Extended Abstracts of the 15th Conference on Solid State Devices and Materials, Tokyo, 1983, pp. 321-324

Invited

Physics of Excitonic Optical Bistability

Eiichi Hanamura

Department of Applied Physics, University of Tokyo,
Hongo, Bunkyo-ku, Tokyo 113, Japan

The optical bistable behavior results from the combined effect of the nonlinear medium and the feedback provided by the Fabry-Perot or Ring cavity to the beams. We propose the optical bistable system due to the coherent dispersive nonlinear polarization of the exciton-excitonic molecule system of CuCl, responding in the short transverse relaxation time of an order of pico-second. Here the microscopic description as well as the dynamics of the nonlinear polarization are required.

§1. Introduction

An optical bistable system is composed of a nonlinear dielectric medium inserted in the Fabry-Perot or the Ring cavity. This system is made to operate in two stable low and high transmitting states under appropriate conditions. Hereafter we call these states an off- and an on-state, respectively. Then it exhibits a hysteresis of transmitted power against change of incident power. This nonlinearity comes from the dispersive and absorptive nonlinear optical response of the medium. These are, respectively, due to the nonlinearity of the real and the imaginary parts of the dielectric constant of the medium. The optical bistability due to the former dispersive nonlinearity is shown to be more easily realized than that due to the latter. Really the dispersive optical bistability has been observed, e.g., by using the nonlinear medium of GaAs[1] and InSb.[2] This optical bistability is considered to be used in the future as essential parts of an optical integral circuit as well as an optical computer, in which light plays the role of electron in large scale integral circuit in modern electronic computer. In order to get the more effective logic and memory devices than very large scale integral circuit or Jesephson junctions, the system is required to switch between the off- and the on-states in an order of pico-second by light pulse with low power of an order of pico-joule.[2] An obstacle against these requirements is that the response time of switching from the on- to the off-state is extremely long, e.g., 40 nano-second for the system of GaAs[1] and of an order of 100 nano-second for that of InSb.[2] They used the dispersive nonlinear response which was brought about by the presence of really excited elementary excitations. Therefore it takes decay time of an exciton in the case of GaAs and that of an electron-hole pair in the case of InSb before the switching-off is completed, although the switching-on time may be reduced to an order of pico-second even in this case. Furthermore, the microscopic mechanism is not yet understood for these nonlinear dispersive responses of the really excited elementary excitations.

The stationary response of the optical bistable system is understood as far as we assume the power-dependent refractive index without knowing the microscopic mechanism. The ratio of the transmitted power P_t to the incident one P_i is given by

$$\frac{P_t}{P_i} = \frac{(1-R)^2}{(1-R)^2+4R\sin^2\delta}, \qquad (1)$$

where R is the reflectivity of the both ends of the resonator, 2δ is the phase change of light after a round traverse in the resonator and $\delta=2\pi n\ell/\lambda$ (ℓ is the length of the medium and λ is the wave length of the light). The refractive

index n is written as a sum of a linear contribution $n_0=\sqrt{\varepsilon_0}$ and the nonlinear one $(4\pi\chi_{NL}/n_0)|E|^2$. (E is the amplitude of the internal electric field and ε_0 is the linear dielectric constant of the medium.) The nonlinear part is proportional to the transmitted power $P_t \equiv (1-R)|E|^2$ so that $\delta = 2\pi n_0 \ell/\lambda + KP_t$. Then we have the following relation:

$$\frac{P_t}{P_i} = \frac{\delta - 2\pi n_0 \ell/\lambda}{KP_i} \quad . \tag{2}$$

The transmitted power P_t is obtained as a function of the incident power P_i by coupling eqs.(1) and (2). Then the hysteresis of the transmitted power against change of the incident one is understood.[3] We can also operate this system as a limiter and a derivative amplifier as well as the optical bistability by controling n_0.[4] In order to get operation of the optical bistability, we need change the value of δ by an order of π, so that the required power $|E|^2$ is of an order of $n_0\lambda/[8\pi\chi_{NL}\ell]$. The incident power may be reduced by increasing the sample length ℓ. However, the increase of the sample length will bring about poor dynamical response as will be discussed later and will also require the higher monocromaticity and coherency of the incident laser light.

In this paper, we wish to discuss the possibility of switching between the on- and the off-states in an order of pico-second by the pulse of an order of pico-joule.[5] When we use the dispersive nonlinearity due to really created elementary excitations, the elementary excitations are easily accumulated in the medium by rather weak incident laser power because of their long life time. However, because of this reason, the switching-off time is determined by the long longitudinal relaxation time as discussed for the cases of GaAs and InSb. We have another contribution to the nonlinear dispersive response from coherent nonlinear optical processes without any real creation of the elementary excitations. The characteristic response time due to this process is determined by the short transverse relaxation time of an order of pico-second in solids or by the response time of the cavity itself. Under usual situations, however, this coherent excitaiton of the nonlinear polarization is possible only under the off-resonant excitation. As a result, this requires higher incident laser power than in the case of the incoherent nonlinearity associated with real excitation of elementary excitations. When we consider the exciton and bi-exciton (excitonic molecule) system, we can describe the detailed dynamics for the nonlinear polarization. Then we find out the extreme enhancement of the nonlinear polarization due to the giant two-photon excitation of the excitonic molecule.[6] This reduces the power required to realize the optical bistability due to the coherent nonlinear processes.

§2. Giant Oscillator Strength and Excitonic Molecule

Excitonic molecule is a bound state of two single excitons.[7] The molecular binding energy ε_m^b is 30meV while the exciton binding energy ε_{exc}^b is 170meV in CuCl. The lowest state of the excitonic molecule in CuCl is described by a linear combination of two optically active Γ_5 excitons and two optically inactive Γ_2 excitons. This is because the binding energy of the excitonic molecule is much larger than the Γ_5-Γ_2 splitting of single excitons in CuCl. This electronic structure gives a hint to create the excitonic molecule directly by two-photon absorption through the optically active part. This contributes to the nonlinear polarization of the medium.

Transition probability of two-photon excitation of an excitonic molecule is expressed by second order perturbation with respect to the electron-photon interaction H_I:

$$W^{(2)}(\omega) = \frac{2\pi}{\hbar}\sum |\langle \Gamma_1^m | H_I^- \sum_i \frac{|i\rangle\langle i|}{E_{ig} - \hbar\omega} H_I^- | 0\rangle|^2 \times \delta(2\hbar\omega - E_m) . \tag{3}$$

$|0\rangle$ is the crystal ground state and $\langle \Gamma_1^m|$ is the final state in which a Γ_1 excitonic molecule is created. H_I^- is the part of H_I which contains the photon annihilation operator. The two-photon transition probability is extremely enhanced by a factor about 10^6 in comparison with an ordinary two-photon absorption due to a band-to-band transition.[6] One reason for this enhancement stems from the effect of a giant oscillator strength. This is due to the fact that in the second photon absorption process which causes the

transition from the intermediate state $|i\rangle$ to the final state $\langle\Gamma_1^m|$, one can excite any valence electron in the range within the large molecular radius around the virtually created first exciton to make an excitonic molecule. The situation is quite different from that which occurs in ordinary two-photon absorption due to band-to-band transition. There the same electron which is excited into the intermediate state has to interact again with the second photon. The further enhancement comes from the resonant effect. This is due to the smallness of the energy denominator in eq.(3). The ratio of $W^{(2)}[\hbar\omega = E_{ex}(0)-\varepsilon_m^b/2]$ to the one-photon transition rate due to a single exciton $W^{(1)}[\hbar\omega=E_{ex}(0)]$ is estimated to be of the order of $10^{-15}(N/V)$ for CuCl, when N/V is the photon density of the exciting light. Therefore, when a dye-laser is used as a tunable excitation source with a photon density $N/V=10^{15}/cm^3$ which corresponds to $10MW/cm^2$, we can expect the two-photon absorption coefficient to be as strong as the one-photon absorption coefficient of a single exciton. The crystal is transparent to the radiation field with the photon energies less than the single exciton. However, when laser light with a reasonable intensity is used, we may expect a sharp absorption peak due to the giant two-photon excitation of excitonic molecules at a position shifted by half the molecular binding energy (ε_b^m=30meV for CuCl) from the exciton absorption peak $E_{ex}(0)$. This sharp line is embedded in rather weak background of the ordinary two-photon absorption due to the band-to-band transition and also of the one-photon absorption tail due to the single exciton.

This giant two-photon excitation of excitonic molecules was observed first by Gale and Mysyrowicz[8] in CuCl just one year after the proposal.[6] Succesively, this has been found in CuCl[9] and CuBr[10] as well as CdS crystals.[11] The strong nonlinear absorption brings about the nonlinear dispersion through the Kramers-Kronig relation. This nonlinearity is used for the optical bistability as will be discussed in §4.

§3. Anomaly of Exciton Polariton

Exciton is a propagating elementary excitation in solids and interacts with the radiation field in the bi-linear form. As a result, they form the hybridized waves which are called the polariton. The dispersion relation is obtained from the relation:

$$\frac{c^2k^2}{\omega^2} = 1+4\pi\chi_L(k,\omega)+4\pi\chi_{NL}(k,\omega)|E|^2 , \qquad (4)$$

where ω and k are the angular frequency and the momentum of the polariton, respectively. χ_L is the linear polarizability and is decomposed into the background contribution $\chi_0(k,\omega)$ and that due to the exciton formation. $\chi_{NL}(k,\omega)$ is the nonlinear polarizability mainly due to the giant two-photon excitation of an excitonic molecule. As a result, the dispersion relation of the polariton is modified depending upon the incident laser power around the incident frequency corresponding to the giant two-photon excitation of excitonic molecule[12] ($\hbar\omega=E_{ex}(0)-\varepsilon_m^b/2$). This effect has been observed, when the incident power increases, as the deviation of the super-Raman scattering peak ω_s versus the incident frequency ω_i from the smooth curve for the linear response.[13] It is this dispersion anomaly due to χ_{NL} that induces the optical bistability. Let us consider the nonlinear medium of CuCl crystal of the length $1\mu m$ sandwiched by the dielectric coating giving reflectivity 0.9. Then we expect the optical bistable response under irradiation of 3.183eV coherent laser light with the power of $1MW/cm^2$ upon the system.

§4. Dynamics of Excitonic Optical Bistable System

In the present work, we wish to study dynamics of the optical bistable system and to search for the possibility of responding in pico-second by the pico-joule pulse. For this purpose, we study equations of motion for polarizations and populations of the exciton and the excitonic molecule under the strong coherent laser irradiation. Then we solve the Maxwell equation for the internal electric field consistently with the equations of the medium. There are two kinds of contributions to the nonlinear dispersive response, i.e., the coherent two photon process described by the off-diagonal density matrix $\rho_{mg} \equiv \langle\Gamma_1^m|\rho(t)|0\rangle$ and the incoherent one described by the diagonal density matrix $\rho_{ee}\equiv\langle exc|\rho(t)|exc\rangle$. ρ_{ee} represents the exciton population and ρ_{mg} does

the coherency between two states of the ground state and the excitonic molecule introduced by two-photon transition. These two processes may be also understood by analogy to the Raman scattering and the luminescence,[14] respectively. The dynamics due to the incoherent nonlinear process is governed by longitudinal relaxation time T_1, while that due to the coherent one is determined by the transverse relaxation time T_2. For the elementary excitation in solids, T_1 is usually of an order of nano-second while T_2 is of an order of pico-second. The optical bistability observed by Gibbs et al. for the GaAs wafer is due to this incoherent process and it takes 40 nano-second to switch off. Really as expected, we can show that not only the switching-on but also the switching-off can be completed in the order of pico-second for the case where the coherent process is dominating. When the incident laser light is nearly resonant to the exciton, the incoherent nonlinear process which is accompanied with creation of excitons prevails. This, however, decreases exponentially as the excitation energy shifts downward from the exciton peak, obeying the Urbach-Martienssen rule.[15] Consequently, the contribution from the incoherent process almost vanishes while that due to the coherent process decreases very gradually only as the inverse of the off-resonance. As a result, under the suitable off-resonant excitation, e.g., at the excitation frequency by 16meV lower than the exciton peak in the CuCl system, the coherent process is shown to be dominant. Usually the higher exciting power is required to fullfill the condition of the optical bistability under the off-resonant excitation. Very fortunately, the transition dipolemoment to excite a single exciton into the excitonic molecule is ten times larger that that of the single exciton through the effect of the giant oscillator strength as discussed in §2. As a result, the required incident power can be reduced by 2 orders of magnitude so that 1MW/cm^2 incident laser power is enough to fullfill the condition under the 16meV off-resonant excitation.

This prediction is not yet experimentally proved. To realize this optical bistable operation, we must satisfy the following four conditions: (1) the pure good crystal of CuCl, (2) forming the Fabry-Perot resonator of CuCl with sufficient precision, (3) the coherent and strong laser source around 3.19eV, and (4) the suitable detector. At present, there is no group which can satisfy these four conditions, but we hope the success under cooperation of two or more groups.

References

1) H.M.Gibbs, S.L.McCall and T.N.C.Venkatesan: Optics News/summer (1979) 6.
2) S.D.Smith and D.A.B.Miller: Proc. Ints. Conf. Phys. Semicond. ed. by S.Tanaka and Y.Toyozawa, (1981) 597.
3) P.W.Smith, E.H.Turner and P.J.Maloney: IEEE J. Quantum Electron. QE-14 (1978) 207.
4) T.N.C.Venkatesan and S.L.McCall: Appl. Phys. Lett. 30 (1977) 282.
5) E.Hanamura: Solid State Commun. 38 (1981) 939.
6) E.Hanamura: Solid State Commun. 12 (1973) 951.
7) See, e.g., E.Hanamura and H.Haug: Physics Reports 33C (1977) 210.
8) G.M.Gale and A.Mysyrowicz: Phys. Rev. Letters 32 (1974) 727.
9) N.Nagasawa, N.Nakata, Y.Doi and M.Ueta: J. Phys. Soc. Japan 38 (1975) 593 and 39 (1975) 987.
10) N.Nakata, N.Nagasawa, Y.Doi and M.Ueta: J. Phys. Soc. Japan 38 (1975) 903.
11) T.Itoh, Y.Nozue and M.Ueta: J. Phys. Soc. Japan 40 (1976) 451.
12) R.März, S.Schmitt-Rink and H.Haug: Z. Phys. B40 (1980) 9.
13) T.Itoh and T.Suzuki: J. Phys. Soc. Japan 45 (1978) 1939.
14) T.Takagahara, E.Hanamura and R.Kubo: J. Phys. Soc. Japan 43 (1977) 802, 811 and 1522, and ibid. 44 (1978) 728 and 742.
15) B.Batlogg and J.P.Remeika: Phys. Rev. Letters 45 (1980) 1126.