

RESEARCH ARTICLE | APRIL 15 1999

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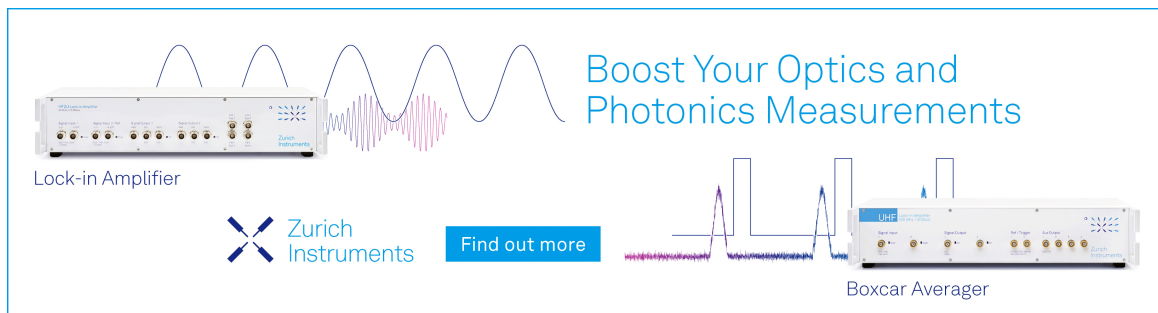
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AIP Conf. Proc. 469, 671–672 (1999)


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High frequency dielectric response of plastic crystals

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Abstract. The dielectric loss in the plastic crystalline phases of ortho-carborane and cyclo-octanol is presented at frequencies 1 MHz - 300 GHz. Both materials show qualitative differences concerning the high-frequency wing of the α -relaxation peak. Near 100 GHz, for both materials a minimum is observed, similar to the findings in structural glass formers.

In plastic crystals (also called rotor or glassy crystals) the centers of mass of the molecules form a regular crystalline lattice but their orientational degrees of freedom are dynamically disordered. Plastic crystals (PCs) are often considered as model systems for structural glass formers (SGFs). Here we present first high-frequency dielectric results on the PC ortho-carborane (o-CA) and compare it with the results on cyclo-octanol (c-OCT) [1]. The MHz-THz frequency region is of special interest due to the predictions of new theoretical and scaling approaches (see, e.g., [2,3]). For experimental details, see [4].

Fig. 1 shows the frequency dependence of the dielectric loss ε'' of o-CA and c-OCT at frequencies above 1 MHz. Marked differences in the high-frequency wing of the α -relaxation peak show up: While in c-OCT an excess wing (a second, more shallow power law, $\varepsilon'' \sim \nu^{-b}$) as known from SGFs [3] seems to be present, no similar feature can be detected in o-CA [5]. However, as was shown earlier [1], the apparent excess wing in c-OCT can presumably completely be ascribed to a secondary relaxation process due to intramolecular degrees of freedom. The o-CA molecule, $B_{10}C_2H_{12}$, forms an almost regularly shaped icosahedron without intramolecular degrees of freedom. From this observation we conclude that in PCs with rigid molecules no high-frequency wing exists and that the excess wing is a universal feature of SGFs only. For a more detailed treatment of the excess wing in PCs, see [5].

For both materials a minimum in $\varepsilon''(\nu)$ is observed. The solid lines in Fig. 1(a) are fits with the superposition ansatz $\varepsilon'' = CD + c_n \nu^n$ with $n = 1$. Here "CD" schematically denotes the empirical Cole-Davidson function for the description of the α -peak. The second term takes account of the low frequency wing of the boson or microscopic peak that can be expected in the THz region. For c-OCT [Fig. 1(b)],

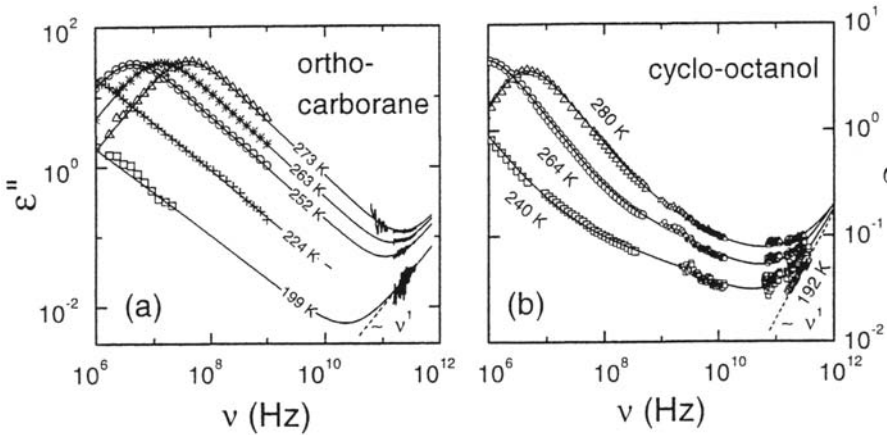


FIGURE 1. $\epsilon''(\nu)$ of o-CA (a) and c-OCT (b) at various temperatures. The solid lines are fits with a phenomenological ansatz (see text). The dashed lines indicate a linear increase towards the boson peak.

the solid lines were calculated with $\epsilon'' = CD + CC + \epsilon_c + c_n \nu$. Here "CC" denotes a Cole-Cole function for the description of the secondary process [1]. The constant loss ϵ_c was included to take account of the rather shallow minimum in c-OCT and may be indicative for additional fast processes in this region. For SGFs, the mode coupling theory (MCT) of the glass transition [2] predicts contributions from a fast process in this region. However, for PCs other scenarios may be applicable [2]. In conclusion, we have provided experimental evidence that the α -relaxation in o-CA can be described by a CD type of spectral shape without an excess wing. High frequency processes may be present in c-OCT, however, more measurements are needed to make a conclusive statement about the presence or absence of additional fast processes in PCs.

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