VOLUME 49, NUMBER 20

result is that, after a rotation by π around \hat{x} , the anisotropy energy remains at its maximum value during a subsequent rotation around \hat{y} , at least as long as an uncontrollable rotation around the axis of the remanent magnetization can be avoided. These properties (characteristic of tridirectional anisotropy) result from the existence of antisymmetric interactions as well as from the macroscopic isotropy of spin-glasses.^{7,9,10} With respect to the problem of ESR in spin-glasses our results confirm the validity of the theoretical approach of Henley, Sompolinsky, and Halperin¹⁰ and Legget.¹¹

³F. Hippert and H. Alloul, J. Phys. (Paris) <u>43</u>, 691 (1982).

⁴H. Alloul, J. Appl. Phys. <u>50</u>, 7330 (1979).

⁵P. Monod and Y. Berthier, J. Magn. Magn. Mater. <u>15-18</u>, 149 (1980).

⁶S. Schultz, E. M. Gullikson, D. R. Fredkin, and M. Tovar, Phys. Rev. Lett. <u>45</u>, 1508 (1980).

⁷W. M. Saslow, Phys. Rev. B <u>22</u>, 1174 (1980), and Phys. Rev. Lett. <u>48</u>, 505 (1982).

⁸A. Fert and P. M. Levy, Phys. Rev. Lett. <u>44</u>, 1538 (1980).

⁹P. M. Levy, C. Morgan-Pond, and A. Fert, J. Appl. Phys. <u>53</u>, 1168 (1982).

¹⁰C. L. Henley, H. Sompolinsky, and B. I. Halperin, Phys. Rev. B <u>25</u>, 5849 (1982).

¹¹A. J. Leggett, private communication.

¹²F. Hippert, H. Alloul, and A. Fert, in Proceedings of the Twenty-Eighth Conference on Magnetism and Magnetic Materials, Montréal, 1982 (to be published).
¹³J. J. Prejean, M. Joliclerc, and P. Monod, J. Phys. (Paris) 41, 427 (1980).

¹⁴A. Fert, to be published.

¹⁵T. Iwata, K. Kai, T. Nakamichi, and M. Yamamoto,

J. Phys. Soc. Jpn. <u>28</u>, 582 (1970).

¹⁶A. Fert and F. Hippert, to be published.

Many-Body Theory of Optical Bistability in Semiconductors

J. P. Löwenau, S. Schmitt-Rink, and H. Haug Institut für Theoretische Physik der Universität Frankfurt, D-6000 Frankfurt am Main, Federal Republic of Germany (Received 6 July 1982)

A unified microscopic theory of the observed optical bistabilities in narrow-gap semiconductors, such as InSb and GaAs, is developed by calculating the complex, nonlinear dielectric function for arbitrary free-carrier concentrations from an integral equation for the polarization function.

PACS numbers: 72.20.Jv, 42.65.-k, 77.40.+i, 78.90.+t

In 1979 dispersive optical bistability was observed in the narrow-gap semiconductors GaAs (Ref. 1) and InSb.² The driving optical nonlinearity in GaAs is connected with the ionization of the exciton, while the nonlinearity in InSb is mainly due to filling of the bands with free carriers. Recently, the observation of optical bistability has been reported also for Te.³ In all these experiments a relatively small number of electron-hole (e-h) pairs is excited in the low-transmission state, while in the high-transmission state a relatively dense e-h plasma is generated. Thus, a microscopic theory of the optical bistability has to connect the low- and high-density regimes. The dominant correlations in the low-density regime are caused by the attractive e-h Coulomb interaction, which gives rise to the formation of

excitons in GaAs. In this substance the exciton binding energy E_0 is — in bulk materials at least at low temperatures and in superlattice structures even at room temperature — larger than its broadening γ . In InSb the broadening always exceeds the binding energy E_0 , so that the exciton resonance is not resolved. In the high-density regime the screening of the Coulomb interaction by intraband scattering is the most important process and is well described by the random-phase approximation (RPA). A consistent theory of the intermediate-density regime is rather difficult, especially for low temperatures and wide-gap semiconductors, where many-exciton effects become important.

In this Letter, we will neglect many-exciton effects and will combine consistently the processes

¹J. S. Kouvel, J. Phys. Chem. Solids <u>21</u>, 57 (1961). ²P. Monod, J. J. Prejean, and B. Tissier, J. Appl. Phys. <u>50</u>, 7324 (1979).

~ ~

which are asymptotically most important. The e-h correlation can be described by the polarization function $P_{\rm e\,h}$, which at the same time determines the optical dielectric function

$$\epsilon(\omega) = \epsilon_{\infty} - \frac{8\pi e^2}{m_0^2} \sum_{\vec{k}, \vec{k}'} M_{vc}(\vec{k}) M_{vc} * (\vec{k}') \times P_{e,b}(\vec{k}, \vec{k}'; \omega).$$
(1)

 ϵ_{∞} is the background dielectric constant; $M_{vc}(\bar{k})$ is the dipole matrix element for direct optical transitions between the valence and the conduction band. Its form is taken from the kp perturbation theory. m_0 and e are the mass and charge of the free electrons, respectively.

The polarization function obeys in the screenedladder approximation a Bethe-Salpeter equation (BSE) of the form⁴

$$\underbrace{\overset{P_{eh}}{\longrightarrow}}_{G_{h}} = \underbrace{\overset{G_{e}}{\longrightarrow}}_{G_{h}} + \underbrace{\overset{G_{e}}{\longrightarrow}}_{G_{h}} \overset{P_{eh}}{\longrightarrow}}_{G_{h}}$$
(2)

In our high-temperature (or narrow-gap) approximation the screening is mainly due to intra-



FIG. 1. Calculated spectra of absorption and refraction for GaAs at various free-carrier densities n. The temperature of the electronic excitations is assumed to be 10 K.

band scattering of free carriers, i.e., contributions of the excitons to the screening are neglected. Thus, V_s and the single-particle Green's function G_i with i = e, h are given by the RPA:

$$V_{s} = V_{q} / \varepsilon_{ii}(q, \omega) = \Longrightarrow = \cdots + \int_{a}^{b_{i}} \int_{$$

$$\stackrel{\scriptstyle \leftarrow}{\underset{\scriptstyle G_{i}}{\Rightarrow}} = \stackrel{\scriptstyle \leftarrow}{\underset{\scriptstyle G_{i}^{\circ}}{\rightarrow}} + \stackrel{\scriptstyle \leftarrow}{\underset{\scriptstyle G_{i}^{\circ}}{\rightarrow}} \stackrel{\scriptstyle \leftarrow}{\underset{\scriptstyle V_{s}}{\rightarrow}} \stackrel{\scriptstyle \leftarrow}{\underset{\scriptstyle G_{i}}{\rightarrow}}$$
(4)

The set of integral equations (2)-(4) has been treated frequently in the low- and high-density limits.⁵ In order to get a solution for arbitrary free-carrier concentrations, we simplify the problem by treating the screening in a quasistatic single-plasmon-pole approximation, which is known to produce relatively good self-energy shifts. For simplicity the damping γ of the pair state is introduced phenomenologically. Furthermore, if only s-wave scattering is considered, Eq. (2) reduces to a one-dimensional integral equation for the auxiliary function $Q(\mathbf{k}, \omega)$ = $\sum M_{vc}^{*}(\mathbf{k}')P_{eh}(\mathbf{k},\mathbf{k}';\omega)$. The integral equation for the associate vertex function can be solved very accurately⁶ by a numerical matrix inversion. The details of the mathematical procedure are described in Ref. 5. The solution yields via Eq. (1) immediately the real and imaginary parts of the dielectric function. Note that a Kramers-Kronig transformation cannot be used to calculate $\epsilon'(\omega)$ from $\epsilon''(\omega)$, if the latter is known only in a very limited frequency range.^{7,8}

The following material parameters have been used: (a) GaAs: $\epsilon_{\infty} = 10.9$, $\epsilon_{0} = 12.35$, $m_{e} = 0.067$



FIG. 2. Experimental absorption spectra for GaAs at various excitation intensities I. The curves with I = 0, I_1 are taken from Ref. 11, the curve with $I = I_2$ is taken from Ref. 10.

 $\times m_{0}$, $m_{h} = 0.54 m_{0}$, $\gamma = 0.05 E_{0}$, $E_{g} = 1519.2$ meV; (b) InSb: $\epsilon_{\infty} = 15.7$, $\epsilon_{0} = 17.05$, $m_{e} = 0.0145m_{0}$, m_{h} = $0.4m_0$, $\gamma = 2.0E_0$, $E_s = 230$ meV. The resulting absorption and refraction spectra $\alpha(\omega)$ and $n(\omega)$, respectively, are shown in Fig. 1 for GaAs for a temperature of 10 K for the electronic excitations and for various free-carrier concentrations n. For the unexcited crystal (n = 0), one sees for the damping $\gamma = 0.05E_0$ the 1s- and 2s-exciton absorption lines well resolved. The ionization continuum for n = 0 agrees well with Elliot's result.⁹ With increasing free-carrier concentration, the ionization continuum shifts to lower energies. The lowest exciton resonance is seen to shift very little, due to the high degree of compensation between the weakening of the e-h binding and the band-gap reduction. The improved compensation as compared to Ref. 6 has been obtained by an accurate calculation of temperature dependence of the single-particle energy. The curve with $n = 5 \times 10^{15}$ cm⁻³ is slightly above the Mott transition, but still a large excitonic resonance is present. This excitonic enhancement would be



FIG. 3. Calculated spectra of absorption and refraction for InSb at various internal excitation intensities I_i . The frequency of the pump beam was assumed to be $\omega_0 = 225$ meV. The electronic temperature is T = 77K. The experimental points of the low-intensity absorption spectrum are taken from Ref. 13.

reduced slightly if the concentration dependence of γ were included. The optical gain ($\alpha < 0$) due to the population inversion for $\omega \leq \mu$, where μ = $\mu_{\rm e} + \mu_{\rm h}$ is the quasichemical potential, cannot be observed in single-beam experiments in which one is limited to the region $\omega \ge \mu$. The associate refraction spectra show that around the 1s level large dispersive changes occur with increasing free-carrier concentration. Especially for ω $\leq E_{1s}$ the refraction index decreases with increasing free-carrier concentration. This optical nonlinearity due to the ionization of the excitons is the mechanism responsible for the observed optical bistability in GaAs.¹ Experimental absorption spectra of GaAs according to Hildebrand, Faltenmeier, and Pilkuhn¹⁰ and Shah, Leheny, and Wiegmann¹¹ are shown for comparison in Fig. 2 for three excitation intensities. One sees that there is good qualitative agreement with the calculated spectra. Quantitatively similar results have been obtained in a single-beam experiment by Gibbs et al.¹² Naturally, in a single-beam experiment one observes a residual absorption even at high light intensities due to the fact that the quasichemical potential μ always stays below ω.

As already discussed, the main difference between InSb and GaAs is the opposite ratio of the damping γ to the exciton Rydberg E_0 . Because of this difference, the exciton resonances are not resolved in InSb at T = 77 K (see Fig. 3), but the excitonic enhancement still causes large devia-



FIG. 4. (a) Carrier density; (b) absorption $\alpha(\omega_0, I_i)$ and refraction $n(\omega_0, I_i)$ at $\omega_0 = 225$ meV vs the internal excitation intensity I_i for InSb at T = 77 K.



FIG. 5. Comparison of the calculated changes of the averaged refraction spectrum -dn/dI for intensities *I* between 30 and 50 W/cm² with the measured coefficient n_2 , defined by $n(\omega) = n_0(\omega) - n_2(\omega)I$, according to Ref. 14 for InSb at 77 K.

tions from the square-root dependence of $\alpha(\omega)$ close to the band edge for small free-carrier concentrations. The experimental values of $\alpha(\omega)$ for $n \rightarrow 0$ are taken from Ref. 13. For InSb, where real bound e-h pairs never exist, the free-carrier concentrations can in the stationary case be expressed directly in terms of the laser intensity via a simple rate equation.⁸ The resulting relation is plotted in Fig. 4(a), where the saturation effects are clearly seen. The crystal bleaches when the quasichemical potential μ approaches the pumping frequency ω_0 . The changes of the absorption and refraction at a fixed frequency with varying laser intensity are shown in Fig. 4(b). The changes of the refraction index with intensity have been measured¹⁴ from the beam-profile distortion at the onset of defocusing between 10 and 40 W/cm². In the linear approximation $n(\omega)$ $=n_0(\omega) - n_2(\omega)I$ of Ref. 14, these changes are described by the function $n_2(\omega)$. In Fig. 5 we compare the experimental values of Miller *et al.*¹⁴ with the calculated first derivative – $dn(\omega)/dI$ averaged for intensities between 30 and 50 W/cm^2 . A surprisingly good agreement is obtained, if one considers that the InSb crystals contained an impurity concentration of the order of 2×10^{14} cm⁻³.

In conclusion, we have shown that an accurate numerical solution of the linear shielded-potential approximation allows an unified description of the optical nonlinearities in GaAs and InSb, which are responsible for the observed optical bistabilities in these narrow-gap materials.

We appreciate valuable comments of Professor S. D. Smith on our manuscript. This work is a project of the Sonderforschungsbereich Frankfurt/ Darmstadt, financed by special funds of the Deutsche Forschungsgemeinschaft.

¹H. M. Gibbs, S. L. McCall, T. N. C. Venkatesan, A. C. Gossard, A. Passner, and W. Wiegmann, Appl. Phys. Lett. <u>35</u>, 45 (1979).

²D. A. B. Miller, S. D. Smith, and A. Johnston, Appl. Phys. Lett. <u>35</u>, 658 (1979); for a recent review see A. Miller, D. A. B. Miller, and S. D. Smith, Adv. Phys. <u>30</u>, 697 (1981).

 3 G. Staupendahl and K. Schindler, Opt. Quantum Electron. <u>14</u>, 157 (1982).

⁴See, e.g., C. Klingshirn and H. Haug, Phys. Rep. <u>70</u>, 315 (1980).

⁵See, e.g., W. F. Brinkman and P. A. Lee, Phys. Rev. Lett. <u>31</u>, 257 (1973); M. Rössler and R. Zimmermann, Phys. Status Solidi (b) <u>67</u>, 525 (1975). Further references are compiled in the review article by Klingshirn and Haug (Ref. 4).

⁶S. Schmitt-Rink, J. P. Löwenau, and H. Haug, Z. Phys. B <u>47</u>, 13 (1982).

⁷T. S. Moss, Phys. Status Solidi (b) <u>101</u>, 555 (1980). ⁸S. W. Koch, S. Schmitt-Rink, and H. Haug, Phys. Status Solidi (b) <u>106</u>, 135 (1981).

⁹R. J. Elliot, Phys. Rev. <u>108</u>, 1384 (1957).

¹⁰O. Hildebrand, B. Faltenmeier, and M. H. Pilkuhn, Solid State Commun. <u>19</u>, 841 (1976).

¹¹J. Shah, R. F. Leheny, and W. Wiegmann, Phys. Rev. B <u>16</u>, 1577 (1977).

¹²H. M. Gibbs, A. C. Gossard, S. L. McCall, A. Passner, W. Wiegmann, and T. N. C. Venkatesan, Solid State Commun. <u>30</u>, 271 (1979).

¹³G. W. Gobeli and H. Y. Fan, Phys. Rev. <u>119</u>, 613 (1960).

 $^{14}\text{D.}$ A. B. Miller, C. T. Seaton, M. E. Price, and S. D. Smith, Phys. Rev. Lett. <u>47</u>, 197 (1981), and private communication.