Quantum beats from nuclei excited by synchrotron pulses

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Synchrotron pulses will excite low-lying nuclear levels whose subsequent decay will exhibit beats with frequencies equal to the nuclear hyperfine splittings. The highly collimated pulses incident on very small enriched single crystals at a Bragg angle will result in an appreciable fraction of the incident radiation in the spectral width of the nuclear resonance being coherently scattered into a highly collimated beam which will exhibit beats corresponding to the difference frequencies emitted by different nuclei, and a decay parameter Γ, with marked time dependence.
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I. INTRODUCTION

Ruby[1] first pointed out that the synchrotron radiation from the storage rings SPEAR and DORIS should be useful for exciting low-lying nuclear levels. To be specific, SPEAR operating at 2.5 GeV and a current of 100 mA produces a flux of about 10^{12} photons/eV sec mrad at 14.4-keV photon energy.[2] Then by means of a bent-crystal monochromator one should be able to get a photon flux on a 1×1 mm target placed ~ 10 m from a tangent to the orbit of electrons of about 10^{12} photons/sec mm^2 at this energy with a bandwidth ~ 1 eV and a collimation $\approx 10^{-4}$ (vertical) $\times 10^{-2}$ (horizontal) sr. The width of the 14.4-keV ^{57}Fe level is $\Gamma \approx 10^{-8}$ eV; therefore, in the synchrotron radiation falling on the target there would be $\sim 10^4$ photons/sec mm^2, which could excite this level. A natural 100% ^{57}Co Mössbauer source gives $\sim 10^{10}$ Mössbauer (γ rays)/sec mm^2, and for the usual Mössbauer-absorption experiments where only a moderate collimation is necessary (say 0.1 sr) the natural source would deliver a flux of about 10^8/mm^2 sec on a target as compared with $\sim 10^4$/mm^2 sec from the storage ring. However, while the brightness of the natural source is $\sim 10^9$ photons/mm^2 sec sr (isotropic) that of the synchrotron radiation is $\sim 10^{10}$/mm^2 sec sr (in the very narrow 10^{-6}-sr collimation), and for experiments involving coherent scattering and transmission through perfect crystals, and interferometry experiments, where high collimation is necessary, the synchrotron source will be superior to the best possible natural source. In addition with wigglers it has been estimated that a $10^3 \times$ increase of the synchrotron flux is attainable[3] and this would make it competitive with the natural source even for low collimation experiments. Furthermore, the synchrotron spectrum is essentially white, and fluxes of the order of those given above can be obtained for exciting any low-lying levels from the nuclear ground state. Many interesting nuclei have no convenient radioactive parents (e.g., ^{40}K, ^{73}Ge,...) and the synchrotron source should be particularly important for studying these substances.

We have discussed elsewhere[4] how filters may be devised which will extract the "Mössbauer slice" from the white synchrotron radiation for use as a source of Mössbauer γ rays. Here we discuss certain new time-differential-perturbed-angular-correlation- (TDPAC) type experiments[5] which could be performed using pulsed synchrotron sources leading to interesting new results.

The synchrotron radiation from SPEAR or DORIS operating in the single-bunch mode consists of sharp pulses of about 10^{-10}-sec duration and about 10^{-6}-sec separation between pulses.[2] Excited nuclear states of energy less ~ 100 keV commonly have lifetimes in the range $\Gamma^{-1} = 10^{-6} - 10^{-10}$ sec. If the pulse, monochromatized to perhaps 1-eV bandwidth at the nuclear transition, impinges upon a small sample containing the resonant nuclei then the electronically scattered x rays, photoelectrons, etc., will emerge promptly during the 10^{-10}-sec pulse, while those processes involving nuclear excitation will be delayed a mean time Γ^{-1}. Therefore, by using a timed detector which can recover from the prompt pulse in a time short compared to Γ^{-1} the resonant and nonresonant events can be separated temporally.[1]

There will be beats in the resonance radiation following the prompt pulse, both in the elastic and inelastic scattered γ rays and the internal conversion electrons as well. Measurement of the beat frequencies will give a direct determination of the hyperfine splittings of the individual nuclei, or of the energy shifts between two different nuclei located in different chemical or magnetic sites.

In the usual TDPAC experiments beats are observed following the population of, say, a first-excited state by the radiative decay of a broad higher state; the beat frequencies give the splittings of the excited state. The present method promises to be much more powerful for a variety of reasons. First the pulse gives a direct excitation of the nuclei from their ground

state. This obviates the need for radioactive sources. More important is this: The synchrotron pulse excites several nuclear excited state sublevels suddenly and coherently which then "oscillate" at their various natural frequencies giving beats at the difference frequencies in the decay probabilities in various directions. The excitation amplitude of this initial state is, for a particular nucleus, independent of the state of atomic binding and the temperature since the synchrotron spectrum is white over many electron volts. However if the recoilless probability, the Mössbauer f, is appreciable then there will be appreciable coherent elastic scattering which will exhibit beats resulting from the interference of waves emitted by *different* nuclei (while if f is small recoil effects will largely wipe out the interference in waves emitted by different nuclei, and the usual TDPAC theory applies).[5,6]

The new and important element in the synchrotron pulse experiment is that the very well collimated pulse produces a spatially coherent excitation of the resonant nuclei in a crystal, and if the pulse direction is set for a Bragg reflection then the Bragg reflected wave will exhibit beats, the measurements of which will give the difference frequencies of the resonance transitions. If the mosaic spread of the crystal is $\lesssim 10^{-3}$ rad, then the reflected beam will have a collimation $d\Omega = 10^{-6} - 10^{-7}$ sr and a very small detector (area ~ 1 mm^2 located ~ 1 m from the target) can be used giving a large signal-to-noise ratio. If f and the concentration of resonant nculei in the crystal are large then *an appreciable fraction of the incident photons in the resonance width will be coherently reflected.* In fact, as we shall see, because of the very low "entropy," or high brightness, of the incident pulse *the very highly collimated coherently scattered γ rays may exceed in number those normally expected (in $\sim 4\pi$ sr) for incoherent scatterers,* and even for small crystals *become comparable to or larger than the number of internal conversion electrons.*

The theory for the beats in the (spatially) incoherent processes is parallel to that developed for ordinary TDPAC experiments, that for the coherent scattering is new.

II. SPATIALLY INCOHERENT PROCESSES

To avoid needless complexity we shall consider a nuclear dipole transition ($E1$ or $M1$). For simplicity, we shall neglect relaxation effects and assume that the ground and excited states are split by static hyperfine (Zeeman, quadrupole) fields. Suppose the nucleus is initially in the mth sublevel of its ground state $|gm\rangle$, and represents the amplitude of the synchrotron pulse incident on the nucleus by $\vec{F}(t)$ with $\vec{F}(t) = 0$ for $t < 0$ and $t > \tau$, then the amplitude $C_{nm}(\tau)$ that the nth sublevel of the excited state $|en\rangle$ has been excited at the end of the pulse is

$$C_{nm}(\tau) = +\frac{i}{\hbar} e^{-i\epsilon_{ne}\tau} \int_0^\tau \langle en|\,\vec{\mu}\,|gm\rangle \times e^{+i\omega_{nm}t}\cdot \vec{F}(t)\,dt \quad , \tag{1}$$

where $\vec{\mu}$ is the dipole operator ($\vec{F} = \vec{E}$ for $E1$ and $\vec{F} = \vec{B}$ for $M1$ transitions) and $\omega_{nm} = \epsilon_{ne} - \epsilon_{mg}$, where the ϵ's are the energies of the indicated states. In (1) we have assumed that $\Gamma\tau << 1$.

We now write $\omega_{nm} = \omega_0 + \Delta_{nm}$, where ω_0 is the mean transition frequency and Δ_{nm} is the splitting. Then if $\Delta_{nm}\tau << 1$ for all n and m, (1) becomes

$$C_{nm}(\tau) \approx \frac{i}{\hbar} e^{-i\epsilon_e\tau} \int_0^\tau \langle en|\,\vec{\mu}\,|gm\rangle \times e^{i\omega_0 t}\cdot \vec{F}(t)\,dt \quad . \tag{2}$$

This assumption is made because it is true for most transitions of interest and because it simplifies the formalism somewhat. The synchrotron pulse consists of many photons emitted by the many electrons in the bunch incoherently one with the other. The pulse from a given electron has a width $\sim \lambda_c/c$ $\sim 10^{-8}/3\times 10^{-10} \sim 10^{-19}$ sec,[2] or if this radiation is filtered to about 1 eV, this coherence time becomes $\sim 10^{-15} << \tau \sim 10^{-1}$ sec. The probability of a given process should be computed starting from (1) where F represents a one-photon amplitude with a duration of the order of the coherence time, and the result summed over all of the photons occurring in the pulse of duration τ, which, with the assumption (2), just amounts to multiplying by the number of photons in the pulse.

From (2) we now have

$$|\psi_e\rangle = \frac{i}{\hbar} \sum_n \vec{\mu}_{nm}\cdot \vec{F}_{\omega_0} e^{-i(\epsilon_{ne} - i\Gamma/2)t}|en\rangle, \quad t > \tau \tag{3}$$

as the excited state amplitude, where

$$\vec{F}_{\omega_0} = \int_{-\infty}^{+\infty} \vec{F}(t) e^{i\omega_0 t}\,dt \quad .$$

The results (1)–(3) were obtained neglecting the vibrational amplitude of the nucleus $\vec{r}$, and the fact that the vibrational state of the crystal $|\chi\rangle$ might change when the quantum was absorbed. The transition amplitude for $|g,m|\chi_0\rangle \rightarrow |en\rangle|\chi_f\rangle$ is, in analogy to (1),

$$C_{nf,m0}(\tau) = \frac{i}{\hbar} e^{-i(\epsilon_{ne} + E_f)\tau} \vec{\mu}_{nm}\cdot \int_0^\tau e^{i(\omega_{nm} + E_f - E_0)t} \langle \chi_f|\vec{F}\left(t - \frac{\hat{n}_0\cdot\vec{r}}{c}\right)|\chi_0\rangle\,dt$$
$$\approx \frac{i}{\hbar} e^{i(\epsilon_{ne} + E_f)\tau} \vec{\mu}_{nm}\cdot \vec{F}_{\omega_0} \langle \chi_f|e^{i\vec{k}_0\cdot\vec{r}}|\chi_0\rangle \quad , \tag{4}$$

where $\hat{n}_0$ is the pulse propagation direction, and where strictly rather than $\vec{k}_0 = c^{-1}\omega_0\hat{n}_0$ in (4), we should have $\hat{n}_0(\omega_{nn'} + E_{f'} - E_0)c^{-1}$, but since $\omega_0 \sim 10^4$ eV, $E_{f'} - E_0 \lesssim 10^{-1}$ eV, the difference is negligible, and similarly $\vec{F}_{\omega_{nn'}} + E_{f'} - E_o \approx \vec{F}_{\omega_0}$ to good approximation. The extension of (3) is now

$$|\psi_c\rangle = \frac{i}{\hbar}\left(\sum_n \vec{\mu}_{nn'}\cdot\vec{F}_{\omega_0}e^{-i(\epsilon_n - i\Gamma/2)t}|en\rangle\right)\sum_f \langle\chi_f|e^{i\vec{k}_0\cdot\vec{r}}|\chi_0\rangle e^{-iE_{f'}t}|\chi_{f'}\rangle 1(t) \quad , \tag{5}$$

where $1(t)$ is the step function, and where we neglect τ (we assume $\Gamma\tau \ll 1$ and $\Delta_{nn'}\tau \ll 1$). The relative amplitudes for the excitation of the various states $|en\rangle$ are independent of the nuclear motions.

The probability per second for emitting a photon in direction $\hat{n}_{f'}$ with the nucleus going to the ground state $|gm'\rangle$ is now given by the usual dipole radiation formula (see Appendix A)

$$\frac{dP_{m'm}}{d\Omega\, dt} = \frac{1}{2\pi}\frac{k_0^3}{\hbar^3}\left|\sum_n \vec{\mu}^{\perp}_{m'n}\vec{\mu}_{nm}\cdot\vec{F}_{\omega_0}e^{-i(\omega_{nm'} - i\Gamma/2)t}\right|^2 \sum_{f'}\left|\sum_f \langle\chi_{f'}|e^{-i\vec{k}_{f'}\cdot\vec{r}}|\chi_f\rangle\langle\chi_f|e^{i\vec{k}_0\cdot\vec{r}}|\chi_0\rangle e^{-iE_{f'}t}\right|^2 ,$$

$$= \frac{1}{2\pi}\frac{k_0^3}{\hbar_3}e^{-\Gamma t}\left|\sum_n \vec{\mu}^{\perp}_{m'n}\vec{\mu}_{nm}\cdot\vec{F}_{\omega_0}e^{-i(\omega_{nm'})t}\right|^2 , \tag{6}$$

where $\vec{\mu}^{\perp}$ is the component of $\vec{\mu}$ perpendicular to $\hat{n}_{f'}$, and in (6) we again have neglected $\Delta_{nn'}$ and $E_{f'} - E_f$ relative to ω_0. The interpretation of (6) is familiar from the theory of angular correlation.[7] The radiation rate in a given direction is independent of the nuclear motion and is proportional to the square of the perpendicular component of the transition-matrix element of the dipole moment operator between $|\psi_c\rangle$ and $|gm'\rangle$ [which appears within the first brackets in (6)]. The magnitude of the transition matrix element of the dipole moment decreases as $e^{-\Gamma t/2}$ but its direction precesses, and as a consequence the cross tems in the square of the sum in (6) do not vanish and give rise to beats (frequencies, $\epsilon_n - \epsilon_{n'}$) in the radiation emitted in a given direction.

Suppose the hyperfine field is axially symmetric. Take the symmetry axis to be $\hat{z}$, and n and m to be the magnetic quantum numbers of an excited state and ground state, respectively. Then

$$\langle en|\mu_p|gm\rangle = [\tfrac{3}{4}(\Gamma_\gamma/k_0^3)]^{1/2}C(j_0 1 j_1; mpn) \quad , \tag{7}$$

where μ_p $(p = 0, \pm 1)$ is a component of $\vec{\mu}$ in the spherical basis, C is the Wigner coefficient, and Γ_γ is the partial width for γ emission. Substituting (7) into (6) we obtain

$$\frac{dP_{m'm}}{d\Omega\, dt} = \frac{9}{32\pi}\frac{\Gamma_\gamma^2}{\hbar^3 k_0^3}e^{-\Gamma t} \times\left|\sum_{p=-1} \hat{e}^{\perp}_{p+m-m'}C(j_0 1 j_1; m', p+m-m') \times \hat{e}_p{}^*\cdot\vec{F}_{\omega_0}C(j_0 1 j_1; mp)\; e^{-i\omega_{m+p,m'}t}\right|^2 , \tag{8}$$

where $\hat{e}_p, p = \pm 1, 0$ are the usual unit vectors in the spherical basis.

As a particular example take $j_0 = \frac{1}{2}$, $j_1 = \frac{3}{2}$ (e.g., ^{57}Fe); furthermore, let $\vec{F}_{\omega_0}$ be perpendicular to $\hat{z}$ say in the $\hat{y}$ direction (the synchrotron radiation is over 90% polarized in the plane of the orbit) then the scattered radiation in the x-y plane can, with a little algebra be shown to be

$$\frac{dP}{d\Omega\, dt} = \frac{7}{8}\frac{\chi_0^2}{(1+\alpha)}\frac{c}{4\pi^2\hbar^2\omega_0}|F_{\omega_0}|^2\left(\frac{\Gamma}{1+\alpha}\frac{\pi}{2}\right) \times\frac{\Gamma}{\hbar}e^{-\Gamma t/\hbar}\{1 + \tfrac{3}{14}[\cos(\Delta_{3/2,-1/2}t - 2\varphi) + \cos(\Delta_{1/2,-3/2}t - 2\varphi)]\} , \tag{9}$$

where α is the internal-conversion coefficient, φ is the azimuthal angle, and $\Delta(n,n') = \hbar^{-1}(\epsilon_n - \epsilon_{n'})$. The factor

$$(c/4\pi^2\hbar^2\omega_0)|F_{\omega_0}|^2 2[\pi\Gamma/2(1+\alpha)]$$

is the photon flux (cm^{-2}) in the energy range $\pi\Gamma/2(1+\alpha)$ in the pulse. (A similar expression can be obtained for the TDPAC internal conversion electron distribution.) If the quadrupole splitting were negligible, then the two frequencies in (9) would be the same and one would obtain a 42% sinusoidal beat amplitude, rather than two 21% sinusoids. In this case of *spatially incoherent* scattering, there is only interference between transitions to the *same* ground state $|gm'\rangle$ and so the beat frequencies reflect only the splitting of the excited state.

III. COHERENT SCATTERING

We now compute the *coherent elastic* scattering. Consider a nucleus whose equilibrium position is $\vec{R}$. We now compute the amplitude

$$|g_i m_i\rangle|\chi_0\rangle \rightarrow |e_i n_i\rangle|\chi_0\rangle$$
$$\rightarrow |g_i m_i\rangle|\chi_0\rangle + \gamma \ .$$

The scattered photon potential at a point $\vec{r}$ and time t is proportional to (see Appendix A)

$$A(\vec{r},t) = \frac{1}{r} e^{-i(\omega_{nm} - i\Gamma/2)t^*} \left\langle \exp\left[-i\left(\omega_{nm} - i\frac{\Gamma}{2}\right)\hat{n}_f \cdot \frac{\vec{R}_i + \vec{r}_i}{c}\right]\right\rangle$$
$$\times \int_{-\infty}^{t^* + \hat{n}_f \cdot (\vec{R}_i + \vec{r}_i)/c} dt' \left\langle F\left(t' - \hat{n}_0 \cdot \frac{\vec{R}_i + \vec{r}_i}{c}\right)\right\rangle e^{i\omega_{nm}t'}$$
$$\approx \frac{1}{r} e^{-i(\omega_{nm} - i\Gamma/2)t^*} e^{-i(\vec{k}_f - \vec{k}_0)\cdot\vec{R}_i} \langle e^{-i\vec{k}_f\cdot\vec{r}_i}\rangle \langle e^{i\vec{k}_0\cdot\vec{r}_i}\rangle F_{\omega_0} 1\left(t^* - \frac{-\hat{n}_f + \hat{n}_0}{c}\cdot R_i\right) \ , \tag{10}$$

where $t^* = t - r/c$ is the retarded time at the origin, $\vec{k}_f = \hat{n}_f \omega_0 c^{-1}$, and the brackets indicate expected values for the crystal vibrational state $|\chi_0\rangle$. The wave from $\vec{R}_i$ is retarded an extra time $(\hat{n}_0 - \hat{n}_f)\cdot\vec{R}_{i/c}$. Generally the crystal thickness will satisfy $2l/c \ll \hbar/\Gamma$ and the waves from all points of the crystal will be recieved before appreciable decay takes place. We now have for the elastic scattered $\vec{E}(E1)$ or $\vec{B}(M1)$ field

$$\vec{F}_{el}(\vec{r},t) \approx \frac{i}{\hbar}\sum_{i,n_i} \frac{e^{-i(\omega_{n_i m_i} - i\Gamma/2)t^*}}{r} f k_0^2 \vec{\mu}^{\perp}_{m_i n_i} \vec{\mu}_{n_i m_i} \cdot \vec{F}_{\omega_0} e^{-i(\vec{k}_f - \vec{k}_0)\cdot R_i} 1(t^*) \ , \tag{11}$$

where $f = e\langle e^{-i\vec{k}_f\cdot\vec{r}_i}\rangle\langle e^{i\vec{k}_0\cdot\vec{r}_i}\rangle$ is the Mössbauer factor, and where we have neglected the transit time through the crystal, for simplicity, and $\vec{F}_{el}(\vec{r},t)$ is the "Feynman field" representing a scattered photon.[8] Our particular interest is in the (spatially) coherent scattering (Bragg scattering). Let there be several sites, $\vec{R}_i + \vec{r}_\alpha$, in each unit cell, $\alpha = 0, 1, \ldots$. Then we have (see Appendix A)

$$\vec{F}_{coh}(r,t) = (k_0^2/r)\vec{M}^{\perp}_{coh}(t^*,\vec{k})S(\vec{k}) \ , \tag{12}$$

where

$$\vec{M}^{\perp}_{coh}(t,\vec{k}) = \frac{i}{\hbar}\frac{Cf}{2j_0+1}\sum_{\alpha,n,m} e^{-i[\omega_{nm}(\alpha) - i\Gamma/2]t}$$
$$\times \vec{\mu}^{\perp}_{nm}(\alpha)\vec{\mu}_{nm}(\alpha)\cdot\vec{F}_{\omega_0} e^{-i\vec{k}\cdot\vec{r}_\alpha} \ , \tag{13}$$

is the analog of the x-ray structure factor, $\vec{k} = \vec{k}_f - \vec{k}_0$, C is the fraction of sites occupied by resonant nuclei, and finally

$$S(\vec{k}) = \sum_{\text{crystal}} e^{-i\vec{k}\cdot\vec{R}_i} \ .$$

If a thin crystal is set at a Bragg angle, then the reflected intensity will be proportional to the square of the form factor $|\vec{M}^{\perp}_{coh}|^2$ [see Eq. (14) or (A10)]. This will result in beats corresponding to the difference frequencies of *all* the allowed transitions ω_{nm}, from which the splittings of the excited *and* ground states may be found, whereas the beats in the incoherent scattering and the internal conversion electron intensity give only the excited-state splitting. The time dependence of the Bragg scattered intensity will be more complicated than that of the incoherent processes, necessarily, since it yields more information. For example for the $\frac{1}{2} \rightarrow \frac{3}{2}$ dipole transition considered above there are six allowed transitions and six ω_{nm}'s. $|\vec{M}_{coh}(t,\vec{k})|^2 e^{\Gamma t}$ will then, in the general case, contain 15 Fourier components. If there is negligible quadrupole splitting, degeneracy reduces this to seven, and further reduction results from properly choosing the polarization of the incident pulse. However, even in the general case, the difference frequencies are simple functions of the magnetic moments and quadrupole moments of the two states and the magnetic field and the electric field gradient at the nucleus and should allow for simple analysis of the data.

Of course, beats with frequency $\omega_{nm} - \omega_{n'm'}$, $m' \neq m$ result from the interference of waves emitted by *different* nuclei in the crystal, and if there are several sites in the unit cell then beats with frequency $\omega_{nm}(\alpha) - \omega_{n'm'}(\alpha')$, $\alpha' \neq \alpha$ are due to the interference of waves from nuclei located at sites α and α'. The latter is of some interest. Let us simplify and assume that there are two unsplit sites at $\vec{R}_i$ and $\vec{R}_i + \vec{r}$ with transition frequencies ω_0 and $\omega_0 + \Delta$, respectively. Then it is clear that $\vec{M}_{coh}$ takes the form

$$\vec{a} e^{-i\omega_0 t}(1 + e^{-i(\Delta t - \vec{k}\cdot\vec{r})}) e^{-\Gamma t/2} \ ,$$

and the Bragg scattered intensity is of the form $A[1 + \cos(\Delta t + \vec{k}\cdot\vec{r})]e^{-\Gamma t}$, so that obviously one can determine the chemical shift Δ between the two sites, and, by measuring at several reflections, determine $\vec{r}$ as well.

It is interesting to note that the scattered intensity actually is *not* of the form $Ae^{-\Gamma t}[1 + \cos(\Delta t + \vec{k}\cdot\vec{r})]$.

Even for very small crystals (e.g., less than 1 μm thick) when they are excited at a Bragg angle the coherent radiation width $\Gamma_{\rm coh}$ (see below) becomes comparable to (perhaps larger than) the incoherent width[9] (incoherent scattering, internal conversion), but in this simple example $\Gamma_{\rm coh} \propto A(1+\cos\Delta t)$, varying from $2A$ to zero, and rather than $e^{-\Gamma t}$ with Γ constant there should appear $e^{-\Gamma(t)t}$, where

$$\Gamma(t) = \Gamma_{\rm incoh} + \Gamma_c + (\Gamma_c/t\Delta)[\sin(\Delta t + \vec{k}\cdot\vec{r}) - \sin(\vec{k}\cdot\vec{r})] \ ,$$

multiplying the coherent and *incoherent* decay probabilities.

The coherent scattering $\sim|\vec{F}_{\rm coh}|^2$ is only appreciable [see Eq. (12)] when $\vec{k}\approx\vec{\tau}$, a reciprocal-lattice vector for the crystal. Let a thin crystal be set at a Bragg angle to the synchrotron pulse. If $\delta(\approx 10^{-4})$ is the angular spread of the incident radiation and Δ is the mosaic spread of the crystal, then in the Born approximation the number of quanta Bragg scattered per second is approximately[10]

$$\frac{dP(\vec{\tau})}{dt} = \frac{k_0^3}{2\pi\hbar}|\vec{M}^{\perp}_{\rm coh}(\vec{\tau},t)|^2 V n_0 \times \frac{n_0\lambda^3}{(\sin2\theta)(\delta^2+\Delta^2)^{1/2}} \ , \qquad (14)$$

where V is the crystal volume illuminated by the pulse, n_0 is the number of unit cells per unit volume, and θ is the Bragg angle. The last factor in (14), $n_0\lambda^3/(\delta^2+\Delta^2)^{1/2}\sin2\theta$, arises from the fact that the coherent scattering from a small perfect crystallite is near zero unless the incident wave vector $\vec{k}_0$ is within the Bragg reflection width δ' of a set of Bragg planes. It is easily shown[11] that $|S(\vec{k}_f-\vec{k}_0)|^2$ integrated over the direction of $\hat{k}_f$ and averaged over the direction of the incident beam $\hat{k}_0$ is $4\pi V n_0$ if $k_0 a >> 1$ (where a is the interatomic distance) independent of the spatial distribution of the scatterers. For a "gas" of scatterers, we have

$$\Sigma(\vec{k}_0) \equiv \int |S(\vec{k}_f-\vec{k}_0)|^2 d^2\hat{k}_f = 4\pi n_0 V \equiv \bar{\Sigma} \ ,$$

independent of $\hat{k}_0$; but for a crystal, $\Sigma(\vec{k}_0)$ is nearly zero except when $\hat{k}_0$ is near a Bragg angle where it then attains a value $\Sigma(\vec{k}_0)\approx\bar{\Sigma}\Delta\theta/\delta' >> \bar{\Sigma}$, where $\Delta\theta >> \delta'$ is a mean spacing between Bragg directions. Formula (14) was derived assuming δ or $\Delta \gtrsim \delta'$. In short, atoms do not scatter independently in a crystal; they interfere with each other, as we all know; and near a Bragg angle, the effective scattering cross section per atom is much larger than the mean atomic scattering cross section.

We now specialize and suppose for simplicity only one atom per unit cell, and again as in (7) that the hyperfine field is uniaxial, we then have from (7), (8), and (13),

$$\vec{M}^{\perp}_{\rm coh} = \frac{i}{\hbar}\frac{3}{4}\frac{\Gamma_\gamma}{k_0^3}\frac{Cf}{2j_0+1}e^{-\Gamma t/2}\sum_{p,m}\hat{e}_p^{\perp}\hat{e}_p^{\,*}\cdot\vec{F}_{\omega_0}C^2(j_0 1 j_1; mp)e^{-i\omega_m + p,p^t} \ , \qquad (15)$$

or specializing to $j_0=\frac{1}{2}$, $j_1=\frac{3}{2}$,

$$\vec{M}_{\rm coh} = A[\tfrac{1}{2}(\hat{x}+i\hat{y})(e^{-i\omega_{3/2,\,1/2}t}+\tfrac{1}{3}e^{-i\omega_{1/2,\,-1/2}t})(\hat{x}+i\hat{y}) + \tfrac{1}{2}(\hat{x}-i\hat{y})(e^{-i\omega_{-3/2,\,-1/2}t}+\tfrac{1}{3}e^{-i\omega_{-1/2,\,1/2}t})(\hat{x}-i\hat{y}) + \tfrac{2}{3}\hat{z}(e^{-i\omega_{1/2,\,1/2}t}+e^{-i\omega_{-1/2,\,-1/2}t})\hat{z}]\cdot\vec{F}_{\omega_0} \ . \qquad (16)$$

For illustrative purposes suppose the quantization axis $\hat{z}$ is perpendicular to the scattering plane and that $\vec{F}_{\omega_0}$ is in the $\hat{z}$ direction, then from (14)–(16) we obtain

$$\frac{dP_{\rm coh}(t)}{dt} = n_0 V\frac{C^2f^2n_0\lambda^3}{(\sin2\theta)(\delta^2+\Delta^2)^{1/2}}\frac{\varkappa^2}{2(1+\alpha)^2}\frac{|F_{\omega_0}|^2}{4\pi^2\hbar^2k_0}\left(\frac{\pi\Gamma}{2}\right)\frac{\Gamma}{\hbar}[1+\cos(\omega_{1/2,\,1/2}-\omega_{-1/2,\,-1/2})t]e^{-\Gamma t} \ . \qquad (17)$$

On the other hand, by squaring $C_{nf,m0}$ [Eq. (4)], summing over n and f, averaging over m and using (7), we obtain

$$\frac{dP_{\rm incoh}(t)}{dt} \approx n_0 V C\frac{4\pi\varkappa^2}{1+\alpha}\frac{|F_{\omega_0}|^2(\frac{1}{2}\pi\Gamma)}{4\pi^2\hbar^2k_0}\frac{\Gamma}{\hbar}e^{-\Gamma t} \qquad (18)$$

for the rate of incoherent processes (internal conversion plus incoherent scattering), and for the ratio of (17) to (18),

$$\Gamma_{\rm coh}/\Gamma_{\rm incoh} \approx [1/8\pi(1+\alpha)][Cf^2n_0\lambda^3/(\sin2\theta)(\delta^2+\Delta^2)^{1/2}](1+\cos\Delta\omega t) \ , \qquad (19)$$

where $\Delta\omega = \omega_{1/2,1/2} - \omega_{-1/2,-1/2}$. For example, for 100% ^{57}Fe taking $Cf^2 n_0 \lambda^3/\sin 2\theta \approx 10^{-1}$, the mosaic spread $\Delta \approx 10^{-3}$, and $1+\alpha = 10$, we have

$$(\Gamma_{\rm coh}/\Gamma_{\rm incoh})_{^{57}{\rm Fe}} \approx 0.4(1+\cos\Delta\omega t) \ , \tag{20}$$

and rather than $\exp(-\Gamma t)$ multiplying (17) and (18) there should appear

$$e^{-\Gamma(t)t} \approx \exp -\Gamma_{\rm inc} t\left[1+0.4+0.4\frac{\sin\Delta\omega t}{\Delta\omega t}\right] \ , \tag{21}$$

where $\Gamma_{\rm inc} \approx \Gamma$ (for this case of large α).

Now (17) and (18) were derived using the Born approximation and the above results are strictly valid only for very thin crystals. We have shown elsewhere[4] that in the case of high-concentration ^{57}Fe a few-thousand-layer crystal will reflect approximately 25% of the "resonance slice" of synchrotron pulse incident at a Bragg angle, and so for even such a thin crystal the Born approximation is not very reliable for quantitative predictions, for which one must use the multiple scattering[8] or "dynamical theory."[12] Nevertheless, the above results are qualitatively correct. For a very thin crystal with small mosaic spread, (i) an appreciable fraction of the resonance radiation will be coherently reflected (in $d\Omega \approx 10^{-7}$ sr for the case considered), (ii) the coherently scattered fraction can be much larger than $1/(1+\alpha)$, and (iii) the coherently scattered fraction, and thus $\Gamma_{\rm coh}/\Gamma$, will exhibit beats and the decay will not be exponential but (approximately) of the form (21).

IV. CONCLUSION

In this paper we have presented the basic theory for the quantum beats which will be observed when nuclei with low-lying excited states are excited by a synchrotron pulse. A new and important effect will be observed when the pulse is incident at a Bragg angle on a good single crystal: beats will be observed in the (spatially) coherently emitted γ rays, and if the concentration of the resonant nuclei is high, and the Mössbauer factor f is appreciable, then an appreciable fraction of the incident photons in the spectral width of the nuclear level will be reflected into a very small solid angle. From the beat frequencies the differences of all the nuclear transition frequencies can be determined, including, if there are more than one resonant nuclei per unit cell, the isomer shifts between the sites. In addition, if the coherently scattered waves are appreciable compared to the incoherent decay modes then the decay "constant" Γ will be strongly time dependent for both the coherent and incoherent (e.g., internal conversion) processes.

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APPENDIX A

Here we outline, in more detail, the derivation of the basic equations (6) and (10).

Because the temporal duration of the incident radiation pulse is short compared to the lifetime of the nuclear resonance, it is legitimate to regard the formation of the intermediate excited state $|\psi_e\rangle$ and its subsequent decay as two independent quantum-mechanical processes.[13] Consequently, we can use the general theory for γ emission given in Paper IIIA.[8]

As in Eq. (1) of IIIA, if the initial state of the crystal $|\psi_0\rangle$ corresponds to the Mössbauer nucleus at $\vec{R}_l$ being in an excited state and the remaining Mössbauer nuclei being in their ground states, then the photon potential $A_\mu(z)$ at the space-time point z due to a transition to a final crystal state $|\psi_f\rangle$ corresponding to all Mössbauer nuclei being in their ground states is given by

$$A_\mu(z) = c^{-1}\langle\psi_f|\sum_l \int d^4x\, D_{\mu\nu}(z,x) J^0_\nu(x,l)|\psi_0\rangle \ , \tag{A1}$$

where $J^0_\nu(x,l)$ is the source transition current of the lth nucleus, and $D_{\mu\nu}(z,x)$ is the photon propagator, which if we neglect scattering is given by the free photon propagator $D^{(0)}_{\mu\nu}(z,x)$,

$$D^{(0)}_{\mu\nu}(z,x) = -4\pi g_{\mu\nu}\int \frac{d^4k}{(2\pi)^4}(k_4^2 - k^2 + i\epsilon)^{-1} \times \exp i[\vec{k}\cdot(\vec{z}-\vec{\psi}) - k_4(t_z - t_\psi)] \ , \tag{A2}$$

where in our metric, $g_{00} = +1$, $g_{xx} = g_{yy} = g_{zz} = -1$, and all other components equal 0. As discussed in IIIA, the source transition current $J^0_\mu(x,l)$ is given to excellent approximation by

$$J^0_\mu(x,l) = e^{iH_0 t_x} j^{(l)}_\mu(\vec{x}) \times e^{-i(H_0 - i\Gamma/2)(t_x - \tau_l)} 1(t_x - \tau_l) \ , \tag{A3}$$

where $j^{(l)}_\mu(\vec{x})$ is the current-density operator for the atom l, Γ is the width of the excited Mössbauer level, and H_0 is the Hamiltonian for the complete crystal with electromagnetic interactions replaced by instantaneous Coulomb and magnetic interactions. In (A3) it is assumed that the excited nuclear state was produced at $t_x = \tau_l$. For a synchrotron pulse arriving at the origin at $t = 0$, then $\tau_l = c^{-1}\hat{n}_0\cdot\vec{R}_l$, where $\hat{n}_0$ is the direction of propagation of the pulse.

Using the time-dependent perturbation theory leading to Eq. (5), the initial excited state produced at R_l at $t = \tau_l$ by the synchrotron pulse is

$$|\psi_c(l)\rangle = \sum_f \frac{i}{\hbar} e^{i\vec{k}_0'\cdot\vec{R}_l}\left(\sum_{n_l}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0}|e_l n_l\rangle\right) \times \langle\chi_f|e^{i\vec{k}_0\cdot\vec{r}_l}|\chi_0\rangle|\chi_f\rangle \ , \tag{A4}$$

where

$$\vec{k}_0' = \hat{n}_0(\omega_0 + E_f - E_0)c^{-1} \approx \hat{n}_0\omega_0 c^{-1} \ .$$

Substituting (A4) and (A2) into (A1) and carrying out the t_N integration gives for the transverse part of the photon potential produced by the transition

$$|\psi_c(l)\rangle \rightarrow |gm_l'\rangle|\chi_{f'}\rangle \ ,$$

$$\vec{A}_\perp(R,t) = \frac{1}{R}1\left(t^* + \frac{\hat{n}_f\cdot\vec{R}_l}{c} - \frac{\hat{n}_0\cdot\vec{R}_l}{c}\right)\sum_f\sum_{n_l} e^{-i(\omega_{n_l m_l'} - i\Gamma/2)t^*} e^{-i(\vec{k}_f' - \vec{k}_0')\cdot\vec{R}_l}$$

$$\times c^{-1}\langle gm_l'|\int dx\, j_1^{(l)}(\vec{x})e^{-i\vec{k}_f'\cdot\vec{x}}|en_l\rangle\frac{i}{\hbar}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0}e^{-i\Delta E_{ff'}t^*}\langle\chi_{f'}|e^{-i\vec{k}_f'\cdot\vec{r}_l}|\chi_f\rangle\langle\chi_f|e^{i\vec{k}_0'\cdot\vec{r}_l}|\chi_f\rangle$$

$$= \frac{1}{R}1\left(t^* + \frac{\hat{n}_f - \hat{n}_0}{c}\cdot\vec{R}_l\right)\sum_f e^{-i(\vec{k}_f' - \vec{k}_0')\cdot\vec{R}_l}(e^{-i\Delta E_{ff'}t^*}\langle\chi_{f'}|e^{-i\vec{k}_f'\cdot\vec{r}_l}|\chi_f\rangle\langle\chi_f|e^{i\vec{k}_0'\cdot\vec{r}_l}|\chi_f\rangle$$

$$\times\left(\frac{\omega_0}{\hbar c}\sum_{n_l} e^{-i(\omega_{n_l m_l'} - i\Gamma/2)t^*}\vec{\mu}^{\perp}_{m_l' n_l}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0}\right) \ . \tag{A5}$$

Here $t^* = t - R/c$ is the retarded time at the origin, $\vec{k}_f' = \hat{n}_f(\omega_0 + E_f - E_f')c^{-1}$, and in the second line of (A5) we have made the $E1$ dipole approximation $j_1(-k_f') \rightarrow -i\omega_0\vec{\mu}^{\perp}$ [for an $M1$ transition, we should of course take $j_1(-k_f) \rightarrow i\vec{k}_f\times\vec{\mu}$]. Also in carrying out the time integrals, we have dropped correction terms which are of order Γ/ω_0 and hence quite negligible.

For the *coherent elastic* scattering we have $|\chi_f\rangle = |\chi_{f'}\rangle = |\chi_0\rangle$, $m_l' = m_l$, and $k_f = k_0$, so in this case the total elastically scattered wave from all sites is

$$\vec{A}_\perp^{\,\mathrm{el}}(\vec{R},t) = \frac{1}{R}1(t^*)\sum_l e^{-i(\vec{k}_f - \vec{k}_0)\cdot\vec{R}_l}\left(\frac{\omega_0}{\hbar c}\sum_{n_l} e^{-i(\omega_{n_l m_l} - i\Gamma/2)t^*}\vec{\mu}^{\perp}_{m_l n_l}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0}\right)$$

$$\times(\langle\chi_0|e^{-i\vec{k}_f\cdot\vec{r}_l}|\chi_0\rangle\langle\chi_0|e^{i\vec{k}_0\cdot\vec{r}_l}|\chi_0\rangle) \ , \tag{A6}$$

which gives Eq. (10) with the appropriate proportionality factors. $\vec{F}(R,t)$ in Eq. (11) and subsequent equations is then the $\vec{E}(E1)$ or $\vec{B}(M1)$ field obtained from $\vec{A}_\perp$ in the usual manner, i.e., $\vec{F}(R,t) = -(1/c)(\partial\vec{A}/\partial t)$ $(E1)$, or $\vec{F}(R,t) = \vec{\nabla}\times\vec{A}$ $(M1)$. Taking the coherent average over initial nuclear ground states m_l and over the fraction of sites occupied by the resonant isotopes and over initial crystal vibration states $|\chi_0\rangle$, gives the *coherent elastic* wave

$$\vec{F}_{\mathrm{coh}}(R,t) = (k_0^2/R)M^{\perp}_{\mathrm{coh}}(t^*,\vec{k})S(\vec{k}) \tag{A7}$$

in the notation of Eqs. (12) and (13).

For the *spacially incoherent* processes, the photon flux [probability/cm^2/sec of finding a photon at $(\vec{R},t)$] is given by Eq. (41), of III A,[8]

$$n(\vec{R},t) = \frac{\omega_0}{2\pi\hbar c}\sum_{f'}\langle|\vec{A}_\perp(R,t;0\rightarrow f')|^2\rangle$$

$$= \frac{c}{2\pi\hbar\omega_0}\sum_{f'}\langle|\vec{F}(R,t;0\rightarrow f')|^2\rangle \ . \tag{A8}$$

Here $A_\perp$ is given by (A5), the angular brackets refer to an average over the ensemble representing the initial state of the crystal, and the sum is over all final phonon states $|\chi_{f'}\rangle$, and over final nuclear ground states m_l', giving

$$n(\vec{R},t) = \frac{k_0^3}{2\pi\hbar^3}e^{-\Gamma t^*}$$

$$\times\sum_{m_l'}\left\langle\left|\sum_{n_l}\vec{\mu}^{\perp}_{m_l' n_l}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0}e^{-i\omega_{n_l m_l'}t^*}\right|^2\right\rangle\frac{1(t^*)}{R^2} \ , \tag{A9}$$

which is the basis of Eq. (6). The ensemble average $\langle\ \rangle$ will be over initial nuclear ground states which reduces to $(2j_0+1)^{-1}\sum_{m_l}$.

For the *coherent elastic* scattering, the scattered photon flux is given by $(c/2\pi\hbar\omega_0)|\vec{F}_{\mathrm{coh}}|^2$, or, making use of (A7),

$$n_{\rm coh}(R,t) = \frac{1(t^*)}{R^2}\,\frac{k_0^3}{2\pi\hbar}\,|M^{\perp}_{\rm coh}(t^*,\vec{k})|^2|S(\vec{k})|^2 \ . \tag{A10}$$

Averaging over the angular spread of the incident synchrotron radiation and over the mosaic spread of the crystal gives Eq. (14).

An alternative method for deriving these results is to treat the problem purely as a scattering problem, using the general theoretical treatment given in Papers I and II.[8] In particular, with the vector potential of the incident pulse given by

$$\vec{A}(t) = \int \vec{a}^{\,0}(\omega)e^{-i\omega t}d\omega \ , \tag{A11}$$

the coherent elastic wave is given by Eq. (24) of I,[8]

$$A^{\rm coh}_{\mu}(\vec{R},t) = \sum_f \int d\omega \frac{e^{i(kR-\omega t)}}{R} \times e^{-i(\vec{k}_f-\vec{k}_0)\cdot\vec{R}_c} M^{\rm coh}_{\mu\nu}(\vec{k}_f,\vec{k}_0;\omega) \times a^0_{\nu}(\omega) \ . \tag{A12}$$

Here

$$\hat{\epsilon}^f_{\mu} M^{\rm coh}_{\mu\nu}(\vec{k}_f,\vec{k}_0;\omega)\epsilon^0_{\nu} = f_{\rm coh}(\vec{k}_f,\hat{\epsilon}_f;\vec{k}_0,\hat{\epsilon}_0)$$

is the coherent elastic scattering amplitude for scattering the photon $|\vec{k}_0,\hat{\epsilon}_0\rangle \rightarrow |\vec{k}_f,\hat{\epsilon}_f\rangle$ by the nucleus and is given for an arbitrary multipole transition by Eq. (4) of II (taking the coherent averge of the elastic scattering amplitude). For an $E1$ transition this gives explicitly

$$f_{\rm coh}(\vec{k}_f,\hat{\epsilon}_f;\vec{k}_0,\hat{\epsilon}_0) = \frac{k_0^2 C e^{-k^2\langle x^2\rangle}}{2j_0+1} \times \sum_{n_l m_l} \frac{\hat{\epsilon}_f\ \vec{\mu}_{m_f n_l}\vec{\mu}_{n_l m_i}\ \hat{\epsilon}_0}{(\omega_{n_l m_l}-\omega)-\frac{1}{2}i\Gamma} \ , \tag{A13}$$

while for an $M1$ transition

$$(\hat{\epsilon}_f\cdot\vec{\mu})(\vec{\mu}\cdot\hat{\epsilon}_0) \rightarrow (\vec{k}_f\times\hat{\epsilon}_f\cdot\vec{\mu})(\vec{k}_0\times\hat{\epsilon}_0\cdot\vec{\mu}) \ .$$

Approximating the incident pulse as a Lorentzian having a width $\gamma \gtrsim 1$ eV $>> \Gamma$, centered about the resonant frequency ω_0, then

$$\vec{a}^{\,0}(\omega) \propto i[2\pi(\omega-\omega_0+i\gamma)]^{-1}\hat{\epsilon}_0 \ ,$$

and (A12) and (A13) give

$$A^{\rm coh}_{\perp}(R,t) = \frac{1(t^*)}{R}S(\vec{k})\frac{k_0 C e^{-k^2\langle x^2\rangle}}{2j_0+1} \times \sum_{n_l m_l} e^{-i(\omega_{n_l m_l}-i\Gamma/2)t^*}\vec{\mu}^{\perp}_{m_l n_l}\vec{\mu}_{n_l m_l}\cdot\vec{F}_{\omega_0} \ , \tag{A14}$$

which is equivalent to (A7). Here

$$\vec{F}_{\omega_0} = 2\pi a^0(\omega_0)\begin{cases} ik_0\hat{\epsilon}_0(E1) \\ i\vec{k}_0\times\hat{\epsilon}_0(M1)\end{cases} = \int \vec{F}(t)e^{+i\omega_0 t}dt$$

as before (and we have taken $\vec{F}_{\omega_{n_l m_l}} \cong \vec{F}_{\omega_0}$), and we have dropped the scattered wave contribution proportional to $e^{-\gamma t^*}$ which emerges promptly and carries only Γ/γ of the total reflected energy.

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