

Presence of negative entropies in Casimir interactions

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Abstract

Negative entropy in connection with the Casimir effect at uniform temperature is a phenomenon rooted in the circumstance that one is describing a nonclosed system, or only part of a closed system. In this paper we show that the phenomenon is not necessarily restricted to electromagnetic theory, but can be derived from the quantum theory of interacting harmonic oscillators, most typically two oscillators interacting not directly but indirectly via a third one. There are two such models, actually analogous to the transverse magnetic (TM) and transverse electric (TE) modes in electrodynamics. These mechanical models in their simplest version were presented some years ago, by J. S. Høye et al., *Physical Review E* **67**, 056116 (2003). In the present paper we re-emphasize the physical significance of the mechanical picture, and extend the theory so as to include the case where there are several mediating oscillators, instead of only one. The TE oscillator exhibits negative entropy. Finally, we show explicitly how the interactions via the electromagnetic field contain the two oscillator models.

I. INTRODUCTION

Let us begin by recapitulating the conventional procedure for calculating the Casimir force between two dielectric media, typically two half-spaces separated by a gap a : One starts from the two-point function for the electric field at two neighboring spacetime points, usually by using the fluctuation-dissipation theorem assuming uniform temperature, and then uses Maxwell's stress tensor to calculate the surface pressure, here called P . Then, the free energy F per unit surface can be found by integration of $P = -\partial F/\partial a$, and the internal energy U per unit area follows from the thermodynamical formula

$$U = \frac{\partial(\beta F)}{\partial\beta}, \quad (1)$$

with $\beta = 1/(k_B T)$. The corresponding entropy S then finally follows from

$$S = \frac{U - F}{T} = -\frac{\partial F}{\partial T}. \quad (2)$$

This procedure is considered in detail at various places, for instance in the standard sources [1–3].

The following point here calls for attention: The theory is based upon the electrodynamics of a *non-closed* physical system. That is, the force is calculated from the difference in the electrodynamic stress tensor between the inside and the outside of a dielectric medium. The properties of the medium itself are not accounted for. The fact that we are dealing with an electromagnetic subsystem makes it not so unreasonable that we can encounter unexpected properties when calculating physical properties of the subsystem such as the Casimir entropy.

Consider for definiteness the two-slab system above, assuming the separation a to be constant. We let the temperature T increase, from zero upwards. We further assume the standard Drude dispersion relation

$$\varepsilon(i\zeta) = 1 + \frac{\omega_p^2}{\zeta(\zeta + \nu)}, \quad (3)$$

where ν is the dissipation parameter. As has been shown in detail by explicit calculations, as long as ν is different from zero as always is the case for a real material, the slope $\partial F/\partial T$ is *zero* at $T = 0$ [4–6]. That is, the Nernst theorem is satisfied for the Casimir entropy. We ought to emphasize this point, because assertions to the contrary have often appeared in

the literature. An ambiguity might occur only if the parameter ν were exactly zero, which is, however, only a fictitious case.

Then for increasing temperature the free energy starts to increase while for high temperatures it decreases in the usual way. This increase means that the entropy $S = -\partial F/\partial T$ becomes negative in this region. This special property has been subject to several studies recently; cf., for instance, Refs. [7, 8] with further references therein.

In particular, for high temperature (with the separation a fixed) the TE contribution is negative, but tends to zero. This means that the TE entropy is negative; in fact, it typically is always negative for all values of T . Whether the total entropy is negative depends on the balance with the TM entropy, which is typically (but not necessarily) positive. More often than not, there is a region of low temperature when the total entropy goes negative.

Most previous studies have considered the negative entropy problem from the standpoint of electrodynamics. This is quite a natural approach, as the effect is related to the circumstance that the relationship between canonical momentum \mathbf{p} for a particle with mass m and charge q and the electromagnetic vector potential $\mathbf{A}(\mathbf{r}, t)$ is $(\mathbf{p} - q\mathbf{A})^2/2m$ (as is known, this is the reason for the absence of classical diamagnetism, the Bohr-van Leeuwen theorem¹). It is, however, possible to describe this effect in a different way which is simpler and does not involve electromagnetism explicitly, namely as an interaction between two quantum mechanical harmonic oscillators 1 and 2, mediated indirectly via a third oscillator 3. Actually we presented this oscillator model in an earlier paper (cf. Sec. IV in Ref. [4]), but it seems that this model has been left largely unnoticed. And then we have come to the main motivations for the present paper:

- to re-emphasize the physical significance of the oscillator model;
- to generalize the theory so as to encompass the case where there are many interacting oscillators, similar to the electromagnetic field, instead of only one;
- to provide a general proof that the TE entropy is negative for high temperature.

¹ However, recall that the Langevin construction gives a reasonable model of diamagnetism for dielectrics—see, for example, Ref. [9].

II. TWO HARMONIC OSCILLATOR MODELS

As mentioned above, we assume the validity of the Drude dispersion relation (3), as this is the most physical one. The competing dispersion relation, the plasma relation, corresponds to setting $\nu = 0$. The introduction of our harmonic oscillator model in Ref. [4] was actually motivated by the current discussion about choosing between the Drude/plasma relations. There are actually two different oscillator models, corresponding to the TM and TE modes of the analogous electromagnetic theory.

Consider first the classical partition function of a harmonic oscillator with energy

$$H = \frac{1}{2m}p^2 + \frac{1}{2}m\omega^2x^2 \quad (4)$$

where x is the position, p is the momentum, ω is the eigenfrequency, and m is the mass. Integrating both momentum and position the classical partition function is found to be

$$Z = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{-\beta H} dp dx = \frac{1}{\hbar\beta\omega} \quad (5)$$

This gives the free energy and its frequency dependency as

$$F = -\frac{1}{\beta} \ln Z = -\frac{1}{\beta} \ln \left(\frac{1}{\hbar\beta\omega} \right) \sim \ln \omega. \quad (6)$$

Thus for three non-interacting harmonic oscillators the inverse partition function is proportional to \sqrt{Q} where

$$Q = a_1 a_2 a_3, \quad a_i = \omega_i^2 \quad (i = 1, 2, 3). \quad (7)$$

By quantization using the path integral method [10, 11], the classical system turns out to be split into a set of classical harmonic oscillator systems described by Matsubara frequencies. Then for each Matsubara frequency expression (7) is replaced by

$$Q = A_1 A_2 A_3, \quad A_i = \omega_i^2 + \zeta^2 = a_i + \zeta^2, \quad (8)$$

where $\zeta = i\omega$. (Depending upon convention $\zeta = -i\omega$ is often used.)

Assume now that there is no direct interaction between oscillators 1 and 2. The interaction between them is mediated entirely by oscillator 3, which can be imagined to be situated in an intermediate position. For simplicity we assume all oscillators one-dimensional. The

interaction can now be represented as cx_ix_j where x_i and x_j are coordinates and c a coupling constant. With this the quantity Q becomes

$$Q = \begin{vmatrix} A_1 & 0 & c \\ 0 & A_2 & c \\ c & c & A_3 \end{vmatrix} = A_1A_2A_3 - c^2(A_1 + A_2) \\ = A_1A_2A_3(1 - D_1)(1 - D_2) \left(1 - \frac{D_1D_2}{(1 - D_1)(1 - D_2)} \right), \quad (9)$$

where

$$D_i = \frac{c^2}{A_iA_3} \quad (i = 1, 2). \quad (10)$$

The quantum free energy F is obtained by summing over the Matsubara frequencies $K = \hbar\zeta = i\hbar\omega = 2\pi n/\beta$ with n integer

$$\beta F = \lim \frac{1}{2} \sum_n \ln Q(\zeta) \quad (+\text{const.}). \quad (11)$$

Here \lim refers to the limit of a discretization procedure. As pointed out in Ref. [4] this must be carefully defined as in Ref. [10] to obtain correctly the well-known result for F . However, we can skip this discussion here as only the last factor of (9) is of interest. The product $A_1A_2A_3$ represents the three non-interacting oscillators. Further the $A_i(1 - D_i)$ ($i = 1, 2$) represent each of the two oscillators with their radiation reaction via the third oscillator. Finally the last factor represents the induced Casimir energy.

The above model represents the situation analogous to the *TM mode*. To model the TE mode we will need another model, which is the analogue to the electromagnetic interaction where the third oscillator interacts with the *momenta* p_i of the other two, i.e., the interaction $(p_i - \text{const} \times x_3)^2/2m_i$ [m_i is the mass, $i = 1, 2$]). By evaluation of the classical partition function one now will find that the interaction has no influence upon thermal equilibrium (as mentioned, this is the analog of classical diamagnetism which is equal to zero). Quantum mechanically the problem is less straightforward. But we can simplify the calculation by exchanging the roles of momenta and coordinates of the first two oscillators, i.e., we use the momentum representation. Then the interaction will get the form

$$\text{const} \times a_i \left(x_i - \frac{c}{a_i} x_3 \right)^2 = \text{const} \times \left(a_i x_i^2 - 2cx_ix_3 + \frac{c^2}{a_i} x_3^2 \right). \quad (12)$$

Compared with the first model considered above an extra x_3^2 term has appeared with the consequence that the previous coefficient a_3 has changed to

$$a_3 \rightarrow a_3 + \frac{c^2}{a_1} + \frac{c^2}{a_2}, \quad (13)$$

and in the quantum case

$$A_3 \rightarrow A_3 + \frac{c^2}{a_1} + \frac{c^2}{a_2}. \quad (14)$$

Inserted in expression (9) this means that the coefficient D_i has changed to

$$D_i = \frac{c^2}{A_3} \left(\frac{1}{A_i} - \frac{1}{a_i} \right) = -\frac{\zeta^2 c^2}{a_i A_i A_3}. \quad (15)$$

Again the free energy due to the interaction follows by summation of the logarithm of the last factor of expression (9). In the classical high temperature limit ($\beta \rightarrow 0$) only the term $\hbar\zeta = 2\pi n/\beta = 0$ is present, but with expression (15) its contribution is zero. This is similar to what happens for the TE zero mode (in the Drude model) for the Casimir effect. For finite temperatures the corresponding free energy must be negative. But since it approaches zero when $T \rightarrow \infty$, there will be a temperature interval for which the Casimir free energy increases with increasing temperature, corresponding to a Casimir entropy $S = -\partial F/\partial T$ being negative.

III. INTERACTIONS VIA MANY OSCILLATORS

In the models of Sec. II two oscillators interacted via a third one. This situation we can extend and generalize to interactions via many oscillators. Such a situation is the analogue of electromagnetic interactions which have a continuum of frequencies. Then the a_3 and A_3 of Eqs. (6) and (8) are generalized to

$$a_3 \rightarrow a_i, \quad A_3 \rightarrow A_i, \quad i = 3, 4, 5, 6, \dots, \quad (16)$$

with $a_i = \omega_i^2$ and $A_i = a_i + \zeta^2$ as before.

Again oscillators 1 and 2 interact via oscillators i ($i \geq 3$) where the coefficient c of Eq. (9) becomes coefficients c_i . [Different coefficients c_{1i} and c_{2i} for the two oscillators 1 and 2 will also be possible.] With this one will find that the inverse partition function will be the

determinant that generalizes Eq. (9) to

$$\begin{aligned}
Q &= \begin{vmatrix} A_1 & 0 & c_3 & c_4 & \cdots \\ 0 & A_2 & c_3 & c_4 & \cdots \\ c_3 & c_3 & A_3 & 0 & \cdots \\ c_4 & c_4 & 0 & A_4 & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{vmatrix} = \begin{vmatrix} A_1(1-D_1) & -A_1D_1 & c_3 & c_4 & \cdots \\ -A_2D_2 & A_2(1-D_2) & c_3 & c_4 & \cdots \\ 0 & 0 & A_3 & 0 & \cdots \\ 0 & 0 & 0 & A_4 & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{vmatrix} \\
&= A_1A_2(1-D_1)(1-D_2) \left(1 - \frac{D_1D_2}{(1-D_1)(1-D_2)}\right) \left(\prod_{i \geq 3} A_i\right), \tag{17}
\end{aligned}$$

where now

$$D_j = \frac{1}{A_j} \sum_{i \geq 3} \frac{c_i^2}{A_i}, \quad (j = 1, 2). \tag{18}$$

Here, to evaluate the determinant, columns $i = 3, 4, \dots$ have been multiplied with c_i/A_i and subtracted from columns 1 and 2.

The second model is again the analogue of the electromagnetic interaction for the TE mode. Then the momenta of oscillators 1 and 2 interact with all the oscillators of the electromagnetic interaction. Thus the interaction will have the form $(p_j - \sum_{i \geq 3} (c_i x_i))^2 / (2m_j)$ ($j = 1, 2$), and again one finds that the interaction has no influence upon the classical partition function. To simplify in the quantum case we again can exchange the roles of momenta and coordinates of oscillators 1 and 2. Like Eq. (12) the interaction then ends up with the form

$$a_j \left(x_j - \frac{1}{a_j} \sum_{i \geq 3} (c_i x_i) \right)^2 = a_j x_j^2 - 2x_j \sum_{i \geq 3} (c_i x_i) + \frac{1}{a_j} \sum_{i \geq 3} \sum_{l \geq 3} (c_i c_l x_i x_l). \tag{19}$$

The coefficients c_i can be extended to the more general c_{ji} ($j = 1, 2$), but to simplify the matrices below a bit this is not done. With Eq. (19) and short hand notations $\mu = 1/a_1 + 1/a_2$ and $q_j = A_j(1/A_j - 1/a_j)$, Eq. (9) will be generalized to ($A_i = a_i + \zeta^2$)

$$Q = \begin{vmatrix} A_1 & 0 & c_3 & c_4 & \cdots \\ 0 & A_2 & c_3 & c_4 & \cdots \\ c_3 & c_3 & A_3 + c_3^2 \mu & c_3 c_4 \mu & \cdots \\ c_4 & c_4 & c_4 c_3 \mu & A_4 + c_4 c_4 \mu & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{vmatrix} = \begin{vmatrix} A_1 & 0 & c_3 & c_4 & \cdots \\ 0 & A_2 & c_3 & c_4 & \cdots \\ c_3 q_1 & c_3 q_2 & A_3 & 0 & \cdots \\ c_4 q_1 & c_4 q_2 & 0 & A_4 & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{vmatrix}. \tag{20}$$

In Eq. (20) rows $j = 1, 2$ have been multiplied with c_i/a_1 and c_i/a_2 respectively and subtracted from rows $i = 3, 4, \dots$. Next, similar to Eq. (17) columns $i = 3, 4, \dots$ are multiplied with $q_j c_i/A_i$ for $j = 1, 2$ and subtracted from columns 1 and 2. The resulting contributions to the inverse partition function is again (17), now with D_j given by

$$D_j = \frac{q_j}{A_j} \sum_{i \geq 3} \frac{c_i^2}{A_i} = -\frac{\zeta^2}{a_j A_j} \sum_{i \geq 3} \frac{c_i^2}{A_i}. \quad (21)$$

Altogether, this is just a straightforward generalization of result (9) for Q with D_j either given by Eqs. (10) or (15) in the two cases. The main difference lies in the quantities D_j that in the present section contain many contributions. Thus with D_j given by Eq. (21) as with Eq. (15) the corresponding Casimir entropy will be negative in an interval as concluded at the end of Sec. II. This constitutes a proof that the TE entropy must always be negative at high temperature. Typically, in fact, it is negative at all temperatures [8, 12].

An additional notable and interesting feature of the inverse partition function, which is the square root of Eq. (17) for each Matsubara frequency, is the product term for $i \geq 3$. Clearly this part is not affected by the presence of oscillators 1 and 2 and their influence upon the resulting eigenfrequencies of the coupled system of all oscillators. Thus oscillators $i \geq 3$ can without any approximation be eliminated or disregarded to be replaced by the interaction quantities D_j at thermal equilibrium. Correspondingly, with polarizable media the quantized electromagnetic field can be eliminated to be replaced by the radiating dipole-dipole interaction. This simplification we have utilized in Ref. [11] and later works.

IV. INTERACTION VIA THE ELECTROMAGNETIC DIPOLE RADIATION FIELD

For two oscillators interacting via the electromagnetic field it should now be possible to identify this situation with Eq. (17) where D_j is expression (18) for the TM mode and expression (21) for the TE mode. The free energy of interaction (Casimir energy) follows from the logarithm of the penultimate factor of (17) when inserted in Eq. (11). As we will see, the radiating dipole interaction has the form and structure consistent with the expressions for D_j .

For two oscillators interacting via the potential $\psi(\mathbf{r})_{s_1 s_2}$ with oscillator coordinates s_i , which can be identified with polarization (here in one dimension for simplicity). The Casimir

free energy F in the classical case is given by Eq. (3.4) in Ref. [11] as

$$\beta F = \frac{1}{2} \ln(1 - (\alpha\psi)^2) \approx -\frac{1}{2}(\alpha\psi(\mathbf{r}))^2 \quad (22)$$

where α is polarizability. In the quantum case one sums over Matsubara frequencies as in Eq. (5.8) of that reference by which

$$\beta F = \frac{1}{2} \sum_K \ln(1 - (\alpha_K \psi_K)^2). \quad (23)$$

With two equal oscillators (same α) it should be possible to make the identification

$$\frac{D_j}{1 - D_j} \rightarrow \alpha_K \psi_K. \quad (24)$$

It is clear that $1/A_j$ corresponds to $\alpha_K \propto 1/(a_j + \zeta^2)$ for a simple oscillator with eigenfrequency $\omega = \sqrt{a_j}$ not interacting with its surroundings. As pointed to below Eq. (9) the $A_j(1 - D_j)$ represent s oscillator $j = 1$ or 2 alone and their interactions with oscillator 3. Thus in Eq. (17) the same factor represent the interaction of oscillator j with the electromagnetic field represented by oscillators $i \geq 3$. So $1/(A_j(1 - D_j))$ corresponds to α_K with radiation reaction taken into account. With this the remaining part $A_j D_j$ of (23) should represent ψ_K . According to Eqs. (18) or (21) this gets contributions from the oscillators through which oscillators 1 and 2 interact. Then the remaining crucial question is whether the radiating dipole interaction ψ_K is consistent with the two expressions for D_j . Thus we must look for the eigenmodes of the electromagnetic field. In free space these modes are plane waves of wave vector \mathbf{k} and frequency

$$\omega = ck \quad (25)$$

where c is light velocity. These waves should, if possible, be identified with the oscillators $i \geq 0$ of Sec. III. And this identification we find from the Fourier transform of the radiating dipole interaction. This interaction is given by Eq. (6.1) in Ref. [13] ($\zeta = i\omega$)

$$\tilde{\phi}(12) = \frac{4\pi}{3} s_1 s_2 \frac{1}{(ck)^2 + \zeta^2} [(ck)^2 \tilde{D}(12) + 2\zeta^2 \hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2] \quad (+\text{const.}), \quad (26)$$

with $\tilde{D}(12) = 3(\hat{\mathbf{k}} \cdot \hat{\mathbf{s}}_1)(\hat{\mathbf{k}} \cdot \hat{\mathbf{s}}_2) - \hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2$. The hats denote unit vectors. Here \mathbf{s}_j are the polarizations of the two oscillators. The constant term can be disregarded as it only contributes to a δ -function $\delta(r)$ in \mathbf{r} -space and is thus zero anyway with $r \neq 0$.

It is now easily seen that expression (26) has precisely the form where both expressions (18) and (21) for $A_j D_j$ are present with A_i given by (8). The $\tilde{D}(12)$ and $\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2$ terms of

expression (26) correspond to expressions (18) and (21) respectively. With Fourier transform (26) the frequency dependent dipole interaction $\psi(k) \rightarrow \phi(12)$ is given by

$$\phi(12) = \frac{1}{(2\pi)^3} \int \tilde{\phi}(12) e^{i\mathbf{k}\mathbf{r}} d\mathbf{k} = s_1 s_2 [\psi_{DK}(r)D(12) + \psi_{\Delta K}(r)\Delta(12)], \quad (27)$$

where from Eq. (5.10) of Ref. [11]

$$\psi_{DK}(r) = -\frac{e^{-\tau}}{r^3} \left(1 + \tau + \frac{1}{3}\tau^2 \right) \quad \text{and} \quad \psi_{\Delta K}(r) = -\frac{2e^{-\tau}}{3r^3} \tau^2 \quad (+\text{const. } \delta(\mathbf{r})), \quad (28)$$

with $\tau = i\omega r/c$.

Thus altogether, interactions via the electromagnetic field contain both the two oscillator models considered in Sec. III. The dipole-dipole interaction (26) is then a sum (\rightarrow integral) of eigenmodes (Fourier components) that induce the resulting interaction between the two oscillators. An implication of this, as we have seen, is that the contribution to the entropy can be negative in some regions.

V. SUMMARY

We have studied the reason for possible negative entropy related to the Casimir interaction between two media. This negative entropy may seem unphysical. To show that this is not so, we have studied two harmonic oscillator models where two oscillators interact via a third one. For one of the models the momenta of the two oscillators interact with the amplitude of the third one in a way similar to the interaction with the electromagnetic vector potential, and in fact corresponds to the TE polarization. Then a negative entropy contribution is found. This shows that this type of behavior is not unphysical. Then the situation with the third oscillator is generalized to a set of oscillators that mediates the induced interaction between the two oscillators. Finally it is noticed that the induced radiating dipole-dipole interaction between a pair of oscillating dipole moments can be identified with a combination of the induced ones of the two oscillator models. This paper gives a proof that the TE contribution to the entropy must be negative for large T , being typically negative for all T . The TM contribution is typically positive. The total entropy, therefore, is likely to contain a negative entropy region.

Appendix A: Field theory approach

The point of this appendix is to show that the considerations of the main text have a close correspondence with the field theoretic approach in quantum electrodynamics. The latter starts from the expression for the free energy as a sum over Matsubara frequencies (here $\hbar = c = 1$)

$$F = -\frac{T}{2} \sum_{n=-\infty}^{\infty} \text{Tr} \ln \mathbf{\Gamma} \mathbf{\Gamma}_0^{-1}, \quad (\text{A1})$$

where $\mathbf{\Gamma}_0$ is the free electromagnetic Green's dyadic, and $\mathbf{\Gamma}$ is that in the presence of bodies which interact with the electromagnetic field, e.g., dielectric or metallic bodies. For the case of dielectrics, we can define a potential in terms of the permittivity ε , $V = \varepsilon - 1$, and then we can readily show for two disjoint bodies, for which $V = V_1 + V_2$, that the free energy is

$$F = \frac{T}{2} \sum_n \text{Tr} \ln[(\mathbf{1} - \mathbf{\Gamma}_0 V_1)(\mathbf{1} - \mathbf{\Gamma}_0 \mathbf{T}_1 \mathbf{\Gamma}_0 \mathbf{T}_2)(\mathbf{1} - \mathbf{\Gamma}_0 V_2)], \quad (\text{A2})$$

in terms of the scattering matrices

$$\mathbf{T}_i = V_i(\mathbf{1} - \mathbf{\Gamma}_0 V_i)^{-1}. \quad (\text{A3})$$

Evidently, Eq. (A2), sometimes called the TGTG formula, is identical with Eq. (17) inserted into Eq. (11), which was derived long before the modern renaissance of multiple-scattering formulations of Casimir problems. Here the A_i 's have been disregarded, as not involving interaction with the electromagnetic field, and the D_i are identified with

$$D_i \leftrightarrow \mathbf{\Gamma}_0 V_i. \quad (\text{A4})$$

And the break-up into electromagnetic modes, detailed in Sec. IV, is just the well-known decomposition

$$\begin{aligned} \mathbf{\Gamma}_0(\mathbf{r}) &= (\nabla\nabla - \mathbf{1}\nabla^2) \frac{e^{-|\zeta_n|r}}{r} \\ &= \left[(3\hat{\mathbf{r}}\hat{\mathbf{r}} - \mathbf{1}) \left(1 + |\zeta|r + \frac{1}{3}\zeta^2 r^2 \right) - \mathbf{1} \frac{2}{3}\zeta^2 r^2 \right] \frac{e^{-|\zeta|r}}{r^3}, \end{aligned} \quad (\text{A5})$$

which restates Eqs. (27) and (28). So the correspondence is not merely analogous, it is precise.

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