MATRIX METHOD OF SOLVING THE BLOCH EQUATIONS FOR THE EXCITING FIELD WITH ARBITRARY AMPLITUDE AND PHASE MODULATION

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A matrix method of solving the Bloch optical equations is proposed. Some particular solutions for modulated electric field are discussed. The advantages of the proposed method over the conventional approaches are demonstrated.

It is well known (see, for example, Ref. 1) that the equations describing the temporal behavior of the density matrix of the two–level system excited by a resonant field with allowance for the collisional and radiative relaxation phenomena in the dipole–interaction approximation form closed system of three first–order differential equations (Bloch equations). In the case of constant amplitude and phase of the exciting field, the coefficients in the Bloch equations are constant as well, and these equations can be easily solved.[2] In the case of sufficiently high amplitude of the exciting field, the solution for the level population difference of the system has the form of exponentially decaying oscillations with the Rabi frequency $\Omega_R = dE/\hbar$, where d is the dipole transition moment and E is the field amplitude. When the field amplitude becomes so much low that the inequality $\Omega_R < \Gamma_2$ holds true, where Γ_2 is the transition linewidth, the population difference decreases exponentially to its equilibrium value before even a single transition occurs. Consequently, the population difference in the two–level system excited by a resonant field oscillates only in the case of a sufficiently high field amplitude. For time–dependent amplitude and phase of the exciting field, the Bloch equations involve variable coefficients, and the general solution for arbitrary functions cannot be obtained. In the particular case of the periodic amplitude modulation in which the field amplitude is a periodic function of time (this field can be interpreted as a set of monochromatic components with equal amplitudes and the same spectral intervals between any adjacent components), the Bloch equations involve periodic coefficients and can be solved, as a rule, by application of the Floquet theorem. In accordance with this theorem, the solution of these equations can be represented by a series in the modulation frequency harmonics.[3] Fenenille and Schweighofer[4] as well as Toptygina and Fradkin[5] pioneered in applying the Floquet theorem to the solution of the Bloch equations for amplitude–modulated exciting field. Now this method is widely used for solving the Bloch equations with periodic coefficients.[6–10] The main drawbacks of application of the Floquet theorem are the following. First, harmonic amplitudes are represented in terms of continued fractions. This necessiates their numerical summation. Second, this theorem is inapplicable for a periodic modulation of the parameters of the exciting field.

The matrix method of solving the systems of linear differential equations used for the solution of the Bloch equations in the case of stochastic phase modulation[15] or periodic amplitude modulation[16] of the exciting field has no above–enumerated drawbacks. It allows one to derive the analytical solution of the Bloch equations not only for periodic coefficients but also for arbitrary coefficients and eliminates continued fractions.

Thus, let us consider the two–level system interacting with the field which can be written down in the form

$$\varepsilon(t) = E(t)\cos(\omega t + \Phi(t)) , \tag{1}$$

where $E(t)$ and $\Phi(t)$ are the field amplitude and phase being arbitrary functions of time and ω is the field cyclic frequency being equal to that of the transition between the levels of the system. In this case the equations describing the temporal behavior of the elements of the density matrix of this system in the dipole–interaction approximation in rotating coordinate system have the form[17]

$$\begin{aligned} \dot{\sigma}_{21}(t) &= -\Gamma_2\,\sigma_{21}(t) - \frac{i\,d\,\varepsilon(t)}{h}\,\mathrm{e}^{i\omega t}\,n(t) , \\ \dot{n}(t) &= -(n - n_0)\,\Gamma_1 - \frac{2\,i\,d\,\varepsilon(t)}{h}\left(\sigma_{21}(t)\,\mathrm{e}^{-i\omega t} - \sigma_{12}(t)\,\mathrm{e}^{i\omega t}\right) , \end{aligned} \tag{2}$$

where σ_{21} and σ_{12} are the slowly varying amplitudes of the off–diagonal elements of the density matrix, $n(t)$ is the level population difference, $n_0 = n_{(t=0)}$, and Γ_2^{-1} and Γ^{-1} are the relaxation times for polarization and population, respectively. By substituting relation (1) into Eq. (2), dropping the terms $\exp(\pm 2\,\omega\,t)$, and transforming to the conventional Bloch variables u and v after introduction of a new variable $\gamma = (u - i\,v)/2$ for which $\sigma^* = \sigma$, we obtain the Bloch equations for the exciting field given by relation (1) in the vector–matrix form

$$\frac{\mathrm{d}X}{\mathrm{d}t} = A(t)\,X(t) + L ,$$

where

$$X = \begin{vmatrix} u \\ v \\ n \end{vmatrix},\ A(t) = \begin{vmatrix} -\Gamma_2 & 0 & -a(t) \\ 0 & -\Gamma_2 & b(t) \\ a(t) & -b(t) & -\Gamma_1 \end{vmatrix},\ L = n_0\,\Gamma_1 \begin{vmatrix} 0 \\ 0 \\ 1 \end{vmatrix} , \tag{3}$$

$$a(t) = \Omega_R(t)\sin\varphi(t),\ b(t) = \Omega_R(t)\cos\varphi(t),\ \Omega_R(t) = dE(t)/\hbar.$$

Given that the commutator $[A(t), \mathrm{e}^{B(t)}]$ is zero, the formal solution of Eq. (3) can be written down in the form[18]

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$$X(t) = e^{B(t)} \left\{ \int_0^t e^{-B(t')} L dt' + \begin{vmatrix} 0 \\ 0 \\ n_0 \end{vmatrix} \right\} . \quad (4)$$

where $B(t) = \int A(t)dt$. To calculate $\exp[B(t)]$, let us make use of the Sylvester formula[3]

$$e^B = e^{\lambda_1} \frac{(B - \lambda_2 \mathbf{I})(B - \lambda_3 \mathbf{I})}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)} + e^{\lambda_2} \frac{(B - \lambda_1 \mathbf{I})(B - \lambda_3 \mathbf{I})}{(\lambda_2 - \lambda_1)(\lambda_2 - \lambda_3)} +$$

$$+ e^{\lambda_3} \frac{(B - \lambda_1 \mathbf{I})(B - \lambda_2 \mathbf{I})}{(\lambda_3 - \lambda_1)(\lambda_3 - \lambda_2)} , \quad (5)$$

where $\lambda_{1,2,3}$ are the eigenvalues of the matrix $B(t)$ and $\mathbf{I}$ is the unit matrix. Without any restrictions on the field amplitude under assumption that $\Gamma_2 = \Gamma_1 = \Gamma$ we derive relations for $\lambda_{1,2,3}$ in explicit forms:

$$\lambda_1 = -\Gamma t , \quad \lambda = -\Gamma t \pm i f(t) , \quad (6)$$

where $f^2(t) = \mathbf{I}_a^2(t) + \mathbf{I}_b^2(t)$, $\mathbf{I}_a(t) = \int_0^t a(t')dt'$, and

$$\mathbf{I}_b(t) = \int_0^t b(t')dt' .$$

After substitution of Eqs. (6) into Eq. (5), we derive

$$e^{\pm B(t)} = \frac{e^{\pm \Gamma t}}{f^2(t)} \times$$

$$\times \begin{vmatrix} \mathbf{I}_b^2 + \mathbf{I}_a^2 \cos f(t) & \mathbf{I}_a \mathbf{I}_b (1 - \cos f(t)) & \pm \mathbf{I}_a f(t) \sin f(t) \\ \mathbf{I}_b \mathbf{I}_a (1 - \cos f(t)) & \mathbf{I}_a^2 + \mathbf{I}_b^2 \cos f(t) & \pm \mathbf{I}_b f(t) \sin f(t) \\ \pm \mathbf{I}_a f(t) \sin f(t) & \pm \mathbf{I}_b f(t) \sin f(t) & f^2(t) \cos f(t) \end{vmatrix} . \quad (7)$$

By substituting Eqs. (7) into Eq. (4), we obtain the solution of the Bloch equations for the field with arbitrary amplitude and phase modulation in the form

$$X(t) = n_0 \Gamma \frac{e^{-\Gamma t}}{b^2(t)} \begin{vmatrix} \mathbf{I}_1(\mathbf{I}_b + \mathbf{I}_a^2 \cos f(t)) - \mathbf{I}_2 \mathbf{I}_a \mathbf{I}_b (1 - \cos f(t)) - (\mathbf{I}_3 + \Gamma^{-1}) \mathbf{I}_a f(t) \sin f(t) \\ \mathbf{I}_1 \mathbf{I}_a \mathbf{I}_b (1 - \cos f(t)) - \mathbf{I}_2 (\mathbf{I}_a^2 + \mathbf{I}_b^2 \cos f(t)) + (\mathbf{I}_3 + \Gamma^{-1}) \mathbf{I}_b f(t) \sin f(t) \\ (\mathbf{I}_1 \mathbf{I}_a + \mathbf{I}_2 \mathbf{I}_b) f(t) \sin f(t) + (\mathbf{I}_3 + \Gamma^{-1}) f^2(t) \cos f(t) \end{vmatrix} , \quad (8)$$

where $\mathbf{I}_1(t) = \int_0^t e^{\Gamma t'} \mathbf{I}_a(t') \frac{\sin f(t')}{f(t')} dt'$,

$$\mathbf{I}_2(t) = \int_0^t e^{\Gamma t'} \mathbf{I}_b(t') \frac{\sin f(t')}{f(t')} dt' , \quad \mathbf{I}_3(t) = \int_0^t e^{\Gamma t'} \cos f(t')\, dt' . \quad (9)$$

Now we can specify the condition under which Eq. (4) is correct. With the use of Eqs. (7) we derive

$$[A(t), e^{B(t)}] = e^{-\Gamma t} \frac{(a \mathbf{I}_b - b \mathbf{I}_a)}{f^2(t)} \times$$

$$\times \begin{vmatrix} 0 & f(t) \sin f(t) & \mathbf{I}_b (1 - \cos f(t)) \\ - f(t) \sin f(t) & 0 & \mathbf{I}_a (1 - \cos f(t)) \\ \mathbf{I}_a (1 - \cos f(t)) & \mathbf{I}_a (1 - \cos f(t)) & 0 \end{vmatrix} . \quad (10)$$

It then follows that the solutions of Eq. (3) given by formula (8) are correct only if one of the following conditions is fulfilled: (1) $t \to \infty$ and (2) $a \mathbf{I}_b = b \mathbf{I}_a$.

The first condition means that the solutions given by formula (8) are correct for arbitrary functions $E(t)$ and $\varphi(t)$ only in the steady state, after any relaxation process is damped out. Physically this means that formula (8) is correct for such values of t for which the inequately $\Gamma t \gg 1$ is true.

The second conditions means that formula (8) is correct for any t' from the interval $[0, t]$, but not for arbitrary $E(t)$ and $\varphi(t)$. Using explicit forms of the functions $a(t)$, $b(t)$, $\mathbf{I}_a(t)$, and $\mathbf{I}_b(t)$, we can show that the second condition is fulfilled in two cases. First, when the field amplitude and phase are nonmodulated, i.e., when $E(t) = \text{const}$ and $\varphi(t) = \text{const}$. Second, when $E(t)$ is arbitrary function while $\varphi(t) = \text{const}$. In the particular case of $\varphi = 0$, we derive

$$a(t) = \mathbf{I}_a(t) = 0,\ b(t) = \Omega_R(t),\ \mathbf{I}_b(t) = \int_0^t \Omega_R(t')dt',\ f(t) = \mathbf{I}_b(t),$$

$$\mathbf{I}_1(t) = 0,\ \mathbf{I}_2(t) = \int_0^t e^{\Gamma t'} \sin \mathbf{I}_b(t')dt',\ \mathbf{I}_3(t) = \int_0^t e^{\Gamma t'} \cos \mathbf{I}_b(t')dt' .$$

Thus, the solutions of the Bloch equations for arbitrary amplitude modulation of the exciting field have the form[19]

$$X(t) = n_0 \Gamma e^{-\Gamma t} \begin{vmatrix} 0 \\ -\mathbf{I}_2(t) \cos \mathbf{I}_b(t) + (\mathbf{I}_3(t) + \Gamma^{-1}) \sin \mathbf{I}_b(t) \\ \mathbf{I}_2(t) \sin \mathbf{I}_b(t) + (\mathbf{I}_3(t) + \Gamma^{-1}) \cos \mathbf{I}_b(t) \end{vmatrix} . \quad (11)$$

From Eq. (11), assuming $E(t) = \text{const}$, we can easily derive the well–known Torrey solutions[2] for monochromatic excitation. Solution (11) can be easily written out in an explicit form for periodically modulated $E(t)$.

Thus, solutions (8) and (11) obtained for the two–level system excited by the resonant field with arbitrary amplitude and phase modulation or with amplitude modulation alone are generalization of the existing solutions for the particular types of modulation of the field parameters (taking into account the above–mentioned assumptions).

In conclusion it should be noted that the problem of investigating the resonance interaction between the two–level system and the field with modulated parameters (in particular, stochastic field) was first formulated by Khermanovich and Kopvillem in the late 70 s.

REFERENCES

1. L. Allen and J. Elerly, *Optical Resonance and Two–Level Atoms* [Russian translation] (Mir, Moscow, 1975), 222 pp.
2. H.C. Torrey, Phys. Rev. **76**, 1059–1071 (1949).
3. V.A. Yakubovich and V.M. Starzhinskii, *Linear Differential Equations with Periodic Coefficients and their Applications* (Nauka, Moscow, 1972), 718 pp.

4. S. Fenenille and M.G. Schweighofer, J. Phys. **B9**, 2003–2011 (1976).
5. G.I. Toptygina and É.E. Fradkin, Zh. Eksp. Tekh. Fiz. **82**, 429–440 (1982).
6. N. Tsykada and T. Nakayama, Phys. Rev. **A25**, 964–973 (1982).
7. A.A. Mak, S.G. Przhebel'skii, and N.A. Chiger', Izv. Akad. Nauk SSSR, ser. Fiz. **47**, 1976–1977 (1983).
8. G.S. Agarwal and N. Nayak, JOSA , 164–169 (1984).
9. L.W. Hillman, I. Krasinski', K. Kosh, et al., JOSA **2**, 211–220 (1985).
10. G.S. Agarwal and N. Nayak, J. Phys. **B19**, 3385–3392 (1986).
11. G.S. Agarwal and N. Nayak, Phys. Rev. **A33**, 391–399 (1986).
12. H. Friedman and A.D. Wilson–Gordon, Phys. Rev. **A36**, 1333–1341 (1987).
13. A.D. Wilson–Gordon and H. Friedman, Phys. Rev. **A38**, 4087–4092 (1988).
14. S. Shakmajian, K. Koch, and C.K. Stroud, JOSA **5**, 2015–2024 (1988).
15. A.V. Alekseev and N.V. Sushilov, Zh. Eksp. Tekh. Fiz. **89**, 1951–1955 (1985).
16. A.V. Alekseev, A.V. Davydov, N.V. Sushilov, et al., J. Phys. **51**, 723–734 (1990).
17. J. Steinfield, ed., *Laser and Coherent Spectroscopy* [Russian translation] (Mir, Moscow, 1978), 629 pp.
18. I.A. Lappo–Danilevskii, *Application of Matrix Functions to Theory of Linear Systems of Ordinary Differential Equations* (State Techical and Theoretical Press, Moscow, 1957), 612 pp.
19. A.V. Alekseev, Yu.A. Zinin, and N.V. Sushilov, Opt. Spektrosk. **69**, 1245–1250 (1990).