Can an ideal gas feel the shape of its container?

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Thermodynamic quantities of an ideal gas enclosed in a finite container are examined. We use an asymptotic expansion for high temperatures to obtain the partition function of an ideal gas, both in two and three dimensions, showing the leading corrections to the internal energy due to a finite container. In the three-dimensional case, the first correction term depends only on the area-volume ratio, but higher order terms depend also on other geometric properties of the container. However, according to recent results, we show that the answer to the question posed in the title is negative. © 1997 American Association of Physics Teachers.

I. INTRODUCTION

In this paper we are interested in examining how the thermodynamic properties of an ideal gas are influenced by the geometry of its container. Perhaps it is not evident, but this problem is closely related to the one formulated by M. Kac as: "Can one hear the shape of a drum?"¹ Both problems can be reduced to the study of the distribution and behavior of the eigenvalues of the Helmholtz equation,

$$\nabla^2 \phi(\mathbf{r}) + \lambda \phi(\mathbf{r}) = 0, \tag{1}$$

with Dirichlet boundary conditions ($\phi=0$ on the boundary) over a domain of defined geometry.

Thermodynamic properties of an ideal gas can be extracted from the partition function

$$\mathcal{Z} = \frac{z^N}{N!},\tag{2}$$

where N is the number of particles and z is

$$z = \sum_{\{\text{states}\}} \exp(-\beta E_n), \qquad (3)$$

with $\beta = (k_B T)^{-1}$, k_B is Boltzmann's constant, and T is the absolute temperature. The eigenvalues (energy levels) of one particle, E_n , are obtained from the stationary states $\psi(\mathbf{r},t) = \phi(\mathbf{r})e^{-iEt/\hbar}$ of the time-dependent Schrödinger equation,

$$-\frac{\hbar^2}{2m}\nabla^2\psi + V(\mathbf{r})\psi = i\hbar \frac{\partial\psi}{\partial t}$$
(4)

with $V(\mathbf{r})=0$. Thus $\phi(\mathbf{r})$ obey the Helmholtz equation (1) with $\lambda = 2mE/\hbar^2$ and Dirichlet boundary conditions.

In order to show the analogy with Kac's problem, let us consider the small oscillations of a field $\varphi(\mathbf{r},t)$ described by the wave equation

$$\nabla^2 \varphi - \frac{1}{c^2} \frac{\partial^2 \varphi}{\partial t^2} = 0, \tag{5}$$

where φ could be, for example, the displacement of a membrane, the vibrations of the electromagnetic field or the pressure of a fluid. If we look for periodic solutions (normal modes) of the form $\varphi(\mathbf{r},t) = \phi(\mathbf{r})e^{-i\omega t}$ in Eq. (5), we obtain the Helmholtz equation (1) with $\lambda = (\omega/c)^2$. The eigenvalues λ_n depend both on the boundary conditions (Dirichlet or Neumann²) and on the shape of the boundary. Inversely, from the complete knowledge of the eigenvalues one may extract information on the geometry of the boundary (for a given boundary condition). This can be done by analyzing the asymptotic expansion of the sum

$$S(t) = \sum_{n=1}^{\infty} \exp(-\lambda_n t) \quad \text{for} \quad t \to 0,$$
(6)

which formally is the same as the one-particle partition function, Eq. (3). Precisely the link between both problems lies here.

A common practice in statistical mechanics textbooks³ is to transform the sum over states (3) into an integral, step which is justified by saying that it is correct "in the limit of the mean de Broglie wavelength being much, much smaller than the dimensions of the container." Recently M. I. Molina,⁴ discussing the validity of such transformation, showed approximately the main correction to the thermodynamic quantities in two particular cases: ideal gas in a box and ideal gas in a sphere.

The purpose of this paper is to use a well-known asymptotic expansion of the sum (6) to derive a general expression for the corrections to the thermodynamic quantities, particularly the energy for an ideal gas due to a large, but yet finite, container volume.

The paper is organized as follows: Section II historically reviews the problem, Sec. III presents the calculation of the internal energy for the ideal gas in the cases of dimension two and three. Finally, Sec. IV summarizes our conclusions and remarks.

II. HISTORICAL BACKGROUND

The problem of the behavior of normal modes in a cavity, that is, basically counting the normal modes, has an interesting history, which has attracted both physicists and mathematicians since the latter half of the nineteenth century. For example, as early as 1882, Arthur Schuster⁵ posed the problem and remarked on its importance.

According to Kac,¹ the problem of counting normal modes in a cavity originated in 1910, when H. A. Lorentz was invited to Göttingen to deliver the Wolfskell Lectures. During the fourth lecture, he presented his work with J. H. Jeans about the characteristic frequencies of the electromagnetic field in a three-dimensional resonant cavity of volume V, giving the following expression for the number $N(\lambda)$ of eigenvalues with less than a given wavelength λ , for certain simple geometries of the cavity:

$$N(\lambda) \approx \frac{V}{6\pi^2} \lambda^{3/2}$$
 when $\lambda \to \infty$. (7)

He posed to the mathematicians there present the problem of proving that this expression is independent of the geometry (the shape) of the cavity, and hence depends only upon the volume. Hilbert, who was in the audience, predicted that this conjecture would not be solved during his lifetime. Surprisingly H. Weyl, a former student of his, only 2 years later was able to prove, using integral equations techniques, the following result:

$$\lim_{\lambda \to \infty} \frac{N(\lambda)}{\lambda^{D/2}} = \frac{V}{(2\pi)^D} B_D, \qquad (8)$$

where *D* is the dimension and $B_D = \pi^{D/2} / \Gamma(D/2+1)$ the volume of the unit sphere in *D* dimensions.⁶

Now, it is known that the study of the above limit is equivalent¹ to the study of the behavior of the following sum, when $t \rightarrow 0$:

$$S(t) = \sum_{n=1}^{\infty} \exp(-\lambda_n t), \qquad (9)$$

where λ_n are the eigenvalues of $-\nabla^2$. Using the result of Weyl in two dimensions, we obtain, when $t \rightarrow 0$,

$$S(t) \approx \frac{A}{4\pi t},\tag{10}$$

where A is the area of the domain. In 1954 Å. Pleijel⁷ obtained, for the case of Dirichlet boundary conditions, a second term for (10), which reads

$$S(t) \approx \frac{A}{4\pi t} - \frac{L}{4} \frac{1}{\sqrt{4\pi t}} \quad \text{for} \quad t \to 0,$$
(11)

where *L* is the perimeter of the domain. Note that due to the classical isoperimetric inequality $L^2 \ge 4\pi A$, it follows that once *S*(*t*) is known (i.e., all eigenvalues are known), it is possible to decide whether the domain is or is not a circle.

The next step toward the solution of this problem was taken by Kac, who together with L. Bers reformulated it under the following suggestive question: Can one hear the shape of a drum?¹ Kac conjectured, using probability techniques, that for a "drum" (i.e., with fixed edges) with smooth borders the sum (9) becomes

$$S(t) \approx \frac{A}{4\pi t} - \frac{L}{4} \frac{1}{\sqrt{4\pi t}} + C \text{ for } t \to 0,$$
 (12)

where $C = \frac{1}{6}(1-r)$ and r is the number of holes in the domain.

Later, in 1967, McKean and Singer⁸ proved Kac's conjecture and showed a general expression for Riemannian manifolds, obtaining the following expression for C in the case of a "polygonal drum":

$$C = \sum_{i=1}^{n} \frac{\pi^2 - \gamma_i^2}{24\pi\gamma_i},$$
 (13)

where *n* is the number of corners and γ its corresponding interior angles.

Applying the results of McKean and Singer to domains with smooth borders, R. Waechter⁹ obtained, for three dimensions (with Dirichlet boundary conditions), the following expansion of the sum (9):

$$S(t) \approx \frac{V}{(4\pi t)^{3/2}} - \frac{A}{16\pi t} + \frac{M}{6\pi (4\pi t)^{1/2}} + \frac{J}{512\pi}, \qquad (14)$$

where M is the surface integral of mean curvature

$$M = \int_{S} \frac{1}{2} (\kappa_1 + \kappa_2) dS, \qquad (15)$$

with κ_1 and κ_2 the principal curvatures at the surface element dS on the body surface, and

$$J = \int_{S} (\kappa_1 - \kappa_2)^2 dS.$$
(16)

In summary,¹⁰ today we know the first five a_i in the following expression of the sum (9) for domains with smooth borders¹¹ in *D* dimensions:

$$S(t) = \frac{1}{(4\pi t)^{D/2}} \sum_{i=0}^{k} a_i t^{i/2} + O(t^{(k-D+1)/2}),$$
(17)

where $O(\cdot)$ is the order of the approximation.¹² In the Appendix we show how the first term of this formula can be obtained in the case of a rectangular parallelepiped, using properties of Jacobi's theta function.

Finally, let us add that the initial question raised by Kac was answered negatively in 1992 by Gordon, Webb, and Wolpert,¹³ who showed explicitly two domains that although having different shapes, have the same eigenvalues (i.e., isospectral domains). Notably, this theoretical result was experimentally verified recently¹⁴ by employing thin microwave cavities shaped in the form of two different domains known to be isospectral.

III. PARTITION FUNCTION FOR AN IDEAL GAS

Consider an ideal monatomic gas composed of N identical particles of mass m, enclosed in a box of volume V, at a temperature T. We talk about an "ideal gas" when the particles are noninteracting and

$$\lambda_{\rm dB} \left(\frac{N}{V}\right)^{1/3} \ll 1,\tag{18}$$

where $\lambda_{dB} = \hbar/\sqrt{2mk_BT}$ is the de Broglie wavelength and \hbar is the Planck constant divided by 2π . Standard derivation of the partition function for a *D*-dimensional ideal gas³ enclosed in a box with sides $a_1, a_2, ..., a_D$ begins with the calculation of the eigenvalues of the Schrödinger/Helmholtz equation with Dirichlet boundary conditions

$$\boldsymbol{\epsilon}_{n_1,n_2,\dots,n_D} = \frac{\hbar^2 \, \pi^2}{2m} \left[\left(\frac{n_1}{a_1} \right)^2 + \left(\frac{n_2}{a_2} \right)^2 + \dots + \left(\frac{n_D}{a_D} \right)^2 \right]. \tag{19}$$

Assuming a gas temperature (density) high (low) enough to apply Boltzmann's statistics, the partition function is given by (2) with

$$z = \prod_{i=1}^{D} \sum_{n_i=1}^{\infty} \exp\left(-\frac{\hbar^2 \pi^2}{2ma_i^2} \beta n_i^2\right).$$
(20)

Transforming the sum (20) into an integral, in the case of a box of side a, we obtain the well-known result

$$\mathscr{Z} = \frac{1}{N!} \frac{1}{2^{ND}} \left[\frac{2ma^2}{\pi\hbar^2 \beta} \right]^{ND/2}.$$
 (21)

In general, when calculating the partition function, the expression one is interested in is Eq. (3). When transforming the sum (3) into an integral one introduces the *density of states* $\rho(E)$. For example, the usual expression for $\rho(E)$ in three dimensions, namely, $\rho(E) = (V/4\pi^2)(2m/\hbar^2)E^{1/2}$, is



Fig. 1. Correction terms of Eq. (25) in the case of argon, for regular polygons from 3 to 12 sides, and a circle. The dots correspond to an area $A = 0.1 \text{ m}^2$ and the diamonds to an area $A = 1 \text{ m}^2$.

obtained by counting the number of states of the Helmholtz equation in a cube. However, this behavior of $\rho(E)$ is the leading behavior of $\rho(E)$ for any bounded domain. Strictly speaking, the density of states $\rho(E)$ will depend on the actual shape of the domain.¹⁵ Following the discussion of Sec. II, we can obtain further information about the shape of the domain by studying the asymptotic value of sum (3) when β goes to zero (i.e., $T \rightarrow \infty$, the ideal gas case). Noting that the eigenvalue problem of the Schrödinger equation is the same as the eigenvalue problem of the wave equation with Dirichlet boundary conditions, we can use directly the expressions of Sec. II, replacing t by $(\hbar^2/2m)\beta$.

In the next subsections, we are going to examine the cases of dimensions two and three, using the asymptotic expansion of sum (3). This allows us to estimate the correction to the standard formula in the case of a finite container and helps us to partially answer the question posed in the title.

A. Ideal gas in two dimensions

Let us now consider the general partition function (2) in two dimensions. Using directly Kac's expansion (12) with $\beta \rightarrow 0$, Eq. (3) gives

$$z(\beta) \approx \left(\frac{2m}{\hbar^2}\right) \frac{A}{4\pi\beta} - \left(\frac{2m}{\hbar^2}\right)^{1/2} \frac{L}{4(4\pi\beta)^{1/2}} + C.$$
(22)

We set out to apply this formula to thermodynamic quantities such as the internal energy $U = -(\partial \ln(\mathcal{Z})/\partial\beta)_{V,N}$, the pressure $P = \beta^{-1}(\partial \ln(\mathcal{Z})/\partial V)_{T,N}$, and the specific heat $C_v = (\partial U/\partial T)_{V,N}$, among others. In the case of the internal energy we obtain



Fig. 2. Isospectral domains in two dimensions (see Ref. 24).

$$U \approx -N \frac{\partial}{\partial \beta} \ln\left(\left(\frac{2m}{\hbar^2}\right) \frac{A}{4\pi\beta} - \left(\frac{2m}{\hbar^2}\right)^{1/2} \frac{L}{4(4\pi\beta)^{1/2}} + C\right).$$
(23)

Now, differentiating, expanding in powers of $\beta = (k_B T)^{-1}$, and using the definition of the thermal wavelength Λ ,

$$\Lambda(T) = \sqrt{\frac{2\pi\hbar^2}{mk_B T}},\tag{24}$$

we obtain

$$U(T) = Nk_B T \left[1 + \frac{1}{8} \frac{L}{A} \Lambda(T) - \left(C - \frac{1}{32} \frac{L^2}{A} \right) \frac{1}{A} \Lambda^2(T) + O(\Lambda^3(T)) \right].$$
(25)

Similar expressions hold for the pressure and the specific heat. It is interesting to note that, in this expression, the first correction term

$$\frac{1}{8}\frac{L}{A}\Lambda(T) \tag{26}$$

is independent of the constant C (related to the smoothness of the perimeter).

As an illustration of the above expression, let us particularize the internal energy to the case of an n-sided regular polygon of fixed area A and apply it to argon at standard conditions.

In this case, the perimeter-area relation is given by

$$\frac{L^2}{A} = 4n \, \tan\left(\frac{\pi}{n}\right),\tag{27}$$

whereas the constant C in Eq. (13) is

$$C = \frac{1}{6} \frac{(n-1)}{(n-2)}.$$
(28)

Table I. Expressions of the terms V, A, M and J of Eq. (29) for a rectangular parallelepiped of sides a_1 , a_2 , a_3 , a cylinder of length l and radius r and a sphere of radius R.

	V	Α	М	J
Parallelepiped	$a_1 a_2 a_3$	$2(a_1a_2+a_1a_3+a_2a_3)$	$\frac{3}{2}\pi(a_1+a_2+a_3)$	-64π
Cylinder	$\pi r^2 l$	$2\pi r(r+l)$	$\frac{\pi}{2}(2l+3\pi r)$	$2\pi \left(\frac{l}{r}-\frac{64}{3}\right)$
Sphere	$\frac{4}{3}\pi R^3$	$4\pi R^2$	$4\pi R$	0



Fig. 3. Correction terms of Eq. (30) in the case of argon, for a cube, a cylinder whose length is twice the radius, and a sphere. The dots correspond to a volume V=0.1 m³, the squares to a volume V=0.5 m³, and the diamonds to a volume V=1 m³.

It is interesting to see what happens in the ideal gas regime with the correction terms (i.e., how sensitive the gas is to "feel" the shape of its container) when we decrease or increase the area A (volume V). The de Broglie wavelength λ_{dB} associated with argon at standard conditions is $\lambda_{dB} \approx 0.045$ Å, and the specific volume is 22.41 m³/kmol, giving a typical interatomic distance of about l=30 Å. Assuming the requirements for a dilute and noninteracting gas when l<10 Å are no longer valid, we can vary, for example, from A=1 m² (V=1 m³) to A=0.1 m² (V=0.1 m³).

Figure 1 shows the corrections to the energy for different regular polygons, from 3 to 12 sides, and a circle, with areas $A = 1 \text{ m}^2$ (diamonds) and $A = 0.1 \text{ m}^2$ (dots). The corrections with respect to an ideal gas are very small, the first-order correction being about 10^{-12} and the next corrections still smaller. The corrections are smallest in the case of a circle and largest in the case of a triangle, which is expected due to the isoperimetric inequality. The difference between the circle and triangle corrections increases as the area decreases.

Of course, according to the results of Gordon, Webb, and Wolpert,¹³ there are domains where, although different in shape, the thermodynamic properties of an ideal gas will be exactly the same, independent of the order of the approximation in (25). An example of this type of domain in shown in Fig. 2. In this sense, an ideal gas in two dimensions cannot feel the shape of its container, although it can "feel" some geometric properties.

B. Ideal gas in three dimensions

Let us go to a more realistic physical example: an ideal gas in three dimensions. Here we need to replace Kac's formula (12) by Waechter's result (14), obtaining

$$z(\beta) \approx \left(\frac{2m}{\hbar^2}\right)^{3/2} \frac{V}{(4\pi\beta)^{3/2}} - \left(\frac{2m}{\hbar^2}\right) \frac{A}{16\pi\beta} + \frac{1}{6\pi} \sqrt{\frac{2m}{\hbar^2}} \frac{M}{(4\pi\beta)^{1/2}} + \frac{J}{512\pi}.$$
 (29)

For the internal energy, after differentiating, expanding in powers of $\beta = (k_B T)^{-1}$, and using the definition of the thermal wavelength Λ , we obtain

$$U(T) = \frac{3}{2} N k_B T \left[1 + \frac{1}{12} \frac{A}{V} \Lambda(T) - \frac{1}{6} \left(\frac{2}{3\pi} - \frac{1}{8} \frac{A^2}{MV} \right) \right]$$

Note that the first correction term of this expansion only depends, aside from $\Lambda(T)$, on the area-volume relation, having the same functional form for any container geometry.¹⁶

The values of V, A, M, and J for a rectangular parallelepiped, a cylinder, and a sphere are given in Table I. From Minkowski's isoperimetric inequalities, $M^2 \ge 4\pi A$ and $A^2 \ge 3VM$, we can estimate the corrections for different figures. Thus, for a fixed volume (or area, or M), the first correction is smallest in the case of a sphere.

We can particularize these results to the case of argon in the ideal gas regime. The corrections to the energy for three different shapes of its container are shown in Fig. 3. Again, as in the two-dimensional case, only the first-order correction term of the expansion is at all significant, although very small, increasing when the volume decreases.

Finally, it is noteworthy that, also in the three-dimensional case, an ideal gas cannot feel the shape of its container, although some geometrical properties can be "felt." In fact, one could build a box with isospectral cross-sectional shapes, as done in Ref. 14 with the surfaces shown in Fig. 2, and fill it with gas. In this case, in spite of the fact that the shapes are different, the expressions V, A, M, and J of Eq. (29) (and all higher order corrections) will be exactly the same, so the thermodynamic properties will not show any differences.

IV. CONCLUDING REMARKS

In this work we have investigated the influence of a finite container on the thermodynamic quantities of an ideal gas. The calculations are based on well-known results from the asymptotic properties of the spectrum of the Laplacian. The energy is obtained, both in two and three dimensions, as an expansion in powers of the thermal wavelength, whose coefficients depend on the geometric properties of the container.

Thus, in principle, an ideal gas could "feel" some aspects of the shape of its container, because its thermodynamic quantities depend on geometric properties such as the volume, area, principal curvature ratios, and so on. Actually, the identification of these in Eq. (30) is not as direct as in Eq. (14). However, a practical example with argon shows that the corrections are very small, so in practice only the firstorder correction is significant. In other words, this fact reinforces the accepted idea that the standard derivation of the partition function of an ideal gas is a very good approximation.

As a final remark, let us say that recently the existence of isospectral domains in two dimensions has been demonstrated, both theoretically and experimentally. By this means, one can also build three-dimensional isospectral domains. Hence, an ideal gas, even with all terms in the expansion of the partition function completely known, is not able to discriminate in these cases between two different shapes. We can conclude that, strictly speaking, the answer to the question posed in the title is negative.

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APPENDIX

Following closely the method employed in Ref. 17, we will show, as an example, how to calculate the asymptotic expansion (9) for a box with sides a_1 , a_2 , and a_3 [see also Ref. 9, Ref. 18 (and references therein), and Ref. 19]. The sum S(t) reads

$$S(t) = \sum_{k=1}^{\infty} e^{-\lambda_k t} = \prod_{i=1}^{3} \sum_{n_i=1}^{\infty} e^{-\pi^2 n_i^2 t/a_i^2}$$
$$= \prod_{i=1}^{3} \frac{1}{2} \left(\theta \left(\frac{\pi t}{a_i^2} \right) - 1 \right),$$

where $\theta(\cdot)$ is Jacobi's theta function.

As the theta function satisfies Jacobi's identity $\theta(t) = (1/\sqrt{t}) \theta(1/t)$ (see Ref. 20), we get

$$S(t) = \frac{1}{8} \prod_{i=1}^{3} \left(\frac{a_i}{\sqrt{\pi t}} \ \theta \left(\frac{a_i^2}{\pi t} \right) - 1 \right).$$

Also, we have

$$\theta\left(\frac{1}{x}\right) = 1 + 2\sum_{k=1}^{\infty} e^{-k^2 \pi/x} \le 1 + 2\sum_{k=1}^{\infty} e^{-k \pi/x} = 1 + O(e^{-\pi/x}).$$

Thus it is clear that

$$S(t) = \frac{a_1 a_2 a_3}{(4 \pi t)^{3/2}} - \frac{2(a_1 a_2 + a_2 a_3 + a_1 a_3)}{16 \pi t} + \frac{(a_1 + a_2 + a_3)}{4(4 \pi t)^{1/2}} - \frac{1}{8} + \exp. \text{ small.}$$

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