COMMENT ON "SELF-INDUCED TRANSPARENCY IN THE EXCITON SYSTEM"

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ABSTRACT

Starting with the Hamiltonian for periodically arranged spin operators with a spin 1/2 we derive the equations of motion of the spin components under influence of an external field. In order to find the possibility of the self-induced transparency in this system a real solution of a pulse-like propagating field is sought. Following the previous work by Haken and Schenzle we obtain a solution which contains the influence of the dispersion and polariton effects of the system. In the absence of these effects the solution corresponds to a well-known hyperbolic-secant solution derived by McCall and Hahn. Discussion is made on the condition for SIT to be present.

I. INTRODUCTION

Self-induced transparency (SIT) first studied by McCall and Hahn[1) is a phenomenon that a short pulse of coherent light above a critical power threshold propagates through a resonantly absorbing medium with anomalously low energy loss. In practice the critical power threshold is characterized by field strengths sufficiently intense enough for 2π pulse, and also the initially applied pulse width should be shorter than any characteristic damping times of the excited state of the medium.

Recently this problem has been studied in connection with the soliton problem.[2) From this point of view the SIT in the exciton system, if exists, is understood as the propagation of distortionless light-pulse through the dispersive material interacting with light fields nonlinearly. In general, in a linear and dispersionless system a pulse-like traveling wave can always exist. However, if the dispersion is introduced to this system, then the various Fourier components of the initial wave will propagate at different velocities, and therefore the initial wave form is not sustained. On the other hand, when a nonlinear interaction is introduced in the dispersion-less system, the pulse energy is continually dissipated into higher harmonic modes by the harmonic generation. Therefore, if SIT is present in the system a delicate balance should exist between these effects.

The study of SIT in the exciton system has been attracting much theoretical interest,[3~5] and what has been shown so far is that the polariton effect would prevent and the saturation effect would increase the possibility of SIT in the exciton system. The main differences from the case of two-level atoms are; 1) there exists the energy dispersion in the exciton system, and the macroscopic polarization which appears in Maxwell's equations should be defined in some artificial way, and 2) the polariton effect would play an essential role in the propagation of light fields in the medium. The presence of these effects and the nonlinear polarization indicates that if SIT exists in this system there should be some critical restrictions on the field intensity and the forms of the exciton dispersion and interaction with light fields.

In the present work, instead of considering the exciton explicitly, we consider a system consisting of two-level atoms locating periodically at lattice sites l's. By assigning to each atom the spin angular momentum operator σ_l with a spin 1/2 and introducing spin-spin interactions between atoms at different lattice sites we derive the equations of motion for σ_l^+, σ_l^- and σ_l^z. We define the macroscopic polarization from these spin operators averaged over a region small compared with the light wavelength. The interaction between light fields and the medium is expressed in terms of σ_l^+, σ_l^- and the circularly polarized vector potential $\tilde{\vec{A}}_\pm$ of propagating light fields. After making usual transformations we derive the equation of motion for the amplitude and phase of $\tilde{\vec{A}}_\pm$ by eliminating all variables associated with the medium. We, then, seek for a distortionless propagating pulse solution of light field and derive formally the conditions for SIT to be present in the system.

II. EQUATIONS OF MOTION

Letting a_l^+, a_l^- and b_l^+, b_l^- be creation and annihilation operators of the ground and excited states of the atom at the l-th site, we define the spin operators σ_l^+ and σ_l^- by

$$\sigma_l^+ = b_l^+ a_l \quad \text{and} \quad \sigma_l^- = a_l^+ b_l \ . \tag{1}$$

With the usual commutation relations σ_l^z is then given by

$$2\sigma_l^z = [\sigma_l^+, \ \sigma_l^-] = b_l^+ b_l - a_l^+ a_l \ . \tag{2}$$

In terms of these spin operators the Hamiltonian of our system is written as[5)]

$$H = \sum_l \omega_o \sigma_l^z + \frac{1}{2} \sum_{ll'}{}' J(l-l')(\sigma_l^+ + \sigma_l^-)\ (\sigma_{l'}^+ + \sigma_{l'}^-)$$

$$- \frac{i}{c} \vec{\mu}\omega_o \cdot \sum_l \vec{A}(l,t)\ (\sigma_l^+ - \sigma_l^-), \tag{3}$$

where $J(l-l')$ is the interaction potential between two atoms at the l-th and l'-th sites. ω_o and μ are the energy difference and the atomic dipole moment between the two states, and $\vec{A}(l,t)$ represents the vector potential of the external field.

In order to write the equations of motion in the rotating frame we express the spin operators $\sigma_l^{\pm}$ and the vector potentials $\vec{A}$ as

$$\sigma_l^{\pm} = \tilde{\sigma}_l^{\pm} \exp \pm i(\omega t - kl) \quad ,$$

and

$$\vec{A}_l^{\pm} = \tilde{\vec{A}}_{\pm}(l,t) \exp \mp i(\omega t - kl) \ ,$$

where $\tilde{\sigma}_l^{\pm}$ and $\tilde{\vec{A}}_{\pm}(l,t)$ are slowly varying functions of t and l in comparison to the exponential part. Then the equations of motion of the spin operators are written as

$$\frac{\partial}{\partial t}\tilde{\sigma}_l^{\ +} = +i\Delta\omega\tilde{\sigma}_l^{\ +} + \frac{1}{c}\omega_o\vec{\mu}_+\tilde{\vec{A}}_-(l,t)\sigma_l^{\ z} - 2i\sigma_l^{\ z}\Sigma' J(l-l')e^{-ik(l'-l)}\tilde{\sigma}_{l'}^{\ +} \quad , \tag{4}$$

$$\frac{\partial}{\partial t}\tilde{\sigma}_l^{\ -} = -i\Delta\omega\tilde{\sigma}_l^{\ -} + \frac{1}{c}\omega_o\vec{\mu}_-\tilde{\vec{A}}_+(l,t)\sigma_l^{\ z} + 2i\sigma_l^{\ z}\Sigma' J(l-l')e^{ik(l'-l)}\tilde{\sigma}_{l'}^{\ -} \quad , \tag{5}$$

$$\frac{\partial}{\partial t}\sigma_l^{\ z} = -\frac{\omega_o}{2c}\{\vec{\mu}_-\tilde{\vec{A}}_+(l,t)\tilde{\sigma}_l^{\ +} + \vec{\mu}_+\tilde{\vec{A}}_-(l,t)\tilde{\sigma}_l^{\ -}\}$$

$$- i\{\tilde{\sigma}_l^{\ +}\Sigma' J(l-l')\tilde{\sigma}_{l'}^{\ -}e^{ik(l'-l)} - \tilde{\sigma}_l^{\ -}\Sigma' J(l-l')\tilde{\sigma}_{l'}^{\ +}e^{-ik(l'-l)}\} \quad , \tag{6}$$

where fast oscillating terms are neglected. Here, we have put $\Delta\omega = \omega_o$- and $\vec{\mu}_\pm = \vec{\mu}_x \pm i\vec{\mu}_y$. From these equations we can obtain the conservation law of the medium by multiplying $\tilde{\sigma}_l^{\ -}$ from the right of eq. (4) and $\tilde{\sigma}_l^{\ +}$ from the left of eq. (5) and making use of eq. (6) as follows;

$$\frac{\partial}{\partial t}(\tilde{\sigma}_l^{\ +}\tilde{\sigma}_l^{\ -}) + \frac{\partial}{\partial t}(\sigma_l^{\ z}\sigma_l^{\ z} - \sigma_l^{\ z}) = 0 \quad . \tag{7}$$

Concerning the last terms of eqs. (4)-(6) we treat $\tilde{\sigma}_{l'}^{\ \pm}$ as to be classical quantities and expand them formally as

$$\tilde{\sigma}_{l'}^{\ \pm} = \tilde{\sigma}_l^{\ \pm} + \frac{\partial\tilde{\sigma}_l^{\ \pm}}{\partial\vec{l}}(\vec{l}'-\vec{l}) + \cdots\cdots \quad .$$

Then, the summation over l' can be approximated by

$$\sum_{l'}{}' J(l-l')\tilde{\sigma}_{l'}^{\ \pm} e^{\mp i\vec{k}(\vec{l}'-\vec{l})} \simeq \tilde{J}(\pm\vec{k})\,\tilde{\sigma}_l^{\ \pm} - i\vec{v}(\pm\vec{k})\frac{\partial\sigma_l^{\ \pm}}{\partial\vec{l}} \quad , \tag{8}$$

where the Fourier transform $\tilde{J}(q)$ and the exciton velocity $\vec{v}(\vec{k})$ are defined by

$$J(l-l') = \frac{1}{N}\sum_q \tilde{J}(\vec{q})\, e^{-i\vec{q}(\vec{l}-\vec{l}')} \quad , \tag{9}$$

and

$$\vec{V}(\pm\vec{k}) = \left.\frac{\partial\tilde{J}(\vec{q})}{\partial\vec{q}}\right|_{q=\pm k} \quad .$$

It should be noticed that $\tilde{J}(\vec{k}) = \tilde{J}(-\vec{k})$ since $J(\vec{l}-\vec{l}')$ depends on $|\vec{l}-\vec{l}'|$

only. The number of lattice sites N is defined in the next section.

III. TRANSFORMATION TO MACROSCOPIC EQUATION

In order to define the macroscopic polarization we introduce a transformation defined by

$$< \tilde{\sigma} >_{\vec{z}} = \sum_{l<\Delta V} \Delta(\vec{z} - \vec{l})\tilde{\sigma}_l \quad , \tag{10}$$

where the summation over l is restricted within a volume ΔV. The dimension of ΔV is taken to be smaller than the wavelength of the light field and yet to be large enough so that the energy dispersion of exciton still has a meaningful effect. In the present case the transformation (10) can be interpreted as defining the average value of $\tilde{\sigma}_l^{\pm}$ or σ_l^z at a point z, since $\tilde{\sigma}_l^{\pm}$ as well as σ_l^z is a slowly varying function of l. Now the Fourier transform of eq. (9) is to be defined in ΔV, and N in eq. (9) should be the number of lattice sites inside the volume ΔV.

With the above procedure and also the assumption that the average of product can be replaced by the product of the average divided by N the equations of motion (4)-(6) are written as

$$\frac{\partial}{\partial t} < \tilde{\sigma}^+ > = +i\Delta\omega < \tilde{\sigma}^+ > + \frac{1}{c}\omega_o \vec{\mu}_+ \vec{\tilde{A}}_- < \sigma^z >$$
$$- 2i \frac{1}{N} < \sigma^z > < \tilde{\sigma}^+ > \tilde{J}(\vec{k}) - 2 \frac{1}{N} \vec{v}(\vec{k}) < \sigma^z > \frac{\partial}{\partial \vec{z}} < \tilde{\sigma}^+ > \quad , \tag{11}$$

$$\frac{\partial}{\partial t} < \tilde{\sigma}^- > = -i\Delta\omega < \tilde{\sigma}^- > + \frac{1}{c}\omega_o \vec{\mu}_- \vec{\tilde{A}}_+ < \sigma^z >$$
$$+ 2i \frac{1}{N} < \sigma^z > < \tilde{\sigma}^- > \tilde{J}(-\vec{k}) + 2 \frac{1}{N} \vec{v}(-\vec{k}) \frac{\partial}{\partial \vec{z}} < \tilde{\sigma}^- > < \sigma^z > \quad , \tag{12}$$

$$\frac{\partial}{\partial t} < \sigma^z > = -\frac{\omega_o}{2c} \{ \vec{\mu}_- \vec{\tilde{A}}_+ < \tilde{\sigma}^+ > + \vec{\mu}_+ \vec{\tilde{A}}_- < \tilde{\sigma}^- > \} - \frac{1}{N} \vec{v}(\vec{k}) \frac{\partial}{\partial \vec{z}} (< \tilde{\sigma}^+ > < \tilde{\sigma}^- >) . \tag{13}$$

Also the conservation law given by eq. (7) is written as

$$\frac{1}{N}\frac{\partial}{\partial t}(<\tilde{\sigma}^{+}>\cdot<\tilde{\sigma}^{-}>) + \frac{1}{N}\frac{\partial}{\partial t}<\sigma^{z}>^{2} - \frac{\partial}{\partial t}<\sigma^{z}> = 0 \quad . \tag{14}$$

Making use of eq. (10) we derive approximately the wave equation satisfied by $\vec{\tilde{A}}_{\pm}(z,t)$ as follows;

$$\frac{\partial}{\partial z}\vec{\tilde{A}}_{\pm} + \frac{\eta^{2}\omega}{kc^{2}}\frac{\partial}{\partial t}\vec{\tilde{A}}_{\pm} \pm \frac{i}{2}\frac{1}{k}(k^{2} - \frac{\eta^{2}}{c^{2}}\omega^{2})\vec{\tilde{A}}_{\pm} = \frac{2\pi\omega}{kc}\vec{\mu}_{\pm}<\tilde{\sigma}^{\mp}> \quad . \tag{15}$$

Here η is the index of refraction of the medium. The third term in the left hand side, which is neglected in the work of Haken and Schenzle, should be included self-consistently when the polariton effect is considered.

To obtain a solution of propagating fields we introduce a variable ζ by $\zeta = t - z/v$, where v is the velocity of the propagating fields and equals c/η in the case of nondispersive media. Then $\vec{\tilde{A}}_{\pm}$ are given as functions of ζ, and eq. (15) is transformed to

$$\frac{d}{d\zeta}\vec{\tilde{A}}_{\pm}(\zeta) \mp \frac{i}{2k\Delta}(k^{2} - \frac{\eta^{2}}{c^{2}}\omega^{2})\vec{\tilde{A}}_{\pm}(\zeta) = -\frac{2\pi\omega}{kc\Delta}\vec{\mu}_{\pm}<\tilde{\sigma}^{\mp}>_{\zeta} \quad , \tag{16}$$

where $\Delta = \frac{1}{v} - \frac{\eta^{2}\omega}{kc^{2}}$. For convenience we put

$\beta = \frac{1}{2k\Delta}(k^{2} - \frac{\eta^{2}\omega^{2}}{c^{2}})$ and $\gamma = \frac{2\pi\omega}{kc\Delta}$, and transform $\vec{\tilde{A}}_{\pm}$ into

$$\vec{\tilde{A}}_{\pm}(\zeta) = \vec{A}(\zeta)\ \exp \pm i(\beta\zeta + \phi(\zeta)) \quad . \tag{17}$$

When β is independent of ζ, it follows immediately from eq. (16) that

$$\begin{aligned}\frac{d}{d\zeta}(\vec{A}\,e^{\pm i\phi}) &= (\dot{\vec{A}} \pm i\vec{A}\dot{\phi})\,e^{\pm i\phi} \\ &= -\gamma\vec{\mu}_{\pm}\,e^{\mp i\beta\zeta}<\tilde{\sigma}^{\mp}>_{\zeta} \quad ,\end{aligned} \tag{18}$$

where we put $d/d = \dot{}$. We use eq. (18) to relate the macroscopic value of $<\tilde{\sigma}^{\pm}>_{\zeta}$ with the amplitude of the vector potential. It should be noticed that by introducing ζ the equations of motion (11) ~ (13) are expressed as ordinary differential equations.

IV. SEEK FOR SOLUTION OF SIT

We now seek for a real solution of $\vec{A}(\zeta)$ from eqs. (11)~(14) and (18). First, substitution of eq. (18) into eq. (13) gives

$$\frac{d}{d\zeta} < \sigma^z > = N\lambda^2 \frac{d}{d\zeta} \vec{A}^2 + \xi \frac{d}{d\zeta} Y \quad . \tag{19}$$

Here Y is defined by $Y = (<\tilde{\sigma}^+ > \cdot < \tilde{\sigma}^- >)/N$, which is equal to $N\lambda^4/g^2 \times(\dot{\vec{A}}^2 + \vec{A}^2\dot{\phi}^2)$ by means of eq. (18). For convenience we have put* $\lambda^2 = \omega_o/2N\gamma c$, $g = \omega_o \, |\mu| \, /2c$ and $\xi = v(\vec{k})/v$. Equation (19) immediately gives

$$\frac{1}{N} < \sigma^z > = \lambda^2\vec{A}^2 + \xi \frac{1}{N} Y + C_1 \quad . \tag{20}$$

Also from eqs. (14) and (18) it follows that

$$\frac{d}{d\zeta} Y = \frac{d}{d\zeta} < \sigma^z > - \frac{1}{N} \frac{d}{d\zeta} < \sigma^z >^2 \quad , \tag{21}$$

and integration of eq. (21) gives

$$Y = < \sigma^z > - \frac{1}{N} (<\sigma^z >)^2 + C_2 \quad . \tag{22}$$

The constant C_2 is chosen so that $Y = A = 0$ at $\zeta = -\infty$. From eqs. (20) and (22), and the definition of Y it follows that

$$(\dot{\vec{A}})^2 = \vec{A}^2[-(\dot{\phi})^2 + \frac{g^2}{\lambda^2} \{ 1-\lambda^2\vec{A}^2-2C_1 \} \{ 1+\xi (1-2\lambda^2\vec{A}^2-2C_1) \}] \; . \tag{23}$$

Here we retain the terms up to the first order in ξ.

By multiplying eq. (11) by $\gamma\vec{\mu}_-\tilde{\vec{A}}_+$ and eq. (12) by $\gamma\vec{\mu}_+\tilde{\vec{A}}_-$, and using eqs. (13) and (18) we obtain for a pulse-shape solution of $\vec{A}$

$$\vec{A}^2(\dot{\phi}+\frac{\beta}{2}) = - \frac{(1-\xi)}{2\lambda^2} \{\Delta\omega (\frac{< \sigma^z >}{N} - C_1) - \tilde{J}(k) \{(\frac{< \sigma^z >}{N})^2 - C_1^2 \} \} \; . \tag{24}$$

* These quantities have the same meaning as in reference 3), but differ by a factor 2.

Substituting eq. (20) we have for $\dot{\phi}$

$$\dot{\phi} = -\frac{\beta}{2} - \frac{\Delta\omega}{2}\{1-\xi(\lambda^2\vec{A}^2+2C_1)\} + \frac{1}{2}\tilde{J}(\vec{k})\{2C_1+(1+\xi)\lambda^2\vec{A}^2\}\ . \tag{25}$$

Finally, by eliminating $\dot{\phi}$ from eqs. (23) and (25), the equation satisfied by $\vec{A}$ is derived as follows;

$$(\dot{\vec{A}})^2 = \vec{A}^2\left[\frac{1}{\tau_o^2} - \frac{1}{\tau_1^2}\lambda^2\vec{A}^2 + \frac{1}{\tau_2^2}\lambda^4\vec{A}^4\right]\ . \tag{26}$$

The constants, τ_o, τ_1 and τ_2, are defined as

$$\frac{1}{\tau_o^2} = \frac{g^2}{\lambda^2}(1-2C_1)(1+\xi(1-2C_1)) - \frac{1}{4}(\beta+\Delta\omega-2C_1\tilde{J}(\vec{k}))^2$$
$$-2C_1\xi\Delta\omega(\beta+\Delta\omega-2C_1\tilde{J}(\vec{k}))\ , \tag{27}$$

$$\frac{1}{\tau_1^2} = \frac{g^2}{\lambda^2}(1+3\xi(1-2C_1)) + \frac{1}{2}(\beta+\Delta\omega-2C_1\tilde{J}(\vec{k}))\tilde{J}(\vec{k})$$
$$+\frac{1}{2}\xi\{(\beta+\Delta\omega-2C_1\tilde{J}(\vec{k}))(\Delta\omega+\tilde{J}(\vec{k}))-C_1\Delta\omega\tilde{J}(\vec{k})\}\ , \tag{28}$$

$$\frac{1}{\tau_2^2} = 2\xi\frac{g^2}{\lambda^2} - \frac{1}{4}\{(\tilde{J}(\vec{k}))^2 + 2\xi\tilde{J}(\vec{k})(\Delta\omega+\tilde{J}(\vec{k}))\}\ . \tag{29}$$

It is clear from eq. (26) that if τ_o^2 is negative there is no real solution of $\vec{A}$. In order to have a pulse-shape solution of $\vec{A}$ the following conditions are found to be required;

$$D = \left(\frac{1}{\tau_1^2}\right)^2 - 4\left(\frac{1}{\tau_o^2}\right)\left(\frac{1}{\tau_2^2}\right) > 0, \text{ and } \frac{1}{\tau_1^2} + \sqrt{D} > 0\ . \tag{30}$$

If these conditions are satisfied, then the solution is given by

$$\lambda^2\vec{A}^2 = \frac{2(1/\tau_o)^2}{(1/\tau_1)^2 + \sqrt{D}\cosh\frac{2}{\tau_o}(\zeta-\zeta_o)}\ . \tag{31}$$

It is noticed that if $(1/\tau_2)^2 = 0$ the above solution becomes identical to that obtained for the two-level system.

V. INITIAL CONDITION AND POLARITON EFFECT

We discuss briefly the polariton effect in connection with choice of the integration constant C_1, which is to be determined by the initial condition of the medium. If we assume for simplicity the weak coupling between adjacent atoms as well as between exciton and light fields, we can put $\xi = 0$ and $\tilde{J}(\vec{k}) = 0$. Then, it follows from eq. (20) that

$$\frac{1}{N} < \sigma^z > = \lambda^2 \vec{A}^2 + C_1 \quad . \tag{32}$$

In general the first term of the right-hand-side is much smaller than unity. Also, the value of $< \sigma^z >/N$ is between -1/2 and 1/2 in the present model. If the medium is in the ground state initially, then C_1 is taken to be -1/2, and this case with an additional condition $\beta = 0$ corresponds to the case treated by Haken and Schenzle.[3]

In the present formalism the polariton effect is reflected in β. As is usually done in the weak field polariton case, if we put $\frac{d}{d\zeta} = 0$ in eqs. (11), (12) and (16), then it follows that

$$\beta = \frac{1}{\Delta\omega} (-2C_1 - \lambda^2 \vec{A}^2) \frac{g^2}{\lambda^2} \quad . \tag{33}$$

It is clear from eq. (33) that the present case for $C_1 = -1/2$ and $\lambda^2 \vec{A}^2 \sim 0$ is essentially similar to the case treated by Hanamura.[4] Furthermore, eq. (33) shows that in the case $C_1 = -1/2$ and $\Delta\omega (= \omega_0 - \omega) > 0$ the polariton effect is reduced as the field intensity increases. This corresponds to the giant polariton case, which is numerically analyzed by Inoue.[5] If this is the case, β becomes a function of ζ through $\vec{A}(\zeta)$ and eq. (18) will be modified by replacing $\dot{\phi}$ by $(\dot{\phi} + \dot{\beta}\zeta)$. However, taking $C_1 = -1/2$ in eq. (33) we obtain from eq. (27)

$$\frac{1}{\tau_0{}^2} = \left(\frac{3}{2} + \lambda^2 \vec{A}^2\right) \frac{g^2}{\lambda^2} - \frac{1}{4} \left\{ \frac{1}{(\Delta\omega)^2} \frac{g^4}{\lambda^4} (1 - \lambda^2 \vec{A}^2) + (\Delta\omega)^2 \right\} \quad , \tag{34}$$

which can be either negative or positive depending on g, λ, and $\Delta\omega$, and can find that if $1/\tau_o^2$ is positive for the weak field case, then it also holds for the giant polariton case.

VI. CONCLUSION

We derived formally the solution of a pulse-like propagating wave in the system with the dispersion and polariton effects, although the possibility of its occurrence in a real system would require more profound analyses of the parameters used. It was shown that SIT possibly takes place under some conditions.

REFERENCES

1) S. L. McCall and E. L. Hahn: Phys. Rev. 183 (1969) 457.
2) See, *e.g.*, A. C. Scott *et al.* : Proc. IEEE 61 (1973) 1443.
3) H. Haken and A. Schenzle: Z. Phys. 258 (1973) 231.
4) E. Hanamura: J. Phys. Soc. Japan 37 (1974) 1554.
5) M. Inoue: J. Phys. Soc. Japan 37 (1974) 1560.