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Physical implications of the Mössbauer effect

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Summary. — The main features of the Mössbauer effect are analyzed, and shown to cause an apparent violation of the uncertainty principle unless an appropriate "coalescence" is introduced, acting between at least 10^9–10^{10} nuclei.

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1. – Introduction

Since its discovery in 1957 [1], the Mössbauer effect (ME) has captured the attention of two generations of scientists, and given origin to a wide community of researchers who currently make of it the basis of their theoretical and experimental activity.

For most of them the basic interest of the ME derives from the fact that it suggests and allows measurements endowed with an unprecedented precision (both in Physics and in other Sciences, such as Chemistry and Biology), requiring in general rather simple and low-cost equipments [2, 3].

For other researchers, however, the main appeal of the ME stands in its interpretation and in its conceptual implications, involving the very foundations of solid-state Physics.

Let us recall here that, during his experiments on the nuclear resonance scattering of the γ-rays emitted in many decays of excited nuclei, Mössbauer observed that a fraction of the emitted photons exhibited an increased resonant absorption in cases where, because of the cooling applied both to the source and to the absorber, no effect at all was to be expected. Most of all, the experimental results showed a peculiar absence of recoil both in the emitting and in the absorbing nuclei, and a drastic reduction of the Doppler broadening in the registered spectra.

As far as the recoil-free character of these processes is concerned, a strong analogy was immediately observed with the scattering of X-rays and slow neutrons by crystal lattices, where the energy exchange, in a computable fraction of cases, turns out to be vanishingly small. The fraction of recoilless nuclei (which may be even close to 100%) was found to be provided by the well-known Debye-Waller formula [4-7], expressing the probability of a zero-phonon process.

Little attention was paid to the reduced Doppler broadening of the observed spectra. Among the few researchers who considered this point, Frauenfelder [8] tried to establish an analogy with the results presented by Dicke [9] on the spectral sharpening found in the radiation emitted in suitable conditions by a gas in the infrared and microwave range. We shall show, however, that this comparison does not appear to be logically justified.

The most challenging question raised by the ME concerns its consistency with energy and momentum conservation. The present paper is due to the widespread feeling, perceived by its authors throughout the specialized literature, that the apparent violations of these conservation laws may be covered, and therefore basically justified, by the indeterminacy level allowed by Heisenberg's principle. We shall see, on the contrary, that the uncertainty principle itself turns out to be largely violated by the ME, unless an appropriate (and *not yet* existing) theory of the structure of matter (and of its internal forces) may be shown to explain the experimental evidence.

2. – Physical implications of the Doppler broadening reduction

Let us begin our considerations by analyzing the absence of Doppler broadening exhibited by the fraction of emitting nuclei which undergo a Mössbauer process.

According to Mössbauer himself [10], if we call x a generic direction through the emitting atom, Δx a length of the order of the average interatomic distance in the crystal lattice and Δp_x the indeterminacy of the relevant momentum component of the radiating nucleus, we must expect that

$$\Delta x\, \Delta p_x \geqslant \hbar \,. \tag{1}$$

Since $\hbar \cong 1.054 \times 10^{-27}$ erg s, and the interatomic distance, in a solid, is of the order of a few ångströms, the relation (1) implies that

$$\Delta p_x \geqslant 10^{-19}\,\text{g cm/s}\,. \tag{2}$$

In order to check this estimate, let us consider here the typical case [11] of the γ-decay of the excited nuclei of ^{67}Zn, where the involved mass is $M \cong 1.086 \times 10^{-22}$ g, the photon is emitted with an energy $E_0 \cong 93.3$ keV $\cong 1.495 \times 10^{-7}$ erg and the decay time of the excited nuclear state is $\tau \cong 1.3 \times 10^{-5}$ s.

The width Γ_0 of the corresponding emission line may be evaluated by means of the usual relation

$$\tau \Gamma_0 \cong \hbar \,, \tag{3}$$

leading to the value

$$\Gamma_0 \cong 5.06 \times 10^{-11}\,\text{eV} \cong 8.106 \times 10^{-23}\,\text{erg}\,. \tag{4}$$

For nuclei subject to Mössbauer decay, the extent of the thermal Doppler broadening (if it exists) may reach at most, consistently with the experimental evidence, a fraction of the observed value Γ_r of the width of the line emitted in the considered reaction.

Since it turns out that $\Gamma_r \approx 2\Gamma_0$, we shall assume, for a numerical estimate, that such a thermal contribution is of the order of $\Gamma_0/2$.

If we call: E_{lab} the photon energy in the laboratory frame, v_{th} the thermal velocity of the nucleus, and $\beta_{th} = v_{th}/c$, we may expect (on the basis of the usual expression of the energy Doppler shift) a value of E_{lab} in the range

$$E_{lab} \cong E_0 \pm \beta_{th} E_0 . \tag{5}$$

This leads to an expected Doppler broadening

$$|E_{lab} - E_0| \cong |\beta_{th} E_0| \cong \Gamma_0/2 \cong 2.53 \times 10^{-11}\,\text{eV} , \tag{6}$$

and to a value $\beta_{th} \cong 2.71 \times 10^{-16}$. Since, in the present case, the uncertainty of the momentum component p_x of the nucleus is of the order of $2p_{th} \equiv 2Mv_{th}$, we see therefore that

$$\Delta p_x \cong 2Mc\beta_{th} \cong 1.76 \times 10^{-27}\,\text{g cm/s} , \tag{7}$$

a value smaller by many orders of magnitude with respect to the one, given by (2), required by the uncertainty principle.

We stress that such a principle is satisfactorily respected when no ME is present. When, in fact, the linewidth Γ_r is due to the usual thermal Doppler broadening, it turns out to be of the order of 0.1 eV, many orders of magnitude larger than Γ_0. The value of β_{th} is then of the order of 10^{-6}, so that $\Delta p_x \cong 1.7 \times 10^{-17}$ g cm/s, in full respect of condition (2).

In the case of the ME, on the other hand, in order not to violate Heisenberg's principle, we should have to associate to the emitting nucleus an effective mass at least 10^9 times larger than M, thus implying a sort of rigid "coalescence" of almost all the nuclei (including the nuclei not subject to the ME) contained in a microcrystal of about 0.4 μm (such as the one employed, for instance, in ref. [11]): no ME is observed, in fact, when the microcrystal is substantially smaller.

Concerning the need for a theory of such a "coalescence" process, we recall that no phonon is created in the ME, which leaves the quantum state of the lattice unperturbed. Since, moreover, no displacement is found in the radiated peak, no force of simply electrostatic nature (as the ones usually assumed to act between the crystal nuclei) may be strong enough to justify the observed nuclear rigidity. The required force should have to be greater, indeed, than the elastic one binding the nucleus to the lattice, even if we assumed an interaction time between the γ-radiation and the emitting nucleus of the order of the decay time τ. The hardest task connected to such a theory is to explain in which way this process may selectively affect only the fraction of nuclei undergoing a ME, while the other nuclei of the same lattice behave in a perfectly usual way, *i.e.* as single and recoiling particles.

3. – Physical implications of the recoil-free emission and absorption

According to Mössbauer [10], if $\mathbf{p}_1$ is the momentum of the nucleus before the emitting process, $\mathbf{k}$ the wave number of the emitted radiation and $\mathbf{p}_\gamma = \hbar\mathbf{k}$, the total energy E_r exchanged by the emitting nucleus is given by

$$E_r = (\mathbf{p}_1 - \mathbf{p}_\gamma)^2/2M - \mathbf{p}_1^2/2M = \mathbf{p}_\gamma^2/2M - \mathbf{p}_\gamma \cdot \mathbf{p}_1/M . \tag{8}$$

In this relation the term $E_{r\gamma} \equiv \mathbf{p}_\gamma^2/2M$, representing the recoil energy imparted by the photon to the emitting nucleus, *does not* depend on the motion of the nucleus itself, and can only cause a displacement of the spectral line.

The term $E_{rp} \equiv \mathbf{p}_\gamma \cdot \mathbf{p}_1/M$, on the other hand, *does* depend on the momentum distribution of the nucleus, and ought to cause a thermal Doppler broadening. In agreement with the arguments of the previous section, we may assume that

$$E_{rp} \approx p_\gamma p_1/M \approx \Gamma_0/2\,. \tag{9}$$

Let us now refer, in order to fix ideas, to the case of the γ emission from the excited nuclei of ^{57}Fe, where the involved mass is $M \cong 9.24 \times 10^{-23}$ g, the emission energy is $E_0 \cong 14.4$ keV $\cong 2.3 \times 10^{-8}$ erg, and $p_\gamma = E_0/c \cong 0.766 \times 10^{-18}$ g cm/s.

By taking again x as a generic direction through the emitting nucleus, we may estimate that

$$\Delta p_x \approx 2p_1 \approx (\Gamma_0/2)(M/p_\gamma) \cong 1.32 \times 10^{-24}\,\text{g cm/s}\,. \tag{10}$$

Even by assuming $\Delta x \cong 0.4\,\mu$m (*i.e.* by attributing to the position of the nucleus an uncertainty ranging all over the employed microcrystal, much larger than the one proposed by Mössbauer himself [10]) we see therefore that

$$\Delta x\,\Delta p_x \approx 5.\,29 \times 10^{-29}\,\text{erg s}\,, \tag{11}$$

a result two orders of magnitude lower than the value required by the uncertainty principle.

We recall, moreover, that in measurements such as the ones performed on ^{67}Zn (by means of a refinement of the Mössbauer spectrometer allowing to reach the present-day energy resolution limit [12]) it is typically found that $E_{r\gamma} \leq 10^{-11}$ eV.

The only possible conclusion is that eq. (8), although correctly describing the usual emission processes, does not hold in the case of the ME, unless we assume, for any single emission process, an effective mass of the order of 10^9 atoms at least. Such a "coalescence" process appears again to be an unavoidable requirement for any future theory of the ME.

4. – Doppler broadening and zero-point energy

As is well known, the basic working principle of a Mössbauer spectrometer [8] is the energy sweeping of the γ-radiation around the emission energy E_0 by means of a mechanical oscillation impressed to the entire microcrystal employed as a radiation source, thus allowing to get a measurement of the absorbed fraction of photons as a function of the instantaneous velocity imposed to the source. This procedure assumes that the radiating nuclei are rigidly bound to the crystal to which they belong: an assumption which, as we have seen, is directly suggested by the experimental evidence.

It is an interesting point to verify here to what extent this evidence is consistent with our physical expectations.

Let E_M be the average kinetic energy of the emitting nucleus, and ΔE_{th} the corresponding thermal broadening of the γ-radiation spectrum. By keeping eq. (5) into account, it is seen that

$$|\beta_{\rm th}| \cong \Delta E_{\rm th}/E_0, \tag{12}$$

so that

$$E_M = M v_{\rm th}^2/2 \cong (\Delta E_{\rm th}/E_0)^2 M c^2/2 . \tag{13}$$

Recalling that, in the case of ^{67}Zn, we have $E_0 \cong 93.3$ keV, and that we previously estimated $\Delta E_{\rm th} \cong 2.53 \times 10^{-11}$ eV, it is seen that

$$E_M \cong 3.5 \times 10^{-33}\,{\rm erg} . \tag{14}$$

Since the Debye frequency of ^{67}Zn is $\omega_{\rm D} \cong 3.06 \times 10^{13}$ rad/s [13], the relevant zero-point energy is given by

$$\hbar\omega_{\rm D}/2 \cong 0.01\ {\rm eV} \cong 1.6 \times 10^{-14}\,{\rm erg} . \tag{15}$$

If we keep into account (according to the Debye model) all the frequencies between zero and $\omega_{\rm D}$, the entire zero-point energy of the nucleus turns out to amount to four times such a value.

We may conclude that the energy content revealed by the observed linewidth is smaller, by many orders of magnitude, than the expected contribution of the zero-point energy, although its presence is required by the uncertainty principle itself.

5. – The zero-average interpretation

As we have already recalled, one of the main difficulties presented by the interpretation of the ME arises from the unobservability of any trace, in the γ-radiation spectrum, of both the recoil and thermal energy of the emitting nucleus.

In striking contrast with this fact, the γ spectrum turns out to be appreciably affected by the energies corresponding to valence bonds, magnetic moments and atomic asymmetries acting, even very loosely, on the nucleus. Referring to the relativistic Doppler shift

$$E_{\rm lab} = E_0(1-\beta^2)^{1/2}(1-\beta\cos\alpha_{\rm lab})^{-1} \tag{16}$$

(where $\beta = v/c$, and $\alpha_{\rm lab}$ is the angle between the instantaneous velocity of the emitting nucleus and the radiation direction), Frauenfelder [8] argues that "the characteristic time for the lattice vibration is much shorter than the lifetime of the excited nuclear state. The linear term $\beta\cos\alpha_{\rm lab}$ in eq. (16) will hence average out to zero, giving rise to the unshifted and sharp ME". In other words, the effect of the oscillations performed by a nucleus during its excited lifetime τ (which typically ranges between 10^{-5}–10^{-8} s), with a frequency given by the Debye value (of about 10^{13} Hz) at most, may be thought to average to zero in the final frequency shift, thus cancelling the so-called "first order" Doppler effect (proportional to β).

A proof of this assertion is found by Frauenfelder in the works published by Dicke [9] on the narrowing of the emission lines occurring, for a gas in suitable conditions, in the microwave field.

The line sharpening described by Dicke (by assuming an ordered structure of the molecular mean free path) does not appear, however, to fit the ME. In Dicke's case, in fact, the radiation frequency is $\nu \cong 10^{10}$ Hz, and the thermal oscillation frequency is about 10^{12} Hz.

In the case of the ME, on the other hand, $\nu \cong 1.5 \times 10^{19}$ Hz: the frequency of the thermal oscillations of the nucleus (which may reach, at most, the Debye frequency value) is not high enough to average their effect to zero even during many periods of the emitted radiation.

The *coup de grace* to Frauenfelder's argument is given by the fact that nuclei of the same kind, with the same decay time τ and making part of the same crystal, but *not* subject to a ME, lead to a regularly broadened and back-shifted spectrum, where no cancellation effect is observed.

Referring to eq. (16), the supporters of the zero-average argument claim that a *second-order* Doppler effect (proportional to β^2) could survive to such an average, thus somewhat confirming the argument itself. Josephson [14] however (as well as other researchers [15] acting along different lines) has clearly excluded the existence of such a second-order effect. The experimental results attributed, at first, to this effect were due in fact to the relativistic loss of mass corresponding to the emitted energy. We may say, with Balko, Cohen and Sparrow [16], that "under certain conditions, the nuclei in the crystalline medium exhibited a behaviour that is equivalent to their having a velocity of zero, and all the recoil velocities of the emitting nuclei were zero".

If, in conclusion, the zero-average were due to the oscillations occurring during a few radiation periods, the frequency of these oscillations should be much larger than the one of the emitted radiation. If, on the other hand, such a zero-average concerned the radiation emitted during the entire decay time τ, it should occur for *any* kind of nuclear oscillations, whatever their origin could be. All on the contrary, no zero-average is observed in those experiments where the entire lattice is forced to vibrate with a period less than the decay time τ, with amplitudes of the order of that of the thermal oscillations. In particular, in the work published by Ruby and Bolef [17] (the first of a long line of research) an experiment is described where atoms of ^{57}Co had previously diffused into a thin sheet of stainless steel. Such a sheet was glued on a piezoelectric crystal, forcing it to vibrate at 20 Mhz. The amplitude of the space oscillation was about 0.2 Å. The final γ-radiation at 14.4 keV turned out to be modulated by the imposed vibration, showing no trace of an average on the instantaneous impressed velocities during the emission time.

It could be easily verified, indeed, that, in this case, a numerical average performed over a time of the order of $10\,\tau$ should lead, if actually present, to an almost complete disappearance of the imposed modulation frequency.

We finally observe here that the zero-average interpretation was very efficaciously excluded by many other authors, such as Lipkin [18] and Eyges [19].

6. – Conclusions

We recall here that every elementary process of Mössbauer emission proceeds from a single and definite nucleus, as is clearly shown, for instance [20], by the reaction chain leading from the excited ^{57}Co to ^{57}Fe. In such a nuclear decay (which is the most currently employed in Mössbauer spectroscopy), the emission of a first (generally non-Mössbauer) photon at 123 keV is followed, in fact, after a decay time $\tau \cong 10^{-7}$ s, by a second photon (at 14.4 keV), clearly coming from the *same* nucleus and generally subject to ME. We observe, incidentally, that the steep oscillation energy increase, which should be exhibited by the nucleus because of the recoil due to the first photon, appears to be completely absent when the second photon is emitted.

The single (and definite) particle character of the Mössbauer process is the basis of the arguments contained in the present paper. Now, the ME belongs to a wider class of phenomena (such as the scattering of X-rays and slow neutrons from crystal lattices, and such as the very presence of a second, unshifted peak in the Compton effect [21]) which leave a physical system in a final state equal to the initial one: any theory of such a behaviour must be able to explain this "rigidity".

If the emission of γ-radiation is described in terms of emitted and absorbed photons (as was done both by Mössbauer himself [10] and in the present paper) the apparent violation of Heisenberg's principle indicates the need of devising a mechanism able to establish a rigid bound between the nucleus and the lattice.

If the γ-radiation, on the other hand, is described (as is done, for instance, in the cited case of Frauenfelder [8]) in terms of wave trains emitted in times even longer than τ, decaying as $\exp[-t/\tau]$ and subject to a zero-average fluctuation effect, the *fluctuation* frequency should be reasonably assumed to be much higher than the *radiation* frequency.

One would also expect a critical dependence on τ of the Debye-Waller formula; and it remains to be explained why this average of the thermal motions affects only the atoms subject to a ME, and why it does not occur in the case of externally imposed mechanical vibrations.

We may conclude that the mechanism underlying the ME still constitutes an unsolved problem.

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