

# Clebsch–Gordan coefficients for scattering tensors in ZnO and other wurtzite semiconductors

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Selection rules for inter- and intra-valley scattering processes between the minima of the lowest conduction band  $\Gamma$ ,  $L$ ,  $M$ , and  $A$  are investigated in wurtzite ZnO. The knowledge of the selection rules is required for the determination of the Clebsch–Gordan coefficients (CGCs). These coefficients diagonalize the

Kronecker products (KPs) of relevant irreducible representations of quasi particles participating in scattering processes. The elements of the scattering tensors are the linear combinations of the CGCs. Within this work, the CGCs for ZnO belonging to the space group  $O_{6v}^4$  are determined.

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**1 Introduction** The study of scattering processes involving phonons provides important information about various physical properties of semiconductors. These processes are usually described by the different scattering tensors which are determined by appropriate experimental techniques. Applying electrical (transport) measurements, tensors like the second rank conductivity, capacitance, and resistivity tensor can be determined. The inelastic light scattering is described by a symmetric second rank tensor which can be derived with high accuracy using Raman spectroscopy. The mechanical properties of crystals depend on tensors like surface stiffness constants, determined by, e.g., Brillouin scattering, or the third rank piezoelectric tensor which involves electron–acoustic–phonon interactions. Particularly, in non-centro-symmetric crystals (zinc blende and wurtzite) the strain tensor and induced electric fields can be expressed as a third rank electromechanical tensor (see, e.g., Ref. [1]).

In this contribution we investigate scattering processes in the wide band gap semiconductor ZnO which is attracting much attention due to potential applications in, e.g., short-wavelength optoelectronic devices and surface acoustic wave modulators [2, 3]. In particular, possible inter- and intra-valley scattering processes at local extrema in the

electronic band structure of ZnO are studied by Clebsch–Gordan coefficients (CGCs). These scattering processes can occur at high symmetry points due to the interaction of carriers with phonons, impurities, and dislocations. The mechanisms have been studied in a wide variety of different semiconductors. Lax and Hopfield [4] and Birman et al. [5] have investigated the selection rules for direct and indirect radiative transitions by intra- and inter-valley scattering in Si and Ge. The Raman scattering tensors in cubic  $\text{Cu}_2\text{O}$  ( $O_h^4$ ) were analyzed by Birman [6]. The mathematical theory of the CGCs was derived by Berenson and Birman [7] who calculated CGCs for diamond ( $O_h^3$ ) and rocksalt ( $O_h^5$ ) [8]. Birman et al. [9] have shown that the matrix elements of the Luttinger–Bir–Picus  $\mathbf{k} \cdot \mathbf{p}$  effective Hamiltonian can be constructed as products of the CGCs times symmetrized tensorial field quantities. They applied the method to the  $\beta$ -wolfram structure ( $O_h^3$ ). Kunert and Suffczyński [10, 11] developed the theory of the CGCs related to the wave vector selection rules (WVSRs) in terms of block structures. They computed the CGCs for the  $O_h^3$  space group. Finally, Berenson [12] discussed the scattering tensors for crystals limited to the point groups  $T_d$  and  $C_{6v}$ . However, the coefficients for ZnO have not been studied up to now. In principal, the here presented results can also be applied to

**Table 1** Clebsch–Gordan coefficient matrix for  $L_1 \otimes M_1 = A_1 \oplus A_5 \oplus L_1 \oplus L_2$ ,  $a = 1/\sqrt{2}$ ,  $b = 1/\sqrt{3}$ ,  $w = -1/2 + i\sqrt{3}/2$ .

WVSRs	block	$A_1$	$A_5$		$L_1$	$L_2$
	$\sigma\sigma'\sigma''$					
$k_L + k_M = k_A$	111	$b$	$b$	$b$	0 0 0	0 0 0
$2k_L + 2k_M = k_A$	221	$b$	$-iwb$	$-iw^*b$	0 0 0	0 0 0
$3k_L + 3k_M = k_A$	331	$b$	$w^*b$	$wb$	0 0 0	0 0 0
$2k_L + 3k_M = k_L$	231	0	0	0	$ia$ 0 0	$-ia$ 0 0
$3k_L + 2k_M = k_L$	321	0	0	0	$-ia$ 0 0	$ia$ 0 0
$k_L + 3k_M = 2k_L$	132	0	0	0	0 $-ia$ 0	0 $ia$ 0
$3k_L + k_M = 2k_L$	312	0	0	0	0 $ia$ 0	0 $-ia$ 0
$k_L + 2k_M = 3k_L$	123	0	0	0	0 0 $ia$	0 0 $-ia$
$2k_L + k_M = 3k_L$	213	0	0	0	0 0 $-ia$	0 0 $ia$

$$U^{-1}(L_1 \otimes M_1)U = \begin{pmatrix} A_1 & 0 & 0 & 0 \\ 0 & A_5 & 0 & 0 \\ 0 & 0 & L_1 & 0 \\ 0 & 0 & 0 & L_2 \end{pmatrix}$$

other wide band gap semiconductors like GaN, CdS, 6H-SiC, BeO, and ZnS which crystallize in the same hexagonal  $C_{6v}^4$  structure.

**2 Symmetry of ZnO band structure** The states of spin less quasi-particles like phonons and excitons are classified according to the single-valued irreducible representations (SV-irrps) of the crystal space group. Recently, Lambrecht et al. [13] calculated the band structure of ZnO using first principles linear muffin-tin orbital density functional theory. In their computation the spin of electrons and holes was disregarded. However, the effect of the time reversal symmetry along the high symmetry line  $\Delta$  and at point  $A$  was taken into account.

Figure 1 in Ref. [13] gives an overview of the ZnO band structure throughout the entire Brillouin zone (BZ). Several minima of conduction bands at high symmetry points can be distinguished, e.g., for  $\Gamma$ ,  $M$ ,  $A$ ,  $L$ , and  $X$ . The corresponding symmetries are  $\Gamma_1$ ,  $M_1$ ,  $A_1$ ,  $L_1$ , and  $X_1$ . The valley  $A_1$  is time reversal degenerate. Therefore, its symmetry is supposed to be  $A_1 \oplus (A_1)^* = A_1 \oplus A_4$  [14]. The maxima of the valence bands at the  $\Gamma$  point and  $A$  point have the symmetries  $\Gamma_5$  and  $A_5$ , respectively.

For direct optical transitions in semiconductors, which preserve the  $k$  vector, an electron can be scattered by a phonon. In such a case the scattering processes can be described by group theoretical scattering selection rules regarding only the SVirrps [4, 5]. In most semiconductors, however, there is also an appreciable and measurable spin-orbit interaction. Consequently, the electron and hole states must be assigned by spinor representations. For ZnO, these are  $\Gamma_7$  for the conduction band and  $\Gamma_{7+}$ ,  $\Gamma_9$ ,  $\Gamma_{7-}$  for the upper valence bands. The spinor representations are also referred to as double-valued irrps (DV-irrps).

**3 The wave vector selection rules: Kronecker product and Clebsch–Gordan coefficients** In this section we discuss the consequences of the momentum

conservation principle, when an electron in the state  $k$  is scattered into another state with  $k'$  due to a phonon in the  $k''$  state. The momentum conservation determines the WVSRs. In ZnO as well as many other hexagonal crystals with the space group  $C_{6v}^4$ , there are three minima at the  $L$  and  $M$  points in the BZ. The first minimum at each of these points is characterized by the wave vectors  $k_L(101)$  and  $k_M(010)$  in the fundamental domain of the BZ [15]. The remaining two minima are derived from the  $k_L$  and  $k_M$  wave vectors to  $2k_L(011)$ ,  $3k_L(-111)$  and  $2k_M(100)$ ,  $3k_M(010)$ , where 1, 2, and 3 are the symmetry operators according to the CDML labeling [15]. Together with  $k_L(101)$  and  $k_M(010)$ , the wave vectors form so-called stars of the high symmetry points  $L$  and  $M$ . Combining these six wave vectors, we obtain the WVSRs of momenta conservation which are listed in Table 1 together with the calculated coupling coefficients. The  $U$  matrices for all tables are created from the elements of the Tables 1–5. For an example, see the listed  $U$  matrix of Table 3.

The description of any scattering process is given by the Kronecker product (KP) of irrps which corresponds to the particles involved in such processes. For example, for

**Table 2** Clebsch–Gordan coefficient matrix for  $(A_1 \oplus A_1^*) \otimes M_1 = [L_1 \oplus L_2]$ .

WVSRs	block	$aa'$	$L_1$			$L_2$		
	$\sigma\sigma'\sigma''$		1	2	3	1	2	3
$k_A + k_M = k_L$	111	11	0	0	0	$\pm i$	0	0
		11	$\pm i$	0	0	0	0	0
$k_A + 2k_M = 2k_L$	122	12	0	0	0	0	$\pm i$	0
		21	0	$\pm i$	0	0	0	0
$k_A + 3k_M = 3k_L$	133	13	0	0	0	0	0	$\pm i$
		31	0	0	$\pm i$	0	0	0

$$U^{-1}((A_1 + A_1^*) \otimes M_1)U = \begin{pmatrix} L_1 & 0 \\ 0 & L_2 \end{pmatrix}$$

**Table 3** Clebsch–Gordan coefficient matrix  $M_1 \otimes M_1 = [\Gamma_1 \oplus \Gamma_5]$ ,  $b = 1/\sqrt{3}$ ,  $w = -1/2 + i\sqrt{3}/2$ .

WVSRs	block			$\Gamma_1$		$\Gamma_5$	
	$\sigma\sigma'\sigma''$	$ad'$	$a'' =$	1	1	2	
$k_M + k_M = k_\Gamma$	111	11		$b$	$b$	$b\sqrt{w^*}$	
$2k_M + 2k_M = k_\Gamma$	221	12		$b$	$bw$	$bw^*\sqrt{w^*}$	
$3k_M + 3k_M = k_\Gamma$	331	13		$b$	$bw^*$	$bw\sqrt{w^*}$	

$$U^{-1}(M_1 \otimes M_1)U = \begin{pmatrix} \Gamma_1 & 0 \\ 0 & \Gamma_5 \end{pmatrix}$$

$$U = b \begin{pmatrix} 1 & 1 & \sqrt{w^*} \\ 1 & w & w^*\sqrt{w^*} \\ 1 & w^* & w\sqrt{w^*} \end{pmatrix}$$

the scattering of an electron from the  $L$  valley into the  $M$  valley, the KP  $L \otimes M$  must be decomposed into irrps of phonons. For this purpose, we use the comprehensive tables for KPs of the 230 space groups by Crackenell et al. [15] (CDML). The CGCs are the elements of a unitary matrix that reduces the direct KP of two reducible or irrps onto a sum of blocks of irrps contained in the product  $D^{\{^*k\}l} \otimes D^{\{^*k'\}l'}$  with

$$U^{-1}D^{\{^*k\}l} \otimes D^{\{^*k'\}l'} U = \sum_{l''} c_{ll''} D^{\{^*k''\}l''}, \quad (1)$$

where all quantities have their usual meaning [7]. The KP reflects different kinds of interactions like electron–hole, electron–phonon, phonon–phonon, etc. which are normally treated as perturbation of an unperturbed Hamiltonian. In other words, the CGCs matrix diagonalizes the perturbation terms. Selected CGCs for ZnO are listed in Tables 1–5. For detailed calculations of CGCs in other space groups, please refer to the Berenson–Birman–Kunert method [7, 10, 11].

**4 Scattering processes in ZnO** Despite the fact that ZnO is a direct band gap semiconductor, inter- and intra-valley scattering processes are possible. Carriers in semiconductors are scattered by their interaction with many different defects, like stacking faults, dislocations, surfaces,

**Table 4** Clebsch–Gordan coefficient matrix  $M_1 \otimes M_1 = [M_4]$ ,  $c = 1/\sqrt{2}$ ,  $w = -1/2 + i\sqrt{3}/2$ .

WVSRs	block			$M_4$		
	$\sigma\sigma'\sigma''$	$ad'$	$a'' =$	1	2	3
$k_M + 2k_M = 3k_M$	111	11		$c$	0	0
$k_M + 3k_M = 2k_M$	123	12		0	$w$	0
$2k_M + k_M = 3k_M$	231	13		0	0	$w^*$
$2k_M + 3k_M = k_M$	222	21		$-c$	0	0
$3k_M + k_M = 2k_M$	333	22		0	$w^*$	0
$3k_M + 2k_M = k_M$	312	23		0	0	$w$

$$U^{-1}(M_1 \otimes M_1)U = [M_4]$$

**Table 5** Clebsch–Gordan coefficient matrix for exciton wave functions originating from high symmetry point  $A$ ,  $A_7^{\text{CB}} \otimes A_9^{\text{VB}} = [\Gamma_5 \oplus \Gamma_6]$ .

WVSRs	block			$\Gamma_5$		$\Gamma_6$	
	$\sigma\sigma'\sigma''$	$ad'$	$a'' =$	1	2	1	2
$k_A + k_A = k_\Gamma$	111	11		0	$\pm i$	0	0
				0	0	$\pm i$	0
				0	0	0	$\pm i$
				$\pm i$	0	0	0

$$U^{-1}((A_7)^{\text{CB}} \otimes (A_9)^{\text{VB}})U = \begin{pmatrix} \Gamma_5 & 0 \\ 0 & \Gamma_6 \end{pmatrix}$$

interfaces, and impurities. Very common impurities with high concentrations in ZnO are, e.g., the group III donors Al, Ga, and In in the neutral ( $I_6, I_8, I_9$ ) and ionized charge state ( $I_0, I_1, I_{2/3}$ ) [16, 17]. Raman scattering in resonance with excitonic states results in a strong increase of the Raman cross-section due to the exciton–phonon interaction in ZnO [18]. In terms of electrical transport, the Boltzmann kinetic equation includes the effects on the distribution function  $f(\mathbf{k})$  due to the applied field, diffusion of carriers, and scattering by phonons and impurities [19, 20]. The scattering processes are determined by appropriate matrix elements of the relevant Hamiltonian. These matrix elements, in turn, depend on the symmetry and obey certain selection rules. From the band structure of ZnO [13], we can easily derive the symmetries of the valence bands and conduction bands at the high symmetry points  $M, L$ , and  $A$ . Here, we determine the CGCs for the intra- and inter-valley scattering processes involving the emission of phonons in ZnO. These CGCs are listed in Tables 1–4. For an excitation energy  $E > 3.8$  eV, the creation of excitons at the high symmetry point  $A$  is possible. The symmetries for these excitons are listed in Table 5.

**5 Tables of coefficient matrices** Following the general introduction about scattering processes in ZnO, we now provide detailed tables of CGCs for various scattering processes. For scattering processes between different valleys of the conduction band, the following KPs are required:  $L \otimes M, L \otimes \Gamma, L \otimes A, M \otimes \Gamma, M \otimes A$ , and  $\Gamma \otimes A$ . In Table 1, we list the CGCs for electron scattering between the  $L_1$  and  $M_1$  valleys resulting in large phonon momenta of  $A_1, A_5, L_1$ , and  $L_2$  symmetries ( $L_1 \otimes M_1 = A_1 \oplus A_5 \oplus L_1 \oplus L_2$ ).

Table 2 displays the CGCs for linear combinations of wave functions of phonons with large momenta  $\hbar k_{L_1}$  and  $\hbar k_{L_2}$  which originate from the scattering of electrons occupying the conduction band minima of  $A_1$  and  $M_1$  symmetry ( $(A_1 \oplus A_1^*) \otimes M_1 = [L_1 \oplus L_2]$ ).

Tables 3 and 4 consist of CGCs for scattering processes of electrons occupying three different sub-valleys with momenta  $\hbar k_M, 2\hbar k_M$ , and  $3\hbar k_M$  which belong to one minimum of the CB valley  $M_1$ . The scattering processes resulting in low momentum ( $\hbar k_\Gamma \approx 0$ ) of  $\Gamma_1$  and  $\Gamma_5$  phonons are shown in Table 3 ( $M_1 \otimes M_1 = [\Gamma_1 \oplus \Gamma_5]$ ). The CGCs

for  $M_4$  phonons with large momentum are listed in Table 4 ( $M_1 \otimes M_1 = [M_4]$ ).

In Table 5, we display an example of the symmetries and CGCs of possible excitons at the  $A$  point for  $(A_7)^{CB} \otimes (A_9)^{VB}$ . The conceivable excitons of  $A$  symmetries have low momentum and the same symmetries as the  $\Gamma$  excitons. It should be noted that the valence band symmetry at the  $A$  point in Table 5 describes a hole state from the  $B$  valence band as has been proven in a recent magneto-optical study of the valence band ordering in ZnO by Wagner et al. [21].

**6 Discussion** The CGCs provided in this work have several important applications. Principally, they are defined as coefficients between basis functions of the irrps contained in the KP and the basis functions of the product of two irrps [7, 11]

$$\psi_{a''}^{k'' \sigma'' l'' \gamma} = \sum_{\sigma a} \sum_{\sigma' a'} \left( \begin{array}{cc|cc} \mathbf{k} & l & \mathbf{k}' & l' \\ \sigma & a & \sigma' & a' \end{array} \middle| \begin{array}{cc|c} \mathbf{k}'' & l'' & \gamma \\ \sigma'' & a'' & \end{array} \right) \Phi_a^{k \sigma l} \Phi_{a'}^{k' \sigma' l'}, \quad (2)$$

where

$$\left( \begin{array}{cc|cc} \mathbf{k} & l & \mathbf{k}' & l' \\ \sigma & a & \sigma' & a' \end{array} \middle| \begin{array}{cc|c} \mathbf{k}'' & l'' & \gamma \\ \sigma'' & a'' & \end{array} \right) = U_{aa'; a''}^{\sigma l \sigma' l'; \sigma'' l'' \gamma} \quad (3)$$

are the matrix elements of the CGCs. The symbols  $a, a',$  and  $a''$  indicate the matrix elements of the KP  $l \otimes l'$  and the representation contained in the KP ( $l''$ ). These symbols  $a, a',$  and  $a''$  enumerate also the basis wave functions of the irrps  $l, l',$  and  $l''$ , respectively (see Eq. (4)). The  $\mathbf{k}, \mathbf{k}'$  and  $\mathbf{k}''$  stand for wave vectors (momenta  $\hbar k$ ) of quasi particles involved in scattering processes such as electrons and phonons. The  $U$  matrix (CGC matrix) has two important physical meanings. On the one hand, it yields suitable linear combinations of the appropriate symmetry allowed basis functions of irrps ( $l''$ ) contained in the KP in terms of product wave functions of  $l \otimes l'$  (see Eqs. (2) and (3)). On the other hand, the  $U$  matrix diagonalizes any KP of two irrps ( $l \otimes l'$ ) and brings the irrps  $l''$  to the matrix diagonal block form (see Eq. (1) and the equations below the Tables 1–5).

For example, using our coefficients listed in Table 3, the appropriate symmetries of the  $\Gamma_5$  wave functions contained in  $M_1 \otimes M_1$  can be determined to:

$$\psi_{11}^{\Gamma_5} = \frac{1}{\sqrt{3}} \left\{ \psi_{11}^{k_M M_1} + w \psi_{22}^{2k_M M_1} + w^* \psi_{33}^{3k_M M_1} \right\},$$

$$\psi_{22}^{\Gamma_5} = \frac{1}{\sqrt{3}} \sqrt{w^*} \left\{ \psi_{11}^{k_M M_1} + w^* \psi_{22}^{2k_M M_1} + w^* \psi_{33}^{3k_M M_1} \right\}. \quad (4)$$

Next, the CGCs can also be used in the determination of the scattering tensors  $\mathbf{P}$ . When expanded in powers of

$kQ$ , we have [20]:

$$\mathbf{P}_{\alpha\beta} = \mathbf{P}_{\alpha\beta}^0 + \sum_{\mu\nu} \mathbf{P}_{\alpha\beta}^{j\nu} \mathbf{k}^j \cdot \mathbf{Q}(\mu\nu) + \dots \quad (5)$$

and in terms of CGCs they are

$$\mathbf{P}_{\alpha\beta}^{j\nu} = \sum_{\mu\nu} c(\mu\nu m) U_{vj, mn}^{l \otimes l'} \mathbf{P}_{\alpha\beta}^{(1)}(\mu\nu), \quad (6)$$

where  $\mathbf{P}$  is the first order scattering tensor, and  $U$  is the matrix of the CGCs. The quantities in the above equations have their usual meanings [20]. The CGCs can also be used in the description of morphic effects [12], higher infrared order moment expansion, diagonalization of phonon dynamical matrices, as well as in the description of Gunn effect (inter-valley electron–phonon interaction) (see Ref. [1] and references therein). The coefficients can essentially be utilized in the construction of the effective Luttinger–Bir–Picus Hamiltonian matrix elements [9]:

$$H(K) = \sum_{\kappa} a_{\kappa} \delta_{\kappa, jj^*} \sum_l X_l^{(\kappa)} K_l^{(\kappa)*}, \quad (7)$$

where  $a$  is the constant and  $K_l$  is Hermitian. The  $X_l$  span the  $l$ -dimensional linear vector space consisting of independent matrices. Birman et al. [9] have shown that the matrices are exactly the CGCs and therefore each element of an effective Hamiltonian matrix is a sum of symmetry adapted components of invariants times a CGC with the reduced matrix given by means of the Wigner–Eckart theorem:

$$H_{\alpha\beta}(K) = \sum_{\kappa} a_{\kappa} \sum_l (U^{-1})_{\alpha\beta l}^{j^* j \kappa} K_l^{(\kappa)*}. \quad (8)$$

Using the CGCs and invariants, Birman et al. [9] constructed the effective Hamiltonian matrix for Diamond ( $O_h^7$ ) with the  $\mathbf{k} \cdot \mathbf{p}$  expansion method to the second order degree in  $k$  of the twofold degenerate  $\Gamma_{12+}$  state. With the here provided CGCs, a similar approach can be applied to construct the effective Hamiltonians for ZnO.

**7 Conclusion** In summary, we have studied the scattering tensors in ZnO by means of CGCs. Starting from existing band structure calculations, the CGCs were derived from selection rules for inter- and intra-valley scattering processes. For scattering processes in the same valley, both cases of low and large momentum of the quasi-particles were considered. In addition, the symmetries and CGCs of excitons at the high symmetry point  $A$  were determined.

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