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Progress Towards an Acoustic/Microwave Determination of the Boltzmann Constant at LNE-INM/CNAM

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Abstract The Boltzmann constant k will be re-determined by using the simple, exact connection between the speed of sound in noble gases (extrapolated to zero pressure) and the thermodynamic temperature T, the molar mass of the gas M, and the universal gas constant R. The speed of sound will be determined in a spherical cavity of known volume V by measuring the acoustic resonance frequencies. This acoustic method led to the CODATA-recommended value of k; however, the CODATA value of k came from measurements using an almost perfectly spherical, stainless-steel-walled cavity filled with stagnant argon. The steel cavity's volume was determined by weighing the mercury of well-known density required to fill it. In contrast, a copper-walled, quasi-spherical cavity (intentionally slightly deformed from a sphere), filled with helium gas that is continuously refreshed by a small helium flow that will mitigate the effects of outgassing, will be used. The volume of the copper cavity will be determined by measuring the microwave resonance frequencies and/or by three-dimensional coordinate measurements. If the microwave method is satisfactory, the measurement of k will be based on the ratio of the speed of sound in helium—obtained by acoustic resonance measurements—to the speed of light, obtained by microwave resonance measurements. This method exploits the theorem that the frequency ratios are independent of the details of the shape of the quasi-spherical cavity. Here, progress at LNE-INM/CNAM towards a better mechanical design and better understanding of the excess of the half-widths of the acoustic and microwave measurements are reported.

Keywords Acoustic resonances · Electromagnetic resonances · Triaxial ellipsoid cavity

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1 Introduction

We report progress in a three-year-long project to re-determine the Boltzmann constant k by using acoustic techniques that improve upon those used at NIST by Moldover et al. [1] in 1988. The 1988 determination of k used a stainless-steel-walled, nearly perfect spherical cavity with stagnant argon. The volume of the cavity was determined by weighing the mercury of well-known density required to fill it. In contrast, we will use a copper-walled, quasi-spherical cavity (intentionally deformed sphere) filled with helium gas that is continuously refreshed by a small helium flow, to mitigate the effects of outgassing. The volume of this cavity will be determined by measuring its microwave resonance frequencies, and/or by three-dimensional coordinate measurements. If the microwave method is satisfactory, the measurement of k would be based on the ratio of the speed of sound in helium—obtained by acoustic resonance frequencies—to the speed of light, obtained by microwave resonance frequencies. This ratio is independent of details of the shape of the quasi-spherical cavity used. In fact, if the cavity deforms, the changes affect the acoustic and microwave frequencies is such a way that their ratios remain nearly constant [2].

The acoustic method can be used to determine the universal gas constant $R = kN_A$, where the Avogadro constant N_A has a relative standard uncertainty of 5.0×10^{-8}. In a noble gas, the speed of sound, extrapolated to zero pressure, has a simple, exact connection to the thermodynamic temperature T, the molar mass of the gas M, and the universal gas constant R. In a quasi-spherical cavity, the speed of sound can be obtained by measuring the acoustic resonance frequencies f_n of the modes with eigenvalues Z_n and by measuring the volume of the cavity V.

The project presented here is divided into six tasks:

1. design and manufacture of the resonator and the thermal enclosure,
2. determination of the volume of the quasi-spherical cavity,
3. determination of the molar mass of the gas,
4. theory and validation of the acoustic and electromagnetic corrections,
5. mapping the temperature of the cavity's wall and its stability, and
6. computing k and establishing the budget of uncertainties.

2 Experimental Setup

The apparatus is sketched in Figs. 1 and 2; however, these sketches are not to scale. For example, the length of the inlet tube was 3 m and the average radius of the quasi-spherical cavity was 5 cm. The cryostat in Fig. 1 has already been used for primary thermometry from 7 K to 273 K, as described in detail in [3]. For measuring k, minor modifications were made to decrease the cryostat's thermal time constants near 273 K. A copper rod was installed between the liquid bath and the shield; it decreased the time constant characterizing the shield's approach to the bath temperature to approximately 5 min.

2.1 The Quasi-sphere

The first task was to build a quasi-spherical cavity that more accurately approximated the cavities that have been modeled mathematically [4]. A 0.5 L copper-walled,

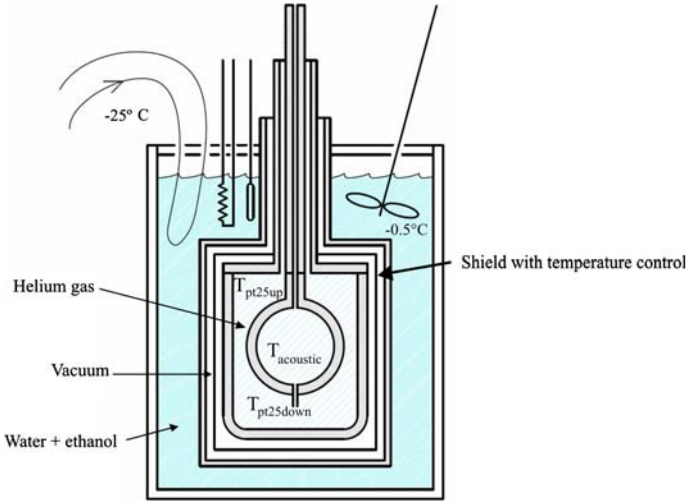

Fig. 1 Schematic diagram of the experimental setup (*Note*: the dimensions are not to scale. For example, the tube that admitted gas to the resonator was 3 m long and the average radius of the quasi-spherical cavity was 5 cm.)

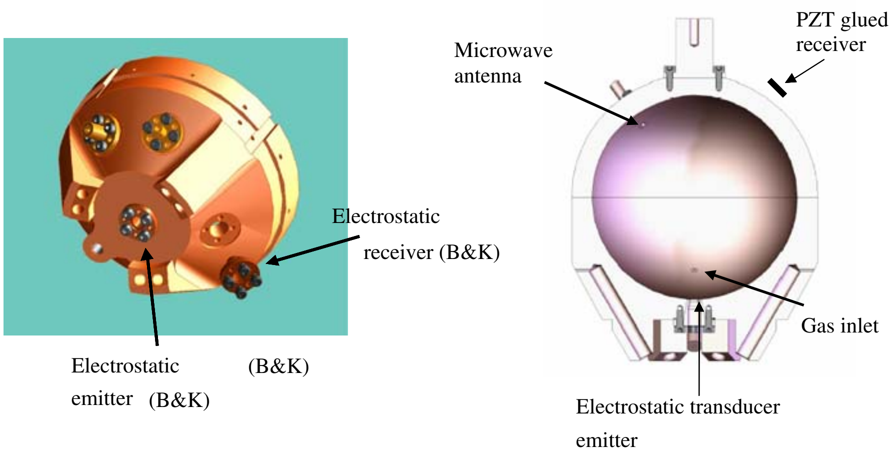

Fig. 2 Schematic drawing of the quasi-sphere indicating the positions of the Bruel and Kjaer microphones (B&K) and the external piezoelectric transducer

quasi-spherical cavity was built. However, its final shape differed from our design, in part, because the high-speed milling machine did not work as well as claimed. A skilled craftsman hand-polished the interior surfaces to a mirror finish. Although the shape of the cavity was not exactly in accord with our drawings, it was one of the best ever made; therefore, we opted to continue the measurements. When the resonator was installed in the cryostat, its temperature was stable to within 0.1 mK.

The cavity was designed to be a triaxial ellipsoid [5] with the shape,

$$\frac{x^2}{(1+\varepsilon_2)^2} + y^2 + \frac{z^2}{(1+\varepsilon_1)^2} = \frac{a^2}{(1+\varepsilon_2)^2\,(1+\varepsilon_1)^2} \tag{1}$$

with $\varepsilon_1 = 0.002$ and $\varepsilon_2 = 0.001$. These values of ε_1 and ε_2 are large enough to lift the degeneracy of the triply degenerate microwave resonances of a perfect sphere, and the expansion of the triaxial ellipsoid in spherical harmonics requires only four terms.

The microwave frequencies of the manufactured cavity were consistent with the values $\varepsilon_1 = 0.00211$ and $\varepsilon_2 = 0.00105$; however, both microwave measurements and dimensional measurements (as described in [6]) made with a coordinate measuring machine (CMM) showed that the shape of the cavity did not meet the specifications. The CMM results showed that both quasi-hemispherical cavities had an unwanted lobe that increased the volume of the 0.5 L cavity by 120 mm^3. This volume increase was easily detected by the microwave measurements. The spherical harmonic expansion of the actual shape required more than the four terms needed to represent a triaxial ellipsoid.

The walls of the cavity were made of copper. Copper's high thermal conductivity reduced the thermal gradients that might propagate inward from the cryostat to the helium in the cavity. Up to seven thermometers could be installed around the quasi-sphere to verify its temperature uniformity. The cavity's copper walls had another advantage; their high electrical conductivity at microwave frequencies reduced the penetration of the microwave fields into the walls. For the TM11 mode at 273 K, $\delta_\mathrm{m}/a = 18.71 \times 10^{-6}$, as calculated from measurements of the widths of the microwave resonances [6]. (Here, δ_m is the microwave penetration length and a is the average radius of the cavity. This ratio is only slightly larger than its theoretical ratio $(\delta_\mathrm{m}/a)_\mathrm{theory} = 18.01 \times 10^{-6}$, as calculated using the low-frequency electrical conductivity of oxygen-free high-conductivity copper.)

Conical holes were machined through the cavity's walls to admit the acoustic transducers (microphones) and the microwave antennae. These holes had a ground finish that minimized the cross-sectional area of crevices between the transducers and the conical holes surrounding them. Crevices generate perturbations to the acoustic field in the cavity that cannot be predicted without knowing many details of the holes and the transducers. As described in [3], cylindrical ducts were used to admit and remove the helium gas. However, the ducts were epoxied into conical fittings that mated with conical holes in the resonator. This made it easy to attach and remove the ducts without adding crevices.

To improve the alignment of the two hemispheres, we used a ring (not shown) with an internal diameter that mated to the outer diameters of the quasi-hemispheres. This ring permitted fine adjustments to the alignment that are not possible with the fixed locating pins that were used to align previous quasi-spheres.

2.2 The Cryostat

In our very first experiments, only two calibrated SPRTs were used [7], one at the top and at the other at the bottom, as shown in Fig. 1. After these thermometers were installed in the resonator, they indicated a temperature difference of 3.7 mK. This difference was too large to be real and was independent of the temperature of the bath and the pressure in the vessel. Because the SPRTs were surrounded by gas, we strongly suspect that the resistance of at least one of the SPRTs changed when the thermometers were installed in the resonator.

The cryostat effectively isolated the helium in the resonator from gradients and fluctuations of the temperature in the liquid bath. When the temperature of the bath was increased by 0.6 K, the relative temperature of the two thermometers changed by (0.02 ± 0.05) mK.

2.3 The Gas Handling System

The gas handling system was similar to that previously described [3,8]; however, it was improved by the addition of the capability to fix the pressure downstream of the resonator. Now, it is possible to have a constant pressure within the cavity, even when the flow is changed. For the moment, the pressure regulation performance is ± 2 Pa (peak to peak) in the range from 20 kPa to 700 kPa with high-purity helium gas.

3 Preliminary Results

3.1 Changes in the Frequencies and the Half-Widths of the Acoustic Resonances

3.1.1 Effect of the Flow

After closing the entire system, the resonator was evacuated to 10 kPa and then filled with helium to 0.5 MPa at least six times to dilute the residual air left in the resonator with helium and then to remove the helium-air mixture. After this flushing procedure, virtual leaks (or outgassing) were still a problem. To prevent the accumulation of impurities, helium continuously flowed through the resonator. To test if the flow rate was large enough, the flow rate was changed by a factor of 10. Figure 3 shows the dependence of the acoustic temperature (i.e., the temperature obtained from acoustic measurements) on the helium flow rate, in the presence of outgassing. We plot ΔT, which is defined as the temperature difference between the acoustic temperature and the temperature measured by a platinum thermometer (the latter is assumed to be independent of the gas flow rate). The values of ΔT are referred to the same arbitrary reference point. (A reference point was needed because the volume was not known and the calibrations of the platinum thermometers were in doubt.) Figure 3 shows that a flow of at least 95 $\mu\text{mol} \cdot \text{s}^{-1}$ was needed before the acoustic temperature was independent of the flow. (The half-widths of the resonances did not change when the flow changed.) Because the accumulation of contaminants is pressure-dependent, a more interesting measure of flow is the time necessary to completely replace the gas in the resonator ($\boldsymbol{\tau} = \boldsymbol{\rho V}/\dot{\boldsymbol{n}}$ where ρ is the molar density, V is the volume of the sphere, and $\dot{\boldsymbol{n}}$ is the flow rate). After three weeks of experiments with a continuous flow of pure helium, we concluded that the gas in the sphere had to be replaced at least once every 1,200 s. However, after three additional weeks, the minimum acceptable replacement rate decreased to once every 4,400 s. This experience demonstrated the

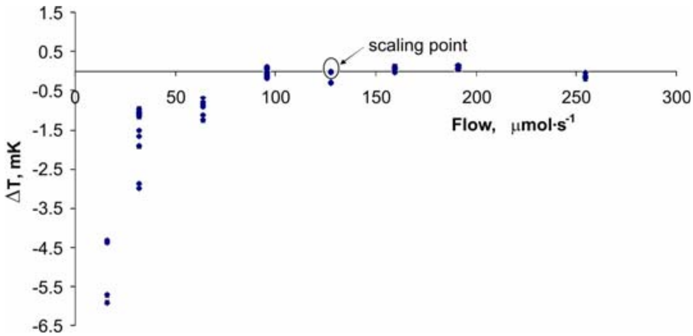

Fig. 3 Temperature differences and acoustic half-widths as a function of helium flow at 272 K and 0.5 MPa. Top: change of the acoustic temperature determined from the (0,4) mode relative to the temperature determined from one platinum thermometer

need for continuous flow and the need to ensure that the flow rate was sufficiently large to compensate for the outgassing.

3.1.2 Effect of the Voltage on the Acoustic Source

We studied the effect of changing the voltage driving the acoustic source transducer while holding constant the temperature of the resonator, the pressure in the resonator, the gas flow rate, and the polarization voltage on the source transducer. (We used a polarizing voltage of 100 V, and the driving and detected frequencies were identical.) We recorded the resistance of one platinum thermometer and the resonance frequency of the (0,4) mode while increasing the excitation voltage applied to the acoustic source transducer from 4 V_{pp} to 104 V_{pp}. (Here, V_{pp} is the peak-to-peak sinusoidal voltage applied to the acoustic source.) Although the sinusoidal voltage increased by a factor of 25, the change of difference between the acoustic temperature and the platinum temperature (0.002 ± 0.080) mK was not significant (Fig. 4).

The half-width of the (0,4) mode was independent of V_{pp} as shown in the bottom of Fig. 4. However, the uncertainty of the half-width decreased as the excitation increased until 40 V_{pp} was reached. Above 40 V_{pp}, the uncertainty was independent of V_{pp}. Therefore, another factor contributed to the uncertainty of the half-width. We suspect that this limiting factor was related to the pressure fluctuations during the flow control. To test this suspicion, we intentionally detuned the pressure feedback loop, thereby increasing the pressure fluctuations by approximately a factor of two. Simultaneously, the uncertainty of the half-width measurements increased by approximately the same factor of two. We are continuing to study this subject.

3.1.3 One Effect of the Shell Motion

A piezoelectric ceramic transducer was glued outside the resonator (Fig. 2). This transducer allowed us to detect oscillations of the coupled shell-gas system. In Rayleigh's theory, the shell of the resonator is rigid, meaning that the shell does not move in response to the oscillations of the gas. However, the piezoelectric ceramic did detect oscillations of the gas, as shown in Fig. 5. This figure displays the spectra detected

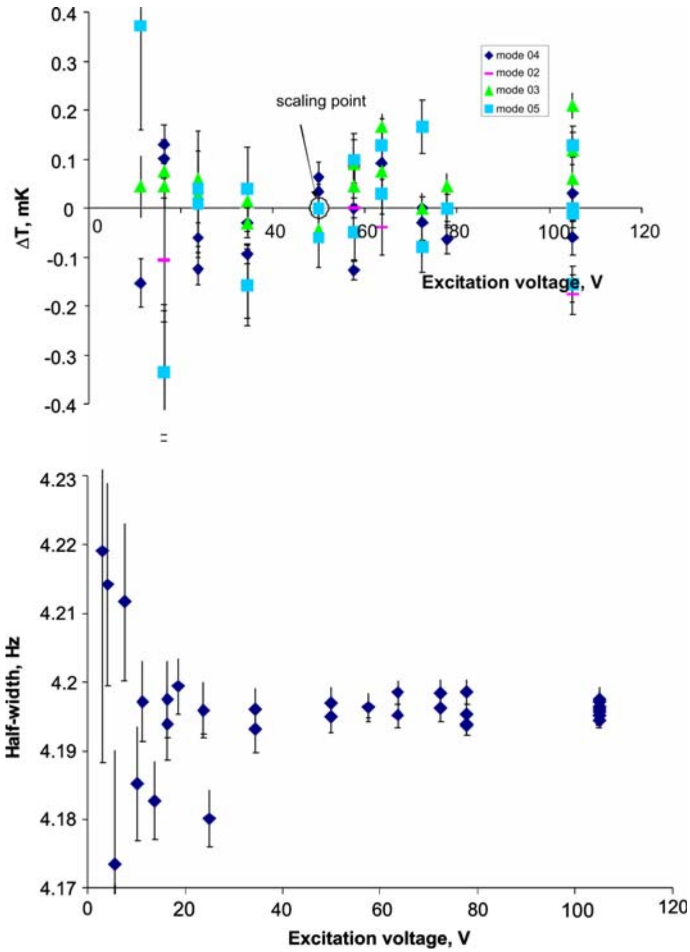

Fig. 4 Temperature differences and half-widths as a function of the sinusoidal voltage applied to the acoustic source at 272 K and 0.5 MPa

simultaneously by the microphone flush with the inner surface of the cavity and by the piezoelectric transducer while the cavity was filled with helium at 0.5 MPa and 272 K. By design, the microphone detects the oscillations of the gas and it is only weakly sensitive to the oscillations of the shell. In contrast, the signal from the piezoelectric ceramic transducer is proportional to the stress on the transducer, which is generated almost entirely by the deformation of the shell. There are at least two different types of resonances in this spectrum, the mechanical ones due to the resonator shell and those of the piezoelectric ceramic itself.

As indicated by the arrow in Fig. 5, the breathing mode of the shell is calculated to be near 18 kHz. The calculation approximates the shell as isotropic and spherical without an equatorial joint and without appendages such as bolts, ducts, supports, and thermometers. Because the appendages add mass without stiffness, they will lower

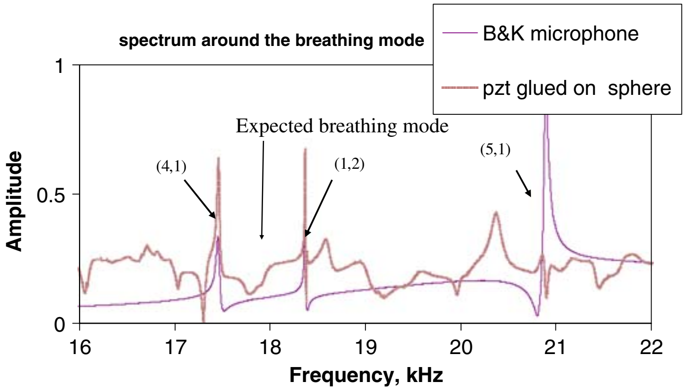

Fig. 5 Acoustic spectra near the predicted breathing mode of the shell. The shell was filled with helium at 0.5 MPa and 272 K. The spectra from a microphone and the piezoelectric (PZT) ceramic transducer were recorded simultaneously. The PZT spectrum shows many resonances. The microphone spectrum shows the non-radial modes (4,1), (1,2), and (5,1)

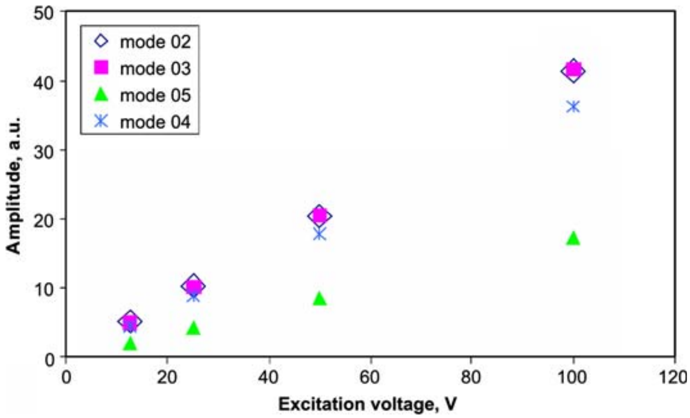

Fig. 6 Amplitude of four radially symmetric acoustic modes detected by the PZT as a function of the excitation voltage at the source transducer

the breathing frequency. Without more information, it is not possible to know which, if any, of the resonances in Fig. 5 is the breathing mode.

Figure 6 shows that the maximum amplitude of the modes (0,2) to (0,5) is a linear function of the excitation voltage. Although the interaction between the fluid and the shell is complex, Fig. 4 shows that even when the amplitude of the shell's motion is increased by a factor of eight, neither the acoustic temperature (i.e., the resonance frequency) nor the half-width change at the level of (0.002 ± 0.080) mK.

Because of the complex spectrum of shell vibrations, and also because helium has a relatively large acoustic second virial coefficient, it might be better to extrapolate to zero pressure at constant frequency instead of following the current practice of extrapolating to zero pressure at constant temperature. The constant frequency extrapolation can be accomplished on a quasi-isotherm by following each pressure change with a temperature change that returns a specific acoustic resonance, for example, the (0,3) mode, to the same frequency. Then, one would correct the resonance frequency for the temperature change required to extrapolate to zero pressure at constant temperature.

3.2 Half-Width in Microwave and Frequency Change

As described in [3], the small cavity for the microwave antenna can perturb the acoustic field, unless the cavity is filled with a material that is opaque to the acoustic field and transparent to the microwave field. For expediency, Pitre et al. [3] filled the cavity with vacuum grease. However, the grease used is soft at 273 K and might migrate during months of measurements. Here, we used Stycast 2850 FT to fill the small cavity with the microwave antenna. This epoxy is easy to use; it has the same coefficient of thermal expansion as copper, and it has a low rate of outgassing (less than $4 \times 10^7\,\mathrm{Pa \cdot L \cdot s^{-1} \cdot cm^{-2}}$). We measured the microwave half-widths and the frequencies before and after the antenna cavity was filled with this epoxy. For the lowest four microwave triplets, the fractional changes of the complex frequencies multiplied by 10^6 were: TM11, $-1.25 + 0.25i$; TE11, $-0.68 + 0.11i$; TM12, $-0.30 + 0.39i$; and TE12, $-0.31 + 0.37i$. The microwave resonance frequencies can be measured with a fractional resolution of 10^{-8}; therefore, the resonance measurements easily detected the effect of the epoxy. For the determination of the Boltzmann constant, we plan to begin with a quasi-sphere with only two holes for the two microwave antennas, and, step by step, to add all the other transducers and holes. By doing this we will be able to measure the changes of resonance frequency and half-width generated by every small perturbation.

4 Conclusion

This progress report presents the work completed very recently at the LNE-INM/CNAM. In this first report, we have started to characterize the quasi-sphere from an acoustic point of view. Minor experimental difficulties (one wire used to heat

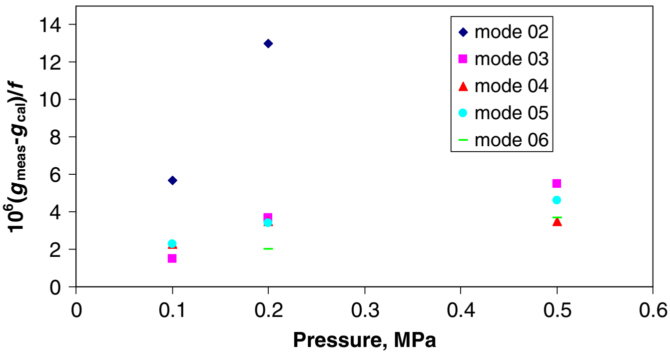

Fig. 7 Excess half-widths of five radially symmetric acoustic modes as a function of pressure

the pressure vessel broke, the thermometer was not stable, and pressure regulation not at its optimum) prevented us from measuring an isotherm that would permit us to re-determine the Boltzmann constant. Nevertheless, the cryostat was stabilized at three different pressures and the acoustic modes (0,2), (0,3), (0,4), (0,5), (0,6), (1,2), (1,3), (1,4), and (1,5) were recorded. (The non-radial (1,*n*) modes will be discussed elsewhere.) Upon extrapolation to zero pressure, the excess of the half-width of the acoustic radial modes does not exceed 3 ppm as shown in Fig. 7. With this 0.5 L sphere, a determination of the Boltzmann constant is scheduled before the end of 2008 and an uncertainty of approximately 10 ppm seems achievable. At the same time, the development of another 0.5 L sphere is planned, which will have a shape conforming to the initial drawing. This will allow us to reach a far lower uncertainty.

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