	Experiment title: The effect of carbon doping on the vibrational properties of amorphous and crystalline GeTe: a microscopic insight on its ultralow thermal conductiviy	**Experiment number**: HC-3371
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Report:

Phase-Change Materials (PCM) exhibit a unique combination of properties from a reversible and ultra-fast amorphous/crystal transition to drastic differences in electronic properties, which put them at the forefront of research for optical and electronic storage. Among emerging non-volatile resistive memories, Phase Change Random Access Memories are at this day the most promising alternative technology in order to replace the current "flash memory". This technology exploits the unique properties of chalcogenide based materials, with an electrical resistance changing by several orders of magnitude upon crystallization and a very low thermal conductivity in both phases, especially important as it allows decreasing the electric power needed for inducing the crystal-to-amorphous transition by Joule heating, and makes further electronic devices miniaturization accessible.

In this context, much attention has been given to binary GeTe, where carbon doping has been found to greatly improve the high temperature retention properties, specifically increasing the crystallization temperature. Surprisingly, doping GeTe with carbon dramatically changes its thermal properties: measurements performed on C-GeTe thin films show that while doping only slightly decreases the thermal conductivity in the amorphous phase, it dramatically reduces the one of the crystalline phase, of a factor larger than 10, making it very similar to the value in the corresponding amorphous phase. Such reduction can only be due to a dramatic change in phonon transport, as the crystalline electronic conductivity is reduced by only 30% for C doping in the 10-15% range (our work).

In previous experiments (HC-2822, HC-3415) we have investigated the vibrational properties of polycrystalline GeTe and C-GeTe with 10% and 15% atomic content of carbon, called here GeTeCy and GeTeCz, by means of inelastic x ray scattering at ID28. Such measurements showed no difference in the longitudinal acoustic dispersion of the three samples, down to an exchanged momentum q=4 nm-1. Lower than that, we could no measure, because of a huge elastic line, whose intensity increases with C-content, hiding the weak inelastic features. Such limitation corresponds to an accessible energy range limited to energies higher than 10 meV.

In this experiment we measured the vibrational density of states using the Inelastic X ray Scattering with Nuclear Resonance Energy analysis technique (IXS-NRA).

The samples were the same used in previous experiments: 1-micron thick polycrystalline layers deposited on a silicon wafer, and capped with a tiny (10nm) amorphous SiN layer for reducing oxidation.
Measurements were done in a grazing incidence geometry, at the energy of the Fe^{57} nuclear resonance. An energy resolution of ~0.8 meV was obtained.
The geometry was optimized not to have any signal from the substrate. In one case we had it and could correct the signal knowing the silicon density of states.

Polycrystalline GeTe was measured at 50 K, temperature low enough for excluding any multi-phonon contribution, 200 K (approximately the Debye temperature) and 300 K. GeTeCz was measured at 50 and 300 K, while GeTeCy, for lack of time, only at 50 K.
Data analysis is still in progress as for the temperature effect, the multi-phonon subtraction being a critical step in the data reduction.
Concerning low temperature data, we do find a difference between the three samples, which is more marked at low energy, E < 7 meV, the energy range that we could not access by IXS. As can be seen in the figure, there is a general lowering and broadening of the density of states features with carbon content. More interestingly, the density of states increases with respect to pure GeTe at low energy, giving raise to something like a Boson Peak. We are presently working for connecting these results to the reported decrease of the thermal conductivity.

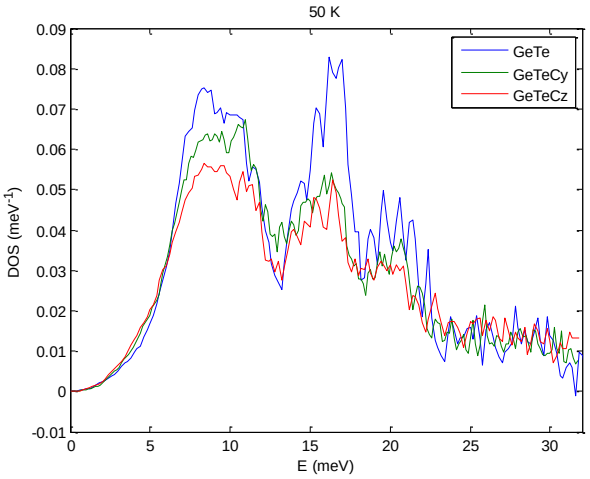

Figure 1: Density of states at 50 K of polycrystalline GeTe and C-doped GeTe after raw data reduction