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Suppression of transition dipole moments in radioactive atoms

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Abstract. Optical emission from radioactive atoms is investigated. It is shown that the radioactivity of the central nucleus results in the suppresion of the optical transition dipole moment in the corresponding atom. The situations are regarded when this suppression is significant and can be experimentally observed.

1. Introduction

Optical research techniques play an important role for nuclear physics [1]. Measuring isotopic shift and hyperfine splitting in atomic spectra provides us information on nuclear mass, shape and spins [2,3]. The optical approach is especially important for short-lived nuclei, where other techniques may appear not applicable. However, in the case of short-lived nuclei the ratio of the mean time of an optical transition and the nuclear half-life comes into play. Obviously, the lifetime of a radioactive nucleus should be larger or at least comparable to the time required for the optical transition to occur. In this paper we will show, that in the case when these times are of the same order, nuclear radioactivity manifests itself in the optical spectrum, giving rise to the suppresion of the optical transition dipole moment, and as a consequence to the decrease of spontaneous emission probability.

2. Natural broadening

It is well known that the natural broadening of a spectral line emitted on the transition from an initial level $|i\rangle$ to a final level $|f\rangle$ is determined by the Breit-Wigner contour:

$$\frac{dW}{d\omega} = \frac{2\pi}{(\omega - \omega_{fi})^2 + \Gamma_{fi}^2/4}, \tag{1}$$

where ω is the photon frequency, ω_{fi} is the atomic transition frequency, Γ_i (Γ_f) is the width of the initial (final) level, and $\Gamma_{fi} = \Gamma_f + \Gamma_i$ is the spectral line width.

An atom represents a bound system consisting of a nucleus and electrons and should be described by a multiparticle wave function. Correspondingly, the width of an atomic level Γ_n ($n = i, f$) is affected both by the nuclear and electronic decay processes and in the simplest case can be represented as the superposition:

$$\Gamma_n = \Gamma_n^{(e)} + \Gamma_0^{(N)}, \tag{2}$$

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where $\Gamma_n^{(e)}$ is the purely electromagnetic decay width and $\Gamma_0^{(N)}$ is the nuclear decay width. Here we suppose the nuclear decay processes to be not affected by the state of the electronic shells, i.e. $\Gamma_0^{(N)}$ is one and the same for any electronic state $|n\rangle$. This suggestion is justifiable, because the nuclear interaction is several orders more intensive than the electromagnetic one. From this it follows, that the spectral line width Γ_{fi} is generally described by the following expression:

$$\Gamma_{fi} = \Gamma_{fi}^{(e)} + 2\Gamma_0^{(N)}, \tag{3}$$

where $\Gamma_{fi}^{(e)} = \Gamma_f^{(e)} + \Gamma_i^{(e)}$ is the width of the electronic transition. Usually, $\Gamma_{fi}^{(e)} \gg \Gamma_N^{(0)}$, i.e. the nuclear lifetime $\tau_N = \hbar/\Gamma_0^{(N)}$ is much greater than the mean time of the electronic transition $\tau_{fi}^{(e)} = \hbar/\Gamma_{fi}^{(e)}$. This ensures that the atom emits the photon till its nucleus decays. The spectral line width in this case is determined by the electronic decay width only: $\Gamma_{fi} = \Gamma_{fi}^{(e)}$. In contrast, when $\Gamma_{fi}^{(e)} \ll \Gamma_N^{(0)}$, i.e. $\tau_{fi}^{(e)} \gg \tau_N$, the nucleus decays before the atom emits and no optical spectrum can be observed.

However, for some atoms created under laboratory conditions the nuclear lifetime coincides in order with the mean time of an optical transition [4,5]. For such atoms the widths in expression (3) are comparable and contribute equally into the total spectral line width. Thus, the natural line broadening of such atoms will be affected by the nuclear radioactivity as well.

3. Generalized Quantum Dynamics

While the electromagnetic interaction is well understood and theoretically described, the nuclear interaction Hamiltonian has not been found yet. Instaed, the effective field theory (EFT) was proposed by Weinberg for the description of nucleon-nucleon (NN) interactions, which implies no potentials at all [6]. At the same time, it has been shown that the Schrödinger equation is not the most general dynamical equation consistent with the current concepts of quantum physics, and a more general equation of motion was derived by Gainutdinov in [7]. Being equivalent to the Schrödinger equation in the case of instantaneous interactions, this equation permits the generalization to the case where dynamics is governed by a nonlocal-in-time interaction. It was successfully applied for solving different problems in atomic [8] and nuclear physics [9–11], as well as in quantum optics [12]. In Ref. [9] it has been shown that the requirement that the NN T-matrix satisfies this equation and the symmetries of quantum chromodynamics, determines the low energy NN operator up to a constant. The interaction operator defined in this way is nonlocal in time. Thus, the low energy NN dynamics is governed by the Gainutdinov equation with the nonlocal-in-time interaction operator. The NN T-matrix obtained by solving this equation was shown to coincide with that derived in EFT of nuclear forces [13]. In this work we use this equation, because its form does not depend on the specific feature of the interaction (the Scrödinger equation contains the interaction Hamiltonian). All the needed dynamical information is contained in the boundary condition for this equation. This feature of the equation allows one to draw general conclusions about the decay of such complex systems as nuclei.

The Gainutdinov equation has been derived as a direct consequence of the basic postulate of the Feynman approach expressing the superposition law for quantum probability amplitudes and the basic postulates of the canonical approach manifesting the ingenious Dirac guess that the physical logic of microworld is adequate to the mathematical logic of Hilbert spaces. In particular, from the Feynman postulate it follows that $\langle\Psi_2|U(t_2,t_1)|\Psi_1\rangle$, being the probability amplitude of finding the quantum system in the state $|\Psi_2\rangle$ at time t_2 if at time t_1 it was in the state $|\Psi_1\rangle$, can be represented as a sum of probability amplitudes for each of the alternative ways in which this event can happen. These alternatives can be divided in different classes. For

example, subprocesses with definite instants of the beginning and end of the interaction in the system can be chosen as such alternatives. In this case the evolution operator can be represented in the form

$$U(t_2,t_1)=\mathbf{1}+\int_{t_1}^{t_2}dt'\int_{t_1}^{t'}dt\ \tilde{S}(t',t), \tag{4}$$

where $\tilde{S}(t',t)$ is an operator whose matrix elements $\langle\Psi_2|\tilde{S}(t',t)|\Psi_1\rangle$ are the probability amplitudes for the alternative ways in which the interaction in the system begins at time t and ends at time t'. For the evolution operator to be unitary for any t_2 and t_1, the operator $\tilde{S}(t',t)$ must satisfy the equation [7]:

$$(t'-t)\tilde{S}(t',t)=\int_t^{t'}dt_4\int_t^{t_4}dt_3(t_4-t_3)\tilde{S}(t',t_4)\tilde{S}(t_3,t). \tag{5}$$

This equation allows one to obtain the operator $\tilde{S}(t',t)$ for any t and t' and hence the dynamics of the system, if $\tilde{S}(t'_4,t'_3)$ corresponding to infinitesimal duration times $\tau=t'_4-t'_3$ of interaction are known. Most of the contribution to the evolution operator in the limit $t_2\to t_1$ comes from the processes associated with the fundamental interaction in the system encoded in the interaction operator $H_{\rm int}(t',t)$. Equation (5) with the boundary condition $\tilde{S}(t',t)\lim_{t'\to t}H_{\rm int}(t',t)$ determines the dynamics of the system. The evolution operator $U(t,0)$ in the Schrödinger picture can be presented in the form:

$$U(t,0)=\frac{i}{2\pi}\int dx\exp(-izt)G(z), \tag{6}$$

where $z=x+iy$, $G(z)=G_0(z)+G_0(z)T(z)G_0(z)$, $G_0(z)=(z-H_0)^{-1}$ is the free Green operator (H_0 is the free Hamiltonian) and $T(z)=i\int_0^\infty d\tau\exp^{(-iz\tau)}\exp(-iH_0t_2)\tilde{S}(t_2,t_1)\exp(iH_0t_1)$. The contribution to the Green operator $G(z)$, which comes from the processes associated with the self-interaction of particles, has the same structure as the free Green operator $G_0(z)$. For this reason it is natural to replace $G_0(z)$ by the propagator $\tilde{G}_0(z)$, which describes the evolution of particles interacting only with vacuum and hence has the structure $\tilde{G}_0(z)=(z-H_0-C(z))^{-1}$, where the operator $C(z)$ has the same eigenvectors as H_0 ($C(z)|n\rangle=C_n(z)|n\rangle$, $H_0|n\rangle=E_n|n\rangle$). Correspondingly, the operator $T(z)$ should be replaced by the operator $M(z)$, which describes the evolution of particles interacting not only with vacuum. These operators are related as follows: $G(z)=G_0(z)+G_0(z)T(z)G_0(z)=\tilde{G}_0(z)+\tilde{G}_0(z)M(z)\tilde{G}_0(z)$. Making use of the equation

$$\frac{dG(z)}{dz}=-G^2(z), \tag{7}$$

being one of the forms of equation (5) in the energy representation [14–18], one can get the following equations for the operators $M(z)$ and $C(z)$:

$$\begin{aligned}\frac{dM(z)}{dz}&=-(1-P)M(z)\tilde{G}_0(z)\tilde{G}_0(z)M(z)\\&-M(z)\tilde{G}_0(z)\frac{dC(z)}{dz}-\frac{dC(z)}{dz}\tilde{G}_0(z)M(z),\end{aligned} \tag{8}$$

$$\frac{dC_n(z)}{dz}=-\langle n|M(z)(\tilde{G}_0(z))^2M(z)|n\rangle, \tag{9}$$

where P is a projection operator: $P\hat{A}=\sum_n|n\rangle\langle n|a_n$ with $a_n=\langle n|\hat{A}|n\rangle$. The boundary conditions for these equations are $C(z)\lim_{|z|\to\infty}PB(z)$ and $M(z)\lim_{|z|\to\infty}(1-P)B(z)$, where $B(z)=i\int_0^\infty e^{-iz\tau}e^{-iH_0t_2}H_{\rm int}(t_2,t_1)e^{iH_0t_1}$.

In the case of a radioactive atom, the interaction operator $B(z)$ is a sum of the operator describing the strong interaction of nucleons $B^{(N)}(z)$ and the Hamiltonian H_I describing the week interaction of the atomic electrons with the electromagnetic field $B(z) = B^{(N)}(z) + H_I$. Solving equations (8) and (9) by expanding the solution in powers of H_I and restricting ourselves to the second order term, we get

$$M(z) = M^{(N)}(z)+H_I+M^{(N)}(z)\tilde{G}_0(z)H_I+H_I\tilde{G}_0(z)M^{(N)}(z)+H_I\tilde{G}_0(z)M^{(N)}(z)\tilde{G}_0(z)H_I, \quad (10)$$

$$C_n(z) = C_n^{(e)} + C_n^{(N)}, \quad (11)$$

where $C_n^{(e)}(z)$ and $C_n^{(N)}(z)$ are the solutions of the equations $dC_n^{(e)}(z)/dz = -\langle n|H_I(\tilde{G}_0(z))^2H_I|n\rangle$ and $dC_n^{(N)}(z)/dz = -\langle n|M^{(N)}(z)(\tilde{G}_0(z))^2M^{(N)}(z)|n\rangle$ respectively with corresponding boundary conditions. $M^{(N)}$ is the solution of equation (8) with the boundary condition $M(z)\lim_{|z|\to\infty}(1-P)B^{(N)}(z)$.

Generally, the self energy function $C_n(z)$ can be represented by its Taylor expansion:

$$C_n(z) = C_n(z = E_n^{(0)}) + \frac{dC_n(z)}{dz}|_{z=E_n^{(0)}}+..., \quad (12)$$

where $E_n^{(0)}$ is the bare energy of the state $|n\rangle$. It is well known that the first term in this expansion represents the energy shift $\Delta E_n^{(0)}$ caused by the vacuum polarization and the decay width Γ_n:

$$C_n(z = E_n^{(0)}) = \Delta E_n^{(0)} - i\Gamma_n.$$

In what follows we will assume that the energy shifts $\Delta E_n^{(0)}$ are included into the energies of the states, i.e. that the renormalized energy values are used.

As for the derivative

$$\eta = \frac{dC_n(z)}{dz}|_{z=E_n^{(0)}},$$

it includes the contributions from the off-shell virtual transitions associated with vacuum fluctuations $\Sigma_n(z)$ and from the real on-shell transitions giving rise to the system dissipation $C_n^{\rm on}(z)$: $\eta = \Sigma_n(z) + C_n^{\rm on}(z)$. The second term in this expression is imaginary and represents a small correction to the decay width Γ_n, which can be neglected in virtue of the exponentiality of the decay law. The off-shell function characterizes the strength of quantum fluctuations, manifesting themselves in virtual transitions of the quantum system into all possible decay states. The fact that in the problem under study the decay widths of the nuclear and electronic states are of the same order of magnitude does not mean that the same is true for the nuclear and electronic off-shell functions $\Sigma_n^{(N)}(z)$ and $\Sigma_n^{(e)}(z)$. Indeed, despite the fact that the interaction of nucleons is strong, the nucleon decay widths that are determined by the on-shell transitions are relatively small. But in virtual transitions the strength of NN interaction may manifest itself explicitly. In contrast, the quantum fluctuations induced by the interaction of electrons with the electromagnetic field are relatively week ($\Sigma_n^{(e)}$ are of the order of $\alpha^4 m_e$, where $\alpha = 1/137$ is the fine structure constant and m_e is the electronic rest mass) and can be neglected. Thus, the self energy function of an atom can be represented in the form

$$C_n(z) = \Sigma_n^{(N)}(z) - i\Gamma_0^{(N)}/2 - i\Gamma_n^{(e)}/2. \quad (13)$$

With (13), the free green operator of the radioactive atom in the state $|n\rangle$ has the following form:

$$G(z) = \frac{1}{z - E_N - \varepsilon_n - H_I - B^{(N)}(z) - \Sigma_n^{(N)}(z) + i\Gamma_0^{(N)}/2 + i\Gamma_n^{(e)}/2},$$

where E_N is the renormalized rest energy of the nucleus and ε_n is the electronic energy in the state $|n\rangle$.

4. Suppression of atomic transition dipole moment

The probability A_{fi} that a photon will be emitted, when the atom transits from the state $|i\rangle$ to the state $|f\rangle$ is proportinal to the cube of the photon frequency ω and the absolute square of the matrix element of transition dipole moment operator d_{fi}:

$$A_{fi} \sim \omega^3 |d_{fi}|^2.$$

The matrix element of the transition dipole moment operator is defined on the basis of the electronic wave functions φ_f and φ_i in the initial and in the final states correspondingly:

$$d_{fi} = e \int \varphi_f^*(\mathbf{r}) \mathbf{r} \varphi_i(\mathbf{r}) d\mathbf{r}. \tag{14}$$

These functions are usually found as solutions of the corresponding Schrödinger (Dirac) equation with the constant central potential.

However, due to quantum fluctuations the unstable nucleus spends a part of time in its bare state $|\Psi_n^0\rangle$, while another part of time it exists in the virtual state of its decay products $|\Psi_{\text{virt}}\rangle$. Thus, the state vector $|\Psi_n\rangle$ of the unstable nucleus can be generally represented as a superposition of two vectors:

$$|\Psi_n\rangle = \mathcal{Z}|\Psi_n^0\rangle + \chi|\Psi_{\text{virt}}\rangle,$$

where $\mathcal{Z} = (1-\eta)^{-1}$ determines the probability to find the nucleus in the bare state, while $\chi = \eta/(\eta-1)$ is the probability to find the nucleus in the state of its decay products. This means, that the atomic electrons spend not the whole time in the field of the bare nucleus potential, when they can be described by the wave functions φ_f and φ_i. Actually, during a certain time they exist in the field of the nuclear decay products. Owing to this, the contribution from φ_f and φ_i to (14) decreases and thus the matrix element of the dipole moment operator is suppressed. It can be shown [19], that the probability for a photon to be emitted with a certain energy ω is defined by the following formula:

$$\frac{dW}{d\omega} \sim \omega \frac{\Gamma}{(\omega - \omega_{fi})^2 + \Gamma^2/4}, \tag{15}$$

with the width

$$\Gamma = 2\Gamma^{(N)} + \mathcal{Z}^2 \Gamma_{fi}^{(e)}. \tag{16}$$

According to the definition, $\mathcal{Z} < 1$, and hence the contribution from the electromagnetic decay width is suppressed.

The coefficients $\mathcal{Z}$ and χ determine the ratio of the time spent by the nucleus in the bare state to the time spent in the state of its decay products. The values of these parameters change from one isotope to another, as well as depend on the nuclear decay type. Usually $\mathcal{Z} \sim 1$, but in some situations this relation may change. One interesting example for this is super-allowed alpha decay observed for certain light nuclei in the vicinity of ^{100}Sn isotope. It is supposed that the possibility of alpha decay in this region is caused by the fact that protons and neutrons occupy identical orbitals leading to a significant increase of the alpha particle preformation probability [20]. This means, that the part of time spent by the nucleus in the state of its decay products is essential. The lightest experimentally observed alpha emitter is the isotope of tellurium ^{105}Te. The measured reduced width of this isotope is $\delta_r \sim 70keV$ [4, 5]. It can be shown that the corresponding strength of quantum fluctuations should have the order of 0.4: $\eta \sim 0.4$. Additionally, the half-life of ^{105}Te is measured to be $\sim$ 700ns, that is the widths $\Gamma^{(N)}$ and $\Gamma_{fi}^{(e)}$ are comparable for certain optical transitions in the atom of this isotope. Thus, according to formulae (15) and (16) the nuclear radioactivity will manifest itself in the optical spectrum, giving rise to the change of natural line broadening.

5. Conclusion

We have considered the influence of nuclear radioactivity on the optical spectrum of corresponding atoms. We have derived a general formula for the spectral line profile, emitted from an atom with a radioactive nucleus, which contains the nuclear decay width as well. We have shown that for certain nuclei quantum fluctuation processes result in the suppression of the atomic transition dipole moment and as a consequence to the change of spectral line natural width. Physically, the reason for this is the fact, that such nuclei spend just a part of time in a bare state, while another part of time they exist in the state of their decay products, and as a consequence the atomic electrons experience the bare nucleus potential not the whole time. This phenomenon can be important for the creation of new optical method for the measurement of nuclear half-life from the optical line width.

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