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Highly localized quantum elastic processes in solids

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Abstract

Most experimental observations of quantum effects in solid state physics deal with the stationary and collective behaviour of large aggregates of elementary mechanical oscillators. The analysis, however, of sudden and highly localized processes occurring at single oscillators may lead to a more complete understanding of the coupling between microscopic and macroscopic quantum properties. © 2002 Elsevier Science B.V. All rights reserved.

1. Quantum rigidity

Let us consider the scattering of, say, X quanta, or electrons, or neutrons, or light neutral atoms, by an aggregate of mechanical oscillators. The process occurs as a sequence of sudden, highly localized events, involving in principle one single oscillator at a time. From a classical point of view, the probability of a non-dissipative scattering is vanishingly small: a perfectly elastic behaviour may only be conceived, indeed, in the limit of perfectly rigid microscopic bonds. From a quantum point of view, on the other hand, there exists a finite probability for the system to be left in its initial level of mechanical oscillation [1]. This feature is confirmed by many experimental evidences.

In Compton's experiment of X-ray scattering by the atoms of a graphite crystal [2] the observed scattered radiation spectrum includes two different peaks: the first peak, corresponding to the standard Compton effect, is upshifted (in wavelength) with respect to the incident radiation, and is currently explained as a photon scattering by the most external, and almost free, orbital electrons; the second peak, in its turn, is centered around the incident wavelength, and is attributed to strongly bound orbital electrons. In this second case the entire atom behaves like a unique rigid body, with an effective scattering mass thousands of times larger than the electron mass.

When an incident particle is scattered by a diatomic molecule, such a molecule often behaves like a rigid body, preserving its initial internal energy level [3,4]. After having mixed, for instance, a diatomic gas with a monoatomic one at a higher temperature, the thermalization time of the two gases is seen to be much shorter than the excitation time of the vibrational internal levels of the diatomic molecules: a clear proof that the diatomic molecule has a high probability of exhibiting a rigid behaviour.

Analogous features were described in Ref. [5], concerning the case of 4-atomic molecules.

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When the number of atoms of the target is very large, as in the case of a macroscopic crystal lattice, a local impulsive process (scattering, absorption or emission) affecting a single nucleus may occur *in a phononless way*, without any energy exchange at all, thus implying a perfectly rigid and elastic behaviour of the whole lattice: the most spectacular case is provided [6,7] by the Mössbauer effect (ME), where no recoil is observed in the emitting/absorbing nuclei, and no Doppler shift is presented by the relevant spectral lines. The fraction f of oscillators undergoing an elastic process may be written in the Debye–Waller (D–W) form [8]

$$f = \begin{cases} \exp\left[-3\dfrac{E_R}{2k_B T_D}\right], & \text{for } T \approx 0\ \mathrm{K}, \\ \exp\left[-6\dfrac{E_R T}{k_B T_D^2}\right], & \text{for } T \gg \dfrac{T_D}{2}, \end{cases} \tag{1}$$

where k_B is the Boltzmann constant, T is the temperature of the system, T_D is the Debye temperature and $E_R = p^2/2M$ (being M the atomic mass and p the transferred momentum) is the free recoil energy.

Phononless transitions, indeed, had already been observed (see, for instance, Refs. [9–15]) several years prior to Mössbauer's discovery, as narrow lines in absorption and emission spectra associated with electron transitions between states pertaining to ion structural defects (the so-called *impurity centers*) in crystals. After the absorption/emission of an optical photon there is in fact a finite probability that the recoil potential energy is not dissipated as heat in the crystal, and that the system is left in its ground state of mechanical vibration. Gurney and Mott [16] suggested that an electron, raised to the excited state by light absorption, can be thrown into the conduction band by lattice vibrations before it has the opportunity to return to the ground state, and that such non-radiative transitions vary with temperature according to a D–W-like expression. The most recent form of this expression, applied to very low temperature lattices, is known as the Huang–Rhys *factor* [17]. Large values of f are frequently observed in lines of laser transitions in ruby and in some impurity transitions in semiconductors. In such instances one typically observes only the sharp zero-phonons transitions, the multi-phonon structure being lost in the noise. Zero-phonon transitions, in which a photon is absorbed with no transfer of energy to the lattice phonons, appear as a line which, in an ideal crystal at absolute zero, would have a breadth determined only by the lifetime of the excited electronic state [18]. The difficulty, however, of distinguishing the signal from the noise when operating on low-energy photons (with line-widths of the fluorescence diagrams many orders of magnitude larger than the characteristic Mössbauer peaks) made the experimental results much less spectacular and self-evident than the ME. The rigidity implied by these results did not attract, therefore, the attention it could deserve.

The rigid behaviour characterizing, in conclusion, so many aspects of physical reality (from elementary to very complex aggregates of mechanical oscillators) cannot find its justification in forces of simply electrostatic nature, such as the ones assumed to act between the atoms of a crystal. One could suspect, moreover, that non-dissipative, recoilles processes which do not perturb a microscopic oscillator (behaving as if it were rigidly bound to its center of oscillation), call into question the very applicability of the uncertainty principle: no destruction of knowledge [19] of the oscillator *momentum*, indeed, is caused by a measurement of its *position* based on the detection of a particle scattered without perturbing the oscillator itself.

The present Letter aims at pointing out, in this interesting and somewhat neglected field of discussion, the physical implications both of the experimental results and of their current treatment and interpretation.

2. Quantum rigidity in momentum transmission

The frequency spectrum of a macroscopic lattice ranges between a minimum frequency $\omega_{\min}$, corresponding to wavelengths of the order of the crystal linear dimensions, and a maximum value $\omega_{\max} \cong \omega_D$ (where $\omega_D = k_B T_D/\hbar$ is the Debye frequency of the lattice), corresponding to wavelengths of the order of the average interatomic spacings. In the presence of an impulsive solicitation one could expect, because of the large number of available low-energy levels, the excitation of at least the lower part of the frequency spectrum, and therefore an energy exchange with at least the less energetic internal oscillation modes, covering a wide region of the crystal itself [20]. The observed evidence, however, that sudden and localized processes may occur without any energy exchange at all, leads to a striking contradiction: on the one hand, the fact that low energy levels are not excited appears to be a proof that an atom undergoing the localized elastic process has not the time to "communicate" with distant atoms of the lattice; on the other hand, the fact that no recoil is exhibited by the single atoms involved in the elastic process suggests that the imparted momentum is immediately shared with the whole lattice.

Let us consider, for instance, the scattering of thermal neutrons ($v \approx 10^6$ cm/s) from heavy nuclei. The crossing time Δt of the neutron through the region defined by the range of the nuclear forces ($\approx 10^{-13}$ cm) may be estimated around 10^{-19} s [21]. Although a mechanical "message" between atoms and lattice can only travel at sound's speed, let us admit for a moment an electromagnetic nature of such a "message" [22]. In the time Δt an electromagnetic signal may cover a distance of the order of 3×10^{-9} cm, even lower than the average spacing between two atoms in a molecule or in a crystal lattice, which is around 1 Å. Not even an electromagnetic message may allow, therefore, the transmission of the (hypothetical) force required to provide the rigidity implied by the absence of recoil.

Let us pass now to a crystal impurity, consisting even of a single atom. Differently from the aforementioned case of (low-energy) electron transitions in color centers, let us consider a high energy process, where the nucleus itself of the impurity is submitted to a strong impulsive solicitation. This solicitation raises anharmonic oscillations whose frequencies do not belong to the eigen-spectrum of the lattice, which should remain therefore, in principle, strictly localized. Let us consider, for instance, in order to fix ideas, the case of an atom of ^{40}K, excited by a high energy neutron impressing to the nucleus a recoil energy of, say, 800 eV [23]. The perturbation causes a considerable radiation damage and local heating, including the expulsion of the atom from its position in the lattice and a (fusion-like) breaking of the crystal bonds, followed by a new crystallization of the damaged zone. All these features must necessarily occur in a time not longer than 1/10 of the life-time (≈ 4 ns) of the excited nuclear state: after 4 ns, in fact, a 29.4 KeV γ-quantum is emitted (and may be observed) which is *not affected* by the previous perturbations, as proven by the fact that the fraction of nuclei undergoing ME is provided by the D–W expression applied to the unperturbed crystal. According to Ref. [24] the recoil energy diffuses out of its initial localization in a time of the order of 10^{-13} s, i.e., much before the Mössbauer emission. In such a time, however, an acoustic message may only travel through 1 Å: a mechanical perturbation would not even have time enough to reach the closest atoms in the lattice.

This apparent disappearance of the local oscillations has been attributed to various mechanisms [25], such as, for instance, the creation and re-absorption of localized phonons, with an overall energy balance close to zero. This interpretation leaves, however, an even more entangled situation. In order to accept it we should renounce, in fact, to the idea of photon emission, in favour of a wave-train behaviour allowing to exchange momentum all along the life-time of the excited nuclear state. It is quite clear, moreover, that if even a small part of the wave-train were submitted to a perturbation, new harmonics would arise which would strongly affect the photon statistics. Ref. [25] suggests the existence of a deep connection between quantum rigidity and zero-point oscillation energy: in the localized oscillations the phonons would be, somehow, quickly eliminated, leaving a strongly deformed zero-point frequency spectrum. According to Ref. [26], however, this suggestion appears to imply the enhancement, due to localized perturbations, of any kind of elastic interaction (ME included), in sharp contrast with the D–W expression written (as mentioned later on in the present Letter) in terms of atomic mean square deviation from the equilibrium position.

3. Quantum rigidity in resonant absorption and reflection of γ radiation

Let us consider, in order to fix ideas, a crystal of ^{57}FeBO$_3$. Were the nuclei of ^{57}Fe in an excited state, they would emit (with a decay-time $\tau \cong 9 \times 10^{-7}$ s) photons at $E_0 \cong 14.41$ KeV. Let us imagine now to launch against such a (*not* previously excited) crystal a flux of photons endowed with energies centered around E_0 and generated, for instance, by means of synchrotron radiation sources [27]. The D–W expression predicts a large fraction of resonant Mössbauer absorptions, which are made possible by the rigid behaviour, within the crystal lattice, of the atoms containing the absorbing nuclei: the absence of this rigidity would compromise, in fact, the resonance itself. No absorption process occurs, however, if the γ radiation hits the crystal at Bragg angles, since it undergoes, in this case, a resonant elastic reflection. For each quantum, the probability (often approaching 100%) of elastic reflection depends on the stationary wave-field distribution "anchored" at the crystal lattice for the entire duration of the wave train accompanying the impinging quantum itself. According to quantum mechanics, in fact, the wave propagation occurs in the general terms of classical electromagnetism, thus implying the formation, due to the collective behaviour of the crystal, of a stationary pattern (generated by the interference of the incident and reflected waves) where the nodes of the electric field in correspondence with the nuclei reduce almost to zero the probability of any absorption process [28]. Should the well ordered structure of such a pattern disappear, a total resonant absorption of the Mössbauer fraction of impinging photons predicted by the D–W expression would occur. Clearly enough, the stationary character of the pattern allowing elastic reflection may only be warranted by the rigidity of the lattice at which the pattern itself is "anchored". Another basic requirement is the rigorous coherence of each wave-train: a property which is only possible for single-photon wave-trains. At high frequencies, in fact, no coherent source may ensure a coherent, laser-like radiation, and the photons of a beam turn out to be uncorrelated. Each photon can only be guided along the interferential pattern by its own wave-train, and the apparent role of this monophotonic wave is to modify (without energy exchange) the statistics of the zero-point electrical oscillations internal to the nuclei.

Any resonant elastic absorption and reflection process require, in conclusion (in different forms) the quantum rigidity of the atoms involved in the interaction.

4. Experimental aspects of the rigidity problem

The phononless character of elastic processes such as the ME was always directly inferred, so far, from the properties of the relevant γ radiation spectra. These properties were neat and reliable enough to allow many kinds of measurements endowed with an unprecedented precision, requiring, moreover, rather simple and low-cost equipments. That is why the ME was considered to provide much more an excellent diagnostic tool in the most disparate branches of physics and chemistry than an interesting (and, in fact, fundamental) topic in itself. It is nowadays possible to utilize, however, for a more complete understanding of the coupling between microscopic and macroscopic properties of a crystal lattice, many recently acquired experimental techniques and facilities, allowing extremely high space and time resolutions [29]. The timing properties necessary for time-resolved nuclear spectroscopy are of the order of some ps, with divergence for an X-ray beam of the order of some µrad.

A possible way to verify the existence (or the absence) of an acoustic recoil phonon due to the absorption/emission of a γ quantum could be based on the piezo-electric creation, in a suitable crystal, of electric charges due to an acoustic wave. The detection of such a wave (composed of a small number of phonons) within a length, at most, of some tens of Ångströms, would require of course very high time and space resolutions. The dynamics of low-intensity phonons has been explored, so far, by means (for instance) of superconducting (Josephson) tunnel junctions [30,31]. Since, in any case, the momentum is finally transmitted to the center of mass of the crystal, the piezo-electric approach would shed light on the details of this transmission.

The techniques nowadays employed for the detection of transient phonons in crystals [32] could allow to detect the phonons generated by the inelastic interaction of single nuclei with resonant X (or γ) radiation, and to point out the phonon absence when the interaction turns out to be elastic. The inelastic interaction of single γ photons with Mössbauer nuclei was observed by Seto et al. [33] and by Chumakov et al. [34] and seen to be accompanied by single, high-energy phonons [35].

Concerning, finally, the measurement of the length of the wave-train associated with a single γ photon, and the self-interferential, single-photon character of the process, many fundamental experimental results, involving in an essential way the rigidity of a fraction of the atoms of a lattice, were summarized in Ref. [28]. An analogous behaviour was observed by means of the so-called EXAFS (extended X-ray absorption fine structure) method [36], applied to the de Broglie waves associated to the electrons extracted, for instance, by an X photon from the K level of an atom. According to the standard quantum approach, these waves base their own reflection (and therefore the electron extraction) on a previous, virtual (i.e., particleless) test of the rigidity (described by a suitable D–W probability factor) of the nearest atoms. This feature is testified by the measurement of the oscillations (around the average value) of the X-ray absorption coefficient of the involved atoms, for energies beyond the minimum excitation threshold of the K level. High precision results were obtained in this field by means of the most recent X-radiation sources (the so-called *undulators*), and are accounted for in Ref. [37].

5. Theoretical aspects of the rigidity problem

The impulsive processes undergone by single atoms of an aggregate of oscillators without modifying the energy of its internal eigen-states still constitute a challenging theoretical problem.

The observed rigidity is often attributed to the role of the so-called reciprocal vector g, a quantity (defined as the product of two suitable vectors characterizing a reticular plane of a crystal) originally employed for the description of light reflection from such a plane [38,39]. When, successively, experiments concerning X-rays were performed, the use of the vector g was maintained [40], as if the recoil due the radiation could still be neglected, thus associating to the crystal a rigidity on whose nature no further question was raised. It was simply assumed as a matter of fact in all cases of elastic scattering, and rejected in the inelastic fraction of cases predicted by the D–W expression. The rigidity suggested by the experimental results is currently, more or less explicitly attributed to the vector g by assuming it as much more than a mere abstraction: in all scattering events, for instance, the quantity $\hbar$g is often assumed to contribute in an essential way the overall momentum balance [41]. It is clear, however, that such a vector cannot be understood as an effective physical reality.

In view of a more serious and consistent treatment of the questions raised by quantum rigidity, let us consider now the possible approaches to the problem.

The dynamics of a crystal lattice may be classically described either by means of the *local* variables, expressing the displacements of the single atoms from their equilibrium positions, or by means of the *normal* variables, provided (under the condition of leading to separable dynamical equations) by suitable linear superpositions of the full set of the local variables [42]. Although the normal variables give a description of the stationary oscillations of the whole crystal around its equilibrium configuration, they always allow, in classical mechanics, a final return to the local variables which describe the individual atomic behaviour [43, 44]. Localized, impulsive events may therefore be explicated in terms of the *local* atomic coordinates after a preliminary set up and solution in terms of the *normal* variables. Classical mechanics, however, does not admit the possibility of impulsive processes leaving the full set of internal eigen-modes in its initial state. Quantum mechanics, on the contrary, is well fit to envisage a finite probability of a rigid behaviour leaving the interacting system in its initial level of mechanical oscillation. A simple example is provided by the case of a single particle with mass M oscillating, with a (single) frequency ω, in a one-dimensional external harmonic potential $kx^2/2$, solicited by an external time-dependent force $F(t)$. In the quantum treatment [45] (characterized, in the absence of perturbations, by a discrete set of levels $n = 0, 1, 2, \ldots$) a transition $n \to n$, leaving the system unperturbed, occurs with a probability

$$P_{n\to n} = \exp\left[-\varepsilon_0(t)\right]\left|L_n\left(\varepsilon_0(t)\right)\right|^2, \qquad (2)$$

with

$$\varepsilon_0(t) = \frac{1}{2M\hbar\omega}\left|\int_0^t F(t)\exp(-i\omega t)\,dt\right|^2. \qquad (3)$$

where L_n is a Laguerre polynomial. If the oscillator is initially moving in the ground state ($n = 0$), its wave function is a Gaussian, and $P_{0\to0}$ may be written in the form

$$P_{0\to0} = e^{-k^2\langle x^2\rangle_0}, \qquad (4)$$

where $\langle x^2\rangle_0$ is the mean square deviation of the position of the oscillator from its equilibrium value in the ground state.

According to Eq. (2) a single oscillator may react to a sudden impulse as a perfectly rigid body (i.e., with a final state coinciding with the initial one), *as allowed by the external harmonic potential, which behaves like* *an infinite absorber of momentum*: a quite academic, indeed, and not very realistic Ansatz.

The more realistic case, however, of an aggregate of mechanical oscillators at temperature T, where a single atom undergoes an impulsive solicitation, appears to be much less simple and academic. Eq. (2) turns out to be replaced [46], in this case, by an expression of the form

$$P_{\bar{n}\to\bar{n}} \approx \exp\left[-R\sum_{=1}^{3N}\frac{a^2}{\hbar\omega}(2n\ +1)\right], \tag{5}$$

where n is the number of phonons at thermal equilibrium in the th mode of oscillation:

$$n\ =\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^{-1}. \tag{6}$$

$\bar{n}$ represents the set of n defining the state of the crystal lattice, R is the recoil energy of a single free atom subject to the perturbing impulse, and a^2 is the weight of such a mode on the motion of the perturbed atom. According to Eq. (5), the probability $P_{\bar{n}\to\bar{n}}$ of a transition leaving the whole aggregate in the initial state is appreciable only in the absence of low energy phonons, in agreement with the previously mentioned observation that no low frequency of the lattice is found to be excited in elastic events. The choice of the physical model allowing to evaluate the weighting factor a^2 is therefore a crucial point of any theoretical approach.

An impulsive solicitation may be due, for instance, to the emission (induced by the absorption of an X quantum) of an electron from the K level of an atom. In this case, the effect of thermal disorder on the EXAFS spectrum [36] is generally taken into account by a parameter (the so-called "EXAFS D–W factor") which has a different value for each shell around the absorber atom. When the distribution function of the surrounding atoms is not a Gaussian, the relevant D–W factor is very close to Eq. (5) with $n \neq 0$.

In the limit $T \to 0$, when all the $3N$ values of n defining the vibration spectrum of the crystal tend to zero, a $0 \to 0$ transition is the only possible elastic one, and Eq. (5) may be put, within the Debye model, in a form analogous to Eq. (4). This form coincides with the archetypal one of the Debye–Waller expression [47], which, for an assembly of oscillators at temperature T, may be reduced, by means of tedious but straightforward passages, to the form (1). The role played by the ground state oscillation level (because of its impossibility to lose any amount of mechanical energy), may provide, indeed, a plausible explication of the observed absence both of spectral Doppler shift and of atomic recoil. No answer is provided, however, by the D–W expression to the problem of the mechanism allowing the instantaneous propagation to the whole solid (or even to the closest atoms) of a local perturbation when any local energy exchange is forbidden.

Even the very formalism to be adopted is far from being self-evident. Although, indeed, the use of the local variables may appear to be the most suitable to describe the recoil of a single atom in a macroscopic lattice and the instantaneous rearrangement of the neighboring atoms, any attempt to face the problem by means of these variables has encountered, up to now, almost insurmountable difficulties. In Ref. [48], for instance, formally correct and self-contained results were obtained in terms of local variables, but turned out to be practically useless from the point of view of numerical computation and handling.

On the other hand, as far as normal variables are concerned, although they turn out to allow an easy quantization of a crystal, they unavoidably view it as a unique physical system submitted to stationary, collective oscillations, in sharp contrast with the features of a local impulsive event. In the quantum case, moreover, no final return is possible to the local variables required by the treatment of local perturbations. In terms of normal variables the quantum rigidity of microscopic bonds is described [46] under the tacit assumption (ignoring the problem of interatomic communications) that an aggregate of mechanical oscillators generates a momentum-absorbing potential analogous to the external potential acting on a single oscillator. In other words, the rigid behaviour of a macroscopic aggregate leads *to assume, rather than to explain*, the rigidity of the crystal potential, which is certainly not implicit in the postulates of quantum mechanics: it is a property, on the contrary, which any consistent model should expressly justify.

6. Conclusions

In conclusion, the role, suggested by the D–W expression (to be viewed as an empirical description, and not as a physical interpretation), of the zero-point mechanical oscillation spectrum [46,49] in all elastic phenomena is quite plausible. While, indeed, the presence of the *thermal* phonon spectrum is limited, below the Debye temperature, to the lowest frequencies, the entire spectrum is always present and active in the *zero-point* (phononless) oscillations. The zero-point background has therefore the possibility, in stationary conditions, of acquainting each atom of the crystal with the characteristics of the whole lattice to which it belongs, including its irregularities, which strongly affect the spectral structure of the crystal oscillations.

Within this logical frame, the main question, in view of an adequate theoretical treatment, concerns the creation, due to the zero-point background, of a coalescence able either to absorb the impulse due to a sudden and localized force acting on a single nucleus, or to transfer it to the center of mass of the crystal: a transfer which must turn out to be both *instantaneous* (thus avoiding the nuclear recoil) and *selective* (i.e., limited to the D–W fraction). The natural zero-point oscillations, through the role of their momentum spectrum, may be responsible of the self-preserving character of the zero-point structure of the lattice. This could avoid (in a fraction of cases predicted by the D–W expression) the excitation of acoustic phonons which would destroy the elastic character of the process.

A subsequent paper should try to make plausible the existence of a zero-point momentum reservoir, providing an instantaneous balance, in any possible direction, to a wide range of suddenly and locally imparted momenta, thus justifying the elasticity of quantum bonds in solid structures.

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