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Simplified theory of optical nonlinearities in spin-polarized semiconductors

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Abstract
The spin degree of freedom is largely disregarded in existing theories of the density-dependent optical properties of an interacting electron–hole plasma in quasiequilibrium. Here, we extended the pair equation, which is applicable to a bulk semiconductor at elevated temperatures, to calculate optical nonlinearities due to a spin-polarized plasma. We obtained agreement with recent circular dichroism data in laser-excited GaAs by using the plasma density alone as the fitting parameter. The simplicity of our theory, based on the analytical pair-equation formula, makes it ideal for conveniently modelling absorption in a carrier spin-polarized semiconductor.

1. Introduction

The application of the spintronics paradigm to optoelectronics has resulted in novel semiconductor devices based on the optical properties of spin-polarized carriers. A prominent example is the spin vertical cavity surface-emitting laser. In this device, the quasiequilibrium plasma of spin-polarized electrons injected for lasing causes the optical constants of the semiconductor to be modified by spin-polarized density-dependent optical nonlinearities (ON). The physics of these ON can be studied by circularly polarized pump–probe experiments. In a recent experiment by Nemec *et al* [1] in bulk GaAs at room temperature, a right circularly polarized pump pulse initially excited electron spins; the simultaneously created hole spins rapidly relax in less than 100 fs. The change in the sample's transmission due to these excess quasiequilibrium carriers was probed later by right (σ^+) and left (σ^-) circularly polarized light. The spin-polarized electrons caused a difference in the absorption of σ^+ and σ^-, i.e. circular dichroism, which showed a change in sign as the probe photon energy was varied. The crossover was attributed to competing spin-polarized ON from phase-space filling (PSF) and bandgap renormalization (BGR). The results of this experiment were very recently explained by an elaborate three-band model for the optical properties of carrier spin-polarized semiconductors [2]. The theory was based on the earlier spin-unpolarized Green's function formalism [3, 4], which requires the time-consuming implementation of a specialized, numerical solution to an integral equation. A less sophisticated version of this theory would be helpful to experimentalists by enabling a direct, quicker interpretation of the findings of circularly polarized, quasiequilibrium pump–probe experiments. Besides, a more analytic theory could lead to insights which may otherwise be masked by elaborate numerics.

Green's function theory gives a microscopic description of the effect of a spin-unpolarized plasma on absorption, in terms of density-dependent ON due to PSF, BGR and screening of the Coulomb enhancement (CE) of absorption. Although at any probe photon energy BGR increases absorption whereas both PSF and the screening of the CE decrease absorption, these mechanisms of ON are incorporated in a way that makes it difficult to disentangle their individual contributions. Nevertheless, the different pump powers used to excite a spin-unpolarized plasma corresponded to plasma densities in the microscopic calculation for density-dependent absorption [5], which compared excellently with experiment [6].

Good agreement with these experimental results was also obtained by the more phenomenological pair-equation model for ON [6–8], elements of which are widely used in studies of saturable absorbers [9, 10] and in laser-excited bulk GaN [11], AlInGaN [12], CuCl [13, 14] and $In_{1-x}Ga_xSb$ [15]. Although the plasma density assumes the role of a fitting parameter in this theory, it has the advantage of offering an analytical, easily computed expression for the density-dependent absorption spectrum, compared to the full microscopic theory. The different ON are also represented by independent terms in this expression, permitting the effect of each one to be isolated [6].

However, PSF is incorporated in a way that is formally consistent but that does not entirely account for its effect on absorption. It is recognized that, at room temperature, PSF is one of the dominant ON (the other being the screening of the CE) [6, 16]. This could explain why, in comparing against the same spin-unpolarized data as the microscopic theory [5], the pair-equation theory [6] used generally higher plasma densities as the fitting parameter. On the other hand, in semiconductors where PSF is very important, for example in the narrow gap InGaSb, this problem has been circumvented by using the full expression for PSF in the pair-equation model to obtain agreement with the data [15].

Here, we merged the closed-form, analytical formula for absorption in the pair-equation model with the spin degree of freedom to yield a simplified theory for spin-dependent ON. It reproduced the key feature of the circularly polarized pump–probe experiment of Nemec *et al*, the crossover in circular dichroism, using only the fitting parameter of the original pair equation, i.e. the plasma density. Within the framework of our theory having separable contributions from each of the ON, the crossover is due to an interplay between spin-dependent versions of the PSF and BGR alone. Furthermore, an analytical understanding of the experimentally observed density- and spin-dependent trends, which was missing in the microscopic theory, is now made possible. The screening of the CE of absorption does not determine the crossover energy since both helicities of the probe experience the same screening by the plasma. Thus, unlike the pair-equation model for spin-unpolarized ON in GaAs, where screening masks the incomplete inclusion of PSF, here it was necessary to use the full (spin-dependent) PSF to capture the crossover in circular dichroism. Besides, we found that this modification of the PSF in the pair-equation model enabled it to compare well with the previous data for the spin-unpolarized, density-dependent absorption of Lee *et al* [6], using plasma densities estimated from the microscopic theory.

After reviewing the spin-unpolarized pair-equation model, we describe below its extension to capture the spin degree of freedom. We also give an expression for the Fourier transform of the Hulthén potential used for screening the electron–hole attraction, so as to facilitate the calculation of spin-dependent BGR. The agreement obtained in this work with the experimental data for circular dichroism suggests that, with a minor modification, the pair-equation model can indeed simplify the calculation of spin-dependent ON in carrier spin-polarized semiconductors.

2. Method

2.1. Spin-unpolarized optical nonlinearities

In spin-unpolarized pump–probe experiments, the pump creates the same number of spin-down (↓) and spin-up (↑) electrons, both of which equally contribute to the probed nonlinearities in absorption. For an arbitrary density of interacting electrons and holes of densities n and p, respectively ($n = p$), the optical response has to be obtained numerically from the microscopic theory. However, by ignoring the effect of PSF (and BGR) on the reduction of the electron–hole attraction and by ascribing it only to plasma screening, the pair-equation approximation offers an analytical solution for density-dependent absorption $\alpha(\omega, n)$ due to an interacting spin-unpolarized plasma. In this approximation [7, 8]

$$\alpha(\omega, n) = \frac{4\pi^2\omega}{n_b c}|d_{cv}|^2 A(\omega, n, p) \sum_{\nu} |\Phi_\nu(\boldsymbol{r} = 0)|^2 \times \delta(\hbar\omega - E_\nu - \Delta E_g), \quad (1)$$

where d_{cv} is the interband dipole matrix element and n_b is the background refractive index. In (1), $|\Phi_\nu(\boldsymbol{r} = 0)|^2$ suppresses absorption by screening the bound states and the CE. $|\Phi_\nu(\boldsymbol{r} = 0)|^2$ and E_ν can be obtained analytically as eigenfunctions and eigenvalues of the modified Wannier equation, provided that the electron and hole attract via the screened Hulthén potential. The term involving the summation over ν in (1), is given in [7] (see equations (44) and (45) there). The spin degeneracy is included in all summations, unless otherwise indicated. BGR is described by ΔEg as $\Delta Eg/E_0 = -1+(1-1/g)^2$ for $g \geqslant 1$ and as $\Delta Eg/E_0 = -1/g$ for $g < 1$, where $g = 12/(\pi^2 a_0 \kappa)$, a_0 is the exciton Bohr radius, E_0 is the exciton Rydberg and κ is the screening wavevector. κ can obtained from equation (46) of [7]. BGR increases absorption by causing the optical transition to occur at larger quasimomenta, where there are more available states. In (1), we have substituted the PSF term in the original pair equation [7], $A_0(\omega, n, p) = \tanh[\beta(\hbar\omega - E_g - \mu_e - \mu_h)/2]$, by $A(\omega, n, p) = 1 - f_e(\omega) - f_h(\omega)$. Here

$$f_j(\omega) = \frac{1}{\exp\left[\beta\left((\hbar\omega - E_g)\frac{m_r}{m_j} - \mu_j\right)\right] + 1} \qquad (j = e, h), \quad (2)$$

where μ_j is the chemical potential (got by solving equation (47) of [7]), $m_r = (1/m_e + 1/m_h)^{-1}$ and $\beta = 1/(k_B T)$. The substitution resulted in $\alpha(\omega, n)$ correctly approaching the noninteracting (free carrier) expression for absorption in the limit of no Coulomb interaction, unlike the original pair-equation model [7, 8]. Moreover, it has been shown [7, 8] that we can write $A(\omega, n, p) = f A_0(\omega, n, p)$, where f varies between 0 and 1. Thus, the suppression of absorption by the PSF is reflected more in $A(\omega, n, p)$ than in $A_0(\omega, n, p)$. Note also that $\alpha(\omega, n \to 0)$ reduces to the well-known Elliott formula, as expected [7].

2.2. Spin-polarized optical nonlinearities

In the following, we extend the pair equation to a spin-polarized plasma. In the two-band model presented here, the selection rules dictate that the σ^+ pump excites from the valence band into the conduction band, three times as many ↓ electrons as ↑ electrons [17]. This results in a net electron spin polarization ξ in the sample given by $\xi = (n_\downarrow - n_\uparrow)/(n_\downarrow + n_\uparrow)$, where n_s is the number of electrons in the $s = \downarrow, \uparrow$ spin bands and $n_\downarrow + n_\uparrow = n$. Since it follows from the selection rules that σ^+ also probes the ↓ electrons three times more sensitively than ↑ electrons (and σ^- mostly probes the ↑ electrons), α^+ (α^-) describing the absorption of σ^+ (σ^-) was expressed as

$$\alpha^+ = \tfrac{3}{4}\alpha^\downarrow + \tfrac{1}{4}\alpha^\uparrow \qquad \alpha^- = \tfrac{1}{4}\alpha^\downarrow + \tfrac{3}{4}\alpha^\uparrow, \quad (3)$$

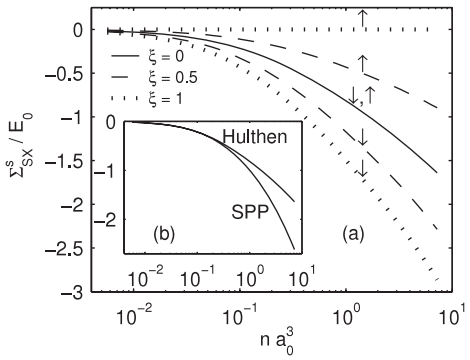

Figure 1. Density dependence of screened exchange energy Σ^s_{SX} for $s = \downarrow, \uparrow$ spin bands, using the (a) Hulthén potential for various spin polarizations ξ and (b) single-plasmon-pole (SPP) potential used in the full microscopic theory [4] (with $C = 4$) for $\xi = 0$. A comparison with the Hulthén potential is also shown.

where α^s is the absorption into the s spin band. Due to spin-dependent ON, the pumped spin imbalance causes $\alpha^\downarrow \neq \alpha^\uparrow$, resulting in circular dichroism, $\Delta\alpha = \alpha^+ - \alpha^-$. We represented the spin-dependent PSF by $A^s(\omega, n_s, p) = A(\omega, 2n_s, p)$, reflecting the differential blocking of states in the $\downarrow$ and $\uparrow$ spin bands. The bandgap is also differentially renormalized for these bands because $n_\downarrow \neq n_\uparrow$. The steps used to calculate this spin-dependent BGR, ΔE^s_g, are outlined below.

ΔE^s_g can be written [4] as a sum over the conduction and valence bands of a ξ-independent (and band-independent) 'Coulomb hole' term, Σ_{CH}, and a ξ-dependent 'screened exchange' term, Σ_{SX}. If $\xi = 0$, $\Delta E^\downarrow_g = \Delta E^\uparrow_g = \Delta E_g$. However, for non-zero ξ, $\Delta E^\downarrow_g \neq \Delta E^\uparrow_g$ due to only the electronic $\Sigma^s_{SX}(\xi)$ term, since the holes are unpolarized. Hence, ΔE^s_g was obtained using

$$\Delta E^s_g = \Delta E_g - \Sigma^s_{SX}(\xi = 0) + \Sigma^s_{SX}(\xi). \tag{4}$$

Note that $\Sigma^\downarrow_{SX}(\xi = 0) = \Sigma^\uparrow_{SX}(\xi = 0)$. We computed a k-independent Σ^s_{SX} from (spin degeneracy excluded from summation): $\Sigma^s_{SX} = -\sum_k V_H(k) f^s_e(k)$, where

$$V_H(k) = i\frac{\pi e^2 a_0 g}{\epsilon_0 k L^3}[\psi'(ika_0 g/2) - \psi'(-ika_0 g/2)] \tag{5}$$

is the Fourier transform of the Hulthén potential $V_H(r)$ given by [8] $V_H(r) = [2e^2/(\epsilon_0 a_0 g)]/[\exp(2r/(a_0 g)) - 1]$. In (5), $\psi'(z)$ is the first derivative of the digamma function. The ξ and density dependence of Σ^s_{SX} is shown in figure 1.

In the simplified model considered here, screening of the CE was spin-independent. Thus, we expressed $\alpha^s(\omega, n, \xi)$ as

$$\alpha^s(\omega, n, \xi) = \frac{4\pi^2\omega}{n_b c}|d_{cv}|^2 A^s(\omega, n, p) \sum_\nu |\Phi_\nu(\boldsymbol{r} = 0)|^2 \times \delta(\hbar\omega - E_\nu - \Delta E^s_g). \tag{6}$$

Note that the separability of the various ON in (1) is retained in (6). If $\xi = 0$, then $A^s(\omega, n_s, p) = A(\omega, n, p)$ and $\Delta E^s_g = \Delta E_g$, which caused $\alpha^\downarrow = \alpha^\uparrow$. This further resulted in $\alpha^+ = \alpha^- = \alpha(\omega, n)$ (cf (3)) in this limit, as expected.

Unlike equation (2) of Nemec *et al* [1] which is also based on the spin modification of a pair equation, our model permitted the actual computation of α^+ and α^- by explicitly accounting for spin-dependent many-body effects. The Fourier transform of the Hulthén potential obtained by us in (5) enabled the self-consistent calculation of spin-dependent BGR (cf (4)). Moreover, our model is valid even at low photon energies in the vicinity of the bandgap E_g since we did not neglect the contribution to absorption from screening of the bound states by the plasma (i.e. summation term in equation (45) of [7]).

2.3. Comparison with experiment

The calculated circular dichroism $\Delta\alpha(\omega, n, \xi)$, using n as the sole fitting parameter, was compared with the experimental data at a probe delay of $t = 7$ ps. The corresponding ξ was got from $\xi(t) = \xi_{max}\exp(-2t/\tau_s)$, describing the relaxation of spin polarization $\xi_{max} = 0.5$ within a characteristic time [1] $\tau_s = 130$ ps after its creation at $t = 0$ by the pump. The unpumped absorption α_0 of the sample was computed from the expression for $\alpha(\omega, n)$ in (1), with the background doping density $n = 10^{15}$ cm^{-3}. The experimental probe spectral width [1] of 30 meV was accounted for by Gaussian averaging about each ω of the calculated absorption spectra. The material parameters used were: exciton Rydberg $E_0 = 4.2$ meV, exciton Bohr radius $a_0 = 124.3$ Å, electron mass $m_e = 0.0665\, m_0$, hole mass $m_h = 0.52\, m_0$, $E_g = 1.424$ eV, $\epsilon_0 = 13.71$ and $T = 295$ K. The broadening of the sample, Γ, was taken to be density-dependent [6]: $\Gamma = (1.25 + 2 \times 10^{-18}\ \text{cm}^3 \times n)E_0$.

3. Results and discussion

We now present the results. The modification of the PSF term of the original pair equation leaves intact its ability to describe spin-unpolarized density-dependent absorption. Moreover, it permits our extension of the pair equation to capture spin-dependent ON revealed in the absorption of circularly polarized light by a spin-polarized plasma. This is achieved by using the plasma density as the sole fitting parameter, as in the original pair equation. We justify these assertions by comparing with experimental data.

The comparison of the modified pair equation using the full PSF in (1) with the density-dependent absorption spectra of Lee *et al* [6] is shown in figure 2. The agreement (cf figure 1 of [6]) shows that the substitution of the original $A_0(\omega, n, p)$ with $A(\omega, n, p)$ is able to reproduce the data using plasma densities estimated by the microscopic theory [5]. These densities were lower than those obtained from the original pair equation [6].

Our simple model generated the experimentally observed circular dichroism induced by the spin-polarized carriers and its main feature—the spectral crossover (figure 3(a)). This crossover was interpreted in terms of the opposing effect of PSF and BGR on the absorption of circularly polarized light. After a majority of $\downarrow$ electrons are pumped by σ^+, PSF suppresses α^+ more than α^- (cf (3)), resulting in $\Delta\alpha$ being negative. On the other hand, the larger BGR experienced by

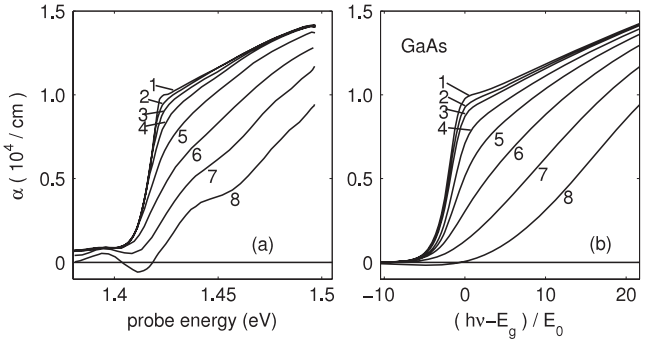

Figure 2. Comparison between experiment and theory for spin-unpolarized optical nonlinearities in absorption. (a) Experimental spectra probed after pumping a 15 μm spot with a power of (1) 0, (2) 0.2, (3) 0.5, (4) 1.3, (5) 3.2, (6) 8, (7) 20 and (8) 50 mW. The oscillations in curves 7 and 8 are an experimental artefact. The data shown are obtained from figure 1(a) of [6]. (b) Calculated spectra for plasma densities (got from the full microscopic theory [5]) of (1) 0, (2) 1.5×10^{16}, (3) 3×10^{16}, (4) 8×10^{16}, (5) 1.8×10^{17}, (6) 4×10^{17}, (7) 8×10^{17} and (8) 1.4×10^{18} cm^{-3}.

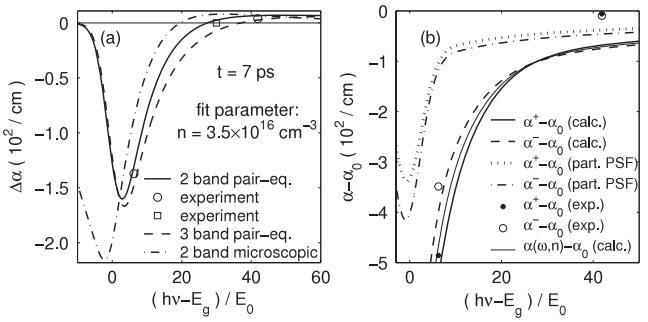

Figure 3. Comparison between theory and experiment for spin-polarized optical nonlinearities. (a) Circular dichroism, $\Delta\alpha$. The estimated experimental plasma densities are 1.3×10^{17} cm^{-3} (circles) and 6×10^{16} cm^{-3} (square). Calculations based on the two-band pair equation of (6) (solid curve) are compared with those of the appropriate two-band version of the existing microscopic theory (dotted–dashed curve) and with a further extension of the pair equation to a three-band model which includes the influence of light holes (dashed curve). The adequate agreement obtained with the data by the simpler (6) justifies its use. (b) Corresponding change (using (6)) in α^+ and α^- from the unpumped absorption α_0, induced by the σ^+ pump with the same parameters as (a). The crossover in $\Delta\alpha = \alpha^+ - \alpha^-$ can be seen. The case of a linearly polarized pump which would create spin-unpolarized carriers is shown (thin solid curve) for reference. Experimental data at the crossover energy (as in (a)) is not available. Also shown is the impossibility of obtaining a crossover by using a theory based on the original partial PSF, because of the consequent dominance of BGR over the entire spectral range. This was verified also at $n = 1.3 \times 10^{17}$ cm^{-3}, which was the estimated experimental density for the data shown.

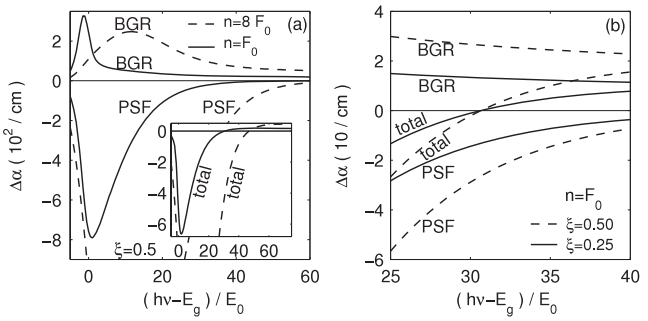

Figure 4. Decomposition into PSF and BGR of total $\Delta\alpha$ for different (a) plasma densities, $F_0 = 1.3 \times 10^{17}$ cm^{-3} and $8F_0$ and (b) spin polarizations, $\xi = 0.5$ and 0.25, in the presence of Coulomb interaction according to (6). The screening of the CE term, $|\Phi_\nu(\boldsymbol{r} = 0)|^2$, identically multiplies the individual PSF and BGR components. According to the pair-equation model, the interplay between these components fully determines the spectral crossover in the corresponding total $\Delta\alpha$ which is indicated in the inset of (a) and in (b). The calculated density- and ξ-dependent trends in total $\Delta\alpha$, which were also observed in experiment [1], can be explained on the basis of the decomposition shown (see text).

the majority $\downarrow$ spin band causes a photon to excite optical transitions into this band at larger quasimomenta, causing α^+ to be greater than α^-. It turns out that the effect of the full PSF dominates over that of BGR at lower energies. However, the exponential fall of the effect of PSF at higher energies lying in the tail of the Fermi function causes the slowly falling change in absorption due to BGR to dominate at higher energies. The influence of the valence band structure from the light holes on our results and a suitable comparison with the microscopic theory [2], shown in figure 3(a), indicate that the two-band pair equation (cf (6)) indeed suffices to satisfactorily fit the experimental data. However, we found that, if the partial PSF $A_0^s(\omega, n, p)$ was used instead of the full $A^s(\omega, n, p)$, BGR would have dominated over PSF for all energies and this theory would fail to predict a crossover (figure 3(b)).

Although the spin-unpolarized ON due to screening of the CE does not cause dichroism, it affects the overall shape of the $\Delta\alpha$ spectrum via the $|\Phi_\nu(\boldsymbol{r} = 0)|^2$ term in (6). Screening has a strong influence on $\Delta\alpha$ (especially in the microscopic description) in the spectral region near the bandgap E_g, where the dip (figure 3(a)) is due to the ionization of the exciton by the plasma. This excitonic feature appears broadened mainly due to the non-negligible probe spectral width considered. The over-screening of the CE by the static Hulthén potential [4] used in the pair equation is not readily apparent in figures 2 and 3(a). However, by plotting the change in absorption from its unpumped value α_0, the effect of over-screening is clearly brought out in figure 3(b), especially at higher energies. This discrepancy with experiment can only be resolved by using dynamic screening, which cannot be incorporated in a simplified model of plasma effects on absorption.

The crossover in total $\Delta\alpha$ calculated by the pair-equation model (cf (6)) shifts to higher photon energies as the plasma density is increased (inset of figure 4(a)), whereas it is independent of ξ (figure 4(b)). Although these trends which were experimentally observed [1] were also obtained by the microscopic theory [2], a detailed understanding is lacking so far. This is because the screening of the CE itself depends on the other ON of PSF and BGR in the microscopic theory which makes it difficult to identify their contributions to total $\Delta\alpha$. However, their separation in (6) allows for an analytical understanding of the crossover energy's density- and ξ-dependent trends. Since the opposition between PSF and BGR results in the crossover according to (6), we consider only these mechanisms of ON below. It can be analytically shown

that, in the crossover region ($h\nu \gg E_g$), the negative $\Delta\alpha$ from PSF has a superlinear density dependence that dominates over the positive $\Delta\alpha$ from BGR whose density dependence is sublinear. This causes the blueshift of the crossover energy as the density is increased. On the other hand, a simultaneous linear dependence of the PSF component (for a non-degenerate plasma when $h\nu \gg E_g$) on ξ and also of the BGR component on ξ results in the crossover energy being ξ-independent. This analysis is supported by calculations based on (6) which suitably resolve total $\Delta\alpha$ into PSF and BGR components for different densities (figure 4(a)) and ξ (figure 4(b)).

4. Conclusion

In conclusion, the simplified theory developed here makes it possible to calculate optical nonlinearities and their effect on the absorption of circularly polarized light in a carrier spin-polarized semiconductor, without resorting to time-consuming numerical methods. The use of this theory was validated by the fresh insights it gave, in the presence of the Coulomb interaction, into the experimentally observed density- and spin-dependent trends of the crossover energy which were not obtainable from the microscopic model. Besides, our modification of the PSF term of the pair-equation model is useful in describing not only spin-polarized but also spin-unpolarized plasma-induced changes in absorption, especially when the nonlinearity due to PSF is dominant.

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