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High-frequency excitations in glassy crystals

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1. Introduction

The high-frequency excitations in glasses and supercooled liquids belong to the great mysteries in the physics of condensed matter. While dielectric spectroscopy is a paramount technique for the investigation of glassy dynamics at lower frequencies, only in recent times experimental advances enabled the systematic investigation of the high-frequency region of about GHz–THz using this technique and thus most experimental results on the high-frequency dynamics in glass formers so far were obtained using scattering techniques as light and neutron scattering. It is well established now that at frequencies around 100 GHz, a minimum appears in the imaginary part of the susceptibility. The existence of an excess contribution in this region can be interpreted as the signature of a fast process originating, e.g., from encaged molecular motion as predicted by the mode coupling theory (MCT) of the glass transition [1], but also other explanations have been suggested [2,3].

Also the origin of the boson peak (BP), dominating at THz frequencies, still is controversially debated and in recent years a vast number of investigations has been devoted to this intriguing phenomenon [4–17]. The term ‘boson peak’ was established to characterize the intensity, in excess of the Debye vibrational density of states (DOS), observed in Raman experiments on disordered matter at low-frequencies, which reveals a temperature dependence in accord with the Bose–Einstein statistics [18]. The existence of excess low-frequency modes in disordered matter is also well established from infrared (IR) spectroscopy. A broad peak in the far IR absorption of dipolar liquids has been detected by Poley [19]. Amongst the early IR works on liquids, supercooled liquids, and glasses, it was Whalley [20] who has stressed the close relation of this excess absorptivity in IR experiments and the Raman scattering results. Since then this characteristic Poley absorption has been detected in a large number of molecular liquids, supercooled liquids, and glasses and was revealed to be closely related to the BP [21]. While a number of IR experiments were carried out on glasses (see, e.g., [22–25]), however no systematic investigations and analyses of the absorption processes in the BP regime have been performed.

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Aside of the canonical glasses, during recent years the so-called plastic crystals (PCs) have attracted considerable interest (see, e.g., [26] for an overview). In PCs the centers of mass of the molecules form a regular crystalline lattice but the molecules are dynamically disordered with respect to their orientational degrees of freedom. Due to their translational symmetry, PCs are much simpler to treat in theoretical and simulation approaches of the glass transition and therefore these materials are often considered as model systems for structural glass formers. Indeed the orientational relaxation dynamics of PCs in many respects resembles the relaxation dynamics of conventional glass formers. Especially, in many cases by sufficiently fast cooling, the high-temperature dynamically disordered state can be supercooled, thus avoiding complete orientational ordering often occurring at low-temperatures, and concomitantly the dynamics shows the continuous slowing down over many orders of magnitude that is typical also for glass-forming liquids. There are also some investigations of the high-frequency dynamics of PCs, using scattering techniques or molecular dynamics simulations (e.g., [27–29]); for example, neutron scattering on ethanol revealed a BP also in its plastic phase [27].

While, as mentioned above, several dielectric investigations of the high-frequency dynamics of canonical glass formers have appeared in recent years [25,30–32], corresponding reports on PCs are scarce and not much is known about the dielectric high-frequency response of PCs [33,34]. Having in mind the well-defined phonon modes in PCs compared to canonical glasses, and the infrared silence of acoustic phonon modes, such investigations can provide valuable hints, e.g., on the origin of the BP. In the present work we have investigated *ortho*-carborane (OCA), a prototypical PC [35,36]. The nearly spherically shaped OCA molecule, $B_{10}C_2H_{12}$, forms a rigid icosahedron whose corners are occupied by ten boron and two carbon atoms. Overall, the carborane molecules are nearly spherically shaped and therefore experience little steric hindrance for reorientational processes, thus favoring the forming of a plastic-crystalline phase. Near 275 K, OCA undergoes a phase transition where the reorientational motion is partly restricted [37–41]. The complete orientational order reported at $T \leqslant 165$ K [39,42] can be easily suppressed already by moderate cooling rates. From the temperature dependence of the α-relaxation time [8] and from calorimetric [35,42] measurements a 'glass' temperature, T_g 134 K can be deduced. Our results provide clear evidence for a fast process and a BP-like feature also in PCs. We compare the results to those obtained in the canonical low-molecular weight glass former glycerol.

2. Experimental procedures

Measurements in an exceptionally broad frequency range were performed by a combination of different experimental techniques, employing coaxial reflection and transmission techniques, submillimeter wave spectroscopy, and Fourier transform infrared spectroscopy.

For the IR experiments, the transmission of commercially available OCA powders, pressed into pellets, was determined by Fourier transform spectroscopy. The phase shift was calculated by a Kramers–Kronig transformation. The required low-frequency extrapolation was based on the measured dielectric data, while at high-frequencies a $\nu^{0.5}$ power law extrapolation was used. By taking into account multiple reflections, the complex permittivity $\varepsilon' \;\; i\varepsilon''$ was obtained. In some cases, especially at low-frequencies, oscillations occur in the data due to spurious multiple reflections, caused, e.g., by the cryostat windows. Due to a very low-transmission, in the immediate vicinity of the detected $\varepsilon''(\nu)$-peaks the absolute values have a large experimental uncertainty.

For details on the dielectric and submillimeter wave experiments, see [43].

3. Results

Fig. 1 shows a comparison of the broadband dielectric loss spectra of the glass-forming liquid glycerol [25,44] and the PC OCA [45] for a variety of temperatures. While the overall behavior is qualitatively similar in many respects, also marked differences show up. Common to both spectra is the dominating α-peak, which marks the characteristic time scale of the reorientational dynamics of the dipolar molecules. In contrast to OCA, in glycerol a second power law shows up on the high-frequency flank

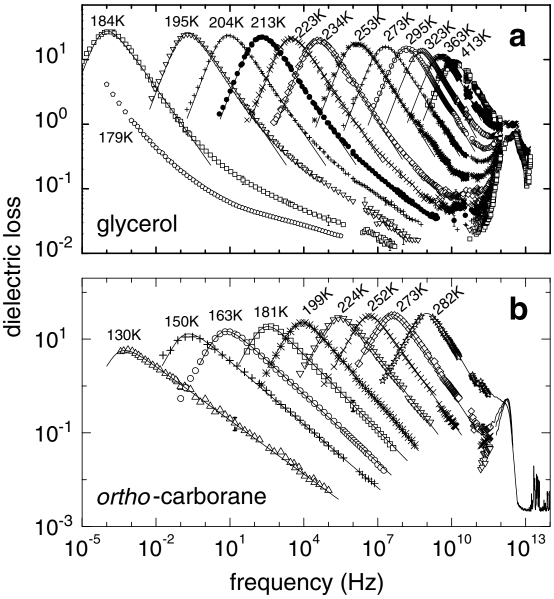

Fig. 1. Frequency dependence of the dielectric loss in glycerol [25,44] (a) and OCA [45] (b) at various temperatures (to keep the figures readable, at high-frequencies not all data points are shown; in (b) at $\nu > 400$ MHz the data are shown as solid lines and at $\nu > 3$ THz only data for 282 K are shown). The sub-T_g curve at 179 K in (a) was measured in thermal equilibrium after five weeks aging time [44]. The lines are fits of the peak region with the CD function.

of the α-peak. The temperature-dependent shift of the α-peaks through the frequency window mirrors the continuous slowing down of the glassy dynamics when approaching the glass temperature [25,26]. It is well known that the α-relaxational dynamics in plastic crystals, reflecting reorientational molecular motions, exhibits analogous behavior to that in canonical glass formers, where it is coupled to translational motions, a fact that led to the establishment of plastic crystals as model systems for canonical glass formers.

As revealed by Fig. 1, this analogy seems to prevail even for the high-frequency dynamics: Around 100 GHz, very similar to glycerol and other canonical glass formers [25,32], OCA exhibits an $\varepsilon^{00}(\nu)$-minimum around 100 GHz Another common feature of both materials is an almost temperature-independent high-frequency peak showing up at about 2 THz. However, its shape seems to be different for both materials, being more symmetric for glycerol but exhibiting a much steeper high-frequency flank for OCA. At higher frequencies, further much sharper peaks show up. These resonance-like modes can be ascribed to intramolecular excitations.

4. Discussion

In Fig. 1, in contrast to OCA, in glycerol a second power law shows up on the high-frequency flank of the α-peak. This feature which was termed ‘excess wing’ in [46], is found in a variety of glass formers [25,47], but is not observed in PCs [26,46]. As there is ample evidence in recent literature that the excess wing is due to a secondary relaxation process [44,48], it seems that in PCs this relaxation is absent or at least has a much lower amplitude than in the canonical glass formers [26]. So far, there is no commonly accepted microscopic explanation of the origin of this relaxation or the reason for its absence in PCs. It also is unclear if it can be identified with the well-known Johari–Goldstein β-relaxation processes [49], observed in many glass formers or if is a separate phenomenon adding another member to the zoo of dynamic processes in glass-forming materials.

As shown in Fig. 2, similar to glycerol, plastic crystalline OCA shows clear indications for an additional fast process in the region of the loss minimum. In contrast to the findings in [34] based on a more restricted dataset, at the lower temperatures a simple superposition of two power laws, determined by the high-frequency wing of the α-peak and the increase towards the peak in the THz region (dashed lines in Fig. 2), cannot describe the experimental data satisfactorily. These results give a strong hint that also in plastic crystals there is an additional fast process contributing to the loss in this region. Indications for such a contribution were also found in the PC cyclo-octanol [33]. To account for the excess intensity in the minimum region without invoking a specific model, the experimental data of Fig. 2 were parameterized by adding a constant to the two power laws (solid lines). It should be noted that such a constant-loss contribution was also employed to describe the high-frequency response in other materials, including PCs [28] and canonical glass formers [3,31]. In the latter materials, the fast process often is ascribed to the motion of molecules in the cage formed by the adjacent molecules, as described in detail by the MCT [1]. Indeed the spectral shape and temperature dependence of the dielectric loss minimum has been shown to be in good accord with the predictions of the MCT [50]. Thus, in light of the results of Fig. 2, one may ask if there is an analogue to the cage effect in PCs and in fact some strong indications for the applicability of the MCT to PCs were obtained recently [29,51]. For a quantitative comparison of the dielectric results with MCT predictions, similar to that performed in glycerol [31], a more complete set of data is necessary.

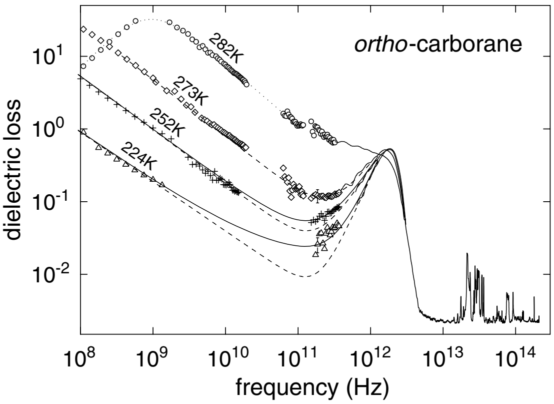

Fig. 2. Dielectric loss spectra of plastic-crystalline OCA for various temperatures. To maintain readability, at $\nu > 400$ GHz the data are shown as solid lines and at $\nu > 3$ THz only data for 282 K are shown. The dashed lines are calculated by a superposition of two power laws; the solid lines comprise an additional constant loss contribution. The dotted line is a guide to the eye.

The peak showing up near 2 THz in OCA (Fig. 2) is rather broad, with a relatively moderately increasing low-frequency wing, and thus clearly is not due to a resonance-like excitation, as, e.g., a phonon mode or an intramolecular vibration. The mere fact of the appearance of this peak also in the spectrum of a PC demonstrates that it is a spectral feature inherent to disordered matter. For glycerol it was shown that this peak corresponds to the BP, observed in scattering experiments [25], which is demonstrated in Fig. 3(a) where the dielectric results in the peak region are compared to the imaginary part of the susceptibility obtained from light scattering experiments by Wuttke et al. [52]. However, one should have in mind that the excess heat capacity, which in canonical glass formers arises due to the boson peak excitations, in partly orientationally disordered OCA is similar to that observed in fully ordered *meta*-carborane [42]. Hence it is clear that librational excitations coupled to acoustic modes must play an essential role in establishing the THz peak in OCA.

In the present work the term ‘boson peak’ is used in a somewhat sloppy way to denote the high-frequency peak universally appearing in the susceptibility spectra of glassy

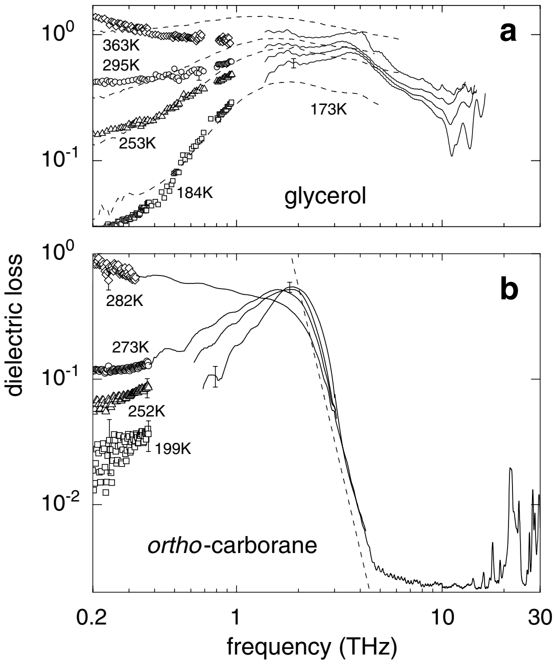

Fig. 3. Dielectric loss spectra in the IR region of the structural glass former glycerol (a) and plastic crystalline OCA (b) at various temperatures. For OCA, at $\nu > 3$ THz only data for 282 K are shown, the temperature dependence in this region being negligible. In (a) additional light scattering data are shown [52] (dashed lines); this data set was shifted vertically to result in a comparable intensity of the peak amplitude. The dashed line in (b) indicates a $\nu^{\ 7}$ decrease.

matter. However, one should have in mind that in literature, the BP is often defined as a peak in the DOS divided by the squared frequency, $g(\omega)/\omega^2$, i.e., denoting an *excess* contribution over the Debye vibrational DOS. Fig. 4 demonstrates for the lowest temperatures investigated, where contributions from α-relaxation and fast process can be neglected, that the detected BP-like feature in PCs indeed also corresponds to a peak in the quantity $\varepsilon'' T/\omega$, which is a good approximation of $g(\omega)/\omega^2$. In most reports on the BP of canonical glass formers, $g(\omega)$ is extracted from neutron or light scattering experiments, which mainly couple to translational degrees of freedom. However, it was demonstrated [25] that IR spectroscopy, despite coupling predominantly to reorientational motions, leads to a very similar spectral shape of the BP.

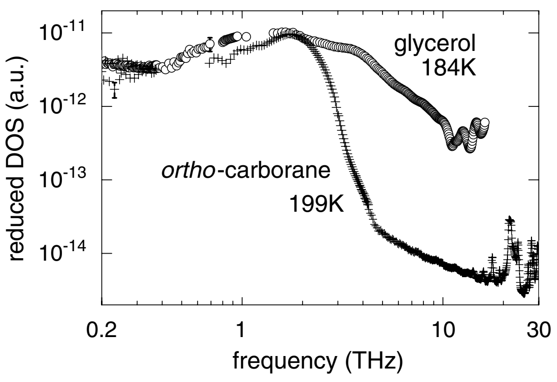

Fig. 4. Spectra of $\varepsilon'' T/\omega$ in the THz region for glycerol and plastic-crystalline OCA at the lowest temperatures investigated. The plotted quantity is approximately proportional to the reduced DOS, divided by the squared frequency, $g(\omega)/\omega^2$.

In Figs. 3 and 4 the BP in the canonical glass former glycerol is compared to the corresponding peak in OCA. At low-temperatures where any influence from processes at lower frequencies can be excluded, the low-frequency wing of the THz-peak for both materials looks quite similar, increasing with an approximate power law $\varepsilon'' \ / \ \nu^n$ with an exponent n of 1–1.5. However, following an approximate power law with exponent of 7 (dashed line in Fig. 3(b)), the high-frequency wing for the plastic crystal exhibits a much steeper decrease than for glycerol, where $\varepsilon''(\nu)$ decreases with similar slope as on the left wing, before the intramolecular modes come into play above about 10 THz. This strong dissimilarity of the spectral shape gives some hint that phonon like excitations play a role in the formation of the BP: At first, in contrast to canonical glass formers, in PCs there are well-defined phonon modes. In addition, OCA can be viewed analogous to a monoatomic crystal: It consists of hard spheres of $B_{10}C_2H_{12}$ with strong intramolecular bonding, which are linked to each other by relatively weak van der Waals bonds. Thus it seems reasonable to expect a purely acoustic Debye phonon DOS for this material, while in the hydrogen-bonded glycerol also optic modes should play a role. It is likely that these differences cause the different spectral shapes of the BPs in both materials. A possible explanation for the strong asymmetry of the peak in OCA emerges invoking ideas that the BP may simply reflect the complete phonon DOS. Then the high-frequency cut-off of the Debye DOS is the cause of the observed strong decrease at the high-frequency wing of the THz-peak. It was speculated already long ago (e.g., [24]) that a coupling of the full acoustic DOS to the IR radiation can be achieved via a disorder-induced breaking of selections rules, the disorder in the present case arising from the orientational degrees of freedom of the molecules. Alternatively one can assume the BP becoming IR active via the strong hybridization of collective phonon modes with local relaxational excitations [15,53,54]. This explains also the occurrence of contributions in *excess* to those expected for a Debye DOS, as commonly detected in Raman, NS, and specific heat experiments, which cannot be rationalized within the first scenario.

In contrast, in canonical glass formers all phonon modes, considerably smeared out due to the lack of long-range translational order, and low-frequency intramolecular modes mix up to form a peak with a much softer high-frequency wing. Also optical phonon modes may play a role here, the observed multi-peak structure (Figs. 3(a) and 4) resulting from the complete vibrational DOS. Nevertheless, it is reasonable that the low-frequency part is determined by the acoustic DOS and thus is similar to that of the plastic crystals. The mere fact that we detect a BP with dielectric spectroscopy, which is only sensitive to molecular reorientations, indicates that it is not exclusively due to acoustic phonon modes. Instead, either local relaxational or collective optic modes must play a role and acoustic modes are involved most likely via hybridization with reorientational modes.

5. Conclusions

In conclusion, we were able to demonstrate that the typical high-frequency behavior of canonical glass formers, namely a relatively broad peak close to 2 THz, preceded by an excess contribution in the minimum region around 100 GHz, also occur in the dielectric loss spectra of the plastic crystal OCA. The latter finding, together with those on cyclo-octanol [33], demonstrates for another class of materials, namely the plastic crystals, that the occurrence of excess intensity in the region of the $\varepsilon''(\nu)$ minimum is indeed a universal property of *all* disordered matter. This notion was promoted in [55], based on measurements on canonical glass formers and strongly doped semiconductors. For glass-forming liquids, the MCT predicts contributions from a fast process in this region, in reasonable accord with the experimental results; and there are indications that this may also be the case for PCs [29,51].

From the excess heat capacity of OCA, it is clear that librational-like modes must contribute to the THz peak [42]. Nevertheless, the detected strong asymmetry of the BP in the orientationally disordered crystal OCA, which is in marked contrast to the spectral shape found in canonical glasses, in accord with arguments in favor of a mainly acoustic character of phonon modes in the investigated PCs, strongly indicate that acoustic modes dominate the BP in OCA. The fact that these modes are infrared active confirm that there is a strong mixing of relaxational localized and phonon-like extended modes, leading to the observed contributions in excess to the Debye DOS. It is reasonable that similar effects are also responsible for the BP of structurally disordered materials.

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