**Clebsch-Gordan coefficients for scattering tensors in Bi2Se3**

**A. G. J. Machatine1, H. W. Kunert1, P. Niyongabo1, M. Govender1, 2, and B. W. Mwakikunga2**

1Department of Physics, University of Pretoria, Pretoria 0002, South Africa

2DST/CSIR National Centre for Nano-Structured Materials, P. O. Box 395, Pretoria 0001, South Africa

E-mail: Augusto.Machatine@up.ac.za

**Abstract.** The analysis of Raman and Brillouin scattering spectra requires the knowledge of scattering tensors. Based on Birman's method, we compute the Clebsch-Gordan coefficients for first-order Raman and Brillouin scattering for Bi2Se3 with  space group symmetry. The linear combination of Clebsch-Gordan coefficients gives the matrix elements of scattering tensors. Our calculations are useful for interpretation of spectra in Raman and Brillouin scattering measurements.

**1. Introduction**

The topological insulator Bi2Se3 crystallizes into the trigonal space group with five atoms per unit cell. It has a layered structure and an energy gap of 0.3 eV. Topological insulators are new quantum states of matter which are insulating in the bulk, but possess “non-trivial” edge conducting surface states with time-reversal symmetry protected against backscattering [1, 2]. Raman studies were conducted on Bi2Se3 nanopallets and showed broadening of peaks [2]. This result and the electron-phonon interactions in the Raman analysis are not well understood. Ab initio calculations of thin films have been performed, and quantum confinement effects were discussed in an attempt to understand the results [3]. It is thus imperative to understand the phonon scattering processes.

Raman and Brillouin spectroscopy are techniques used to understand the nature of the phonons in a sample. In these scattering processes, photons excite the crystal under investigation, causing the quasi-particles such as phonons, polarons, magnons and polaritons to be either created or annihilated, and thereafter reemit photons with a frequency shift. It is well known that the states of particles and quasi-particles in the crystal are classified according to the irreducible representation (irreps) of the space group of a crystal [4].

The Raman scattering involving one-phonon processes are related to the zone centre optical phonons. One-phonon Brillouin scattering arises from the interaction of light with acoustic phonons close the zone centre. Birman et al. [5] and Berenson [6] have shown that the first-order Raman scattering are precisely linear combinations of certain Clebsch-Gordan coefficients (CGC’s). They also showed that near the highest symmetry point , the Brillouin tensor is proportional to the strain tensor [7]. In order to calculate the CGC’s, we need the generators of matrix representations of the groups. These generators are contained in the Cracknell, Davies, Miller, Love (CDML) tables, along with the Kronecker product (KP) of symmetrized, anti-symmetrized squares and cubes for the 230 space group irreps [8]. In this paper, we focus on the calculation of the CGC’s and construction of first-order Raman and Brillouin scattering tensors.

**2. Scattering tensors and Clebsch-Gordan coefficients**

In this section we present a summary of CGC’s theory that will be used for determination of Raman and Brillouin scattering tensors. The complete theory of CGC’s for a crystal space group has been outlined by several authors [7]. Particularly, Kunert et al. [4], Birman [5] and Berenson [6] have calculated the CGC’s and scattering tensors for ,  and  point groups, respectively. In this work, we extend our calculation to  space symmetry group.

If the incident light has unit polarization vector  with component and the unit polarization vector for the scattered light  with component  , the intensity of the scattered light is then given as

, (1)

where  is the scattering tensor and  a constant. The scattering tensor operator involved in different processes are expanded in terms of Taylor series with respect to different coordinates. In the next subsection we give the expansion for the Raman scattering tensor and the Brillouin scattering tensor operators.

**2.1 Raman scattering tensors**

For the Raman scattering process, the operator  is expanded in terms of the normal coordinate  to give

. (2)

The coefficients of these series correspond to the Raman scattering tensors for various orders. The linear term is the first-order (one-phonon) Raman scattering tensor and the bilinear term is the second-order Raman scattering tensor. Birman et al. showed that coefficients of the expansion are related to the CGC’s. The first-order tensor is written as  where  is a constant, the equilibrium ion coordinate in the unit cell, j () is the allowed Raman irrep with degeneracy  and  is the unitary matrix element (CGC’s) [6].

The Raman active modes are contained in the symmetrized squares of the vector representation . For  space symmetry group, the vector representation is given by

, (3)

and the symmetrized square is given by

. (4)

The ordinary product  is not contained in the symmetrized square and therefore does not appear in the Raman scattering tensor. Similar features were observed in ZnO with symmetry  [4] when the  is a direct sum of irreps. The calculated CGC’s are tabulated in Table 1.

**Table 1.** Clebsch-Gordan coefficients for Raman Scattering tensors for.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  |  |  |  | |
| xx | 0 |  | 0 | 0 |
| xy |  |  | 0 | 1 |
| yx |  |  | 1 |  |
| yy | 0 |  | 0 | 0 |
| zz | 1 |  | 0 | 0 |

The first-order Raman scattering tensors are matrices constructed from Table 1 in the matrix form , and are listed in Table 2.

**Table 2.** First-order Raman scattering tensors for.

|  |  |  |
| --- | --- | --- |
| ; | ; | . |

In the next subsection, we construct the first-order Brillouin scattering.

**2.2 Brillouin scattering**

For Brillouin scattering, the operators are expanded in terms of acoustic phonon polarization  with the polarization  and phonon  resulting in

, (3)

where  is the first order Brillouin scattering tensor. The Raman tensor  is the first-order Brillouin tensor, and is a linear combination of the first-order Raman scattering tensor*.* The matrix and  are the unitary matrices for the symmetrised square and ordinary KP, respectively [9].

Using the obtained first-order Raman scattering tensors, we construct the first-order Brillouin scattering tensor as linear combinations of CGC’s from Table 1. As an example, an acoustic phonon incident in the *x*-direction is scattered in in the *y*-direction resulting in a Brillouin tensor. The Brillouin tensors  and so

. (4)

The non-zero Brillouin scattering tensors are listed in Table 3 where  and  are arbitrary constants subject to measurements.

**Table 3.** First-order Brillouin Scattering Tensors for .

|  |
| --- |
| ; ; |
| . |

**5. Discussion**

In summary, we have calculated the CGC’s from which the matrix elements of the first-order Raman and Brillouin scattering are constructed. In the symmetrised square of the V-rep the product representation  is excluded. Gachter [10] attributed this interaction in  symmetry to resonance Raman which is different from first-order Raman. The results given here are applicable to the class of materials belonging to space group. Koster et al. [11] and Loudon [12] have tabulated CGC’s and scattering tensors for 32 point groups using different bases, and so it is pertinent to compare and extend the use of scattering tensors. We used CDML bases which contain irreps for the 230 space groups and their subgroups for high symmetry points and lines in the respective Brillouin zones. Our calculations of CGC’s based on Birman’s method, are useful for the construction of higher order Raman and Brillouin scattering tensors for morphic effects. Scattering tensors are used in polarization and experimental intensity studies due to phonon scattering. Also invariants needed in phase transition studies as well as effective Hamiltonians [13] can be easily derived from our results.

**6. Conclusion**

We have calculated the Clebsch-Gordan coefficients for the first-order Raman and first-order Brillouin scattering tensors. We show that the first-order Brillouin scattering tensors are obtained from the first-order Raman scattering tensors in. Our results will be used in the interpretation of polarized of Raman scattering and Brillouin spectroscopy. From these experimental assignments, the intensity of phonons and elastic constants of materials can be determined.

**References**

1. H. Zhang, C. Liu, X. Qi. X. X. Dai, Z. Fang, and S. Zhang, Nature **5** (2009) 438
2. J. Zhang, Z. Peng, A. Soni, Y.Zhao, Y. Hiong, B. Peng and J. Wang, Nano Lett. **11** (2011) 2407
3. W. Cheng, Phys. Rev. B 83 (2011) 094301-1
4. M. Cardona, in Light Scattering in Solids edited by M. Cardona, G. Gunthevodt, (Springer-Verlag) ISBN 038711913 (1983).
5. H.W. Kunert et al. Phys. Status Solidi **B** (2010) 1.
6. J.L. Birman and R. Berenson, Phys. Rev. **B**  **9** (1974) 4512
7. R. Berenson, J. Phys. Chem. Solids **42** (1981) 391.
8. A.P. Cracknell, B.L. Davies, S.C. Miller, W.F. Love, Kronecker Product Tables, (Ifl/Plenum, New York, 1979) Vol. 1-4.
9. J.L. Birman, Theory of Crystal Space Groups and Lattice Dynamics (Springer-Verlag, Berlin) (1984).
10. B. Gachter, J. Mol. Spect. **63** (1976) 1
11. G.F. Koster, Phys. Rev. **109** (1958) 227
12. R. Loudon, Adv. Phys. **13** (1964) 13
13. J.L. Birman and T. Lee, **14** (1976) 318