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APPLICATION OF THE PHASE REPRESENTATION TO THE THEORY OF RESONANCE FLUORESCENCE

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In this paper we use the phase representation formalism which was introduced by I. Białynicki-Birula, Z. Białynicka-Birula, *Phys. Rev.* **A14**, 1101 (1976) and applied in subsequent publications to the calculation of energy levels of a dressed atom (*Bull. Acad. Pol. Sci. Ser. Sci. Math. Astron. Phys.* **26**, 1501 (1978) and *Acta Phys. Pol.* **A57**, 599 (1980)). The algorithm for the calculation of the density matrix proposed earlier will be used here to study the resonance fluorescence.

1. Introduction

In a series of recent[1] papers [3, 4] we have applied a new simple numerical method to the calculation of the energy levels of a dressed atom and used it to study different resonances phenomena. Our methods have been based on the phase representation [1, 2] which is very useful in the analysis of quantum processes taking place in the presence of intense photon beams. As it was shown in [1] and [4], the phase representation is also useful in the study of atomic density matrices.

In this paper I shall apply the algorithm introduced in II to the study of resonance fluorescence.

In Section 2 I give a brief presentation of our algorithm introduced in I. Its application to resonance fluorescence is presented in Section 3. Section 4 contains conclusions.

2. The algorithm for the calculation of atomic density matrices

For the sake of simplicity I assume that the atom is dressed by the field polarized linearly and the atom has two levels. Our algorithm can also be used in the case of the dressing field with an arbitrary polarization.

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[1] I will refer to Ref. [3] and [4] as I, II and to the formulas of [3, 4] as (I. 1), (II. 1) etc.

I shall start from the equation which has been introduced in the previous papers (I.3),

$$i\frac{\partial\psi(\phi)}{\partial\phi}=H_\phi\psi(\phi) \tag{1}$$

and its formal solution

$$\psi(\phi)=U(\phi)\psi(0), \tag{2}$$

where

$$U(\phi)=\exp\left[-i(N-\varepsilon_\psi)\phi\right]P\exp\left(-i\int_0^\phi \mathscr{H}\,d\phi'\right) \tag{3}$$

and $\mathscr{H}$ was defined by (I.3), (II.27). Similarly as in the case of the time evolution, one can introduce in this "ϕ-picture" a ϕ-dependent density operator

$$\varrho(\phi)=U^\dagger(\phi)\varrho(0)U(\phi). \tag{4}$$

Decomposing $\varrho(\phi)$ into a sum of Pauli matrices

$$\varrho=\tfrac{1}{2}+\boldsymbol{p}\cdot\boldsymbol{\sigma},$$

one obtains

$$\varrho(\phi)=\tfrac{1}{2}+\boldsymbol{p}\cdot\boldsymbol{\sigma}(\phi),$$

where

$$\boldsymbol{\sigma}(\phi)=P\exp\left(i\int_0^\phi \mathscr{H}\,d\phi'\right)\boldsymbol{\sigma}(0)P\exp\left(-i\int_0^\phi \mathscr{H}\,d\phi'\right).$$

One the other hand, using the equation

$$i\frac{\partial\varrho}{\partial\phi}=[H_\phi,\varrho(\phi)],$$

one obtains the equation for $\boldsymbol{\sigma}(\phi)=(\sigma_1(\phi),\sigma_2(\phi),\sigma_3(\phi))$:

$$\frac{\partial\sigma_1}{\partial\phi}=\frac{\omega_0}{\omega}\sigma_2$$

$$\frac{\partial\sigma_2}{\partial\phi}=-\frac{\omega_0}{\omega}\sigma_1+4\frac{\lambda}{\omega}\cos\phi\sigma_3$$

$$\frac{\partial\sigma_3}{\partial\phi}=-4\frac{\lambda}{\omega}\cos\phi\sigma_2 \tag{5}$$

with the initial condition $\boldsymbol{\sigma}(0)=\boldsymbol{\sigma}$ ($\boldsymbol{\sigma}$ are the usual Pauli matrices).

Eq. (5) has a formal solution

$$\sigma(\phi) = P\exp\left[-i\int\limits_0^{\phi}\left(\frac{\omega_0}{\omega}\Sigma_3+4\frac{\lambda}{\omega}\cos\phi\Sigma_1\right)d\phi'\right]\sigma(0). \tag{6}$$

Now, as before [3, 4], I can again calculate the expression containing the ϕ-ordered exponential operator, dividing the interval $[0, \phi]$ into l subintervals. Denoting $\phi_m = m\phi/l$ I have

$$P\exp\left(-i\int\limits_0^{\phi}\mathscr{K}d\phi'\right) = \exp(-i\Delta\phi\mathscr{K}_l)\ldots\exp(-i\Delta\phi\mathscr{K}_1), \tag{7}$$

where

$$\mathscr{K}_m = x\Sigma_3+4y\cos\phi_m\Sigma_1.$$

The product (7) can again be interpreted as a composition of "small" rotations and the result is obtained in the same manner as before for the case of spin 1/2-representation

$$\mathscr{K}_m = \tfrac{1}{2}x\sigma_3+2y\sigma_1\cos\phi_m,$$

only now the interval $[0, 2\pi]$ is replaced by $[0, \phi]$. The resulting rotation is specified by the vector $\boldsymbol{\alpha}(\phi)$

$$\boldsymbol{\sigma}(\phi) = \exp(-i\boldsymbol{\alpha}\cdot\boldsymbol{\Sigma})\boldsymbol{\sigma}(0). \tag{8}$$

The expression for $\exp(-i\boldsymbol{\alpha}\cdot\boldsymbol{\Sigma})$ has been given by (II.42):

$$\exp(-i\boldsymbol{\alpha}\cdot\boldsymbol{\Sigma}) = \frac{\boldsymbol{\alpha}\otimes\boldsymbol{\alpha}}{|\boldsymbol{\alpha}|^2}+\left(1-\frac{\boldsymbol{\alpha}\otimes\boldsymbol{\alpha}}{|\boldsymbol{\alpha}|^2}\right)\cos|\boldsymbol{\alpha}|-i\frac{\boldsymbol{\alpha}}{|\boldsymbol{\alpha}|}\boldsymbol{\Sigma}\sin|\boldsymbol{\alpha}|, \tag{9}$$

where $(\boldsymbol{\alpha}\otimes\boldsymbol{\alpha})_{ij} = \alpha_i\alpha_j$.

Formulas (8) and (9) give a very simple algorithm for the calculation of $\sigma_i(\phi)$ and hence also for $\varrho(\phi)$. For the case of circular polarization of dressed field with the use of this expression I can obtain an exact analytical formula for $\boldsymbol{\sigma}(\phi)$.

3. *Application of the algorithm to the theory of resonance fluorescence*

3.1. Brief review of the theory of resonance fluorescence

The problem of resonance fluorescence has been analysed both experimentally and theoretically. The pioneering experiment has been done by Wood (1913). A lot of spectroscopic information (g-factor, fine or hyperfine structures, radiative lifetimes ...) is obtained by resonance fluorescence experiments [5].

At first the resonance fluorescence has been studied in the limit of a weak electromagnetic field (the intensity of the incident beam is very low) [6]. The development of lasers gave a new impulse in the study of this problem. Special properties of laser radiation (monochromatic property, high intensity etc.) ensure new experimental conditions for the study of resonance fluorescence in different regions of frequencies of the incident electromagnetic wave and in the limit of strong field.

It is well-known [6] that for a weak incident field the fluorescence spectrum is of the type of the δ-function. The first theoretical paper considering the limit of strong field was that of Mollow [8]. He obtained the three-peaked distribution of the fluorescence spectrum. The observation of the Mollow spectrum was first made by Schuda et al. [9]. Recently these results were confirmed by Walther [10]. References to recent theoretical papers can be found there. Generally in the vicinity of an exact resonance the predicted spectrum consists of a strong central component at the wavelength of the incoming laser radiation and two weaker side components which have 1/3 of the intensity of the central line. The separation of the side components is determined by the Rabi nutation frequency.

Cohen–Tanoudji [7] et al. calculated this spectrum treating the fluorescence as a spontaneous emission of dressed atoms. They obtained the same spectrum as before. It has been shown also that Fermi's golden rule approach to this problem is not adequate, because from this rule we obtain that the height of the central components is two times greater than that of the two side components. It is only from an exact analysis of the master equation [7] that one obtains the two sidebands to be three times smaller than the height of the central components (effect of cascades).

Experimental observations indicate the presence of an effect which is not in good agreement with the simple theory: the spectrum is not symmetrical as predicted by the theory. This was explained by different arguments e.g. by including the interaction with other levels [11] or by introducing a non-Markovian behavior in two-level atom fluorescence [12].

All of the papers mentioned above used the rotating wave approximation (RWA), so that the Bloch-Siegert shift was neglected.

I shall use a new very simple algorithm for calculating the transition probabilities between the levels of dressed atom. We shall see that even using only Fermi's golden rule, and neglecting the effect of interaction with other levels but without RWA, one obtains an asymmetrical fluorescence spectrum for the two-level atom with the one sideband having height three times smaller than the height of the central component in a certain domain of laser frequency and laser intensity. Thus we cannot neglect Bloch–Siegert shifts in the calculation, especially in the radiospectroscopic case.

3.2. New algorithm for numerical calculations of transition probabilities

For the sake of simplicity I consider again only a two-level atom dressed by the electromagnetic field polarized linearly, perpendicular to the static field. Then the transition amplitude for the spontaneous emission from a state ψ_i to a state ψ_f in the first order of perturbation theory is proportional to the matrix element (see Fig. 1) $\langle\psi_f|\sigma_1|\psi_i\rangle$, or in the phase representation

$$\langle\psi_f|\sigma_1|\psi_i\rangle = \int_0^{2\pi} \frac{d\phi}{2\pi}\, \psi_f^\dagger(\phi)\sigma_1\psi_i(\phi). \tag{10}$$

It is worth noting that Sambe [13] used similar Hilbert space with a similar scalar product as in the phase representation, but in the semiclassical treatment. From the form of the wave function (2) given in the previous section one obtains

$$\langle\psi_f|\sigma_1|\psi_i\rangle = \int_0^{2\pi} \frac{d\phi}{2\pi}\, \psi_f^\dagger(0)\sigma_1(\phi)\psi_i(0)\exp\left[i(\varepsilon_i-\varepsilon_f+n_f-n_i)\phi\right], \tag{11}$$

where $\sigma_1(\phi)$ was calculated in Section 2.

Fig. 1

For the calculation of (11) we must find $\psi_f(0)$ and $\psi_i(0)$, which satisfy the following equation

$$\left(\exp(i2\pi\varepsilon)\exp\left(\frac{i}{2}\boldsymbol{\alpha}\cdot\boldsymbol{\sigma}\right)-1\right)\psi_\varepsilon(0) = 0.$$

It follows further that

$$\boldsymbol{\alpha}\cdot\boldsymbol{\sigma}\psi_\varepsilon(0) = -|\boldsymbol{\alpha}|\psi_\varepsilon(0) \quad \text{if} \quad \varepsilon = \frac{|\boldsymbol{\alpha}|}{4\pi},$$

$$\boldsymbol{\alpha}\cdot\boldsymbol{\sigma}\psi_\varepsilon(0) = |\boldsymbol{\alpha}|\psi_\varepsilon(0) \quad \text{if} \quad \varepsilon = \frac{-|\boldsymbol{\alpha}|}{4\pi}. \tag{12}$$

Hence we have the following solutions of (12)

$$\psi_\varepsilon(0) = \frac{1}{\sqrt{2(n_3+1)}}\begin{pmatrix}-n_1+in_2\\ n_3+1\end{pmatrix} \quad \text{for} \quad \varepsilon = \frac{|\boldsymbol{\alpha}|}{4\pi}, \tag{13a}$$

$$\psi_\varepsilon(0) = \frac{1}{\sqrt{2(n_3+1)}}\begin{pmatrix}n_3+1\\ n_1+in_2\end{pmatrix} \quad \text{for} \quad \varepsilon = \frac{-|\boldsymbol{\alpha}|}{4\pi}, \tag{13b}$$

where

$$\boldsymbol{\alpha} = (\alpha_1, \alpha_2, \alpha_3),$$

$$n_i = \frac{\alpha_i}{|\boldsymbol{\alpha}|}.$$

Finally, I obtain the following algorithm for the calculation of the matrix element (11)

— Divide the interval $[0, 2\pi]$ into l subintervals.

— Calculate $\sigma_1(\phi_k) = [\exp(-i\alpha(\phi_k)\cdot\Sigma)]_1^i\sigma_i$ by the algorithm described in Section 2, where $\phi_k = \dfrac{2\pi k}{l}$. Then

$$A = \langle\psi_f|\sigma_1|\psi_i\rangle = \frac{1}{l}\sum_{k=1}^{l}\sigma_1(\phi_k)\exp[i(n_f-n_i+\varepsilon_i-\varepsilon_f)\phi_k]. \tag{14}$$

Taking the square of A we obtain the transition probability for the spontaneous emission from ψ_i to ψ_f.

We consider now the energy diagram calculated before [3, 4]. From Fig. 2 we have the following three transitions corresponding to the three-peaked spectrum of resonance fluorescence

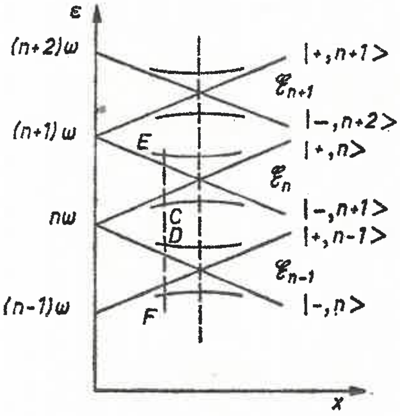

Fig. 2

(a) the transitions corresponding to the central component, *ED* and *CF*. Then $n_f-n_i = -1$ and $\varepsilon_i-\varepsilon_f = 0$. It is easy to see that the rates for these transitions are identical so we must calculate the transition probability only for one of them:

1° for the transition *CF*, $\varepsilon_i = \varepsilon_f = -\dfrac{|\alpha|}{4\pi}$, therefore from (13b) one has

$$\psi_f^\dagger(0)\sigma_1\psi_i(0) = n_1,$$
$$\psi_f^\dagger(0)\sigma_2\psi_i(0) = n_2,$$
$$\psi_f^\dagger(0)\sigma_3\psi_i(0) = n_3. \tag{15}$$

2° for the transition *ED*, $\varepsilon_i = \varepsilon_f = \dfrac{|\alpha|}{4\pi}$, therefore from (13a) one has

$$\psi_f^\dagger(0)\sigma_1\psi_i(0) = -n_1,$$
$$\psi_f^\dagger(0)\sigma_2\psi_i(0) = -n_2,$$
$$\psi_f^\dagger(0)\sigma_3\psi_i(0) = -n_3. \tag{16}$$

On the other hand, from previous section we have

$$\sigma_1(\phi) = [\exp(-i\alpha\cdot\Sigma)]^i_1\sigma_i,$$

so that we can write

$$\sigma_1(\phi) = f_1(\phi)\sigma_1+f_2(\phi)\sigma_2+f_3(\phi)\sigma_3. \tag{17}$$

Substituting (15)–(17) into (14) one obtains

$$\langle\psi_\mathrm{f}|\sigma_1|\psi_\mathrm{i}\rangle_\mathrm{CF} = -\langle\psi_\mathrm{f}|\sigma_1|\psi_\mathrm{i}\rangle_\mathrm{ED}.$$

Thus the matrix elements for the transitions *CF*, *ED* give the same transition probability.

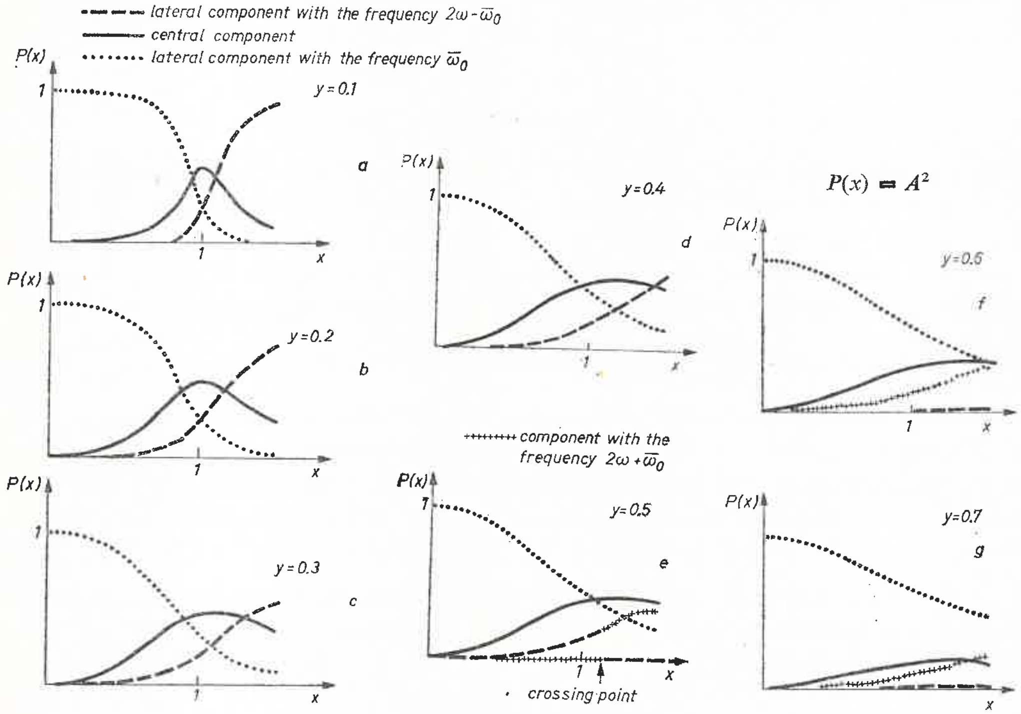

Fig. 3a–g

(b) the transition *CD* corresponding to the lateral component with the frequency $\bar{\omega}_0 = \dfrac{|\alpha|}{2\pi}$:

$$\varepsilon_\mathrm{i}-\varepsilon_\mathrm{f} = +\frac{|\alpha|}{2\pi}, \quad n_\mathrm{f}-n_\mathrm{i} = 0$$

(c) the transition *EF* corresponding to the lateral component with the frequency $2\omega-\bar{\omega}_0$:

$$\varepsilon_i-\varepsilon_f = \frac{|\alpha|}{2\pi}, \quad n_f-n_i = -2.$$

The numerical results are given in Fig. 3 for different values of y.

We see immediately from these calculations that the lateral components are not symmetrical and near the resonance $(\omega_0/\omega = x \approx 1)$ we have the height of one lateral component three times smaller than the height of the central component for certain value of y $(y \approx 0.5)$.

When we are going through the crossing points of the energy levels we must choose suitable branches of the wave functions involved in the formula for the transition probability. This remark becomes more important when we consider the atom dressed by an electromagnetic field with an arbitrary polarization because then the mechanism of the crossing is more complicated.

4. *Conclusions*

I have presented a new application of the phase representation: an algorithm based on this representation has been used in the study of the resonance fluorescence. Our formalism is useful, because of the two advantages of the phase representation in comparison with the standard approach. Firstly, the order of differential equations involved is lower (it is easy to see this if we take e.g. the operator $a^{\dagger}a$, which in analogy with the mechanical oscillator has the form $\frac{-d^2}{dx^2}+x^2$ in conventional representation, but $n+\frac{1}{i}\frac{\partial}{\partial\phi}$ in the phase representation). Secondly, because the phase ϕ varies only in the interval $[0, 2\pi]$, the boundary conditions for the problem considered are simpler than in other representations. This enables us to calculate numerically the matrix elements in a very simple way, which was illustrated in previous publications [3, 4] and here, so we hope that our approach may be very useful in the general time-dependent problem.

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