HD-187: Study of High-Pressure High-Temperature fluid carbon dioxide by inelastic x-ray scattering -- Allocated time: 18 shifts at ID16

The present experiment was the continuation of a previous one (HD-73) whose first aim was the study of the pressure and temperature dependence of the high frequency dynamics in fluid carbon dioxide. The interest of this study resides first of all in the fact that the relaxation dynamics of this kind of systems (linear non-polar molecule with quadrupolar interaction) has not been investigated yet, and, second, in the possibility of extracting the viscosity from a viscoelastic analysis of the data in a (P,T) range of geophysical interest, i.e. the upper mantle conditions (P < 10 GPa and T < 1500 K).

The measurements were performed on samples loaded in diamond anvil cells at a constant temperature of 650 K. We investigated two pressure points, 2.6 and 7 GPa. The former was chosen in order to isolate the temperature dependence, as it has the same density of the highest pressure point measured in the previous experiment, i.e. 2 GPa at 450 K. The latter was measured on the over-compressed fluid, beyond the freezing point and it corresponded to a density increment as large as 20%. Two sets of exchanged momentum q were measured at the lowest pressure and three at the highest, up to 23.5 nm^{-1}, for an integration time of 800 s per energy point. Two shifts of the allocated beamtime were devoted to the empty cell measurements.

While in the previous experiment we found that the empty cell contribution to the elastic line was dominant, this time it turned out to be nearly absent. Given that the diamonds, though different, were of the same quality, the reasons of such a difference are still unknown, but they could reside in a dramatic dependence of the signal on the diamonds orientation. A clear understanding of the factors affecting the empty cell contribution is however still missing and requires further studies.

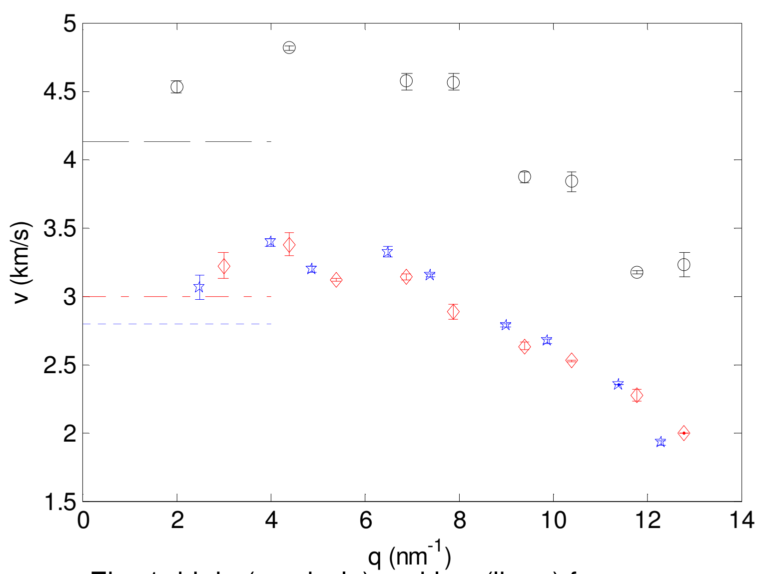

Fig. 1: high (symbols) and low (lines) frequency sound velocity: circles, dashed line: 650 K 7 GPa, diamonds, dot-dashed line: 650 K 2.6 GPa, stars, dotted line: 450 K 2 GPa

The spectra were fitted to a model with two relaxation times, as there was evidence of the presence of a structural relaxation at both the pressure points.

From a preliminary analysis we found that the phonon dispersion is temperature independent at least in the temperature range here explored. Indeed, a good correspondence is observed between our lowest pressure point and the highest pressure point investigated in the previous experiment, which are at the same density (Fig. 1). The high frequency velocity is therefore only density-dependent, differently from the low frequency one, and so is the relaxation time.

Conversely, the pressure affects both the amount of the positive dispersion observed in the sound velocity, and the relaxation time which increases with density, as expected.

A more refined analysis to obtain the density and temperature dependence of the viscosity is under way.