



Quantum elasticity in Debye solids

R. Giovanelli, A. Orefice^{*†}

Universita' di Milano, DI.PRO.VE., Via Celoria, 2, 20133 Milano, Italy

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Abstract

The rigid behaviour of a crystal lattice submitted to sudden and localized events (such as emission, absorption and scattering of quanta and/or light particles) is currently treated in terms of the well known Debye–Waller expression. Starting from first principles, and referring to solids described according to the Debye model, we present here a rigorous and general expression of the fraction of recoilless events, of which the Debye–Waller form is shown to represent only a first approximation, holding in the case of very low temperature solids.

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1. Introduction

In the emission, absorption and scattering processes of electromagnetic quanta, electrons, slow neutrons and light neutral atoms by single atoms of a material aggregate there exists a finite probability of occurring in a recoilless, non-dissipative way. This feature, implying a rigid behaviour of the lattice as a whole, is confirmed by the experimental evidence of many phenomena such as, for instance, the Mössbauer effect. It is nowadays well known that such a recoilless behaviour has a quantum nature, basically due to the finite probability P_{mm} of a perturbed quantum oscillator of remaining in its initial m th level of mechanical oscillation.

The fraction of recoilless events, for a crystal lattice submitted to short and localized impulses, is currently assumed to be given by the Debye–Waller (D–W) expression, whose original formulation [1,2] dates back to years when such a quantum nature was not yet clear enough.

Although the D–W expression was often reconsidered for many years [3–7], and although its quantum nature was clearly pointed out [8,9], its current accounts, even in standard textbooks [10,11], still present traces of this origin, particularly in what concerns its level of approximation. When, however, the Mössbauer effect (ME) was discovered in 1957, the adoption by Mössbauer himself (in order to evaluate the fraction of ME events) of the current D–W expression induced most researchers to believe that it was the exact and definitive one, forgetting and neglecting the involved and approximate character of its deduction. It was not noticed, in particular, that the problem could be faced in a much more satisfactory way thanks to a work published in 1958 by Kerner [12]. Kerner, indeed, had found the exact solutions of the Schrödinger equation in the case of harmonic oscillators submitted to forces with arbitrary intensity and time dependence. The corresponding exact expression of the probabilities P_{mm} , when inserted into the context of a Debye solid, could easily allow to get a new and general expression of the fraction of recoilless events. This fact was finally observed and published in Ref. [13]. At that stage, however, the comparison with the standard D–W expression still remained somewhat obscure.

^{*} Corresponding author. Tel.: +39 250316586; fax: +39 250316575.

E-mail address: adriano.orefice@unimi.it (A. Orefice).

In the present paper we get a double result. The first result is the deduction of the same general expressions obtained in Ref. [13] by a quite different method, giving them, therefore, a double warranty. The second, and most important, result is the demonstration that the standard D–W expression can be recovered as a particular, approximate case from our general expression, in the limit of very low temperatures and perturbation energies of the crystal lattice.

Aiming here at a fully self-contained account, we describe in Section 2 the Debye model underlying both our considerations and the standard ones.

Section 3 provides the exact form of the probability P_{mm} obtained, for a single quantum oscillator, both according to our new procedure and according to the previous one, based on Kerner’s work.

This expression of P_{mm} is inserted, in Section 4, into the context of a Debye solid (submitted to short and localized perturbations of arbitrary amplitude), thus obtaining our Eq. (18), which provides an exact and general expression of the fraction of elastic events in such a perturbed solid.

Finally, Section 5 is devoted to the demonstration that the standard Debye–Waller fraction is a particular, approximate case of our Eq. (18) (holding for very low crystal temperatures and perturbation energies), and to a comparison between the respective numerical results.

2. The Debye model

In order to fix ideas, we shall refer here to nuclear emission processes from a crystal lattice whose mechanical eigenoscillation spectrum involves frequencies $f_{osc} \approx 10^{13} \text{ s}^{-1}$, and to nuclei which, after a suitable excitation, decay with a life-time $T_{life} \approx 10^{-7} \text{ s}$, by the emission of γ radiation at frequencies $f_{rad} \approx 10^{19} \text{ s}^{-1}$.

Let us consider, now, a ‘regular’ crystal lattice composed of N atoms. Such a lattice admits, in general, an eigen-spectrum of $3N$ mechanical oscillations, ranging between a minimum (angular) frequency ω_{min} , corresponding to wavelengths of the order of the crystal linear dimensions, and a maximum frequency ω_D (the Debye frequency of the lattice) corresponding to a wavelength of the order of twice the equilibrium interatomic spacings. In the so-called Debye model the spectral distribution of the mechanical eigen-frequencies of the lattice is approximated as a continuum by means of a (Rayleigh–Jeans) function $S(\omega)$ proportional to ω^2 in the form

$$S(\omega) = 3 \frac{\omega^2}{\omega_D^3} \tag{1}$$

such that

$$\int_0^{\omega_D} S(\omega) d\omega = 1$$

having assumed $\omega_{min} \equiv 0$. In thermal equilibrium, the

average number of phonons with eigen-frequency ω is given by the expression

$$\bar{n}(\omega, T) = \frac{1}{\exp(h\omega/k_B T) - 1} \equiv \frac{1}{\exp\left(\frac{\omega \hbar \omega_D}{T T_D}\right) - 1} \tag{2}$$

where k_B is the Boltzmann constant, T is the temperature of the system and $T_D = \hbar \omega_D / k_B$ is the so-called ‘Debye temperature’ of the lattice.

We shall make the assumption that the mechanical energy levels, m , of the emitting oscillators are low enough to allow a harmonic approximation of their motion. We shall introduce moreover, for each oscillator involved in the emission process, the probability

$$P_m(\omega; T) = \frac{\bar{n}^m}{(1 + \bar{n})^{m+1}} \tag{3}$$

of starting from the m th energy level [14].

Let now the nuclei of crystal atoms be subject to random emission processes of γ -radiation with wave vector \mathbf{k} . By assuming a Gaussian space distribution of the oscillators, and taking the x -axis in the direction of the wave vector, a well-established tradition [6,7] provides the expression of the mean square path of an atom along such a direction in the form

$$\langle x^2 \rangle = \frac{3\hbar^2}{Mk_B T_D} \left[\frac{1}{4} + \left(\frac{T}{T_D} \right)^2 \int_0^{T/T_D} dx \frac{x}{e^x - 1} \right] \tag{4}$$

where M is the mass of the atom. Since, the emission of a quantum with a momentum $\hbar \mathbf{k}$ imparts an equal and contrary impulse to the emitting nucleus, and $E_C = (\hbar k)^2 / 2M$ represents the free recoil energy of the emitter, we find it useful to introduce the adimensional ratio

$$\varepsilon_D = \frac{E_C}{\hbar \omega_D} \tag{5}$$

thus obtaining the relation

$$k^2 \langle x^2 \rangle = 6\varepsilon_D \left[\frac{1}{4} + \left(\frac{T}{T_D} \right)^2 \int_0^{T/T_D} dx \frac{x}{e^x - 1} \right] \tag{6}$$

whose role will turn out to be crucial later on in the present paper.

The values of the parameter ε_D , representing a measure of the intensity of the sudden shock due to the emission process, typically range between $\varepsilon_D = 0.05$ in the case of the Mössbauer effect (ME) in Fe^{57} and $\varepsilon_D = 3.5$ in the case of the ME in Zn^{67} .

3. Elastic processes of a single quantum oscillator

Let us consider now, to begin with, a single linear oscillator, with mass M , whose classical behaviour is described by an equation of the form

$$M\ddot{x}_c + kx_c = 0 \tag{7}$$

where $x_c(t)$ is the classical position, oscillating around $x_c = 0$, and kx_c is the elastic restoring force. The standard stationary solutions of the corresponding Schrödinger equation may be written in the form

$$\psi_m(x) = N_m \exp\left(-\frac{\alpha^2 x^2}{2}\right) H_m(\alpha x) \quad (m = 0, 1, 2, \dots) \quad (8)$$

where

$$\alpha = \sqrt{\frac{M\omega_c}{\hbar}}; \quad \omega_c = \sqrt{\frac{k}{M}}; \quad N_m = \sqrt{\frac{\alpha}{2^m m! \pi^{1/2}}}$$

and H_m is an Hermite polynomial of order m .

Let the oscillator undergo a short and sudden impulse, due to the emission of a γ -quantum with wave vector \mathbf{k} , along which we assume the direction of the x -axis. If $\psi_m(x)$ is the initial wave function of the oscillator, starting from the m th stationary state, the wave function after the emission process may be written, with excellent approximation, in the form

$$\psi_f \equiv \psi_m \exp(i\mathbf{k}r) \equiv \psi_m \exp(ikx) \quad (9)$$

so that (making use of Ref. [15]) the probability of finding the oscillator in the same m th state after emission takes the form

$$\begin{aligned} P_{mm} &= \left| \int dx \psi_m^* \psi_f \right|^2 = \left| \int dx |\psi_m|^2 \exp(ikx) \right|^2 \\ &= N_m^4 \left| \int_{-\infty}^{\infty} dx \exp(-\alpha^2 x^2 + ikx) H_m^2(\alpha x) \right|^2 \\ &= \exp\left(-\frac{k^2}{2\alpha^2}\right) \left| L_m\left(\frac{k^2}{2\alpha^2}\right) \right|^2 \end{aligned} \quad (10)$$

where

$$\frac{k^2}{2\alpha^2} = \frac{(\hbar k)^2 / 2M}{\hbar \omega_c} \quad (11)$$

L_m is a Laguerre polynomial of order m , and the asterisk (*) denotes a complex conjugation.

The same result could, indeed, be recovered from the exact solutions of the Schrödinger equation obtained in Ref. [12] for an oscillator submitted to a space-independent force of arbitrary amplitude. Starting from Ref. [12], in fact, the probability P_{mm} of an elastic ($m \rightarrow m$) process was shown, in Ref. [13], to evolve with time according to the exact expression

$$P_{mm} = P_{mm}[\varepsilon(t)] = \exp[-\varepsilon(t)] |L_m[\varepsilon(t)]|^2 \quad (12)$$

where

$$\varepsilon(t) = \frac{E(t)}{\hbar \omega_c} \quad (13)$$

and $E(t)$ is the classical value of the energy reached by the oscillator (under the action of the force) at the time t . If the classical oscillator of Eq. (7) is submitted to a constant and

uniform force \mathbf{F} for a time τ , the value of the function $\varepsilon(t)$ may be expressed in the form [16]

$$\varepsilon(t) = \frac{2}{(2\pi q)^2} \frac{E_c}{\hbar \omega_c} \left[1 - \cos\left(2\pi q \frac{t}{\tau}\right) \right] \quad (14)$$

where $q = \tau / t_c$, $t_c = 2\pi / \omega_c$ and $E_c = (F\tau)^2 / 2M$. The final value $\varepsilon(t = \tau)$ turns out, therefore, to be close to zero when $\tau \gg t_c$, while it reaches the maximum possible value

$$\varepsilon_{\text{Max}} = \frac{E_c}{\hbar \omega_c} \quad (15)$$

when the force acts for a time shorter than, say, $0.1 t_c$. Such a condition is certainly verified in the case of the emission of a single γ -quantum, whose momentum $\hbar k$ is equal and opposite to the impulse $\mathbf{F}\tau$ imparted to the emitter, so that

$$\varepsilon(t = \tau) = \varepsilon_{\text{Max}} = \frac{(\hbar k)^2 / 2M}{\hbar \omega_c} \quad (16)$$

a value which, inserted into Eq. (12), leads once more to Eq. (10), whose demonstration is, therefore, warranted by two different and independent methods.

4. Elastic processes of a crystal lattice

The physical features characterizing a single quantum oscillator may now be extended to short and sudden elastic processes occurring at single atoms of the crystal lattice considered in Section 2, represented as an aggregate of mechanical oscillators with frequencies belonging to the spectrum $S(\omega)$ of Eq. (1). We shall assume isotropic emission processes, and let θ (with $0 \leq \theta \leq \pi$) be the angle between the momentum $\hbar k$ and the instantaneous oscillation direction of the emitting nucleus, thus reducing the transferred energy ratio of Eq. (16) to

$$\left(\frac{(\hbar k)^2 / 2M}{\hbar \omega_c} \right) \cos^2 \theta \equiv \frac{\varepsilon_D \cos^2 \theta}{\omega / \omega_D} \quad (17)$$

By making use of the spectral distribution (1), the probability of a recoilless process of a lattice at temperature T may then be written in the compact form

$$\begin{aligned} P_{\text{el}}(T; \varepsilon_D) &= \frac{1}{\pi} \int_0^\pi d\theta \int_0^1 d\left(\frac{\omega}{\omega_D}\right) \left[3 \left(\frac{\omega}{\omega_D}\right)^2 \right] \\ &\quad \times \sum_m P_m(\omega; T) P_{mm}(\omega; \theta; \varepsilon_D) \end{aligned} \quad (18)$$

where P_m is provided by Eq. (3), the sum \sum_m is extended to the energy levels involved in the emission process at temperature T , and, from Eqs. (12) and (17),

$$P_{mm}(\omega; \theta; \varepsilon_D) = \exp\left(-\frac{\varepsilon_D \cos^2 \theta}{\omega / \omega_D}\right) \left| L_m\left(\frac{\varepsilon_D \cos^2 \theta}{\omega / \omega_D}\right) \right|^2 \quad (19)$$

Fig. 1 presents the plots the probability $P_{\text{el}}(T, \varepsilon_D)$ of a recoilless process as a function of the ratio T/T_D , for a typical value ($\varepsilon_D = 1$) of the shock energy ratio ε_D . As shown

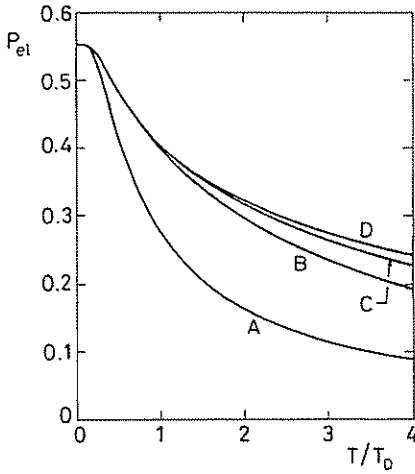


Fig. 1. Plot versus T/T_D of the probability P_{el} of recoilless emission according to Eq. (18), for $\epsilon_D=1$. The sum over the mechanical energy levels m is performed, respectively, with $m=0$ alone in curve A, and over the values $m \leq 4$ in curve B, $m \leq 8$ in curve C, $m \leq 12$ in curve D.

by Fig. 1, a sum over the values $m \leq 8$ turns out to provide, in the usual range of the relevant physical parameters ($T/T_D \leq 4$, $\epsilon_D \leq 4$), adequately converging values of the plotted function.

A simplified, but still significant, approximation of Eq. (18) may be obtained by means of the so-called Einstein model, where the oscillation spectrum is reduced to a single average ‘Einstein frequency’ $\omega_E \approx \alpha \omega_D$, with a suitable choice of the parameter α : a careful best fit suggests the assumption of $\alpha \approx 0.7$. In such an approximation, the probability of an elastic process may be written in the simple form

$$P_{el}^{(E)}(T, \epsilon_D) = \frac{1}{\pi} \int_0^\pi d\theta \sum_m P_m(\omega_E, T) P_{mm}(\omega_E; \theta; \epsilon_D) \quad (20)$$

5. Comparison with the Debye–Waller approximation

If the temperature T of the lattice is low enough to involve only, or mainly, oscillators in the ground state ($m = 0$), we may assume, in Eq. (18), $P_0 \approx 1$ (and, therefore, $P_m \approx 0$ for $m > 0$) thus limiting the main contribution to the term containing the $0 \rightarrow 0$ probability alone:

$$P_{00}(\omega; \theta; \epsilon_D) = \exp\left(-\frac{\epsilon_D \cos^2 \theta}{\omega/\omega_D}\right) \quad (21)$$

Observing that, in the case of a quantum oscillator in the ground state, its space distribution takes up the Gaussian form

$$|\psi_0(x)|^2 = \frac{\alpha}{\sqrt{\pi}} \exp(-\alpha^2 x^2) \quad (22)$$

whose flexes are placed at

$$x = \pm \Delta x = \pm \frac{1}{\alpha \sqrt{2}} = \pm \sqrt{\frac{\hbar^2}{2M\hbar\omega_c}} \quad (23)$$

we have

$$k^2(\Delta x)^2 = \frac{(\hbar k)^2/2M}{\hbar\omega_c} = \frac{\epsilon_D}{\omega/\omega_D} \quad (24)$$

The reasonable assumption [17]

$$k^2(\Delta x)^2 \cong k^2 \langle x^2 \rangle \quad (25)$$

with such a term provided by Eq. (6) (based itself, as we may recall, on a Gaussian space distribution), leads now to perform, within Eq. (18), the replacement

$$\sum_m P_m(\omega, T) P_{mm}(\omega; \theta; \epsilon_D) \rightarrow \exp(-k^2 \langle x^2 \rangle \cos^2 \theta) \quad (26)$$

It may be observed that, in such a limit, the replaced term loses its ω dependence, and depends on T through the function $\langle x^2 \rangle$, as shown by Eq. (4).

Recalling then that

$$\int_0^1 d\left(\frac{\omega}{\omega_D}\right) \left[3\left(\frac{\omega}{\omega_D}\right)^2\right] = 1$$

we get

$$P_{el} = P_{DW}(T, \epsilon_D) = \frac{1}{\pi} \int_0^\pi d\theta \exp(-k^2 \langle x^2 \rangle \cos^2 \theta) \quad (27)$$

which is the standard Debye–Waller expression. By observing moreover [15] that

$$\begin{aligned} \int_0^\pi d\theta \exp(-a \cos^2 \theta) &= \int_0^2 dt \frac{\exp(-at/2)}{\sqrt{t(2-t)}} \\ &= \pi \exp(-a/2) I_0(a/2) \end{aligned} \quad (28)$$

where I_0 is the modified Bessel function of order 0, we may put Eq. (27) in the alternative and compact form (to be compared with our general expression (18))

$$P_{DW}(T, \epsilon_D) = \exp\left(-\frac{k^2 \langle x^2 \rangle}{2}\right) I_0\left(\frac{k^2 \langle x^2 \rangle}{2}\right) \quad (29)$$

with $k^2 \langle x^2 \rangle$ provided by Eq. (6). We present, in Fig. 2, the plots (versus T/T_D) of the functions P_{el} and P_{DW} provided, respectively, by Eqs. (18) and (29) (i.e. by our general expression and by the standard D–W approximation), for some typical values ($\epsilon_D = 0.05; 0.3; 3.5$) of the parameter ϵ_D , representing a measure of the intensity of the sudden shock due to the emission process.

It is clearly seen that the D–W expression, because of its limitation to the (mechanical) ground state of the emitting nuclei, systematically underestimates the fraction of events leaving the lattice in the same initial energy level, thus underestimating the quantum rigidity of the solid. The differences between P_{el} and P_{DW} , almost negligible at low values of the emissive shock, are seen to become

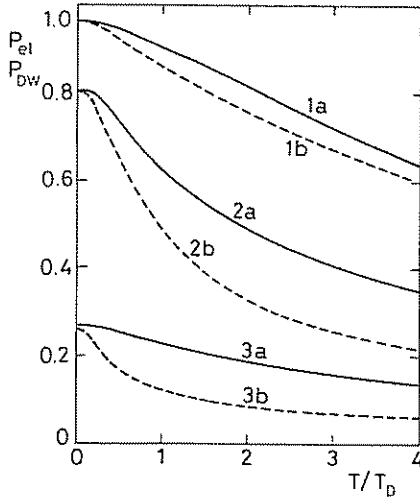


Fig. 2. Comparison of the probabilities of recoilless emission P_{et} (curves 1a, 2a, 3a) and P_{dw} (curves 1b, 2b, 3b) plotted versus T/T_D according, respectively, to Eqs. (18) and (29). Curves 1a, 1b correspond to a shock parameter $\varepsilon_D=0.05$; curves 2a, 2b to $\varepsilon_D=0.3$; curves 3a, 3b to $\varepsilon_D=3.5$.

increasingly appreciable with increasing values of both ε_D and T ; this is, in conclusion, the most noticeable contribution of the present paper, providing, therefore, a new and more general approach to the problem of quantum rigidity.

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