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The harmonic lattice, recoilless transitions, and the coherent state

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The probability for recoilless transitions, relevant for the understanding of x-ray scattering from atoms bound in a crystal (applicable also to elastic scattering of neutrons from solids and to the Mössbauer effect), given by the Debye–Waller factor, is derived in a novel manner using the coherent state basis for the normal mode oscillators describing the harmonic lattice, a method which, while being simple and elegant, also reveals the relationship to a heuristic classical discussion of the problem.

I. THE PROBABILITY FOR RECOILLESS TRANSITIONS

The intensity distribution in x-ray diffraction spots from atoms in a lattice (made diffuse by thermal vibrations) is given by the Debye–Waller factor.¹ The result also applies to the determination of the fraction of gamma rays emitted (or absorbed) resonantly (without change in energy due to recoil) by a nucleus bound in a crystal that occurs in the celebrated Mössbauer effect as well as for the case of elastic scattering of neutrons from solids. The probability that a nucleus in the lattice emits (or absorbs) a photon of momentum $\mathbf{p}$ without any change in the state of the lattice is given by the square modulus of the overlap between the state vector $e^{i\mathbf{p}\cdot\mathbf{X}/\hbar}|L_i\rangle$, corresponding to the lattice with a “site,” displaced by $\mathbf{X}$ with respect to its equilibrium position and recoiling with momentum $\mathbf{p}$, and the state $|L_i\rangle$ representing the undisturbed lattice. Of course, with a solid at temperature T it is also essential to include the thermal average (shown below through a second set of angular brackets) thus yielding the basic formula

$$f = |\langle\langle L_i|e^{i\mathbf{p}\cdot\mathbf{X}/\hbar}|L_i\rangle\rangle_T|^2, \quad (1)$$

from which the Debye–Waller factor f is to be calculated. Here, it needs to be noted that p the photon momentum is a number while $\mathbf{X}$ is an operator. Lattice vibrations in a solid, in the harmonic approximation, are described in terms of the normal modes, with corresponding coordinates $\{\xi_s\}$, and hence by an ensemble of oscillators with frequencies $\{\omega_s\}$. The state of the lattice can then be specified by providing the oscillator quantum numbers (occupancies) of the different modes, viz., $|n_s\rangle$. We can now express the component of $\mathbf{X}$ in the direction of recoil, $\hat{\mathbf{p}}\cdot\mathbf{X}$ (where $\hat{\mathbf{p}}$ is the unit vector along $\mathbf{p}$), in terms of the normal-mode coordinates ξ_s for the lattice:

$$\hat{\mathbf{p}}\cdot\mathbf{X} = \sum_{s=1}^{3N} c_s \xi_s, \quad (2a)$$

with the normalization condition

$$\sum_{s=1}^{3N} c_s^2 = 1, \quad (2b)$$

on the expansion coefficients, where $3N$ is the number of degrees of freedom for N atoms in three dimensions. Expressing the displacement of the nucleus in terms of the normal modes of the lattice (made possible by the fact that

the normal modes are but combinations of the displacements of the nuclei) enables us to discuss the response of the lattice as a whole to the decay or scattering event. The underlying theory and the associated calculation to obtain the probability for such recoilless processes were performed by Lamb² for neutron scattering from nuclei of atoms bound in a solid, and the corresponding situation for photons has been considered by Mössbauer³ and by Visscher.⁴ A simplified derivation for the Mössbauer effect was presented by Lipkin⁵ and also by Weisskopf.⁶ Singwi and Sjölander⁷ used an approach developed by van Hove⁸ to arrive at the result. Interestingly, a discussion of this problem based on a classical picture for the process provided by Shapiro has also yielded an identical formula, a fact that has been remarked upon as being “unexpected” in the book on Mössbauer effect by Frauenfelder.⁹ The quantal calculations of f , referred to above, employ the Fock space (occupation number) representation and involve manipulations of Hermite polynomials and Bessel functions. Furthermore, some of the papers arrive at the exact result, despite certain approximations, which were later shown¹⁰ to be unnecessary on the basis of mathematical identities involving Hermite polynomials. The present derivation, using the device of coherent states, will avoid these unnecessary complications (using, as we shall see, nothing more complicated than Gaussian integrals) and the “mystery” of the unexpected agreement with the classical result will be clarified.

II. THE COHERENT STATE

The energy eigenstates of the proto-typical one-dimensional harmonic oscillator, described by the Hamiltonian

$$H = p^2/2m + \frac{1}{2}m\omega^2 x^2, \quad (3)$$

are best developed through the introduction of the annihilation and creation operators:

$$a \equiv (p - im\omega x)/\sqrt{2m\omega\hbar}, \quad (4a)$$

$$a^\dagger \equiv (p + im\omega x)/\sqrt{2m\omega\hbar}, \quad (4b)$$

whereby the normalized n th excited state (or the state of n quanta), $|n\rangle$, may be generated from the ground (or ‘vacuum’) state $|0\rangle$ through the repeated application of the creation operator:

$$|n\rangle = (a^\dagger)^n/\sqrt{n!}|0\rangle. \quad (5)$$

A set of nonstationary normalized states, known as coherent states,¹¹ first introduced by Schrödinger,¹² is obtained through a superposition of the occupation number states, thus

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle, \quad (6a)$$

where α is a complex number. These are minimum uncertainty wavepackets as the variance of the position and the momentum for an oscillator in such a state obtainable from

$$(\Delta x)^2 = \langle \alpha | x^2 | \alpha \rangle - \langle \alpha | x | \alpha \rangle^2 = \hbar/2m\omega \equiv l^2, \quad (6b)$$

$$(\Delta p)^2 = \langle \alpha | p^2 | \alpha \rangle - \langle \alpha | p | \alpha \rangle^2 = \hbar m\omega/2 = \hbar^2/4l^2, \quad (6c)$$

satisfy $\Delta x \Delta p = \hbar/2$. These states have been widely applied to nonlinear optics and laser physics.¹³⁻¹⁵ Their domain of usefulness has been extended to the description of the superfluid state,¹⁶ pion production,¹⁷ nuclear structure,¹⁸⁻²⁰ infrared problems in quantum electrodynamics,²¹ quantum theory of noise,²² plasma physics,²³ elucidation of classical correspondences,^{24,25} and to various other areas of physics.²⁶

These coherent states, which are in fact eigenstates of the annihilation operator a , belonging to the eigenvalue α , form a complete (actually an overcomplete) set:

$$\int |\alpha\rangle \langle \alpha| \frac{d^2\alpha}{\pi} = 1, \quad (7)$$

where the integration is over the plane of complex α . This "decomposition of the identity" is very useful, in that various operators, and in particular the density operator²⁷ ρ (which we shall need when performing thermal averages) can be conveniently incorporated into the formalism, through the so-called P representation:^{13,14}

$$\rho = \int \frac{d^2\alpha}{\pi} P(\alpha) |\alpha\rangle \langle \alpha|, \quad (8a)$$

which is but the density operator expressed in the basis of the coherent states. For oscillators in thermal equilibrium at a temperature T

$$P(\alpha) = (1/\langle n \rangle) e^{-|\alpha|^2/\langle n \rangle}, \quad (8b)$$

where the average occupancy $\langle n \rangle$ is given by

$$\langle n \rangle = e^{-\hbar\omega/kT} / (1 - e^{-\hbar\omega/kT}), \quad (8c)$$

k being the Boltzmann constant. These expressions for P are derived in both Refs. 13 and 14. The thermal average of any quantity evaluated in the coherent state basis is thus found by using $P(\alpha)$ as a weight function in determining the mean.

Another great advantage of the coherent state representation resides in the ease with which classical limits can be realized. Thus taking cognizance of the nonstationary character of the state, and putting in the time dependence appropriate for each stationary component ($|n\rangle$), we have, but for an irrelevant overall phase factor:

$$|\alpha\rangle_t = |\alpha(t)\rangle = |\alpha e^{-i\omega t}\rangle, \quad (9)$$

where the notation used above expresses the fact that with the elapse of time the coherent state retains its form, though the eigenvalue α of the annihilation operator a ,

which labels the state will have changed to $\alpha e^{-i\omega t}$. Now writing $\alpha = -i|\alpha|e^{-i\theta}$, it can be verified that

$$\langle \alpha | x | \alpha \rangle_t = 2|\alpha| \sqrt{\hbar/2m\omega} \cos(\omega t + \theta), \quad (10)$$

wherein the limit

$$\alpha \rightarrow \infty \text{ and } \hbar \rightarrow 0: 2|\alpha| \sqrt{\hbar/2m\omega} \rightarrow A, \quad (11)$$

yields, in an explicit manner, the classical correspondence for the time-dependent displacement for an oscillator, viz., $A \cos(\omega t + \theta)$, and moreover the quantal fluctuations expressed through the uncertainties [refer Eqs. (6a) and (6b)] go to zero in this limit.

III. CALCULATION OF f IN THE COHERENT STATE BASIS

In order to evaluate the matrix element occurring in f [see Eq. (1)] it is appropriate to express the exponent therein in terms of the normal mode coordinates $\{\xi_s\}$ [vide Eq. (2a)], and, furthermore, to convert these to the corresponding phonon annihilation and creation operators a_s and $a_s^\dagger$ [using relations analogous to Eqs. (4a) and (4b)]:

$$\xi_s = i l_s (a_s - a_s^\dagger), \quad (12)$$

where for the sake of brevity we have introduced $l_s = \sqrt{\hbar/2m\omega_s}$, with ω_s the modal angular frequency. The matrix element in question therefore becomes

$$\langle \{\alpha_s\} | e^{i p \cdot X / \hbar} | \{\alpha_s\} \rangle = \prod_s \langle \alpha_s | e^{-(p l_s / \hbar) c_s (a_s - a_s^\dagger)} | \alpha_s \rangle, \quad (13)$$

where the lattice state is being described through the multimode coherent state $|\{\alpha_s\}\rangle \equiv \prod_s |\alpha_s\rangle$. It is at this stage that the immense advantage of the coherent state basis begins to manifest itself; for realizing that the kets are eigenstates of the annihilation operator while the bras of the creation operators, the matrix element can be readily evaluated, provided one can move the a_s to the right of the $a_s^\dagger$ s. This is accomplished through the Baker-Hausdorff formula, which generalizes the formula $e^x \cdot e^y = e^{x+y}$ to the case when the exponents are operators which do not commute. In the special case where two operators A and B are such that their commutator $[A, B]$, is an ordinary number (viz. not an operator), one has:

$$e^{A+B} = e^A \cdot e^B \cdot e^{-(1/2)[A, B]}. \quad (14)$$

Thus the operator on the right-hand side of Eq. (13) may be more conveniently expressed, using the Baker-Hausdorff formula, as

$$e^{-(p l_s / \hbar) c_s (a_s - a_s^\dagger)} = e^{(p l_s / \hbar) c_s a_s^\dagger} \cdot e^{-(p l_s / \hbar) c_s a_s} \cdot e^{-(p^2 l_s^2 / \hbar^2) c_s^2 / 2}, \quad (15)$$

where use has been made of $[a, a^\dagger] = 1$. Accordingly,

$$\langle \alpha_s | e^{-(p l_s / \hbar) c_s (a_s - a_s^\dagger)} | \alpha_s \rangle = e^{-(p^2 l_s^2 / \hbar^2) c_s^2 / 2} \cdot e^{(p l_s / \hbar) c_s (\alpha_s^* - \alpha_s)}, \quad (16)$$

where we have exploited the fact that the coherent states are eigenstates of the annihilation operators. Implementing Eq. (16) in Eq. (13) we arrive at

$$\langle \{\alpha_s\} | e^{i\mathbf{p}\cdot\mathbf{X}/\hbar} | \{\alpha_s\} \rangle = e^{-2i\sum_s c_s (p_{\parallel s}/\hbar) \text{Im}(\alpha_s)} \cdot e^{-\sum_s (c_s^2/2) (p_{\parallel s}/\hbar)^2}, \quad (17)$$

which in turn [using Eq. (6b) for each mode] leads to

$$\begin{aligned} & |\langle \{\alpha_s\} | e^{i\mathbf{p}\cdot\mathbf{X}/\hbar} | \{\alpha_s\} \rangle|^2 \\ &= e^{-p^2 (\sum_s c_s^2 \bar{r}_s^2)/\hbar^2} \\ &= e^{-p^2 (\langle X^2 \rangle - \langle X \rangle^2)/\hbar^2}, \end{aligned} \quad (18)$$

a generalization of van Hove's result^{8,9} appropriate to a basis with $\langle \mathbf{X} \rangle \neq 0$. To obtain the thermal-averaged probability we use the P representation of the density matrix, taking the P -weighted mean, employing the weighting provided by Eq. (8b), to get

$$\begin{aligned} & \langle \langle \{\alpha_s\} | e^{i\mathbf{p}\cdot\mathbf{X}/\hbar} | \{\alpha_s\} \rangle \rangle_T \\ &= e^{-(1/2)\sum_s c_s^2 (p_{\parallel s}/\hbar)^2} \prod_s \int \frac{d^2\alpha_s}{\pi \langle n_s \rangle} e^{-|\alpha_s|^2/\langle n_s \rangle} \\ & \quad \times e^{-2i(p_{\parallel s}/\hbar)c_s \text{Im}(\alpha_s)}, \end{aligned} \quad (19a)$$

with

$$\langle n_s \rangle = e^{-\hbar\omega_s/kT} / (1 - e^{-\hbar\omega_s/kT}). \quad (19b)$$

The Gaussian integrals [over $\text{Re}(\alpha_s)$ and $\text{Im}(\alpha_s)$] are readily performed [refer to the Appendix] to yield

$$\begin{aligned} & \langle \langle \{\alpha_s\} | e^{i\mathbf{p}\cdot\mathbf{X}/\hbar} | \{\alpha_s\} \rangle \rangle_T = e^{-(1/2)\sum_s c_s^2 (p_{\parallel s}/\hbar)^2 (1+2\langle n_s \rangle)} \\ &= e^{-(R/2)\sum_s (c_s^2/\hbar\omega_s) \coth(\hbar\omega_s/2kT)}, \end{aligned} \quad (20)$$

where we have substituted the recoil energy R for $p^2/2m$. Now the summation over the phonon modes may be replaced by a frequency integral at the cost of introducing a spectral weight function $g(\omega)$, and consequently the Debye-Waller factor becomes

$$f = e^{-R/3N\int [g(\omega)/\hbar\omega] \coth(\hbar\omega/2kT) d\omega}, \quad (21)$$

where the expansion coefficients c_s , being frequency independent,⁹ have been put equal to $1/3N$ in deference to the normalization condition [Eq. (2b)].

In the Einstein model of the solid there is a single frequency ω_E and consequently the normalized frequency distribution being

$$g(\omega) = 3N\delta(\omega - \omega_E), \quad (22a)$$

the Debye-Waller factor becomes

$$f = e^{-[R/kT_E] \coth(T_E/T)}, \quad (22b)$$

where T_E , the characteristic temperature of the oscillator, is defined through $\hbar\omega_E = kT_E$. In the Debye model of the solid, on the other hand,

$$g(\omega) = (9N/\omega_D^3) \omega^2 \Theta(\omega_D - \omega), \quad (23a)$$

where Θ is the Heaviside step function, which is zero for negative values of its argument and unity for positive values. A change of variables, $x = \hbar\omega/kT$, enables us to obtain:

$$f = \exp \left[-\frac{3R}{2kT_D} \left[1 + 4 \left(\frac{T}{T_D} \right)^2 \int_0^{T_D/T} \frac{x dx}{(e^x - 1)} \right] \right], \quad (23b)$$

wherein the Debye temperature has been introduced via $\hbar\omega_D = kT_D$. This is the expression for the Debye-Waller factor as it appears in the literature. Thus we have demonstrated how the use of coherent states allows the derivation of these well-known results in a straightforward and graceful manner.

The classical derivation of the fraction of unshifted emission from an oscillating source, originally due to Shapiro, as discussed in Frauenfelder's textbook,⁹ depends on the basic notion that radiation from an oscillating source (vibrating with frequency ω) suffers Doppler shifts, and the amplitude of the unshifted line (of frequency Ω), and that of the Doppler wings (of frequencies $\Omega \pm \omega$, $\Omega \pm 2\omega$, etc.), could be obtained from the expansion and collection of corresponding terms from:

$$\exp[-i\Omega t + i\mathbf{K}\cdot\mathbf{X}_0 + i\mathbf{K}\cdot\mathbf{A}\cos(\omega t + \theta)], \quad (24)$$

which represents the wave emitted (with wave vector $\mathbf{K}$) by this source which is oscillating with amplitude A about its mean position $\mathbf{X}_0$ with a single frequency ω (as is the case with the Einstein solid). The amplitude of the unshifted line may be extracted from this expression through an integration with respect to the phase θ (a process that would integrate out to zero all the Doppler wings). It is interesting to observe that the result of this calculation agrees with that arrived at through the method based on the proper quantum treatment of the problem. That this is expected to be the case is made evident in our approach via the coherent state basis, which is ideally suited to expose just such classical correspondences, when it is recognized that Eq. (16), expressing the relevant probability amplitude, is the quantal analog of Eq. (24). If one were now to insert the time-dependent parametrization of α_s , as introduced through Eqs. (9) and (10), the exact parallelism of the procedure of averaging over A and θ for the classical calculation and the integration over the modulus and phase of the complex variable α in the quantum case provides the underlying reason for the mathematical equivalence between the two approaches, at least where $P(\alpha)$ is a function of $|\alpha|$, which holds when we are concerned with the system in thermal equilibrium.

Thus we have demonstrated the efficacy of the coherent state basis for the calculation of the probability for recoilless transitions in crystal lattices, through the ease and elegance of the derivation, and have also exhibited the inherent advantage in making contact with the classical description. It is suggested that the use of the coherent state basis in problems involving phonons could also prove to be advantageous in other problems of solid-state physics too.

APPENDIX

The process of thermal averaging when we are working in the coherent state basis involves carrying out integrations in the complex α -plane using the weighting corresponding to an equilibrium distribution. Thus, for example, carrying out the integration on the right-hand side of Eq. (19a) may be instructive. Writing $\alpha_s = \xi + i\eta$ the relevant integral becomes

$$\begin{aligned} & \int \frac{d^2\alpha_s}{\pi\langle n_s \rangle} e^{-|\alpha_s|^2/\langle n_s \rangle} e^{-2i(p l_s/\hbar) c_s \text{Im}(\alpha_s)} \\ &= \frac{1}{\pi\langle n_s \rangle} \int_{-\infty}^{+\infty} d\xi e^{-\xi^2/\langle n_s \rangle} \\ & \quad \times \int_{-\infty}^{+\infty} d\eta e^{-\eta^2/\langle n_s \rangle - 2i(p l_s/\hbar) c_s \eta}. \end{aligned}$$

Completing the square in the quadratic form in η in the second exponent in the integrand by writing it as

$$\frac{1}{\langle \eta \rangle} \left(\eta + i c_s \frac{p l_s}{\hbar} \langle n_s \rangle \right)^2 + c_s^2 \left(\frac{p l_s}{\hbar} \right)^2 \langle n_s \rangle,$$

and then displacing the variable of integration, $\eta' \equiv \eta + i c_s p l_s \langle n_s \rangle / \hbar$, both the integrals adopt the standard form of the Gaussian integral. Accordingly, the above integral yields:

$$e^{-c_s^2 (p l_s / \hbar)^2 \langle n_s \rangle}.$$

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SHAKESPEARE AND THE SECOND LAW OF THERMODYNAMICS

A good many times I have been present at gatherings of people who, by the standards of the traditional culture, are thought highly educated and who have with considerable gusto been expressing their incredulity at the illiteracy of scientists. Once or twice I have been provoked and have asked the company how many of them could describe the Second Law of Thermodynamics. The response was cold: it was also negative. Yet I was asking something which is about the scientific equivalent of: *Have you read a work of Shakespeare's?*

C. P. Snow, *The Two Cultures and the Scientific Revolution* (Cambridge, U. P., New York, 1959), pp. 15–16.