	Experiment title: Electron-phonon coupling in CaC_6: a low-temperature study of its phonons	**Experiment number**: HS-3998
Beamline: ID28	**Date of experiment**: from: 3rd July 2009 to: 7th July 2009	**Date of report**: 31st August 2009
Shifts: 12	**Local contact(s)**: Mr Andrew Walters, Dr Alexei Bossak	*Received at ESRF:*

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Report:

In 2005 the Graphite Intercalation Compounds (GICs) CaC_6 and YbC_6 were discovered to have superconducting transition temperatures (T_C) over an order of magnitude higher than anticipated (11.5 K and 6.5 K respectively) [1,2]. Early studies suggested that the superconducting mechanism could be highly exotic [3], but more recently the theoretical consensus has been that the superconductivity is mediated by phonons [4,5].

However, several experimental measurements are inconsistent with these Density Functional Theory (DFT) descriptions of the superconductivity in GICs. One of the most notable of these is the measurement of a large Ca isotope effect ($\alpha_{Ca} \approx 0.5$) in CaC_6 [6], at variance with the predicted values of $\alpha \approx 0.25$ for both α_{Ca} and α_C. Within the BCS model, the experimental value of $\alpha_{Ca} \approx 0.5$ means that all of the electron-phonon coupling occurs via Ca phonons, and no C phonons are involved at all. Of course, as there is no published measurement of α_C in CaC_6 this statement is difficult to justify, as the carbon isotope effect α_C could also be substantial.

Another experimental measurement at odds with the theory is the observation in CaC_6 of strong electron-phonon coupling to high energy (≈ 160 meV) graphitic phonons, seen using Angle-Resolved Photo-Emission Spectroscopy (ARPES) [7]. The electron-phonon coupling observed here is so strong that it is more than capable of explaining the observed T_C itself. These conclusions from the ARPES measurements are quite controversial, and there is a long-standing debate about how electron-phonon coupling can be reliably extracted from ARPES data.

These ARPES measurements provided the core motivation for this Inelastic X-ray Scattering (IXS) experiment: namely, to study the temperature dependence of the high-energy graphitic phonons above and below T_C. In particular we wanted to study the phonons around the M-K line (as described within the graphite Brillouin zone), as this was the trajectory along which the observed electron-phonon coupling was maximised in the ARPES measurement. We planned to be sensitive to any sudden temperature dependent changes in linewidth, providing evidence for a change in the electron-phonon coupling below T_C, as well as any other change in the phonons which could be a crucial signature of the true nature of the superconductivity in CaC_6.

However, in reality this experiment was somewhat problematic, as the extreme air-sensitivity of the samples resulted in the first batch of samples being of insufficient purity. Consequently the first two days of allotted beamtime were taken up by Dr Alexei Bossak, whilst new CaC_6 samples were shipped from the sample growers (hence only 12 shifts being taken up so far). These new samples proved to be superior, not just to the first batch of samples for this experiment, but also to the sample used in our first ID28 experiment on CaC_6 [8]. Whereas the first experiment also observed additional intensity at higher than anticipated energies (see Figure 1(a) in Ref. [8]), equivalent scans in this beamtime revealed no such intensity. It is likely that this intensity arose directly from a small graphite impurity in that sample.

We measured once again at all six of the momenta presented in Figure 1(a) in Ref. [8], thereby providing the detailed dispersion of the intense phonon in this energy range from M to K. The width of this feature does not appear to change as a function of Q, so there is no clear evidence for electron-phonon coupling which is heavily localised in Q. This would be the nature of the type of electron-phonon coupling suggested by the ARPES measurement.

As well as the sample-related issues, we also had difficulty with the cryogenic equipment that we used on the experiment. These problems meant that only a few scans were done at low temperatures, and all of these scans have fewer statistics than the equivalent high temperature scans. In Figures 1 and 2 we plot representative data measured at identical Q at both ambient temperature and at low temperatures. In both cases it is clear that there is no measurable temperature dependence.

In summary - despite the considerable technical difficulties - we were able to provide convincing evidence in this experiment that there is no significant temperature dependence in the high-energy graphitic phonons in CaC_6. It may be that the electron-phonon coupling manifests itself in the phonons solely within a Kohn anomaly which exists over all the entire temperature range that we have studied thus far. Subsequent IXS studies will attempt to locate such a Kohn anomaly, thereby providing complementary evidence to the ARPES measurements [7].

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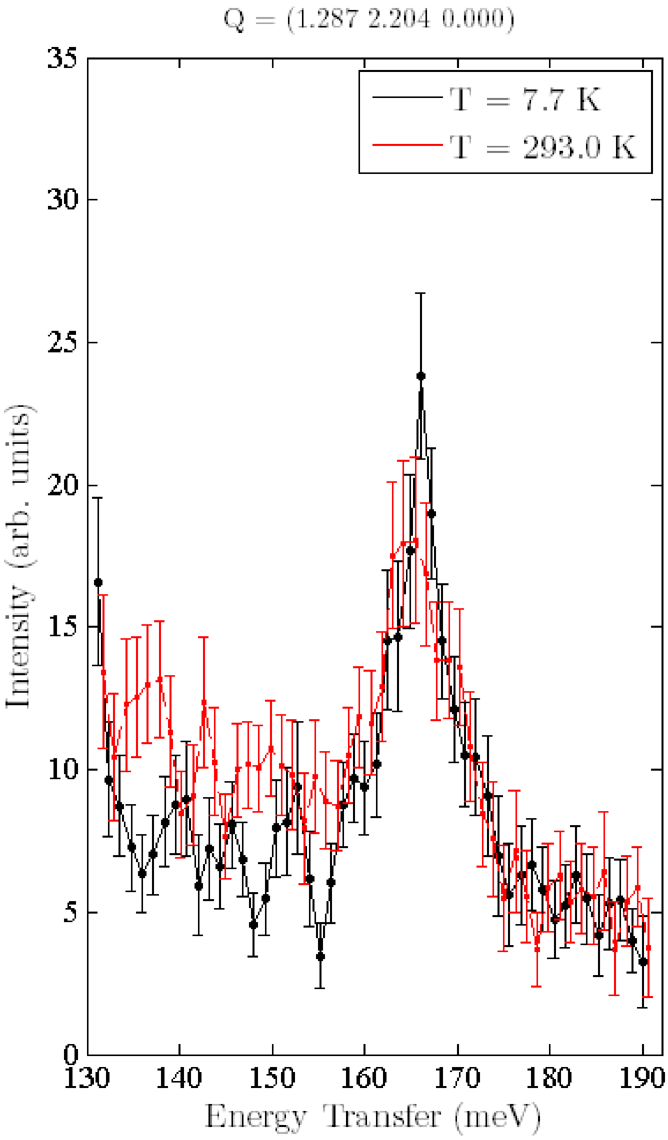

Figure 1 Representative data measured below and above the superconducting transition temperature in CaC_6. As observed in all scans, there is no observable temperature dependence.

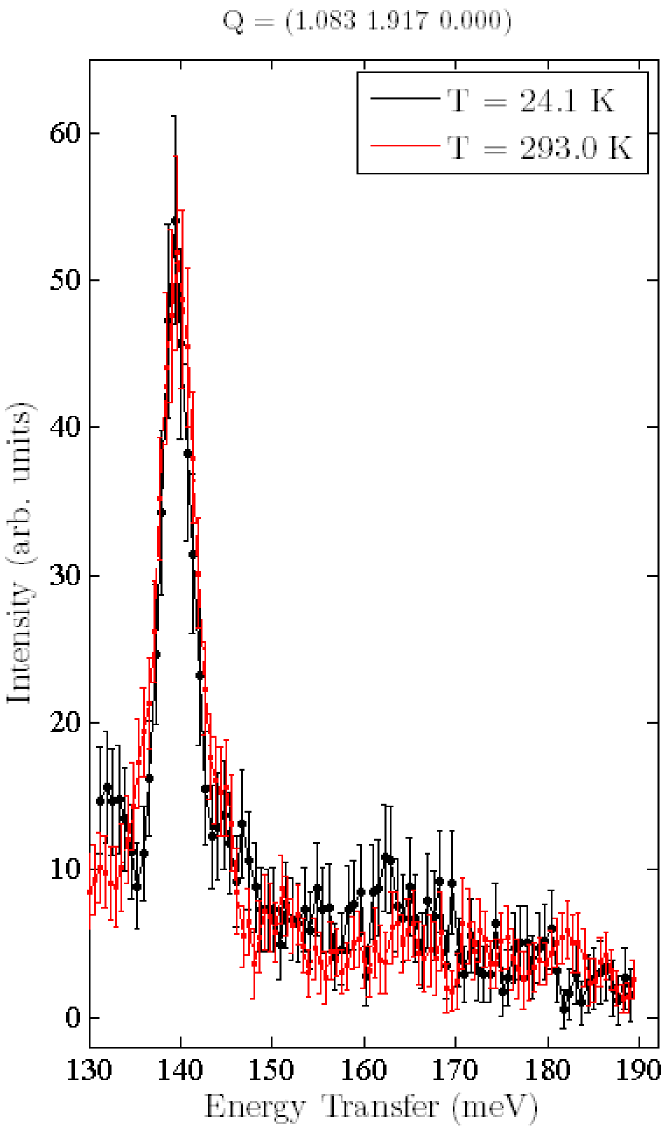

Figure 2 Representative data measured just above and well above T_C. Once again, as in Figure 1, there is no discernable temperature dependence.