Author preprint

Second-order boundary corrections to the radial acoustic eigenvalues for a spherical cavity

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Submitted to Metrologia: 15 June 2012
Accepted as Short Communication: 24 July 2012

Abstract
We calculated the eigenvalues of the radially-symmetric acoustic modes of a gas-filled, spherical cavity to order $(\delta_t/a)^2$, where δ_t is the thickness of the thermal boundary layer and a is the radius of the cavity. Our results explain an anomaly revealed by high-precision acoustic measurements made to re-determine the Boltzmann constant.

The recent decision to re-define the kelvin has renewed interest within the metrological community to re-determine the Boltzmann constant k_B with lower uncertainty [1]. To date, the most accurate determinations of k_B are based on measurements of the speed of sound in a noble gas contained in a spherical or quasi-spherical cavity [2,3][1]. The theory for the radially-symmetric gas oscillations including the effects of thermal dissipation and shape deformations is well established [2,4,5]. Mehl [6] calculated to second order the effect on the eigenvalues due to arbitrary shape deformations using boundary shape perturbation theory and showed that the radial-mode eigenvalues are unchanged to first order when the deformations preserve the volume. The effect of thermal dissipation at the cavity's wall on the resonance frequencies and half-widths has been calculated to first order using perturbation theory [4] and verified from the exact solution for spherical geometry [5]. Perturbation theory treats the thermal boundary layer as a small, effective surface admittance that shifts the resonance frequencies by $\Delta f_t^{(1)}$ and increases the half-widths by $g_t^{(1)}$ from their values when the boundary layer is neglected. Importantly, the first-order theory for the boundary layer predicts that the resonance frequency shifts downward and the half-width increases by the same fractional amount, *e.g.*

$$\frac{\Delta f_t^{(1)}}{f_{0n}^{(0)}} \approx -(\gamma-1)\frac{\delta_t}{2a} \tag{1a}$$

$$\frac{g_t^{(1)}}{f_{0n}^{(0)}} \approx (\gamma-1)\frac{\delta_t}{2a} \tag{1b}$$

for the $(0,n)$ radial mode in a spherical cavity. Here, $\gamma = C_p/C_V$ is the ratio of the isobaric and isochoric heat capacities, $\delta_t = (D_t/\pi f)^{1/2}$ is the thickness of the thermal boundary layer, D_t is the thermal diffusivity, f is the frequency, and a is the radius of the cavity. The superscripts in parentheses indicate the order of the approximation.

As part of the effort to re-determine k_B, de Podesta, *et al.* [7] and Gavioso, *et al.* [8] measured the resonance frequencies and half-widths of the radial acoustic modes of gas-filled quasi-spherical cavities with extraordinary precision. In [7] the gas was argon, and in [8] the gas was helium. Surprisingly, both groups found that, at low pressures and low frequencies, the measured half-widths g_m were narrower than the half-widths g_{calc} calculated using the first-order boundary layer correction and other, smaller perturbations. For the (0,2) mode in argon at

[1] Note: in Sec. 5.1.3 of [3], Pitre *et al*. state that their "largest value of $1/Q^2$ was 4.04×10^{-8}, obtained for the (0,2) mode at 50 kPa…" The value 4.04×10^{-8} is erroneous. The maximum value of $1/Q^2$ from the thermal boundary loss is 4.6×10^{-7}, which is consistent with Pitre *et al.*'s figure 28 and is comparable to the second-order corrections discussed here.

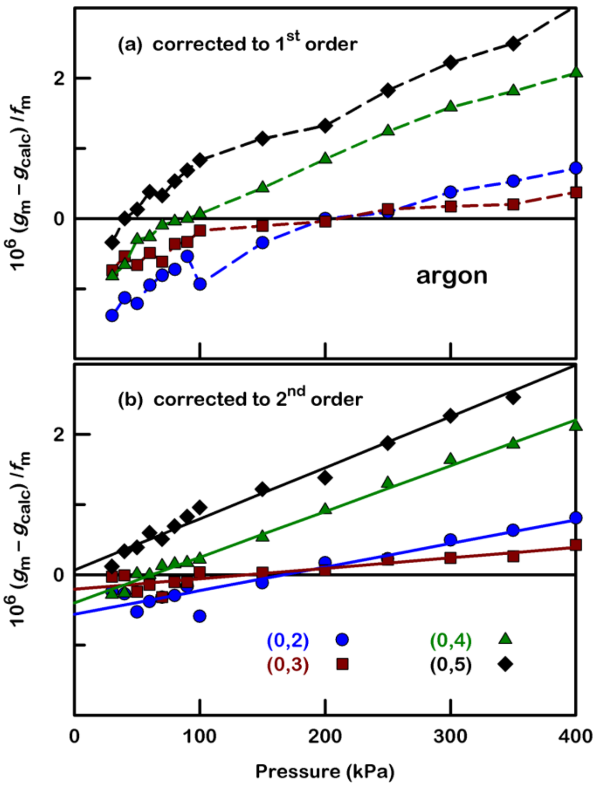

Figure 1. The scaled, measured excess half-widths $10^6(g_{\mathrm{m}} - g_{\mathrm{calc}}) / f$ for the lowest 4 acoustic radial modes of argon in a quasi-spherical cavity [7] as a function of pressure. (a) g_{calc} includes the thermal boundary correction to $O(\delta_{\mathrm{t}}/a)$. (b) g_{calc} includes the thermal boundary correction to $O(\delta_{\mathrm{t}}/a)^2$. The solid lines are linear fits to the data.

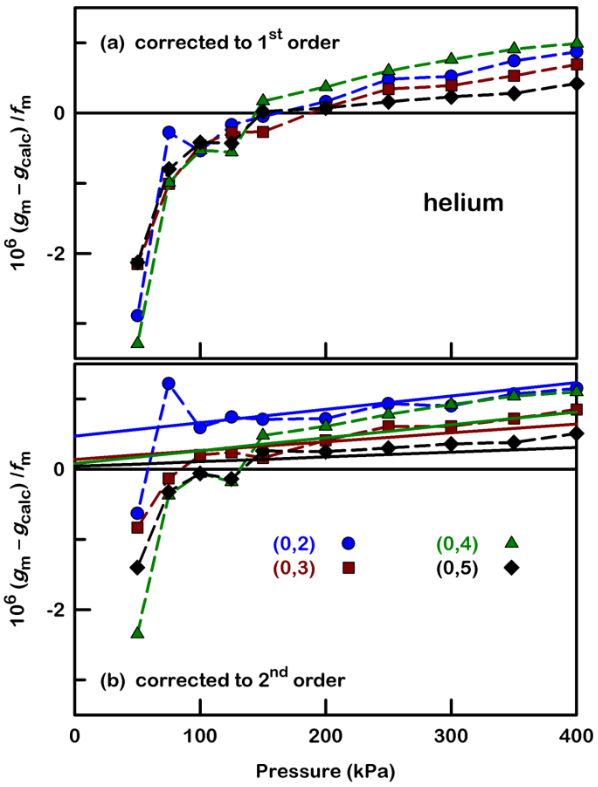

Figure 2. Same as figure 1 for the radial modes of helium from [8]. The solid lines are linear fits to the data up to 1 MPa, but omitting the data at 0.05 MPa.

30 kPa from [7], $\delta_{\mathrm{t}}/a \approx 1.2\times10^{-3}$ and $g_{\mathrm{m}}/f_{\mathrm{m}} \approx 400$ ppm (1 ppm = 1×10^{-6}). For the (0,2) mode in helium at 50 kPa from [8], $\delta_{\mathrm{t}}/a \approx 1.7\times10^{-3}$ and $g_{\mathrm{m}}/f_{\mathrm{m}} \approx 600$ ppm. Figures 1(a) and 2(a) show that the scaled excess half-widths $10^6 \times \left(g_{\mathrm{m}} - g_{\mathrm{calc}}\right)/f$ for the first 4 radial modes have a downward curvature and are negative at low pressures. This anomalous behavior is most pronounced for the (0,2) mode and more pronounced in helium than in argon. To the extent that this result is not understood, it reduces our confidence in acoustic determinations of k_{B}.

We calculated the effect of the thermal boundary layer on the resonance frequencies and half-widths to second order $O(\delta_{\mathrm{t}}/a)^2$ for the radial modes in a spherical cavity and applied a correction to the data (see figures 1(b) and 2(b)). The solid lines in figure 1(b) and 2(b) serve to guide the eye. The second-order correction has no adjustable parameters, and it does not change the resonance frequencies. Nevertheless, it removes the curvature from the argon data and significantly reduces the negative intercepts to much less than 1 ppm. For helium, the second-order correction removes most of the curvature, and if the lowest pressure data (0.05 MPa) are ignored, the intercepts from linear fits are positive and closely grouped near zero. The small residual curvature in the helium data may indicate the presence of additional second-order effects.

Linear dependence on pressure of the excess half-widths has been reported in many publications on acoustic measurements in spherical resonators, yet there is no quantitative model to explain the observed slope. If the linear dependence is due to the gas motion driving a mechanical motion of the shell or its support, then the linear dependence will extrapolate to zero at zero pressure. The small negative intercepts in the data remaining after the second-order correction is applied could have several causes. Three are mentioned here. (1) Nonzero intercepts could be caused by an error in the value of the gas's thermal conductivity used to calculate g_{t}/f. Recently, Cencek *et al.* [9] reported *ab initio* calculations of the viscosity and thermal conductivity of helium with an extremely low relative uncertainty of 2×10^{-5}. From Cencek's calculations and the recent work by May *et al.* [10], who measured the ratio of the viscosities of argon to helium and calculated the Prandtl number of argon, we estimate that the relative uncertainty in the thermal conductivity of argon is only about 3×10^{-4}. The low uncertainties in the transport properties of argon and helium affect the zero-pressure intercepts for the excess half-widths by significantly less than 0.1 ppm; therefore, they will be unimportant in this context. (2) Nonzero intercepts could be caused by an error in the pressure measurement. Because $\delta_{\mathrm{t}} \propto (\text{pressure})^{-1/2}$, an offset of 80 Pa in the pressure gauge changes the excess half-width by 0.3 ppm to 0.5 ppm for the lowest pressures in figures 1(b) and 2(b). Therefore, with high quality pressure gauges and careful measurement techniques, pressure errors are not likely to cause significant nonzero intercepts. (3) Nonzero intercepts could be caused by errors in the fitting of the acoustic data used to determine f_{m} and g_{m}. Here, we refer to systematic errors produced by a limitation of the function used to fit the resonance line shape. This important point is discussed at the end of this article.

The data in [7] and [8] are the first measurements with sufficiently high precision and quality to show clear deviations from linearity at low pressures and low frequencies, and they are the first to require the small correction presented here to restore that linearity. In the remainder of this paper, we give a brief description of our calculation. We will present a detailed derivation of second-order boundary layer corrections for spherical and cylindrical geometries in a forthcoming publication.

Starting from the linearized Navier-Stokes and continuity equations, the heat diffusion equation, and assumed $e^{i\omega t}$ time dependence, we follow the development in [5,11,12] to obtain, without further approximation, the bi-quadratic equation for the temperature oscillation $\tilde{T}$

$$\left(\nabla^2 + k_{\mathrm{ac}}^2\right)\left(\nabla^2 + k_{\mathrm{t}}^2\right)\tilde{T} = 0 \qquad (2)$$

where the wavenumbers k_{ac} and k_{t} are known exactly as functions of ω/c, the thermal diffusivity, the kinematic viscosity, and the bulk viscosity. The oscillatory fields for pressure $\tilde{p}$, density $\tilde{\rho}$, and longitudinal (curl-free) velocity $\tilde{\mathbf{u}}_l$ must satisfy same the bi-quadratic operator as the temperature. Equation (2) supports two independent wave modes: one is a propagating (acoustic) mode with a (mostly real) wavenumber k_{ac}, and the other mode is a diffusive (thermal) mode with wavenumber $k_{\mathrm{t}} \approx (1-i)/\delta_{\mathrm{t}}$ to lowest order. The divergence-free (shear wave) velocity $\tilde{\mathbf{u}}_{\mathrm{v}}$ is zero for radially-symmetric modes, which are considered here.

In spherical coordinates, equation (2) has an exact analytical solution that intrinsically includes the acoustic and thermal waves [5]. We solved equation (2) for the exact eigenfunctions that describe the radially-symmetric modes $(0,n)$ of a gas in a rigid spherical cavity with radius a, including thermal conduction near the wall. Here, we neglect thermal accommodation and assume the cavity wall has infinite thermal conductivity. When we imposed the boundary conditions on the temperature $[\tilde{T}(a) = 0]$ and on the velocity $[\hat{\mathbf{r}}\cdot\tilde{\mathbf{u}}(a) = 0]$, we obtained an expression for the resonance frequencies f_{0n} and half-widths g_{0n} as a series expansion in powers of the small parameter δ_{t}/a:

$$f_{0n} = f_{0n}^{(0)} + \Delta f_{\mathrm{t}}^{(1)} + \Delta f_{\mathrm{t}}^{(2)} + \ldots \qquad (3a)$$

$$g_{0n} = g_{\mathrm{bulk}} + g_{\mathrm{t}}^{(1)} + g_{\mathrm{t}}^{(2)} + \ldots \qquad (3b)$$

We retained terms through $O(\delta_{\mathrm{t}}/a)^2$. Our result agreed with previous results to $O(\delta_{\mathrm{t}}/a)$ and included the well-known contribution to the half-width from viscous and thermal losses in the bulk (g_{bulk}). The second-order correction due to the boundary layer reduces the half-width, but does not shift the resonance frequency $(\Delta f_{\mathrm{t}}^{(2)} = 0)$

$$\frac{g_{\mathrm{t}}^{(2)}}{f_{0n}^{(0)}} = -\tfrac{1}{2}\left(\gamma-1\right)\left(2\gamma-1\right)\left(\frac{\delta_{\mathrm{t}}}{a}\right)^2 = -\frac{2\left(2\gamma-1\right)}{\gamma-1}\left(\frac{g_{\mathrm{t}}^{(1)}}{f_{0n}^{(0)}}\right)^2. \qquad (4)$$

In practice, the excess half-widths are presented as a fraction of the measured resonance frequency f_{m} instead of a fraction of the unperturbed frequency $f_{0n}^{(0)}$ as in equation (4). If we use f_{0n}, from equation (3a), to calculate the boundary layer corrections, then we must multiply equation (4) by $\tfrac{1}{2}\left(3\gamma-1\right)/\left(2\gamma-1\right) \approx \tfrac{6}{7}$ to obtain the total second-order correction.

Using equation (4) with $\gamma = 5/3$, we corrected de Podesta's data by adding the quantity $+7(g_{\mathrm{m}}/f_{\mathrm{m}})^2$ to the half-width data in figure 1(a). The correction was largest for the (0,2) mode at 30 kPa where its value was 1.1×10^{-6}. We estimate that the approximation $g_{\mathrm{t}}^{(1)}/f^{(0)} \approx g_{\mathrm{m}}/f_{\mathrm{m}}$ introduced a systematic error of 2% or less in the magnitude of the correction for the argon data. For the helium data, we used equation (4) with values for δ_{t}/a supplied by the authors. The boundary condition that the amplitude of the temperature oscillation vanishes at the wall is approximate because the thermal conductivity of the resonator's wall is not infinite. The effect of the wall's thermal effusivity on the resonance frequencies and half-widths was taken into account in [7] according to equations (3) and (4) of [13]. For helium in the copper resonator used in [8], we estimate that penetration of the thermal wave into the wall would increase the half-width for the (0,2) mode by only 0.12 ppm independent of pressure. The increase is only 0.02 ppm for argon in a similar resonator.

The quasi-sphere used by de Podesta *et al.* is characterized by radii in the ratio $1:1+\epsilon:1-\epsilon$, where $\epsilon \approx 0.0005$. The surface area-to-volume ratio of the quasi-sphere differs from that of a perfect sphere of equal volume by a term of order ϵ^2. Thus the shape correction to g_{0n} is $O(\epsilon^2)$. In the range of the data in figure 1, $\epsilon^2 \ll g_{\mathrm{t}}^{(2)} / f_{0n}^{(0)}$; therefore, we ignored the difference between a quasi-sphere and a sphere in the calculation of equation (4).

Presently, we are conducting similar calculations for cylindrical resonators to account for terms of $O(\delta_{\mathrm{t}}/a)^2$ and the analogous term $O(\delta_{\mathrm{v}}/a)^2$, where δ_{v} is the thickness of the viscous boundary layer. The details of the calculations for the spherical and cylindrical geometries will be presented in a forthcoming publication.

Lastly, we mention that the standard resonance function (equation 2.42 in [2]) used to fit the experimental data in [7] and [8] and used by the rest of the acoustical metrology community was derived under the assumption that the resonance half-width does not change as the frequency is swept through the resonance. However, since the thickness of the boundary layer is proportional to $f^{-1/2}$, the fitting function introduces a systematic error of order $O(\delta_{\mathrm{t}}/a)^2$ in the fitted values of f_{m} and g_{m}. This fitting error is the same order as the correction calculated here and will also show up in the excess half-widths. This effect on helium data is 3 times larger than the effect on argon data from the same resonator, since δ_{t}/a is 1.7 times larger in helium than argon at the same pressure for the same mode. Further discussion about the fitting function and its limitations is beyond the scope of this paper. We are studying this effect and will address this issue elsewhere.

Acknowledgements

The author thanks Michael de Podesta for sharing his data before publication, Roberto Gavioso for reminding me of his published helium data, Michael Moldover for encouraging and supporting this work, Jim Mehl and XiaoJuan (Gloria) Feng for useful discussions, and Laurent Pitre for calling attention to the related calculation that yielded a different result in [12].

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