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Franz-Keldysh effect on Landau levels and magnetoexcitons in quantum wells

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We consider the optical properties associated with the photoexcitation and radiative recombination of electron-hole pairs and excitons in intrinsic quantum wells in an unusual high-field regime, namely, in the presence of a strong dc electric field in the quantum-well plane and a crossed static magnetic field aligned with the growth direction. We find Landau-level-like oscillatory structure in the optical density well below the lowest Landau level or magnetoexciton when the electric field is present.

The Franz-Keldysh effect (FKE) describes the modification to the optical density (OD) in an intrinsic semiconductor in the presence of a dc electric field **F**.[1] With **F** applied, a nonvanishing OD below the band gap develops as well as oscillations in the OD above the gap. The theory for bulk and quantum wells (QW's) is outlined in Ref. 2. In the present study we consider the FKE first on free electron-hole (e-h) pairs and then on excitons in the presence of a magnetic field **B**. We first consider free e-h pairs in a QW for which the FKE is known to be especially dramatic, taking $\mathbf{F}=F\hat{\mathbf{x}}$ and $\mathbf{B}=B\hat{\mathbf{z}}$ with $\hat{\mathbf{x}}$ and $\hat{\mathbf{z}}$ unit vectors lying in the QW plane and aligned with the growth direction. Finally, we include the e-h Coulomb interaction V, which is responsible for excitonic effects, in an exactly solvable model in the high-field limit. We emphasize that **F**, **B**, and V are included nonperturbatively (though approximations are made to arrive at an exactly solvable model). Although space limitations preclude further discussion, our Green-function approach makes explicit the relationship between the dynamics of e-h pairs in *strong* fields and the spectroscopic properties—an area extensively explored in atomic physics.[3] Moreover, our treatment accounts self-consistently for propagation effects of the optical field. (For the single QW such effects are insignificant; however, the method points the way to how to treat other cases, such as Bragg structures where effects associated with radiative coupling may dominate.) We are thus able theoretically to access an unusual and, to the best of our knowledge, until now unexplored high-field regime.

Magnetospectroscopy of QW's is certainly not new, and well developed theoretical techniques to calculate their electronic structure and optical properties have been published.[4] *Weak* in-plane **F** fields have been used to couple (zero-field) angular-momentum excitonic eigenstates leading to the appearance in optical spectra of features associated with transitions from the crystal ground state that are dipole forbidden at zero field in a perturbation-theory picture.[5] Also in this geometry, optical spectra in the presence of ac in-plane fields have recently been studied.[6,7] Optical spectroscopy of QW's with **F** and **B** in the $\hat{\mathbf{z}}$ direction[8] and with $\mathbf{B}\|\hat{\mathbf{x}}$ and $\mathbf{F}\|\hat{\mathbf{z}}$ (Ref. 9) have to some extent been investigated. A very few works[10] deal with the geometry considered here. Reference 10 was spectroscopic in nature and involved the expansion of the exciton envelope functions in a set of hydrogenic radial wave functions. Continuum states, however, were not included; their inclusion is necessary in the limit of large **F** fields as well as to explore parameter space. The Green-function approach permits us to treat the fields **F** and **B** nonperturbatively (though we do make simplifications—see below) and then to include excitonic effects self-consistently in a simple but sensible model. Our treatment thus fully includes exciton ionization within the model. The approach is extremely convenient since nowhere need we refer explicitly to Landau levels or to any other basis. Among our results, we find clear Landau-level-like structure in the OD well below the lowest Landau level in the presence of **F**. With V included we reproduce magnetoexciton spectra in the absence of **F**,[11,12] while with **F** applied unique spectral line shapes are predicted. We can thus map out the crossover of the spectroscopic properties from magnetoexcitons ($\mathbf{F}=\mathbf{0}$) to the FKE ($\mathbf{B}=\mathbf{0}$) in this fundamental regime of semiconductors in the presence of strong fields.

Let a weak optical field [plane wave, amplitude $I(t)$] be incident normally on the QW in the vicinity of an excitonic resonance. The reflected $R(t)$ and transmitted $T(t)$ optical fields are self-consistently determined by[13]

$$T(t)=I(t)+\int_{-\infty}^{t}dt'\,\chi(t-t')T(t') \tag{1}$$

and $R=T-I$, where χ is the optical susceptibility. (We have assumed an optically thin QW.) Thus, the spectral response is given by $T(\omega)=[1-\chi(\omega)]^{-1}$ and $R(\omega)=\chi(\omega)T(\omega)$, where $f(\omega)=\int_{-\infty}^{\infty}dt\,e^{i\omega t}f(t)$. In addition to T and R, it is useful to compute the OD, $w(\omega)=-2\,\mathrm{Re}\,\chi(\omega)$ and the radiatively renormalized OD, $w_r(\omega)=1-|R(\omega)|^2-|T(\omega)|^2=w(\omega)|1-\chi(\omega)|^{-2}$. This definition of absorption, as that part of the intensity neither reflected not transmitted, is sensible since the QW is optically thin.

The optical susceptibility χ is simply related to the propagator K and retarded Green function D for free e-h pairs in the combined presence of **F** and **B**. (We include the e-h Coulomb potential later.) For $\mathbf{B}=\mathbf{0}$, the response to **F** is given by that of a free particle of reduced mass μ. For $\mathbf{B}\neq\mathbf{0}$, however, the electron and hole response must be treated independently since the cyclotron frequencies for the two types of particles differ due to the lighter electron effective mass m. The full expression written as a Lehmann expansion accounting for both electron and hole cyclotron motion in bulk (with $\mathbf{F}=\mathbf{0}$) is given in Eq. (7) of Ref. 12. Although such an exact expression for the e-h propagator for the QW (even with $\mathbf{F}\neq\mathbf{0}$) can be easily written down, we would be hard put subsequently to integrate it due to its complexity. Instead, we make use of the fact that typically, the hole mass considerably exceeds that of the electron. We therefore treat the hole as fixed. This is *prima facie* a good approximation if the hole mass is much greater than the electron mass or if the dephasing rate exceeds the hole cyclotron frequency. Be this as it may, we shall assume the hole is fixed in the following; our results shall justify this assumption. Although we will not be able to reproduce the details of the spectra (associated with hole cyclotron motion, band mixing, nonparabolicity, etc.), the treatment brings to the fore other aspects, such as those associated with light propagation and exciton ionization. Moreover, an extensive exploration of parameter space is possible with our model, which will facilitate the identification of interesting effects that have hitherto gone unidentified.

If $I(t)$ is circularly polarized, it couples to e-h pairs of definite spin σ. This will pick out spectral features associated with an individual spin state; we therefore assume in the following that the incident optical field $I(t)$ is circularly polarized. We then have ($\hbar=1$) $\chi(t)=2\pi i A m^{-1}D(0,0;t)$,

$$D(\mathbf{r},\mathbf{r}';t)=-i[K(\mathbf{r},\mathbf{r}';t)+\text{c.c.}]\theta(t), \tag{2a}$$

$$K(\mathbf{0},\mathbf{0};t)=\frac{m\eta}{2\pi i\sin\eta t}e^{-i(\varepsilon'_{\pm}+\omega_0)t}e^{i\omega_0\eta t^2\cot\eta t} \tag{2b}$$

to the extent that we can treat the QW as a two-dimensional (2D) system. Here $A=(mg|S|^2d^2\alpha\varepsilon_0/\sqrt{\epsilon_b})$ ($\approx10^{-4}$ for GaAs QW's), ed is the interband dipole matrix element, g is a spin-orbit factor ($=\frac{1}{2}$ for e-h pairs involving the heavy-hole band, $\frac{1}{6}$ for the light-hole band), S (≈1) is the overlap integral of the electron and hole single-particle envelope functions, $\alpha=e^2/\hbar c$ is the fine-structure constant, ε_0 is the frequency of the edge of the 2D e-h continuum density of states, $\varepsilon_\pm=\varepsilon_0\mp\mu_0 B$, $\varepsilon'_\pm=\varepsilon_\pm-i\gamma$, $\omega_0=(e^2F^2/2m\omega_c^2)$, $\eta=\omega_c/2$, $\pm$ denotes the helicity of the light (spin), γ is an energy- and state-independent nonradiative width, μ_0 is the Bohr magneton, $\omega_c=eB/m$ is the electron cyclotron frequency, e is the electron charge, and ϵ_b is the background dielectric constant at optical frequencies; $\mathbf{r}$ and $\mathbf{r}'$ are e-h relative coordinates. $K(\mathbf{r},\mathbf{r}';t)$ is obtained straightforwardly by the techniques of Ref. 14.

We proceed to calculate the cw optical properties of the QW in the crossed fields $\mathbf{F}$ and $\mathbf{B}$. We have, as a function of normalized frequency $\overline{\omega}=\omega/\eta$,

$$\chi(\overline{\omega})=2A\int_0^\infty dx\,\csc x\,e^{i(\overline{\omega}+i\overline{\gamma})x}\sin[(\overline{\varepsilon}_\pm+\overline{\omega}_0)x-\overline{\omega}_0x^2\cot x], \tag{3}$$

where the overbar denotes division by η.[15] This expression is deceptively simple; great care must be exercised in its numerical evaluation. We can see from a purely analytic viewpoint how various limiting behaviors result from this expression. First, in the zero-$\mathbf{F}$ (zero-$\overline{\omega}_0$) limit, i.e., the limit of pure Landau levels, χ can be expressed simply in terms of the Euler-ψ (digamma) function, giving rise to an infinite set of equispaced features bounded below. In the parametrization of Eq. (3), the effect of $\overline{\gamma}$ is to modulate the impact of $\mathbf{B}$; $\overline{\gamma}\gtrsim1$ has the effect of suppressing the Landau levels while leaving features associated with the FKE. This is because for large $\overline{\gamma}$, we can make the replacements $\csc x\rightarrow1/x$ and $\cot x\rightarrow(1/x)-(x/3)$ in the integral leading to the pure FKE. In a regime where $\overline{\gamma}$ is not large, both Landau levels and the FKE appear. In the presence of $\mathbf{F}$, the cotangent term generates destructive interference at the poles of the cosecant leading to a spreading of the Landau levels with increasing $\mathbf{F}$. These features are all clearly seen below in our numerical results.

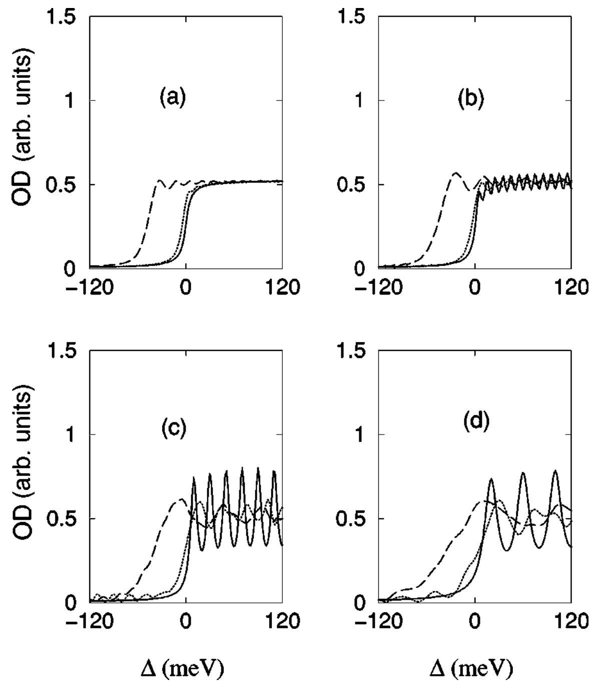

FIG. 1. OD $w(\overline{\omega})$ as a function of detuning $\Delta=\omega-\varepsilon_\pm$ from the paramagnetically shifted band edge. The parameters are (a) $\eta=2$ meV ($B\approx1$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); (b) $\eta=5$ meV ($B\approx3$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); (c) $\eta=10$ meV ($B\approx6$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); and (d) $\eta=20$ meV ($B\approx12$ T), $\gamma^{-1}=400$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed).

In Fig. 1 is shown the OD $w(\omega)$ as a function of detuning $\Delta=\omega-\varepsilon_\pm$ from the paramagnetically shifted band edge. For realistic values of γ, $w(\omega)\approx w_r(\omega)$ for a single QW, i.e., propagation effects are not so important. The material parameters have been chosen to be typical of $GaAs/Al_xGa_{1-x}As$ QW's at cryogenic temperatures and low optical excitation density. The fields are chosen such that in Fig. 1(a) $\eta=2$ meV ($B\approx1$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); in Fig. 1(b) $\eta=5$ meV ($B\approx3$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); in Fig. 1(c) $\eta=10$ meV ($B\approx3$ T), $\gamma^{-1}=800$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed); and in Fig. 1(d) $\eta=20$ meV ($B\approx12$ T), $\gamma^{-1}=400$ fs, $\omega_0=0$ (solid), η (dotted), 10η (dashed). Thus, Fig. 1(a) corresponds to the pure FKE (the solid curve is essentially the zero-field optical density, i.e., the 2D OD at the band edge softened by the dephasing). As ω_0 is increased, the band edge shifts to the red, softens further, and oscillations above the band edge appear; these effects are well known.[2] For Figs. 1(b)–1(d) the magnetic field is of increasing magnitude to show the interplay between the FKE and Landau levels. The solid curves show the Landau levels for insignificantly small values of **F**. For the larger values of ω_0 the internal motion of an e-h pair strictly speaking knows no ground state. This leads to a nonvanishing subgap OD. Because there is literally no band edge, the Landau levels also have no lowest state, thus leading to the oscillatory Landau-like structure below the band edge, as is evident in the dotted curves of Figs. 1(c) and 1(d). This is the crossover regime where **F** and **B** are comparable. Note that a recent semiclassical (WKB) treatment of the density of states of a free electron in combined **F** and **B** was inadequate to reveal the rich spectral structure we find from a purely quantum-mechanical standpoint.[16] For strong **F** and **B** fields [dashed curves of Figs. 1(c) and 1(d)], the lowest large Landau-FK resonance shows a substantial broadening with respect to either the pure Landau level or the oscillatory structure in the FKE. This reflects the fact that in the combined fields, e-h pairs initially at zero separation are stripped apart even faster than in the individual fields. In this regime, the line shapes, however, are highly complex, as a consequence of the coherent dynamics of the e-h pair in the **F** and **B** fields. Above the band edge, there is an interplay between the oscillatory structure associated with Landau levels and that associated with the FKE.

Thus far, the e-h Coulomb interaction $V(r)$, which for the zero-field case leads to excitons and the Sommerfeld factor, has been excluded from consideration; **F** and **B**, however, have been accounted for nonperturbatively. The retarded Green function D_v in the presence of V satisfies the Dyson equation $D_v(\mathbf{r},\mathbf{r}';\overline{\omega})=D(\mathbf{r},\mathbf{r}';\overline{\omega})+\int d^2r''\,D(\mathbf{r},\mathbf{r}'';\overline{\omega})V(r'')D_v(\mathbf{r}'',\mathbf{r}';\overline{\omega})$. Perturbation theory in V therefore corresponds to iterating to a given order. Instead of this route, we consider a model which can be solved to all orders which is expected to be valid at high fields **F** or **B** (i.e., when the relevant classical paths far exceed the exciton Bohr radius). We take $V(r)=v\,\delta^2(\mathbf{r})$ sufficiently near the band edge where v $(v<0)$ is a fitting parameter. (Working in real space obviates difficulties that can occur with the contact potential in k space.) Then, since the optical properties only depend on D_v at $\mathbf{r}=\mathbf{r}'=0$, we have for the susceptibility in the presence of V

$$\chi_v(\overline{\omega})=\frac{\chi(\overline{\omega})}{1+iv(2\pi A)^{-1}\chi(\overline{\omega})}, \qquad (4)$$

and $R(\overline{\omega})$, $T(\overline{\omega})$, $w(\overline{\omega})$, and $w_r(\overline{\omega})$ are given by the above expressions with χ replaced by χ_v. Although this model may lead in some regimes (cf. Ref. 17) to artificial line shapes, we have verified (see below) that we obtain reasonable line shapes near the band edge for limiting cases of **F** and **B** where a comparison with more detailed calculations is possible.

In Fig. 2 is shown the OD as a function of Δ including the e-h Coulomb interaction with $v/2\pi A=-0.36$. For Fig. 2(a) $\gamma^{-1}=800$ fs, $\omega_0=0$, and $\eta=0$ ($B=0$ T, solid), 5 meV ($B\approx 3$ T, dashed), 10 meV ($B\approx 6$ T, dotted), and 20 meV ($B\approx 12$ T, chain). Note that our model for V appears to be valid semiquantitatively down to zero fields in the presence of dephasing [Fig. 1(a), solid curve]; the contact potential supports a single bound state. In practice, dephasing merges higher-lying bound states with the continuum edge. Note that the contact potential leads to a slightly different relationship between the excitonic oscillator strength and excitonic binding energy compared with the true expression. Nevertheless, the model zero-field spectrum near the band edge is reasonable. For $\mathbf{B}\neq\mathbf{0}$, the features of the magnetoexciton spectra well reproduce those calculated in Ref. 11 and elsewhere. In fact, we expect our model to improve with increasing field strength as exciton ionization becomes more marked and the lifetime becomes shorter. Figures 2(b)–2(d) make use of the same parameters as those employed in Figs. 1(b)–1(d); in the present case, however, V is included. For substantial values of ω_0, the magnetoexcitons broaden, but retain signatures associated with the magnetic field, i.e., Landau-level-like features. In particular, for large **F** and **B** there is even stronger broadening, as well as a redshift of the major band-edge feature. The redshift is determined primarily by **F** (see Fig. 1). We see that [cf. Figs. 2(c) and 2(d)] subgap Landau-level-like structure is retained even including V. We would like to remark that the spectral line shapes fully incorporate the effects of exciton ionization within the model. In particular, ionization does not simply result in a Lorentzian broadening of the exciton or magnetoexciton lines, but in highly asymmetric line shapes possessing a significant degree of structure. Also note that we have included an interband dephasing rate (albeit energy- and state-independent dephasing) which is appropriate for very high quality QW's at low temperature and excitation density.

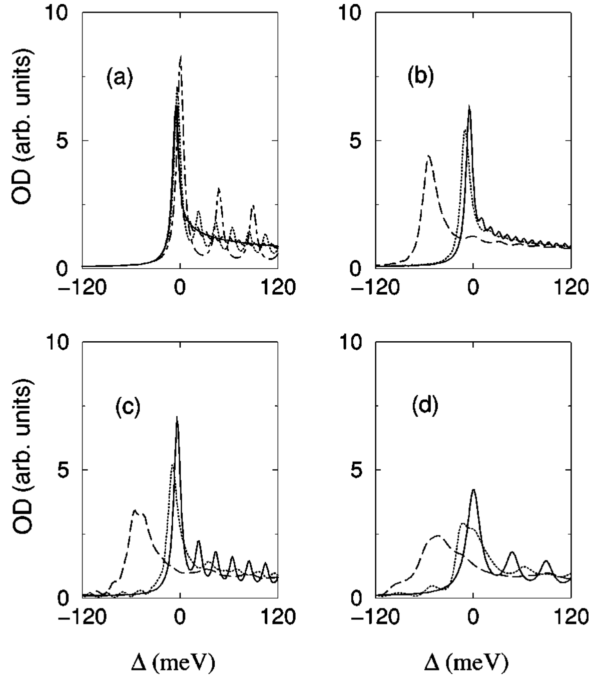

FIG. 2. OD as a function of Δ including the e-h Coulomb interaction $V(r)=v\,\delta^2(\mathbf{r})$ with $v/2\pi A=-0.36$. For (a) $\gamma^{-1}=800$ fs, $\omega_0=0$, and $\eta=0$ ($B=0$ T, solid), 5 meV ($B\approx 3$ T, dashed), 10 meV ($B\approx 6$ T, dotted), and 20 meV ($B\approx 12$ T, chain). (b)–(d) use the same parameters as those in Figs. 1(b)–1(d); in the present case, however, V is included.

To conclude, we have calculated the Franz-Keldysh effect on Landau levels and magnetoexcitons in QW's accounting for **F** and **B** nonperturbatively. A model *e-h* Coulomb interaction *V* was included self-consistently. This model is expected to be correct in the limit of large fields, but also gives apparently sensible results in the low-field limit as well. Thus, the model presented here is useful to carry out an exploration of parameter space at minimum computational cost, something for which the band-structure approaches hitherto implemented are ill suited. We find Landau-level-like structure below the paramagnetically shifted band gap or lowest magnetoexciton in the presence of **F**. With *V* present, we predict unusual spectral line shapes—strong asymmetry and nontrivial structure—of magnetoexcitons which incorporate the effects of exciton ionization. The study of the physics of excitons in **F** and **B** fields provides additional flexibility and may shed light on related atomic and molecular problems.[3]

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