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Recent Results in Relaxation Theory: Non-Stationary
Processes and Polarization Effects

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M. Blume

Brookhaven National Laboratory*, Upton, New York 11973 USA

and State University of New York at Stony Brook†

Stony Brook, New York 11794 USA

Abstract

A summary is given of theoretical results concerning line-shape in the presence of non-stationary perturbations, and of the polarization of radiation emitted by a nucleus influenced by time-dependent fields.

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I. Introduction

In this paper a brief review is given of two topics in relaxation theory, both of which have some history but which have been of recent interest. The first topic, which is discussed in Section II, concerns the Mössbauer line-shape when the emitter is influenced by non-stationary random fields. Most of the theories of relaxation effects have been concerned with stationary processes, i.e. those random fields which on average do not change with time. Non-stationary processes are typically found as "after effects", as in an emitter which is initially Fe^{3+} and which subsequently captures an electron to become Fe^{2+} . The theory for these processes is straightforward but is more complex than that for stationary processes.

In Section III a discussion is given of a separate problem: what is the polarization of radiation which is emitted by a nucleus that is subject to random fields? The results presented here give a straightforward generalization of the correlation function expression for the Mössbauer line shape which answers this question. There are as yet no experimental data on this subject, but the experiments, while difficult, will be worthwhile in some circumstances to differentiate relaxation effects from other phenomena which cause unusual line-shapes.

II. Non-Stationary Relaxation Processes

The expression for the line-shape in the presence of non-stationary processes was derived in a previous paper¹ and is simply repeated here:

$$I = \int_0^\infty dt' \int_0^\infty dt'' e^{i\omega(t'-t'')} - \frac{\Gamma}{2} (t'+t'') \frac{1}{\langle V^{(-)}(t') V^{(+)}(t'') \rangle}. \quad (1)$$

Here ω is the frequency of the emitted radiation, Γ is the natural linewidth of the nuclear line, and $V^{(+)} (V^{(-)})$ is the nuclear operator for emission (absorption) of a photon. The time-development of this operator is governed by a Hamiltonian $\mathcal{H}(t)$ which, in the general case we consider, may be explicitly time-dependent. Thus

$$V^{(+)}(t) = \exp_{-} \left(i \int_0^t \mathcal{H}(\tau) d\tau \right) V^{(+)} \exp_{+} \left(-i \int_0^t \mathcal{H}(\tau) d\tau \right), \quad (2)$$

where the subscripts $\pm$ indicate positive and negative time-ordering of the exponential.

In the correlation function in eq. (1), the angular brackets denote an average over the initial state of the system:

$$\langle A \rangle = \text{tr} (\sigma A),$$

where σ is the density matrix for the system. The bar denotes an average over any stochastic elements in the time development.

If the random process is stationary and the Hamiltonian commutes with the density matrix, eq. (1) can be transformed into the familiar form¹

$$I = \frac{2}{\Gamma} \text{Re} \int_0^\infty d\tau e^{i\omega\tau} - \frac{\Gamma}{2} \tau \frac{1}{\langle V^{(-)} V^{(+)}(\tau) \rangle}, \quad (3)$$

which is widely used in Mössbauer line-shape calculations.

It should be emphasized that eq. (3) is not correct for explicitly time dependent non-stationary processes, and it is necessary to calculate with eq. (1) in these cases.

We consider here a simple class of Hamiltonian which is characteristic of a large class of non-stationary processes. We take

$$\mathcal{K}(t) = \mathcal{K}_a \epsilon(T - t) + \mathcal{K}_b \epsilon(t - T) \quad (4)$$

where $\epsilon(x) = 1, x > 0; 0, x \leq 0$. This Hamiltonian thus has the form $\mathcal{K}_a$ for $t < T$ and $\mathcal{K}_b$ for $t > T$. This models the $\text{Fe}^{3+} \rightarrow \text{Fe}^{2+}$ transition mentioned above, and, depending on the nature of $\mathcal{K}_a$ and $\mathcal{K}_b$, can represent many different physical processes. A generalization of this form would allow for a sequence of many different Hamiltonians, rather than just two.

In order to calculate the line shape for the form (4), it is substituted in eqs. (2) and (1). The stochastic average in (1) is, in this case, an average over the time T at which the transition takes place:

$$\overline{A} = \gamma \int_0^\infty dT e^{-\gamma T} A(T),$$

with γ the inverse of the lifetime of the state with Hamiltonian $\mathcal{K}_a$ (not to be confused with the inverse nuclear lifetime Γ).

The principal formula needed is

$$\exp\left(i \int_0^t \mathcal{K}(\tau) d\tau\right) = \exp(i\mathcal{K}_a t) \epsilon(T-t) + \exp(i\mathcal{K}_a T) \exp(i\mathcal{K}_b(t-T)) \epsilon(t-T). \quad (5)$$

With this expression the intensity, eq. (1), becomes

$$I = A + B + C + D,$$

with

$$A = \gamma \int_0^\infty dT e^{-\gamma T} \int_0^T dt' \int_0^T dt'' e^{i\omega(t'-t'')} - \frac{\Gamma}{2}(t'+t'')$$

$$x \langle e^{i\mathcal{K}_a t''} v(-) e^{-i\mathcal{K}_a t''} e^{i\mathcal{K}_a t'} v(+) e^{-i\mathcal{K}_a t'} \rangle,$$

$$B = \gamma \int_0^{\infty} dT e^{-\gamma T} \int_T^{\infty} dt' \int_T^{\infty} dt'' e^{i\omega(t'-t'')} - \frac{\Gamma}{2} (t'+t'')$$

$$x < e^{i\mathcal{K}_a T} e^{i\mathcal{K}_b(t''-T)} V(-) e^{-i\mathcal{K}_b(t''-T)} e^{i\mathcal{K}_b(t'-T)} V(+) e^{-i\mathcal{K}_b(t'-T)} e^{-i\mathcal{K}_a T} >,$$

$$C = \gamma \int_0^{\infty} dT e^{-\gamma T} \int_0^T dt' \int_T^{\infty} dt'' e^{i\omega(t'-t'')} - \frac{\Gamma}{2} (t'+t'')$$

$$x < e^{i\mathcal{K}_a T} e^{i\mathcal{K}_b(t''-T)} V(-) e^{-i\mathcal{K}_b(t''-T)} e^{-i\mathcal{K}_a T} e^{i\mathcal{K}_a t'} V(+) e^{-i\mathcal{K}_a t'} > ,$$

$$D = C^*.$$

The contributions A and B represent the lines due to $\mathcal{K}_a$ and $\mathcal{K}_b$, respectively, while C and D are interference terms involving both Hamiltonians (note that $\mathcal{K}_a$ drops out of B entirely if $[\mathcal{K}_a, \sigma] = 0$). The terms C and D are characteristic of non-stationary processes. They occur even in the simplest of cases, but they are more complex (and more interesting) if $\mathcal{K}_a$ and $\mathcal{K}_b$ do not commute with one another.

A straightforward calculation gives a result recently derived in a different manner by Kankaleit⁽²⁾. We take $\mathcal{K}_a = \mathcal{K}_0 + D P_{\text{ex}}$, $\mathcal{K}_b = \mathcal{K}_0$, where $\mathcal{K}_0$ is the unperturbed Hamiltonian for the nucleus and P_{ex} is a projection operator on the excited nuclear state. The nucleus thus finds itself initially in a state in which it experiences an isomer shift D, but at time T a transition occurs to surroundings which no longer produce the shift. Straightforward calculation then gives

$$A = \frac{2\gamma + \Gamma}{\gamma + \Gamma} \frac{1}{(\omega - \omega_0 - D)^2 + (\gamma + \frac{\Gamma}{2})^2},$$

$$B = \frac{\gamma}{\gamma + \Gamma} \frac{1}{(\omega - \omega_0)^2 + \frac{\Gamma^2}{4}},$$

$$C+D = - \frac{2\gamma}{\gamma + \Gamma} \frac{(\omega - \omega_0)(\omega - \omega_0 - D) - \frac{\Gamma}{2}(\gamma + \frac{\Gamma}{2})}{((\omega - \omega_0 - D)^2 + (\gamma + \frac{\Gamma}{2})^2)((\omega - \omega_0)^2 + \frac{\Gamma^2}{4})}.$$

The resulting expression has the correct limiting values:

$\gamma \rightarrow 0$ (infinite lifetime for the isomer shifted state) yields

$$I \sim \frac{1}{(\omega - \omega_0 - D)^2 + \frac{\Gamma^2}{4}},$$

$\gamma \rightarrow \infty$ yields the unshifted line, and $D \rightarrow 0$ also gives the unshifted line for all values of γ . The interference terms C and D produce asymmetries in the spectra.

The procedure followed here has been used to calculate more complex spectra (S. Banerjee, Ph.D. thesis, SUNY at Stony Brook; S. Banerjee and M. Blume to be published). In some cases a different stochastic averaging procedure may be indicated by the physical processes involved, but this is easily included in the calculations.

III. Polarization Effects

It is clear that relaxation phenomena can affect the polarization of emitted radiation. This is most easily seen by considering the familiar example of radiation emitted by a nucleus in a magnetic field which fluctuates between values $\pm h$ along a given axis. In the case of slow relaxation the Zeeman-split lines will, when observed at (say) 90° to the axis, show linear polarization. For fast relaxation, on the other hand, the spectrum collapses into a single

unpolarized line, and we then expect a complex situation for intermediate values of the relaxation time.

In order to derive the required expressions for the polarization we must look in more detail at the polarization dependence of the nuclear matrix elements. We begin with the Wigner-Weiskopf expression for the probability of a transition by the nucleus from state λ to state α with emission of a photon in the direction $\hat{k}$ with frequency ω and polarization p :

$$W \sim \frac{|\langle \alpha; \hat{k} \omega p | V | \lambda \rangle|^2}{(\omega + E_\alpha - E_\lambda)^2 + \frac{\Gamma^2}{4}} \quad (6)$$

with $p = \pm 1$. $p = +1$ stands, in the basis we use, for left circular polarization, while $p = -1$ stands for right circular polarization. If we are interested in general elliptic polarization and not simply in left or right circular polarization, we may use the fact that arbitrary polarization can be made up of a linear combination of these two:

$$|\pi\rangle = \sum_p c_p |p\rangle,$$

where the coefficients c_p determine the particular type of polarization. The probability of emission of this general type of polarization is then

$$W \sim \frac{|\sum_p c_p \langle \alpha; \hat{k} \omega p | V | \lambda \rangle|^2}{(\omega + E_\alpha - E_\lambda)^2 + \frac{\Gamma^2}{4}} \quad (7)$$

$$= \sum_{pp'} c_p c_{p'}^* \frac{\langle \lambda | V_{p'}^{(-)} | \alpha \rangle \langle \alpha | V_p^{(+)} | \lambda \rangle}{(\omega + E_\alpha - E_\lambda)^2 + \frac{\Gamma^2}{4}}$$

Here $V_p^{(+)}$ is the nuclear operator for emission of a photon with polarization p . The explicit matrix element is given by⁽³⁾

$$\begin{aligned} \langle I_0 m_0 | V_p^{(+)} | I_1 m_1 \rangle &= \alpha \sum_{LM} i^L (2L+1)^{\frac{1}{2}} D_{Mp}^{*(L)}(\hat{k}) \\ &\times (M_L + i p E_L) C(I_0 L I_1; M_0 M m_1), \end{aligned} \quad (8)$$

where M_L and E_L are the magnetic and electric 2^L pole strengths, C is a Clebsch-Gordan coefficient, and D is a rotation matrix. (In the formulas written in section II the polarization dependence of the operators was suppressed and it was assumed that a sum over polarization was taken.)

Equation (7) can now be converted into a correlation function form in the usual manner⁽¹⁾. Averaging over initial nuclear states λ , summing over the final states α , and averaging over the different polarizations emitted leads to the expression

$$I = \sum_{pp'} \overline{c_p c_{p'}} \cdot \frac{2}{\Gamma} \operatorname{Re} \int_0^\infty d\tau e^{-i\omega\tau} - \frac{\Gamma}{2\tau} \overline{\langle V_{p'}^{(-)} V_p^{(+)}(\tau) \rangle}, \quad (9)$$

where we have assumed, for simplicity, a stationary process. [For non-stationary processes the expression is simply a generalization of eq. (1), namely

$$I = \sum_{pp'} \overline{c_p c_{p'}^*} \int_0^\infty dt' \int_0^\infty dt'' e^{i\omega(t'-t'')} - \frac{\Gamma}{2}(t'+t'') \overline{\langle V_{p'}^{(-)}(t'') V_p^{(+)}(t) \rangle}$$

We will discuss only eq. (9) in the following.]

The only new quantity to be found in (9) is the 2x2 matrix $\overline{c_p c_p^*}$, which contains a complete description of the polarization of the emitted radiation. Because it is a 2x2 matrix it can be written as a linear combination of the unit matrix and the three Pauli matrices:

$$\overline{c_p c_p^*} \equiv \rho = \frac{1}{2} (1 + \underline{P} \cdot \underline{\sigma}). \quad (10)$$

The three constants P_ξ , P_η , P_ζ are in fact the Poincaré-Stokes vector of the polarization. (For a more complete discussion of this vector see ref. (3) and articles quoted therein.) For unpolarized radiation $\underline{P} = 0$, and eq. (9) reduces to the previously derived results.

The radiation of polarization $\underline{P}$ is then given by

$$I(\underline{P}) = \text{tr}(\rho M), \quad (11)$$

where ρ is given in (10) and

$$M_{p'p} = \frac{2}{\Gamma} \text{Re} \int_0^\infty d\tau e^{i\omega\tau} - \frac{\Gamma}{2} \frac{\tau}{\langle v_{p'}^{(-)} v_p^{(+)}(\tau) \rangle} \quad (12)$$

A full measurement of the intensity and the three components of the Poincaré-Stokes vector will thus determine four correlation functions $M_{p'p}$ rather than the single combination $M_{++} + M_{--}$ determined by detectors insensitive to the polarization of the radiation. It should be pointed out that eqs. (11) and (12) are useful even for time-independent fields.

Detailed calculations for simple relaxation models will be presented separately (S. Banerjee and M. Blume, to be published).

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References

1. BLUME, M., in Hyperfine Structure and Nuclear Radiations
(North-Holland Publishing Co., Amsterdam, 1968) p. 911.
2. KANKELEIT E., Z. Physik A275 (1975) 119.
See also GAL J., HADARI Z., BAUMINGER E.R., NOWIK I., OFER, S.,
Sol. St. Comm. 15 (1974) 1805.
3. BLUME M., and KISTNER O.C., Phys. Rev. 171 (1968) 417.