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Interband optical spectra of magnetoexcitons in semiconductor nanorings: Electron-hole spatial correlation

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An analytic model [R. A. Römer and M. E. Raikh, Phys. Rev. B **62**, 7045 (2000); K. Moulopoulos and M. Constantinou, *ibid.* **70**, 235327 (2004)] for magnetoexcitons in nanoscale semiconductor rings is extended to calculate directly the linear optical properties. The spectroscopic properties exhibit pronounced $\Phi_0 = hc/e$ excitonic Aharonov-Bohm oscillations in the threading magnetic flux Φ when the ring radius R is less than the effective exciton Bohr radius a_0. The electron-hole spatial correlation induced by an optical field as a function of nanoring radius and threading magnetic flux is studied.

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The excitonic (x) (Ref. 1) Aharonov-Bohm (AB) effect[2] in the optical properties of semiconductor nanorings (NR) has attracted intense recent interest.[3–14] The x-AB effect is manifested in principle in a periodic modulation of the NR optical properties with period $\Phi_0 = hc/e$ in the magnetic flux Φ threading the NR.[1] In practice, however, these oscillations have been found to be weak for the dominant spectroscopic feature, which is associated with the lowest-lying exciton. For NR radii R larger than the excitonic Bohr radius a_0, the resulting x-AB effect is weak due to the fact that the exciton as a composite object orbits around the NR, and since the exciton is charge neutral, the x-AB effect is negligible. Nonetheless, schemes to circumvent this limitation have been proposed. Such approaches typically either polarize the exciton in the NR plane so that the electron and hole at least partially sample the AB phase around the NR independently[9] or polarize the exciton in the vertical direction, and then exploit the differences in the effective radii of the electron and hole wave functions in typical NR structures.[11]

With advances in nanoscale e-beam lithography and in self-organized growth, however, it is likely that NR dimensions with $R \sim a_0$ or even $R < a_0$ will soon be fabricated; alternatively, exploiting smaller band-gap materials using structures with R similar to what has been demonstrated can potentially access the same regime. Another possibility involves magnetoexcitons in carbon nanotubes, which may exhibit an x-AB effect.[15] In the following, we extend an exactly solvable model[6,7] to calculate in a direct fashion the linear optical properties of magnetoexcitons in NR's assuming a point-contact potential for the electron-hole Coulomb interaction. The model permits easy exploration of the crossover region $R \sim a_0$. We find that the excitonic optical properties of NR's can show pronounced x-AB oscillations provided $R < a_0$. Moreover, one can directly obtain the spatial electron-hole correlation associated with magnetoexcitons in NR's as a function of photon energy, threading magnetic flux, and NR parameters.

The basic theory is discussed in Ref. 7; however, for the sake of making the present study self-contained, we include the outlines of a derivation here. Our aim is to obtain the Green function (GF) for the electron-hole Schrödinger equation; as we show, this is related to the solution of the polarization equation of the semiconductor Bloch equations[16] (SBE) in the low-density limit, which determines the linear optical properties. We begin with the time-independent Schrödinger equation for an electron and a hole in a NR. The full Hamiltonian for the electron and hole is denoted H_T, which includes terms for the electron and hole kinetic energy, and the three-dimensional confinement potential. H_T depends on the three-dimensional electron and hole coordinates $\mathbf{r}_e$ and $\mathbf{r}_h$. We shall not need the explicit form of H_T. We assume that the subband motion associated with the radial direction (ρ_e and ρ_h in cylindrical coordinates) and the direction normal to the NR plane (z_e and z_h) is frozen out, and can be described by the electron and hole single-particle envelope functions $f_e(\rho_e, z_e)$ and $f_h(\rho_h, z_h)$, respectively.

Once the ρ_e, ρ_h, z_e, z_h dependence is integrated out, the only explicit degrees of freedom are the axial angles, φ_e and φ_h, respectively, associated with the electron and hole positions around the NR. Having integrated out the ρ and z motion, we can thus write the exciton envelope function as $\psi(\varphi_e, \varphi_h)$, which satisfies $H\psi = E\psi$ with

$$H = \frac{\hbar^2}{2m_e R^2}\left(-i\frac{\partial}{\partial\varphi_e} + f\right)^2 + \frac{\hbar^2}{2m_h R^2}\left(-i\frac{\partial}{\partial\varphi_h} - f\right)^2 + V_C(\varphi_e - \varphi_h), \tag{1}$$

where $m_e(m_h)$ is the electron (hole) effective mass, $f = \Phi/\Phi_0$, and $V_C(\varphi_e - \varphi_h)$ is the electron-hole Coulomb interaction where the carriers are constrained to lie on the NR. Note that $V_C(\varphi_e - \varphi_h)$ in fact depends on $f_e(\rho_e, z_e)$ and $f_h(\rho_h, z_h)$, but below we choose a model potential that obviates the need for an explicit expression. The Hamiltonian H explicitly contains only φ_e and φ_h dependence; the quantization energies associated with $f_e(\rho_e, z_e)$ and $f_h(\rho_h, z_h)$ are constant insofar as the subsequent discussion is concerned, and play no role in the ensuing development; they will therefore be dropped.

Previously,[9] we have used the very good approximation[17,18] $V_C(\varphi_e - \varphi_h) = -(e^2/\epsilon_b)(2R\sin(|\varphi_e - \varphi_h|/2) + a)^{-1}$ with ϵ_b the background static dielectric constant and a a constant proportional to R to account for the ρ- and z-oriented widths of the single-particle envelope functions. In the following, we approximate V_C by the point-contact potential, $V_C(\varphi_e - \varphi_h) = v\,\delta(\varphi_e - \varphi_h)$ $(v < 0)$, where the angular arguments are understood to be modulo 2π and $v=v'/(2\pi R)$. The fitting parameter v' is fixed for a given material and radial width of the NR channel, and is chosen to ensure that the energy of the lowest-lying exciton with respect to the effective band gap (i.e., including subband quantization) agrees with the potential employed in Ref. 9. The reason for this procedure is that we assume that the dominant contribution to the exciton binding energy comes from the subband quantization, which is not explicitly treated; thus, the fitting ensures that the exciton binding energy is not a strong function of R.

We next exploit the NR rotational symmetry to simplify the problem vastly. To this end, we make the substitutions $\Theta=(m_e\varphi_e+m_h\varphi_h)/(m_e+m_h)$ is the center-of-mass coordinate and $\varphi=\varphi_e-\varphi_h$ in Eq. (1) where φ is the relative coordinate. At this stage, H can be written as a sum of terms that depend on Θ and φ independently; thus, the Hamiltonian of Eq. (1) is separable, and the eigenstates can be expressed as products of $(2\pi)^{-1/2}\exp(i\nu\Theta)$ with ν and integer and of a function of φ. The factor $(2\pi)^{-1/2}\exp(i\nu\Theta)$ describes the center-of-mass variation of the wave function's phase around the NR. Again, the center-of-mass energy does not depend on φ, and thus it can be disregarded in computing the relative motion. In fact, we show below that only for $\nu=0$ do the states contribute to the optical properties (momentum conservation in interband transitions).

We have for the relative-motion Hamiltonian

$$\tilde{H} = (\hbar^2/2\mu R^2)(-i\partial/\partial\varphi + f)^2 + V_{\mathrm{C}}(\varphi), \tag{2}$$

where $\mu=m_e m_h/(m_e+m_h)$ is the electron-hole reduced mass. At this stage, we can assemble the eigenfunctions of H_T. They are of the form $\Psi(\mathbf{r_e},\mathbf{r}_h)=R^{-1}f_e(\rho_e,z_e)f_h(\rho_h,z_h)\eta(\varphi)\times(2\pi)^{-1/2}\exp(i\nu\Theta)$, where the factor R^{-1} ensures that $f_e(\rho_e,z_e)f_h(\rho_h,z_h)$ is normalized over $\rho_e d\rho_e \rho_h d\rho_h dz_e dz_h$ and the exciton envelope function $\eta(\varphi)$ is assumed normalized over $d\varphi$.

The eigenfunctions $\eta(\varphi)$ of the Hamiltonian $\tilde{H}$ of Eq. (2) can be obtained by substituting an arbitrary linear combination of $\nu=0$ states $(2\pi)^{-1/2}\exp(in\varphi)$ with n an integer into the time-independent Schrödinger equation; alternatively, we can use the method of GF's. Let the Hamiltonian, in the absence of the electron-hole Coulomb potential, be $\tilde{H}_0=\lim_{v\to 0}\tilde{H}$. The retarded GF $G_0(\varphi,\varphi';E)$ associated with $\tilde{H}_0$ satisfies

$$(E+i0^+ - \tilde{H}_0)G_0(\varphi,\varphi';E) = \delta(\varphi-\varphi'). \tag{3}$$

The spectral representation (expansion in eigenfunctions of H_0) of the unperturbed GF $G_0(\varphi,\varphi';E)$ is

$$G_0(\varphi,\varphi';E) = \frac{1}{2\pi}\sum_{n=-\infty}^{\infty}\frac{e^{in(\varphi-\varphi')}}{E+i0^+ - \alpha(n+f)^2}, \tag{4}$$

where $\alpha=\hbar^2/2\mu R^2$. One shows, using standard techniques commonly employed for the single-impurity model of solid-state physics, that for general values of v, the GF G satisfies the Dyson equation

$$G(\varphi,\varphi';E) = G_0(\varphi,\varphi';E) - v\int_0^{2\pi} d\varphi'' G_0(\varphi,\varphi'';E) \times\delta(\varphi'')G(\varphi'',\varphi';E). \tag{5}$$

This is solved to give

$$G(\varphi,\varphi';E) = G_0(\varphi,\varphi';E) - v\frac{G_0(\varphi,0;E)G_0(0,\varphi';E)}{1+vG_0(0,0;E)}. \tag{6}$$

This concludes the derivation of the GF. It remains to relate the GF to optically relevant quantities.

The linear optical properties of the NR are determined by the polarization equation of the SBE in the low-density limit,[19]

$$(E+i0^+ - H_T)\mathcal{Q}(\mathbf{r}_e,\mathbf{r}_h) = -d_{cv}\mathcal{E}\delta^3(\mathbf{r}_e-\mathbf{r}_h); \tag{7}$$

d_{cv} is the interband dipole matrix element between the bulk p-like valence-band and s-like conduction-band Bloch states, $\mathcal{Q}(\mathbf{r}_e,\mathbf{r}_h)$ is the interband polarization per unit volume that will eventually lead to the induced interband dipole moment associated with the optical generation of excitons,[16] and $\mathcal{E}$ is the amplitude of the incident optical field whose corotating term is $\mathcal{E}\exp(-iEt/\hbar)$. As above, the ρ- and z-oriented motion of the electron and hole in the NR is frozen out in the single-particle envelope functions $f_e(\rho_e,z_e)$ and $f_h(\rho_h,z_h)$. Writing $\delta^3(\mathbf{r}_e-\mathbf{r}_h)=\rho_e^{-1}\delta(\varphi_e-\varphi_h)\delta(\rho_e-\rho_h)\delta(z_e-z_h)$, multiplying Eq. (7) on the left by $f_e^*(\rho_e,z_e)f_h^*(\rho_h,z_h)$, integrating over ρ_e, ρ_h, z_e, and z_h, and dropping the energy associated with subband quantization, we have

$$(E+i0^+ - H)\mathcal{P}(\varphi_e,\varphi_h) = -d_{cv}U^*\mathcal{E}(1/2\pi R)\delta(\varphi_e-\varphi_h), \tag{8}$$

where $U=\int d\rho dz\, f_e(\rho,z)f_h(\rho,z)$ is the overlap integral of the electron and hole single-particle envelope functions. $\mathcal{P}(\varphi_e,\varphi_h)$ is now the interband polarization per unit length around the NR circumference. Finally, the interband polarization $P(0)$ of the NR (assuming the R is much less than the wavelength of photons near resonance with the interband optical transition) is obtained by integrating Eq. (8) over the center-of-mass coordinate around the NR

$$P(\varphi) = R\int_{-\pi}^{\pi} d\Theta\, \mathcal{P}(\varphi_e,\varphi_h). \tag{9}$$

This picks out the $\nu=0$ center-of-mass eigenstates of H, and is equivalent to the statement that the incident photon carries negligible momentum, and thus the center-of-mass momentum of the exciton must also be zero. One can also view this selection rule as follows:[6] The factor $(2\pi)^{-1/2}\exp(i\nu\Theta)$ describes the center-of-mass variation of the wave function's phase around the NR. Noting that, inasmuch as R is much less than the relevant optical wavelength in the vicinity of the interband transitions of interest, if this phase variation corresponds to $\nu=0$, then the interband polarization around the NR adds constructively, and thus the polarization for the transition does not sum to zero. If, instead, $\nu\neq 0$, the coherent sum of the interband polarization around the NR vanishes.

From Eq. (8), we therefore have,

$$(E+i0^+ - \tilde{H})P(\varphi) = -d_{cv}U^*\mathcal{E}\delta(\varphi). \tag{10}$$

The induced interband dipole moment of the NR is $d_{cv}UP(0)$. A comparison of Eqs. (3) and (10) shows that the interband polarization is given by $P(\varphi)=-d_{cv}\mathcal{E}U^*G(\varphi,0;E)$ and thus the dipole moment associated with the recombination of the optically generated excitons is

$-d_{cv}^2\mathcal{E}|U|^2G(0,0;E)$. Consequently, the solutions of $G^{-1}(0,0;E)=0$ give the dipole-allowed optical resonances (eigenstates) of the system.

Thus, $G(\varphi,0;E)$ and specifically $G(0,0;E)$ play important parts in the theory. Based on the foregoing, we have for $G(0,0;E)$

$$G(0,0;E)=[G_0(0,0;E)]/[1+vG_0(0,0;E)]. \tag{11}$$

Finally, performing the summation in Eq. (4) for $\varphi=\varphi'=0$ gives the explicit expression

$$G_0(0,0;E)=\frac{1}{4\alpha\beta^{1/2}}\{\cot[\pi(f+\beta^{1/2})]-\cot[\pi(f-\beta^{1/2})]\}, \tag{12}$$

where $\beta=E/\alpha$. Expressions for other arguments are given in Ref. 7.

In the following, we plot $-\mathrm{Im}G(0,0;E+i\gamma)$, which is proportional to the optical density,[19] where $\gamma>0$ is a phenomenological broadening. We employ parameters appropriate to GaAs-based structures; viz. $m_e=0.07m_0$ and $m_h=0.4m_0$, with m_0 the free-electron mass. We further take $\gamma=0.2$ meV and $v=-9425$ meV Å. Figure 1 shows $-\mathrm{Im}\,G(0,0;E+i\gamma)$ for $R=60$ Å and $0<f<0.5$. The solid curve in Fig. 1(a) is for $f=0$; the dashed curve is for $f=0.5$. A pronounced dependence of the lowest-lying excitonic feature on f is seen. Excited dipole-allowed states, which lie at positive energies, exhibit much stronger f dependences, since for these states V_C plays a much smaller role. The lowest-lying exciton becomes blueshifted and grows in strength with f at the expense of the resonance at ~ 10 meV at $f=0$.[6] In Fig. 2 are plotted the results for $R=200$ Å. Note that we have chosen the strength of the effective Coulomb interaction to ensure that the binding energy of the lowest-lying exciton does not depend strongly on R for $f=0$. The solid curve in Fig. 2(a) is for $f=0$; the dashed curve is for $f=0.5$. For $R=200$ Å, the f dependence of the lowest-lying exciton is entirely negligible. Higher peaks shift [difficult to see in Figs. 2(a) and 2(b), but evident in Fig. 2(c)] as the internal electron-hole motion of the excited states is more distributed around the NR. There is, however, some redistribution of oscillator strength with f amongst the states for $E>0$, since these states, being less bound than the lowest-lying exciton, are more sensitive to changes in f. Also note in Fig. 2(a) that the excited-state optical densities are suppressed somewhat for those states closest to the ground state—an effect closely related to the Sommerfeld factor in quasi-one-dimensional systems.[18] The growth of the lowest-lying peak at the expense of the first excited state as f increases from 0 to 0.5, over which range the energy splitting of these states decreases, is a manifestation of the quasi-one-dimensional Sommerfeld factor. The assumption of a δ-function potential is thus adequate to produce a range of effects related to the Sommerfeld factor.

From the spectral representation, it is clear that $|G(\varphi,\varphi;E)|$ at equal angular arguments is proportional to the probability density for eigenstates of the Hamiltonian of Eq. (2) for E sufficiently near the respective eigenenergies. Another useful and physically significant quantity is $|G(\varphi,0;E)|^2$ [which coincides with the wave function in Eq. (10) of Ref. 6; their Δ_{N_0} is a solution of $G^{-1}(0,0;\Delta_{N_0})=0$],

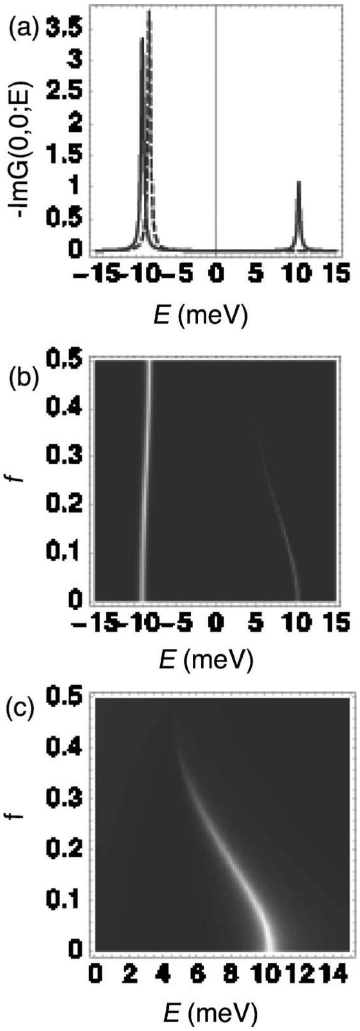

FIG. 1. $-\mathrm{Im}\,G(0,0;E)$ for a $R=60$ Å GaAs NR. This quantity is proportional to the optical density (absorption). (a) In this frame, $f=0$ (solid curve), $f=0.5$ (dashed curve). (b) This frame shows a density plot of $-\mathrm{Im}\,G(0,0;E)$ as a function of E and f. Relative feature heights are as given in frame (a). (c) As in frame (b), but for $E>0$.

which gives the probability density for relative electron-hole separation φ for optical excitation with photons of energy E. Though the full GF of Eq. (6) is given in Ref. 7, the expression will not be included here. In Fig. 3 is plotted $\ln|G(\varphi,\varphi;E)|$ [left column; (a), (c), (e)] and $\ln|G(\varphi,0;E)|^2$ [right column; (b), (d), (f)] for a GaAs NR with $f=0$, 0.25, and 0.5 for a NR with $R=60$ Å. Focusing on the left column, as f increases from 0 to 0.5, the lowest-lying exciton becomes more localized about zero electron-hole separation (as evidenced by the more closely spaced contour lines), consistent with the observations above on the growth of the corresponding peak in the optical spectrum with increasing f in this range. This increased binding is concomitant with the blueshift of the exciton line commented upon above. One notes, however, that the degree of binding should be measured not with respect to the $f=0$ NR band gap, but with respect to the band gap for the various values of f. The internal motion of the first excited state has a different behavior. It becomes increasingly localized at $\varphi=\pi$ as f in-

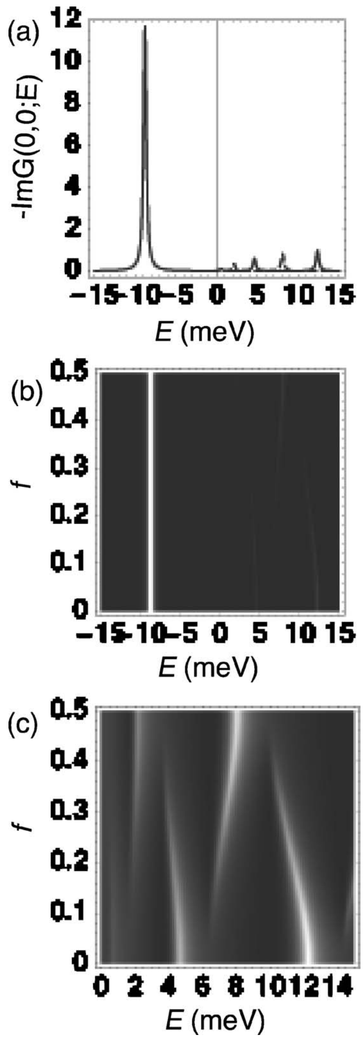

FIG. 2. $-\text{Im}\,G(0,0;E)$ for a $R=200$ Å GaAs NR. (a) In this frame, $f=0$ (solid curve), $f=0.5$ (dashed curve). (b) This frame shows a density plot of $-\text{Im}\,G(0,0;E)$ as a function of E and f. Relative feature heights given in frame (a). (c) As in frame (b), but for $E>0$.

creases in this range of values. As is evident from the right column [as well as from Figs. 1(b) and 1(c)], the excited states play little role in the linear optical properties. Note that the first excited state, evident in Fig. 3(e), is almost entirely absent from Fig. 3(f).

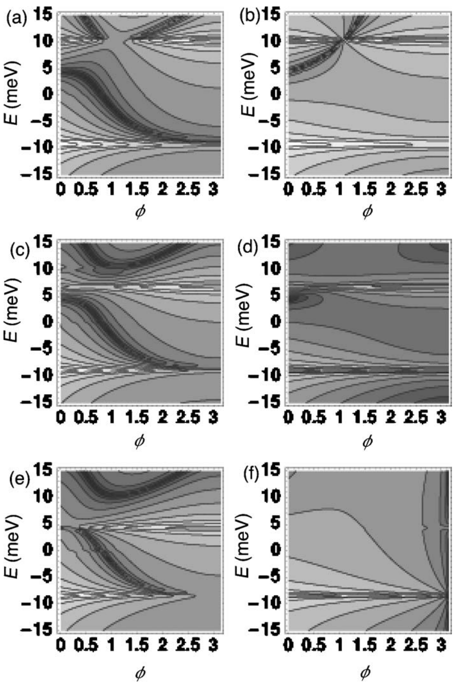

FIG. 3. $\ln|G(\varphi,\varphi;E)|$ [left column; (a), (c), (e)] and $\ln|G(\varphi,0;E)|^2$ [right column; (b), (d), (f)] for a $R=60$ Å GaAs NR for (a) and (b) $f=0$, (c) and (d) 0.25, and (e) and (f) 0.5.

To conclude, we have extended the model of Refs. 6,7 to treat the magneto-optical properties of semiconductor NR's. Specifically, a GF-based approach straightforwardly yields all relevant quantities pertaining to the optical properties. Moreover, the approach enables one to calculate directly the relative electron-hole spatial properties of magnetoexcitons in NR's. It further permits an efficient exploration of parameter space, and offers insight into the physics underlying magneto-optical effects in these structures. Of course, once one has the GF in hand, it is straighforward also to compute various transport properties (such as the current around the NR) as well.

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