Materials Science Forum Vols 433-436 (2003) pp 257-260 *Online: 2003-09-15*

doi:10.4028/www.scientific.net/MSF.433-436.257

Lattice Dynamics of 4H-SiC by Inelastic X-ray Scattering

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Keywords: 4H Silicon Carbide, phonon dispersion relations, inelastic x-ray scattering

Abstract. We have measured the phonon dispersion relations in 4H-SiC by inelastic x-ray scattering (IXS) using monochromatized synchrotron radiation. The **q**-space directions Γ–K–M, Γ–M, and Γ–A were mapped out. Lattice dynamical calculations that allowed the prediction of phonon eigenvectors, as well as their symmetries, also helped in choosing the best scattering geometries. The IXS phonon data are compared with those previously obtained from low temperature photoluminescence measurements and from laser Raman spectroscopy.

1 Introduction

Silicon carbide is a wide band gap material which crystallizes in a large number of polytypes [1], exhibiting properties, such as high thermal conductivity, that make it a good candidate for optoelectronic applications. Despite the large number of articles dedicated to the vibrational properties of SiC [2, 3, 4], little experimental information is available covering the whole Brillouin zone (BZ). Such information has been recently obtained for 6H-SiC [5] by inelastic neutron scattering (INS), and for 3C-SiC by inelastic x-ray scattering (IXS) [6], a relatively new technique which employs highly monochromatized synchrotron radiation [7].

IXS allows the investigation of the phonon dispersion relations of crystals of submilimeter size, as demonstrated for the wide band gap materials diamond [8], AlN [9], and GaN [10], where large size samples are not available.

In [6] we reported IXS data for 3C-SiC along several directions of the BZ and along the Γ–A direction for 4H-SiC. We present here more detailed results for the phonon dispersion of 4H-SiC along the Γ–K–M, Γ–M and Γ–A directions of the BZ obtained by IXS. Data extracted from photoluminescence for the M-point and from Raman spectroscopy for the Γ-point are analyzed and compared with our IXS results and with Born-von-Kármán lattice dynamical calculations [11]. These calculations were used to elucidate the best scattering geometries for our experiments and to characterize the symmetry of the different modes at the Γ and M points.

2 Experimental details and calculation procedure

IXS measurements were performed for several samples with orientation $(1\bar{2}10)$ and (0001), and typical sizes of $10 \times 5 \times 0.2$ mm, cut from the same single crystal. In order to decrease dead time due to sample mounting and alignment we kept the sample at room temperature.

The experiments were carried out at the European Synchrotron Radiation Facilities (ESRF) in Grenoble (ID28). Details of the resolution and calibration procedure can be found in [6].

In order to calculate the dispersion relations of 4H-SiC by the Born-von-Kármán method, and the corresponding eigenvectors, we used force constants obtained by interpolation and extrapolation of those calculated *ab initio* for 3C (zinc blende) and 2H (wurtzite) SiC using linear response and the local density approximation of density functional perturbation theory. The details of the *ab initio* calculations for 3C- and 2H-SiC are given in [6, 12] whereas those involved in the determination of the force constants of 4H-SiC will be given in [11]. We obtain a good agreement with a phonon dispersion calculated in [13] using the bond-charge model.

3 Results and Discussion

Figure 1 shows the theoretical phonon dispersion curves of 4H-SiC, together with experimental points obtained by IXS (•), along the high symmetry directions Γ–K–M (q_x), Γ–M (q_y), measured on the $(1\bar{2}10)$ surface, and Γ–A (q_z), measured on the (0001) surface; for a sketch of the BZ of the hexagonal SiC polytypes see [5, 14]. The latter corresponds to a double backfolding of the Γ–L direction of the 3C-SiC. The concept of folding along Γ–A has been used in the

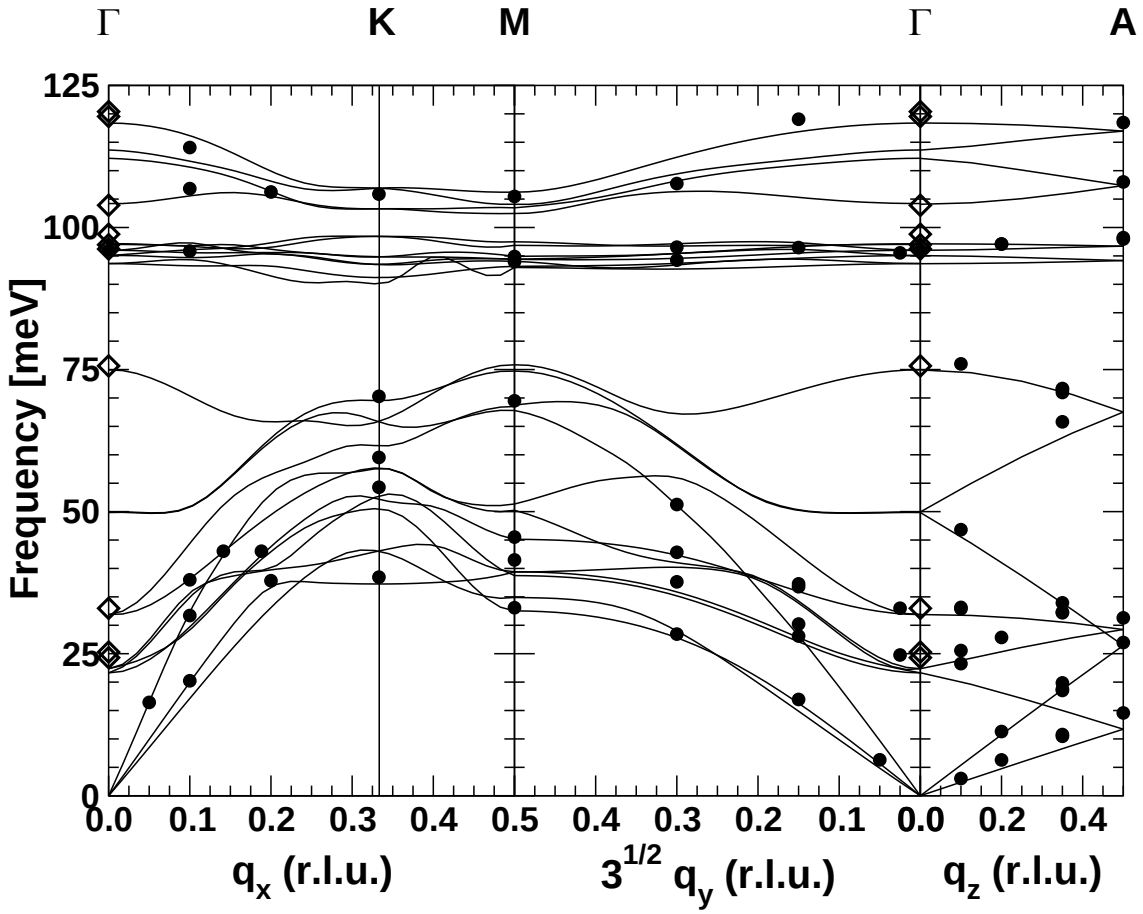

Fig. 1. Phonon dispersion relations of 4H-SiC. IXS data (•) are displayed together with calculated phonon dispersion (solid lines) and Raman data (◇) from Ref. [13]. The **q**-axes are given in reciprocal lattice units (r.l.u.). For a comparison of the M-point frequencies with photoluminescence data see Table 1.

past to predict phonon frequencies [2, 3] and eigenvectors [4] of 3C-SiC along Γ–L which agree

with IXS data [6]. The Raman data collected in [13] are also displayed in Fig. 1 by open diamonds. Notice that along the Γ–A direction the calculated frequencies are slightly lower than the measured ones. This underestimate is also reflected in Table 1, where we list the theoretical and IXS data together with those obtained from the indirect gap photoluminescence (LTPL) at 2 K [15, 14]. The LTPL spectra exhibit phonon peaks corresponding to indirect transitions between the maximum of the valence band at Γ, and the minima of the conduction band, at the M-point, aided by phonon emission.

We have assigned in Table 1 the symmetry of the different modes at the M-point on the basis of the calculated eigenvectors. The M_1 and M_3 symmetry phonons are emitted in LTPL processes polarized along the c-axis whereas M_2 and M_4 phonons are emitted for the perpendicular polarization [14]. The agreement between symmetry assignments and the observed polarizations is excellent.

Symmetry	[a]IXS	[b][15]	[b][14]		theory	Symmetry	[a]IXS	[b][15]	[b][14]		theory
M_2	33.1(2)	33.0	33.2	$\perp$	32.5	M_2	94.0(3)	–	94.5	$\perp$	93.0
M_1		36.5	36.6	$\parallel$	34.9	M_3	94.9(3)	96.1	94.7	$\parallel$	93.2
M_3		40.6	40.6	$\parallel$	38.7	M_4		97.3	96.0	$\perp$	94.0
M_4		41.0	41.1	$\perp$	39.4	M_3		97.8	96.5	$\parallel$	94.4
M_3	41.5(2)	41.7	41.9	$\parallel$	39.4	M_2		99.1	–	$\perp$	94.4
M_2	45.5(2)	46.1	46.3	$\perp$	45.1	M_1		101.6	–	$\parallel$	94.9
M_4		50.8	50.9	$\perp$	50.2	M_1		105.6	103.5	$\parallel$	96.9
M_1		52.7	52.7	$\parallel$	51.3	M_4		100.3	98.9	$\perp$	97.5
M_4		67.8	68.1	$\perp$	67.8	M_4		106.3	103.9	$\perp$	102.5
M_1	69.5(2)	68.7	68.8	$\parallel$	68.5	M_1	105.5(5)	108.2	106.3	$\parallel$	103.5
M_4		76.6	76.3	$\perp$	74.7	M_1		108.7	109.5	$\parallel$	104.1
M_1		78.5	78.0	$\parallel$	75.8	M_4		–	106.5	$\perp$	106.2

Note. [a]Data at 300 K.
Note. [b]LTPL data at 2 K.

Table 1. Frequencies of the phonons of 4H-SiC in meV (1 meV $\approx$ 8.07 cm^{-1}) at the M-point of the Brillouin zone determined by inelastic X-ray scattering (IXS), compared with experimental data from low temperature photoluminescence (LTPL) and our lattice dynamical calculations. After the LTPL frequencies we have listed the corresponding polarizations ($\parallel$ or $\perp$ with respect to the c-axis) of the emitted light as given in [14].

The frequencies calculated for the folded LO phonons at Γ are 118.4, 113.6, 112.2 and 104.1 meV. The highest and lowest of these frequencies have been determined by Raman spectroscopy to be 119.5 and 103.9 meV. Note that the calculated discontinuity in the highest LO frequency near Γ, depending on whether the phonons are polarized along c or perpendicular to c, is 0.03 meV, too small to be seen in Fig. 1. The same can be said for the TO phonon discontinuities. The folded TO phonon frequencies at Γ are 97.1 (Γ_5 symmetry), 96.0 (Γ_6), 95.0 (Γ_6), and 93.6 meV (Γ_5) as compared with the Raman results: 98.8, 97.2, and 96.2 meV [3]. Raman data in [16] show a splitting of 2.5 meV between the uppermost, TO(Γ_5) mode and a Γ_6 mode which is compatible with our calculated TO(Γ_5) and TO(Γ_6) frequencies. Splittings of $\approx$ 1 meV, also compatible with our calculations, have been recently observed for the TO modes on nanosized powders of SiC with a nonstoichiometric excess of carbon [17].

Inelastic neutron scattering measurements on a large single crystal of 4H-SiC are now being performed at the Laue Langevin Institute (Grenoble) to determine with high resolution the acoustic and longitudinal optic phonon branches along the Γ–M and Γ–K–M directions in the BZ [18].

4 Acknowledgements

We thank M. Krisch and T. Ruf for fruitful discussions. The use of excellent facilities and instruments at the ESRF is gratefully acknowledged.

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Silicon Carbide and Related Materials - 2002
10.4028/www.scientific.net/MSF.433-436

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