NoN analyticity of Hanle effect at zero magnetic field in a quantum dot

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Abstract

Non analytic behaviour of Hanle effect in InGaAs quantum dots is described in terms of a simple 4-level model. Despite simplicity the model makes it possible to explain the observed fracture of Hanle curve at zero magnetic field and obtain quantitative agreement with the experiment.

1 Introduction

In this note we suggest a possible explanation of nonanalytic form of Hanle curves measured for an ensemble of InGaAs/GaAs quantum dots (QD) in [1]. These curves (i.e. the dependance of the degree of circular polarization of luminescence on magnetic field in Voigt geometry) were obtained using the protocol of excitation providing suppression of nuclear polarization in QD. Typical curve (taken from [1]) presented at Fig.1a reveal a ”fracture” at zero magnetic field. High quality of Hanle curves obtained in [1] make this
nonanalytic character unambiguous. Below we present an explanation of this feature in the frame of a simple model of energy structure of QD. Similar model with some complications was used, for example, in [2].

2 The model

This model consider QD as an “island” of some semiconductor material surrounded by a material with a broader band gap (in our case it is InGaAs QD surrounded by GaAs). Electron motion within QD is quantized and we take into account only one level of spatial quantization in valence zone (we call it valence zone level) and one level in conduction zone (conduction zone level). Each of these levels can be occupied by two electrons (with spin-up and spin-down). We assume that the ground state |0⟩ of uncharged QD is correspond to the presence of two electrons at the valence zone level and to the absence of electrons at the conduction zone level. Lowest excitations of QD can be obtained by transition of one electron to the conduction zone level (appearance of hole at the valence level). So, four lowest excited states (equal to the number of spin configurations) are possible |s_z, −μ_z⟩, where s_z = ±1/2 and μ_z = ±1/2 are spins of electron at the conducting and valence zone levels respectively. These four states we denote as | + +⟩, | + −⟩, | − +⟩, | − −⟩ (Fig.2). Consider the possible channels of luminescence of QD. We assume that recombination of electron and hole with opposite spin is the only possible processes giving rise to the luminescence and that both of these processes have the same probability. Therefore

$$\left| \langle + + | \hat{d}_\pm |0\rangle \right|^2 = \left| \langle - - | \hat{d}_\pm |0\rangle \right|^2 = 0,$$

(1)

and

$$\left| \langle + - | \hat{d}_\pm |0\rangle \right|^2 = \left| \langle - + | \hat{d}_\pm |0\rangle \right|^2 \equiv D^2,$$

(2)

here \(\hat{d}_\pm\) is the dipole operator of interaction with electromagnetic field of \(\sigma_\pm\) polarization. The recombination of electron with spin +1/2 (-1/2) and hole with spin -1/2 (+1/2) is accompanied by emission of \(\sigma_+\) (\(\sigma_-\)) photon. Below we consider the rate of luminescence to be proportional to \(D^2\)

$$W = kD^2$$

(3)
3 Luminescence and Hanle effect

Within the frame of the above model an arbitrary excited state $\Psi$ of the QD can be presented as a linear combination of the above basis states:

$$\Psi = C^{++}|++\rangle + C^{+-}|+-\rangle + C^{-+}|-+\rangle + C^{--}|--\rangle$$ (4)

If $n$ QD’s are prepared in such a state then within $dt$ time interval $dn_+$ photons in $\sigma_+$ polarization will be emitted and

$$dn_+ = nk|\langle \Psi|d_+|0\rangle|^2 dt = nW|C^{+-}|^2 dt$$ (5)

Analogously in $\sigma_-$ polarization $dn_-$ photons will be emitted

$$dn_- = nk|\langle \Psi|d_-|0\rangle|^2 dt = nW|C^{-+}|^2 dt$$ (6)

If emitted photons are registered by a differential polarimetric detector whose output current $i$ is proportional to the difference of the photon currents in polarizations $\sigma_+$ and $\sigma_-$ (i.e. $dn_+/dt - dn_-/dt$) with corresponding coefficient of proportionality being the quantum efficiency $\xi$ of the photoreceivers, then the contribution of these $n$ QD’s to the output current of such detector can be calculated as

$$i = e\xi nW \left( |C^{+-}|^2 - |C^{-+}|^2 \right)$$ (7)

where $e$ is the electron charge. In this formula the number $n$ of QD’s and coefficients $C^{+-}$ and $C^{-+}$ can be time dependent.

Now we consider the Hanle effect i.e. the dependance of degree of polarization of luminescence on magnetic field. We imply the steady state regime when the reasons exciting the luminescence do not depend on time. For the process of QD’s excitation we accept the following model. Assume that at the time moment $T$ there are $N_0$ QD’s in the ground state and there is some action under which these dots (with probability per time unit $P$) can be excited in state $\Psi_0$:

$$\Psi_0 = C_0^{++}|++\rangle + C_0^{+-}|+-\rangle + C_0^{-+}|-+\rangle + C_0^{--}|--\rangle$$ (8)

with coefficients $C_0^{++}$, $C_0^{+-}$, $C_0^{-+}$, $C_0^{--}$ supposed to be known. (For example if this action is irradiation by light with $\sigma_+$ polarization then the only non zero coefficient is $C_0^{+-}$). So, in the case of this regime of excitation $n_0 = PN_0dT$ QD’s in state $\Psi_0$ are generated within the time interval from $T$ to $T + dT$. Let us consider the contribution of this set of dots to the output current of
the above differential receiver. After excitation these QD’s begin to emit photons and their contribution to the output current is described by Eq. (7) in which the temporary dependance of coefficients $C^+(t)$ and $C^-(t)$ is defined by solution of Shrődinger equation for the wave function $\Psi(t)$ of QD with initial condition Eq. (8) at $t = T$. The relevant Hamiltonian has the form:

$$H = g_e\beta B s + g_h\beta B \mu$$

(9)

where $s$, $\mu$ and $B$ are the operators of electron and hole (electron at the valence zone level) spins and magnetic field. In this formula we take into account that g-factors of electron $g_e$ and hole $g_h$ can be different: $g_e \neq g_h$.

The temporary dependance of the number of excited QW’s is governed by the equation which is obtained by summation of Eq (5) and (6):

$$\frac{dn}{dt} = -nW\left(\left|C^+(t)\right|^2 + \left|C^-(t)\right|^2\right)$$

(10)

If $n(t = T) = n_0$ then one can write for $n(t)$ the following expression:

$$n(t) = n_0 \exp\left\{ -W \int_T^t d\tau \left(\left|C^+(\tau)\right|^2 + \left|C^-(\tau)\right|^2\right) \right\}$$

(11)

Consequently, the contribution $di_T(t)$ of set of QD’s created in state Eq. (8) in temporary interval $[T, T + dT]$ to the output current of differential photodetector is defined by the following expression (see Eq. 7):

$$di_T(t) = \xi e PN_0 W dT \exp\left\{ -W \int_T^t d\tau \left(\left|C^+(\tau)\right|^2 + \left|C^-(\tau)\right|^2\right) \right\} \times$$

$$\times \left(\left|C^+(t)\right|^2 - \left|C^-(t)\right|^2\right) \Theta(t - T)$$

(12)

Assume that we have calculated the contribution $di_0(t)$ produced by QD’s excited at $T = 0$. Due to the stationarity of $P$ and $N_0$ (steadystate regime) the following relationship should hold:

$$di_T(t) = di_0(t - T)$$

(13)

To calculate the total output current $I$ of differential photodetector one should integrate contributions of all QD’s excited at an arbitrary time moments:

$$I = \int_{-\infty}^{+\infty} \frac{di_T(t)}{dT} dT$$

(14)

Taking into account the Eq’s (12) and (13) and making the replacement of variable we obtain:

$$I = \xi e PN_0 W \int_0^{+\infty} dT \exp\left\{ -W \int_0^T d\tau \left(\left|C^+(\tau)\right|^2 + \left|C^-(\tau)\right|^2\right) \right\} \times$$

$$\times \left(\left|C^+(t)\right|^2 - \left|C^-(t)\right|^2\right) \Theta(t - T)$$

(15)
Functions $C^+(t)$ and $C^-(t)$ entering this equation should be obtained by solution of temporary Shrödinger equation with initial conditions Eq. (8) at $t = 0$. Now we have to find the value of $N_0$ – the number of QD’s in the ground state in steadystate regime. Let us for a moment consider the photoreceiver (not the above differential one) whose output current is proportional to the total photon current in both polarizations. If we denote this current as $I_\Sigma$ then:

$$I_\Sigma = \xi e P N_0$$

If the intensity $P$ of excitation is small and the effects of saturation can be neglected then one can set the value of $N_0$ be equal to the total number $N$ of QD’s in the region of irradiation. If it is not the case then for calculation of $N_0$ the total number of dots $N$ should be reduced by a number $N^*$ of dots excited to an arbitrary (because of the steadystate regime) time moment (say, $t$):

$$N_0 = N - N^*$$  \hspace{1cm} (16)

The value of $N^*$ can be obtained by summation of the numbers of QD’s excited at all time moments $T$ with $T < t$ (see formula Eq. (11)):

$$N^* = P N_0 \int_{-\infty}^{t} dT \exp \left\{-W \int_{T}^{t} d\tau \left( |C^+(\tau)|^2 + |C^-(\tau)|^2 \right) \right\}$$  \hspace{1cm} (17)

Using the fact that functions $C(\tau)$ depend on the difference $\tau - T$ and making the relevant replacing of variables in the integral one can obtain:

$$N^* = P N_0 \int_{0}^{\infty} dT \exp \left\{-W \int_{0}^{T} d\tau \left( |C^+(\tau)|^2 + |C^-(\tau)|^2 \right) \right\}$$  \hspace{1cm} (18)

Substituting this into Eq. (16) we obtain for $N_0$ the following expression:

$$N_0 = N \left[ 1 + P \int_{0}^{\infty} dT \exp \left\{-W \int_{0}^{T} d\tau \left( |C^+(\tau)|^2 + |C^-(\tau)|^2 \right) \right\} \right]^{-1} \hspace{1cm} (19)$$

It is convenient to introduce the following functions:

$$\Phi(T) \equiv \exp \left\{-W \int_{0}^{T} d\tau \left( |C^+(\tau)|^2 + |C^-(\tau)|^2 \right) \right\}, \quad F(T) \equiv |C^+(T)|^2 - |C^-(T)|^2$$  \hspace{1cm} (20)

then

$$I = \xi e P N_0 W \int_{0}^{+\infty} dT \Phi(T) F(T)$$  \hspace{1cm} (21)

$$N_0 = N \left[ 1 + P \int_{0}^{\infty} dT \Phi(T) \right]^{-1} \hspace{1cm} (22)$$
To find the current $I$ one should calculate the temporary dependance of coefficients $C^{++}(t)$, $C^{+-}(t)$, $C^{-+}(t)$, $C^{--}(t)$. We now solve this problem for the magnetic field having only $x$ component i.e. $\mathbf{B} = (B, 0, 0)$. We are going to analyse the Hanle effect in Voigt geometry, so this is the only case of interest for us. If we arrange the basis functions corresponding to the presence of one electron-hole pair in QD in the way described above then the matrix of the Hamiltonian $H$ Eq. (9) will take the following form:

$$
H = \frac{\hbar}{2} \begin{pmatrix}
0 & \nu_h & \nu_e & 0 \\
\nu_h & 0 & 0 & \nu_e \\
\nu_e & 0 & 0 & \nu_h \\
0 & \nu_e & \nu_h & 0
\end{pmatrix}
$$

(23)

where $\hbar \nu_{e(h)} \equiv g_{e(h)} \beta B$. All four states in the absence of magnetic field have the same energy (degenerated). This constant energy can be omitted in the Shrödinger equation which has the form:

$$
\frac{i}{\hbar} \frac{dC^{++}}{dt} = \frac{1}{2} [\nu_h C^{+-} + \nu_e C^{-+}] \\
\frac{i}{\hbar} \frac{dC^{--}}{dt} = \frac{1}{2} [\nu_e C^{+-} + \nu_h C^{-+}] \\
\frac{i}{\hbar} \frac{dC^{+-}}{dt} = \frac{1}{2} [\nu_h C^{++} + \nu_e C^{--}] \\
\frac{i}{\hbar} \frac{dC^{-+}}{dt} = \frac{1}{2} [\nu_e C^{++} + \nu_h C^{--}]
$$

(24)

Introduce new variables:

$$
X \equiv C^{++} + C^{--}, \ Y \equiv C^{++} + C^{--}, \ Z \equiv C^{++} - C^{--}, \ G \equiv C^{++} - C^{--}
$$

(25)

and

$$
\Omega \equiv \frac{1}{2} \frac{B \beta (g_h + g_e)}{\hbar} = \frac{1}{2} (\nu_h + \nu_e), \ \omega \equiv \frac{1}{2} \frac{B \beta (g_h - g_e)}{\hbar} = \frac{1}{2} (\nu_h - \nu_e)
$$

(26)

Then the general solution of Eq. (24) has the form:

$$
X = A \cos \Omega t + B \sin \Omega t, \ Y = i[B \cos \Omega t - A \sin \Omega t] \\
Z = A_1 \cos \omega t + B_1 \sin \omega t, \ G = i[B_1 \cos \omega t - A_1 \sin \omega t],
$$

(27)

(28)

where constants $A, A_1, B, B_1$ defined by the the initial conditions. $C$-functions we are interesting in can be expressed as:

$$
C^{+-} = \frac{1}{2}(X + Z), \quad C^{-+} = \frac{1}{2}(X - Z)
$$

(29)
4 Excitation into the allowed for the luminescence state

In this case only $C^{+−} = 1$ is non-zero at $t = 0$ and one can write down the following initial conditions:

\[ X(t = 0) = 1, \quad Y(t = 0) = 0, \quad Z(t = 0) = 1, \quad G(t = 0) = 0 \quad (30) \]

Consequently:

\[ X(t) = \cos \Omega t, \quad Y(t) = -i \sin \Omega t, \quad Z(t) = \cos \omega t, \quad G(t) = -i \sin \omega t \quad (31) \]

and

\[ C^{+−} = \frac{1}{2} (\cos \Omega t + \cos \omega t), \quad C^{−+} = \frac{1}{2} (\cos \Omega t - \cos \omega t) \quad (32) \]

For the combinations of $C$-functions we are interesting in we obtain:

\[ \left| C^{+−} \right|^2 + \left| C^{−+} \right|^2 = \frac{1}{2} (\cos^2 \Omega t + \cos^2 \omega t) \quad (33) \]

and

\[ \left| C^{+−} \right|^2 - \left| C^{−+} \right|^2 = \cos \Omega t \cos \omega t \quad (34) \]

In this case the functions Eq. (20) (we supply them by a mark $b$ (bright)) have the form:

\[ \Phi_b(T) = \exp \left\{ - \frac{W}{2} \left( T + \sin \frac{2\Omega T}{4\Omega} + \sin \frac{2\omega T}{4\omega} \right) \right\} \quad (35) \]

\[ F_b(T) = \cos \Omega T \cos \omega T \]

Now it is possible to calculate the output current of differential photodetector by means of formulas Eq. (22,21). The field dependance (due to the field dependance of $\Omega$ and $\omega$) of the output current in this case is the curve with wide maximum (nearly plane-like in the vicinity of zero field) strongly differing from Lorentz curve (Fig3. top).

5 Excitation into the forbidden for the luminescence state

This corresponds to the initial condition with only non-zero $C^{++} = 1$. Consequently (see Eq. (25))

\[ X(t = 0) = 0, \quad Y(t = 0) = 1, \quad Z(t = 0) = 0, \quad G(t = 0) = 1 \quad (36) \]
and then:

\[ X(t) = -\imath \sin \Omega t, \quad Y(t) = \cos \Omega t, \quad Z(t) = -\imath \sin \omega t, \quad G(t) = \cos \omega t \]  

(37)

and further:

\[ C^{+-} = -\frac{1}{2} (\sin \Omega t + \sin \omega t), \quad C^{-+} = -\frac{1}{2} (\sin \Omega t - \sin \omega t) \]  

(38)

For the combinations of \( C \)-functions we are interesting in we obtain:

\[ \left| C^{+-} \right|^2 + \left| C^{-+} \right|^2 = \frac{1}{2} (\sin^2 \Omega t + \sin^2 \omega t) \]  

(39)

and

\[ \left| C^{+-} \right|^2 - \left| C^{-+} \right|^2 = \sin \Omega t \ \sin \omega t \]  

(40)

In this case the functions Eq. (20) (we supply them by a mark \( d \) (dark)) have the form:

\[ \Phi_d(T) = \exp \left\{ -\frac{W}{2} \left( T - \frac{\sin 2\Omega T}{4\Omega} - \frac{\sin 2\omega T}{4\omega} \right) \right\} \]  

(41)

\[ F_d(T) = \sin \Omega T \ \sin \omega T \]

Calculations of output current of differential photodetector by formulas Eq. (21, 22) shows that current field dependance is non-analytic in the vicinity of zero field – a kind of ”fracture” is appeared (Fig3. bottom). Note that the experimental Hanle curve also has the peculiarity of this type.

6 General case

Let us consider now the general case. It means that in the steadystate regime there are \( N_0 \) dots in the ground state and \( P^{+-} \) is the probability of excitation of QD in state \(| + - \rangle \) and \( P^{++}, P^{-+} \) and \( P^{--} \) are the same probabilities for states \(| ++ \rangle, | - + \rangle \) and \(| -- \rangle \). In this case the calculation analogous to presented above give the following expression for output current of the differential photodetector in terms of functions Eq. (35) and Eq. (41)

\[ I = \xi eW N_0 \left[ \left( P^{+-} - P^{-+} \right) \int_0^\infty dT \Phi_b(T) \Phi_b(T) + \left( P^{++} - P^{--} \right) \int_0^\infty dT F_d(T) \Phi_d(T) \right] \]  

(42)

\[ N_0 = N \left[ 1 + \left( P^{++} + P^{-+} \right) \int_0^\infty \Phi_b(T) dT + \left( P^{++} + P^{--} \right) \int_0^\infty \Phi_d(T) dT \right]^{-1} \]

The commonly used quantity measured in experiment is the degree of polarization (we denote it \( \rho \)). To calculate \( \rho \) one should divide the obtained above differential photocurrent on total photocurrent \( I_\Sigma \) in both polarizations:

\[ I_\Sigma = \xi eN_0 (P^{+-} + P^{-+} + P^{++} + P^{--}) \]
and consequently:

\[
\rho = W \left[ \left( P^{++} - P^{-+} \right) \int_0^\infty dT F_b(T) \Phi_b(T) + \left( P^{++} - P^{-+} \right) \int_0^\infty dT F_d(T) \Phi_d(T) \right] \frac{P^{-+} + P^{++} + P^{-+} + P^{-+}}{P^{++} + P^{++} + P^{-+} + P^{-+}}
\]

\[(43)\]

7 Comparison with the experiment

Now let us apply the obtained results to the experiments related to Hanle effect in InGaAs QD. First of all we note that the polarized luminescence is observed from the ensembles of charged QD’s \[1, 3\]. The degree of polarization of luminescence of uncharged QD was found to be weak. In charged QD there are three electrons – two electrons occupy the valence zone level and one the conducting zone level.

In terms of above model the reason of weak polarization of the luminescence of uncharged QD’s may be as follows. In uncharged QD both electrons occupy the valence zone level. The external excitation generate the electron-hole pairs in the barrier. So, to emit photon the uncharged QD should simultaneously trap electron and hole from the barrier. This may take much more time as compared with that required for trapping only electron (or only hole). Therefore the initial polarization of the electron-hole pair (created by the external polarized pumping) can decay and the luminescence of QD appears to be weakly polarized in this case.

Now let QD be charged by a single electron (resident electron) as it is the case for the experiments described in \[1\]. In this case of negatively charged QD trapping of the hole (positively charged) from the barrier is likely due to the Couloumn attraction. For this reason trapping of the hole can be so fast that its polarization does not decay and is defined by the polarization of pumping (say \(\sigma_+\)). Therefore if the spin of resident electron is random then we have the case of excitation of QD states \(|+−>\) and \(|−−>\) with equal probabilities. The scheme of this process is presented at fig.4. So, to calculate the Hanle signal one can use formula \((43)\) with \(P^{−−} = P^{++} \neq 0\) and \(P^{++} = P^{−+} = 0\). The result of such calculation is presented at fig.1b. The values of fitting parameters are presented at figure caption. One can see that theoretical and experimental curves have much in common. This justify the calculation presented in this note. Despite the fact that the only important fitting parameter in our calculations was the ratio \(g_e/g_h\) the degree of polarization at zero field (17%) is also in good agreement with the
The most important feature of the observed Hanle curve is, in our opinion, the presence of a fracture in the vicinity of zero field. Within the above simple model this fracture appeared due to the excitation of QD in the state $|++\rangle$ forbidden for the luminescence (see section V). The second important reason of this is temporal dependance of the probability of the luminescence after excitation (see Eq. (10)). Similar dependance was studied, for example, in [4].

Finally it should be mentioned that one of the curious features of the luminescence of QD is that its polarization has the sign inverse to that of excitation [1, 2]. We are not ready to discuss this phenomenon in details but it sounds like true that the hole can change spin polarization during trapping by the QD.

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Sorry for not perfect English.
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Figure 1: (a) – the experimental Hanle curve for InGaAs QD ensemble, (b) – the same calculated. The fitting parameters are: \( g_h = 0.28, g_e = 0.57, P_{-+} = 1, P_{+-} = 1, P_{--} = P_{++} = 0, W = 0.25 \cdot 10^{10} \text{ sec}^{-1} \).
Figure 2: The QD’s ground and excited states.
Figure 3: Top – the Hanle curve for the case of excitation in $|+ - >$ state, bottom – the same for the case of excitation in $|++ >$ state.
Figure 4: Scheme of QD excitation.