

Particle on a Ring Spectroscopic Selection Rules Determined by Group Theory

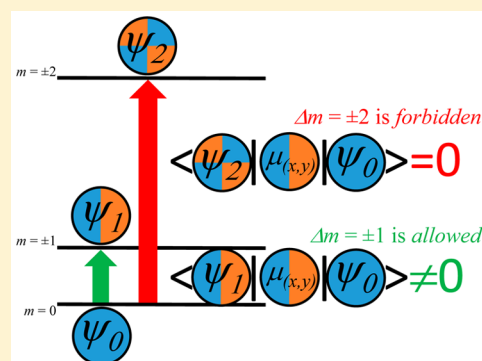
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S Supporting Information

ABSTRACT: Early introduction of symmetry concepts in the Physical Chemistry curriculum has been shown to help students understand the relative simplicity of spectroscopic selection rules. Most Quantum Mechanics and Spectroscopy textbooks begin with various one-dimensional problems. Application of symmetry arguments to the particle-in-a-box problem is presented in some books, but no sources have been found where symmetry arguments are used to determine the selection rules for particle-on-a-ring spectroscopic transitions. This hinders the early introduction of symmetry concepts. This article removes this hindrance by deriving the particle-on-a-ring rotational selection rules using group theory symmetry arguments. The $D_{\infty h}$ character table is used to define rotational wave function symmetry, and the $D_{\infty h}$ direct product table is used to determine the nonzero behavior of the transition dipole moment integral. A survey of the symmetry of allowed transitions leads to the well-known rotational selection rules of $\Delta m = 0$ for Rayleigh scattering, $\Delta m = \pm 1$ for direct absorption and emission, and $\Delta m = \pm 2$ for Raman scattering. This approach will allow the use of symmetry arguments for spectroscopic selection rule determination for the one-dimensional problems that establish the early foundation in Quantum Mechanics and Spectroscopy courses.

KEYWORDS: Upper-Division Undergraduate, Graduate Education/Research, Physical Chemistry, Analogies/Transfer, Problem Solving/Decision Making, Group Theory/Symmetry, Quantum Chemistry, Raman Spectroscopy, Spectroscopy



INTRODUCTION

This communication is written to the physical chemistry professor for the benefit of the student to clarify the connection between spectroscopic measurement, molecular symmetry, group theory, and quantum mechanics. When introducing quantum theory, every major physical chemistry text begins by citing various failures of classical physics.^{1–3} The overwhelming majority of these failures involve spectroscopy. The transition dipole moment (TDM) integral is central to these failures and is at the heart of all spectroscopic measurements as it defines the wave function interaction in a transition. In a spectrum, the selection rules record which transitions can occur through interaction with light.⁴ These rules do not distinguish the intensity that the transition will exhibit but merely list which transitions are allowed or forbidden. Understanding how the TDM integral governs the selection rules will open up many spectroscopic techniques to the student, and will give them an understanding of the unshakable connection between the atomic-level wave function and the laboratory-level measurement of spectroscopic transitions.

The selection rules can be calculated using analytical calculus for 1D systems such as the particle in a box (PB), the particle on a ring (PR), and the harmonic oscillator (HO). But these analytical solutions are moderately long, and the students get lost in understanding the original purpose. Student comments

supporting this conclusion are available as [Supporting Information](#).

It is precisely here that symmetry finds its greatest use. The symmetry of the PB and HO problems is obvious, and the odd–even symmetry selection rules developed in these 1D cases can be used to explain molecular electronic and vibrational selection rules.

One problem exists with this early introduction approach: the inability to use symmetry arguments for rotational selection rules. The 1D PR selection rules traditionally have been justified using the conservation of angular momentum. The professor must either state this condition by fiat, or take a detour introducing a separate concept of angular momentum for rotational systems. In an effort to avoid this detour, and unify defining selection rules for 1D systems in a simplistic manner, this article has derived the rotational selection rules based on symmetry.

Previous articles have covered many aspects of spectroscopic transitions. Such aspects include the definition of the rotational energy levels of the PR system, and the basic derivation of selection rules deduced from the electric dipole transition.^{5,6} Since the derivation of rotational selection rules can be daunting, some have rationalized the selection rules for student simplicity.⁷ Other articles have used group theory to derive

these selection rules for three-dimensional rotation⁸ and labeled symmetry species for rotational levels within full symmetry groups.⁹ Some articles have shown the selection rules through symmetry conservation without using the TDM integral, while others have given credit for the justification of selection rules to the quantization of light.^{10,11} However, none of these sources present the PR selection rule derivation at a level suitable for early introduction in the semester.

■ TRANSITION DIPOLE APPROXIMATION

Selection rules are based on the approximation that the intensity of a transition is proportional to the square of the TDM integral (eq 1). Typical examples involve an electric dipole (as in electronic spectroscopy, permanent dipoles in rotating polar molecules, or fluctuating dipoles in vibrating molecules), but other transition moments can be studied such as the polarizability tensor in Raman scattering. The electric dipole function is represented by μ in eq 1, ψ_n is the upper wave function, and ψ_n is the lower wave function in the evaluated transition.¹ The TDM integral is an assessment of the interaction between the levels in a transition and the dipole moment operator.¹² Those transitions with an integral that equals zero are forbidden, and those with nonzero integrals are allowed.¹ A shorthand representation of the same TDM integral with Dirac notation is used for simplicity (eq 2).

$$I \propto [\int \psi_n \mu \psi_n dx]^2 \quad (1)$$

$$I \propto \langle n' | \mu | n \rangle^2 \quad (2)$$

■ SYMMETRY-DEFINED SELECTION RULES

To determine the selection rules, symmetry is used to predict the resulting value (either zero or nonzero) of the transition dipole moment integral (eqs 3 and 4).¹²

$$\begin{aligned} \langle \text{Even} | \text{Odd} | \text{Even} \rangle &= \langle \text{Even} | \text{Odd} \rangle \\ &= \langle \text{Odd} \rangle \\ &= 0 \therefore \text{Forbidden} \end{aligned} \quad (3)$$

$$\langle \text{Odd} | \text{Odd} | \text{Even} \rangle = \langle \text{Odd} | \text{Odd} \rangle = \langle \text{Even} \rangle \neq 0 \therefore \text{Allowed} \quad (4)$$

Each wave function in the integrand is identified as having an odd or even symmetry based on the properties of even and odd functions. If $f(x)$ equals $f(-x)$, then $f(x)$ is an even function. If $f(-x)$ equals $-f(x)$, then $f(x)$ is an odd function.⁴ The direct product of the irreducible representations of each wave function in the transition and the dipole operator will yield the overall symmetry of the integrand (eqs 3 and 4).

■ ROTATIONAL SELECTION RULES

Due to the circular nature of the coordinate system, the even or odd symmetry of the wave function is difficult to establish. The PR wave function has real and imaginary portions. Since Euler's relationship defines both portions as having the same symmetry merely shifted by $\pi/2$ from each other, the evaluation of only the real portion is enough to identify rotational wave function symmetry. The normalized PR wave function is shown in eq 5.⁴ The wave functions are denoted by the rotational quantum number m and the rotational angle on the ring in radians, ϕ . The quantum number can be positive and negative integers including zero. Thus, when m is negative, the arguments inside

the cos and sin functions are negative. This is interpreted as rotation in the opposite direction as positive m wave functions. This is also an example of degenerate energy states for $m \neq 0$.

$$\psi_m(\phi) = \sqrt{\frac{1}{2\pi}} [\cos(m\phi) + i \sin(m\phi)] \quad (5)$$

For this case when manipulating the 1D PR system, it will be understood as an electron orbiting on a ring with a fixed radius confined to the x, y plane therefore giving a single variable, ϕ , the angle of rotation. In this specific case, the rotational selection rules would be called the electronic angular momentum selection rules. Figure 1 defines the coordinate system of the ring and the angle of rotation.

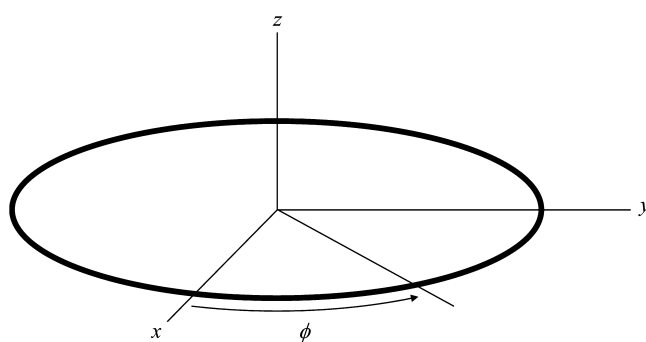


Figure 1. One dimensional ring with defined coordinate system and rotational angle.

■ RING SYMMETRY

The symmetry of the ring-shaped coordinate system may be used to determine the point group, and the subsequent symmetry of each rotational wave function is derived from the character table of that point group (Table 1). Since the ring has the same symmetrical properties of a cylinder with an inversion center, the point group of $D_{\infty h}$ denotes its symmetry.⁴ Another view is through extrapolation from cyclopropane (D_{3h}) to benzene (D_{6h}) to a ring ($D_{\infty h}$) with infinite C_2 and σ_v giving the 1DPR system a $D_{\infty h}$ symmetry.⁴

■ ASSIGNING SYMMETRY TO ROTATIONAL WAVE FUNCTIONS

Each wave function is determined to be even (gerade) or odd (ungerade) by inspecting its behavior upon inversion through aerial observation of the positive and negative lobes on opposing sides of the ring (Figures 2 and 3).¹⁵ Wave functions with lobes that are unchanged by inversion are gerade, g, while those which are inverted are ungerade, u (Table 1). Further wave function classification is determined by recognizing that the characters under the rotational symmetry element ($2C_{\infty}^{\phi}$) are in fact the real portion of the 1D PR wave function (eq 5).¹⁴ For example, the E_1 character $2 \cos(\phi)$ is similar to the real part of the $m = \pm 1$ wave function, while the E_2 character $2 \cos(2\phi)$ is similar to the real part of the $m = \pm 2$ wave function. The electric dipole μ of a charged particle is typically represented as the x -axis (μ_x). However, in the $D_{\infty h}$ character table, x and y are inseparable and thus the dipole moment operator $\mu_{(x,y)}$ will have the E_{1u} irreducible representation. If this were a 3D system, μ_z (A_{1u}) would play a role. But with the electron confined to the xy -plane, a dipole along the z -axis is impossible.

Table 1. $D_{\infty h}$ Character Table¹²

$D_{\infty h}$	E	$2C_{\infty}^{\phi}$	...	$\infty\sigma_v$	i	$2S_{\infty}^{\phi}$	...	∞C_2	
$A_{1g} = \sum_g^+$	1	1		1	1	1		1	x^2+y^2, z^2
$A_{2g} = \sum_g^-$	1	1		-1	1	1		-1	R_z
$E_{1g} = \prod_g$	2	$2\cos(\phi)$		0	2	$-2\cos(\phi)$		0	$(R_x, R_y), (xz, yz)$
$E_{2g} = \Delta_g$	2	$2\cos(2\phi)$		0	2	$2\cos(2\phi)$		0	(x^2-y^2, xy)
E_{3g}	2	$2\cos(3\phi)$		0	2	$-2\cos(3\phi)$		0	
E_{4g}	2	$2\cos(4\phi)$		0	2	$2\cos(4\phi)$		0	
E_{mg}	2	$2\cos(m\phi)$		0	2	$(-1)^m 2\cos(m\phi)$		0	
$A_{1u} = \sum_u^+$	1	1		1	-1	-1		-1	z
$A_{2u} = \sum_u^-$	1	1		-1	-1	-1		1	
$E_{1u} = \prod_u$	2	$2\cos(\phi)$		0	-2	$2\cos(\phi)$		0	(x, y)
$E_{2u} = \Delta_u$	2	$2\cos(2\phi)$		0	-2	$-2\cos(2\phi)$		0	
E_{3u}	2	$2\cos(3\phi)$		0	-2	$2\cos(3\phi)$		0	
E_{4u}	2	$2\cos(4\phi)$		0	-2	$-2\cos(4\phi)$		0	
E_{mu}	2	$2\cos(m\phi)$		0	-2	$(-1)^{m+1} 2\cos(m\phi)$		0	

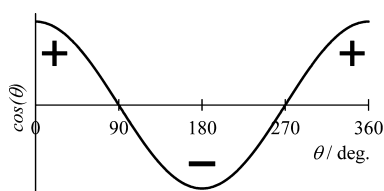


Figure 2. Rotational wave function $|m| = 1$ plotted from 0 to 2π (0° to 360°). Positive and negative lobes establish the positive and negative wedges in a top-down plot of the wave functions.

As every rotational wave function is assigned an irreducible representation, trends appear making it simpler to further label all higher-level wave functions (Figure 3). Above the ground state, the number in each irreducible representation corresponds to the quantum number, and ungerade is assigned to odd quantum numbers while gerade is assigned to even quantum numbers. This is summarized in eqs 6.1–6.3.

$$A_{1g} \text{ for } m = 0 \quad (6.1)$$

$$E_{|m|g} \text{ for even } |m| > 0 \quad (6.2)$$

$$E_{|m|u} \text{ for odd } m \quad (6.3)$$

EXPANDING DIRECT PRODUCT TABLE

The product of the given irreducible representations in the transition will determine the overall symmetry of the TDM integrand. We found it necessary to expand the direct product tables to E_6 ($m = \pm 6$) to be sure of the selection rule trends. This expansion to produce Table 3 is illustrated in Table 2.

$$E_1 \otimes E_2 = E_1 \oplus E_3 \quad (7)$$

SELECTION RULE DETERMINATION

The symmetry of the integrand can be determined using the $D_{\infty h}$ direct product table. An allowed transition (i.e., nonzero value for the TDM) is obtained only if the symmetry of the integrand contains the totally symmetric irreducible representation A_{1g} in the $D_{\infty h}$ character table.¹² The following equations present the results for $\Delta m = \{\pm 1, \pm 2, \text{ and } \pm 3\}$ which is enough to establish a trend that precludes any allowed higher Δm transitions.

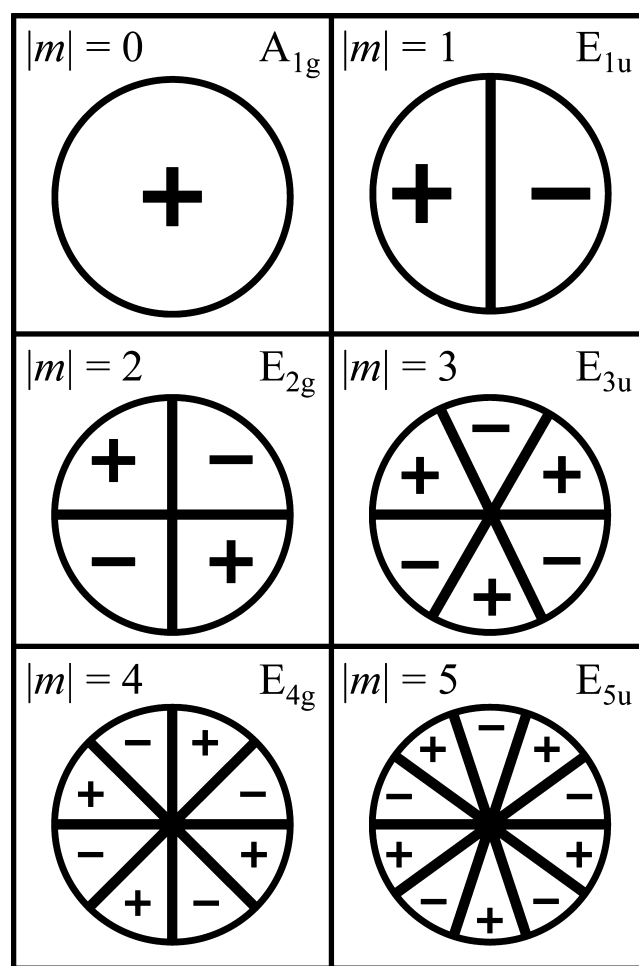


Figure 3. Top-down plot of the real portions of the first six PR wave functions and their irreducible representations.

The A_{1g} symmetry in the integrand in the direct product results in Table 4 yields an allowed transition, confirming the selection rule of $\Delta m = \pm 1$.

The transitions with $\Delta m = \{\pm 2, \pm 4, \dots\}$ are all forbidden due to the g–u product rules ($\Gamma_g \otimes \Gamma_g = \Gamma_g$; $\Gamma_u \otimes \Gamma_u = \Gamma_g$; $\Gamma_g \otimes \Gamma_g = \Gamma_u$). The resulting integrand will have u symmetry and cannot possibly contain A_{1g} .

The results in Table 5 show the transitions from $m = 0$ to $\{\pm 3, \pm 5, \dots\}$. The $\Delta m = \pm 3$ transitions in Table 5 require some

Table 2. Example Showing the Validity of Eq 7^a

Representation	E	$2C_{\infty}^{\phi}$	$\infty\sigma_v$	i	$2S_{\infty}^{\phi}$	∞C_2
E_{1g}	2	$2 \cos(\phi)$	0	2	$-2 \cos(\phi)$	0
E_{2g}	2	$2 \cos(2\phi)$	0	2	$2 \cos(2\phi)$	0
E_{3g}	2	$2 \cos(3\phi)$	0	2	$-2 \cos(3\phi)$	0
$E_1 \otimes E_2$	4	$2 \cos(\phi) + 2 \cos(3\phi)$	0	4	$-2 \cos(\phi) - 2 \cos(3\phi)$	0
$E_1 \oplus E_3$	4	$2 \cos(\phi) + 2 \cos(3\phi)$	0	4	$-2 \cos(\phi) - 2 \cos(3\phi)$	0

^aRow 4 shows the left side of eq 7, and row 5 shows the right side of eq7.

analysis of the trends seen in the direct product table (Table 3). They are forbidden because the only way to yield an A_{1g} irreducible representation would be for the direct product of $E_{1u} \otimes E_m$ to yield an $E_{(m+3)}$ irreducible representation. This is impossible because E_1 can only increase the subscript by one ($E_1 \otimes E_m = E_{(m+1)} \oplus \dots$). This same limitation will occur for all higher Δm transitions, making $\Delta m = \pm 1$ the only allowed transition type in the 1DPR system for direct absorption and emission spectroscopies.

The Rayleigh ($\Delta m = 0$) and Raman ($\Delta m = \pm 2$) scattering selection rules can be determined using this method. These scattering phenomena utilize the polarizability operator (α), which transforms as all the second-order terms ($x^2 + y^2, xz$, etc.) in the character table ($A_{1g} \oplus E_{1g} \oplus E_{2g}$).

$$I \propto \langle m' | \alpha | m \rangle^2 \quad (9)$$

$$I \propto \langle m' | A_{1g} \oplus E_{1g} \oplus E_{2g} | m \rangle^2 \quad (10)$$

The solution of eq 10 for the Rayleigh scattering selection rule ($\Delta m = 0$) is shown in Table 6. The first row of Table 6 yields an allowed transition because of the product of three A_{1g} irreducible representations. The second and third rows show that Rayleigh scattering is allowed for $m \neq 0$ because the E_m on the right survives left-multiplication by the A_{1g} portion of the polarizability operator and the direct product of any irreducible representation with itself ($E_m \otimes E_m$) will contain A_{1g} .

The allowed transitions for Raman scattering ($\Delta m = \pm 2$) are shown in Table 7. The first row in Table 7 yields an allowed transition because the E_{2g} in the polarizability operator survives multiplication on the right by A_{1g} and yields A_{1g} when left-multiplied by E_{2g} . The second and third rows in Table 7 show that Raman scattering is allowed for all $m \neq 0$ because $E_2 \otimes E_m = E_{m-2} \oplus E_{m+2}$ and $E_{m+2} \otimes E_{m+2}$ will contain A_{1g} .

The polarizability operator is even (gerade) while the electric dipole operator is odd (ungerade). Therefore, the $\Delta m = \{\pm 1, \pm 3, \dots\}$ transitions fail the g-u symmetry requirement under the polarizability operator just as the $\Delta m = \{\pm 2, \pm 4, \dots\}$

Table 4. Analysis Showing That $\Delta m = \pm 1$ Transitions Are Allowed by Symmetry

Transition		Symmetries		Result (All $\neq 0$)
$\langle 1 \mu_{(xy)} 0 \rangle$	=	$\langle E_{1u} E_{1u} A_{1g} \rangle$	=	$\langle A_{1g} \oplus A_{2g} \oplus E_{2g} \rangle$
$\langle m + 1_{\text{odd}} \mu_{(xy)} m_{\text{even}} \rangle$	=	$\langle E_{(m+1)u} E_{1u} E_{mg} \rangle$	=	$\langle A_{1g} \oplus E_{2g} \oplus E_{2mg} \oplus A_{2g} \oplus E_{2(m+1)g} \rangle$
$\langle m + 1_{\text{even}} \mu_{(xy)} m_{\text{odd}} \rangle$	=	$\langle E_{(m+1)g} E_{1u} E_{mu} \rangle$	=	$\langle A_{1g} \oplus E_{2g} \oplus E_{2mg} \oplus A_{2g} \oplus E_{2(m+1)g} \rangle$

Table 5. Analysis Showing That $\Delta m = \pm 3$ Transitions Are Forbidden by Symmetry

Transition		Symmetries		Result (All = 0)
$\langle 3 \mu_{(xy)} 0 \rangle$	=	$\langle E_{3u} E_{1u} A_{1g} \rangle$	=	$\langle E_{2g} \oplus E_{4g} \rangle$
$\langle m + 3_{\text{odd}} \mu_{(xy)} m_{\text{even}} \rangle$	=	$\langle E_{(m+3)u} E_{1u} E_{mg} \rangle$	=	$\langle E_{(m+3)u} \oplus E_{(m+1)u} \oplus E_{(m-1)u} \rangle$
$\langle m + 3_{\text{even}} \mu_{(xy)} m_{\text{odd}} \rangle$	=	$\langle E_{(m+3)g} E_{1u} E_{mu} \rangle$	=	$\langle E_{(m+3)g} \oplus E_{(m+1)g} \oplus E_{(m-1)g} \rangle$

Table 6. Analysis Showing That Rayleigh Scattering $\Delta m = 0$ Is Allowed by Symmetry

Transition		Symmetries		Result (All $\neq 0$)
$\langle 0 \alpha 0 \rangle$	=	$\langle A_{1g} A_{1g} \oplus E_{1g} \oplus E_{2g} A_{1g} \rangle$	=	$\langle A_{1g} \oplus E_{1g} \oplus E_{2g} \rangle$
$\langle m_{\text{even}} \alpha m_{\text{even}} \rangle$	=	$\langle E_{mg} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mg} \rangle$	=	$\langle A_{1g} \oplus \dots \rangle$
$\langle m_{\text{odd}} \alpha m_{\text{odd}} \rangle$	=	$\langle E_{mu} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mu} \rangle$	=	$\langle A_{1g} \oplus \dots \rangle$

transitions fail the g-u symmetry requirement under the electric dipole operator.

Table 8 shows that $\Delta m = \pm 4$ transitions are forbidden even though they pass the g-u symmetry requirement. The only way to yield an A_{1g} irreducible representation would be for the direct product of $\alpha \otimes E_m$ to yield an $E_{(m+4)}$ irreducible representation. The polarizability tensor can only increase the subscript by two ($E_2 \otimes E_m = E_{(m+2)} \oplus \dots$). This same limitation will occur for all higher Δm transitions, making $\Delta m = \pm 2$ (+2 for Stokes shift; -2 for anti-Stokes shift) the only allowed transition type in the 1DPR system for Raman scattering.

CONCLUSION

Introduction of symmetry concepts early in the Physical Chemistry curriculum when students are working on the 1D

Table 3. $D_{\infty h}$ Expanded Direct Product Table

$D_{\infty h}$	A_1	A_2	E_1	E_2	E_3	E_4	E_5	E_6
A_1	A_1	A_2	E_1	E_2	E_3	E_4	E_5	E_6
A_2		A_1	E_1	E_2	E_3	E_4	E_5	E_6
E_1			$A_1 + A_2 + E_2$	$E_1 + E_3$	$E_2 + E_4$	$E_3 + E_5$	$E_4 + E_6$	$E_5 + E_7$
E_2				$A_1 + A_2 + E_4$	$E_1 + E_5$	$E_2 + E_6$	$E_3 + E_7$	$E_4 + E_8$
E_3					$A_1 + A_2 + E_6$	$E_1 + E_7$	$E_2 + E_8$	$E_3 + E_9$
E_4						$A_1 + A_2 + E_8$	$E_1 + E_9$	$E_2 + E_{10}$
E_5							$A_1 + A_2 + E_{10}$	$E_1 + E_{11}$
E_6								$A_1 + A_2 + E_{12}$

Table 7. Analysis Showing That Raman Scattering $\Delta m = \pm 2$ Is Allowed by Symmetry

Transition		Symmetries		Result (All $\neq 0$)
$\langle 2l\alpha 0 \rangle$	=	$\langle E_{2g} A_{1g} \oplus E_{1g} \oplus E_{2g} A_{1g} \rangle$	=	$\langle A_{1g} \oplus \dots \rangle$
$\langle m + 2_{\text{even}} \alpha l m_{\text{even}} \rangle$	=	$\langle E_{(m+2)g} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mg} \rangle$	=	$\langle A_{1g} \oplus \dots \rangle$
$\langle m + 2_{\text{odd}} \alpha l m_{\text{odd}} \rangle$	=	$\langle E_{(m+2)u} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mu} \rangle$	=	$\langle A_{1g} \oplus \dots \rangle$

Table 8. Analysis Showing That Raman Scattering with $\Delta m = \pm 4$ Is Forbidden by Symmetry

Transition		Symmetries		Result (All = 0)
$\langle 4l\alpha 0 \rangle$	=	$\langle E_{4g} A_{1g} \oplus E_{1g} \oplus E_{2g} A_{1g} \rangle$	=	$\langle E_{2g} \oplus E_{3g} \oplus E_{4g} \oplus E_{5g} \oplus E_{6g} \rangle$
$\langle m + 4_{\text{even}} \alpha l m_{\text{even}} \rangle$	=	$\langle E_{(m+4)g} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mg} \rangle$	=	$\langle E_{(m+4)g} E_{(m+2)g} \oplus \dots \rangle$
$\langle m + 4_{\text{odd}} \alpha l m_{\text{odd}} \rangle$	=	$\langle E_{(m+4)u} A_{1g} \oplus E_{1g} \oplus E_{2g} E_{mu} \rangle$	=	$\langle E_{(m+4)u} E_{(m+2)u} \oplus \dots \rangle$

problems is a good way to convince them of the value of symmetry in the prediction of the selection rules of spectroscopic phenomena. This article presents symmetry arguments to derive the rotational selection rules so that symmetry can be used for all of the standard 1D systems.

By associating the rotational symmetry of the PR problem with the $D_{\infty h}$ character table and assigning the wave functions to irreducible representations, the direct product table may be utilized to compute the symmetry of the TDM integrand and the nonzero behavior of all possible TDM integrals. As a result, the selection rules for Rayleigh scattering ($\Delta m = 0$), direct absorption and emission ($\Delta m = \pm 1$), and Raman scattering ($\Delta m = \pm 2$) were derived for the 1DPR system.

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available on the ACS Publications website at DOI: 10.1021/ed500699q.

Nonedited results of a questionnaire requesting student input on the utility of symmetry in their understanding of spectroscopic concepts (PDF, DOCX)

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Notes

The authors declare no competing financial interest.

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