Ravindran and Arora Reply: In a recent Letter [1] we reported mode Grüneisen parameters of optical phonons in zirconium tungstate and showed that modes of energies much higher than 10 meV *also contribute substantially* to negative thermal expansion (NTE) in the material. In their Comment [2] Chaplot and Mittal (CM) repeat the arguments already mentioned in [3] to arrive at the conclusion that the temperature (T) dependence of thermal expansion coefficient (α) *could arise only* from modes below 10 meV.

CM argue that the temperature dependence of the contribution α_i to the thermal expansion arising from Einstein modes of energy E_i is the same as that of C_{Vi}. However, it must be emphasized that the T dependence of the total α could be quite different from that of the total C_V due to the multiplicative prefactors γ_i in the summation. In addition, γ_is could be either positive or negative leading to the cancellation of contributions of different phonons to the total α. Hence the argument of CM about α_i and C_{Vi} is not extendable to total α. In order to substantiate our claim regarding the phonons of energy higher than 10 meV, we show in Fig. 1 the cumulative contribution of phonons of energies up to E to the thermal expansion $\alpha(E)$ calculated at 70 and 300 K from the observed optical phonon frequencies and the number of degrees of freedom obtained using a procedure described earlier [1]. Because of lack of data the contribution of acoustic and optic phonons below 5 meV could not be considered. It is evident that even at 70 K phonons of energies above 10 meV contribute substantially to α and at 300 K their contribution is as high as 60%. It is important to point out that at 300 K the value of α calculated using our model (10×10^{-6} K^{-1}) is in excellent agreement with the measured value ($11 \pm 2 \times 10^{-6}$ K^{-1}) and it differs from the reported data only below 150 K. A part of this difference may have its origin in the contribution of the acoustic phonons to NTE. This was clearly stated earlier [1]. Further, the C_V calculated using our model is also in good agreement (inset of Fig. 1) with the reported T dependence [4].

CM claim that α calculated from their lattice dynamical model [5] fits better to the reported data. Their calculation shows that over 40% of the total α arises from two transverse acoustic phonons near the Brillouin-zone center and almost all the NTE arises from phonons below 8 meV. These lattice dynamical calculations have limitations, because they are based on empirical potentials that are fitted to the equilibrium crystal structure parameters, and thus are not guaranteed to describe the volume dependence of phonon modes accurately. Further, the predicted extremely large (~ -40) mode Grüneisen parameters of the acoustic phonons are yet to be confirmed experimentally. Although their model takes into account all the acoustic and optic phonons, the calculated value of α is far too low, $\sim$70% of the reported $\alpha(T)$ above 70 K (Fig. 1 of CM). In fact, their $\alpha(T)$ shows good agreement with the data of David *et al.* [6] only below 30 K. In view of this the conclusions drawn from their lattice dynamical calculation, which needs refinement, are questionable.

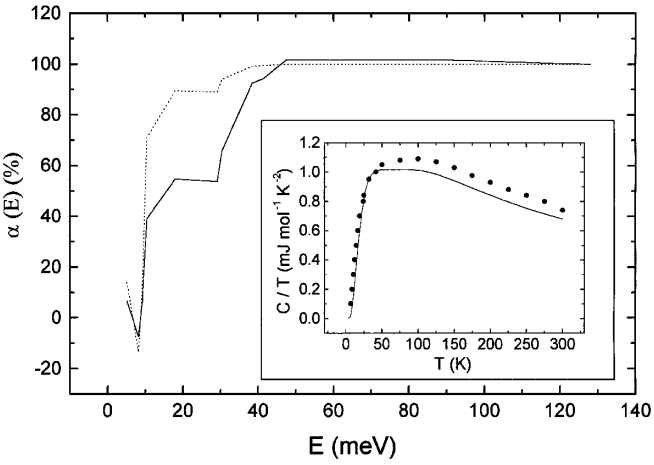

FIG. 1. Cumulative contribution to thermal expansion coefficient as a function of phonon energy at different temperatures. Full curve: 300 K; dashed curve: 70 K. The inset shows the calculated specific heat (full curve) and the data of Ref. [4].

Figure 2 of CM [2] presents the average Grüneisen parameter calculated using the expression of α. Any agreement or otherwise basically originates from the temperature dependence of α and C_V, which is already contained in their Fig. 1 [2].

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