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2004 J. Phys.: Condens. Matter 16 223

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# Density of vibrational states and light-scattering coupling coefficient in the structural glass and glassy crystal of ethanol

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Received 27 September 2003

Published 9 January 2004

Online at [stacks.iop.org/JPhysCM/16/223](http://stacks.iop.org/JPhysCM/16/223) (DOI: 10.1088/0953-8984/16/3/003)

## Abstract

The density of vibrational states in the terahertz regime of ethanol in both the structural glass and the glassy crystal has been extracted from low-temperature specific heat data published previously via the Tikhonov regulation method. In addition, low-frequency light-scattering (LS) spectra (boson peak) have been measured for these two phases at low temperature (11 K) applying a tandem Fabry–Pérot interferometer. By comparing the LS data and the density of states, the light-vibration coupling coefficient is extracted. It is shown that the coupling coefficient is essentially the same for both disordered phases, obeying a linear frequency dependence down to 200 GHz.

## 1. Introduction

Vibrational motion in glasses around and below the terahertz (THz) range is associated with a number of interesting features: a non-Debye behaviour of the density of vibrational states at low frequencies giving rise to a peak in the heat capacity  $C_p/T^3$  at low temperatures (say,  $T \sim 5\text{--}10\text{ K}$ ) [1, 2], and an intense broad peak in the Raman spectrum as well as in the infrared absorption at low frequencies (boson peak) [3, 4]. These features coexist with an apparent linear dispersion curve of sound-like excitations [5], as well as a *universal* thermal conductivity qualitatively different from that found in crystalline solids [6, 7]. In other words, an excess density of states at low frequencies with respect to a Debye law is a characteristic feature of glasses. Understanding the THz dynamics of glasses has thus attracted the attention of many research groups and is currently one of the most challenging problems of glass physics.

An interesting direction in order to solve this problem is the investigation of positionally ordered, but orientationally disordered, solids, namely glassy crystals. We will use the term ‘glassy crystal’ [8] for kinetically frozen orientational disorder to distinguish it from the term

‘orientational glass’ traditionally employed for mixed crystals [9], where the orientational disorder has no kinetic origin. One of the most appealing substances in this respect is ethanol, which can be obtained as a fully ordered crystal, as an orientationally disordered crystal (glassy crystal) and as a structural (amorphous) glass [10]. The THz dynamics of the different phases of ethanol has been studied in several papers [11–14], but important questions about its vibrational dynamics remain unsettled. For example, accurately obtaining the vibrational spectrum from inelastic neutron scattering at low frequencies is a difficult task, and hence the calculation of low-temperature specific heat from the density of states as revealed by neutron scattering agrees only moderately well with experimental specific-heat data [12]. Here, knowledge of the low-frequency light-scattering (LS) spectrum measured at low enough temperature, for which relaxational motion may be ignored, offers another route for testing the vibrational density of states characteristic of glasses. Determination of the light-vibration coupling coefficient containing information about the vibrational correlator [15] is therefore of great interest.

The present work addresses the following issues: evaluation of the density of vibrational states from low-temperature specific-heat data and measurement of the low-temperature LS spectrum at low frequencies for the two disordered phases of ethanol. It will be shown that the coupling coefficient for the glassy crystal of ethanol is essentially the same as for the structurally disordered glass.

## 2. Raman coupling coefficient

Knowledge of the vibrational density of states and the LS spectrum allows one to determine the light-vibration coupling coefficient  $C(\nu)$  that is related to the LS intensity via the Shuker–Gammon formula [15, 16]

$$I(\nu) = C(\nu)g(\nu)\frac{n+1}{\nu}, \quad (1)$$

where the Stokes side of the spectrum is considered and  $n = 1/[\exp(h\nu/k_B T) - 1]$  is the Bose–Einstein occupation number for frequency  $\nu$  and temperature  $T$ .

Since the Raman coupling coefficient contains information about the vibrational correlator (further, if the total  $q$ -dependence of the coupling coefficient were known, the vibrational correlator could be found just by Fourier transformation [17]; unfortunately, in the experimentally accessible range of  $q$  the  $q$ -dependence is extremely weak [18]), there is a great deal of experimental and theoretical work devoted to the investigation of  $C(\nu)$  behaviour. Although, in general, without knowledge of the whole  $q$ -dependence there is no one-to-one correspondence between  $C(\nu)$  and the vibrational correlator, the experimentally found  $C(\nu)$  serves as a good test of different models. For example, shortly localized vibrations are expected to have  $C(\nu) \simeq \text{constant}$  [15], whilst a polarizability disorder for the case of scattering from acoustic plane waves predicts  $C(\nu) \propto \nu^2$  at low frequencies [19, 20], as well as in the case of scattering from spatially attenuated acoustic waves if the attenuation length  $l$  is  $l^{-1} \propto \nu^4$  [3, 21]; again  $C(\nu) \simeq \text{constant}$  in the case of the last mechanism, but if  $l^{-1} \propto \nu^2$  [21, 22], then within the fracton-like vibrational excitations model  $C(\nu) \propto \nu^x$  is predicted [23]. According to the results of [24],  $C(\nu) \propto 1/\nu$  for a model of isolated sphere-like clusters. The soft potential model in its simplest approximation [25] predicts  $C(\nu) \simeq \text{constant}$ , if only the excess over the Debye contribution is used as the density of states. The wide scatter in the frequency dependence of  $C(\nu)$  predicted in the different approaches allows one to make the distinction between the different models and explains the great importance of the study of  $C(\nu)$ . In particular, the transition between different frequency behaviours of  $C(\nu)$  can serve as a marker of the transition from one type of vibration to another (for example, in [26] the transition

to  $C(\nu) \simeq \text{constant}$  behaviour at  $110 \text{ cm}^{-1}$  in silica is interpreted as the onset of phonon localization).

Experimentally, it was found that the coupling coefficient of the boson peak in glasses has approximately linear frequency dependences in all cases [27–33]. Moreover, in a recent detailed study of  $C(\nu)$  in silica [34] it was discovered that the coupling coefficient should be described more precisely as  $C(\nu) \propto \nu + \text{constant}$ . In later studies this behaviour near the boson peak maximum was also found for a number of glasses [35, 36]. It is important to note that this behaviour can hardly be extended to zero frequency, since it was found that at frequencies below one-half or one-third of the boson maximum position this linear behaviour turns into a superlinear one [35]. We also note that there exists a difference in  $C(\nu)$  for polarized and depolarized spectra [37], but in the range of the boson peak maximum this difference has marginal influence for the whole picture of  $C(\nu)$  behaviour (for example, the polarized and depolarized  $C(\nu)$  for  $\text{SiO}_2$  glass are presented in [38], where it can be seen that this difference is not a dominant effect for the frequency dependence). Here it is remarked that the linear behaviour of  $C(\nu)$  has been found for glasses with very different levels of the Debye contribution [35]. Moreover, the ratio between the frequency-independent contribution and the linear term in the coupling coefficient at the boson peak maximum is approximately the same for different glasses, for which  $C(\nu)$  obeys a  $\nu + \text{constant}$  behaviour [35, 39]. This result supports the idea that all vibrations around the boson peak are hybridized and cannot be separated into propagating (Debye contribution) and localized ones.

The experimentally found linear frequency behaviour of the coupling coefficient challenges most of the theoretical models cited above. At the present time, the linear frequency prediction of  $C(\nu)$  is given mainly by the models with fractal-like or diffuse-like behaviour for vibrations [40–43]. Similar behaviour has been found in numerical calculations of percolator dynamics [44]. Thus, the linear behaviour of  $C(\nu)$  near the boson peak maximum can be mainly associated with the diffuse-like behaviour of the vibrations, although alternative viewpoints also exist [45].

*A priori*, it is not clear what the behaviour of the low-frequency vibrations should be in glassy crystals. Having a more ordered structure, glassy crystals could be characterized by *plane-wave* acoustic excitations as in fully ordered crystals. In this case, the mechanism of Raman scattering would be related to fluctuations of the polarizability, predicting a square dependence of  $C(\nu)$ . A linear behaviour of the coupling coefficient would suggest a similarity between the vibrational wavefunctions for the cases of the glassy crystal and structural glass. In the latter case, particular attention should be paid to the level of the frequency-independent contribution to  $C(\nu)$ : an increase or decrease of this contribution for the case of the glassy crystal in comparison with the usual glass would provide valuable information on its origin.

### 3. Experimental details

Ethanol was purchased from Aldrich (water content  $< 0.001\%$ ). The samples were sealed in Duran cuvettes after several freeze–pump degassing cycles, and then mounted in a helium cryostat. The structural glass of ethanol was obtained by cooling through the melting and glass transition temperatures ( $T_m = 159 \text{ K}$  and  $T_g = 97 \text{ K}$ , respectively) with a cooling rate about  $\approx 10 \text{ K min}^{-1}$ . In order to achieve the plastic crystal, the supercooled liquid was annealed at  $T = 106.5 \text{ K}$ . The transformation process was monitored by means of Brillouin-line measurements [46].

The difficulty with low-frequency, low-temperature LS experiments with ethanol is related to its low scattering power due to the absence of chemical groups with high anisotropic polarizability. We used the advantage of multichannel accumulation in a six-pass Sandercock

tandem Fabry–Pérot interferometer (TFPI) [47] that allows one to accumulate even weak LS signals, and at the same time has a high level of suppression of the elastic line. An Ar<sup>+</sup> laser at a wavelength of 514.5 nm and with a power of 250 mW was used as the pump source; details of the experimental setup are described elsewhere [48, 49]. The depolarized LS experiments with the TFPI were performed at  $\sim 170^\circ$  scattering angle. Double scan with a free spectral range (FSR) of 750 GHz was used in order to obtain the spectra in the 75–1500 GHz range.

As mentioned above, an important advantage of a Sandercock interferometer is its high suppression of the elastically scattered line. This was checked experimentally by measuring the spectra from a metal bar. The suppression obtained was better than  $2 \times 10^{-11}$  at a frequency shift higher than 15% of the FSR. The lowest level of inelastically scattered light from the ethanol sample in the Stokes side of all experimental spectra was higher than  $10^{-9}$  of its elastic line intensity. Therefore the elastic line suppression was good enough for our LS experiment.

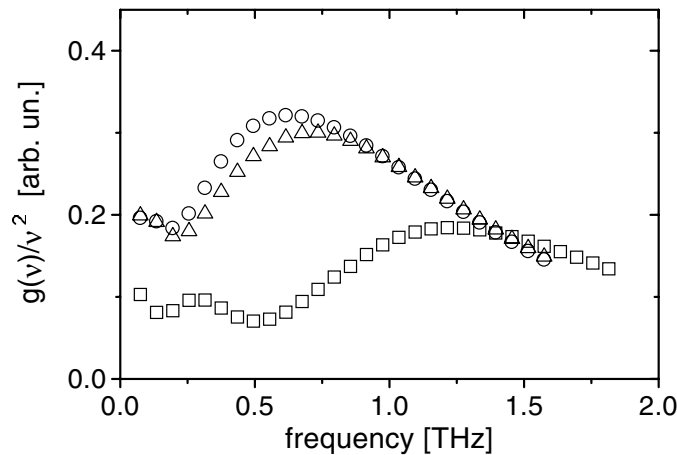
Another experimental problem caused by the low level of recorded LS is possible contributions from the glass material of the cuvette, especially in the near backscattering geometry used in our LS experiment. In order to reduce this effect we performed an *in situ* alignment in polarized geometry with a low FSR, so that the Brillouin lines from the ethanol and the cuvette are easily seen. Since they have quite different spectral positions, they are clearly distinguishable. The alignment of the optical scheme was then selected in such a way that we observed a maximal Brillouin line from the ethanol sample and a minimal one from the cuvette material. At the lowest temperature, an intensity ratio of 400 was achieved for both lines. The whole polarized spectrum for the glass cuvette covering both the Brillouin-line and boson peak was recorded separately. This comparison allowed us to conclude that for our best alignment the boson peak spectrum of the material of the cuvette contributed less than 1/250 to the ethanol spectrum. The real temperature in the illuminated part of the sample volume was determined from the Stokes/anti-Stokes ratio. The lowest temperature achieved was 11 K.

The vibrational density of states for the different ethanol phases was extracted from the low-temperature specific-heat data obtained by Talón *et al* [12, 14], by solving the corresponding integral equation using the Tikhonov regulation method as described in [38, 50]. The experimental specific-heat data reported in [12] were measured up to 30 K, which allowed us to extract a reliable spectrum of the density of vibrational states  $g(\nu)$  up to about  $\nu \approx 1.5$  THz.

#### 4. Results and discussion

The corresponding vibrational density of states  $g(\nu)/\nu^2$  for the three different ethanol phases as obtained from the specific-heat data [12, 14] is shown in figure 1. The displayed curves  $g(\nu)/\nu^2$  of the two disordered phases, namely the structural glass and glassy crystal, exhibit a boson peak maximum at  $\nu = 0.63$  and 0.71 THz, respectively. The density of states for the glassy crystal phase is somewhat lower than for the structural glass at frequencies below the boson peak maximum, which occurs at a slightly lower frequency for the structural glass. The corresponding maximum of the Debye-normalized density of states for the ordered crystalline phase is found at significantly higher frequencies (around 1.25 THz). This maximum reflects the van Hove singularity observed by inelastic neutron scattering at 6 meV [11]. (Strictly speaking the van Hove singularity is defined for an extremum in the density of states [51], but obviously it has some counterpart in the presentation  $g(\nu)/\nu^2$ .)

The three curves  $g(\nu)/\nu^2$  seem to follow the same qualitative behaviour: with increasing order the boson peak maximum shifts to higher frequencies and its magnitude decreases. This behaviour may be regarded as reminiscent of the idea of [52], where it was suggested that the boson peak in disordered systems is associated with the lowest van Hove singularity in their acoustic-like vibrational spectrum. Here we note that this approach of associating the boson

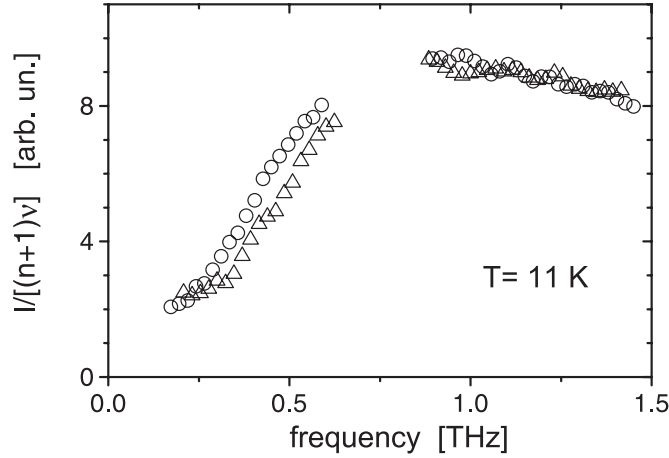


**Figure 1.** Density of vibrational states over frequency squared,  $g(\nu)/\nu^2$ , as obtained by deconvoluting specific-heat data for the different phases of ethanol. Squares correspond to the ordered crystal, triangles to the glassy crystal and circles to the structural glass.

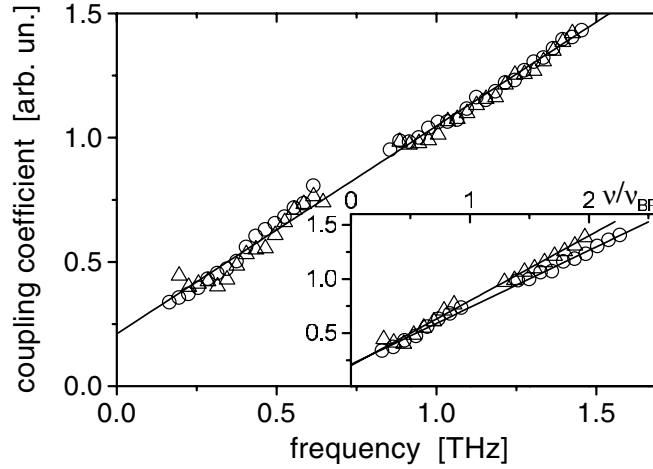
peak with a van Hove singularity has some relation to models where the boson peak is associated with the fundamental modes of structural nanoclusters in the framework of the approach of non-continuous glass nanostructure [53, 54]. Indeed, in a rough approximation the dynamics of a nanocluster mimics that of a pseudocrystal, whose lattice constant is equal to the nanocluster size [55]. By a pseudocrystal we mean here a regular packing of nanoclusters of approximately the same size. In this case, the lowest van Hove singularity will have a frequency about that of the fundamental mode of the nanoclusters. We should also mention here that figure 11 of [12] shows a scaling plot of the data  $(C_p/T^3)/P_c$  versus  $T/T_{\max}$ , where  $P_c$  is the height of the maximum in  $C_p/T^3$  and  $T_{\max}$  its position, for both normal and deuterated ethanol. This scaling follows the one employed by Liu and von Löhneysen [56] for many different glasses, and shows that the data for the two types of disordered solids are superimposed in the whole temperature region. In contrast, the curves for the ordered crystal show a far narrower shape, in agreement with similar findings in other materials [57]. This fact indicates the different nature of the low-energy vibrational spectra of glasses and ordered crystals.

Figure 2 displays the results of our LS experiments for the two disordered phases of ethanol, presented in spectral density form, that is  $I(\nu)/[\nu(n+1)]$  versus  $\nu$ . The spectral hole near 750 GHz corresponds to the first order of TFPI transmission that was cut from the spectra. Similarly to the density of states, the LS spectra of the structural glass and the glassy crystal are approximately the same in the range above the boson peak maximum; at lower frequencies the spectrum of the glassy crystal is weaker than that of the structural glass of ethanol.

The coupling coefficient  $C(\nu)$  for the disordered phases of ethanol was calculated from the LS spectra and the vibrational density of states via equation (1). The extracted coefficients are displayed in figure 3. It can be seen that  $C(\nu)$  for the two disordered phases of ethanol exhibits the same frequency dependence and the same amplitude. Moreover, the spectral dependence of  $C(\nu)$  for both ethanol phases is reproduced by a linear behaviour (the solid line in figure 3). A linear behaviour of the coupling coefficient for the vibrational spectrum at frequencies close to the boson peak maximum was found to be a universal feature of the structural glasses studied previously [35]. Here, we have found that the coupling coefficient for a substance characterized by long-range order, but with orientational disorder, exhibits the same behaviour as structural glasses. Since the coupling coefficient is related to the vibrational wavefunction [15], the



**Figure 2.** Low-frequency LS spectra of the two disordered phases of ethanol at  $T = 11$  K: glassy crystal (triangles) and structural glass (circles). The spectral intensity is normalized at the high-frequency part of the spectrum.



**Figure 3.** Coupling coefficient  $C(\nu)$  of the two disordered phases of ethanol: glassy crystal (triangles) and structural glass (circles). The solid line is a linear fit. The inset shows  $C(\nu)$  versus the frequency scaled by the boson peak position  $\nu_{BP}$ . Solid lines are linear fits:  $C(\nu) \propto \nu/\nu_{BP} + B$ , where  $B = 0.4$  and  $0.33$  for structural glass and glassy crystal, respectively.

present result demonstrates that the vibrations corresponding to the boson peak have essentially the same wavefunction in both glassy crystals and structural glasses.

In previous work [35], it was shown that the linear spectral dependence of the coupling coefficient studied in several glasses can be scaled by the frequency of the boson peak maximum in  $g(\nu)/\nu^2$ . Two typical cases were found for the linear dependence of  $C(\nu)$  if scaled by the boson peak maximum  $\nu_{BP}$  according to

$$C(\nu) \propto \nu/\nu_{BP} + B. \quad (2)$$

One case is when  $B \approx 0.5$  (glasses of type I) and the second case is when  $B \approx 0$  (glasses of type II). The inset of figure 3 shows the coupling coefficient for the two disordered phases of

ethanol in the scaled presentation. It can be seen that the structural glass could be ascribed to type I, since it has a non-zero constant  $B \approx 0.4$ . The coupling coefficient of the glassy crystal also behaves similarly to glasses of type I, but has a lower constant  $B \approx 0.33$ .

Note that the coupling coefficients of the two phases of ethanol superpose on each other when they are plotted versus absolute frequency (figure 3), but they look different when presented versus the scaled frequency  $\nu/\nu_{BP}$  (inset of figure 3). This feature distinguishes the present case from the changes in the vibrational density of states observed in ordinary glasses induced by thermal annealing [39] or permanent densification [58, 59]. Indeed, at first glance the transformation of the vibrational density of states implied by the transition from the structural glass to the glassy crystal phase (figure 1) is very similar to the changes in  $g(\nu)$  induced by permanent densification of  $\text{SiO}_2$  glasses [58, 59] or by long annealing near the glass transition temperature of  $\text{B}_2\text{O}_3$  glasses [39]. However, as shown in [39],  $C(\nu)$  is different for structural glasses with different extents of annealing but falls onto a single spectral curve after the frequency is scaled by the boson peak position. This means that if one takes the position  $\nu_{BP}$  as a parameter influenced by the thermal treatment, then the boson peak wavefunction can be scaled by this parameter. The case of comparing a structural glass with a glassy crystal appears to be different, however. It seems that the correlation between the boson peak wavefunction and the boson peak position is slightly different for glassy crystals than for structural glasses, and does not exhibit the above-mentioned scaling with  $\nu_{BP}$  (figure 3).

## 5. Conclusion

We have presented a study of the THz vibrations (boson peak) in the different phases of ethanol. The vibrational density of states has been extracted from the corresponding low-temperature specific-heat data published previously and compared with the low-frequency light-scattering spectra as measured by applying tandem Fabry–Pérot interferometry. It was found that the vibrational spectra are very similar for both disordered phases of ethanol, namely the structural glass and glassy crystal, the latter exhibiting only slightly lower values at the low-frequency side of the boson peak. The coupling coefficient obtained is basically the same for both phases. If scaled by the boson peak position  $\nu_{BP}$ , it is well described by the linear frequency dependence  $C(\nu) \propto \nu/\nu_{BP} + B$ , where  $B \approx 0.4$  and  $0.33$  for the structural glass and glassy crystal, respectively. This result leads to the conclusion that the boson peak wavefunction in the glassy crystal and that in the structural glass are essentially the same.

## Acknowledgments

This work was supported by RFFI Grant No 02-02-16112. MAR acknowledges financial support from MCyT (Spain) within project BFM2000-0035-C02.

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