# Experimental report

14/02/2017

**Proposal:** 6-01-330 **Council:** 4/2016

**Title:** Collective dynamics of fluid Argonin the supercritical regime

**Research area:** Physics

**This proposal is a resubmission of 6-01-329**


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**Samples:** 36Ar

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| IN6 | 7 | 6 | 09/11/2016 | 15/11/2016 |


**Abstract:**

We propose to measure the dynamic structure factor of fluid Ar in the supercritical regime along two isotherms, namely 295 K and 250 K, and for pressures between 0.1 and 3 kbar. We want to verify the correlation between the appearance of the positive sound dispersion (the difference between the unrelaxed and the hydrodynamic value of the longitudinal sound velocity) and the crossing of the Widom line
recently observed by MD simulations (G. Simeoni, et al., Nature Phys. 6 (7), 503 (2010)).

We have performed an INS experiment on supercritical natural Neon measuring the inelastic spectrum at several densities along two isotherms at 300 K and 100 K, as shown in figure 1, to check if the positive sound dispersion (PSD), i.e. the enhancement of the THZ sound velocity with respect to its adiabatic value, could allow distinguishing between gas-like and liquid-like behavior in the supercritical regime (ref. 1-5). The critical point of Neon is located at: Critical Temperature= 44.34 K; Critical Pressure = 26.79 bar. This means that the two isotherms are in deep supercritical conditions: T/Tc= 6.7 and 2.2. We did not measure the proposed purely coherent neutron scatterer $Ar^{36}$ (as mentioned in the proposal) because the actual price of this isotope was extremely high and we thus chose natural Ne because of its low values of both incoherent and absorption cross sections ($s_{inc}$=0.008 and $s_{abs}$ = 0.039, respectively, while $s_{coh}$ = 2.628 barn). The measured thermodynamic points are showed in the attached plot showing the density evolution with pressure along the two isotherms according to NIST and the locations of the Frenkel line ($C_V=2k_B$) (ref. 6,7) and extrema of thermodynamic response functions like isobaric heat capacity, thermal diffusivity and kinematic shear viscosity (Widom line).

The purpose of the experiment was to detect the onset of the positive sound dispersion with pressure along the two isotherms. This is expected to take place at densities where the supercritical fluid properties change from gas-like to liquid-like. Unfortunately due to kinematic constraints, to the presence of a broad quasi elastic peak and to the damping of the acoustic peak at low densities, we were unable to properly measure the acoustic peak at any pressure.

Nevertheless we observe changes in the spectrum with pressure at both temperatures. In particular we were measuring two types of spectra: spectra at low densities are different from spectra at high densities. More in detail the INS spectra show a broad and quite symmetric profile at low densities, while at high densities they show an overall intensity decrease, with a pronounced peak only on the anti Stokes side (see figure 2). It is important to notice that these spectral

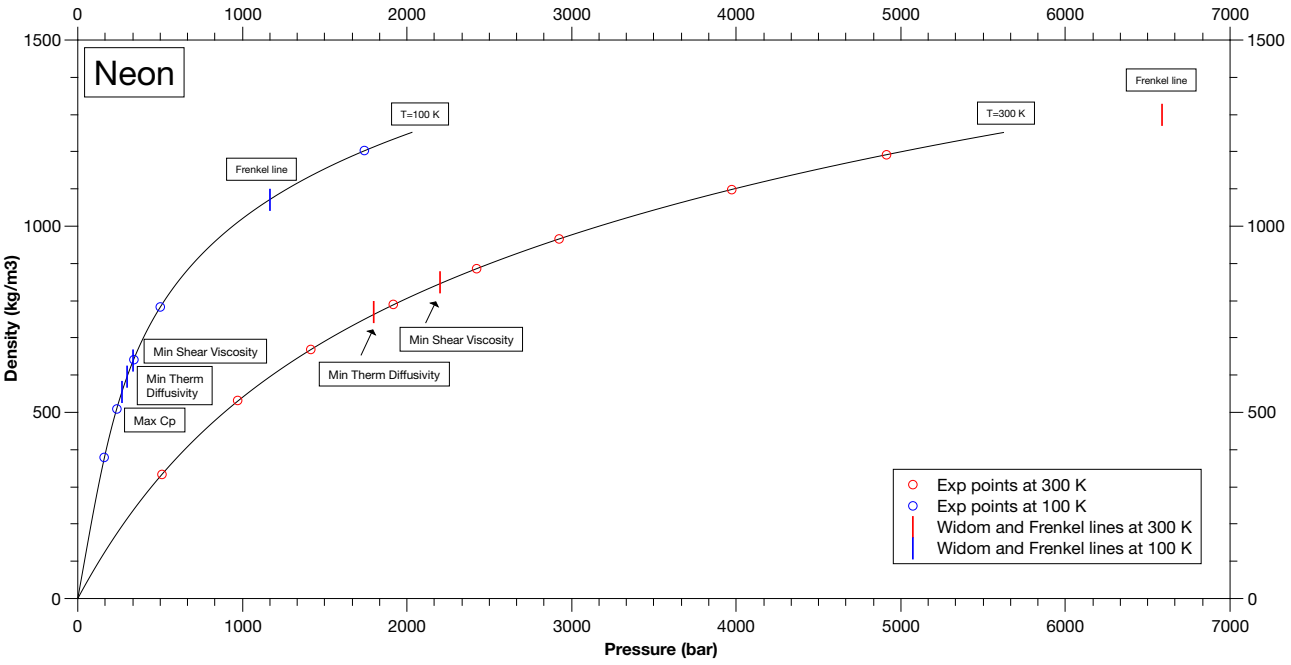


***Figure 1. Measured experimental points along the two isotherms at 100 and 300 K.***

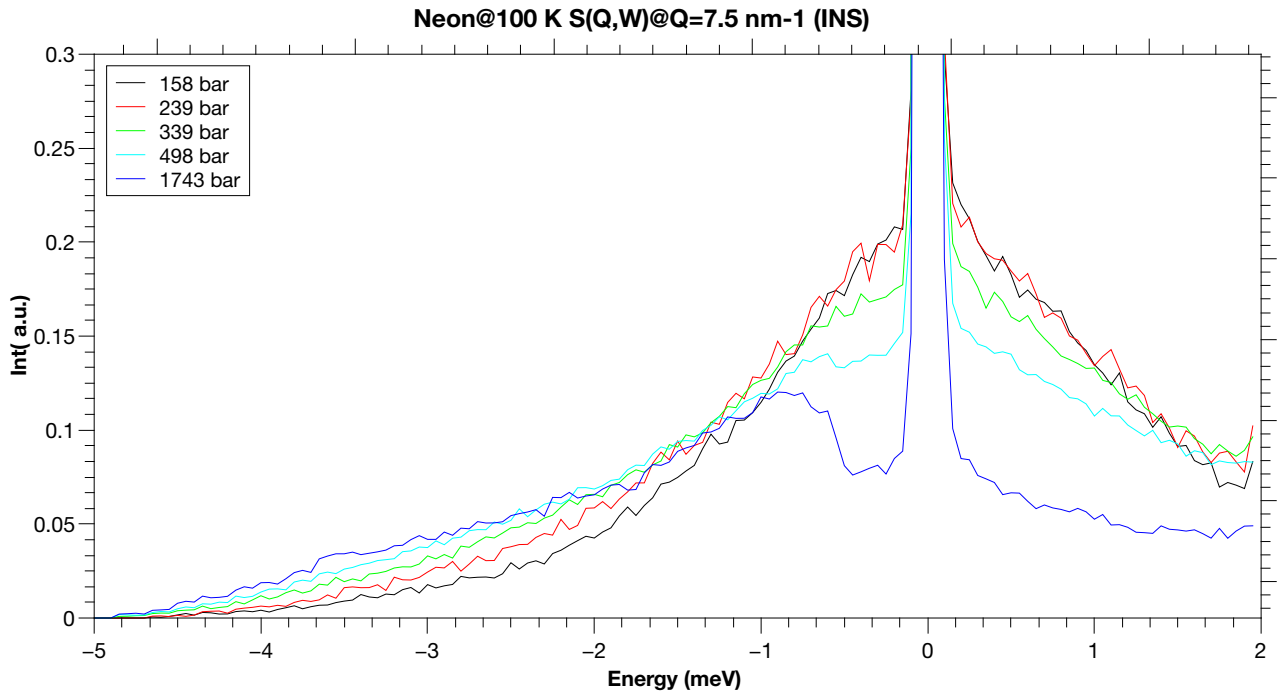


***Figure 2. Measured spectra at Q=7.5 nm$^{-1}$ along the isotherm at 100 K.***

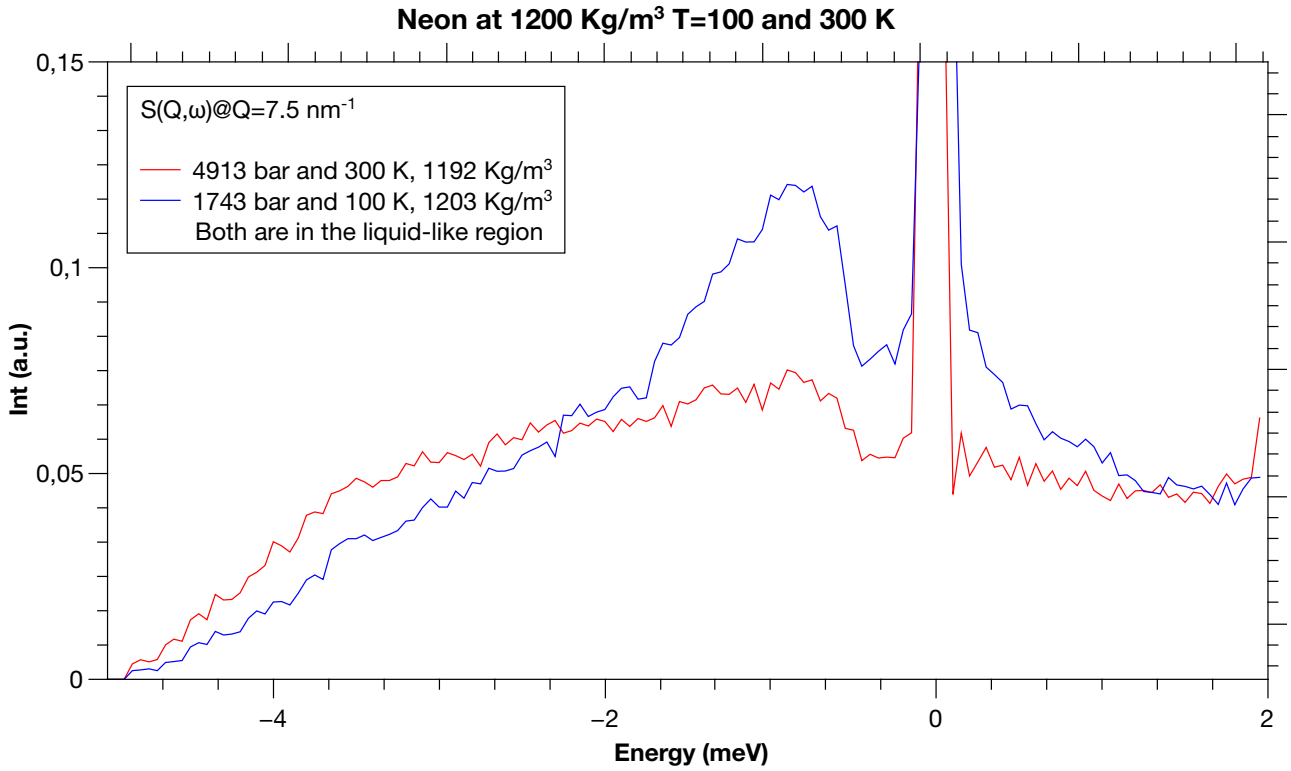


***Figure 3. Measured spectra at similar density (1200 Kg/m$^3$) at 100 and 300 K at Q=7.5 nm$^{-1}$***

changes depend on the thermodynamic state of the sample and are reversible at both temperatures. The peak in the high density spectrum appears only on the anti Stokes side so the absence of the Stokes counterpart could be either an instrumental artifact or indicate a true excitation in a out-of equilibrium sample. The observed peak is located at about the same energy at similar density at 100 and 300 K but it is much more prominent at 100 K as shown in the figure 3.

Looking more carefully at the isochoric data shown in figure 3 (1200 Kg/m$^3$) and figure 4 (785 Kg/m$^3$) we can see that the changes are correlated to the location of the thermodynamic point in the liquid-like or gas-like regions. The spectra shown in figure 3 have a very similar quasi elastic profile

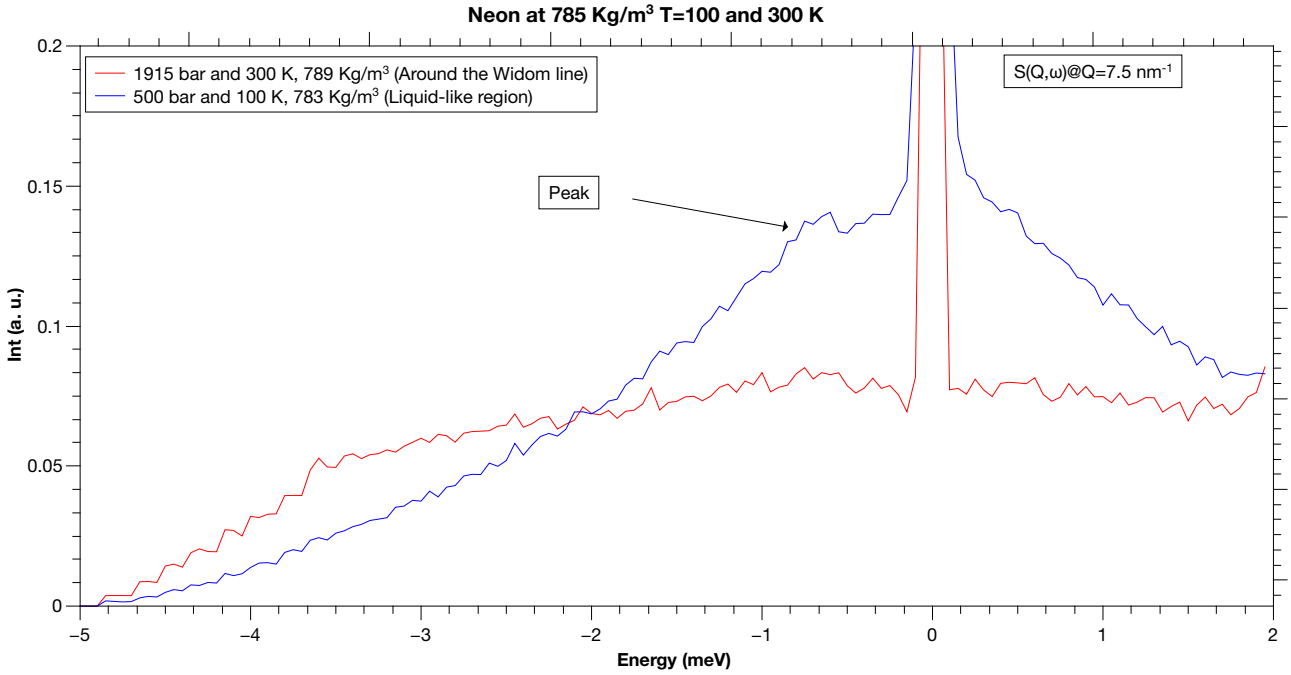


***Figure 4. Measured spectra at similar density (785 Kg/m$^3$) at 100 and 300 K at Q=7.5 nm$^{-1}$***

apart from the more prominent peak at low temperature and they are both in the liquid-like region with respect to the Widom line. On the contrary the spectra shown in figure 4 have a very different quasi elastic spectrum as the room temperature one is at about the “Widom” line, while the one at 100 K is inside the liquid-like region.

Several possible hypothesis have been tested to classify this signal as a spurious. But, as a matter of fact the position of the new excitation in the liquid like region would correspond to elastic scattering coming from an object at about 20 cm from the sample center in the detector direction. This is beyond any geometry limit of the sample environment. The solar collimator installed on the instrument reduces the effective optic volume to a cylinder of 10 cm in diameter. Multiple scattering events delay the signal thus in case of multiple scattering the signal should appear on the right hand side of the elastic peak.

This puzzling result call for further investigation and a repetition of the experiment is mandatory.


1) *F.A.Gorelli, et al., Phys. Rev. Lett. 97, 245702 (2006).*
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3) *G. Simeoni, et al., Nature Phys. 6 (7), 503 (2010).*
4) *F.A. Gorelli, et al., Sci. Rep. 3, 1203; DOI:10.1038/srep01203 (2013).*
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7) *Brazhkin et al PRL 111, 145901 (2013)*