

**Comment on “Raman Scattering and the Low-Frequency Vibrational Spectrum of Glasses”**

In a recent Letter [1], Schmid and Schirmacher reported a model of Raman scattering in glasses in the terahertz spectral range where the spectrum is dominated by the boson peak. In this model the light scattering arises due to spatial fluctuations of elasto-optic (Pockels) constants  $p(\mathbf{r}) = p^{(0)} + \Delta p(\mathbf{r})$ , while the main, nonfluctuating part of the constants  $p^{(0)}$  leads only to Brillouin scattering and Raman scattering from high-frequency optical modes. The authors claim that the Raman intensity  $I(\omega)$  is not proportional to the vibrational density of states  $g(\omega)$ , but to a convolution of Pockels-constant correlation functions with the dynamic strain susceptibilities of the glass.

In this Comment we would like to point out that the theory of Ref. [1] estimates only a part of the light scattering intensity, the part due to “electrical disorder.” There is another, “mechanical disorder” contribution that in fact may be dominating. This contribution corresponds to the nonfluctuating part of the Pockels constants  $p^{(0)}$  that was neglected in Ref. [1]. The light scattering on vibrations in the boson peak region is allowed even with the nonfluctuating Pockels constant if vibrations are not plane waves with definite wave vectors. Density and elastic constants fluctuations in glasses lead to deviations of the vibrational eigenfunctions from the plane waves. These deviations are not essential for long wavelength phonons because underlying Hamiltonian is translational invariant in average.

However, translational invariance is not held at small scales comparable to the size of glass heterogeneities or correlation radius of elastic constants fluctuations. This is manifested, in particular, by appearance of the boson peak. The mechanical disorder leads to the Shuker-Gammon expression for the light scattering intensity

$$I_{ijkl}(\omega) = C_{ijkl}(\omega)g(\omega)[n(\omega) + 1]/\omega. \quad (1)$$

Here,  $C_{ijkl}(\omega)$  is light-to-vibration coupling coefficient defined by the equation

$$C_{ijkl}(\omega)[n(\omega) + 1]/\omega \propto \sum_{mnpq} p_{ijmn}^{(0)} p_{klpq}^{(0)} \Lambda_{mnpq}(\omega), \quad (2)$$

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

where  $s_{mn}^{\alpha}(\mathbf{r})$  is the strain tensor corresponding to eigen-

mode  $\alpha$  with the Bose-factor included [see, e.g., Eqs. (15)–(17) in Ref. [2]. We note that in Ref. [2]  $\Lambda_{mnpq}$  is defined differently: it includes the factor  $g(\omega)$ ].

So defined coupling coefficient was estimated for the case of distorted plane waves in Ref. [3] and for non-propagating vibrations in Refs. [4,5]. The proportionality of the Raman intensity to the vibrational density of states was established also in Ref. [6]. Let us note that the Shuker-Gammon expression (1) is also valid in the model of Ref. [1]. In this model the fluctuating Pockels constants appear in the angle brackets in the denominator of Eq. (3) instead of  $p_{ijmn}^{(0)} p_{klpq}^{(0)}$  in Eq. (2). Although the mechanical contribution to the light scattering occurred less in magnitude than electrical disorder part in the case of distorted plane waves [3], even in this case it can be equal or larger for individual tensor components. Moreover, estimations of Ref. [2] showed that the mechanical disorder alone is enough to predict the right ratio of the boson peak to Brillouin line intensity in silica glass. Anyway, neglecting the mechanical disorder contribution to the boson peak intensity in light scattering makes the Letter [1] incomplete.

V.N. Novikov<sup>1</sup> and E. Duval<sup>2</sup>

<sup>1</sup>IA&E

Russian Academy of Sciences  
Novosibirsk, 630090, Russia

<sup>2</sup>Université de Lyon

Université Lyon-1, UMR5620 CNRS

Laboratoire de Physico-Chimie des Matériaux Luminescents  
Domaine scientifique de la Doua  
10 rue Ampère 69622 Villeurbanne France

Received 4 November 2008

DOI:

PACS numbers: 78.30.Ly, 65.60.+a, 78.20.-e

- [1] B. Schmid and W. Schirmacher, Phys. Rev. Lett. **100**, 137402 (2008).
- [2] V.N. Novikov, E. Duval, A. Kisliuk, and A.P. Sokolov, J. Chem. Phys. **102**, 4691 (1995).
- [3] J. Jäckle, in *Amorphous Solids: Low-Temperature Properties*, edited by W.A. Phillips (Springer, Berlin, 1981).
- [4] E. Duval, V.N. Novikov, and A. Boukenter, Phys. Rev. B **48**, 16785 (1993).
- [5] E. Duval, L. Saviot, N. Surovtsev, J. Wiedersich, and A. J. Dianoux, Philos. Mag. B **79**, 2051 (1999).
- [6] M. Montagna, G. Viliani and E. Duval, J. Raman Spectrosc. **27**, 707 (1996).