Oscillatory screening of the dc electric field in the Si-SiO$_2$ multiple quantum wells probed by second-harmonic generation

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Abstract

DC-electric field, being screened in 3D semiconductors, normally decays monotonically in space. Experimental studies of the DC electric field screening in Si-SiO$_2$ multiple quantum wells by electric field induced optical second-harmonic generation show a non-monotonic, oscillatory-like decay. The model of electrons localized inside quantum wells, with the first subband occupied, allows a description of the phenomenon. Interwell Coulomb interaction, a finite value of the electron charge and strong effective-mass anisotropy result in a crucial difference from 3D Fermi liquid.
Properties of the degenerate electron subsystem of a solid are basically determined by an interplay between Coulomb interaction and quantum effects, which is revealed in correlation and exchange interaction. The corresponding typical energy scales are given by characteristic magnitude of the Coulomb interaction \( \varepsilon_C \propto e^2 n^{-1/3} \) and the Fermi energy for noninteracting electrons \( \varepsilon_F \propto \hbar^2 m_0^{-1} n^{-2/3} \), where \( n \) is the electron density, \( \varepsilon \) denotes the lattice dielectric constant, \( m_0 \), \( e \) and \( \hbar \) are fundamental constants. If \( \varepsilon_F / \varepsilon_C >> 10^{-2} \div 10^{-1} \) the system has the properties of a normal Fermi liquid \([1]\), whereas in the opposite case a dielectric bandgap forms \([2]\). An Anderson-Mott transition occurs at the specific value of \( \varepsilon_F / \varepsilon_C \)[3].

Significant attention has been paid, over recent decades, to spatially confined semiconductor-based systems. Advanced modern technologies allow the fabrication of structures with a predetermined morphology, and therefore control over their electronic properties. Amongst 2D semiconductor-based systems (superlattices and multiple quantum wells) two extreme cases should be pointed out. While epitaxial structures like GaAs/AlGaAs are characterized by relatively close bandgaps of the components, the bandgap difference in amorphous Si-SiO\(_2\) based systems are much higher than the Si bandgap.

The anisotropic dispersion law of electrons requires the consideration of two parameters, \( (\varepsilon_F / \varepsilon_C)_{\parallel} \) and \( (\varepsilon_F / \varepsilon_C)_{\perp} \), which describe tangential and transversal electron motion, respectively. For the Si layer with a carrier density of \( 10^{16} \div 10^{18} \) cm\(^{-3}\), at the zero temperature and with \( m \approx m_0 \) we have \( (\varepsilon_F / \varepsilon_C)_{\parallel} \approx 10^{-2} \div 10^{-1} \), i.e. lateral properties of the system can be described in terms of a Fermi-liquid. The parameter \( (\varepsilon_F / \varepsilon_C)_{\perp} \) is governed by the transparency of potential barriers in the superstructure. For high potential barriers the parameter \( (\varepsilon_F / \varepsilon_C)_{\perp} \) is very small, and the interlayer electron motion is suppressed by the Coulomb repulsion. Electrons are localized within the layers and the structure can be treated as a set of independent quantum wells (QW), i.e. as multiple QW’s (MQW). The Mott-like transition is expected at a certain transparency of the barrier.

Properties of the electron liquid reveal themselves in the response of the system to external fields. For example, Friedel oscillations appear in the screening cloud in the degenerate
Fermi-liquid \cite{4}. The period of these oscillations is governed by the typical de-Broglie wavelength in the system. 0D systems weakly interacting with their environment show a Coulomb blockade phenomenon, which is due to a finite value of the elementary charge \cite{5}. Analysis of four coupled quantum dots \cite{6} shows that properties of an array of interacting quantum dots depend on the interdot tunnel coupling. For weak coupling this system behaves as a set of independent dots, at intermediate coupling the Mott insulator forms (the so-called collective Coulomb blockade) and for strong coupling the Coulomb blockade is completely destroyed. It is worth noting that for the collective Coulomb blockade the screening has an essentially collective nature, but it still shows non-monotonic features. Particularly, the carrier density in neighboring quantum dots is \textit{anti-correlated}.

On the other hand, the DC-electric-field screening in bulk semiconductors shows monotonic spatial distribution of the voltage drop across the space charge region \cite{7}. We conclude that nonmonotonic peculiarities in the DC-screening indicate that the electron subsystem essentially differs from the bulk 3D case.

Optical second-harmonic generation (SHG) is a versatile optical probe of surfaces and interfaces \cite{8}. This technique is applicable to all interfaces accessible by light, including buried interfaces \cite{9}, and has inherently high spatial \cite{10}, spectral \cite{11} and temporal \cite{12} resolutions. The sensitivity of SHG arises from the strong symmetric selection rules for the second-order susceptibility. Namely, the second-order susceptibility vanishes in the dipole approximation for media with inversion symmetry, and the quadrupole term becomes prevalent. In the presence of the DC-electric field the dipole contribution into the second-order polarization \(P^{2\omega}\) arises \cite{13}:

\[
P^{2\omega} = i\chi^{(2Q)}k\omega E^\omega E^\omega + \chi^{(3D)}E^{DC} E^\omega E^\omega, \tag{1}
\]

where \(E^\omega\) and \(E^{DC}\) are the fundamental and DC fields, respectively; \(k\omega\) is the fundamental wavevector; nonlinear susceptibilities \(\chi^{(2Q)}\) and \(\chi^{(3D)}\) are responsible for the \(2\omega = \omega + \omega\) and \(2\omega = 0 + 0 + \omega\) processes, respectively. Two terms in (1) are comparable at \(E^{DC} \approx 10^5\) V/cm. This sensitivity to the DC-fields has been experimentally demonstrated for semiconductor-
electrolyte and semiconductor-dielectric interfaces [13].

In this paper, the DC electric field screening in Si-SiO$_2$ MQW is studied by DC-field induced SHG. The magnitude of parameter $(\varepsilon_F/\varepsilon_C)_{\perp}$ is smaller than $10^{-3}$ for the structures studied. The experimental bias dependence of the SHG intensity for MQW reveals the essentially non-monotonic, oscillatory-like behavior. This is attributed to the essential quantum effects in the highly anisotropic layered system with a Coulomb interaction between layers.

MQW structures were fabricated by RF magnetron sputtering on a vicinal n-Si(100) wafer [14]. MQW structures consist of 40 double amorphous layers of Si-SiO$_2$. Two MQW samples were studied with SiO$_2$ layers 3 and 5 nm thick. The Si layers were 1.1 nm thick for both samples. The perfect periodic layered structure of the MQW is studied by X-ray diffraction and on-line Auger-electron spectroscopy. The amorphous structure of the Si layers is confirmed by Raman spectroscopy [14].

The output of a Q-switched YAG:Nd$^{3+}$ laser at a wavelength of 1064-nm was used as fundamental radiation. A pulse duration of 15 ns and an intensity of 1-10 MW/cm$^2$ avoid laser damage of the sample and other undesired photo-induced effects. The experimental setup is described in detail elsewhere [15]. The transparency of the Si substrate at the fundamental wavelength allows the study of SHG in transmission. The advantage of this geometry is that only a few components of the nonlinear susceptibility tensor contribute to the signal, which simplifies the analysis.

DC-electric-field is applied to the MQW by imposing a bias voltage between the Al back-side electrode and the In-Ga ring gate electrode on the top of MQW. The free carriers easily move in the in-plane direction, while the QW’s are well isolated from each other, therefore the voltage distribution inside the structure does not depend on the in-plane coordinates, although the cap electrode is not planar.

The in-plane symmetry of the samples was analyzed by measuring the azimuthal SHG anisotropy. Figure 1 shows the dependence of the SHG intensity on the azimuthal angle $\psi$. The two-fold symmetry of the dependence indicates the in-plane anisotropy of the sample attributed to the initial miscut of the substrate from the (100) direction. From the micro-
scopic viewpoint, this means the “terrace” structure of the interface. We expect that these terraces are reflected in the morphology of several Si and SiO$_2$ layers, closest to the substrate, which possess the 2/m point-group symmetry (Figure 1). The deposition of further layers erodes the anisotropy. Therefore the symmetry of the rest of layers is expected to be $\infty/m$.

Since both 2/m and $\infty/m$ layers possess an inversion symmetry, $P^{2\omega}$ is given by Eq. (1). Symmetry of the layer governs the selection rules for the components of the fourth-rank tensors $\chi^{(2Q)}$ and $\chi^{(3D)}$. Take the y-axis to be parallel to the terraces, and the z-axis normal to the surface. For the transmission geometry $xzxx, xzyy$, and $yzxy = yzyx$ components of $\chi^{(2Q)}$ and $\chi^{(3D)}$ contribute to the SHG intensity for the 2/m layers, and there is no contribution from $\infty/m$ layers. Consequently, the DC-electric-field effects in the buried Si-SiO$_2$ layers near the vicinal Si substrate are purely detected.

Figure 2 shows the dependence of the SHG intensity on the applied bias measured at the maximum of the rotational anisotropy. Clear oscillatory-like behavior is observed. According to Eq.(1) this is related to the oscillatory dependence of the DC-electric-field on the bias.

The following model is developed to obtain the DC-field distribution. First, we suppose that there is no charge coupling between the upper Si layer and the metal electrode. Therefore, the current through the structure is zero, the Fermi level is the same for all QW’s and the Si substrate. Second, since $(\varepsilon_F/\varepsilon_C)_\perp << 10^{-3}$ the structure should be considered as a set of charged QW’s, with the 2D Fermi-gas of electrons in each well.

We consider the dependence of $E^{DC}$ and the electrostatic potential $\varphi$ in the middle of the Si layer and the 2D charge density $n$ as functions of the discrete variable the number of well $l$. The following discrete analogue of the electrostatic equations with appropriate boundary conditions is used:

$$
\begin{cases}
-(E^{DC}(l) - E^{DC}(l - 1)) = -2\pi(n(l) + n(l - 1))/\varepsilon \\
\varphi(l - 1) - \varphi(l) = (\varepsilon d + \varepsilon D_1)(E^{DC}(l) + E^{DC}(l - 1))/2\varepsilon ,
\end{cases}
$$

where $d$ and $D$ are the thicknesses of the Si and SiO$_2$ layers, respectively; $\varepsilon$ and $\varepsilon_1$ are static
dielectric constants of these layers, $U_0$ is a bias voltage.

We take into account only the first quantized sub-band of the electron energy spectrum of the quantum well. The expression for $n(\varphi)$ is obtained by the same procedure as for the 3D case [16] and is given by:

$$n(\varphi) = -\frac{m_e Te_e}{\pi \hbar^2} \ln[1 + \exp((e_e \varphi + \mu)/T)] + \frac{m_h Te_h}{\pi \hbar^2} \ln[1 + \exp(-(e_h \varphi - \mu + \Delta)/T)],$$  \hspace{1cm} (3)

where $\mu$ is the Fermi energy, $\Delta$ is the bandgap, $T$ is temperature, $m$ is the effective mass of the carrier, subscripts $e$ and $h$ stand for electrons and holes.

In the limit of $T = 0$ and $\Delta = 0$ the system (2) is linear and $\varphi(l)$ takes the form:

$$\varphi(l) = C_1 A^l + C_2 A^{-l},$$  \hspace{1cm} (4)

where $C_1$ and $C_2$ are constants and

$$A = \left(1 - \sqrt{(\varepsilon d + \varepsilon_1 D)/\varepsilon^2 a_0}\right) \left(1 + \sqrt{(\varepsilon d + \varepsilon_1 D)/\varepsilon^2 a_0}\right)^{-1}.$$  \hspace{1cm} (5)

$a_0$ denotes the Bohr radius $\hbar^2 (m e^2)^{-1}$. If $(\varepsilon d + \varepsilon_1 D) > \varepsilon^2 a_0$, then $A$ is negative and $\varphi(l)$ reveals the oscillatory dependence. This type of dependence remains for the case of the finite magnitude of bandgap and temperature, as Eqs. (2) should be solved numerically. Thereafter the SHG bias dependence can be obtained straightforwardly. As discussed below, the oscillations in $\varphi(l)$ result in oscillations in $E^{DC}(U_0)$ for the buried $2/m$ layers, which give rise to the SHG signal, accordingly in Eq.(1).

The total SHG intensity is composed of the DC-field-induced dipole and field-independent quadrupole terms (Eq. 1). The relative values of the amplitude and phase of the quadrupole term as well as the flat-bend potential are used as adjustable parameters. The best approximation is shown in Fig. 2 by solid curves with arrows indicating the bias interval where only the first quantized electron sub-band is occupied. Outside this interval the potential dependence of carrier density $n(\varphi)$ should be modified with the account of the occupation of the second sub-band. The shift of the bias region of the model validity is a consequence of the initial bend banding in the silicon substrate. For the thicker structure the adjusted position of the flatband potential is naturally shifted to the higher voltage [7].
In discussing the origin of the oscillations, it is useful to compare the system studied above with the classical plasma. In the latter the spatial distribution of charge density is monotonic, and the screening is well described by the kinetic equation for the electron distribution function. This description by the distribution function requires averaging of fields over the elementary volume which includes a large number of particles (the so-called physically infinitesimal volume). This procedure allows the replacement of the field acting on a particle by the average field \[17\]. Generally, the mean-field potential acting on a particular carrier is formed by all charges in the system except the carrier itself. This exclusion is not important for the 3D Fermi-liquid, in which electron wave functions are delocalized in space. Another situation occurs in the MQW structure where the carriers are confined inside the QW’s. This results in the distinct difference between the acting and average fields. We consider the particular case when the Fermi energy for the 2D electron liquid in QW’s is much smaller than the DC potential. This means that the interaction between electrons of the same QW is negligible in comparison with their interaction with other charges of the system. Consequently, the acting potential for the electrons in \(l\)-th well is the DC-potential formed by external charges all the wells, except the \(l\)-th. On the other hand, the average potential is still contributed by all the wells.

Figure 3 demonstrates the appearance of oscillations in \(\varphi(U_0)\) due to the discussed effect. At small bias the occupation of all sub-bands in all QW’s is zero (thin solid line). Consider the potential \(U_{0,1}\), when electrons accumulate in well 1. The potential which determines the occupation number is purely the DC potential \(\varphi_{el}\) of charges at electrodes from the bias supply (the medium dashed line). This is worth noting again that the charges in well 1 do not contribute to this potential. On the other hand, they do contribute to the complete DC potential \(\varphi\) in the system (medium solid line). The kink in this line is determined by the charge of well 1, \(i.e.\) by \(\varphi_{el}\). For the certain layer thickness this kink can result in the negative \(\varphi\) in well 1. Similarly, at higher potential \(U_{0,2}\) holes are accumulated in well 2, which is illustrated by the solid thick curve. One can note from Figure 3, that \(\varphi\) and \(E^{DC}\) in buried layers appear to be oscillatory functions of bias \(U_0\) in the considered situation.
In conclusion, the DC-electric-field screening in the Si-SiO$_2$ MQW layered structure is studied both experimentally and theoretically. The dependence of DC-electric-field $E_{DC}$ on the applied bias voltage probed in the deep buried Si wells by DC-electric-field-induced SHG shows essentially non-monotonic, oscillatory-like behavior. This behavior which is unexpected for 3D Fermi liquids, indicates the importance of the correlations in electron sub-system for the strongly anisotropic layered system. The observed phenomenon is explained using the theoretical description, which takes into account the strong electron Coulomb localization in the quantum wells due to the high anisotropy of the effective mass. Because of this localization the electron subsystem of MQW differs from the 3D Fermi liquid essentially.

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**Figure Captions**

Fig. 1. Upper panel: the schematic of MQW structure with vicinal substrate, and the configuration of the experiment. Lower panel: Azimuthal dependences of the SHG intensity $I^{2\omega}(\psi)$ for MQW with $d = 1.1 \, nm$ and $D = 5 \, nm$, measured in transmission geometry for parallel polarizations of the fundamental and SH waves, for two biases: ($\bullet$) - +2.6 V, ($\circ$) - +1.26 V. Solid lines are fits to data by Eq.(1) with components of $\chi^{(2Q)}$ and $\chi^{(3D)}$, corresponding to the $2/m$ symmetry, as adjustable parameters. Upper panel: schematics of the sample and the geometry of experiment.

Fig. 2. Bias dependences of the SHG intensity for MQW with $D = 3 \, nm$ (upper panel) and $D = 5 \, nm$ (lower panel). Solid lines are fits by the model. Arrows indicate the region of validity of the model, which is determined by neglecting the second and upper sub-bands.

Fig. 3. Lower panel: the schematic of the spatial distribution of potential $\varphi$ (solid lines) across MQW for three monotonically increased values of $U_0$: $U_{0,0}, U_{0,1}, U_{0,2}$. The second
subscript indicates the number of wells participating in the screening of the external DC-electric field. Directions and relative values of DC-field in buried layers $E^{DC}$ corresponding to these values of $U_0$ are shown by arrows. Dashed lines are $\varphi_{el}$ (see text). The upper panel shows the theoretical dependence of the DC-electric-field in buried $2/m$ layers $E^{DC}$ as a function of the external bias $U_0$ for MQW with $d = 1.1 \, nm$ and $D = 5 \, nm$. 
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$U_0 = 1.26 \text{ V}$

$U_0 = 2.6 \text{ V}$
