Comment on "The Electrocaloric Effect in Fluids ..." by K. H. Wang and R. M. Herman

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A misinterpretation by Wang and Herman of the earlier treatment by Batra, Enns, and Pohl of the electrocaloric effect and STS-I (stimulated thermal scattering-I) is discussed.

In a recent paper by Wang and Herman,¹ reference is made to an earlier treatment of the electrocaloric effect and stimulated thermal scattering-I (STS-I) by Batra, Enns, and Pohl.² To quote from Wang and Herman's paper, "In still another treatment,² Eq. (3) is taken to be valid and then, without elaboration, the authors allow that it is acceptable to employ Eq. (2) when describing STS-I." As the paper by Batra et al.² was designed primarily to be a review of STS-II, perhaps not enough stress was placed on the conclusions reached in that paper on STS-I. The quotation above indicates that our treatment of STS-I has been misinterpreted. In point of fact, we arrived at the same major conclusions as Wang and Herman¹ with exactly the same mathematical reasoning. We did not mysteriously or erroneously replace Eq. (3) by Eq. (2) in midcalculation, as implied by the quotation.

Aside from inadvertently omitting a minus sign (which makes no practical difference to the results, as we shall comment on shortly), we employed Eq. (3) throughout our calculation. To see where the misinterpretation arose, let us briefly recapitulate what was done in Ref. 2. Using the notation and equation numbers of that paper, the total gain involved the sum of Eqs. (2.113) and (2.24). We then set the absorption coefficient $\alpha = 0$ (no STS-II) and first considered $\omega^* \equiv \omega/\omega_B \simeq 0$. As stated clearly in the paper we made use of the well-known thermodynamic relation $(\partial \epsilon / \partial T)_{\rho}$ $= (\partial \epsilon / \partial T)_{\rho} + \beta \gamma^{\rho}$, and further noted that $(\partial \epsilon / \partial T)_{\rho}$ $\ll (\partial \epsilon / \partial T)_{p}$. Thus, the dominant term was found to be proportional to $(\beta \gamma^{e})^{2} \simeq (\partial \epsilon / \partial T)_{p}^{2}$, and Eq. (2.117) of Ref. 2 resulted. This last step was taken so as to compare with the earlier work of Starunov.³ Apparently, it is in this last step that the misinterpretation took place. We should have *stressed* the essentially electrostrictive nature of the gain for $\omega^{*} \simeq 0$. The missing minus sign does not affect the final result for $\omega^{*} \simeq 0$ [i.e., Eq. (2.117)] at all. The only difference in the calculation is that one does not have to use the inequality above, although it is true. The gain is found to be proportional to

$$(\beta\gamma^e)^2 - 2\beta\gamma^e \left(\frac{\partial\epsilon}{\partial T}\right)_{\rho} + \left(\frac{\partial\epsilon}{\partial T}\right)_{\rho}^2 = \left(\frac{\partial\epsilon}{\partial T}\right)_{\rho}^2$$

i.e., $(\partial \epsilon / \partial T)_p^2$ is the result of a perfect square.

For $\omega^* \simeq \pm 1$, we did point out the disagreement of our result with that of Starunov.³ Our conclusion, like that of Wang and Herman, was that only a *small* correction to the SBS gain resulted by including Eq. (3). The minus sign changes the details slightly; viz., to the SBS peaks at $\omega^* = \pm 1$ one must add a small Lorentzian of the same (not the opposite, as stated in Ref. 2) sign. Eq. (2-119) of Ref. 2 should be replaced by the estimate $\tilde{\gamma}/\gamma^e = (2R/\beta\rho_0)(\gamma - 1)$ where $R/\beta\rho_0$ has been defined and tabulated for a few common liquids by Batra and Enns.⁴ For CCl₄, the correction is 2.8%. In closing, let it be emphasized that the reasoning and conclusions reached by Batra *et al.*² and by Wang and Herman¹ are the same.

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¹K. H. Wang and R. M. Herman, Phys. Rev. A <u>7</u>, 1248 (1973).

²I. P. Batra, R. H. Enns, and D. Pohl, Phys. Status Solidi B <u>48</u>, 11 (1971).

³V. S. Starunov, Phys. Lett. A <u>26</u>, 428 (1969); Zh. Eksp. Teor. Fiz. <u>57</u>, 1012 (1969) [Sov. Phys.-JETP <u>28</u>, 1085 (1969)].

⁴I. P. Batra and R. H. Enns, Phys. Rev. <u>185</u>, 396 (1969).