ESRF	**Experiment title:** Effects of isotope disorder on phonons in diamond	**Experiment number:** HS-1068
Beamline: ID28	**Date of experiment:** from: 02-February-2000 to: 10-February-2000	**Date of report:** 2-October-2000
Shifts: 24	**Local contact(s):** Dr. Matteo D'Astuto, Dr. Michael Krisch	*Received at ESRF:*

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Report:
Inelastic X-ray scattering (IXS) is nowadays a viable alternative for studying dispersion and self-energy effects of elementary excitations in solids. This is due to the recent advent of dedicated beamlines at high-brilliance third-generation synchrotrons. However, while a substantial amount of work has been devoted to liquids and glasses [1], IXS has been used much less to investigate the lattice-dynamical properties of *crystals* [2]. In the work reported here, we have exploited the unique advantage of IXS to study very small crystals and performed inelastic scattering experiments which are presently impossible with neutrons. The data were taken using excitation energies of 17.79 keV and 15.82 keV with an instrumental resolution of 2.5 meV and 5.5 meV (FWHM), respectively. The high energy resolution and crystal-momentum selectivity of beamline ID28 offers the possibility to determine *self-energies* $\Sigma(\omega) = \Delta(\omega) + i\Gamma(\omega)$ *of individual phonons* across the whole Brillouin zone.

We have measured the longitudinal acoustic and optic phonon branches along the Γ–L direction in natural diamond and an isotopically mixed $^{12}C_{0.53}{}^{13}C_{0.47}$ crystal. In Fig. 1 we display the IXS spectra corresponding to the L point of the Brillouin zone. The LA peak was used as an internal energy reference for each sample. Furthermore, errors due to possible long-time drifts of the energy scale were minimized by using a "double goniometer" arrangement, i. e., we simultaneously mounted both diamonds on the same sample holder and aligned them along the (1 1 1) direction. The upper spectrum ($^{12}C_{0.53}{}^{13}C_{0.47}$) in Fig. 1 is as measured; the lower one (natural diamond) has been scaled by a factor of 0.9790 in order to make the two LA peaks coincide at the energy of 127.9 meV, measured for the isotopically mixed sample.

This factor is in very good agreement with the frequency ratio $\sqrt{12.011/12.475} = 0.9812$, expected from the average isotope masses of the two crystals. Self-energy effects on the LA phonon can therefore be neglected within the experimental accuracy. Lorentzian lineshape fits to the spectra in Fig. 1 (solid lines) yield energies of 152.6 meV (153.5 meV) and linewidths (FWHM) of 2.6 meV (3.5 meV) for the LO_L phonon in natural (isotopically mixed) diamond. Taking into account the intrinsic FWHM (Lorentzian) of 2.5 meV of the 17.79 keV excitation at ID28 (operated with the Si (9 9 9) Bragg reflection) leads to an isotope-disorder induced broadening of $\Gamma(LO_L) = 0.9$ meV and a frequency shift of $\Delta(LO_L) = 0.9$ meV for the isotopically mixed $^{12}C_{0.53}{}^{13}C_{0.47}$ sample compared to natural diamond (almost isotopically pure). These data are in good agreement with theoretical predictions of about 0.6 meV for both quantities [3].

During this experiment we performed also a preliminary study of the phonon dispersion of wurtzite GaN, in order to use the alloted beamtime during the couple of days required to set up and adjust the double goniometer. This study demonstrated the feasibility of the measurement of the phonon dispersion of such material, which is currently of great interest for optoelectronic applications at blue and near-ultraviolet wavelengths. The lattice-dynamical properties of GaN have been studied intensively by theoretical methods and also by Raman scattering, but unfortunately no information is available for phonons of wavevector $\vec{q} \neq \vec{0}$. The main reason for the lack of phonon dispersion information in GaN is that single crystals large enough for inelastic neutron scattering do not exist. However, this limitation can now be overcome with IXS. We have measured a few $\vec{q}$-points mainly along the Γ–A direction, obtaining reliable information about the "silent" mode B_1. The knowledge of the B_1 mode frequencies is rather important, since their energy at Γ determines whether mode anticrossings with other dispersion branches may occur inside the Brillouin zone. Note that the observation of this "silent" mode is only possible by IXS, since it is neither Raman nor infrared active. This mode allows one to check the different lattice-dynamical models and, in the case of CdS, insufficient information on these frequencies has been the origin of strong deviations of phenomenological models from inelastic neutron scattering data [4].

We have performed *ab-initio* calculations of the phonon dispersion along several high-symmetry directions in order to explain the experimental results. Figure 2 displays the phonon dispersion along the $\langle 0\,0\,1 \rangle$ direction. The filled circles are measured phonon energies from IXS, and the open diamonds at $\vec{q} = \vec{0}$ are the Raman data from Ref. [5]. The solid lines are the results of *ab-initio* lattice-dynamical calculation; this theoretical results have been scaled by a factor of 0.97 to obtain optimum agreement with the experiment.

As a result of the measurements performed in the beamline ID28 at ESRF, we have submitted the following contributions:

Phonon dispersion curves in wurtzite-structure GaN determined by inelastic X-ray scattering, T. Ruf *et al.*, submitted to Phys. Rev. Lett. (June 30th, 2000).

Inelastic X-ray scattering study of phonons in GaN and diamond, T. Ruf *et al*, *Proceedings of the 17th International Conference on Raman Spectroscopy*, Ed. by S. L. Zhang and B. F. Zhu (Wiley, New York, 2000), p. 428.

The phonon dispersion of wurtzite-structure GaN studied by inelastic X-ray scattering, T. Ruf *et al*, *Proceedings of the 25th International Conference on the Physics of Semiconductors*, Osaka, 2000, accepted for publication.

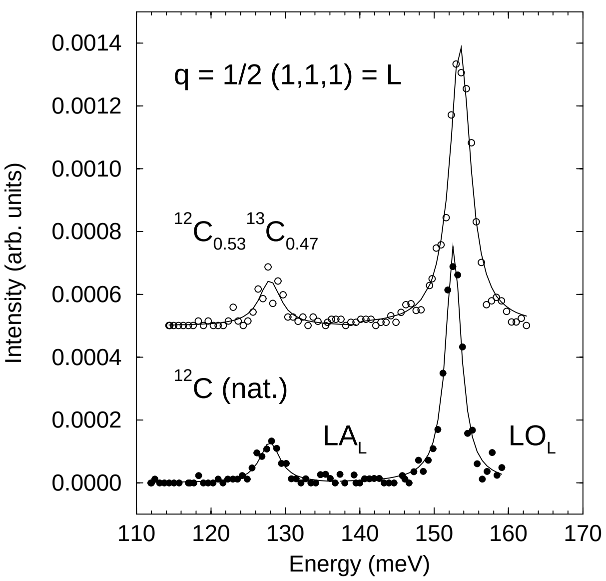

Figure 1

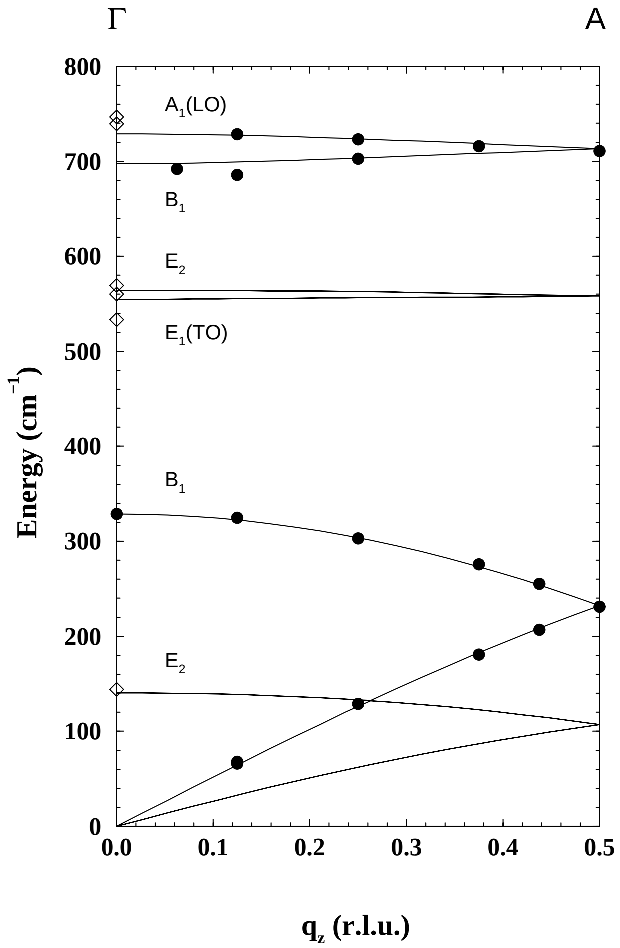

Figure 2

References

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[5] J. M. Zhang *et al.*, Phys. Rev. B **56**, 14399 (1997).