Radiative Width of Molecular-Cluster States

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Molecular states are characterized by enhanced electromagnetic deexcitations of many different multipolarities. The expected enhancement of $E1$, $E2$, and $E3$ transitions is examined by deriving molecular sum rules for radiative deexcitation widths and via a dimensionality approach. The enhancement of the $E1$ transitions is the most striking.

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The single-particle shell model and the collective (quadrupole) degrees of freedom are clearly evident in the low-lying level structure of nuclei. In 1960 it was suggested[1] that high-lying resonance states in ^{12}C + ^{12}C correspond to quadrupole vibration-rotation excitations of dinuclear molecular states in the composite nucleus ^{24}Mg. Recently,[2] it was suggested that "diatomic nuclear molecular" states arise from excitation of a new degree of freedom—a dipole degree of freedom—described by the relative separation vector connecting the centers of the two clusters. The existence of such a collective degree of freedom can be deduced from several observations.

One such observation is the spin and parity of members of an apparent rotational band built on a fixed intrinsic molecular state. When the two nuclear clusters involved are not identical, rotation by 180° around an axis perpendicular to the symmetry axis is not a symmetry operation so that both even and odd spins are allowed in the $K=0$ band. However, reflection in a plane containing the symmetry axis is still a symmetry, leading to a connection between parity and spin.[3] Thus, a $K=0$ molecular band will contain the states 0^+, 1^-, 2^+, 3^-, ... in an alternating parity sequence.

Another observation indicative of a molecular structure is that of enhanced $E1$ transitions (within a rotational band).[2] This is possible only when the ratio of the charges of the two clusters is different from that of their masses; under these conditions the center of charge does not coincide with the center of mass,[4] resulting in a nonvanishing *intrinsic* dipole moment. While $E1$ transitions are expected to be enhanced in such a situation, the $B(E1)$ may still be only a few percent of the single-particle estimate. A central goal of this paper has been to find new measures for such molecular radiative decay widths. To this end we have derived a molecular dipole sum rule to be discussed below.

A third indication of a molecular cluster may be found in the cluster decay width and its relation to the Wigner sum rule. The two sum rules of decay widths can be used simultaneously to yield measures of the probable existence of a molecular structure. For light nuclei, cluster decay widths are readily available as some of the higher-lying states in a proposed band may be observed as scattering resonances.

Recently,[5] a very similar cluster approach has been suggested for heavy ($Z>82$) nuclei. In this case, cluster widths are deduced from the ground-state α-particle decay widths, and from relative hindrance factors for decay to excited states. These have long been known to be small for the low-lying 1^- states in the Ra isotopes.[6]

Recent experimental investigation suggests the existence of an α + ^{14}C molecular band in the light nucleus ^{18}O,[7] as well as an α + ^{214}Rn one in the heavy nucleus ^{218}Ra.[8] In particular, in ^{18}O the proposed cluster band involves a 0^+, 1^-, 2^+, 3^- sequence based on the four-particle, two-hole (4p-2h) 0^+ state at 3.63 MeV. Enhanced $E1$ transitions [$B(E1)\sim 10^{-2}$ Weisskopf unit (W.u.)] and $E2$ crossover transitions [$B(E2)\sim 20$ W.u.] were also found,[7] as shown in Fig. 1.

A first rough estimate of the $E1$ decay rate can be obtained, in the spirit of Gell-Mann and Telegdi,[4] by replacing the nuclear radius in the definition of the Weisskopf unit with the equilibrium displacement of the center of charge from the center of mass. In this way a new "molecular Weisskopf unit" (m.W.u.) for $E1$ transitions is obtained. For example, for α + ^{14}C states in ^{18}O

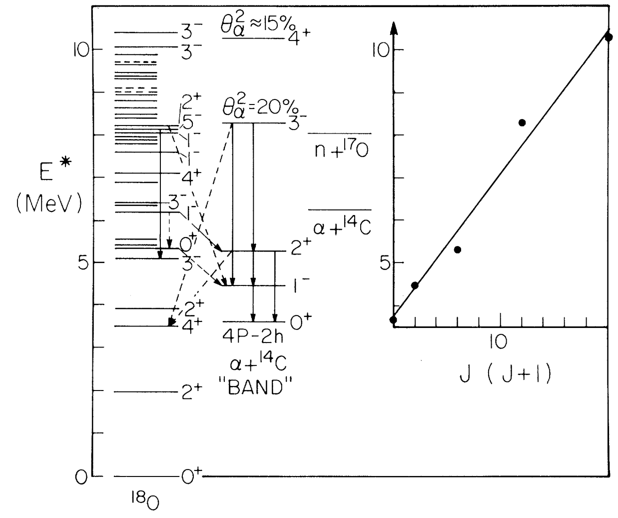

FIG. 1. Natural-parity states in ^{18}O. In solid arrows we show all the enhanced $E1$ ($\sim 10^{-2}$ W.u.) and enhanced $E2$ (~ 20 W.u.) of ^{18}O. They were only recently determined (Ref. 7). Inset: the energy of the 0_2^+, 1^-, 2_3^+, 3_3^-, and 4_3^+ states plotted vs $J(J+1)$.

we find 1 m.W.u. $= 1.79 \times 10^{-3}$ W.u. Thus, $B(E1) > 2 \times 10^{-3}$ W.u. could be considered enhanced on the molecular scale.

While the renormalization of the W.u. to molecular units can be used to measure enhancement of $B(E1)$, it still does not suggest any maximum allowed $B(E1)$ between molecular states. Thus we derive a sum rule for "molecular" $E1$ transitions. The energy-weighted electric dipole sum rule for a nucleus (A, Z) is given by

$$S_1(E1) \equiv \sum_f (E_f - E_i) B(E1; i \to f) = \frac{9}{4\pi} \frac{NZ}{A} \frac{\hbar^2 e^2}{2m}.$$

Here i stands for the fixed initial state and f for the final states (to be summed over). E_i and E_f are the corresponding energies and m the nucleon mass.

If we now only consider transitions in bands based on a "molecular" intrinsic state, excitations of the individual clusters are not allowed. It is, therefore, reasonable to assume that the molecular dipole sum rule (reflecting excitation in the relative motion of the clusters only) is obtained from the total nuclear sum rule by simply subtracting the contributions of the individual clusters—as expressed by their own nuclear sum rules. Explicitly, if the nucleus (A, Z) is decomposed into two clusters (A_1, Z_1) and (A_2, Z_2), then the corresponding energy-weighted "molecular" sum rule is

$$S_1(E1; A_1 + A_2) = S_1(E1; A) - S_1(E1; A_1) - S_1(E1; A_2) = \left(\frac{9}{4\pi}\right) \frac{(Z_1 A_2 - Z_2 A_1)^2}{A A_1 A_2} \left(\frac{\hbar^2 e^2}{2m}\right). \tag{1}$$

As an example, for α-particle clustering $S_1(E_1; \alpha + A_2) \propto (N-Z)^2/A(A-4)$ which includes the correct isospin dependence explicitly. For $\alpha + {}^{14}$C we find the above molecular sum rule to be considerably smaller ($\frac{1}{280}$) than the nuclear sum rule. Thus the measured $B(E1{:}1^- \to 0_2^+) = 2.8 \times 10^{-2}$ W.u. in ^{18}O exhausts a significant fraction (13%) of the molecular sum rule. This $B(E1)$ corresponds to 15.6 m.W.u.

An exact microscopic derivation of the sum rule (1) follows from a decomposition of the nuclear dipole operator $\vec{D}$ [defined as $\vec{D} = e\sum_p (\vec{r}_p - \vec{R})$, where $\vec{r}_p$ and $\vec{R}$ are the vector positions of a proton and center of mass of the nucleus (A, Z), respectively]: $\vec{D} = \vec{D}_1 + \vec{D}_2 + \vec{D}_M$. Here $\vec{D}_i$ $(i = 1, 2)$ is the (nuclear) dipole moment of the ith cluster with respect to its own center of mass: $\vec{D}_i = e\sum_p (\vec{r}_{i,p} - \vec{R}_i)$, and $\vec{D}_M$ is the "molecular" dipole operator,

$$\vec{D}_M = eZ_1(\vec{R}_1 - \vec{R}) + eZ_2(\vec{R}_2 - \vec{R}) = e[(Z_1 A_2 - Z_2 A_1)/A]\vec{S}, \tag{2}$$

which is the dipole moment of (A, Z) when the center of mass coincides with the center of charge in each of the clusters. $\vec{S}$ in (2) is the vector connecting the clusters' centers of mass. If we call $\vec{P}$ the conjugate momentum of $\vec{S}$, the center-of-mass kinetic energy K has a similar decomposition: $K = K_1 + K_2 + \vec{P}^2/2\mu$, where K_i is the internal kinetic energy of (A_i, Z_i) and $P^2/2\mu$ is the kinetic energy in the clusters' relative motion (μ is the reduced mass).

The nuclear sum rule is known to be given by

$$S_1(E1) = \frac{1}{2(2I_i + 1)} \sum_{m_i, m} \langle i, m_i | [D_m, [H, D_m^\dagger]] | i, m_i \rangle,$$

where the total Hamiltonian H can be replaced by the kinetic energy K for velocity- and isospin-independent potentials. We immediately find

$$\langle [D, [K, D^\dagger]] \rangle = \langle [D_1, [K_1, D_1^\dagger]] \rangle + \langle [D_2, [K_2, D_2^\dagger]] \rangle + \langle [D_M, [P^2/2\mu, D_M^\dagger]] \rangle, \tag{3}$$

corresponding to an exact decomposition of the total nuclear sum rule. The first two terms on the right-hand side of Eq. (3) describe $E1$ excitations in each nuclear cluster separately while the last term describes molecular $E1$ excitations. The operator $\vec{\mathrm{D}}_M$ excites modes where the center of charge fluctuates with respect to the center of mass, but at the same time the center of charge in each cluster is fixed with respect to the c.m. of that cluster. Note that Eq. (1) is just (3) when rearranged. The molecular sum rule can thus be calculated as in (1), or directly by evaluating the double commutator of $\vec{\mathrm{D}}_M$ with $P^2/2\mu$, leading to the same exact expression. Clearly the sum rule is *model independent* and no assumption regarding the existence of molecular bands is necessary for its validity.

Approximate molecular sum rules for higher electric multipolarity transitions can be worked out, by taking only the one-body part of the corresponding multipole operator in the center of mass. In such an approximation the nuclear sum rule (for 2^λ-pole) is

$$S_1(E\lambda) = \frac{\hbar^2}{2m}\frac{\lambda(2\lambda+1)^2}{4\pi}\langle \sum e_\alpha^2 r_\alpha^{2\lambda-2}\rangle ,$$

where e_α is the effective charge of a proton or a neutron. A molecular sum rule is then calculated as in (1). For instance, the $E2$ molecular sum rule is

$$S_1(E2, A_1+A_2) = \frac{25}{2\pi}\frac{1}{Z}\left[Z_1Z_2 + \left(Z_1\frac{A_2}{A} - Z_2\frac{A_1}{A}\right)^2\right] S_0^2\,\frac{\hbar^2 e^2}{2m} . \tag{4}$$

The molecular $E2$ sum rule (4) is of the same order of magnitude as the regular isoscalar energy-weighted $E2$ sum rule. Thus we expect enhanced (with respect to the Weisskopf unit) $E2$ crossover transitions in the molecular band, as usually found for collective states in nuclei. The crucial new feature of a molecular band is the enhancement of $E1$ transitions.

From a geometric viewpoint, this enhancement of the higher multipoles can be considered as a reflection of the fact that the $\alpha \otimes (A-4)$ molecular shape, when expanded in spherical harmonics, requires substantial higher-order terms.

A third measure of molecular radiative widths can be obtained from the assumption that the states involved are members of a pure "molecular" rotational band. The radiative transitions can then be easily related to the intrinsic static moments $(q_{\lambda 0})$ of an axially symmetric molecular state, as is the case with ordinary quadrupole bands.[3]

The moments $q_{\lambda 0}$ can be calculated in a simple classical picture if we assume a "molecular clustering" intrinsic state with an equilibrium separation S_0. The $q_{\lambda 0}$ can be expressed in terms of the intrinsic moments of equal or lower multipolarites of the individual clusters and the separation S_0. This can be achieved with the aid of a *new* moment expansion, as is explained below.

Let O and O' be two points along the symmetry axis of a charge distribution, separated by a distance a. The moment $q_{\lambda 0}$ with respect to O can then be expressed in terms of a and the moments $q_{\mu 0}'$ $(\mu \leq \lambda)$ with respect to O' in the following expansion:

$$q_{\lambda 0} = \sum_{\mu=0}^{\lambda} \binom{\lambda}{\mu} q_{\mu 0}' a^{\lambda-\mu} . \tag{5}$$

This expansion can be formally expressed as a binomial: $q_{\lambda 0} = (q_0' + a)^\lambda$, where $q_0'^{\mu}$ is understood to be $q_{\mu 0}'$. Equation (5) is an immediate consequence of similar expansion[9] for the Legendre polynomial

$$r^\lambda P_\lambda(\cos\theta) = \sum_{\mu=0}^{\lambda} \binom{\lambda}{\mu} r'^{\mu} P_\mu(\cos\theta') a^{\lambda-\mu} ,$$

where primed and unprimed quantities refer to O' and O, respectively.

As an example, we have

$$q_{20} = \sum_{i=1}^{2} [q_{20}^{(i)} + 2a_i q_{10}^{(i)} + Z_i a_i^2] ,$$

where $q_{\lambda 0}^{(i)}$ is the 2^λ-pole moment of cluster i, and $a_1 = (A_2/A)S_0$, $a_2 = -(A_1/A)S_0$. It is clear that the two contributions of the clusters (first and third term) are both positive. The cross term, which arises from a polarization of the cluster, is positive too. Thus, again enhanced $E2$ transitions are expected, arising from the increase in q_{20} for the molecular state. This is in contrast to the case of atomic molecules, which comprise both positive and negative charges, so that the molecular polarization gives rise to a smaller quadrupole moment contribution.

In the classical picture S_0 can be estimated from the experimentally known rotational moment of inertia. When the cluster overlap is small, S_0 can be estimated from the cluster size, with use of $r_i = r_0 A_i^{1/3}$ with $r_0 = 1.4$ fm. Therefore, all $B(E\lambda)$ can be estimated in this classical picture.

Thus we calculate for ^{18}O, $B(E1{:}1^- \to 0_2^+) = 0.17$ W.u.; the observed $E1$ is 17% of that expected transition rate calculated for a multipole expansion of a pure molecular state. It also exhausts 13% of the $E1$ molecular sum rule. The suggested $E2$ transition in the molecular band in ^{18}O of $B(E2{:}2_3^+ \to 0_2^+) = 26$ W.u.[7] consists of 23% of the molecular sum rule for $E2$ radiative width. The $J^\pi = 3_3^-$ state at 8.29 MeV in ^{18}O, a member of the proposed[7] $\alpha + ^{14}$C molecular band, has an α width which is 20%[7] of the Wigner sum rule. In the sum-rule analysis of radiative widths we only consider $B(E\lambda{:}0^+ \to \lambda)$; however, as shown in Fig. 1, all five $E1$ and $E2$ in-band transitions are enhanced,[7] as expected, since they are related to the $E\lambda{:}\lambda \to 0^+$ transition. The α width of the 4_3^+ resonant state is also large, as shown in Fig. 1. We consistently find that the radiative sum rules for $E1$ and for $E2$, the Wigner sum rule for α-particle widths, and the classical multipole expansion all suggest that the proposed band in ^{18}O indeed has a large parentage based upon the proposed $\alpha + ^{14}$C dipole molecular band.

For light nuclei extended (many $\hbar\omega$) shell-model microscopic calculations can be done. It would be interesting to determine whether such an enhancement of both the $B(E1)$ and $B(E2)$ together is predicted by such calculation, and if indeed it arises from clustering.[10]

In heavy nuclei such shell-model calculations are beyond present computer capabilities. Thus a phenomenological[5] approach is vital. The experimental investigation of radiative width in the Ra-Th isotopes is now in progress.[8] Branching ratios for $E1/E2$ in ^{218}Ra have been reported,[8] and the $B(E2)$ for ^{218}Ra can be estimated from the systematics of the Ra isotopes. In this way enhanced $E1$ transitions have been found in ^{218}Ra [$B(E1) \sim 10^{-2}$ W.u.].

It thus appears that the dipole molecular collective degree of freedom plays a role in the structure of both light and heavy nuclei. The radiative-width sum rule, derived here, provides a measure of the extent to which molecular states are present; together with the cluster-decay-width sum rule (the Wigner limit), it can provide a crucial test for the existence of nuclear molecular states.

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[1] D. A. Bromley, J. A. Kuehner, and E. Almquist, Phys. Rev. Lett. 4, 365 (1960).

[2] F. Iachello, Phys. Rev. C 23, 2778 (1981).

[3] A. Bohr and B. R. Mottelson, *Nuclear Structure* (Benjamin, New York, 1975), Vol. 2.

[4] M. Gell-Mann and V. L. Telegdi, Phys. Rev. 91, 169 (1953).

[5] F. Iachello and A. D. Jackson, Phys. Lett. 108B, 151 (1982).

[6] F. S. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. 100, 1543 (1955).

[7] M. Gai, M. Ruscev, A. C. Hayes, J. F. Ennis, R. Keddy, E. C. Schloemer, S. M. Sterbenz, and D. A. Bromley, Yale Report No. Yale-3074-719 (to be published).

[8] M. Gai, J. F. Ennis, A. C. Hayes, M. Ruscev, E. C. Schloemer, B. Shivakumar, S. M. Sterbenz, N. Tsoupas, and D. A. Bromley, in *Proceedings of the International Conference on Nuclear Structure, Amsterdam, 1982,* edited by A. Von Derwordt and B. J. Verhaar (European Physical Society, Petit-Lancy, Switzerland, 1982), Vol. I, p. 138.

[9] To prove it, note that the left-hand side is an harmonic function and therefore can be expanded in a complete set of harmonic functions around O': $r'^{\mu} Y_{\mu m}(\Omega')$, $\mu \leq \lambda$, and because of axial symmetry only $m = 0$ are allowed. The expansion coefficients are then determined by taking a point along the symmetry axis where $\theta = \theta' = 0$, $r = r' + a$ and using $P_\mu(1) = 1$.

[10] D. J. Millener, private communication.

[11] M. Danos and L. C. Maximon, J. Math. Phys. (N.Y.) 6, 766 (1965).