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Interband magneto-optical transitions in a layer of semiconductor nano-rings

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Abstract. – We have developed a quantitative theory of the collective electromagnetic response of layers of semiconductor nano-rings. The response can be controlled by means of an applied magnetic field through the optical Aharonov-Bohm effect and is ultimately required for the design of composite materials. We demonstrate that the local field is of paramount importance in the description of such layers. The changes in ellipsometric parameters are 2 to 3 orders of magnitude above the detection limit of a modern ellipsometer.

Modern propositions for composite materials with negative refractive index made from ring elements [1], urge for immediate efforts to create such materials also from layered nano-ring structures for the optical regime. Semiconductor nano-rings are nanosized objects resembling artificial atoms [2]. Topologically, they are characterized by a non simply connected geometry. The consequence of this unique topology is that the center hole enables trapping of magnetic flux quanta. This property of the nano-rings leads to quantum oscillating behavior of the magnetic response of the nano-ring for varying magnetic field, the Aharonov-Bohm (AB) effect [3]. For a simultaneously applied optical beam this gives rise to the optical AB effects [4,5], which can occur only in nano-rings. Varying material properties by means of a magnetic field is an inherent aspect of the AB effects, including the optical one. This option is ultimately required to manipulate these properties, like the making of negative-refractive-index materials [1].

Yet, most of the investigations done already in the field of the optical AB effects in nano-rings has been about far-infrared (FIR) spectroscopy or magneto-photoluminescence (MP) [6–10]. In these methods, an additional stimulus has been used, apart from the electromagnetic beam, to determine the response of the rings. These stimuli can be the creation of an extrinsic carrier population (FIR) or an electromagnetic beam of higher frequency (MP).

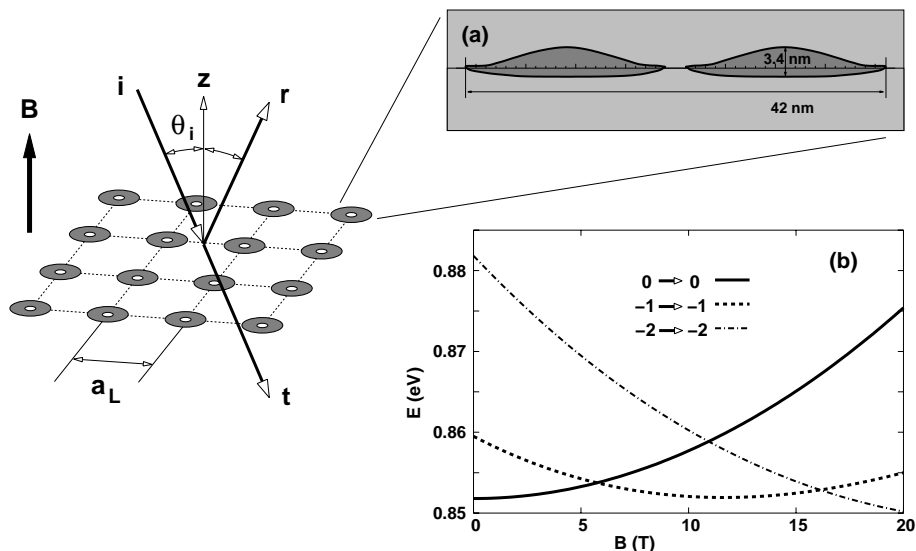


Fig. 1 – Schematic diagram of magneto-optical phenomena in a layer of nano-rings. (a) Schematic InAs/GaAs “eye”-shaped nano-ring (after TEM picture). (b) Transition energy as a function of magnetic field $\mathbf{B} = (0, 0, B)$ for $m_e = m_h = 0, -1, -2$ ($m_h \rightarrow m_e$) optical transitions.

In this sense those methods use pre-excitation. The data obtained by these methods are very important, but actually only return averaged single nano-ring information. For a quantitative characterization of the optical properties of nano-ring-based composite materials, a highly developed optical spectroscopy, like ellipsometry, and an advanced theoretical description are indispensable. Proper understanding and modeling of the collective electromagnetic response of nano-ring layers requires a correct approach, taking into account their composite and discrete character [11, 12].

The purpose of this letter is to show quantitatively that the collective electromagnetic properties of layers of semiconductor nano-rings can be controlled by means of an applied magnetic field through optical AB oscillations. To reach this goal, we develop here the theory of optical AB effects beyond the single nano-ring picture and show that these AB effects are clearly within the range of a modern ellipsometric setup.

The system to be investigated here is a square lattice of InAs/GaAs nano-rings of lattice parameter a_L (fig. 1). The basic element of this lattice is an “eye”-shaped ring as obtained in recent experiments [13], shown in fig. 1(a). It was found recently, that experimentally relevant simulations of the behavior of nano-rings can only be obtained with three-dimensional models using the experimentally determined shape, strain and composition of the rings [5, 7, 8]. We calculate the electron and hole energies/envelope functions, adjacent to the energy gap, for a nano-ring in the presence of a magnetic field \mathbf{B} using the approximate one-band Hamiltonian for electrons and holes as described in detail in [8]. However, the approximations involved, will not affect substantially the treatment of the optical response of layers of nano-objects, as presented here.

For frequencies ω close to the energy gap of the nano-ring, the size of the ring is small as compared to the wavelength and the dipole approximation can be used. Therefore, it is possible to use the Kramers-Heisenberg expression [14] for the polarizability tensor $\hat{\alpha}$ of a

single ring:

$$\hat{\alpha}(\omega) = \hat{\alpha}_S + \frac{e^2}{\hbar} \sum_{i,f} \left(\frac{\omega_{fi}}{\omega} \right) \left[\frac{\langle i|\mathbf{r}|f\rangle \langle f|\mathbf{r}|i\rangle^T}{\omega_{fi} - \omega - i\gamma} \right]. \quad (1)$$

This is an approximate expression, where negative frequencies have been ignored (the near resonance condition). The $\hat{\alpha}_S$ is the static part of the polarizability to be treated later. The dynamic part of the polarizability consists of the sum over states in (1). The possible transition energies $\hbar\omega_{fi} = E_f^e - E_i^h$ are determined by the energies E_f^e, E_i^h for the excited (electron) and ground (hole) states, respectively, as obtained in the previously mentioned calculation. The labels f, i are shorthand notations for the quantum number pairs $\{n_e, m_e\}, \{n_h, m_h\}$, where $n_{e,h}$ is the main and $m_{e,h}$ is the azimuthal quantum number for the electron (hole) states. Using the selection rules $m_e = m_h$ for linearly polarized light, described in [5], enables us to consider near the quantum ring absorption edge only the optical transitions shown in fig. 1(b) as functions of magnetic field B . The sum over states in (1) is limited to transitions from hole states with $n_h = 1$ to electronic states with $n_e = 1$ under the condition that $m_h = m_e = 0, -1, -2$, because only the near absorption edge transitions have the appropriate sensitivity. The crossing of those transition energy curves in fig. 1(b) is the essence of the optical AB effect [4] for a single ring (the large lattice parameter a_L used here, allows for that). γ is the damping factor chosen to be independent of frequency for the selected energy interval. The spatial part of the polarizability is determined by the optical matrix elements $\langle i|\mathbf{r}|f\rangle$, the expectation value of the position vector $\mathbf{r} = (x, y, z)$. Excitonic and Faraday/Kerr or Cotton/Mouton-type magneto-optical effects are not taken into account here.

Because of the cylindrical symmetry of the ring and following the selection rules, we use for our calculations the following optical transition matrix elements [5, 15]:

$$\begin{aligned} \langle i|z|f\rangle &\approx 0, \\ |\langle i|x|f\rangle| &= |\langle i|y|f\rangle| = |\langle S|x|X\rangle| I_{m_e m_h} \quad (m_e = m_h), \\ I_{m_e m_h} &= \frac{1}{\sqrt{2}} \int \rho d\rho dz F_{m_e}^e(\boldsymbol{\rho}, z) F_{m_h}^h(\boldsymbol{\rho}, z), \end{aligned} \quad (2)$$

where $I_{m_e m_h}$ is the electron-hole overlap integral, $F(\boldsymbol{\rho}, z)$ the envelope functions and $\boldsymbol{\rho} = (x, y)$. The matrix element $\langle S|x|X\rangle$ can be presented in the conventional notation of the Kane parameter P ,

$$\langle S|x|X\rangle = \frac{P}{\sqrt{2} \hbar \omega_{fi}}, \quad (3)$$

where m_0 is the free-electron mass [15]. This important value one can also take from direct experimental results available for InAs/GaAs nano-structures [16].

The separate addition of the static polarizability tensor $\hat{\alpha}_S$ is necessary to compensate the losses in the real part when the subsequent sum over states in (1) is limited to a small number of levels. We approximate this static polarizability by the polarizability of a homogeneous dielectric ellipsoid of rotation, for which an analytical expression has been given in [17]:

$$\begin{aligned} \alpha_{uu} &= \epsilon_0 V \frac{\epsilon - 1}{1 + N_u(\epsilon - 1)}, \\ N_z &= \frac{1}{1 - \zeta^2} \left(1 - \frac{\zeta \cos^{-1} \zeta}{\sqrt{1 - \zeta^2}} \right), \end{aligned} \quad (4)$$

where ϵ is the optical dielectric constant of the ring material, $u = x, y, z$ and $\zeta = c/a$, the aspect ratio with a the long and c the short, z -oriented, axis of rotation of the ellipsoid. V is

the volume of the ring. For reasons of symmetry, the relation $2N_x + N_z = 1$ can be used to determine N_x .

The polarizability tensor determines the reflection (r_{ss} , r_{pp}) and transmission (t_{ss} , t_{pp}) coefficients of a layer of nano-rings. For a square-lattice plane, Vlieger has shown that these coefficients are given by [12]

$$\begin{aligned} r_{ss} &= \frac{f_k}{A_y \cos \theta_i - f_k}, \\ r_{pp} &= \frac{f_k \cos \theta_i}{A_x - f_k \cos \theta_i} - \frac{f_k \sin^2 \theta_i}{A_z \cos \theta_i - f_k \sin^2 \theta_i}, \\ t_{ss} &= 1 + r_{ss}, \\ t_{pp} &= \frac{f_k \cos \theta_i}{A_x - f_k \cos \theta_i} + \frac{A_z \cos \theta_i}{A_z \cos \theta_i - f_k \sin^2 \theta_i}. \end{aligned} \quad (5)$$

Here subscripts “s” and “p” refer to light polarization perpendicular and parallel to the plane of incidence, respectively, θ_i is the angle of incidence (fig. 1), $k = c^{-1}\omega$ is the wave vector, c is the speed of light. In (5) we have made use of the following definitions:

$$\begin{aligned} A_u &= \alpha_0 \alpha_{uu}^{-1} - \mathbf{f}_{S,uu}, \\ f_k &= 2\pi i a_L k \end{aligned} \quad (6)$$

with u as defined before. The values for the diagonal part of the intraplanar transfer tensor $\hat{\mathbf{f}}_S$ for a two-dimensional square dipole lattice are $\mathbf{f}_{S,xx} = \mathbf{f}_{S,yy} = -4.51681$, $\mathbf{f}_{S,zz} = -2 \mathbf{f}_{S,xx} = 9.03362$ [12]. The scaling factor $\alpha_0 = 4\pi\epsilon_0 a_L^3$ is commonly used in discrete dipole calculations [12].

Magnetic-field-dependent optical response. The optical properties of the layer of nano-rings in the presence of a magnetic field \mathbf{B} will be represented by the standard ellipsometric angles Ψ , Δ , defined by [18]

$$\frac{r_{pp}}{r_{ss}} = \tan \Psi e^{i\Delta}. \quad (7)$$

These angles are shown in fig. 2, where for the layer of InAs/GaAs nano-rings a lattice parameter of $a_{L0} = 80$ nm has been used, which yields $\alpha_0 = 5.70 \cdot 10^{-32}$ fm². For the calculation of the static polarizability tensor we have used the optical dielectric constant $\epsilon = 12.2$ for InAs and an aspect ratio of $\zeta = 0.081$. The requirement that the volume of the ellipsoid has to be the same as that of the real ring yields a long axis of $a = 18.46$ nm. The components of the static polarizability tensor are then $\alpha_{S,xx} = \alpha_{S,yy} = 2.26 \cdot 10^{-3} \alpha_0$ and $\alpha_{S,zz} = 3.40 \cdot 10^{-4} \alpha_0$. Further we use for the matrix element $\langle S|x|X \rangle = 0.6$ nm [16]. With these choices of parameters the values for the optical properties of the layer will be absolute.

The angles Ψ , Δ represent directly measurable quantities and are as such far beyond the averaged characterization of the optical response of a single ring. In fig. 2 we show Ψ , Δ for a value of $\hbar\gamma$ of 2 and 5 meV. These two values cover the range of γ found in experiment [10] and used in the theory of semiconductor nanostructures. The dynamic range of Ψ , Δ for the energies and magnetic fields used, is several degrees, being 2 to 3 orders of magnitude above the detection limit of a modern ellipsometer [18]. The figure clearly demonstrates strong oscillations in both Ψ and Δ , but the peaked response is most outspoken for Δ . These peaks correspond to the crossings of the transition energies, characteristic of the optical AB effect. The oscillations dominate the magnetic-field dependence of the optical response and have to be distinguished from the weaker Faraday/Kerr and Cotton/Mouton effects.

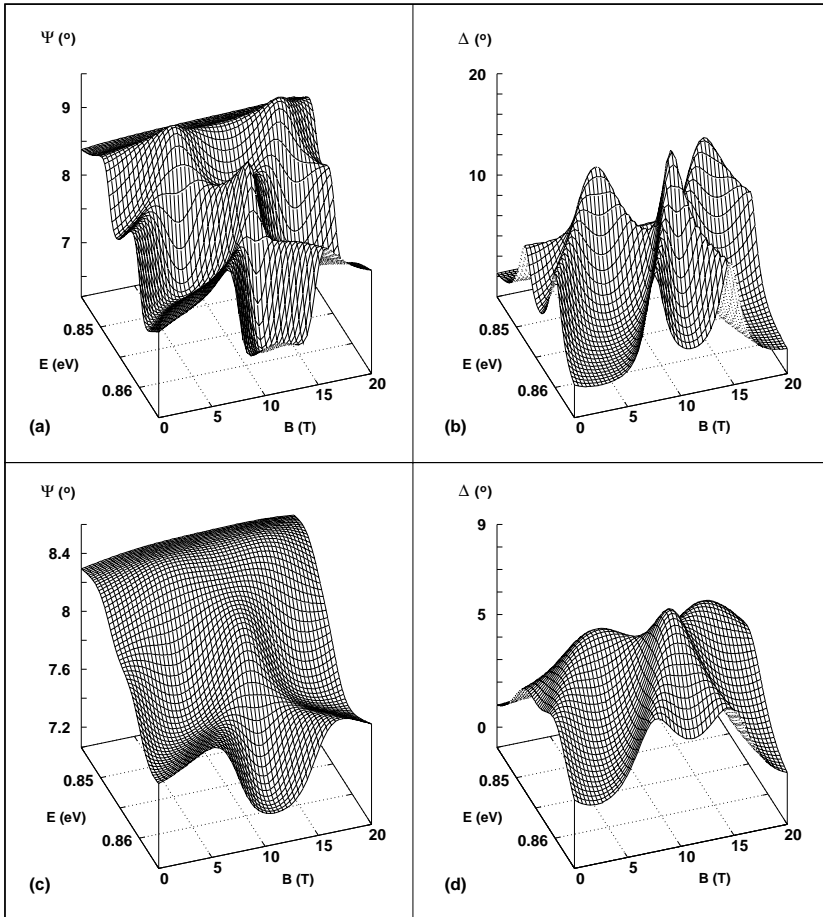


Fig. 2 – Ellipsometric angles Ψ , Δ for a monolayer of InAs/GaAs nano-rings. (a), (b) Ψ , Δ for $\theta_i = 60^\circ$ and $\hbar\gamma = 2$ meV. (c), (d) Ψ , Δ for $\theta_i = 60^\circ$ and $\hbar\gamma = 5$ meV.

Figure 3(a) shows the absorbance of the layer of rings, which is defined as

$$A = 1 - |r|^2 - |t|^2. \quad (8)$$

The absorbance inherits the oscillatory behavior of the ellipsometric angles Ψ , Δ shown in fig. 2. Actually, the interpretation should be the other way around. Absorbance is the optical quantity which is theoretically closest to the quantum-mechanical transitions in the layer of rings. We stress that the magnitude depends strongly on the average concentration of the rings in the layer (fig. 3(b)). This is a clear demonstration for the collective character of the response described. At the same time, it shows that ellipsometric detection of the optical AB effect requires a high lattice density. Fortunately, this condition is already obeyed by the existing samples. Experimentally, the absorbance has to be obtained through calculation from the experimental reflection and transmission coefficients. Yet the role of the absorbance goes further. The absorbance peaks in fig. 3(a) point out the hotspots of energy exchange between electromagnetic field and the charge carriers of the lattice of rings. This is potentially useful for magnetically controllable lasers. Figure 3(c) shows that even without varying the energy, a magnetic field is able to change the value of the absorbance.

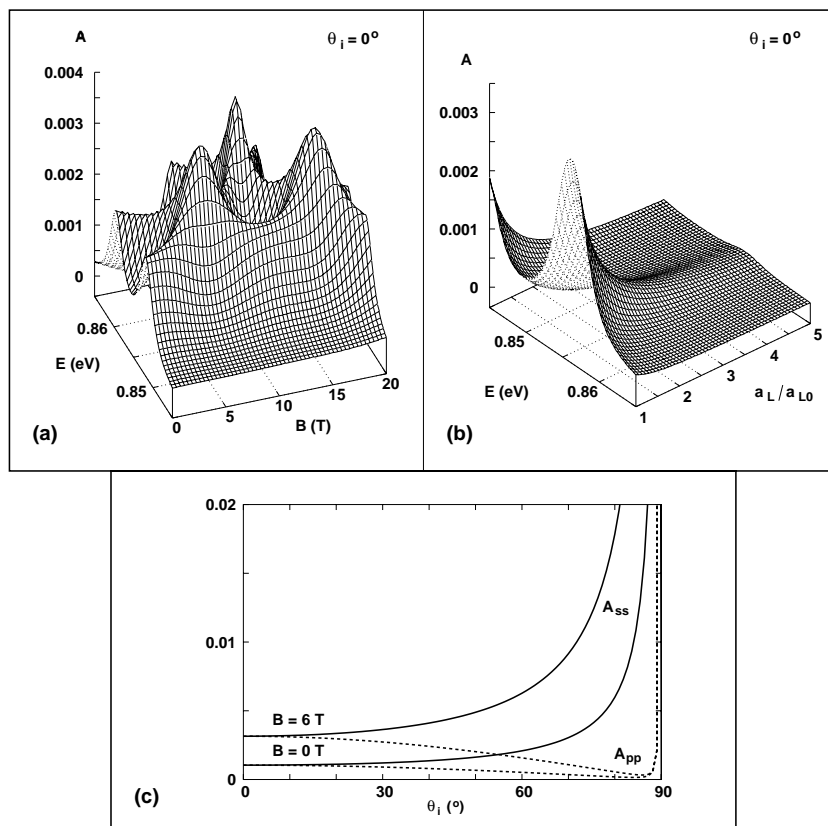


Fig. 3 – Optical absorbance by a layer of InAs/GaAs nanorings ($\hbar\gamma = 2$ meV). (a) Absorbance A for $\theta_i = 0^\circ$. (b) Peak value of ss -type absorbance A as shown in (a) for varying lattice parameter a_L and energy $\hbar\omega$ ($B \approx 6$ T). (c) As (b), but now for varying θ_i only ($\hbar\omega \approx 0.854$ eV and magnetic field B varies from 0 T to 6 T).

In addition, the same panel shows an interesting feature of the absorbance. There is a large difference in the dependence of A_{ss} and A_{pp} on the angle of incidence (fig. 3(c)). This difference has nothing to do with Brewster-like phenomena and therefore can be rightly called angular-dependent dichroism. It is due to the high anisotropy of the polarizability tensor, as can be deduced from (1) and (2), since this difference totally disappears when we do a reference calculation with $\alpha_{zz} = \alpha_{xx}$.

In this letter we have developed a quantitative discrete theory of optical AB effects beyond the single nano-ring picture and within a realistic three-dimensional approach. We conclude that averaged information about single nano-rings is not enough to describe and predict properties of composite materials made from nano-rings. Apart from the quantum-mechanical processes inside the ring, the geometry of the material has to be taken into account. This becomes clear from the expression for induction of an embedded (*e.g.* in a layer) single ring, given by

$$\mathbf{p}_s = \hat{\alpha} \mathbf{E}_L,$$

where \mathbf{p}_s is the dipole moment of the nano-ring and \mathbf{E}_L the local field acting upon the ring and reflecting the system geometry as a whole. For a different geometry this local field will change.

This turns the local field into the key parameter for the design of composite materials. In the context of this letter this means that the AB effect enters through the polarizability $\hat{\alpha}$. Only the local field \mathbf{E}_L , however, can turn the dipole strength \mathbf{p}_s into the full optical response. The effect is an inherently collective one. The optical AB effects predicted in this theory turn out to be easily measurable for any modern ellipsometric setup. It has been made clear that the optical AB effect is sufficiently strong to be used to control the collective electromagnetic properties of layers of semiconductor nano-rings by means of magnetic fields in the optical regime. This is potentially useful for the design of composite materials with negative refractive index. We emphasize that this option is not available for other systems of semiconductor nano-objects. We invite the experimentalists to conduct the measurements necessary to verify our findings.

* * *

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