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Nguyen Ba An

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Le Thi Cat Tuong

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Nguyen Ba An *
International Centre for Theoretical Physics, Trieste, Italy

and

Le Thi Cat Tuong
Institute of Physics, Nghia Do, Tu Liem, Hanoi, Vietnam.

ABSTRACT

A resonant monochromatic pump laser generates coherent excitons in an n-type doped semiconductor. Both exciton–exciton and exciton–donor interactions come into play. The former interaction can give rise to the appearance of optical bistability which is heavily influenced by the latter one. When optical bistability occurs at a fixed laser frequency both its holding intensity and hysteresis loop size are shown to decrease with increasing donor concentration. Two possibilities are suggested for experimentally determining one of the two parameters of the system – the exciton–donor coupling constant and the donor concentration, if the other parameter is known beforehand.

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* Permanent address: Institute of Theoretical Physics, P.O. Box 429 Boho, Hanoi 1000, Vietnam.

1 Introduction

Optical bistability, a very interesting far-from-equilibrium phenomenum, can take place in many physical systems thanks to the combined effect of optical nonlinearity and feedback (For comprehensive overviews see e.g. [1-5]). To this phenomenum in pure semiconductors has been devoted a great deal of works which have studied various kinds of nonlinearity and feedback. In [6-10] the exciton-exciton interaction is taken for nonlinearity while in [11-18] it is the mutual transition between two-photon and biexciton states. As for feedback it may be supplied by different configurations of external cavities [11,15,19-21] or brought about by intrinsic mechanisms due to the balance between optically active quasiparticle excitation and their recombination [22,23]. Hysteresis loops of optical bistability have been shown to develop in a quite different manner and have multiform shapes including those of a two- or three-winged bow [24].

Recently, a systematic analysis [25] has been performed of main man-controlled parameters that may affect the thermally induced optical bistability. Among the extrinsic parameters on which the occurrence and feature of optical bistability depend there is impurity concentration. By doping samples with a definite impurity concentration one can optimize the operation of a bistable device for a given input light frequency. This, as pointed out in [26], opens new aspects of technological engineering of sample properties for optical bistability operation.

In this paper we attempt to present first theoretical calculations for excitonic optical bistability in n-type doped semiconductors basing on the simplest possible approximations. Though so, the results indicate a marked decrease in both holding intensity and hysteresis loop size of the optical bistability that occurs at a given input light frequency when the donor concentration increases. Moreover, possibilities will be discussed of how qualitatively measuring optical bistability characteristics might be helpful for determining such physical parameters as exciton-donor interaction constant or concentration of donors that dope the semiconductor.

2 Model for calculations

The model we use in this paper for theoretical calculations is a n-type doped semiconductor which is subjected to an externally driving laser field with given wave-vector **k**, complex amplitudes $\mathcal{E}_{\mathbf{k}}^{(\pm)}$ and circular frequency Ω_k. The laser frequency is assumed to be resonant with the exciton energy so that a direct connection between the laser and the exciton holds. At high enough optical excitation many excitons are generated in the semiconductor which interact among themselves and with donors. As will be seen, the exciton-exciton interaction is responsible for the occurrence of optical bistability which in turn is influenced by the exciton-donor coupling. In our consideration, concentrations of donors and excitons should be understood not too high so that the screening of their mutual interactions could not cause the formation of an electron-hole plasma with positive donor-ions distributed in it . We thus can neglect any effect of screening and phase transition.The temperature is also anticipated low enough to prevent both excitons and donors from possible thermal ionization. The model system Hamiltonian can then be written in the form (Below and lateron we apply the unit system with Planck constant $\hbar$ and light velocity c equal to unity)

$$(1) \qquad H = \sum_{\nu} g_{\nu k}\left[\mathcal{E}_{\mathbf{k}}^{(-)} e^{-i\Omega_k t} a_{\nu\mathbf{k}}^{+} + h.c.\right] + \sum_{\nu\mathbf{p}} \omega_{\nu p} a_{\nu\mathbf{p}}^{+} a_{\nu\mathbf{p}} +$$

$$+ \frac{1}{V} \sum_{\mathbf{pql}\nu\mu\varsigma\xi} F_{\nu\mu\varsigma\xi}(\mathbf{p},\mathbf{q},\mathbf{l})\, a_{\nu\mathbf{p}+\mathbf{l}}^{+} a_{\mu\mathbf{q}-\mathbf{l}}^{+} a_{\varsigma\mathbf{q}} a_{\xi\mathbf{p}} +$$

$$+ \sum_{n} E_n d_n^{+} d_n + \frac{1}{V} \sum_{\mathbf{pq}\nu nm\mu} U_{\nu nm\mu}(\mathbf{p},\mathbf{q})\, a_{\nu\mathbf{p}}^{+} d_n^{+} d_m a_{\mu\mathbf{q}}$$

In Eq.(1) $a_{\nu\mathbf{k}}$ (d_n) stands for the annihilation operator of an exciton (a donor) with energy $\omega_{\nu k}$ (E_n) and in quantum state ν (n). $g_{\nu\mathbf{k}}$, $U_{\nu nm\mu}$ and $F_{\nu\mu\varsigma\xi}$ denote respectively the exciton-light, exciton-donor and exciton-exciton couplings. V labels the sample volume. In writing H we have ignored the interaction of the exciton with the positive donor-ion because of the low mobility of the latter. For simplicity we restrict ourselves just to the lowest exciton energy level and thus we shall drop the indices ν, μ, ... as well as the summation over them in all the further formulations.

The Heisenberg equation of motion for $< a_{\mathbf{k}} >$ ($< ... >$ means an average over the **k**-mode that is assumed coherent over the whole system) after phenomenologically adding the exciton relaxation time $\tau_{\mathbf{k}}$ will look :

$$(2) \qquad -\left[\frac{d}{dt} + \frac{1}{\tau_{\mathbf{k}}} + i\omega_k\right] < a_{\mathbf{k}} > = i g_k \mathcal{E}_{\mathbf{k}}^{(-)} e^{-i\Omega_k t} +$$

$$+ \frac{i}{V} \sum_{\mathbf{pq}} F(\mathbf{p},\mathbf{q},\mathbf{k}-\mathbf{p}) < a_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{+} a_{\mathbf{q}} a_{\mathbf{p}} > +$$

$$+ \frac{i}{V} \sum_{\mathbf{p}nm} U(\mathbf{k},\mathbf{p}) < d_n^{+} d_n a_{\mathbf{p}} >$$

At this moment, instead of setting up further equations for the averages $< a^{+}aa >$ and $< d^{+}da >$ we resort to the commonly accepted in the literature approximation that is known as the Hartee-Fock splitting and formally means :

$$(3) \quad < a_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{+} a_{\mathbf{q}} a_{\mathbf{p}} > \approx < a_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{+} a_{\mathbf{q}} >< a_{\mathbf{p}} > + < a_{\mathbf{p}+\mathbf{q}-\mathbf{k}}^{+} a_{\mathbf{p}} >< a_{\mathbf{q}} > =$$

$$= \delta_{\mathbf{pk}} < N_{\mathbf{q}} >< a_{\mathbf{q}} > + \delta_{\mathbf{qk}} < N_{\mathbf{p}} >< a_{\mathbf{p}} >$$

$$(4) \qquad < d_n^{+} d_m a_{\mathbf{p}} > \approx < d_n^{+} d_m >< a_{\mathbf{p}} > = \delta_{nm} < D_n >< a_{\mathbf{p}} >$$

where $N_{\mathbf{p}} = a_{\mathbf{p}}^{+} a_{\mathbf{p}}$ and $D_n = d_n^{+} d_n$ label the occupation number operators of the **p**-mode exciton and the n-state donor electron. Inserting Eqs.(3,4) into Eq.(2) gives :

$$(5) \quad \left[\frac{d}{dt} + \frac{1}{\tau_{\mathbf{k}}} + i\omega_k + \frac{i}{V}\sum_{\mathbf{p}} [F(\mathbf{k},\mathbf{p},0) + F(\mathbf{p},\mathbf{k},\mathbf{k}-\mathbf{p})] < N_{\mathbf{p}} >\right] < a_{\mathbf{k}} > =$$

$$= i g_k \mathcal{E}_{\mathbf{k}}^{(-)} e^{-i\Omega_k t} + \frac{i}{V} \sum_{n\mathbf{p}} U_{nn}(\mathbf{k},\mathbf{p}) < D_n >< a_{\mathbf{p}} >$$

To analytically handle Eq.(5) we have to know explicit expressions of function F and U. For F one can see e.g. [29,30] where their dependences on exciton momenta, transferred momentum, electron-hole effective mass ratio and two-exciton state spin symmetry are derived approximately and demonstrated graphically for different physical situations. As for functions U, they may be formulated as [27,31] :

$$U_{nm}(\mathbf{p},\mathbf{q}) = \frac{e^2}{\varepsilon V^2}\sum_{\mathbf{sl}}\left[\left(\frac{f^*(\mathbf{s}-\beta\mathbf{q}) - f^*(\mathbf{s}-\mathbf{q}+\alpha\mathbf{p})}{|\mathbf{p}-\mathbf{q}|} + \frac{f^*(\mathbf{s}-\beta\mathbf{q}) - f^*(\alpha\mathbf{p}-\mathbf{l})}{|\mathbf{p}-\mathbf{l}-\mathbf{s}|}\right)\varphi_n^*(\mathbf{l}-\mathbf{q}+\mathbf{p})\varphi_m(\mathbf{l})f(\mathbf{s}-\beta\mathbf{q})\right] \tag{6}$$

where e and ε are the electron charge and the static dielectric constant of the semiconductor, $\alpha = 1 - \beta = m_e/(m_e + m_h)$ with the electron (hole) effective mass denoted by m_e (m_h). Envelope functions f and φ describe the motion of an electron relative to a hole and to a positive donor-ion. It is well-known that functions F and U, in general, can by no means be calculated. However, in the long wave-length limit they are exactly calculable. In this limit F depends on a_x (exciton Bohr radius) and E_x^b (exciton binding energy) whereas U depends also on a_d (donor electron Bohr radius) and E_d^b (donor electron binding energy). Since the aim of this paper is to outline just qualitative aspects, we can take for U the interaction between an exciton and a free electron. Then, for the low temperature region, where the quasiparticle momenta are small, functions F and U have the following simplified forms [32] :

$$F(\mathbf{k},\mathbf{p},\mathbf{q}) \approx F(0,0,0) \equiv F = \frac{26}{3}\pi a_x^3 E_x^b \tag{7}$$

$$U_{nn}(\mathbf{k},\mathbf{p}) \approx U_{1S1S}(0,0) \equiv U = 24\pi a_x^3 E_x^i \tag{8}$$

Substituting Eqs.(7,8) into Eq.(5), we get

$$-\left[\frac{d}{dt} + \frac{1}{\tau_{\mathbf{k}}} + i\omega_k + 2iF\sigma + iU\rho\right] < a_{\mathbf{k}} > = ig_k\mathcal{E}_{\mathbf{k}}^{(-)}e^{-i\Omega_k t} \tag{9}$$

where $\sigma = V^{-1}\sum_{\mathbf{p}} < a_{\mathbf{p}}^+ a_{\mathbf{p}} >$ and $\rho = V^{-1}\sum_n < d_n^+ d_n >$ denote the exciton and donor densities. In obtaining Eq.(9) we have used the equality :

$$\sum_{\mathbf{p}} < a_{\mathbf{p}} > = < a_{\mathbf{k}} > \tag{10}$$

In fact, the l.h.s. of Eq.(10) equals to $< a_{\mathbf{k}} > + \sum_{\mathbf{p}\neq\mathbf{k}} < a_{\mathbf{p}} >$. However, the $\mathbf{p}$ ($\neq \mathbf{k}$)-modes are incoherent and their averages must vanish [28]. The $\mathbf{k}$-mode governed by the monochromatic classical light field behaves as a coherent and macroscopically occupied one. This allows us to solve Eq.(9) by seeking for a particular solution of Eq.(2) in the form :

$$< a_{\mathbf{k}} > = A_{\mathbf{k}}e^{-i\Omega_k t} \tag{11}$$

with $A_{\mathbf{k}}$ being a c−number which is to be found from the following equation (the index $\mathbf{k}$ will be for brevity suppressed from now on) :

$$\left[(\Omega - \omega - 2F\sigma - U\rho) + \frac{i}{\tau}\right] A = g\mathcal{E}^{(-)} \tag{12}$$

Combining Eqs.(11) and (12) yields

$$\sigma\left[(2F\sigma + U\rho - \Omega + \omega)^2 + \tau^{-2}\right] = g^2|\mathcal{E}^{(-)}|^2 \equiv g^2 I \tag{13}$$

Since Eq.(13) is a cubic (with respect to σ) equation, it might exhibit optical bistability. If $F = 0$, Eq.(13) becomes a linear one (with respect to σ) and no bistability can take place. So, it is the exciton-exciton interaction F that is responsible for the optical bistability occurrence. Physically, this interaction induces an energy shift of the exciton that brings about discontinuous density jumps when the light intensity is sweeping back and forth. The donor parameters U and ρ enter Eq.(13) just in capacity as factors influencing the ($\sigma - I$)-dependence. For convenience we introduce dimensionless normalized light intensity $\tilde{I} = 2F\tau^3 g^2 I$, dimensionless normalized exciton density $\tilde{\sigma} = 2F\tau\sigma$, dimensionless normalized donor density $\tilde{\rho} = \tau U\rho$ and dimensionless normalized laser detuning $\tilde{\delta} = \tau(\Omega - \omega)$ which transform Eq.(13) to the following dimensionless normalized equation :

$$\tilde{\sigma}\left[\left(\tilde{\sigma} - \tilde{\delta} - \tilde{\rho}\right)^2 + 1\right] = \tilde{I} \tag{14}$$

3 Donor-influenced optical bistability

Though Eq.(14) is a cubic equation with respect to $\tilde{\sigma}$, optical bistability could occur only under a certain condition. With some algebraic manipulations, it is easy to find from Eq.(14) the above-mentioned condition which simply reads

$$\tilde{\delta} > \tilde{\rho} + \sqrt{3} \tag{15}$$

Obviously, the presence of $\tilde{\rho}$ in the condition (15) already speaks of the donor influence on the excitonic optical bistability. But to answer the question how namely the influence is, we have to make a more careful analysis. Soppose we handle a laser which emits a coherent light beam with the frequency falling on the resonant spectral region of the exciton and on the region of well-developed optical bistability occurring in pure semiconductors. This means

$$\sqrt{3} \ll \tilde{\delta} < \tau E_x^b \text{ or the same } \sqrt{3}\tau^{-1} \ll \Omega - \omega < E_x^b \tag{16}$$

For CdS, for instance, $E_x^b = 33\,meV$ and $\tau = 7\,ps$ ($\sqrt{3}/\tau \approx 0.16\,meV$), the above spectral requirements are reasonably satisfied. Denote by $\tilde{I}_m$ and $\tilde{I}_M$ ($\tilde{I}_m < \tilde{I}_M$) the boundary values of the laser intensity domain within which optical bistability takes place. We can then derive their expressions in the case of n-type doped semiconductors as

$$\left.\begin{matrix}\tilde{I}_m \\ \tilde{I}_M\end{matrix}\right\} = \frac{2}{27}\left[2\left(\tilde{\delta}-\tilde{\rho}\right) \pm \sqrt{\left(\tilde{\delta}-\tilde{\rho}\right)^2 - 3}\right] \cdot \left[3 + (\tilde{\delta}-\tilde{\rho})^2 \mp (\tilde{\delta}-\tilde{\rho})\sqrt{\left(\tilde{\delta}-\tilde{\rho}\right)^2 - 3}\right] \tag{17}$$

If we fully neglect fluctuations , our system, when optical bistability occurs, can be used as a memory element for an optical computer with the lower and the upper stable states of exciton density being the 0 and the 1 of the memory element. In reality, fluctuations always exist and, as a consequence, they cause undesirable spontaneous and random jumps between

the two above-said states, now quasistable, leading to the loss of memory. Therefore, in a bistable device, the operating value of the incident light intensity must be chosen so that the jumping probability is as minimal as possible. This value intensity is called "holding intensity" $\tilde{I}_h$. The holding intensity, of course, lies in the bistable domain, i.e. $\tilde{I}_m < \tilde{I}_h < \tilde{I}_M$, but its exact value depends upon a concrete treatment of fluctuations that may come from different origins. Just for simplicity, we here conventionally put $\tilde{I}_h = \frac{1}{2}(\tilde{I}_m + \tilde{I}_M)$. As for optical bistability hysteresis loop size we formally define it by the quantity $\Delta\tilde{I} = \tilde{I}_M - \tilde{I}_m$. In Figs. 1 and 2 we plot $\tilde{I}_h$ and $\Delta\tilde{I}$ as functions of donor concentration $\tilde{\rho}$ for various given values of detuning $\tilde{\delta}$. Transparently, for a fixed detuning both holding intensity and hysteresis loop size are decreased when the degree of doping increases. Also, it can be seen from the Figures that the further the detuning (i.e. the larger $\tilde{\delta}$) the higher the rate of the decreasing (i.e. the bigger the slope of the curves in the Figures). From a practical viewpoint, two requirements, among others, for an ideal optical bistable device are low holding intensity and fast switching time. Since the latter depends on the hysteresis loop size [33], one may, as followed from above, optimize the optical bistability operation for a given input frequency in a doped semiconductor by selecting appropriate way and degree of doping, i.e. by preparing samples with proper kind and concentration of donors. Note that one can also change the size of the hysteresis loop by many other ways, for example, by applying the so-called squeezed light [34].

4 Determination of the parameter $\tilde{\rho}$

In this section, contrary to the previous one, we shall shortly discuss a somewhat inverse question. Namely, how can we experimentally determine parameters specifying the donor nature of the system under investigation by studying optical bistability in n-type doped semiconductors ? It is worth noticing at this moment that similar kinds of questions have been dealt with e.g. in [8,35]. The authors of [8] make use of optical bistability to solve the principal problem concerning the sign of the exciton-exciton coupling constant, whereas in [35] we have suggested a means to estimate relative magnitudes between coupling constants of the exciton-exciton and exciton-biexciton interactions in dense exciton-biexciton systems. The effective dynamical exciton-exciton interaction constant in molecular crystals might also be determined by measuring dispersion curves of the giant polariton energy lower-branches at different levels of excitation [36]. Here we try to propose two experimental methods for determining the parameter $\tilde{\rho}$ that enters Eq.(14).

Firstly, it seems quite easy to measure the hysteresis loop size from the experimental curve $\tilde{\sigma} = \tilde{\sigma}(\tilde{I})$ of well-developed optical bistability at a certain, large enough, detuning $\tilde{\delta}$. On the other hand, from Eqs.(17) we can derive the analytic expression for the hysteresis loop size $\Delta\tilde{I}$ which looks quite simple

$$\Delta\tilde{I} = \frac{4}{27}\left[\left(\tilde{\delta}-\tilde{\rho}\right)^2 - 3\right]^{3/2} \tag{18}$$

and so convenient to be inversed into

$$\tilde{\rho} = \tilde{\delta} - \sqrt{\left(\frac{27}{4}\Delta\tilde{I}\right)^{2/3} + 3} \tag{19}$$

Relation (19) reveals that the parameter $\tilde{\rho}$ can be determined from the experimentally known quantities $\tilde{\delta}$ and $\Delta\tilde{I}$. The former, as an external parameter, is known from the laser frequency used, while the latter is to be found from the measured curve $\tilde{\sigma} = \tilde{\sigma}(\tilde{I})$ of the optical bistability observed.

Secondly, the parameter $\tilde{\rho}$ can alternatively be determined by another manner of carrying out experiments. Suppose we are observing a well-developed optical bistability at some detuning $\tilde{\delta}$. We now tune the laser frequency down in order that the phenomenum will undergo the critical situation under which the optical bistability just begins to disappear, i.e. the curve $\tilde{\sigma} = \tilde{\sigma}(\tilde{I})$ just begins to turn from the S-shape form to that with an inflection point. Calling $\tilde{\delta}_c$ the critical value of the detuning at which such a critical situation happens, we have (see Eq.(15))

$$\tilde{\delta}_c = \tilde{\rho} + \sqrt{3} \tag{20}$$

or the same

$$\tilde{\rho} = \tilde{\delta}_c - \sqrt{3} \tag{21}$$

Equality (21) again allows us to determine $\tilde{\rho}$ from the value $\tilde{\delta}_c$ which is to be found experimentally. It is preferable to carry out both kinds of the experiments and then compare the results for correction. Knowing $\tilde{\rho}$ we could estimate the magnitude of ρ (*or* U), if U (*or* ρ) had been given beforehand (e.g. by other kinds of experiments). As the donor concentration can be known from the doping procedure, we may make use of the above suggestions to determine the effective exciton-donor coupling constant which is a very important parameter in physics.

In practice, one measures the output light intensity but not the exciton density in dependence on the input light intensity. The model for calculations must then be improved to involve also the internal photon and, as a result, the hysteresis loop may be very complicated [24]. Fortunately, as proved in [9], the conditions for and the hysteresis loop size of both kinds of optical bistability curves (output-input intensity curves and exciton density-input intensity curves) coincide. So the two possibilities discussed above of determining $\tilde{\rho}$ do not depend on what kind of the characteristic curves is being measured.

5 Conclusion

As a remark we stress that our model is valid only for donor concentration as low as $\tilde{\rho} \ll \tau E_x^b - \sqrt{3}$ (E_x^b is the exciton binding energy). For high concentration of donors the condition (15) is difficult to be met because to witness optical bistability one has then to blue tune the laser frequency that may violate the resonance requirement mentioned in the begining of Section 2. For an estimation, we take CdS with a_x (exciton Bohr radius) $\doteq 25.10^{-8} cm$, $E_x^b = 33\, meV$ and $\tau = 7\, ps$. If U is approximately defined by Eq.(8), we get $U \approx 39.10^{-21}\, eV.cm^3$ yielding the validity condition for the donor concentration as $\rho \ll 8.5.10^{17}\, cm^{-3}$. We thus expect that our model would work well e.g. for ρ up to $10^{16}\, cm^{-3}$.

In conclusion, we have studied optical bistability of excitons in n-type doped semiconductors. It has been shown that by choosing the way ($\propto U$)

and by adjusting the degree ($\propto \rho$) of doping one may control hysteresis loop size as well as holding intensity of the optical bistability which takes place at a given laser frequency. This would provide a means for optimizing the bistable device operation. On the other side, measuring the optical bistability curves under the well-developed and under the critical situations one may get useful estimation of the concentration of donors and their interaction with excitons. The model proposed and the approximations used for calculations are the simplest possible . However, we think that the obtained results, though only qualitative and still very far from full reliance, seem rather helpful. It is hoped that they could serve as a reference for theoretists, experimentalists and also for technologists towards more realistic considerations and fruitful applications along this research direction.

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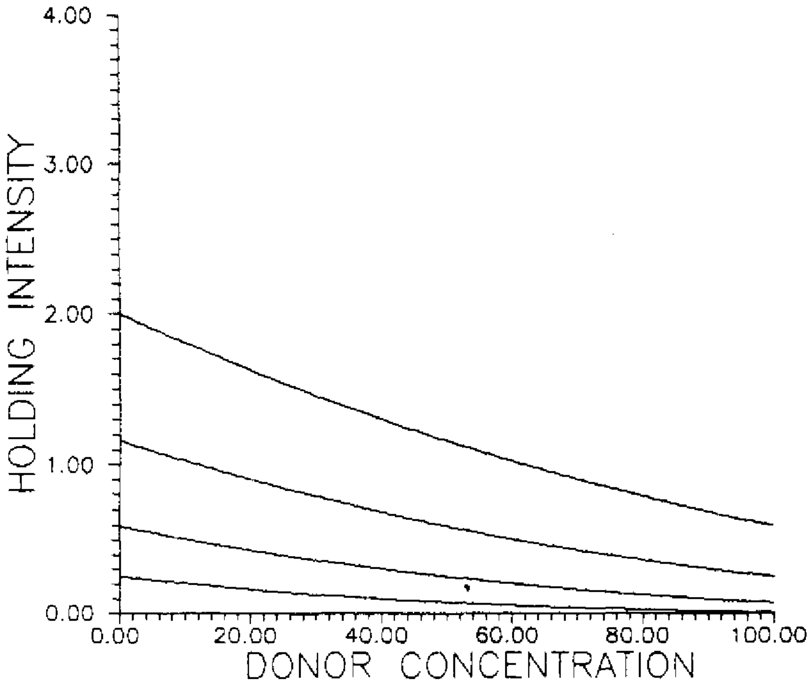
Figure captions

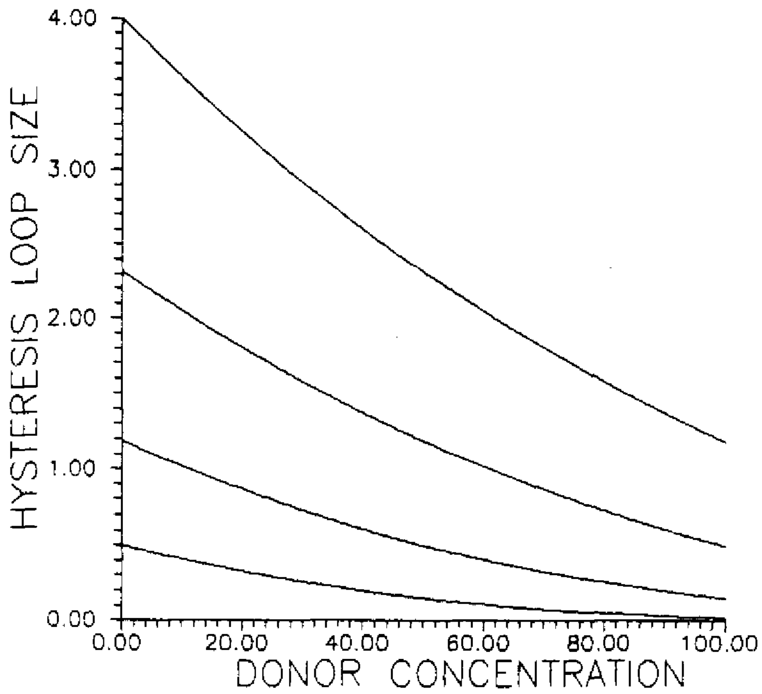
Fig.1 : Dimensionless normalized holding intensity $\tilde{I}_h.10^{-6}$ as a function of dimensionless normalized donor concentration $\tilde{\rho}$ for several demensionless normalized detuning $\tilde{\delta}$ ($\tilde{\delta} = 300$, 250, 200 and 150 downwards).

Fig.2 : The same as in Fig. 1, but for dimensionless normalized hysteresis loop size $\Delta\tilde{I}.10^{-6}$.



4.00
3.00
2.00
1.00
0.00
HOLDING INTENSITY
0.00
20.00
40.00
60.00
80.00
100.00
DONOR CONCENTRATION

Fig.1



4.00
3.00
2.00
1.00
0.00
HYSTERESIS LOOP SIZE
0.00
20.00
40.00
60.00
80.00
100.00
DONOR CONCENTRATION

Fig.2