

Schmid and Schirmacher Reply: We have recently published a theoretical investigation [1] of Raman scattering in disordered solids based on a model in which both the elastic constants of the solids as well as the elasto-optic (Pockels) constants are assumed to fluctuate in space. This leads to an incoherent scattering contribution which can be represented as a convolution in \mathbf{k} space of the spatial Pockels-constant correlation functions with the strain susceptibilities. It has been shown that the measured data of glasses in the boson peak frequency range ($\sim 20 \text{ cm}^{-1}$) can be well accounted for by this treatment. Within this model the measured intensity is *not* proportional to the vibrational density of states, as assumed in many other model treatments quoted in Ref. [1].

In their Comment [2] the authors point out that there is also a coherent contribution solely produced by the “mechanical disorder.” They furthermore claim that for this contribution the Shuker-Gammon-like expression [3] $I_{ijkl}^c(\omega) \propto [n(\omega) + 1]C_{ijkl}(\omega)\frac{g(\omega)}{\omega}$ holds, where $g(\omega)$ is the vibrational density of states, $n(\omega) = 1/[e^{\hbar\omega/k_B T} - 1]$ the boson occupation number and $C_{ijkl}(\omega)$ are light-vibration coupling coefficients [4] $C_{ijkl}(\omega) \propto \sum_{mnpq} p_{ijmn} p_{klpq} \Lambda_{mnpq}(\omega)$,

$$\Lambda_{mnpq}(\omega) = \int d^3\mathbf{r}_1 d^3\mathbf{r}_2 \frac{\langle \frac{1}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) s_{mn}^{\alpha}(\mathbf{r}_1) s_{pq}^{\alpha*}(\mathbf{r}_2) \rangle}{\langle \frac{1}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) \rangle}.$$

Here p_{ijmn} are the Pockels constants, $s_{ij}^{\alpha}(\mathbf{r})$ are the strains corresponding to an eigenmode of frequency ω_{α} and N is the number of modes. The fact that the light couples to the strains $s_{ij}^{\alpha}(\mathbf{r})$ and *not* to the displacements $u_i^{\alpha}(\mathbf{r})$ [3] is dictated by *global* translational invariance. By this we mean the property that the Hamiltonian is invariant with respect to a coordinate transformation $\mathbf{r} \rightarrow \mathbf{r} + \mathbf{R}$.

Let us consider one of the spatial integrals

$$\int d^3\mathbf{r} s_{ij}^{\alpha}(\mathbf{r}) = \frac{1}{2} \int d^3\mathbf{r} \left(\frac{\partial}{\partial x_i} u_j^{\alpha}(\mathbf{r}) + \frac{\partial}{\partial x_j} u_i^{\alpha}(\mathbf{r}) \right). \quad (1)$$

Here $u_i^{\alpha}(\mathbf{r})$ are the displacements of a vibrational mode α . This integral is zero except for boundary terms, which vanish if, e.g., periodic boundary conditions are used. This means that the coherent term suggested in [2] is zero.

However, one can write down a coherent Shuker-Gammon-like expression if one takes into account the nonvanishing light scattering wave vector \mathbf{k} :

$$\begin{aligned} I_{ijkl}^c(\mathbf{k}, \omega) &\propto [n(\omega) + 1] C_{ijkl}(\mathbf{k}, \omega) \frac{g(\omega)}{\omega} \quad \text{with} \\ C_{ijkl}(\mathbf{k}, \omega) &\propto \sum_{mnpq} p_{ijmn} p_{klpq} \Lambda_{mnpq}(\mathbf{k}, \omega) \quad \text{and} \\ \Lambda_{mnpq}(\mathbf{k}, \omega) &= \int d^3\mathbf{r}_1 d^3\mathbf{r}_2 e^{i\mathbf{k}(\mathbf{r}_2 - \mathbf{r}_1)} \\ &\times \frac{\langle \frac{1}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) s_{mn}^{\alpha}(\mathbf{r}_1) s_{pq}^{\alpha*}(\mathbf{r}_2) \rangle}{\langle \frac{1}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) \rangle}. \quad (2) \end{aligned}$$

The numerator of the integrand is the out-of-phase strain susceptibility $\chi_{mnpq}^{\prime\prime}(\mathbf{r}_1, \mathbf{r}_2, \omega)$, multiplied by ω , whereas

the normalization denominator is the density of states $g(\omega) = \langle \frac{1}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) \rangle$. Therefore the density of states drops out of the formula for the scattering intensity. This normalization denominator is also present in the expression given by [2] [their Eqs. (2) and (3)], so $g(\omega)$ drops out, and there is no proportionality between $I^c(\omega)$ and $g(\omega)$ even if $\mathbf{k} \neq \mathbf{0}$. One can rewrite the intensity, say, for polarized scattering to obtain

$$I_{VV}^c(\mathbf{k}, \omega) \propto [n(\omega) + 1] \chi_L^{\prime\prime}(\mathbf{k}, \omega) = S(\mathbf{k}, \omega), \quad (3)$$

where $\chi_L(\mathbf{k}, \omega)$ is the longitudinal strain susceptibility and $S(\mathbf{k}, \omega)$ is the usual scattering law. Because the susceptibility is proportional to k^2 this contribution is strongly suppressed in comparison with the incoherent one. This holds also for the other tensor components of $I_{ijkl}(\mathbf{k}, \omega)$.

In the Comment [2] it is pointed out that a disordered system does not exhibit *local* translational invariance and this is the origin of the boson peak. This is true and is, in fact, the basis of our model [5–8]. There it is shown that the boson peak marks the frequency, beyond which the vibrational excitations are no more weakly scattered plane waves.

We conclude by noting that the coherent contribution to the Raman intensity $I^c(\omega)$ proposed in [2] is zero. A generalized expression taking $\mathbf{k} \neq \mathbf{0}$ into account is strongly suppressed in comparison with the incoherent one, produced by fluctuating Pockels constants. Like the incoherent intensity $I^c(\mathbf{k}, \omega)$ is *not* proportional to the vibrational density of states.

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