ΔI = 2 staggering in rotational bands of diatomic molecules as a manifestation of interband interactions

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Abstract

It is shown that the recently observed ΔI = 2 staggering seen in superdeformed nuclear bands is also occurring in certain electronically excited rotational bands of diatomic molecules. In the case of diatomic molecules the effect is attributed to interband interactions (bandcrossings).

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I. Introduction

Several staggering effects are known in nuclear spectroscopy [1]:
1) In rotational $\gamma$ bands of even nuclei the energy levels with odd angular momentum $I$ ($I=3, 5, 7, 9, \ldots$) are slightly displaced relatively to the levels with even $I$ ($I=2, 4, 6, 8, \ldots$), i.e. the odd levels do not lie at the energies predicted by an $E(I) = AI(I + 1)$ fit to the even levels, but all of them lie systematically above or all of them lie systematically below the predicted energies [2].

2) In octupole bands of even nuclei the levels with odd $I$ and negative parity ($I^\pi=1^-, 3^-, 5^-, 7^-, \ldots$) are displaced relatively to the levels with even $I$ and positive parity ($I^\pi=0^+, 2^+, 4^+, 6^+, \ldots$) [3].

3) In odd nuclei, rotational bands (with $K=1/2$) separate into signature partners, i.e. the levels with $I=3/2, 7/2, 11/2, 15/2, \ldots$ are displaced relatively to the levels with $I=1/2, 5/2, 9/2, 13/2, \ldots$ [4].

In all of the above mentioned cases each level with angular momentum $I$ is displaced relatively to its neighbours with angular momentum $I \pm 1$. The effect is then called $\Delta I = 1$ staggering.

A new kind of staggering ($\Delta I = 2$ staggering) has been recently observed [5, 6] in superdeformed nuclear bands [7, 8, 9]. If $\Delta I = 2$ staggering is present, then, for example, the levels with $I=2, 6, 10, 14, \ldots$ are displaced relatively to the levels with $I=0, 4, 8, 12, \ldots$, i.e. the level with angular momentum $I$ is displaced relatively to its neighbours with angular momentum $I \pm 2$.

Although $\Delta I = 1$ staggering of the types mentioned above has been observed in several nuclei and certainly is an effect larger than the relevant experimental uncertainties, $\Delta I = 2$ staggering has been seen in only a few cases [3, 8, 11, 14] and, in addition, the effect is not clearly larger than the relevant experimental errors.

There have been by now several theoretical works related to the possible physical origin of the $\Delta I = 2$ staggering effect [12, 13, 14, 15, 16, 17, 18], some of them [13, 21, 22, 23, 24] using symmetry arguments which could be of applicability to other physical systems as well.

On the other hand, rotational spectra of diatomic molecules [25] are known to show great similarities to nuclear rotational spectra, having in addition the advantage that observed rotational bands in several diatomic molecules [26, 27, 28, 29] are much longer than the usual rotational nuclear bands. We have been therefore motivated to make a search for $\Delta I = 2$ staggering in rotational bands of diatomic molecules. The questions to which we have hoped to provide answers are:

1) Is there $\Delta I = 2$ staggering in rotational bands of diatomic molecules?
2) If there is, what are its possible physical origins?

In Section II of the present work the $\Delta I = 2$ staggering in superdeformed nuclear bands will be briefly reviewed. Evidence from existing experimental data for $\Delta I = 2$ staggering in rotational bands of diatomic molecules will be presented in Section III and discussed in Section IV, while Section V will contain the conclusions drawn.

II. $\Delta I = 2$ staggering in superdeformed nuclear bands

In nuclear physics the experimentally determined quantities are the $\gamma$-ray transition energies between levels differing by two units of angular momentum ($\Delta I = 2$). For these the symbol

$$ E_{2,\gamma}(I) = E(I + 2) - E(I) $$

(1)
is used, where $E(I)$ denotes the energy of the level with angular momentum $I$. The deviation of the $\gamma$-ray transition energies from the rigid rotator behavior can be measured by the quantity

$$\Delta E_{2,\gamma}(I) = \frac{1}{16}(6E_{2,\gamma}(I) - 4E_{2,\gamma}(I - 2) - 4E_{2,\gamma}(I + 2) + E_{2,\gamma}(I - 4) + E_{2,\gamma}(I + 4)). \tag{2}$$

Using the rigid rotator expression

$$E(I) = AI(I + 1), \tag{3}$$

one can easily see that in this case $\Delta E_{2,\gamma}(I)$ vanishes. In addition the perturbed rigid rotator expression

$$E(I) = AI(I + 1) + B(I(I + 1))^2, \tag{4}$$

gives vanishing $\Delta E_{2,\gamma}(I)$. These properties are due to the fact that Eq. (2) is a (normalized) discrete approximation of the fourth derivative of the function $E_{2,\gamma}(I)$, i.e. essentially the fifth derivative of the function $E(I)$.

In superdeformed nuclear bands the angular momentum of the observed states is in most cases unknown. To avoid this difficulty, the quantity $\Delta E_{2,\gamma}$ is usually plotted not versus the angular momentum $I$, but versus the angular frequency

$$\omega = \frac{dE(I)}{dI}, \tag{5}$$

which for discrete states takes the approximate form

$$\omega = \frac{E(I + 2) - E(I)}{\sqrt{(I + 2)(I + 3)} - \sqrt{I(I + 1)}}. \tag{6}$$

For large $I$ one can take the Taylor expansions of the square roots in the denominator, thus obtaining

$$\omega = \frac{E(I + 2) - E(I)}{2} = \frac{E_{2,\gamma}(I)}{2}. \tag{7}$$

Examples of superdeformed nuclear bands exhibiting staggering are shown in Figs 1–2. We say that $\Delta I = 2$ staggering is observed if the quantity $\Delta E_2(I)$ exhibits alternating signs with increasing $\omega$ (i.e. with increasing $I$, according to Eq. (7)). The following observations can be made:

1) The magnitude of $\Delta E_2(I)$ is of the order of $10^{-4}–10^{-5}$ times the size of the gamma transition energies.

2) The best example of $\Delta I = 2$ staggering is given by the first superdeformed band of $^{149}$Gd, shown in Fig. 1a. In this case the effect is almost larger than the experimental error.

3) In most cases the $\Delta I = 2$ staggering is smaller than the experimental error (see Figs 1b, 2a, 2b), with the exception of a few points in Fig. 1b.

III. $\Delta I = 2$ staggering in rotational bands of diatomic molecules

In the case of molecules the experimentally determined quantities regard the $R$ branch ($(v_{lower}, I) \to (v_{upper}, I + 1))$ and the $P$ branch ($(v_{lower}, I) \to (v_{upper}, I - 1))$, where
\( v_{\text{lower}} \) is the vibrational quantum number of the initial state, while \( v_{\text{upper}} \) is the vibrational quantum number of the final state. They are related to transition energies through the equations [30]

\[
E^R(I) - E^P(I) = E_{v_{\text{upper}}}(I + 1) - E_{v_{\text{upper}}}(I - 1) = DE_{v_{\text{upper}}}(I),
\]

\[
E^R(I - 1) - E^P(I + 1) = E_{v_{\text{lower}}}(I + 1) - E_{v_{\text{lower}}}(I - 1) = DE_{v_{\text{lower}}}(I),
\]

where in general

\[
DE_{v}(I) = E_v(I + 1) - E_v(I - 1).
\]

\( \Delta I = 2 \) staggering can then be estimated by using Eq. (2), with \( E_{2\gamma}(I) \) replaced by \( DE_{v}(I) \):

\[
\Delta E_{2,v}(I) = \frac{1}{16}(6DE_{2,v}(I)-4DE_{2,v}(I-2)-4DE_{2,v}(I+2)+DE_{2,v}(I-4)+DE_{2,v}(I+4)).
\]

Results for several rotational bands in different electronic and vibrational states of various diatomic molecules are shown in Figs 3–9. We say that \( \Delta I = 2 \) staggering is observed if the quantity \( \Delta E_{2}(I) \) exhibits alternating signs with increasing \( I \) (\( I \) is increased by 2 units each time). The magnitude of \( \Delta E_{2}(I) \) is usually of the order of \( 10^{-3} \)–\( 10^{-5} \) times the size of the interlevel separation energy. Several observations can be made:

1) In all cases shown, the “upper” bands (which happen to be electronically excited) exhibit (Figs 3, 4, 7-9) \( \Delta I = 2 \) staggering which is 2 to 3 orders of magnitude larger than the experimental error, while the corresponding “lower” bands (which, in the cases studied, correspond to the electronic ground state of each molecule), show (Figs 5, 6) some effect smaller than the experimental error.

2) There is no uniform dependence of the \( \Delta I = 2 \) staggering on the angular momentum \( I \). In some cases of long bands, though, it appears that the pattern is a sequence of points exhibiting small staggering, interrupted by groups of 6 points each time showing large staggering. The best examples can be seen in Figs 3a, 3b, 7a, 7b. In Fig. 3a (odd levels of the \( v = 1 \) C1\( \Sigma^+ \) band of YD)) the first group of points showing appreciable \( \Delta I = 2 \) staggering appears at \( I = 13–23 \), while the second group appears at \( I = 27–37 \). In Fig. 3b (even levels of the \( v = 1 \) C1\( \Sigma^+ \) band of YD) the first group appears at \( I = 12–22 \), while the second group at \( I = 26–36 \). In Fig. 7a (odd levels of the \( v = 0 \) A6\( \Sigma^+ \) band of CrD) the first group appears at \( I = 15–25 \), while the second at \( I = 27–37 \). Similarly in Fig. 7b (even levels of the \( v = 0 \) A6\( \Sigma^+ \) band of CrD) the first group appears at \( I = 14–24 \), while the second group at \( I = 26–36 \).

3) In all cases shown, the results obtained for the odd levels of a band are in good agreement with the results obtained for the even levels of the same band. For example, the regions showing appreciable staggering are approximately the same in both cases (compare Fig. 3a with Fig. 3b and Fig. 7a with Fig. 7b, already discussed in 2) ). In addition, the positions of the local staggering maxima in each pair of figures are closely related. In Fig. 3a, for example, maximum staggering appears at \( I = 19 \) and \( I = 31 \), while in Fig. 3b the maxima appear at \( I = 18 \) and \( I = 32 \).

4) In several cases the \( \Delta I = 2 \) staggering of a band can be calculated from two different sets of data. For example, Figs 3a, 3b show the \( \Delta I = 2 \) staggering of the \( v = 1 \) C1\( \Sigma^+ \) band of YD calculated from the data on the 1–1 C1\( \Sigma^+ \)–X1\( \Sigma^+ \) transitions, while Figs 3c,
3d show the staggering of the same band calculated from the data on the 1–2 C\(^1\Sigma^+\)-X\(^1\Sigma^+\) transition. We remark that the results concerning points showing staggering larger than the experimental error come out completely consistently from the two calculations (region with \(I = 13–23\) in Figs 3a, 3c; region with \(I = 12–22\) in Figs 3b, 3d), while the results concerning points exhibiting staggering of the order of the experimental error come out randomly (in Fig. 3a, for example, \(I = 11\) corresponds to a local minimum, while in Fig. 3c it corresponds to a local maximum). Similar results are seen in the pairs of figures (3b, 3d), (4a, 4c), (4b, 4d), (6a, 6c), (6b, 6d), (9a, 9c), (9b, 9d). The best example of disagreement between staggering pictures of the same band calculated from two different sets of data is offered by Figs 6b, 6d, which concern the \(v = 2\) X\(^1\Sigma^+\) band of YD, which shows staggering of the order of the experimental error.

5) When considering levels of the same band, in some cases the odd levels exhibit larger staggering than the even levels, while in other cases the opposite is true. In the \(v = 1\) C\(^1\Sigma^+\) band of YD, for example, the odd levels (shown in Fig. 3a, corroborated by Fig. 3c) show staggering larger than that of the even levels (shown in Fig. 3b, corroborated by Fig. 3d), while in the \(v = 2\) C\(^1\Sigma^+\) band of YD the odd levels (shown in Fig. 4a, corroborated by Fig. 4c) exhibit staggering smaller than that of the even levels (shown in Fig. 4b, corroborated by Fig. 4d).

IV. Discussion

The observations made above can be explained by the assumption that the staggering observed is due to the presence of one or more bandcrossings \[31, 32\]. The following points support this assumption:

1) It is known \[33\] that bandcrossing occurs in cases in which the interband interaction is weak. In such cases only the one or two levels closest to the crossing point are affected \[34\]. However, if one level is influenced by the crossing, in the corresponding staggering figure six points get influenced. For example, if \(E(16)\) is influenced by the crossing, the quantities \(DE_2(15)\) and \(DE_2(17)\) are influenced (see Eq. (10)), so that in the corresponding figure the points \(\Delta E_2(I)\) with \(I = 11, 13, 15, 17, 19, 21\) are influenced, as seen from Eq. (11). This fact explains why points showing appreciable staggering appear in groups of 6 at a time.

2) It is clear that if bandcrossing occurs, large staggering should appear in approximately the same angular momentum regions of both even levels and odd levels. As we have already seen, this is indeed the case.

3) It is clear that when two bands cross each other, maximum staggering will appear at the angular momentum for which the energies of the relevant levels of each band are approximately equal \[34\]. If this angular momentum value happens to be odd, then \(\Delta E_2(I)\) for even values of \(I\) in this region (the group of 6 points centered at this \(I\)) will show larger staggering than the \(\Delta E_2(I)\) for odd values of \(I\) in the corresponding region, and vice versa.

4) The presence of staggering in the “upper” (electronically excited) bands and the lack of staggering in the “lower” (electronic ground state) bands can be attributed to the fact
that the electronically excited bands have several neighbours with which they can interact, while the bands built on the electronic ground state are relatively isolated, and therefore no bandcrossings occur in this case. In the case of the CrD molecule, in particular, it is known \cite{27} that there are many strong Cr atomic lines present, which frequently overlap the relatively weaker (electronically excited) molecular lines. In addition, Ne atomic lines are present \cite{27}. Similarly, in the case of the YD molecule the observed spectra are influenced by Y and Ne atomic lines \cite{27}, while in the case of the CrH molecule there are Ne and Cr atomic lines influencing the molecular spectra \cite{28}.

5) The fact that consistency between results for the same band calculated from two different sets of data is observed only in the cases in which the staggering is much larger than the experimental error, corroborates the bandcrossing explanation. The fact that the results obtained in areas in which the staggering is of the order of the experimental error, or even smaller, appear to be random, points towards the absence of any real effect in these regions.

It should be noticed that bandcrossing has been proposed \cite{35, 36, 37} as a possible explanation for the appearance of $\Delta I = 2$ staggering effects in normally deformed nuclear bands \cite{18, 35, 37} and superdeformed nuclear bands \cite{36}.

The presence of two subsequent bandcrossings can also provide an explanation for the effect of mid-band disappearance of $\Delta I = 2$ staggering observed in superdeformed bands of some Ce isotopes \cite{10}. The effect seen in the Ce isotopes is very similar to the mid-band disappearance of staggering seen, for example, in Fig. 3a.

V. Conclusion

In conclusion, we have found several examples of $\Delta I = 2$ staggering in electronically excited bands of diatomic molecules. The details of the observed effect are in agreement with the assumption that it is due to one or more bandcrossings. In these cases the magnitude of the effect is clearly larger than the experimental error. In cases in which an effect of the order of the experimental error appears, we have shown that this is an artifact of the method used, since different sets of data from the same experiment and for the same molecule lead to different staggering results for the same rotational band. The present work emphasizes the need to ensure in all cases (including staggering candidates in nuclear physics) that the effect is larger than the experimental error and, in order to make assumptions about any new symmetry, that it is not due to a series of bandcrossings.

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Figure captions

Fig. 1 $\Delta E_2(I)$ (in keV), calculated from Eq. (2), versus the angular frequency $\omega$ (in MeV), calculated from Eq. (7), for various superdeformed bands in the nucleus $^{149}$Gd [5]. a) Band (a) of Ref. [5]. b) Band (d) of Ref. [5].

Fig. 2 $\Delta E_2(I)$ (in keV), calculated from Eq. (2), versus the angular frequency $\omega$ (in MeV), calculated from Eq. (7), for various superdeformed bands in the nucleus $^{194}$Hg [6]. a) Band 1 of Ref. [6]. b) Band 2 of Ref. [6].

Fig. 3 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the YD molecule [26]. a) Odd levels of the $v = 1 C^1\Sigma^+$ band calculated from the data of the 1–1 $C^1\Sigma^+–X^1\Sigma^+$ transitions. b) Even levels of the previous band. c) Odd levels of the $v = 1 C^1\Sigma^+$ band calculated from the 1–2 $C^1\Sigma^+–X^1\Sigma^+$ transitions. d) Even levels of the previous band.

Fig. 4 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the YD molecule [26]. a) Odd levels of the $v = 2 C^1\Sigma^+$ band calculated from the data of the 2–2 $C^1\Sigma^+–X^1\Sigma^+$ transitions. b) Even levels of the previous band. c) Odd levels of the $v = 2 C^1\Sigma^+$ band calculated from the 2–3 $C^1\Sigma^+–X^1\Sigma^+$ transitions. d) Even levels of the previous band. The experimental error in all cases is ±0.006 cm$^{-1}$ and therefore is hardly or not seen.

Fig. 5 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the YD molecule [26]. a) Odd levels of the $v = 1 X^1\Sigma^+$ band calculated from the data of the 1–1 $C^1\Sigma^+–X^1\Sigma^+$ transitions. b) Even levels of the previous band.

Fig. 6 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the YD molecule [26]. a) Odd levels of the $v = 2 X^1\Sigma^+$ band calculated from the data of the 1–2 $C^1\Sigma^+–X^1\Sigma^+$ transitions. b) Even levels of the previous band. c) Odd levels of the $v = 2 X^1\Sigma^+$ band calculated from the 2–2 $C^1\Sigma^+–X^1\Sigma^+$ transitions. d) Even levels of the previous band.

Fig. 7 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the CrD molecule [27]. a) Odd levels of the $v = 0 A^6\Sigma^+$ band calculated from the data (R2, P2 branches) of the 0–0 $A^6\Sigma^+–X^6\Sigma^+$ transitions. b) Even levels of the previous band. The experimental error in all cases is ±0.006 cm$^{-1}$ and therefore is not seen.

Fig. 8 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the CrH molecule [28]. a) Odd levels of the $v = 0 A^6\Sigma^+$ band calculated from the data (R2, P2 branches) of the 0–0 $A^6\Sigma^+–X^6\Sigma^+$ transitions. b) Even levels of the previous band. The experimental error in all cases is ±0.004 cm$^{-1}$ and therefore is not seen.

Fig. 9 $\Delta E_2(I)$ (in cm$^{-1}$), calculated from Eq. (11), for various bands of the CoH molecule [29]. a) Odd levels of the $v = 0 A^3\Phi_4$ band calculated from the data (Ree, Pee branches) of the 0–1 $A^3\Phi_4–X^3\Phi_4$ transitions. b) Even levels of the previous band. The experimental error in all cases is ±0.01 cm$^{-1}$ and therefore is not seen.
$^{149}\text{Gd}$

Angular Frequency (MeV)

(a)
\[ \Delta E^2 (\text{keV}) \]

Angular Frequency (MeV)

\[ ^{149}\text{Gd} \]
\[ \Delta E^2 (\text{keV}) \] versus Angular Frequency (MeV) for $^{194}\text{Hg}$.
Angular Momentum $I$
Angular Momentum $I$
$\Delta E_2$ (cm$^{-1}$)

Angular Momentum $J$

(c) YD
\( \Delta E_2 \) (cm\(^{-1}\))

Angular Momentum \( J \)

(a)
Angular Momentum $J$
\[ \Delta E_2 \ (\text{cm}^{-1}) \]

Angular Momentum \( J \)
(a) Angular Momentum $I$
YD

Angular Momentum $I$

(b)
Figure (b) shows the variation of YD with Angular Momentum J. The data points with error bars indicate fluctuations in the YD values across different angular momentum values.
(d) Staggering $\Delta I=2$ Angular Momentum $J$
Angular Momentum $N$

Graph showing $\Delta E_2$ (cm$^{-1}$) vs. Angular Momentum $N$ for CrH.
CoH

ΔE_2 (cm\(^{-1}\))

Angular Momentum J

(a)
