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# Sample-gas thermal contact resistance and the photoacoustic signal generation

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By invoking long standing results from studies on heat transfer across a two-media interface, the claim for the non-negligible character of the effects of the solid-gas thermal contact resistance on the photoacoustic signals is amply discussed. Existing experimental evidences are presented which result in strong support for such a claim.

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In recent papers<sup>1-3</sup> we have reported on the effects of the thermal contact resistance between two media on the photoacoustic (PA) signal buildup. In particular it was conclusively demonstrated that when the surface thermal resistance of the solid sample is taken into account, the dependence of the PA signal on both the chopping frequency and the gas thermal properties are considerably modified as compared with those predicted by the classical work of Rosencwaig and Gersho (RG).<sup>4</sup> For instance, for both thermally thin and thermally thick samples, the pressure fluctuations in the cell are predicted in Ref. 1 to vary inversely proportional to the heat capacity of the gas, whereas the standard RG theory predicts that the acoustic signal varies with the square root of the gas thermal diffusivity (i.e.,  $\alpha_g^{1/2}$ ). This prediction that the acoustic signal varies inversely proportional to the gas heat capacity was experimentally observed<sup>1</sup> by taking measurements of the PA signal of a germanium sample 480  $\mu\text{m}$  thick in a wide range of frequencies, changing, however, the cell's gas. The gases used were helium and air, so that, according to Ref. 1, the ratio of the PA signal of our Ge sample for the helium-filled cell to that of the air-filled cell should give us the same value (i.e., 1.3) at both high and low frequencies. In contrast, the RG theory predicts that this ratio should be equal to 2.7, both at high and low frequencies, a fact that was not observed in our experiments.<sup>1,2</sup>

In a recent communication, however, Quimby and Yen<sup>5</sup> (QY) have criticized our results on the effect of the thermal contact resistance on the PA-signal production by presenting what they believe to be correct treatment of the solid-gas contact resistance. The conclusion that they come to is that the value we have assumed for the thermal surface conductance  $H$  ( $\sim 10^{-4}$ – $10^{-3}$  W/cm<sup>2</sup> °K) is four or three orders of magnitude smaller than the values predicted by their theory. As a consequence it is then claimed by these authors that the effects of the surface resistance, as predicted in Refs. 1–3, are expected to be negligible.

The purpose of this is to present a more detailed discussion concerning the surface thermal resistance and show that the estimate of the heat transfer coefficient by Quimby and Yen<sup>5</sup> is, contrary to those authors expectation, incorrect. In

fact, existing PAS experimental evidence is brought to attention which not only confirms the non-negligible character of such effects, but also strongly supports the predictions from the theory in Ref. 1.

According to QY's argument, heat is exchanged between the solid and gas when gas molecules initially at temperature  $T_g$  strike the solid (at temperature  $T_s$ ) and are ejected from the solid at a new temperature  $T'_g$ . This heat flow is limited by the number of molecules colliding with the surface per second, and the average energy picked up or lost by the molecules per collision, so that the heat flux  $J$  can be written as<sup>6,7</sup>

$$J = \frac{P(C_v + \frac{1}{2}K_B)}{(2\pi m K_B T)^{1/2}} (T'_g - T_g), \quad (1)$$

where  $P$  is the gas pressure,  $C_v$  is the heat capacity per molecule of gas,  $K_B$  is the Boltzmann constant, and  $m$  is the mass of the gas molecule. Next, they used typical steady values<sup>7</sup> of the thermal accommodation coefficient,  $(T'_g - T_g)/(T_s - T_g)$ , to estimate  $T'_g - T_g$ , and consequently the heat transfer coefficient  $H = J/(T_s - T_g)$ . This procedure, however, is obviously incorrect. The quantity  $T'_g - T_g$  in Eq. (1) is essentially the temperature fluctuation  $\phi_g$  in the gas.

Hence what QY should have done was, in light of their expression for the heat flux, to write the temperature boundary conditions at the solid-gas interface as

$$-k_s \phi'_s(0) = \frac{P(C_v + \frac{1}{2}K_B)}{(2\pi m K_B T)^{1/2}} \phi_g(0) \quad (2)$$

in place of Eq. (1) of Ref. 1, namely,

$$-k_s \phi'_s(0) = H(\phi_s - \phi_g)_{x=0}.$$

Here the prime denote spatial differentiation, and  $k_s$  and  $\phi_s$  are the thermal conductivity and the temperature fluctuation in the solid, respectively. However, using Eq. (2) together with the heat-flux continuity condition at  $x = 0$  [i.e.,  $k_s \phi'_s(0) = k_g \phi'_g(0)$ ] the result one gets is that there is no temperature fluctuation in the PA cell. This can be seen by writing, in the usual RG notation,  $\phi_s(x) = Ue^{-\sigma x} + Ve^{\sigma x} - Ee^{\beta x}$  and  $\phi_g(x) = \theta e^{-\sigma x}$ , and substituting them into Eq. (2) and the heat-flux continuity condition. Even though this

result may look surprising, a closer analysis shows that it is consistent with the QY formulation of heat transfer mechanism as outlined above. This microscopic view of the heat transfer, of course, refers to the equilibrium situation. The actual physical situation taking place while the PA signal builds up is far more involved than that and the simple-flux formulation by QY does not apply.<sup>8-10</sup> We now take a closer look into that matter in what follows.

The heat transfer between two bodies takes place essentially via two modes, namely, diffusion and radiation. In diffusion, heat is transferred through a medium or from one to another of two media in contact, if there exists a nonuniform temperature distribution in the medium or between the two media. From a microscopic point of view the mechanism of diffusion is visualized as the exchange of kinetic energy amongst the molecules in the regions of high and low temperatures, as aptly noted by Quimby and Yen. The heat transport by radiation occurs, on the other hand, as a result of the emission of radiation by a medium and its subsequent absorption by the other one. Here we have not considered convection to be a distinct mode of heat transfer. Actually, convection is motion of the medium which facilitates heat transfer by diffusion and (or) radiation. For customary reasons only, one usually distinguishes between the diffusion of heat in rigid bodies, which is called conduction, and the diffusion of heat in moving deformable bodies, which is called convection. In the case of a solid-gas interface, the heat transfer occurs essentially by convection (in the latest sense) and radiation, and assuming that the temperature changes are not very large, one may say that the heat flux is proportional to the temperature differences between the solid and the gas, namely,  $J = H(\phi_s - \phi_g)$ . This is the so-called linear approximation for the heat transfer referred to in Ref. 1. The heat transfer coefficient  $H$ , similarly to but more strongly than the thermal conductivity, depends in a complex manner on a number of variables.<sup>8-10</sup> These may include the space, time, geometry, flow conditions, and physical properties. A large amount of work has been devoted to both experimental and theoretical evaluation of  $H$  under many different physical conditions.<sup>11-15</sup> For instance, if air is blown past a flat surface at a high speed, relatively large values of  $H$  are observed and it is independent of  $T_s - T_g$ . If air is not blown past the surface, but rises slowly owing to buoyancy effects, small values of  $H$  are observed and they are roughly proportional to  $(T_s - T_g)^{1/4}$ . As for the effects of pressure upon the free convection in air they have been studied since 1936 by Saunders.<sup>11</sup> The resulting values of commonly encountered heat transfer coefficients are<sup>8-10</sup> of the order to 5–25 W/m<sup>2</sup> °K for free convection in common gases, and typically one to two orders of magnitude greater (i.e., ~100 W/m<sup>2</sup> °K) for free convection in water. As for forced convection in gases,  $H$  is roughly of the order of few tens W/m<sup>2</sup> °K. In the case of heat transfer by radiation,  $H$  is usually one order of magnitude smaller than that of free convection in gases, but it may become comparable to it depending on the particular emissivities and temperature of the bodies. For bodies exposed to air at normal temperatures,<sup>8</sup> free convection and radiation give values of  $H$  of the same order of magnitude. These well established values<sup>8-10</sup> of  $H$  for the

free-convection heat transfer coefficient (~5 W/m<sup>2</sup> °K) are the ones we have used in Refs. 1–3, and they are radically different from the values (~10<sup>4</sup> W/m<sup>2</sup> °K) proposed by Quimby and Yen.<sup>5</sup> In this context two comments should be added. First, we note that such large values of  $H$ , as proposed by QY,<sup>5</sup> are typically of the same order to magnitude as the ones found in extremely efficient heat transfer situations like problems involving changes of phase<sup>8-10</sup> (e.g., boiling liquids and condensing vapors). Consequently, it is unlikely that in the usual PAS experiments  $H$  can assume these values proposed by QY. Second, even under the most favorable thermal contact, such as that of optically flat metallic surfaces pressed together, one does not get values of  $H$  as proposed by Quimby and Yen. This was demonstrated by Jacobs and Starr<sup>15</sup> who have measured the heat transfer coefficient for optically flat silver surfaces pressed with 2 atm and found  $H \approx 3000$  W/m<sup>2</sup> °K. This physically untenable situation of having a solid-gas interface realizing a better thermal contact than two highly polished metal surfaces pressed together is strongly indicative of the incorrectness of QY's formulation.

Now, as for solid-solid interfaces, the thermal contact resistance is of quite a different nature from that for solid-gas interfaces, discussed above. Here there are essentially two typical situations, namely, that of a nearly soldered joint and that of solids just placed in contact. The first case corresponds to sample films deposited on the backing, a situation reported in connection to some PAS experiments. Here the appropriate temperature boundary condition to be used is the temperature continuity condition, because  $H$  is very large. However, in the case of solids just fitted against one another one faces a rather more involved situation, which, as a matter of fact, is found in the majority of the PAS experimental setups. To describe this situation one may assume that between the two solids one has a thin gas skin of thermal conductivity  $k_g$  and thickness  $\delta$ . Hence the effective backing-sample thermal contact conductance  $H_{bs}$  is then given by<sup>8-10</sup>

$$H_{bs} = \frac{1}{1/H_b + \delta/k_g + 1/H_s}, \quad (3)$$

where  $H_b$  and  $H_s$  are the backing-gas and the solid-gas heat transfer coefficients, respectively, and  $\delta/k_g$  is the gas skin thermal resistance. Assuming that the gas skin thickness is of the order of the roughness of a finished (polished) surface

TABLE I. Ratio of the PA signal (5) for different filler gases and comparison with the predictions of Ref. 1 and RG's theory. The experimental results were taken from Kirkbright *et al.*<sup>17</sup> measurement on carbon black at  $\lambda = 570$  nm.

Carbon black PA-signal (S) Ratio	Experimental data <sup>17</sup>	Our theory	RG theory
$S(\text{He})/S(\text{Ar})$	1.13	1.03	2.93
$S(\text{He})/S(\text{N}_2)$	1.35	1.36	2.86
$S(\text{He})/S(\text{CO}_2)$	1.62	1.70	4.04

TABLE II. Thermal diffusivities ( $\alpha$ ) and thermal conductivities ( $k$ ) of gases.

Gases	$\alpha(\text{cm}^2/\text{s})$	$k(\text{mW}/\text{cm}^\circ\text{K})$
Helium	1.8	1.5
Argon	0.21	0.18
Nitrogen	0.22	0.25
Carbon Dioxide	0.11	0.15

(i.e., 10–100  $\mu\text{m}$ ), and noticing that  $k_g \sim 0.1 \text{ W}/\text{m}^\circ\text{K}$  one gets an upper limit for the skin thermal resistance of the order of  $10^{-4} - 10^{-3} (\text{W}/\text{m}^2^\circ\text{K})^{-1}$ . Since  $(H_b)^{-1} \sim (H_s)^{-1} \sim 0.2 (\text{W}/\text{m}^2^\circ\text{K})^{-1}$ , it then follows from Eq. (3) that

$$H_{bs} \simeq H_b H_s / (H_b + H_s) \sim H_b. \quad (4)$$

In other words, the backing-solid heat transfer coefficient is, in this case, roughly of the same order of magnitude as that of the solid-gas interface.

Though we did assume in Ref. 1 (and maintain, in view of what has been said above) that as far as the estimated values of  $H$  were concerned the processes of radiative and convective heat transfer could face, under certain conditions, comparable thermal resistances, no strong case was made there as to the radiative heat transfer being the relevant acting mechanism. In fact, the assumed linear heat transfer mechanism across the media interface as well as the assumed approximation  $h \ll g \ll b$  leading to Eq. (4) in Ref. 1 hold equally well under a dominantly convective transfer. Nevertheless, in QY's paper the case against the theory formulated in Ref. 1 was mainly based on their wrong supposition that said theory rests on the assumption of a purely radiative heat transfer across the solid-gas interface. They seem to have missed the whole point of our statement, namely, that we were just comparing order of magnitude estimates of  $H$  for the two processes. We were, of course, quite aware that their concurrence would require special conditions to be met at the solid-gas interface. In the PA problem it is obvious that the commonly encountered interface conditions favor the heat transfer via free convection, but with a value of  $H$  which, contrarily to QY's contention, is of the same order of magnitude as is commonly found in heat exchange mediated by radiation. Our aim in such comparison was to put forward the important role played by the surface thermal resistance in heat transfer problems and thereby argument as to the appropriateness of its inclusion in the formulation of a theory for the PA effect.

Next, we would like to comment on the effects of the surface thermal resistance upon the PA-signal dependence on the chopping frequency. Quimby and Yen mentioned that for a thermally thick sample in the unsaturated regime they measured the frequency dependence of the PA signal and found it to be  $f^{-3/2}$  in agreement with the RG theory and not  $f^{-2}$  as predicted in Ref. 1. This fact is also claimed by QY to be the definite experimental evidence for the incorrectness of our assumption of  $H \sim 5 \text{ W}/\text{m}^2^\circ\text{K}$ . Unfortunately, no details (such as type of sample, frequency range, gas, sample's length, etc.) regarding their experiment are present-

ed in Ref. 5. In any event, the PA-signal frequency dependence alone cannot be considered as the definite experimental proof for their claim. The reason is that, in the general case, the PA signal is a sum of contributions<sup>3,16</sup> of the heat generated in the bulk and at the surface. In particular for optically transparent and thermally thick samples the PA pressure fluctuations is, according to Ref. 3, given by

$$\delta P = \frac{\gamma P_0 I_0 \alpha_g}{2\pi T_0 l_g k_g} \left( \frac{\beta'}{f} - j \frac{(1 - \beta') B H \alpha_s}{2\pi k_s f^2} \right) \times \exp[j(\omega t - \frac{1}{2}\pi)] \quad (5)$$

where  $\beta'$  and  $\beta$  are the surface and the bulk optical absorption coefficients, respectively, and the other quantities have the same physical meaning as in Refs. 1–5. It follows from Eq. (5) that the frequency dependence of the PA signal is given by the sum of two terms, one varying as  $f^{-1}$  and the other one varying as  $f^{-2}$ . Consequently, working on experimental data, one can eventually adequately fit an overall  $f^{-3/2}$  dependence where an  $af^{-1} + bf^{-2}$  should be expected, especially when the two terms have comparable magnitudes. Here we add that, especially in the case of unsaturated samples, the surface absorption in general can not be neglected as demonstrated by Bennett and Forman.<sup>16</sup> Indeed, for highly transparent materials,<sup>16</sup> such as laser windows, the surface optical absorption is the dominant heat source for the PA-signal buildup. Hence the PA frequency dependence alone cannot be taken as the definite check of our theory. As we mentioned above a more dramatic demonstration of the influence of the thermal contact resistance on the production of the PA signal is, of course, the dependence of the acoustic signal on the gas thermal properties. This is what was done in Ref. 1; according to our theory the acoustic signal should vary inversely proportional to the heat capacity of the gas (i.e.,  $\alpha_g/k_g$ ), whereas RG theory predicts that it varies as  $\alpha_g^{1/2}$ . Hence for an optically opaque and thermally thick sample the ratio of the PA signal from a He-filled cell to an Air-filled cell should be equal to 1.3, instead of 2.7, as predicted by the RG theory. It should be emphasized at this point that well-distinguishable ratios are predicted in view of the difference in the dependences on the cell gas parameters between ours and RG's theory. In fact, while the dependence on the chopping frequency for the case of an optically transparent and thermally thick solid (e.g.,  $af^{-1} + bf^{-2}$  in our theory and  $cf^{-3/2}$  in RG's) might require careful and precise measurements and data fitting to be experimentally told apart, especially when  $a \sim b$ , the ratio of the PA signals at fixed modulation frequency for two different gases inside the cell can be easily measured. Since the two different dependences on the gas thermal properties arise from our taking into account the surface thermal resistance, the experimental verification reported in Ref. 1 of our predictions can be an argument in support of the non-negligible effects of such resistance. In fact, even stronger support to our views comes from a previous work by Kirkbright and co-workers,<sup>17</sup> who experimentally investigated the variation of the PA signal for carbon black with the nature of the cell filler gas. Their spectra for carbon black, in the region 500–650 nm, using different filler gases, are shown in Fig. 7 of Ref. 17. In Table

I, we used the experimental data of Ref. 17 to compute the ratio of the carbon black PA signal (at  $\lambda = 570$  nm) for different gases and compare these values to those calculated according to our  $(\alpha_g/k_g)$  and RG's  $(\alpha_g^{1/2})$  predictions. The values for thermal diffusivity and thermal conductivity of the gases, used in our calculations are given in Table II. No doubt the good agreement between our predictions and the experimental data strongly supports our claim regarding the non-negligible effects of the solid-gas thermal contact resistance on the PA-signal buildup.

In conclusion, we believe that, in view of the foregoing, the QY versus Ref. 1 dispute is definitely settled in our favor. However, should the value of  $H$  in a given PA experiment be needed, we propose that an independent, *in situ* measurement, as suggested below, be carried out rather than relying on tabulated data. The reason for this is that sample handling and treatment may introduce potentially important additional sources (e.g., grease skins, oxidation, etc.) for the effective thermal contact resistance. Such an independent evaluation of the heat-transfer coefficient can be accomplished by analyzing the time evolution of the PA signal when the sample is excited by a single light pulse. This and several other aspects relating to the time-domain investigation of the PA effect of solids are currently being explored by our group and will be the subject of forthcoming publications.

## EDITORIAL NOTE

The Communications section of this issue contains two papers related to the foregoing paper by Lima, Miranda, and Santos. The first, by Quimby and Yen, was originally submitted to the Physical Review as a Comment on a Letter by C. L. Cesar *et al.*, its Ref. 1. In the course of the review of this paper, events conspired to elicit the second paper, by Rosencwaig, which is, as its title indicates, a Comment on the

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- <sup>2</sup>C. L. Cesar, H. Vargas, J. A. Meyer, and L. C. M. Miranda, paper presented at the Photoacoustic Spectroscopy Meeting, Ames, Iowa, Aug 1-3, 1979 (unpublished).
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present paper by Lima *et al.* All of the authors concerned have agreed that these papers should appear together. Although the circumstances leading to this mode of publication are unusual, the mode of publication is in accord with our usual policy of affording the various parties an occasion to state their views on a controversial subject. There has been no attempt to resolve any differences, and the absence of a reply does not imply acceptance of a criticism.