A model of excitonic nonlinearity in semiconductors

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A model is proposed to describe the nonlinear excitonic resonance in semiconductors (specifically in quantum wells). This model includes correlation effects in the simplest way possible, along with phenomenological parameters that characterize the shift of the exciton resonance and its relaxation. The equations that describe these phenomena are solved in various cases.

1. INTRODUCTION

Nonlinear optics properties of semiconductor layered structures have been the subject of a large number of papers (see, e.g., the review in Ref. 1), in which exciton effects have also been studied. These exciton effects are the topic of this paper.

We first pause to briefly discuss the difficulties that arise in solving this problem. The objects of interest are excitons, which can be created in a quantum well; our task is to find the nonlinear response of this system to an electromagnetic field with a frequency close to the exciton resonance.

Ideally, everything about this problem should be clear. Excitons are generated by a resonance field; provided that the exciton concentration is not too large we can forget about their internal structure and treat them as indivisible particles of Bose type which interact among themselves in some fashion. It is this interaction that is the source of the nonlinearity.

One complicating circumstance is the possibility of forming a biexciton. However, for excitons created by irradiation with a specific polarization, which are all of the same type, it is not necessary to discuss biexcitons because single-type excitons apparently repel each other (recall the conditions for formation of a hydrogen molecule). This repulsion has a radius on the order of the size of an exciton, and may plausibly be treated approximately as a hard-sphere interaction; as for the Van der Waals attraction, it is ordinarily weak and need not be included (once again we recall the situation for hydrogen).

Thus, the following model suggests itself: Bose particles with a repulsion plus their interaction with an external field (both creation of excitons and their destruction by this field). This model has one significant deficiency: it can be analyzed only in the limit of low-exciton concentrations (the gas limit), i.e., for sufficiently small resonant field intensities. The situation is especially unfavorable for the gas approximation in the two-dimensional case (i.e., excitons in a quantum well), where the requirement for this approximation to be applicable is that the gas parameter be not simply small, but exponentially small.

Usually such problems are solved within the Hartree–Fock approximation, an approach that is discussed in considerable detail in the review mentioned previously.[1] In this approach the role of correlation effects remains unclear; we would like to understand this, and also obtain at least some idea of the role of relaxation.

In this paper a model is proposed in which correlation effects are included in the simplest possible way. A further advantage of this model is the ease with which relaxation effects can also be taken into account, as is also done here. It would appear that while the model reproduces the usual overall picture of what happens during the excitation of exciton resonances, it also allows us to obtain a number of results without a great deal of work.

The equations of motion for the model are formulated in Sec. 2. In the sections that follow, various problems are discussed: the response to a strong resonance field (Sec. 3), bistability (Sec. 4), and the properties of a weak probe signal superposed on an intense pump (Sec. 5). Finally, additional considerations and comments are presented in the last section.

In order to avoid misunderstandings, I would like to emphasize that whereas the review (Ref. 1) describes results of investigations which (although very approximate) were used over a wide interval of frequencies and external field intensities, the problem treated here is narrower in scope: only the neighborhood of the exciton resonance is investigated, and for intensities that are low enough that only excitons in the ground state are present. Thus it is permissible to ignore, e.g., the creation, of unbound electron-hole pairs.

2. THE MODEL

Let us begin with a model system that only remotely resembles the one of interest to us. We will take from this system only those things that serve to further our goal, and add whatever it does not encompass later. The result will be the model we want.

Let us consider an assembly of two-level atoms described by the Dicke model in an external field. Such a system is more appropriate for describing Frenkel excitons than Wannier–Mott excitons. However, the Dicke model has one attractive property: it is impossible to place two excitations on a single atom. This property mimics the impenetrability of the excitons in our problem, which are treated as hard spheres. It is this trait of an ensemble of two-level atoms that is of interest to us; in fact, it is the only trait that is really necessary for formulating the model.

To each atom we may associate creation and annihilation operators a_m^+, a_m (where m labels the atom) with Fermi commutation relations

$$a_m{}^+ a_m + a_m a_m{}^+ = 1, \tag{2.1}$$

while the operators of differing atoms commute. Then the ground state of an atom corresponds to the vacuum function $|0\rangle$, while the excited states are the functions $a_m^+|0\rangle$. The Hamiltonian of such a system has the form

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$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_{int}, \tag{2.2}$$

where $\mathcal{H}_0$ is the Hamiltonian of the free atoms:

$$\mathcal{H}_0=\mathcal{E}_0\sum_m a_m^+ a_m \tag{2.3}$$

(here $\mathcal{E}_0$ is the spacing between levels), and $\mathcal{H}_{int}$ describes their interaction with an electromagnetic field; in the dipole approximation

$$\mathcal{H}_{int}=-\hat{\mathbf{d}}\mathbf{E}, \tag{2.4}$$

where $\hat{\mathbf{d}}$ is the dipole moment operator of the system; $\mathbf{E}$ is the electric field of a wave:

$$\hat{\mathbf{d}}=\mathbf{d}\sum_m (a_m^+ + a_m),$$
$$\mathbf{E}=\mathbf{E}_0\cos\omega t. \tag{2.5}$$

Here $\mathbf{d}$ is the matrix element of the atomic dipole moment (we may treat it as a real quantity). The external field is assumed to be monochromatic (the generalization to the arbitrary case is obvious), and we will not take into account its dependence on coordinates (i.e. the wavelength is large compared to the distance between adjacent atoms).

Substituting Eqs. (2.3)–(2.5) into (2.2) and retaining only the resonance terms, we obtain the Hamiltonian in the resonance approximation:

$$\mathcal{H}=\mathcal{E}_0\hat{n}-{}^1/_2\Delta(a^+e^{-i\omega t}+ae^{i\omega t}), \tag{2.6}$$

where we introduce the notation

$$\Delta=\mathbf{d}\mathbf{E}_0, \quad \hat{n}=\sum_m a_m^+ a_m,$$
$$a=\sum_m a_m, \quad a^+=\sum_m a_m^+. \tag{2.7}$$

Let us investigate that part of the Hamiltonian (2.6) that describes the interaction with the field. Out of all the atomic degrees of freedom only one is described by the operator a, which may be thought of as a collective operator corresponding to an excitation that is "smeared out" over all the atoms (and is not localized on an individual atom) according to the definition of the operators a^+, a in (2.7). The same picture also holds for the exciton when it is coupled to the electromagnetic field while in a single definite state, with a momentum that coincides with the photon momentum. In addition, only one excitation is allowed on one atom, which recalls the impenetrability of excitons treated as hard spheres. Therefore, if the concentration of atoms is such that the distance between neighbors is on the order of the size of the exciton, the system of excitations of such atoms corresponds qualitatively to our excitonic system.

As for the first operator on the right side of Eq. (2.6), its correspondence with the total exciton energy is obvious. However, this model does not predict any shift in the resonant frequency as the concentration of excitations increases. Since such shifts should appear in a system of excitons, we are forced to include them artificially if we want to describe the exciton system (more of this later).

Let us transform the Hamiltonian (2.6) in such a way that only one collective degree of freedom appears explicitly, as we discussed earlier, which we represent by operators a^+, a. This implies that only states of the type

$$\Phi_n=A_n(a^+)^n|0\rangle, \tag{2.8}$$

are of interest to us, where n is the number of excitations, $0\leqslant n\leqslant N$ (N is the number of atoms; how all this applies to excitons will be clear in what follows). The set of functions (2.8) with all possible values of n is by no means complete; however, it is important that these functions form a closed subspace, i.e., when the Hamiltonian (2.6) acts on the function (2.8) we obtain a function of the same type.

We now present the results of how the various operators in (2.6) act on the function (2.8). First, however, we must calculate the normalization coefficient A_n; it is not difficult to verify that

$$A_n=\left[\frac{(N-n)!}{n!\,N!}\right]^{1/2}. \tag{2.9}$$

Using this expression we obtain

$$a^+\Phi_n=[(n+1)(N-n)]^{1/2}\Phi_{n+1},$$
$$a\Phi_n=[n(N-n+1)]^{1/2}\Phi_{n-1}, \tag{2.10}$$
$$\hat{n}\Phi_n=n\Phi_n.$$

These relations coincide with the relations for the angular momentum operator $\hat{\mathbf{L}}$, with the following substitutions:

$$a^+\to\hat{L}^{(+)}=\hat{L}_x+i\hat{L}_y,$$
$$a\to\hat{L}^{(-)}=\hat{L}_x-i\hat{L}_y, \tag{2.11}$$
$$\hat{n}\to L+\hat{L}_z, \quad L=N/2.$$

As a result the Hamiltonian (2.6) in the subspace of function (2.8) can be written in the following fashion:

$$\mathcal{H}\to\mathcal{E}_0 L-\hat{\mathbf{L}}\mathbf{H}_0. \tag{2.12}$$

where we have introduced the "magnetic field"

$$\mathbf{H}_0=(\Delta\cos\omega t,\ \Delta\sin\omega t,\ -\mathcal{E}_0). \tag{2.13}$$

The origin of the moment $L=N/2$ is clear: as we started with a collection of two-level atoms, each of which has a moment, as it were, of spin 1/2. As we see, there is an interaction with the external field only in the state with maximum possible total spin $N/2$.

Although the Hamiltonian (2.12) forms the basis for the model, it is not yet the whole model. In what follows it is convenient to work not with the Hamiltonian but with the equations of motion, which coincide with the equations for magnetic resonance:

$$d\mathbf{L}/dt=[\mathbf{L}\mathbf{H}_0]. \tag{2.14}$$

Here the moment can even be considered a classical vector.

We now can proceed to correct Eq. (2.14), using terms that (1) take into account the resonance shift mentioned previously and (2) mediate relaxation processes (the necessity and possibility of including these is due to the convenience of the equation of motion).

We will include the resonance shift phenomenologically. The dependence of the system energy on exciton number n can be described by a correction to the Hamiltonian

$$\delta\mathcal{H}=f(\hat{n}), \tag{2.15}$$

where the form of the function f is unknown, so that in spe-

cific calculations we must make some assumptions about it (in Secs. 4 and 5). The equation of motion including the correction (2.15) has the same form as (2.14) but with the replacements

$$\mathscr{E}_0 \to \mathscr{E} = \mathscr{E}_0 + df(n)/dn, \qquad \mathbf{H}_0 \to \mathbf{H} = (\Delta \cos \omega t, \Delta \sin \omega t, -\mathscr{E}). \tag{2.16}$$

Now for the relaxation. In our case of exciton resonance it is apparently more correct to approach this problem by describing the relaxation in the Landau–Lifshitz form (see, e.g., Ref. 2); in this picture the magnitude of the moment $|\mathbf{L}|$ remains constant, while the change of the vector $\mathbf{L}$ in the relaxation process is orthogonal to $\mathbf{L}$ and directed along the magnetic field $\mathbf{H}$. If we adopt this mechanism for relaxation and also take into account the changes connected with the resonance shift (2.16), then in place of (2.14) we finally obtain the following equation of motion:

$$\frac{d\mathbf{L}}{dt} = [\mathbf{L}\mathbf{H}] + \lambda L \left\{ \mathbf{H} - \frac{(\mathbf{H}\mathbf{L})\mathbf{L}}{L^2} \right\}. \tag{2.17}$$

Here λ is a relaxation parameter, which should be rather small so that we can speak of the exciton resonance.

Equation (2.17) is now the equation of the model. As is clear from the process used to derive it, a fundamental property of this equation is that it allows us to treat correlations although in a very simple form. The equation contains the phenomenological parameters ($\mathscr{E} - \mathscr{E}_0$) and λ; as for the quantity N, which determines the length $\mathbf{L}$ ($L = N/2$), we will say more about its connection with the excitonic parameters in the next section.

To conclude this section it is necessary to raise the following question regarding the form of the relaxation term adopted here. It is known that both the solution itself and the limit to which it reduces for an arbitrarily small relaxation depend on how the latter is described, (see, e.g., Refs. 3–5). In the theory of paramagnetic resonance (and optical resonance in a system of two-level atoms) the Bloch equations are usually used, which differ from (2.17) by relaxation terms that do not conserve L. A significant difference between our case and that of paramagnetic resonance is the fact that in the case of paramagnetic resonance we can indicate in principle how the spin relaxes, while in the exciton resonance this cannot be done due to the identity of the excitons, and we can only say that the state of the system changes from Φ_n to Φ_{n-1}; in this case, only Landau–Lifshitz relaxation is appropriate, which we have used. This would be all we needed if nothing more occurred in the system than a change in n.

In reality this is not all that happens. Let us consider, e.g., the following relaxation event: due to their mutual scattering, a pair of excitons convert to another branch ("active" excitons, i.e., those that interact with a given pump, are converted into "passive" excitons that do not interact with the pump). It would seem that all remains as before: the state Φ_n is simply converted into the state Φ_{n-2}. However, these passive excitons occupy a part of the volume, so that the limiting value of N for our "active" excitons is found to be different (N becomes small). However, we expect that as long as the change in N is small compared to N itself this change can be ignored, i.e., we can use Eq. (2.17) as before; that is, the accumulation of passive excitons should not be too large. We should note that the latter also affects the shift in the resonant frequency. This effect also must be included; otherwise the equations can be used only in cases where the number of passive excitons is small compared to the number of active ones.

Finally, it is still possible to excite internal degrees of freedom of the exciton subsystem, which may or may not change the number of excitons. Since the internal degrees of freedom generally do not appear in this model, our description is appropriate only for sufficiently low temperatures or for times that are not too long, i.e., during which the number of excitations in the exciton system is small. If we use the arguments adopted for superfluid systems, we can say that our model is appropriate only for those times when the density of the normal component is considerably smaller than the density of the superfluid component.

Now, it is not possible to derive a rigorous criterion for the applicability of the model—apparently, we would have to use a microscopic approach to do this. It is only important to note that the model does indeed appear to have some region of applicability.

3. SUSCEPTIBILITY

In solving Eq. (2.17) it is convenient to introduce three mutually orthogonal vectors:

$$\begin{aligned} \mathbf{h}_0 &= (0, 0, -\mathscr{E}), \\ \mathbf{h}_1 &= (\Delta \cos \omega t, \Delta \sin \omega t, 0), \\ \mathbf{h}_2 &= [\mathbf{h}_0 \mathbf{h}_1]/h_0, \quad \mathbf{H} = \mathbf{h}_0 + \mathbf{h}_1, \end{aligned} \tag{3.1}$$

where $h_0 = |\mathbf{h}_0| = \mathscr{E}$. We will seek the solution in the form

$$\mathbf{L} = L(\alpha \mathbf{h}_0 + \beta \mathbf{h}_1 + \gamma \mathbf{h}_2). \tag{3.2}$$

Substituting (3.2) into (2.17) and equating the coefficients of $\mathbf{h}_0$, $\mathbf{h}_1$, $\mathbf{h}_2$ on the left- and right-hand sides, we obtain respectively

$$\begin{aligned} \dot{\alpha} + \alpha \frac{\dot{\mathscr{E}}}{\mathscr{E}} &= -\gamma \frac{h_1^2}{h_0} + \lambda [1 - \alpha(\alpha h_0^2 + \beta h_1^2)], \\ \dot{\beta} + \gamma\omega &= \gamma h_0 + \lambda [1 - \beta(\alpha h_0^2 + \beta h_1^2)], \\ \dot{\gamma} - \beta\omega &= (\alpha - \beta) h_0 - \lambda\gamma(\alpha h_0^2 + \beta h_1^2). \end{aligned} \tag{3.3}$$

The solution of interest to us must satisfy the condition $\mathbf{L}^2 = L^2$, i.e.,

$$\alpha^2 h_0^2 + (\beta^2 + \gamma^2) h_1^2 = 1. \tag{3.4}$$

It is not difficult to verify that this condition coincides with Eq. (3.3).

We will look for a steady state, for which α, β, γ (and $\mathscr{E}$) do not depend on time. Let us express α and β in terms of γ using (3.4) and the third of equations (3.3); this gives

$$\begin{aligned} \alpha &= -\frac{\beta}{h_0} \frac{(\omega - h_0) - \lambda h_1^2 \gamma}{1 - \lambda h_0 \gamma}, \\ \beta^2 \left\{ \left[\frac{(\omega - h_0) - \lambda h_1^2 \gamma}{1 - \lambda h_0 \gamma} \right]^2 + h_1^2 \right\} &= 1 - h_1^2 \gamma^2. \end{aligned} \tag{3.5}$$

Taking into account this expression in the first of equations (3.3), we obtain

$$\frac{\gamma}{\lambda h_0} = \beta^2 + \gamma^2 + \frac{\beta^2}{h_0} \frac{(\omega - h_0) - \lambda h_1^2 \gamma}{1 - \lambda h_0 \gamma}. \tag{3.6}$$

At this point we have exact equations; it turns out we

can simplify them. First of all, we can neglect the quantity $\lambda h_1^2\gamma$ compared with $\omega - h_0$, simply because the frequency shift $\lambda h_1^2\gamma$ is always small compared to the interval of frequencies of interest to us. Secondly, we can neglect the last term on the right in Eq. (3.6). We can verify the correctness of these statements after obtaining the result.

After these simplifications, we have in place of (3.5)

$$\alpha = -\frac{\omega-\mathscr{E}}{\mathscr{E}}\frac{\beta}{1-\delta\gamma},$$
$$\beta^2\left\{\left(\frac{\omega-\mathscr{E}}{1-\delta\gamma}\right)^2+\Delta^2\right\}=1-\Delta^2\gamma^2. \tag{3.7}$$

Here we have used the previous notation: $h_0 = \mathscr{E}$, $h_1 = \Delta$, and introduced a new one:

$$\delta=\lambda\mathscr{E}. \tag{3.8}$$

In place of (3.6), taking (3.7) into account, we obtain a quadratic equation for γ, the appropriate solution to which can be written in the following way:

$$\gamma = \frac{1}{2\delta\Delta^2}\{[(\omega-\mathscr{E})^2+\Delta^2+\delta^2]$$
$$-([(\omega-\mathscr{E})^2+\Delta^2+\delta^2]^2-4\delta^2\Delta^2)^{1/2}\}. \tag{3.9}$$

Here the minus sign is used in front of the root, i.e., $\Delta^2\gamma^2$ should be less than unity (see (3.7)); this solution gives the correct limit as $\gamma\to 0$ when $\lambda\to 0$.

Let us evaluate the expression for γ in the following limits. If the effect of attenuation is small compared to the field effect (i.e., $\delta \ll \Delta$) or large ($\delta \gg \Delta$), then for γ we have

$$\gamma \approx \frac{\delta}{(\omega-\mathscr{E})^2+\Delta^2+\delta^2}. \tag{3.10}$$

We must still find a criterion for picking the sign in (3.7), a problem that turns on the question of stability (see Sec. 5). As it turns out, in the stable regime we should have

$$\alpha>0. \tag{3.11}$$

This implies that the projection of $\mathbf{L}$ on the direction of the largest component of the "magnetic field" $\mathbf{h}_0$ should be positive, which is completely natural.

Now we have everything we need to calculate the susceptibility $\varkappa(\omega)$, which is defined in the usual way

$$\mathbf{P}(\omega)=\varkappa(\omega)\mathbf{E}(\omega),$$

where $\mathbf{P}(\omega)$ is the Fourier component of the polarization. According to the definition of the polarization (the dipole moment per unit area in the two-dimensional case), we have

$$\mathbf{P}=\frac{1}{S}\langle\hat{\mathbf{d}}\rangle,$$

where S is the area. Taking into account (2.5), (2.11), and (3.2), this gives

$$\mathbf{P}=\frac{1}{S}\cdot 2\mathbf{d}L_x=\frac{L}{S}\cdot 2\Delta\mathbf{d}(\beta\cos\omega t+\gamma\sin\omega t). \tag{3.12}$$

In the isotropic case we have $\mathbf{d}\|\mathbf{E}_0$; therefore, we can write

$$\Delta\mathbf{d}\to d^2\mathbf{E}_0.$$

The coefficient for $1/2\ \mathbf{E}_0\exp(-i\omega t)$ in (3.12) is the susceptibility we are looking for:

$$\varkappa(\omega)=\frac{N}{S}d^2(\beta+i\gamma). \tag{3.13}$$

Here we have taken into account that $L = N/2$.

Let us investigate this equation in the limiting cases of small and large intensities. Taking into account (3.7), (3.10), and (3.11) as $\Delta\to 0$ we obtain

$$\varkappa(\omega)=-\frac{N}{S}\frac{d^2}{(\omega-\mathscr{E}_0)+i\delta}. \tag{3.14}$$

This is the right contour around the pole, which confirms the correctness of condition (3.11). As for the residue, it now appears possible to make the meaning of N more precise; as we noted previously, we should expect that $(N/S)^{-1/2}$ is on the order of the exciton size; and indeed it is: comparing with the exact results for excitons in the linear regime[1] gives

$$N/S\to\psi^2(0), \tag{3.15}$$

where $\psi(0)$ is the normalized wave function with respect to motion when the electron and hole coordinate values coincide.

The limit of large intensities $\Delta \gg \delta$ is interesting; in this case,

$$\varkappa(\omega)\approx\frac{N}{S}d^2\left\{\frac{\operatorname{sign}(\mathscr{E}-\omega)}{[(\mathscr{E}-\omega)^2+\Delta^2]^{1/2}}+i\frac{\delta}{(\mathscr{E}-\omega)^2+\Delta^2}\right\}. \tag{3.16}$$

We might expect that in the limit of high intensities a result of the form (3.14) should be obtained, but with renormalization of $\mathscr{E}_0$ ($\mathscr{E}_0\to\mathscr{E}$) and δ. As is clear from (3.16), this is not so: $\operatorname{Re}\varkappa$ is greatly changed.

Note that the quantity $\mathscr{E}$, which is a function of Δ and ω, remains to be found; we will return to this problem in the next section.

In conclusion, we present the expression for calculating the energy W absorbed per second per unit surface area:

$$W=\overline{\mathbf{P}\dot{\mathbf{E}}}\to\frac{N}{S}\frac{E_0^2}{2}d^2\omega\gamma$$

(the bar denotes a time average).

4. BISTABILITY

The appearance of bistability in a nonlinear system is not terribly unusual, and occurs in our case as well. In order to understand its appearance it is necessary to know the properties of the function $f(n)$, which defines the shift in the resonance frequency [see Eq. (2.16)]. We will assume that the average energy of the exciton increases with exciton density (see Secs. 1 and 2) and evaluate the shift in frequency using the specific expression:

$$\mathscr{E}=\mathscr{E}_0+\mathscr{E}_1\rho, \tag{4.1}$$

where $\rho = n/N$, $\mathscr{E}_1 > 0$. This simple law apparently is not too bad for small ρ.

A problem unrelated to bistability arises regarding the dependence of $\mathscr{E}$ on the parameters Δ and ω of the problem. In principle this problem is resolved simply: n is coupled to L_z in (2.11), L_z with α in (3.2), and α is determined by expressions (3.7) and (3.9), which themselves contain n through their dependence on $\mathscr{E}$. As a result we obtain an

equation for n which must be solved in order to find the required dependence of $\mathscr{E}$ on Δ and ω.

First of all, let us assume that the damping is small, i.e., $\delta \ll \Delta$, and ignore the contributions associated with it for the moment. As $\delta \to 0$ we obtain the following expression for L_z:

$$L_z = -L\frac{|\mathscr{E}-\omega|}{[(\mathscr{E}-\omega)^2+\Delta^2]^{1/2}}. \tag{4.2}$$

Squaring the left- and right-hand sides of (4.2), expressing L_z in terms of n, and changing to dimensionless variables, we obtain in place of (4.2) the expression

$$(1-2\rho)^2 = \frac{(\varepsilon-\rho)^2}{(\varepsilon-\rho)^2+\bar{\Delta}^2},$$
$$\varepsilon = (\omega-\mathscr{E}_0)/\mathscr{E}_1,\quad \bar{\Delta}=\Delta/\mathscr{E}_1. \tag{4.3}$$

This equation can be analyzed graphically by constructing curves on the interval $0<\rho<1/2$. It is clear from Fig. 1, on which we show the case $0<\varepsilon<1/2$, that bistability is possible in this interval of ε for values of $\bar{\Delta}$ that are not too large. Under these circumstances there are three roots $\rho_1<\rho_2<\rho_3$; as a rule, the middle root corresponds to an unstable solution (see the next section).

It is not difficult to determine the interval of bistability; we present the result:

$$0<\varepsilon<{}^1/_2,$$
$$0<\bar{\Delta}<\bar{\Delta}_0,$$
$$\bar{\Delta}_0^2 = {}^1/_4[1-(1-2\varepsilon)^{2/3}]^3. \tag{4.4}$$

We must still discuss the role of attenuation, which is extremely important for small ε. Let us use expression (3.10) for γ, treating it as an interpolation for arbitrary ratios between Δ and δ. Then in place of (4.3) we obtain

$$(1-2\rho)^2 = \frac{(\varepsilon-\rho)^2+\bar{\delta}^2}{(\varepsilon-\rho)^2+\bar{\Delta}^2+\bar{\delta}^2}, \tag{4.5}$$

where $\bar{\delta}=\delta/\mathscr{E}_1$. It is not difficult to analyze this equation for small values of all quantities entering into it. As a result we obtain in place of (4.4) the following interval of bistability:

$$\bar{\Delta}_1<\bar{\Delta}<\bar{\Delta}_2,$$
$$\bar{\Delta}_{1,2}^2 = {}^8/_{27}[\varepsilon\mp(\varepsilon^2-\bar{\delta}^2)^{1/2}][2\varepsilon\pm(\varepsilon^2-\bar{\delta}^2)^{1/2}]^2 \tag{4.6}$$

(the upper sign for $\bar{\Delta}_1^2$, the lower for $\bar{\Delta}_1^2$). From this it is clear that we should have $\varepsilon>\bar{\delta}$ [in contrast to (4.4)].

The magnitude of the polarization $|\mathbf{P}|$ follows the usual

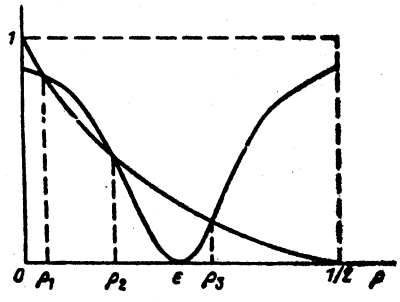

FIG. 1.

S-shaped curve for bistability as a function of the pump intensity (which enters in through Δ^2). When the intensity is increased to the point $\bar{\Delta}_2$, $|\mathbf{P}|$ jumps to a lower value; if the intensity is then decreased there is another jump in $|\mathbf{P}|$ to a higher value at the point $\bar{\Delta}_1$.

5. PROBE SIGNALS

In experimental investigations of nonlinear properties of excitonic systems, a weak (probe) signal is usually used in the presence of an intense pump. It is necessary to clarify how this pump affects the behavior of the weak signal.

To solve this problem let us use the equations of motion (2.17), substituting the following expressions into them in place of $\mathbf{H}$ and $\mathbf{L}$:

$$\mathbf{H}\to\mathbf{H}+\mathbf{H}',\quad \mathbf{L}\to\mathbf{L}+\mathbf{L}', \tag{5.1}$$

and linearizing them with respect to the corrections $\mathbf{H}'$, $\mathbf{L}'$. We obtain

$$\frac{d\mathbf{L}'}{dt} = [\mathbf{L}'\mathbf{H}]+[\mathbf{L}\mathbf{H}']+\lambda L\left\{\left[\mathbf{H}'-\frac{(\mathbf{H}'\mathbf{L})\mathbf{L}}{L^2}\right]-\frac{1}{L^2}[(\mathbf{H}\mathbf{L}')\mathbf{L}+(\mathbf{H}\mathbf{L})\mathbf{L}']\right\}. \tag{5.2}$$

The correction $\mathbf{H}'$ consists of two parts:

$$\mathbf{H}'=\mathbf{H}_1'+\mathbf{H}_2'. \tag{5.3}$$

The first part $\mathbf{H}_1'$ is directly connected with the field of the weak signal analogous to (2.13), i.e.,

$$\mathbf{H}_1'=(\Delta'\cos\Phi,\ \Delta'\sin\Phi,\ 0),$$
$$\Delta'=\mathbf{d}\mathbf{E}_0',\quad \Phi=\Omega t+\Phi_0, \tag{5.4}$$

where Ω is the frequency and $\mathbf{E}_0'$ is the amplitude of the weak signal. The second part $\mathbf{H}_2'$ is determined by the change in $\mathscr{E}$ (see (2.16)) caused by the weak signal:

$$\mathbf{H}_2' = \left(0,0,-\frac{d^2f}{dn^2}L_z'\right). \tag{5.5}$$

It is convenient to seek the vector $\mathbf{L}'$ in a form that explicitly includes its orthogonality to the vector $\mathbf{L}$. For this we must turn to a system of coordinates rotating together with $\mathbf{L}$. This can be done, e.g., in the following way. Let us introduce the unit vectors

$$\boldsymbol{\xi} = \frac{[\mathbf{L}\mathbf{h}_0]}{|[\mathbf{L}\mathbf{h}_0]|},\quad \boldsymbol{\eta}=[\boldsymbol{\zeta}\boldsymbol{\xi}],\quad \boldsymbol{\zeta}=\mathbf{L}/L. \tag{5.6}$$

In terms of these unit vectors $\mathbf{L}'$ can be written as follows:

$$\mathbf{L}'=\mu\boldsymbol{\xi}+\nu\boldsymbol{\eta}. \tag{5.7}$$

From Eq. (5.2) it is necessary to obtain an equation for μ, ν. For this we must know the derivatives of the unit vectors we have introduced, which are not difficult to find if we take into account that the vector $\mathbf{L}$ rotates with frequency ω in the direction ξ. Then we obtain for the derivatives

$$\dot{\boldsymbol{\xi}}=-\omega(\boldsymbol{\eta}\cos\theta+\boldsymbol{\zeta}\sin\theta),$$
$$\dot{\boldsymbol{\eta}}=\omega\boldsymbol{\xi}\cos\theta,$$
$$\dot{\boldsymbol{\zeta}}=\omega\boldsymbol{\xi}\sin\theta. \tag{5.8}$$

We recall that θ is the angle between $\mathbf{L}$ and $\mathbf{h}_0$.

In addition, it is necessary to express the vectors $\mathbf{H}$, $\mathbf{H}'$

entering into (5.2) in terms of the vectors (5.6). We present the result:

$$\mathbf{h}_1=\frac{\Delta^2}{\sin\theta}[\gamma\xi+\beta\cos\theta\cdot\eta+\beta\sin\theta\cdot\zeta],$$
$$\mathbf{h}_2=\frac{\Delta^2}{\sin\theta}[-\beta\xi+\gamma\cos\theta\cdot\eta+\gamma\sin\theta\cdot\zeta],$$
$$\mathbf{h}_0=h_0[-\sin\theta\cdot\eta+\cos\theta\cdot\xi], \qquad (5.9)$$
$$\mathbf{H}_1'=\frac{\Delta'}{\Delta}[\cos(\Phi-\varphi)\cdot\mathbf{h}_1-\sin(\Phi-\varphi)\cdot\mathbf{h}_2],$$
$$\mathbf{H}_2'=\frac{\mathbf{h}_0}{h_0}\frac{d^2f}{dn^2}\nu\sin\theta.$$

Here $\varphi=\omega t$, $\Phi=\Omega t+\Phi_0$.

Substituting (5.7)–(5.9) into (5.2) and equating the coefficients of ξ, η, ζ on the left- and right-hand sides, we obtain the required equations. They are given here in the simplest form, neglecting contributions to (5.2) that contain the product $\lambda\mathbf{H}'$:

$$\dot\mu+\delta_1\mu+(\omega_0-\omega_1)\nu=-L\Delta'\cos\theta\cos(\Phi-\varphi+\chi),$$
$$\dot\nu+\delta_1\nu-\omega_0\mu=L\Delta'\sin(\Phi-\varphi+\chi). \qquad (5.10)$$

Here we introduce the notation

$$\delta_1=\lambda(\alpha h_0^2+\beta\Delta^2)\approx\delta\cos\theta,$$
$$\omega_0=(\omega-\mathscr{E})\cos\theta-\beta\Delta^2\approx[(\omega-\mathscr{E})^2+\Delta^2]^{1/2}\operatorname{sign}(\omega-\mathscr{E}), \qquad (5.11)$$
$$\omega_1=L\frac{d^2f}{dn^2}\sin^2\theta\to\frac{1}{2}\mathscr{E}_1\sin^2\theta,$$
$$\beta+i\gamma=(\beta^2+\gamma^2)^{1/2}e^{i\chi}, \qquad \beta^2+\gamma^2=\frac{\sin^2\theta}{\Delta^2}.$$

We can also write an expression for ω_1 which is convenient when relation (4.1) holds.

Both equations (5.10) are obtained by setting equal the coefficients of ξ and η respectively; as for the third equation, it is satisfied identically.

Especially noteworthy is the fact that the quantity δ_1, which determines the damping of μ and ν, must be positive for stability:

$$\delta_1>0, \qquad (5.12)$$

which immediately gives condition (3.11) (if we do not consider a small neighborhood near $\theta=\pi/2$). Of course, the corresponding frequency Ω_0 should be real:

$$\Omega_0^2=\omega_0(\omega_0-\omega_1)>0. \qquad (5.13)$$

In the opposite case the state is unstable, which occurs immediately for the root ρ_2 (see Sec. 4); this is verified as $\delta\to0$.

In the final analysis what we are interested in is the correction $\mathbf{P}'$ to the polarization due to the field of the probe signal $\mathbf{E}'$; we can calculate this correction using expression (3.12):

$$\mathbf{P}'=\frac{2\mathbf{d}}{S}L_x'=\frac{2\mathbf{d}}{S}(\mu\xi_x+\nu\eta_x). \qquad (5.14)$$

We can find the projections ξ_x, η_x using (5.9):

$$\xi_x=-\sin(\varphi-\chi), \quad \eta_x=\cos\theta\cos(\varphi-\chi). \qquad (5.15)$$

The expression obtained after solving Eq. (5.10) for $\mathbf{P}'$ can be written in the form

$$\mathbf{P}'=\mathbf{p}\{A\sin\Phi+B\cos\Phi+C\sin[2(\varphi-\chi)-\Phi]+D\cos[2(\varphi-\chi)-\Phi]\}, \qquad (5.16)$$

where for the vector $\mathbf{p}$ and coefficients A, B, C, D we have

$$\mathbf{p}=-(2L/S)\Delta'\mathbf{d}\{[(\Omega-\omega-\Omega_0)^2+\delta_1^2][(\Omega-\omega+\Omega_0)^2+\delta_1^2]\}^{-1},$$
$$A=\delta_1\{-[(\Omega-\omega)^2+\Omega_0^2]\cos\theta+(\Omega-\omega)[(\omega_0-\omega_1)+\omega_0\cos^2\theta]\},$$
$$B=[(\Omega-\omega)^2-\Omega_0^2]\{(\Omega-\omega)\cos\theta-{}^1/_2[(\omega_0-\omega_1)+\omega_0\cos^2\theta]\},$$
$$C=\delta_1(\Omega-\omega)(\omega_0\sin^2\theta-\omega_1), \qquad (5.17)$$
$$D={}^1/_2[(\Omega-\omega)^2-\Omega_0^2](\omega_0\sin^2\theta-\omega_1).$$

Here we have included powers of δ_1 in the coefficients A, B, C, D no higher than the first (small attenuation).

From the expressions given here it is clear that in addition to harmonics with frequency Ω, $\mathbf{P}'$ contains harmonics with frequency $2\omega-\Omega$ as well, which is one of the consequences of nonlinearity; hence, radiation appears at this frequency (two pump photons are converted into a probe photon and a photon with frequency $2\omega-\Omega$). Other frequencies should not appear in the resonance approximation. In the weak-pumping limit ($\Delta\to0$, $\theta\to0$) the nonlinear effects disappear and we obtain the usual linear expression for $\mathbf{P}'$ [see (3.14)].

The response has a resonant character (i.e., resonances at frequencies $\Omega=\omega\pm\Omega_0$); in the absence of a resonance shift (i.e., for $\omega_1=0$), the eigenfrequency Ω_0 coincides with the Rabi frequency, which is natural.

It follows from these expressions that the probe signal may be amplified as well as damped. To verify this, we must find what work the field $\mathbf{E}'$ does above the average. We can calculate the corresponding power W' just as in Sec. 3, i.e., using the expression

$$W'=\overline{\mathbf{P}'\mathbf{E}'}\to{}^1/_2\mathbf{p}\mathbf{E}_0\Omega A.$$

Let us present the value of the quantity A at resonance:

$$A(\Omega=\omega\pm\Omega_0)=\delta_1\{-2\Omega_0^2\cos\theta\pm\Omega_0[\omega_0-\omega_1+\omega_0\cos^2\theta]\}.$$

A comparison of the magnitudes of the various terms in the curly brackets shows that the magnitude of the second term is small; this implies that absorption of the probe signal occurs for one of the resonance frequencies and amplification at the other (the amplification is always smaller).

6. DISCUSSION

As far as I know, the equations of the form (2.17) postulated in this model have not been used previously to investigate nonlinear exciton resonance. A number of arguments that are made in this paper are known in the literature (see the review Ref. 1), although not all of them, e.g., bistability was not discussed in review Ref. 1. The model allows us to obtain predictive results in all the cases of interest; undoubtedly this is its advantage.

Considerations of the applicability of the model were presented in Sec. 2. I emphasize once again that the model is suitable only for studying a purely excitonic resonance, implying that the parameter Δ which characterizes the effect of the pump on the system should be considerably smaller than the binding energy of the exciton; the same thing applies to the magnitude of the detuning $|\omega-\mathscr{E}|$ as well. However, the shift in resonance frequency $\mathscr{E}$ need not be small.

Incidentally, the only expressions that are suitable for describing the shift in resonance frequency are (2.15) and (2.16), because in fact we are dealing with the ground state

of the exciton system (in a model formulation). As for the quantity $\mathscr{E}_1$ in (4.1), apparently it is on the order of the binding energy of the exciton.

One of the fundamental effects contained in the model is the effect of saturation: it is impossible to pump more than a certain limiting number of excitons into the system. This is completely natural for those intensities at which the model itself has meaning (recall the limitation on the magnitude of Δ mentioned above).

In particular, questions regarding the role of inhomogeneous broadening of the exciton level remain outside the framework of the model, although such broadening apparently can play an important role in real quantum wells. However, there are several cases where the effects of inhomogeneous broadening could have been taken into account using the results shown here.

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