Mössbauer sum rules for use with synchrotron sources

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The availability of tunable synchrotron radiation sources with millivolt resolution has opened prospects for exploring dynamics of complex systems with Mössbauer spectroscopy. Early Mössbauer treatments and moment sum rules are extended to treat inelastic excitations measured in synchrotron experiments, with emphasis on the unique conditions absent in neutron scattering and arising in resonance scattering: prompt absorption, delayed emission, recoilfree transitions, and coherent forward scattering. The first moment sum rule normalizes the inelastic spectrum. Sum rules obtained for higher moments include the third moment proportional to the second derivative of the potential acting on the Mössbauer nucleus and independent of temperature in the harmonic approximation. Interesting information may be obtained on the behavior of the potential acting on this nucleus in samples not easily investigated with neutron scattering, e.g., small samples, thin films, time-dependent structures, and amorphous-metallic high pressure phases.

I. INTRODUCTION

The recent development of tunable synchrotron radiation sources using the Mössbauer effect provide tools for investigating properties of complex condensed matter systems. The use of these tunable sources for studying different types of elastic and inelastic transitions in crystals[1,2] has opened a field of millivolt spectroscopy for investigation of the dynamics of complex systems which are not accessible to other techniques like neutron scattering. One example is the extension to millivolt spectroscopy for systems previously studied by the Mössbauer technique for nanovolt spectroscopy; e.g., amorphous-metallic high-pressure phases.[3]

For ^{57}Fe at 14.413 keV it is possible to prepare a monochromatic beam with 6 MeV bandpass, tunable over a few hundred eV.[4–7] Furthermore it may be possible to improve the energy resolution to 2 MeV levels using different sets of crystals at an undulator based beamline.[5,8]

The synchrotron source offers possibilities beyond the conventional Mössbauer spectroscopy[9] by exciting nuclear resonances with an incident pulse much shorter than the natural lifetime of the resonance. The disappearance of the incident pulse and all prompt background scattered radiation before the detection of the signal leads to both an enormous background suppression and the possibility of observing forward scattered radiation completely separated from the incident beam, not possible with other techniques. Prompt Rayleigh scattering and scattering by other nonresonant atoms give no background. Forward scattering from many different nuclei is coherent by analogy with Bragg scattering, but is essentially independent of the structure of the sample. This allows the study of coherent radiation from many nuclei in a sample, with the interesting time behavior of speedup and quantum beats,[9] simply by looking at forward radiation without the need to choose Bragg angles, and without even the need for an ordered structure.

In addition there is the well established difference of the Mössbauer technique from other techniques; e.g., neutron scattering, by being sensitive to a particular nucleus like ^{57}Fe in a sample, and having a high cross section. It therefore gives information on the forces and possible localized vibration modes and local forces in the vicinity of the iron or other Mössbauer nuclei in complicated and small samples, and allows investigation of this information as a function of changes in the composition or structure of the system and behavior near phase transitions.

Many of the effects observable in Mössbauer experiments with synchrotron radiation have been discussed in detail by Hannon and Trammell.[9] In this paper we focus on one point not discussed in this excellent review, the information on inelastic excitations produced by hitting nuclei at specific positions in the lattice with a synchrotron pulse. We shall see that sum rules originally derived for moments of the energy spectrum of gamma rays emitted from Mössbauer transitions[10] have now acquired a significance in data analysis from synchrotron radiation and have already been used in recent experiments.[1,2]

Moment sum rules have been applied in many other areas of physics where a sudden momentum transfer occurs on an effectively pointlike constituent in a bound system, from x-ray and neutron scattering[11–13] to lepton-pair emission by heavy quarks bound in hadrons.[14] The general formulation is essentially the same for all processes but the applications to data analyses for individual processes can be very different. Mössbauer scattering of photons from a synchrotron source has the unique features of being a two-step process with prompt absorption and delayed reemission and with frequently occurring recoil-free transitions including coherent forward scattering from different nuclei. These features, which are completely absent in neutron scattering play a crucial role in data analysis. Our purpose here is to use moment sum rules to take them into account and obtain meaningful results for experimental data.

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A central problem arising in understanding the spectrum of inelastic excitations produced by a synchrotron pulse on a sample is the separation of the elastic and inelastic cross sections; i.e., the determination of the Debye-Waller or Mössbauer f factor. Samples used in these experiments must be sufficiently thick so that there is appreciable inelastic absorption from the radiation off resonance. With such thick crystals the radiation at the resonance which is absorbed and scattered elastically is both enhanced by coherent scattering from different nuclei and attenuated by absorption in passing through the sample. The coherent elastic scattering is concentrated into a sharp forward peak to give a very different angular distribution from that of the inelastic scattered radiation. The relative normalization of the elastic and inelastic cross sections and the value of the Lamb-Mössbauer f factor are not obtainable from the data alone. The sum rules provide both a method of separating and normalizing the elastic and inelastic data without detailed analysis[2] and of obtaining localized information about forces on and motion of the Mössbauer nucleus.

II. GENERAL FEATURES OF ELASTIC AND INELASTIC TRANSITIONS

We first summarize some general features of the different types of transitions that occur in the excitation of nuclear resonance levels in a crystal by a pulse of synchrotron radiation which is much shorter than the lifetime of the nuclear state. The nuclei in the crystal will be in a complicated state of excitation after the pulse is over, and the subsequent radiation will be a mixture of several different types of transitions. The absorption and emission processes can both be either elastic or inelastic. In elastic processes a photon is absorbed or emitted by the internal degrees of freedom of the nucleus with no change in the other degrees of freedom of the system. Inelastic processes involve energy transfer to the other degrees of freedom. In both absorption and emission inelastic momentum and energy transfer can occur to the lattice degrees of freedom via nuclear recoil. In emission there is also the possibility of internal conversion with the emission of an electron and subsequent x rays rather than a gamma ray.

The synchrotron radiation pulse will generally have a broad enough energy spectrum to excite both the elastic and inelastic transitions. There will also be tunable sources within this spectrum to enable separation of different types of elastic and inelastic transitions. The elastic excitation can be coherent over many nuclei in the crystal with a subsequent speedup in the decay lifetime. We can thus expect to observe two lifetimes in the detected radiation, the normal lifetime for decays of nuclei produced by inelastic excitation and a speeded-up lifetime produced by the decay of the coherent or superradiant state.

The emitted radiation would therefore consist of the following components:

(1) Purely inelastic transitions giving photons with the inelastic spectrum and also conversion electrons and x rays, with the decay lifetime and angular distributions of single nuclear excitations.

(2) Purely elastic transitions giving a coherent spectrum with the speeded up lifetime and a broadened natural linewidth produced by the speedup and a sharply peaked angular distribution in the forward and/or Bragg direction.

(3) Inelastic excitation and elastic emission. This will give a photon spectrum with the natural linewidth and the natural lifetime and the angular distribution of single nuclear excitations.

(4) Elastic excitation and inelastic emission. This will give the inelastic spectrum and also conversion electrons and x rays, with the angular distributions of single nuclear excitations, but with the speeded-up lifetime.

For general orientation we note the very different energy scales arising in synchrotron Mössbauer physics. The natural linewidths of nuclear transitions and nuclear hyperfine and quadrupole splittings are in the nanoelectronvolt range. This nanovolt spectroscopy is studied with tunable Doppler-shifted Mössbauer lines from radiative sources and by observing time-dependent quantum beats following an excitation from photons generated by a synchrotron pulse. In a completely different domain are the typical lattice energies; e.g., characteristic temperatures like Debye temperatures, which are in the range of tens of millivolts. In this range one also finds the free recoil energy which characterizes the inelastic spectrum of nuclear transitions as well as room temperature thermal energies. The inelastic spectrum in this range has not been experimentally explored in detail. The synchrotron radiation pulse will generally have a broad enough energy spectrum to excite both the elastic and inelastic transitions.

III. BASIC THEORY OF EXCITATION BY SYNCHROTRON RADIATION

All the physics needed to understand the Mössbauer effect had been published long before Mössbauer's discovery.[12,11,13] That photons could be scattered by atoms in a crystal without energy loss due to recoil was basic to all work in x-ray diffraction and crystallography. All the quantitative calculations including the definition and evaluation of the Debye-Waller factor were well known but not interpreted as a probability that a photon could be scattered by an atom in a crystal without energy loss due to recoil. In the wave picture of radiation the Debye-Waller factor written as $\exp(\langle -k^2x^2\rangle)$ described the loss of intensity of coherent radiation because the atoms were not fixed at their equilibrium positions and their motion introduced random phases into the scattered wave.

The relation between Lamb's treatment[11] of neutron capture in crystals and Ott's x-ray treatment[12] was first pointed out by Kaufman[15,16] and reported in detail in a history of these developments.[17] A general formulation including these and other processes of momentum transfer to bound systems is given in the quantum mechanics book[10] which shows the relation of the dual wave-particle descriptions of similar phenomena.

The first article to use the name Mössbauer effect[18] appeared at a time when the physics community either did not believe in the effect or felt that it was not important enough to be called by its discoverer's name. At that time a number of sum rules[19] were derived along with other results[20,21] which remain pedagogically useful today for teaching basic principles of quantum mechanics to graduate students.[10] The general state of confusion on this issue can be seen in the panel discussion which took place at the Second International Mössbauer conference.[22] Further applications of the basic theory[12,11,13] for the Mössbauer effect and for neutron scattering are reported in Ref. 2.

We first consider the application of this basic theory to the case of resonance excitation of a single bound nucleus by a broad beam x-ray source. The cross section for this excitation as a function of the incident gamma ray energy will contain a peak at the resonance energy corresponding to the elastic or no-recoil Mössbauer transition, and a spectrum on both sides of the resonance energy corresponding to inelastic transitions in which the state of the lattice is changed. Consider a transition for photon absorption between some initial lattice state denoted by $|i\rangle$ and a final state denoted by $|f\rangle$. We denote the cross section for this transition as $\sigma_{i\to f}(E)$ and note that its integral over the entire relevant energy interval can be written

$$\int \sigma_{i\to f}(E)dE = \bar{\sigma}|\langle f|e^{i\vec{k}\cdot\vec{r}_\mu}|i\rangle|^2 , \tag{1}$$

where $\vec{k}$ denotes the photon wave number, $\vec{r}_\mu$ is the coordinate in the lattice of the nucleus being excited, and $\bar{\sigma}$ is normalized to give the total integrated cross section over all final states,

$$\bar{\sigma} = \sum_f \int \sigma_{i\to f}(E)dE . \tag{2}$$

In the normal Mössbauer effect, the probability that the transition takes place without any change in the state of the lattice is given exactly by the Debye-Waller factor. In excitation by synchrotron radiation the same Debye-Waller or Mössbauer fraction factor appears in the cross section for the elastic transition in which the lattice remains in its initial state,

$$\int \sigma_{i\to i}(E)dE = \bar{\sigma}|\langle i|e^{i\vec{k}\cdot\vec{r}_\mu}|i\rangle|^2 . \tag{3}$$

IV. SUM RULES FOR MOMENTS OF THE INELASTIC SPECTRUM

Interesting properties of the inelastic transitions are obtainable by generalizing sum rules originally developed for Mössbauer emission[19] to apply to the moments of the excitation energy spectrum,

$$\langle (E_f-E_i-R)^n\rangle \equiv \sum_f \frac{1}{\bar{\sigma}}\int dE\,(E_f-E_i-R)^n\sigma_{i\to f}(E) = \sum_f (E_f-E_i-R)^n|\langle f|e^{i\vec{k}\cdot\vec{r}_\mu}|i\rangle|^2 , \tag{4}$$

where the moments are defined relative to the centroid of the spectrum, denoted by R, which is known[19] to be just the free recoil energy for a nucleus of mass M,

$$R = \frac{(\hbar k)^2}{2M} . \tag{5}$$

The moments can be rewritten by using closure,

$$\langle (E_f-E_i-R)^n\rangle = \langle i|e^{i\vec{k}\cdot\vec{r}_\mu}(H-E_i-R)^n e^{-i\vec{k}\cdot\vec{r}_\mu}|i\rangle , \tag{6}$$

where H is the Hamiltonian describing the lattice dynamics,

$$H = \sum_{\mu=1}^{N}\frac{\vec{p}_\mu^{\,2}}{2M_\mu} + \sum_{\mu,\nu=1}^{N} V_{\mu\nu}(\vec{x}_\mu,\vec{x}_\nu) . \tag{7}$$

N is the number of atoms in the lattice, M_μ is the mass of the atom which may be different from the mass M of the Mössbauer nucleus for other atoms in the lattice, and $V_{\mu\nu}(\vec{x}_\mu,\vec{x}_\nu)$ is some interaction potential depending only upon the coordinates $(\vec{x}_\mu,\vec{x}_\nu)$ of the atoms and not on their momenta.

Substituting the Hamiltonian (7) into the expression (6) gives

$$\langle (E_f-E_i-R)^n\rangle = \langle i|\left[H-E_i-\frac{\hbar\vec{k}\cdot\vec{p}_\mu}{M}\right]^n|i\rangle \tag{8a}$$

and we note that

$$\langle i|(H-E_i) = (H-E_i)|i\rangle = 0 . \tag{8b}$$

Then

$$\langle (E_f-E_i-R)^n\rangle = -\langle i|\left[\frac{2Rp_{z\mu}}{\hbar k}\right]\left[H-E_i-\frac{\hbar k p_{z\mu}}{M}\right]^{(n-1)}|i\rangle , \tag{9a}$$

$$\langle (E_f-E_i-R)^n\rangle = \left[\frac{2R}{M}\right]\langle i|p_{z\mu}\left[H-E_i-\frac{\hbar k p_{z\mu}}{M}\right]^{(n-2)}p_{z\mu}|i\rangle , \tag{9b}$$

where we have chosen our z axis in the direction of the photon momentum $\vec{k}$ so that

$$\left[\frac{\hbar\vec{k}\cdot\vec{p}_\mu}{M}\right] = \left[\frac{\hbar k p_{z\mu}}{m}\right] = \left[\frac{2Rp_{z\mu}}{\hbar k}\right] . \tag{9c}$$

The well-known results for the first and second moments are immediately obtained:

$$\langle (E_f-E_i-R)\rangle = 0 , \tag{10a}$$

$$\langle (E_f-E_i-R)^2\rangle = \left[\frac{2R}{M}\right]\langle i|p_{z\mu}^2|i\rangle = 4R\bar{T}_{z\mu} , \tag{10b}$$

where

$$\bar{T}_{z\mu}\equiv\frac{\langle i|p_{z\mu}^2|i\rangle}{2M} \tag{11a}$$

is the mean kinetic energy in the z direction for nucleus μ. We have assumed that there is no correlation between the directions of $\vec{k}$ and $\vec{p}_\mu$, and therefore that the expectation values of all odd powers of $p_{z\mu}$ must vanish,

$$\langle i|(p_{z\mu})^{(2r+1)}|i\rangle=0\ . \tag{11b}$$

This is generally true in all cases of interest, since it follows from time reversal invariance of the interactions, and holds automatically for a harmonic interaction.

These first two moments depend only on the free recoil energy R and the mean kinetic energy $\bar{T}_{z\mu}$ and are equal to the classical expressions for these two moments for the case of nuclear excitation in a noninteracting gas. The first moment is completely independent of the dynamics of the system and the temperature, while the second moment is proportional to the mean kinetic energy and is thus a monotonically increasing function of the temperature.

V. SUM RULES FOR THE THIRD AND FOURTH MOMENTS

Higher moments were not previously considered, since they did not seem to be relevant to feasible experimental tests at the time. Their evaluation also appeared to be more complicated, since the Hamiltonian H appears explicitly in Eq. (9b) sandwiched between factors $p_{z\mu}$ with which H does not commute. In contrast to the first two moments which are equal to the values obtained simply from classical billiard-ball kinematics for a noninteracting gas, the higher moments depend upon properties of the dynamics, i.e., upon the values of parameters in the Hamiltonian (7) and introduce effects of quantum mechanics, expressed by the explicit appearance of commutators proportional to $\hbar$. We shall see that they provide interesting information on properties of the lattice.

The relevant commutators needed to evaluate the higher moments are

$$[p_{z\mu},H]=-i\hbar\frac{\partial}{\partial z_\mu}\sum_{\rho,\nu=1}^{N}V_{\rho\nu}\ , \tag{12a}$$

$$[p_{z\mu},[p_{z\mu},H]]=-\hbar^2\frac{\partial^2}{\partial z_\mu^2}\sum_{\rho,\nu=1}^{N}V_{\rho\nu}\ . \tag{12b}$$

For the case of a harmonic crystal, the potential energy is a polynomial of second order in all coordinates, which can be written

$$\sum_{\rho,\nu=1}^{N}V_{\rho\nu}(\vec{x}_\rho,\vec{x}_\nu)=V_{\mu\mu}^{zz}(z_\mu)^2+\cdots\ , \tag{13a}$$

where all the terms beyond the first do not contribute to the double commutator (12b). The potential energy can also be expressed in terms of the coordinates ξ_j and the frequencies ω_j of the normal modes,

$$\sum_{\mu,\nu=1}^{N}V_{\mu\nu}=\sum_{j=1}^{3N}\frac{1}{2}M\omega_j^2\xi_j^2\ . \tag{13b}$$

From the orthonormality of the linear transformation between nucleus coordinates z_μ and the normal coordinates ξ_j we obtain the useful relations

$$\sum_{j=1}^{3N}\left[\frac{\partial\xi_j}{\partial z_\mu}\right]^2=1\ , \tag{14a}$$

$$\sum_{\mu=1}^{N}\left[\frac{\partial\xi_j}{\partial x_\mu}\right]^2+\left[\frac{\partial\xi_j}{\partial y_\mu}\right]^2+\left[\frac{\partial\xi_j}{\partial z_\mu}\right]^2=1\ . \tag{14b}$$

For a harmonic crystal the commutators (12) become

$$[p_{z\mu},H]=-i\hbar\sum_{j=1}^{3N}M\omega_j^2\xi_j\frac{\partial\xi_j}{\partial z_\mu}\ , \tag{15}$$

$$[p_{z\mu},[p_{z\mu},H]]=-\hbar^2\sum_{j=1}^{3N}M\omega_j^2\left[\frac{\partial\xi_j}{\partial z_\mu}\right]^2\ , \tag{16a}$$

$$[p_{z\mu},[p_{z\mu},H]]=-\hbar^2\frac{\partial^2}{\partial z_\mu^2}\sum_{\rho,\nu=1}^{N}V_{\rho\nu}=-2\hbar^2V_{\mu\mu}^{zz}\ . \tag{16b}$$

Note that the double commutator (16) depends only upon the force on the coordinate z_μ which is expressed in terms of normal mode variables as a function of the frequencies ω_j and the expansion coefficients of the coordinate z_μ in the normal coordinates ξ_j. The value of the double commutator for a harmonic lattice thus depends only on the parameters of the Hamiltonian and is completely independent of the state of the lattice and of the temperature. This feature is particularly interesting because the third moment of the energy spectrum can be seen to be proportional just to this double commutator,

$$\begin{aligned}\langle(E_f-E_i-R)^3\rangle&=\left[\frac{R}{M}\right]\langle i|[p_{z\mu},H]p_{z\mu}+p_{z\mu}[H,p_{z\mu}]|i\rangle\\&=-\left[\frac{R}{M}\right]\langle i|[p_{z\mu},[p_{z\mu},H]]\ .\end{aligned} \tag{17a}$$

Thus

$$\langle(E_f-E_i-R)^3\rangle=\left[\frac{R}{M}\right]\left\langle i\left|\hbar^2\frac{\partial^2}{\partial z_\mu^2}\sum_{\rho,\nu=1}^{N}V_{\rho\nu}\right|i\right\rangle\ . \tag{17b}$$

For a harmonic lattice this becomes

$$\langle(E_f-E_i-R)^3\rangle=\hbar^2R\sum_{j=1}^{3N}\omega_j^2\left[\frac{\partial\xi_j}{\partial z_\mu}\right]^2=R\hbar^2\overline{\omega_z^2}\ , \tag{18a}$$

where

$$\overline{\omega_z^2}\equiv\sum_{j=1}^{3N}\omega_j^2\left[\frac{\partial\xi_j}{\partial z_\mu}\right]^2 \tag{18b}$$

is a weighted mean square average lattice frequency, and the subscript z denotes that it is determined by normal modes with motion in the z direction. The weighing factors are seen from the normalization relation (14a) to be normalized to unity. For a harmonic crystal the third moment can also be expressed in terms of the force on the coordinate z_μ

$$\langle (E_f - E_i - R)^3 \rangle = \left[\frac{2\hbar^2 R}{M} \right] V^{zz}_{\mu\mu} \ . \tag{19}$$

This result is independent of the lattice wave function $|i\rangle$ and therefore also of the temperature. For an isotropic crystal this result is a simple function of the characteristic temperature of the lattice; e.g., the Debye or Einstein temperature. For an anisotropic crystal, the result will depend upon the angles between the crystal axes and the photon direction, and can give information about the parameters of the anisotropic lattice. If the crystal is not harmonic, the expression (17b) depends upon the coordinates of various atoms in the lattice and therefore on the temperature.

The fourth moment can now be evaluated using the same commutators. We immediately discard expectation values of all odd powers of $p_{z\mu}$ and obtain

$$\begin{aligned} \langle (E_f - E_i - R)^4 \rangle &= \left[\frac{2R}{M} \right] \langle i | \left[\frac{2R}{M} \right] p^4_{z\mu} + [p_{z\mu}, H](H - E_i) p_{z\mu} | i \rangle \\ &= \left[\frac{2R}{M} \right] \langle i | \left[\frac{2R}{M} \right] p^4_{z\mu} - [p_{z\mu}, H]^2 | i \rangle \\ &= \left[\frac{2R}{M} \right] \langle i | \left[\frac{2R}{M} \right] p^4_{z\mu} + \hbar^2 \left[\frac{\partial}{\partial z_\mu} \sum_{\rho,\nu=1}^{N} V_{\rho\nu} \right]^2 | i \rangle \ . \end{aligned} \tag{20a}$$

For a harmonic lattice this becomes

$$\langle (E_f - E_i - R)^4 \rangle = \left[\frac{2R}{M} \right] \langle i | \left[\frac{2R}{M} \right] p^4_{z\mu} + \hbar^2 \sum_{j=1}^{3N} M^2 \omega_j^4 \xi_j^2 \left[\frac{\partial \xi_j}{\partial z_\mu} \right]^2 | i \rangle \ . \tag{20b}$$

VI. USE OF SUM RULES FOR THICK SAMPLES WITH MANY NUCLEI

These results are easily generalized to the case where thee are many nuclei in the lattice which can be excited by the Mössbauer transition, but the intensity of the incident beam is sufficiently weak so that only one photon is absorbed. Since the single excitation cross sections for inelastic transitions by individual nuclei are independent, the total rate for inelastic transitions in an experiment is obtained in the usual manner by summing over all nuclei in the target.

The elastic transitions require special attention. Coherent effects[23] like superradiance can enhance the transition matrix element for photon emission.[24] The coherence changes the angular distribution of the radiation and the enhancement produces a corresponding speedup in the lifetime of the excited state. These effects were originally predicted by Trammel,[24] further developed[25] theoretically and observed in very beautiful experiments.[26,27] An excellent review of these developments has been given in Ref. 9, where the particular coherent state is called a "nuclear exciton."

The speedup of the elastic transition also broadens the linewidth of the nuclear exciton and therefore affects the integrated total cross section (2). However, this coherence does not affect the inelastic excitation spectrum in a lattice by synchrotron radiation. The moments can still be obtained from by the above analysis and Eqs. (17)–(20) but corrections are necessary to the normalization procedure, both because of the enhanced elastic contribution and because radiation in the resonance peak can be attenuated in passing through a sample which is still sufficiently thin to leave radiation off resonance unaffected. Since these effects at resonance are all at $(E_f - E_i) = (E_f - E_i)^n = 0$, we can write

$$\langle (E_f - E_i)^n \rangle_{\text{ex}} = \frac{\langle (E_f - E_i)^n \rangle_1}{K_{\text{en}}} \ , \tag{21}$$

where $\langle (E_f - E_i)^n \rangle_{\text{ex}}$ denotes the experimental moment, defined relative to $(E_f - E_i) = 0$, observed when coherence effects may be present, $\langle (E_f - E_i)^n \rangle_1$ denotes the moment for the case of a single nucleus, and K_{en} denotes a normalization correction factor which is the same for all moments.

The normalization factor K_{en} can be determined experimentally from the first moment and then applied to expressions for the higher moments. Substituting the result for the first moment (10a) we obtain

$$\langle (E_f - E_i) \rangle_{\text{ex}} = \frac{\langle (E_f - E_i) \rangle_1}{K_{\text{en}}} = \frac{R}{K_{\text{en}}} \ . \tag{22a}$$

Thus

$$\langle (E_f - E_i)^n \rangle_{\text{ex}} = \langle (E_f - E_i) \rangle_1 \frac{\langle (E_f - E_i) \rangle_{\text{ex}}}{R} \ . \tag{22b}$$

These moments can now be measured by using tunable sources with an energy bandwidth small compared with the free recoil energy R. The moments can be normalized either by integrating over the entire spectrum, or by integrating only over the inelastic spectrum and removing the elastic peak. The normalization factor K_{en} will be different in the two cases, but the result (22b) applies to both cases.

The value of the factor K_{en} can be calculated theoretically for samples sufficiently thin so that there is no at-

tenuation of the resonance radiation in passing through the sample, from the values of the speedup factor denoted by F^0_{speed} for the superradiant component or nuclear exciton and the Debye-Waller or Mössbauer fraction factor commonly denoted by f. Note, however, that in experiments where the nuclear excitation is detected by the decay of the excitation into a particular decay channel the value of K_{en} can depend upon the branching ratio for the decay into the observed channel.

This is particularly important in the case where the detector sees only the inelastic channels like conversion electrons or x rays. The speedup factor changes the relative branching ratios for decays into different channels, since the speedup applies only to the Mössbauer fraction of the radiation which is proportional to the Debye-Waller or Mössbauer fraction factor f. The total decay rate includes also the incoherent processes of inelastic gamma ray emission, proportional to $1-f$, and internal conversion which is described by the internal conversion coefficient α. The partial widths for incoherent gamma emission and internal conversion are not enhanced by superradiance and are the same as for a single excited nucleus. Thus the total decay width Γ_{tot} is

$$\Gamma_{\text{tot}}=[F^0_{\text{speed}}f+(1-f)+\alpha]\Gamma_\gamma\ , \tag{23}$$

where F^0_{speed} denotes the speedup factor for the superradiant component and Γ_γ denotes the partial width for gamma decay by a single nucleus. The relative probabilities or branching ratios for incoherent gamma emission and internal conversion are correspondingly reduced,

$$\frac{\Gamma_{\text{incoherent}}}{\Gamma_{\text{tot}}}=\frac{1-f}{F^0_{\text{speed}}f+(1-f)+\alpha}\ , \tag{24a}$$

$$\frac{\Gamma_{\text{intconv}}}{\Gamma_{\text{tot}}}=\frac{\alpha}{F^0_{\text{speed}}f+(1-f)+\alpha}\ . \tag{24b}$$

The net speedup factor for the decay rate relative to that of a single nucleus is

$$F_{\text{speed}}=\frac{F^0_{\text{speed}}f+(1-f)+\alpha}{1+\alpha}\ . \tag{25}$$

The decay rate for the elastically excited coherent state is the sum of the normal inelastic decay rate and the enhanced elastic decay rate. In this case the loss due to internal conversion is reduced, because the probability that a given excited nucleus will emit a photon rather than ejecting an electron has been increased.[24,26,27]

Thus in experiments where the detector sees only the inelastic channels like conversion electrons or x rays, the branching ratios for these detection modes are seen from Eqs. (24a) and (24b) to be reduced relative to that of the speeded-up coherent forward radiation. This bias must be taken into account in calculating the factor K_{en}.

Since the coherent effects do not change the inelastic excitations, the normalization factor K_{en} should be the same for the coherent case as for a single nucleus if the experimental normalization for the moments is calculated only by integrating over the inelastic spectrum. For this case the integral is just the total inelastic probability for the single nucleus case; namely $1-f$. Thus we obtain

$$K_{\text{en}}^{\text{inelastic}}=\frac{1}{1-f}\ . \tag{26}$$

VII. CONCLUSIONS

The development of tunable synchrotron radiation sources in the millivolt range provides the possibility of experimental tests and applications for moment sum rules originally derived for the emission of Mössbauer resonance radiation and hitherto used primarily for pedagogical purposes. These sum rules are shown to be very useful in obtaining crucial information from inelastic resonance scattering data.

The first moment sum rule enables the normalization of the inelastic scattering data and the determination of the Lamb-Mössbauer f factor. This moment is equal to the free recoil energy of the resonant nuclear transition and is independent of the structure of the bound system, its wave function, and the temperature.

The second moment is proportional to the mean kinetic energy of the resonant nucleus and is a function of the temperature and the wave function describing the motion of the nucleus in the bound system.

The third is proportional to the second derivative of the potential acting on the resonant nucleus. This is the force constant seen by this nucleus if the forces are harmonic. For a harmonic system this moment depends only upon the force constants in the Hamiltonian of the system and is independent of the wave function of the system or the Hamiltonian.

The fourth moment is the sum of two terms. One term is proportional to the mean value of the fourth power of the resonant nucleus momentum; the second term is a function of the constants appearing in the system Hamiltonian and of the wave function describing the motion of the resonant nucleus in the bound state. The theoretical value of this moment is easily calculated for any particular model.

These sum rules can be particularly useful for Mössbauer synchrotron experiments performed in cases where inelastic excitations are not accessible to other experimental techniques like neutron scattering. The advantages of the synchrotron Mössbauer technique are discussed in detail in Ref. 2, and include the ability to use small samples and thin films, the speed of the measurement with the possibility of investigating short-lived structures and the time development of phase transitions, and the ability to focus on a comparatively rare constituent in a sample.

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