 a} & v_1 d_1 \sqrt{a/c} & 0 & 0 & 2v_1 d_1 \sqrt{a/c}/R & 2v_1 d_1 \sqrt{a/c}/\eta \\
-v_3 d_1 & -v_3 a & -v_3 \sqrt{a^2/c} & -v_2 \sqrt{a^2/c}/(\phi^2) & 0 & 0 \\
\end{pmatrix},
$$
(A12)

[44] The Fourier integrals are defined as $x(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{x}(\omega) e^{-i\omega t} d\omega$ and $\tilde{x}(\omega) = \int_{-\infty}^{\infty} x(t') e^{i\omega t'} dt'$.