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Electric-field-induced second harmonic microscopy

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Electric-field-induced second harmonic microscopy

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Electric-field-induced second harmonic (EFISH) generation was used to image and characterize electric fields near semiconductor interfaces with submicron spatial resolution. Both amplitude and phase of the EFISH light were measured.

Three configurations of the EFISH microscope were demonstrated. We first set up a confocal scanning microscope. Using a normal-incidence configuration, this type of microscope is suitable for measuring the transverse components of the electric field. Thus we use it to image the electric field near the interface of metal-semiconductor Schottky junctions. We acquired EFISH images of the electrodes from -20V to +20V applied bias. Bias-dependent "hot spots" (i.e. tiny region of unusually intense EFISH generation) were detected, showing the possibility of using the EFISH microscope to detect metallization defects in modern semiconductor devices with submicron resolution, which is beyond the capability of most macroscopic electric tests.

In order to measure the vertical components of the electric field in devices like metal-oxide-semiconductor (MOS) capacitors, we changed to an oblique-incidence configuration, and used stationary projection imaging mode. We obtained clear EFISH images of the vertical dc field beneath semitransparent electrodes biased from -8V to +8V, and quantified the spatial variations of normalized second-harmonic generation (SHG) intensity with submicron spatial resolution, showing the capability of EFISH microscope to measure the vertical electric field components microscopically. We discuss the advantages and disadvantages of these two imaging modes.

To completely characterize electric field at the interfaces, we combined the EFISH microscope in stationary projection, oblique-incidence mode with frequency-domain interferometric second-harmonic (FDISH) spectroscopy technique. By measuring EFISH amplitude and phase together, we are able to measure vertical and transverse field components simultaneously. Phase-sensitive EFISH images of MOS capacitors were obtained and analyzed. Micron-space-resolved variations in the EFISH phase-amplitude revealed microscopic inhomogeneities of the vertical and transverse components of the space-charge field that are inaccessible to conventional electric techniques.

Table of Contents

Acknowledgments	v
Abstract	vii
List of Figures	xi
Chapter 1. Introduction	1
1.1 Overview of nonlinear optical microscopy	1
1.2 Electric-field induced second harmonic generation (EFISH) . .	6
Chapter 2. Amplitude EFISH microscopy	12
2.1 Confocal scanning EFISH imaging of coplanar arrays of Metal-Si Schottky contact sample	12
2.1.1 Theoretical background and Electric-field distribution in Metal-Si junction space-charge region	12
2.1.2 Experimental set-up	17
2.1.3 Results and discussion	20
2.2 Stationary projection EFISH imaging of patterned metal-oxide-semiconductor (MOS) sample	22
2.2.1 DC-electric-field spatial distribution in MOS sample and a model of EFISH calculation	22
2.2.2 Experimental set-up	24
2.2.3 Results	26
2.3 Summary	33
Chapter 3. Phase-sensitive EFISH microscopy	34
3.1 Introduction	34
3.2 Experimental set-up	36
3.3 Results and discussion	38

Chapter 4. Conclusions	50
Appendix	55
Appendix 1. Medici simulation for electric field distribution in MOS space-charge region	56
Bibliography	67
Vita	73

List of Figures

1.1	<p>(A) A stereo image pair is synthesized from a stack of 6 cross sections (xy sections) with an axial (z) increment of $3\mu m$. Blue ($380nm \leq \lambda \leq 445nm$) fluorescence excited by two-photon ($630nm$) absorption was detected to record these images of a cluster of fluorescent beads with an confocal scanning microscope. The latex beads are volume-stained with the dye Coumarin 138 and have their measured absorption and emission maxima at $365nm$ and $415nm$, respectively. The data comprise 10 averages for each section with no background subtraction or image enhancement. The total data acquisition time was less than 2 min. (B) The topmost 6 of the images, xy sections, used to synthesize the stereo pair in (A). Scale bar, $50\mu m$.</p>	3
1.2	<p>Two-photon photobleaching in a latex bead is confined to the plane of focus. A section perpendicular to the focal plane (xz section) of a $6\mu m$ latex bead that was continually imaged for 6 min at full excitation power with a small image field ($26\mu m \times 17\mu m$) with the focus slightly below the center of the bead. The dark zone just below the center coincides with the location of the focal plane. The oblong shape of the image of a bead, which is in fact spherical, is due to refractive distortion. The total dose of red laser was $\sim 2mJ/\mu m^2$. The number of photons emitted from each fluorophore before it bleached was about several thousand. Scale bar, $2\mu m$.</p>	4
1.3	<p>(a) linear and (b)-(f) second harmonic images of the magnetic domain structure in (210) oriented garnet film. Input polarization was (b) 0° (c) 10° (d) 35° (e) 90° (f) 145°. (g) Magnetic directions in 4 different domains, as derived from the images of (a)-(f). Symmetry plane m coincides with the side-face plane.</p>	5
1.4	<p>Principle of SHG</p>	7
2.1	<p>An energy diagram of a metal-semiconductor junction at thermal equilibrium. V_{bi} is the same quantity as V_0 used in the text.</p>	13
2.2	<p>Setup for confocal laser-scanning EFISH microscopy.</p>	19
2.3	<p>(a) Images of SHG with scan mirrors stationary and (b) mirror scanning over interdigital pattern of Au electrodes (light regions) with 10V bias between upper and lower array.</p>	19

2.4	Scanning confocal EFISH microscopic images of an interdigital electrode pattern on Si biased at -20V (upper left), and at increasing 5V increments, ending at +20V (lower right). . . .	21
2.5	Experimental set-up for stationary projection EFISH microscopy	25
2.6	Stationary projection EFISH microscopy of a triangle pattern. Bias is starting at 0.5V, and at increasing 0.5V increments, ending at +8V. Then bias is starting over at -0.5V, and at decreasing 0.5V a step, ending at -8V.	27
2.7	Bias-dependence of normalized SHG intensity $I_{SH}(V)/I_{SH}(0)$ along five lines within a $1.3 \times 3\mu m$ area (solid box in panel (a) and inset of (b)) straddling the edge of the electrode.	28
2.8	Spatial variations of $I_{SH}(V)/I_{SH}(0)$ at +5.5V within the $7.5 \times 6\mu m$ area indicated by the dashed box in Fig.2.7(a).	29
2.9	The coherence imaging diagram for the stationary projection imaging mode.	30
2.10	The normalized EFISH intensity distribution in image plane. .	32
3.1	(a) Experimental set-up and (b) microscopic image of the intensity of SH light reflected from 2 semi-transparent Cr electrodes on Si/SiO ₂ with dc bias +8V between the Cr electrodes and an ohmically contacted Al backplane. The upper electrode is $5\mu m$ across, the lower one $20\mu m$ across. Rectangles show portions of the image selected by spectrometer slit for phase-sensitive FDISH measurements. Panel (c) shows same image with dc bias -4V.	39
3.2	Frequency-domain interference fringes at +8V bias from portion of image selected by spectrometer slit positions 1 (left), 2 (middle), 3 (right) shown in Fig.3.1 (b). Note shift of fringes between upper and lower electrodes in position 3. (b) Diagram showing direction of the incident optical field and the DC field at each of the 3 sampled locations.	43
3.3	x and z components of electric field at $V = 8, x = 5.5$, horizontal axis is the depth from Si-Oxide interface.	44
3.4	Line-outs of frequency-domain interference fringes at 6 selected vertical positions for +8V (gray) and -8V (black) bias, showing both spatial and bias-dependent phase shifts.	44
3.5	Fitting curves (dashed line) and experimental fringes (solid line) at vertical electrode location (e) shown in Fig.3.3. There are 204 data points for each curve. DC bias -8V, fitting standard deviation: 115.	45

3.6	Fitting results for +8V and -8V at 6 different locations of the electrodes shown in Fig. 2b. Each stripe is about $0.8 \mu m$ wide, so the space between adjacent locations here is about $0.8 \mu m$. The error bars were decided by changing the parameter to degrade the standard deviation by 5%. The error bars for amplitude are bigger than the error bars for phase.	46
3.7	FDI fringes at different biases: starting at +8V (upper left), and at decreasing 2V a step, ending at -8V (lower right). . . .	48
3.8	Fitting results of phase (a) and amplitude (b) at different bias.	49
4.1	Spatiotemporal evolution of a 10-fs optical pulse propagating through an uncoated near-field fiber probe. The peak intensity of the pulse arrives at the taper entrance ($z = 2\mu m$) at a time $t = 36.7fs$. (a) s polarization: Normalized energy density $ E_x ^2/A_0^2$ at $t = 40fs$ (left) and $t = 52fs$ (right, maximum value of $ E_x ^2$ at fiber exit). The linear gray scale is proportional to $ E_x ^2/A_0^2$. (b) s polarization: Cross section through (a) (right) at two different z coordinates: at the taper exit z_{exit} (left) and at $z_{exit} + 25nm$ (right). (c) and (d) same as (a) and (b), respectively, but for p polarization.	51
4.2	A proposed pump-probe experimental setup to measure EFISH generated by laser wakefields.	53
4.3	Ion-channels for cell membrane.	54
1.1	Electric field contour at V=0	59
1.2	Electric field contour at V=8	60
1.3	z component of electric field at V=8, x=5, horizontal axis is the depth from Si-Oxide interface.	61
1.4	x component of electric field at V=8, x=5, horizontal axis is the depth from Si-Oxide interface.	62
1.5	z component of electric field at V=8, x=5.5, horizontal axis is the depth from Si-Oxide interface.	63
1.6	x component of electric field at V=8, x=5.5, horizontal axis is the depth from Si-Oxide interface.	64
1.7	z component of electric field at V=8, x=4.5, horizontal axis is the depth from Si-Oxide interface.	65
1.8	x component of electric field at V=8, x=4.5, horizontal axis is the depth from Si-Oxide interface.	66

Chapter 1

Introduction

1.1 Overview of nonlinear optical microscopy

Nonlinear optics is the study of phenomena that occur when the response of a material system to an applied optical field depends nonlinearly on the strength of the optical field. Usually this means the polarization $\mathbf{P}$, of a material system depends on the strength $\mathbf{E}$ of the applied optical field in a nonlinear manner [1]:

$$\mathbf{P} = \chi^{(1)} \cdot \mathbf{E} + \chi^{(2)} : \mathbf{E}\mathbf{E} + \chi^{(3)} : \mathbf{E}\mathbf{E}\mathbf{E} + \dots \quad (1.1)$$

where $\chi^{(1)}$ is the linear susceptibility, and $\chi^{(2)}$ and $\chi^{(3)}$ are second- and third-order nonlinear optical susceptibilities, respectively.

Since the properties of the nonlinear interactions differ essentially from those in linear optics, nonlinear optics can add a significant new dimension to microscopy. The first nonlinear optical microscope was reported in 1974 by Hellwarth and Christensen [2], followed by a similar effort in 1978 by Gannaway and Sheppard [3]. After those early efforts, various types of nonlinear optical microscopy has been developed, including coherent anti-stokes Raman scattering microscopy [4], second harmonic microscopy of biological tissue [5]

and adsorbates on various surfaces [6] and, much more recently, dynamic third harmonic microscopy studies of chara plant rhizoids [7].

Among these applications, a few good examples are worth describing in more detail to illustrate the significance of nonlinear optical microscopy.

Watt Webb *et al.* reported a two-photon laser scanning fluorescence microscope [8]. In their experiment, two-photon excitation photobleaching occurred only in a slice about $2\mu m$ thick of a specimen of chromosomes around the focal plane. Such localization shows a distinct advantage of nonlinear two-photon excitation over linear one-photon excitation, where the entire specimen is bleached even when only a single plane is imaged. A much better resolution in the axial or z dimension can be achieved even without a pinhole for a confocal microscope, and it can be used for fast and localized release of biologically active chemicals. Their results are shown in Fig.1.1-Fig.1.2.

This localization can be explained briefly by the quadratic dependence of signal on two-photon excitation intensity, which falls off strongly above and below the focal plane (as $\frac{1}{z^2}$ for large z), while the one-photon excitation intensity does not vary along the z direction.

Th. Rasing *et al.* reported a combined linear and nonlinear magneto-optical microscopy [9]. They used both linear response and magnetization induced optical second harmonic generation (MSHG) to visualize domain structures in epitaxially grown magnetic garnet films. In their experimental configuration, only the magnetization component perpendicular to the film surface

Figure 1.1: (A) A stereo image pair is synthesized from a stack of 6 cross sections (xy sections) with an axial (z) increment of $3\mu m$. Blue ($380nm \leq \lambda \leq 445nm$) fluorescence excited by two-photon ($630nm$) absorption was detected to record these images of a cluster of fluorescent beads with a confocal scanning microscope. The latex beads are volume-stained with the dye Coumarin 138 and have their measured absorption and emission maxima at $365nm$ and $415nm$, respectively. The data comprise 10 averages for each section with no background subtraction or image enhancement. The total data acquisition time was less than 2 min. (B) The topmost 6 of the images, xy sections, used to synthesize the stereo pair in (A). Scale bar, $50\mu m$.

Figure 1.2: Two-photon photobleaching in a latex bead is confined to the plane of focus. A section perpendicular to the focal plane (xz section) of a $6\mu m$ latex bead that was continually imaged for 6 min at full excitation power with a small image field ($26\mu m \times 17\mu m$) with the focus slightly below the center of the bead. The dark zone just below the center coincides with the location of the focal plane. The oblong shape of the image of a bead, which is in fact spherical, is due to refractive distortion. The total dose of red laser was $\sim 2mJ/\mu m^2$. The number of photons emitted from each fluorophore before it bleached was about several thousand. Scale bar, $2\mu m$.

could be directly visualized with the fundamental light (via the linear magneto-optical Faraday effect), whereas in-plane components were probed by MSHG. Thus the combination gives the complete magnetic domain information. Their result is shown in Fig.1.3.

Figure 1.3: (a) linear and (b)-(f) second harmonic images of the magnetic domain structure in (210) oriented garnet film. Input polarization was (b) 0° (c) 10° (d) 35° (e) 90° (f) 145°. (g) Magnetic directions in 4 different domains, as derived from the images of (a)-(f). Symmetry plane m coincides with the side-face plane.

Our work aims to extend nonlinear optical microscopy to the imaging of dc electric fields. Although our work focus exclusively on measuring dc fields in submicron semiconductor devices, the same techniques could in principle be used to imaging electric fields across biological cell membranes, or in laser produced plasmas. Our microscope relies on the nonlinear optical phenomenon

of electric-field-induced second harmonic (EFISH) generation. Thus we now turn to the background of EFISH.

1.2 Electric-field induced second harmonic generation (EFISH)

Optical second harmonic generation (SHG) is a nonlinear optical process that is electric-dipole forbidden in bulk centrosymmetric media such as crystalline Si (Fig.1.4).

Consequently SHG becomes extremely sensitive to the surface and interface, where the structural and/or field discontinuity breaks the centrosymmetry, as well as to bulk regions where electric fields, strain gradients or other factors break centrosymmetry. In particular, locally enhanced SHG in regions pervaded by an electric field $\mathbf{E}^{DC}$ is of special interest, since electric fields play an important role in many semiconductor, electrochemical and plasma systems. This electric-field-induced second-harmonic (EFISH) process can be modeled in the dipole approximation by treating the applied electric field $\mathbf{E}^{DC}$ as an additional electric field contributing to the polarization density $\mathbf{P}^{EFISH}$, along with the laser field $\mathbf{E}(\omega)$. The applied field couples with the driving laser field through a third-order nonlinear susceptibility tensor [10]. The polarization density is

$$P_i^{EFISH}(2\omega, r) = \chi_{ijkl}^{(3)}(2\omega; \omega, \omega, 0) : E_j(\omega, r)E_k(\omega, r)E_l^{DC}(0, r) \quad (1.2)$$

where the $E_j(\omega)E_k(\omega)$ are the components of the laser field, $E_l^{DC}(0)$ is the applied DC field, and $\chi^{(3)}$ is the fourth-rank cubic susceptibility tensor.

Figure 1.4: Principle of SHG

In semiconductors, this model is valid for the region where nonlocality caused by dispersion of the fundamental laser field is small and the screening length of the DC electric field is much larger than a lattice parameter. Usually in the Si transparency region the dispersion is small, and apart from strong inversion and accumulation regimes, the screening length of the DC electric field is on the order of micron, so Eq. (1.2) is adequate for our purpose. More sophisticated expressions for $\mathbf{P}^{EFISH}$ that the effects of nonlocality and short screening length are described in the reference [10].

The EFISH signal originates from the noncentrosymmetric dipole moment of the bulk material across which the applied field drops. In centrosymmetric material, such as Si, the EFISH signal is the only SHG signal generated from bulk dipole moment. Hereafter we denote $\mathbf{P}^{EFISH}$ as $\mathbf{P}^{BD}$.

In real measurements, the total SHG signal also includes contributions from surface dipole polarization and bulk quadrupole polarization. The surface

dipole contribution in the plane wave approximation can be expressed as [12]:

$$\begin{aligned} P_i^S(2\omega, \mathbf{r}) &= \chi_{eff,ijk}^{(2),S}(2\omega) : E_j(\omega, z = 0_+) E_k(\omega, z = 0_+) \\ &= \delta(0_+) \int_0^{z_{surf}} \chi_{ijk}^{(2)}(z') E_j(z', \omega) E_k(z', \omega) dz' \end{aligned} \quad (1.3)$$

where $\chi_{eff}^{(2)}$ is a third-rank effective quadratic susceptibility of the surface layer resulting from the integration of the right hand side of Eq.(1.2), which includes an inhomogeneous susceptibility $\chi^{(2),S}(z')$ of a subsurface layer with thickness z_{surf} , and $z = 0_+$ denotes a position near the interface just inside the semiconductor. The thickness z_{surf} of the subsurface layer is determined by the region where either electron motion remains sensitive to the breakdown of inversion symmetry or where the normal optical electric-field component changes from the outside surface to that inside the surface. Heinz et al. showed the surface contribution to SHG signal is almost entirely from the reconstructed surface layer with a thickness within 2-3 topmost atomic layers [13].

The bulk quadrupole contribution in the plane wave approximation is given by

$$P_i^{BQ}(2\omega, \mathbf{r}) = \chi_{ijkl}^{(2),BQ}(2\omega) : E_j(\omega, \mathbf{r}) i k_k(\omega) E_l(\omega, \mathbf{r}) \quad (1.4)$$

where $\chi^{(2),BQ}$ is a fourth-rank tensor which represents the quadrupole contribution to the quadratic nonlinear susceptibility, and $k(\omega)$ is the wave vector of the fundamental radiation in the semiconductor.

The total quadratic polarization is thus given by the summation of these three contributions:

$$\mathbf{P}^{(2)}(2\omega, \mathbf{r}) = \mathbf{P}^S(2\omega, \mathbf{r}) + \mathbf{P}^{BQ}(2\omega, \mathbf{r}) + \mathbf{P}^{BD}(2\omega, \mathbf{r}) \quad (1.5)$$

This SH polarization is not directly observed. We observe the SH that it radiates. The SH electromagnetic field

$$\mathbf{E}(\mathbf{R}, t) = \mathbf{E}(\mathbf{R}, 2\omega) \exp[i(2\omega t - \mathbf{k}_{2\omega} \mathbf{R})] \quad (1.6)$$

where $\mathbf{E}(\mathbf{R}, 2\omega)$ is the amplitude of the SH field at the point of observation $\mathbf{R}$, can be found by solving the inhomogeneous wave equation for propagation of the SH wave with $\mathbf{P}^{(2)}$ as the source term [11]. The solution can be written formally in terms of the tensorial Green function $\overleftrightarrow{G}(\mathbf{R}, \mathbf{r}', 2\omega)$, which is defined to be the solution of the wave equation with a point source at $\mathbf{r}'$. Then the SH field $\mathbf{E}(2\omega)$ in the point of $\mathbf{R}$ is given by

$$\mathbf{E}(\mathbf{R}, 2\omega) = \int \overleftrightarrow{G}(\mathbf{R}, \mathbf{r}', 2\omega) \mathbf{P}^{(2)}(\mathbf{r}', 2\omega) d\mathbf{r}' \quad (1.7)$$

where integration is taken over the bulk of the semiconductor.

If translational symmetry in the interface plane is present, the DC-field-induced part of the SH field is given by

$$\mathbf{E}^{BD}(\mathbf{R}, 2\omega) = F_{2\omega} F_{\omega}^2 \chi_{eff}^{BD} I_{\omega} \mathbf{p} \exp(i\mathbf{k}_{2\omega} \mathbf{R}) \int_0^{+\infty} E_0(z') \exp[i(k_{2\omega,z} + 2k_{\omega,z})z'] dz' \quad (1.8)$$

where $\mathbf{k}_{2\omega}$ is the SH wave vector, the scalar factor χ_{eff}^{BD} is a linear combination of components of $\chi^{(3),BD}$ which depends on the experimental geometry, I_{ω} is the intensity of the fundamental radiation, and $k_{\omega,z}$ and $k_{2\omega,z}$ are the normal wave-vector components of the fundamental and SHG radiation, respectively, in the semiconductor. The unit vector $\mathbf{p}$ defines the polarization of the EFISH

field, and F_ω and $F_{2\omega}$ are the transmission factors which include Fresnel coefficients and, for an oxidized interface, a correction for multiple reflections in the SiO_2 at both ω and 2ω . Equation (1.8) properly takes into account retardation, the penetration depth of the fundamental wave, the escape length of the SH wave, and multiple reflection interference effects in the oxide layer.

Historically, EFISH was discovered in 1967 by Lee *et al.*[14] at Si- and Ag-electrolyte interfaces in electrochemical cells. The 1981 discovery of surface-enhanced SHG by Shen *et al.*[15] rejuvenated interest in this effect. Since 1984, EFISH has been systematically studied at Si(111)-electrolyte interfaces [16], and to a lesser extent at other semiconductor-electrolyte interfaces: $\text{Cd}_3\text{P}_2(111)$ [17], $\text{CdIn}_2\text{S}_4(111)$ [18], $\text{GaN}(001)$ [19], and TiO_2 [20]. The dc electric field was applied electrochemically in these experiments, and showed the limitation of interface electrochemical reactions, such as oxidation of a Si surface at anodic potential. To overcome this difficulty, EFISH generation studies were extended to systems with bias applied directly by gate electrodes or Schottky-type metal-semiconductor strip contacts. Since 1994, Aktsipetrov *et al.* studied Si- SiO_2 MOS structures with bias applied by a ring metal [21] and a semitransparent Cr gate electrode [22]. Ohlhoff *et al.* studied the field dynamics at Schottky-type contact metal-semiconductor interfaces, utilizing EFISH generation to monitor the internal electric field distribution in Si millimeter-wave circuits [23]. Dadap *et al.* have done similar work, focusing on the possibility of mapping the vector structure of electric fields in silicon using EFISH with a homodyne detection technique [24].

The potential to use EFISH to map microscopical spatial variations of electric fields remains largely untapped [25]. Indeed the electric fields at metal-semiconductor junctions, biological membranes, fusion plasmas and other contexts are often strongly inhomogeneous over micron or submicron scale lengths. Thus it is natural to think about utilizing the phenomenon of EFISH to observe and measure electrical fields in those systems on a microscopic scale. In this work, we demonstrate EFISH microscopy of electric fields in semiconductor devices, taking advantage of both amplitude and phase measurements of the EFISH fields.

Chapter 2

Amplitude EFISH microscopy

2.1 Confocal scanning EFISH imaging of coplanar arrays of Metal-Si Schottky contact sample

2.1.1 Theoretical background and Electric-field distribution in Metal-Si junction space-charge region

Metal-semiconductor junctions are of substantial interest to the microelectronics and photonics community as well as MOS structure. We turn now to an examination of the steady-state electrical characteristics of metal-semiconductor junctions.

We consider the details of this problem for a junction between Au and n -type silicon. The junction and all relevant quantities are shown in Fig.2.1.

Here we are presenting a 1-dimensional analysis for simplicity. Near the edge of electrodes, transverse components of electric fields also become important. This situation is discussed further in Chapter 3.

Imagine a metal and a n -type silicon as a large number of electrons moving in a lattice of positively-charged atoms. When these two materials are brought into immediate contact, electrons diffuse across the interface in both directions. Since the Fermi energy level of n -type silicon is higher than the Fermi energy level of Au, more electrons go into metal than electrons go into

Figure 2.1: An energy diagram of a metal-semiconductor junction at thermal equilibrium. V_{bi} is the same quantity as V_0 used in the text.

Si. Very quickly, there forms a region in Si near the interface which has an excess of positive charge. This region is called depletion region or space-charge region (SCR), and has a characteristic width W which is dependent on doping concentration and material properties and, as we shall see, on any external applied bias.

In SCR, there is an electric field E between the depleted positive ions and the electrons at the metal interface. When this electric field screens the electrons in the bulk of Si from moving into SCR, a thermal equilibrium is achieved. At thermal equilibrium, the chemical potential throughout the sample, including across the junction, must be constant, i.e., there must be no net charge transfer across the junction. There are only two mechanisms for charge transfer in the metal-semiconductor junction: carrier diffusion and electric field drift. These must therefore exactly cancel at thermal equilibrium, as expressed by the following equation for current flow across the junction:

$$J_n = q\mu_n(nE + \frac{\partial n}{\partial x}) = 0 \quad (2.1)$$

where q is charge, μ_n is the carrier mobility, E is the electric field present at thermal equilibrium, usually referred to as the contact electric field, nE represents field induced carrier drift, and the $\partial n/\partial x$ term represents concentration gradient-induced diffusion [26]. Note that the chemical potential can be related to the Fermi level E_F simply by taking into account temperature variation of the chemical potential, which can be expressed as

$$\mu = E_F \left[1 - \left(\frac{1}{12}\right)\left(\frac{\pi}{\beta\mu}\right)^2 + \left(\frac{139}{2880}\right)\left(\frac{\pi}{\beta\mu}\right)^4 + \dots \right] \quad (2.2)$$

where $\beta = 1/kT$. Therefore, for $kT \ll \mu$, $\mu \sim E_F$. Hence, the requirement that the chemical potential is constant throughout the sample, including the junction, leads to the requirement that the Fermi level E_F be constant throughout the sample, including the junction. Thus the Fermi level in Si and the Fermi level in Au must match at the junction.

The requirement of no net current across the junction leads to the existence of an electric field as described earlier. Equation(2.2) can be rearranged to give

$$qE = \frac{kT}{q} \frac{1}{n} \frac{\partial n}{\partial x} \quad (2.3)$$

Associated with this field is the contact potential V_0 , so that $E = -\frac{\partial V_0}{\partial x}$. We can thus write

$$-q \frac{\partial V_0}{\partial x} = \frac{kT}{q} \frac{1}{n} \frac{\partial n}{\partial x} \quad (2.4)$$

and, integrating each side over appropriate limits, we obtain

$$V_0 = \frac{kT}{q} \ln \frac{n_m}{n_s} \quad (2.5)$$

where n_m is the charge carrier concentration in the metal side and n_s is the charge carrier concentration in the semiconductor side. Thus we see that the condition of no net current across the junction leads to the presence of a contact potential. This potential maintains the condition of no net current across the junction and allows charge neutrality to be preserved throughout the entire sample.

We proceed now to examine the electric field distribution in the SCR.

We begin with Poisson's equations

$$-\frac{\partial^2 V}{\partial x^2} = \frac{\rho}{\epsilon} \quad (2.6)$$

Since the field region is mainly depleted of charged carriers, the space charge will be due mainly to the ionized donor atoms in the semiconductor, and will have a magnitude equal to qN_d , where N_d is the number of ions contributing to the charge. We can therefore write

$$-\frac{\partial^2 V}{\partial x^2} = \frac{qN_d}{\epsilon} \quad (2.7)$$

Integrating this equation once, remembering that $E = -\frac{\partial V}{\partial x}$, and that the field falls to zero at $x = W$, then we obtain the electric field distribution:

$$E(x) = -\frac{\partial V}{\partial x} = \frac{qN_d}{\epsilon}(x - W) \quad (2.8)$$

where we have defined W as the point of zero potential. The width of the depletion region can be obtained by evaluating this last equation at any x . Evaluating at $x = 0$, we obtain

$$W = \left(\frac{2\epsilon}{qN_d} V_0 \right)^{1/2} \quad (2.9)$$

where V_0 is just the contact potential as derived earlier.

Applying a negative external bias to the metal will tend to lengthen the SCR in the semiconductor by driving conduction electrons (the same electrons that provide the field-screening) further into the bulk of the semiconductor. Likewise, applying a positive external bias to the metal will tend to shrink the

SCR by drawing conduction electrons out of the semiconductor and over the potential barrier into the metal. This behavior can be understood simply by adding a term V_{ap} underneath the squareroot in Eq.(2.9):

$$W = \left[\frac{2\epsilon}{qN_d}(V_0 - V_{ap}) \right]^{1/2} \quad (2.10)$$

Note that we have neglected the condition of majority-carrier distribution tails, which will add a term of order kT/q inside the Eq.(2.10).

To summarize, Eq.(2.10) describes the width of the SCR formed at the junction between a metal and an n -type Si. N_d is the doping level, V_0 is the contact potential, V_{ap} is the applied bias, q is the electronic charge, and ϵ is the dielectric function of the semiconductor. An expression for the contact potential V_0 may be obtained by considering the quantities in Fig.2.1 (in this figure, V_{bi} is the built-in potential, the same quantity as V_0). Evidently,

$$V_0 = \phi_m - (\chi + E_F - E_C) \quad (2.11)$$

where ϕ_m is the metal's work function, χ is the semiconductor's electron affinity, and E_F and E_C are the Fermi level and conduction level, respectively.

2.1.2 Experimental set-up

To implement EFISH microscopy, a confocal laser-scanning EFISH Microscope was first set up. The diagram of the set up is shown in Fig.2.2. Such systems in various configurations form the basis for almost all commercially available confocal scanning optical microscopes (CSOMs)[27]. The scanning

system works as follows. The laser beam is incident on the first galvanometer-mounted scan mirror which swivels about an axis parallel to the mirror surface. A system of two lenses make a one-to-one image of the scanned spot onto the second scan mirror, which swivels about axis perpendicular to that of the first mirror. These mirrors are driven by two scanner drivers (GSI-Lumonics models AX-200 and CX-660) which have ramp waveforms as inputs. A second system of lenses makes another unity magnification telescope imaged this scanned point into the back pupil plane of a 40X objective lens. This scanned spot is stationary, but the light beam actually scans around this point, so that the focus spot on the sample moves with the scanners. Thus the spot in the back pupil plane of the objective doesn't move, allowing the full numerical aperture of the objective to be used at all times. The fundamental beam was focused by the objective at normal incidence onto the sample to a near-diffraction-limited spot radius $w_0 \sim 1\mu m$ (Fig.2.3a) . The focused spot raster scanned with the galvo-mirrors over a $30 \times 30\mu m$ area on the stationary sample. The same microscope objective then collected the generated SH light in an approximately confocal geometry and imaged it onto the CCD camera, where it traced out a SHG image of the scanned sample area. Fig.2.3b shows a typical scanned image of the total SHG signal from interdigital Al electrodes (light regions) on Si (darker regions). Adequate signals were obtained with total scan time $\sim 100s$.

Figure 2.2: Setup for confocal laser-scanning EFISH microscopy.

Figure 2.3: (a) Images of SHG with scan mirrors stationary and (b) mirror scanning over interdigital pattern of Au electrodes (light regions) with 10V bias between upper and lower array.

2.1.3 Results and discussion

Fig.2.4 shows differential confocal scanning EFISH images of an interdigital, coplanar pattern of Al electrodes (see inset), with bias from -20 to +20 V applied between the upper and lower arrays. For each image in Fig.2.4, the scanned SHG signal from the un-biased structure has been subtracted from the total scanned signal (which in each case resembles Fig.2.3b.), leaving only bias-dependent features. In the upper left image (-20 V) the electrodes are outlined by the EFISH signal from the $\sim \mu m$ -wide SCR. As the bias increases, the differential image fades, disappearing near 0 V, then recovers at positive bias. For the images in Fig.2.4, neither the electrode edges nor the linearly-polarized incident field were oriented along the crystalline [100] or [010] axes of the Si substrate. Consequently EFISH signals were generated with comparable efficiency from dc fields of all orientations via the various non-zero tensor components of $\chi^{(3)}$. Because of normal incidence, the background signal $E_{FI}(2\omega)$ from the Si substrate is much weaker than for the oblique-incidence data. Consequently an enhanced signal $|E_{TOT}(2\omega)|^2 \sim |E_{FD}(\omega)|^2$ is observed from the SCR at both negative and positive biases.

Several bias-dependent “hot spots” are clearly evident in the image of Fig.2.4. Two of the most prominent are marked by arrows in the Fig.2.4. These spots result from local whiskering, or other damage, in the metallization. Such high-field defects represent potential failure points in integrated circuits, and are not easily detected by macroscopic electrical tests. Their straightforward detection in these images may therefore be an important practical application

Figure 2.4: Scanning confocal EFISH microscopic images of an interdigital electrode pattern on Si biased at -20V (upper left), and at increasing 5V increments, ending at +20V (lower right).

of EFISH microcopy.

One of the major drawbacks of confocal scanning EFISH microscopy is that it is based on focusing a femtosecond laser pulse very tightly, resulting in high light intensity on the sample. As a result, the dc-field is partly screened by laser-induced carriers. Laser induced electron-hole pairs move in such a way that the applied electric field decreases. Details of dynamics of photoinduced field screening can be found elsewhere [28]. Also, tight focus of the laser beam may cause heat damage to the sample. So the power of the fundamental laser should be carefully controlled.

2.2 Stationary projection EFISH imaging of patterned metal-oxide-semiconductor (MOS) sample

2.2.1 DC-electric-field spatial distribution in MOS sample and a model of EFISH calculation

(1) DC field spatial distribution

According to equation(1.8), the spatial distribution $E_0(z)$ in the space-charge-region (SCR) will decide the EFISH signal. Again, we use a 1-dimensional analysis for simplicity. In the planar MOS configuration, the spatial distribution of the electrostatic potential $\varphi(z)$ is governed by Poisson equation:

$$\frac{\partial}{\partial z} \left(\epsilon \frac{\partial}{\partial z} \varphi \right) = -4\pi n \quad (2.12)$$

where $\epsilon = \epsilon_{SC}(\epsilon_d)$ is the static dielectric constant of the semiconductor and $n = n(z)$ is the space-charge density.

The boundary conditions are:

$$\varphi(+\infty) = \mu \quad (2.13)$$

$$\varphi(-D) = \mu + U \quad (2.14)$$

where μ is the chemical potential of the semiconductor, D is the thickness of the oxide layer, and U is the applied bias.

If the spatial variation of potential φ is slow enough, which is true except at very high biases, $n(z)$ depends only on the potential φ at point z , i.e., $n(\varphi) = n[\varphi(z)]$. Since the variable z does not enter into equation (2.12) explicitly, and the charge density n depends on z via $\varphi(z)$, the equation (2.12) has the first integral

$$E_0^2(\varphi) = \frac{8\pi}{\varepsilon} \int_{\varphi}^{\mu} n(\varphi') d\varphi' \quad (2.15)$$

After we find the space-charge density distribution, the spatial DC field and potential distribution can be found from the first integral $E_0(\varphi)$ of the Poisson equation by integrating $z = \int_{\varphi}^{\varphi_{int} + \mu} [E_0(\varphi')]^{-1} d\varphi'$, with $\varphi_{int} = \varphi(z = 0_+) - \mu$.

(2) EFISH calculation

After we got the DC electric field distribution, we can substitute $E_0(z)$ into equation (1.8), and get the integral

$$I(U) \equiv I_1 + iI_2 = \int_0^{+\infty} E_0(z) \exp[i(k_{2\omega,z} + 2k_{\omega,z})z] dz \quad (2.16)$$

This integral can be numerically evaluated.

2.2.2 Experimental set-up

Samples were prepared on nominally undoped Si substrates ($700\mu m$ thick, resistivity $8 \times 10^3 \Omega cm$). Patterned metal-oxide-semiconductor (MOS) samples were prepared by: (1) growing a $90\text{\AA}$ thermal oxide; (2) patterning an array of several- μm wide gate electrodes using standard photo-lithographic techniques; (3) evaporating a semitransparent $64\text{\AA}$ Cr film onto the oxide, and (4) lifting off the pattern with Acetone.. A uniform $3000\text{\AA}$ Al electrode was evaporated onto the backside of the substrate. Bias was applied between Cr and Al electrodes. In these samples, fields were oriented primarily perpendicular to the surface, and were located beneath, and probed through, the semitransparent Cr electrodes. Cr generates much weaker SH signals than Si, and thus does not obscure EFISH generation from the underlying space-charge region (SCR) significantly [29].

Fig.2.5 illustrates the set-up for stationary projection EFISH microscopy. The fundamental laser beam was focused at $f/12.5$, 45° incidence, p-polarization to a spot radius $\sim 10\mu m$ on the sample surface. SH light ($\lambda_{SH} = 0.4\mu m$) was generated from the sample collinearly with reflected fundamental light, spectrally filtered, then imaged with about $140\times$ magnification onto a charge-coupled device (CCD) with a microscopic objective. Focusing was optimized by imaging the electrode pattern using an incident test beam at λ_{SH} . Depth-of-field ($\sim 50\mu m$) was sufficient that the entire illuminated region was in focus. The stationary beam method acquires images in parallel with no moving parts, and achieves essentially diffraction-limited lateral resolution. However, spot

Figure 2.5: Experimental set-up for stationary projection EFISH microscopy

radius (and thus field of view) was usually limited to $\sim 30\mu m$ by low count rates (0.1 to 1 count/pixel/s over the illuminated area of metallized silicon), which necessitated data acquisition times of 10 to 100 s to obtain high quality EFISH images.

2.2.3 Results

Fig.2.6 shows the projection EFISH images acquired with 150 seconds averaging time near the tip of a pointed Cr gate electrode biased from -8V to 8V, 0.5V a step. The enhanced SHG from the SCR under the positively-biased electrode is clearly evident. Similarly decreased SHG intensity relative to the adjacent oxidized Si is also evident under the negatively biased electrode. Near the sharp edges of the electrodes, interference fringes with period $\sim \lambda_{SH}$ are clearly visible. These result from diffraction of part of the generated SH light out of the collection aperture of the imaging lens by the sharp edge of the electrode, and are a common feature of microscopy with coherent light [30].

Fig.2.7 shows the bias-dependent, normalized SHG intensity at 5 different perpendicular distances $-0.66 < x < +0.66\mu m$ from the edge ($x = 0$) of the electrode at the location indicated by the solid box in panel (a). For each curve, signal from a line of ~ 10 pixels parallel to the edge of the electrode was averaged, in order to suppress artifacts caused by pixel-to-pixel variations in the detector. Normalizing $I_{SH}(x, V)$ to $I_{SH}(x, V = 0)$ removes the dependence of the signal on the Gaussian intensity profile of the focused laser spot, leaving variations with bias V and positions x on the sample. Curves 1 and 2 were

Figure 2.6: Stationary projection EFISH microscopy of a triangle pattern. Bias is starting at 0.5V, and at increasing 0.5V increments, ending at +8V. Then bias is starting over at -0.5V, and at decreasing 0.5V a step, ending at -8V.

acquired from beneath the electrode, and show nearly identical dependence on bias: enhanced SHG at $V > 0$, reduced at $V < 0$. In this area, the total SH field $E_{TOT}(2\omega, \vec{r}) = E_{FD}(2\omega, \vec{r}) + E_{FI}(2\omega, \vec{r})$ is a coherent sum of the EFISH (i.e. FD) field and the field-independent (FI) background SH field. The enhanced signal at positive bias results from constructive interference between $E_{FD}(2\omega)$ and $E_{FI}(2\omega)$, and the reduced signal at negative bias results from destructive interference between $E_{FD}(2\omega)$ and $E_{FI}(2\omega)$ [10]. The saturation of the signal $|E_{TOT}(2\omega)|^2$, and thus $E_{FD}(2\omega)$, at large negative bias is consistent with the low (unintentional) doping, and thus lack of accumulation regime, in our substrate.

Figure 2.7: Bias-dependence of normalized SHG intensity $I_{SH}(V)/I_{SH}(0)$ along five lines within a $1.3 \times 3\ \mu m$ area (solid box in panel (a) and inset of (b)) straddling the edge of the electrode.

Curve 3 was acquired at the edge of the electrode, and clearly shows a weaker bias-dependence than curves 1 and 2. Curve 4 was acquired at the first dark fringe, and shows a still weaker bias dependence of reversed sign. Curve 5 was acquired at the first bright fringe, where bias dependence has nearly disappeared. The scale length $0.5\mu m$ over which bias-dependence disappears is consistent with the length W of the depletion region for nominally undoped Si.

Figure 2.8: Spatial variations of $I_{SH}(V)/I_{SH}(0)$ at $+5.5V$ within the $7.5 \times 6\mu m$ area indicated by the dashed box in Fig.2.7(a).

Fig.2.8. shows the spatial variations in $I_{SH}(\vec{r}, V = +8V)/I_{SH}(\vec{r}, V = 0V)$ in the area under the Cr electrode indicated by the dashed box in 2.8(a). The signal is quite uniform, as expected, but $\pm 10\%$ variations are observed.

These variations may result from μm -scale variations in defect density at the Si/SiO₂ interface or in dopant density, which would alter the distribution $E_0(z)$ of the dc field in the SCR, and thus $E_{FD}(2\omega)$. Such fine-scale variations are a unique output of EFISH microscopy that cannot be observed in macroscopic electrical tests.

Coherent artifacts appear when sharp boundaries are present in the image area. In Fig.2.6. the interference fringes are clearly seen at the edge of the electrode. This can be explained in detail using coherent imaging theory [30].

According to the experiment configuration, the coherence imaging diagram can be drawn like fig.2.9.

Figure 2.9: The coherence imaging diagram for the stationary projection imaging mode.

Coherence illumination Fraunhofer diffraction:

$$V(x', y') = C_1 C_2 \int \int_A \int \int_B F(x, y) I(V) \exp \left\{ -i \frac{k}{f} \left[\left(x + \frac{f}{D'} x' \right) \xi + \left(y + \frac{f}{D'} y' \right) \eta \right] \right\} dx dy d\xi d\eta \quad (2.17)$$

Here we can use 1-D approximation:

$$V(x') = C_1 C_2 \int \int F(x) I(V) \exp \left\{ -i \frac{k}{f} \left[\left(x + \frac{f}{D'} x' \right) \xi \right] \right\} dx d\xi \quad (2.18)$$

Assume the object transmission function $F(x)$ is constant:

$$V(x') = C_3 \int_{-b}^b \int_{-w}^w I(V) \exp \left\{ -i \frac{k}{f} \left[\left(x + \frac{f}{D'} x' \right) \xi \right] \right\} dx d\xi \quad (2.19)$$

Denote $\frac{k}{f} \left[\left(x + \frac{f}{D'} x' \right) \right]$ by u :

$$\begin{aligned} V(x') &= C_3 \int_{-b}^b \int_{-w}^w I(V) \exp(-iu\xi) dx d\xi \\ &= C_3 \int_{\frac{k}{f}(-b + \frac{f}{D'} x')}^{\frac{k}{f}(b + \frac{f}{D'} x')} 2I(V) \frac{f \sin(uw)}{k u} du \end{aligned} \quad (2.20)$$

Denote uw by v :

$$\begin{aligned} V(x') &= C_4 \int_{w \frac{k}{f}(-b + \frac{f}{D'} x')}^{w \frac{k}{f}(b + \frac{f}{D'} x')} I(V) \frac{\sin(v)}{v} dv \\ &= C_4 I(V) \left\{ Si \left[w \frac{k}{f} \left(b + \frac{f}{D'} x' \right) \right] - Si \left[w \frac{k}{f} \left(-b + \frac{f}{D'} x' \right) \right] \right\} \end{aligned} \quad (2.21)$$

Use parameters in units of micron:

Half width of the object: $b = 20$

Half width of the aperture of the back focal plane: $w = 2500$

Wave number: $k = \frac{2\pi}{0.4}$

Focal length: $f = 5100$

Distance between the back focal plane and the imaging plane: $D' = 800000$

Figure 2.10: The normalized EFISH intensity distribution in image plane.

The interference fringes near the edge are present.

2.3 Summary

Confocal scanning EFISH microscopy uses a tightly focused beam and thus yields higher signal and the potential for modulation microscopy. However, it requires a more complicated scanning mirror configuration, yields somewhat lower spatial resolution, and tends to screen the dc fields by carrier generation. Stationary projection imaging yields diffraction-limited lateral spatial resolution, involves no moving parts, and detects μm -scale lateral field variations related to Si/SiO₂ interface characteristics such as defect and doping density. However, the images offer a limited field of view, are complicated by coherent artifacts and require acquisition times too long for modulation techniques. Both methods can be improved by incorporating techniques developed in non-microscopic contexts. Introducing a strong, independent SH local oscillator (homodyne detection) that coincides spatially and temporally with the EFISH beam generates a signal that is linear in dc field E_0 and can be calibrated straightforwardly to yield absolute field strengths [31]. Careful control of incident and EFISH polarizations with respect to crystalline axes will enable microscopic mapping of selected vector components of $\mathbf{E}_0$ [24].

Chapter 3

Phase-sensitive EFISH microscopy

3.1 Introduction

Previous EFISH microscopy has measured only amplitude of the SH light. To completely characterize the dc field, we must also measure phase variations of the SH light within the image. Frequency-domain interferometric second-harmonic (FDISH) spectroscopy allows the acquisition of full phase spectra with only one measurement [43]. Combining FDISH with EFISH microscopy enables the phase of the SH field to be measured with submicron spatial resolution. The principle of frequency domain interferometry is that two coherent pulses delayed in time can interfere with each other after being spectrally dispersed by the grating in spectrometer, to yield fringes at the detection plane. Since the delay is related to the frequency, the fringes are actually in frequency domain. These two pulses must be combined enter the spectrometer collinearly and sequentially. The array detector then records the intensity

$$\begin{aligned} I(\omega, \tau) &= \left| \frac{1}{2\pi} \int_{-\infty}^{\infty} [E_{sig} + E_{ref}(t - \tau)] e^{i\omega t} dt \right|^2 \\ &= |E_{sig}(\omega) + E_{ref}(\omega) e^{-i\omega\tau}|^2 \\ &= |E_{sig}(\omega)|^2 + |E_{ref}(\omega)|^2 + |E_{sig}(\omega)| |E_{ref}(\omega)| \cos(\Delta\phi(\omega) + \omega\tau) \end{aligned}$$

(3.1)

The complete analysis technique for extracting a phase difference from frequency domain interferograms is well established [44]. First, because a grating spectrometer with a planar array detector measures on intervals that are nearly equally spaced in wavelength, the spectra must be splined to a constant frequency spacing. Splining to a reduced number of data points is a convenient way to make the array of data conform to the 2^n size used for Fast Fourier Transforms (FFT). The splined spectrum is inverse Fourier transformed into the time domain where, because the spectral measurement is necessarily real, a symmetric function is produced with peaks at time $t = 0$ and $t = \pm\tau$. A windowing function is used to select only one of the delayed peaks setting all other parts of the time domain data to zero. The proper peak must be selected which represents the correct sequential order for the signal and reference. This array is transformed back to the frequency domain where the phase difference between the two pulses can be calculated as $\phi = \arctan(\frac{\Im E}{\Re E})$. The phase can then be unwrapped by an algorithm that adds or subtracts 2π to keep discontinuities in the data to less than π . The resulting spectral phase will be primarily a linear function of frequency with the slope equal to the delay, τ . If this delay is known it can be subtracted from the end result or eliminated by translating the time domain array such that the windowed peak is shifted to $t = 0$. This will leave a spectral phase that is the true phase difference between the two pulses.

An alternative fitting technique to extract the phase difference can also

be used. In this work, we used Origin 6 nonlinear curve fitting procedure, which defines a least squares χ^2 merit function and determines best-fit parameters by its minimization. With nonlinear dependences, the minimization must proceed iteratively. We use Levenberg-Marquardt (LM) algorithm for the nonlinear regression, given trial values for the parameters. The details of L-M algorithm can be found in reference [32].

3.2 Experimental set-up

Fig.3.1(a) shows the experimental set-up. As before, MOS sample consisted of patterned semi-transparent gate Cr electrodes (30 Å thick) deposited on Si(100)/SiO₂ with 90 Å thermal oxide by conventional photolithography. DC fields were again located beneath, and probed through, the semi-transparent Cr electrode, which generate negligible SH light themselves, by focusing 25fs, 5 nJ pulses from a mode-locked Ti:S laser at f/25, 45° incidence, p-pol. onto the sample, then imaging the p-pol SH light onto a charge-coupled device (CCD) with a microscope objective. Figs 3.1(b) and (c) show typical 140×-magnified amplitude-only EFISH images of two adjacent Cr electrodes, recorded with CCD at position 1. Brighter EFISH generation is evident beneath the Cr electrode biased at +8V with respect to a 3000 Å Al electrode on the back of the substrate (Fig.3.1b) than beneath the same electrode -4V (Fig.3.1c). To measure SH phase, we inserted a transparent reference $\chi^{(2)}$ film (TCV) before the sample. A SH reference pulse was generated, then delayed by $\tau \sim 1$ ps from the fundamental pulse by group delay in the ~ 1 mm thick glass

substrate of the reference film. The reference SH pulse then reflected from the sample, and was imaged together with the sample SH pulse onto the slit of an imaging spectrometer at position 2. The reference and sample SH pulses propagated collinearly, and interacted with identical optical elements. The slit selected y -directed lineouts of the image, such as rectangles (1), (2) and (3) on Fig.3.1b. By translating the sample along the x -direction, data from different selected lines was acquired serially. Diffraction-limited resolution was retained along the slit, but at each y -position, SH light was spectrally dispersed onto the CCD, which recorded the frequency-domain interference pattern [33]:

$$\begin{aligned}
I(2\omega, \tau) &= \left| E_{ref} + E_{FD}e^{i(\phi_{FD}+\omega\tau)} + E_{FI}e^{i(\phi_{FI}+\omega\tau)} \right|^2 - \left| E_{ref} + E_{FI}e^{i(\phi_{FI}+\omega\tau)} \right|^2 \\
&= 2E_{ref}E_{FD} \cos(\phi_{FD} + \omega\tau) + 2E_{FI}E_{FD} \cos(\phi_{FD} - \phi_{FI}) + |E_{FD}|^2
\end{aligned} \tag{3.2}$$

where E_{ref} is the reference SH field, taken to be purely real, $E_{FI}e^{i\phi_{FI}}$ and $E_{FD}e^{i\phi_{FD}}$ are the dc-field -independent and dc-field-dependent parts, respectively, of the SH field from the sample. The term $\left| E_{ref} + E_{FI}e^{i(\phi_{FI}+\omega\tau)} \right|^2$ represents a FDI image at flatband bias that was subtracted from each image.

A unique advantage of the phase-sensitive EFISH microscope over the amplitude EFISH microscope is that the pure EFISH signal can be isolated from all field-independent SHG. For amplitude-only EFISH microscopy, even with the background subtraction (Eq.(3.2)), since there is a cross term $2E_{FI}E_{FD} \times \cos(\phi_{FD} - \phi_{FI})$, we still can not determine E_{FD} and E_{FI} . But

if we can acquire the phase information ϕ_{FD} and ϕ_{FI} through phase-sensitive EFISH microscope, we are able to completely distinguish E_{FD} and E_{FI} .

The FDISH spectroscopy is actually a variation of “homodyne” technique described by Dadap *et al.*[45]. They used a normal-incidence configuration and time-domain interference technique to linearize the SH intensity dependence on the bias and increase the signal-noise ratio. With a little modification their setup can also be used to measure the phase of SH light and therefore isolate the EFISH signal from the background field-independent SH signal. However, the FDISH technique involves no moving parts, can be used for both normal-incidence and oblique-incidence configurations, and more importantly, can take the whole phase spectra with only one measurement, saving a lot of time and trouble.

3.3 Results and discussion

Fig.3.2 (a) show FD interferograms collected from sampling slits (1) through (3), corresponding to $1.4\mu m$ wide strips of the sample located at the left edge (1), near the center (2) and the right edge (3) of the lower electrode, respectively. Pulses were incident in the $x - z$ plane from the $-x$ direction, as shown in Fig.3.2 (b). High-contrast fringes were obtained in ~ 1 minute. Higher spatial resolution along x could be achieved by using a narrower slit, at the expense of longer data acquisition time. In slit positions (1) and (2), the fringes from the thin and wide electrodes are mutually aligned. However, in position (3), the fringes from the thin electrode shift significantly with respect

Figure 3.1: (a) Experimental set-up and (b) microscopic image of the intensity of SH light reflected from 2 semi-transparent Cr electrodes on Si/SiO₂ with dc bias +8V between the Cr electrodes and an ohmically contacted Al backplane. The upper electrode is 5 μ m across, the lower one 20 μ m across. Rectangles show portions of the image selected by spectrometer slit for phase-sensitive FDISH measurements. Panel (c) shows same image with dc bias -4V.

to those from the wide electrode. This demonstrates a phase shift of EFISH light generated from the beneath the right edge of the lower electrode relative to EFISH generated beneath its center and left edge, only a few microns away, and beneath the wide electrode. EFISH amplitude, by contrast, varies only slightly between positions (1) and (3).

The phase shift at position (3) can be understood with the help of Fig.3.2 (b). In the center of the electrode (position (2)), only a vertical DC field component E_z^0 is present, so the p-polarized EFISH field is

$$E_{FD}^p(E_z^0) = F_{2\omega}^p \{ [\chi_{xzzz}^{(3)} E_z^\omega E_x^\omega + \chi_{xxzz}^{(3)} E_x^\omega E_z^\omega] \cos \theta_T^\omega + [\chi_{zzzz}^{(3)} (E_z^\omega)^2 + \chi_{zzxx}^{(3)} (E_x^\omega)^2] \sin \theta_T^\omega \} E_z^0 \quad (3.3)$$

where $F_{2\omega}^p$ is a Fresnel factor, θ_T^ω is the angle between the transmitted fundamental ray and the surface normal, $E_{x,z}^\omega$ are the x, z components of the fundamental field inside the silicon, and the $\chi_{ijkl}^{(3)}$ are the independent non-zero components of the third-order nonlinear susceptibility for a cubic crystal. The terms in brackets are proportional to the components $P_{FD}^x(2\omega)$ and $P_{FD}^z(2\omega)$ of the EFISH polarization. Near the edges of the electrode, the transverse DC field (E_{DC}^x) generates an additional p-polarized EFISH field

$$E_{FD}^p(E_x^0) = F_{2\omega}^p \{ [\chi_{xxxx}^{(3)} (E_x^\omega)^2 + \chi_{xzzx}^{(3)} (E_z^\omega)^2] \cos \theta_T^\omega + [\chi_{zzxx}^{(3)} E_x^\omega E_z^\omega + \chi_{zzxx}^{(3)} E_z^\omega E_x^\omega] \sin \theta_T^\omega \} E_x^0 \quad (3.4)$$

that adds coherently to $E_{FD}^p(E_z^0)$. For cubic Si, each $\chi^{(3)}$ component in Eq.(3.4) is equal to its counterpart in Eq.(3.3) with $x \leftrightarrow z$. The phase of $E_{FD}^p(E_x^0)$ differ by π at the two edges, since E_x^0 changes sign. In the geometry of Fig.3.2 (b), $E_{FD}^p(E_z^0)$ and $E_{FD}^p(E_x^0)$ add in phase at (1) and π out of phase at (3). We then measure the phase of the total SH field $E_{FI}e^{i\phi_{FI}} + E_{FD}e^{i\phi_{FD}}$ via Eq.(3.2).

A phase reversal of $E_{FD}^p(E_x^0)$ will affect overall SH phase significantly only if $|E_{FD}^p(E_x^0)|$ becomes comparable to $|E_{FD}^p(E_z^0)|$. Because the high refractive index of Si ($n \approx 3.7$) creates predominantly transverse optical fields E_x^ω in the SCR, this occurs even for $|E_x^0| \ll |E_z^0|$. From Snell's law, we get $E_z^\omega/E_x^\omega = \tan \theta_T^\omega \approx 0.2$, $\cos \theta_T^\omega \approx 0.98$ and $\sin \theta_T^\omega \approx 0.18$ for 45° incidence on Si. The relative magnitudes of $\chi_{ijkl}^{(3)}$ in Eqs.(3.3) and (3.4) are determined by the calibration EFISH measurements with $\mathbf{E}^\omega$ and $\mathbf{E}^0$ oriented along crystalline axes. At 800 nm, these measurements yield $\chi_{ijij}^{(3)} \approx \chi_{iijj}^{(3)} \approx \chi_{ijji}^{(3)}$ and $\chi_{iiii}^{(3)} \simeq 3\chi_{ijij}^{(3)}$. Substituting these values in Eq.(3.3), we find $E_{FD}^{p'} \approx 15(E_x^0/E_z^0)E_{FD}^p$, so the contributions become equal in magnitude for $|E_x^0/E_z^0| \approx 0.06$ for 45° incidence. Sensitivity to small E_x^0 is easily tuned by adjusting angle of incidence.

To calculate the real value of $|E_x^0/E_z^0|$ for our sample, we used Medici program to simulate the electric-field distribution in the space-charge region. The input file of the simulation and complete results are in Appendix. Fig.3.3 shows the results of x and z components of electric fields strength in the space-charge region near the right edge. The average of strength of x component is about a quarter of that of z component. So $|E_{FD}^p(E_x^0)|$ is comparable to

$|E_{FD}^p(E_z^0)|$, therefore a phase reversal near the right edge is possible.

The fringes in Fig.3.2 appear straight, indicating spatial phase uniformity along y under each electrode, except near the edges. To quantify the y -dependence, we analyzed line-outs of FDI fringes at selected y -positions. Fig.3.3 shows line-outs (~ 7 -pixels, or $\sim 0.8\mu m$, wide) of FDI fringes from slit position (3) at 6 selected y -positions indicated in Fig.3.2c for +8V and -8V bias. A phase shift of $\sim \pi$ (*i.e.* interchange of peaks and valleys) between +8V and -8V is evident at each position, reflecting the reversal of the space-charge field direction. We fitted each line-out to Eq.(3.2), using E_{FD} , E_{FI} , ϕ_{FD} and ϕ_{FI} as parameters. The model laser pulse profiles were Gaussians matched to the measured profiles. Fig.3.4 compares the fitting curve and the experimental data for a typical line-out. The values of 4 fitted parameters at 6 locations and 2 different biases (+8V and -8V) are plotted in Fig.3.5. The field-independent parameters E_{FI} and ϕ_{FI} didn't change significantly with either position and bias.

Also, the difference of amplitudes of EFISH of 6 locations is within 10%. This indicates the amplitude of the electric field underneath the electrodes are mostly uniform. As mentioned before, the EFISH phase from the upper and lower electrodes is significantly different. The fitting procedure indicates the difference is about 0.83π . The EFISH phase difference for +8V and -8V is about 0.95π .

Fig.3.7. shows the FD fringes at 8 different biases (+8V,+6V,+4V,+2V,-2V,-4V,-6V,-8V). We did similar analysis for each bias applied, and get con-

Figure 3.2: Frequency-domain interference fringes at +8V bias from portion of image selected by spectrometer slit positions 1 (left), 2 (middle), 3 (right) shown in Fig.3.1 (b). Note shift of fringes between upper and lower electrodes in position 3. (b) Diagram showing direction of the incident optical field and the DC field at each of the 3 sampled locations.

Figure 3.3: x and z components of electric field at $V = 8$, $x = 5.5$, horizontal axis is the depth from Si-Oxide interface.

Figure 3.4: Line-outs of frequency-domain interference fringes at 6 selected vertical positions for +8V (gray) and -8V (black) bias, showing both spatial and bias-dependent phase shifts.

Figure 3.5: Fitting curves (dashed line) and experimental fringes (solid line) at vertical electrode location (e) shown in Fig.3.3. There are 204 data points for each curve. DC bias -8V, fitting standard deviation: 115.

Figure 3.6: Fitting results for +8V and -8V at 6 different locations of the electrodes shown in Fig. 2b. Each stripe is about $0.8\ \mu\text{m}$ wide, so the space between adjacent locations here is about $0.8\ \mu\text{m}$. The error bars were decided by changing the parameter to degrade the standard deviation by 5%. The error bars for amplitude are bigger than the error bars for phase.

sistent results except for $\pm 2V$. For $\pm 2V$, the signal to noise ration is too low and we didn't get any decent fitting results using Eq.(3.2). For other biases, the field-dependent parameters are plotted in Fig.3.8.

Figure 3.7: FDI fringes at different biases: starting at +8V (upper left), and at decreasing 2V a step, ending at -8V (lower right).

Figure 3.8: Fitting results of phase (a) and amplitude (b) at different bias.

Chapter 4

Conclusions

We have demonstrated an EFISH microscope that measures both amplitude and phase of imaged second-harmonic light with sub-micron spatial resolution using an unamplified fs laser. Some finely-resolved variations in the space charge field are studied, demonstrating a new method to characterize the field and showing capabilities beyond conventional electrical characterization techniques.

One direction for future extension of this work could be more highly-resolved study of electric field inhomogeneities, by incorporating near-field scanning optical microscope (NSOM) into the EFISH technique. The combination of NSOM and femtosecond spectroscopy has attracted considerable interest as it opens up a new direction for studying ultrafast spatiotemporal dynamics of optical excitations in many material systems on nanometer length scales [34]. Yet, little is known about the effect of near-field fiber probes on ultrashort light pulses. Recently, Roland Muller *et al.* reported a simulation of propagation of femtosecond pulses through uncoated and metal-coated near-field fiber probes [35]. They used finite-difference time domain methods (FDTD) [36], and showed that pronounced spectral shifts and temporal

broadening effects for metal-coated aperture probes, while for uncoated fiber probes, in the near-field region, the spectral shifts and temporal broadening are not significant. Their results are shown in Fig.4.1.

Figure 4.1: Spatialtemporal evolution of a 10-fs optical pulse propagating through an uncoated near-field fiber probe. The peak intensity of the pulse arrives at the taper entrance ($z = 2\mu m$) at a time $t = 36.7fs$. (a) s polarization: Normalized energy density $|E_x|^2/A_0^2$ at $t = 40fs$ (left) and $t = 52fs$ (right, maximum value of $|E_x|^2$ at fiber exit). The linear gray scale is proportional to $|E_x|^2/A_0^2$. (b) s polarization: Cross section through (a) (right) at two different z coordinates: at the taper exit z_{exit} (left) and at $z_{exit} + 25nm$ (right). (c) and (d) same as (a) and (b), respectively, but for p polarization.

This simulation shows the uncoated fiber probe is a promising candidate for near-field femtosecond spectroscopy studies. Spatial resolution down to

$\lambda/4$ has been achieved for uncoated fiber probes [37]. So incorporating near-field technique will greatly improve the spatial resolution and may enable the EFISH microscope to detect the finest field inhomogeneities in modern semiconductor devices.

Another future direction would be pump-probe EFISH microscopy to study femtosecond time-resolved phenomena. One good example is to study the dynamics of photoinduced field screening. As mentioned in Chapter 2, one of the major drawbacks of confocal scanning EFISH microscope is that the dc-field is partly screened by laser-induced carriers, which happens in femtosecond time scale. This photogenerated carrier screening process is of critical importance in recombination kinetics and electron transfer process at semiconductor interfaces [38], and has been studied by various optical methods [39]. Combining pump-probe technique, EFISH microscopy is a new promising tool to study the photogenerated carrier dynamics.

Finally, EFISH microscopy can be applied to other material systems, such as small laser-excited plasmas and biological cell membranes. Y.R. Shen *et al.* reported a picosecond studies of optical SHG in atomic vapor in 1994 [40]. SHG in atomic vapor is a highly forbidden nonlinear optical process because plasma is a centrosymmetric isotropic media. Yet many experiments have demonstrated that SHG in alkali or alkali-earth vapors can be easily detected when an exciting laser pulse is tuned to a two-photon resonance [41]. This is because of laser-induced ionization-initiated EFISH. They observed ~ 100 *photons/pulse* for a peak intensity of 10^{10} W/cm^2 in a K(0.6Torr)/Ar(20Torr)

mixture vapor. Thus it may be possible to use EFISH microscopy to image and measure laser wakefields in plasma. Fig.4.2 shows a possible pump-probe imaging configuration.

Figure 4.2: A proposed pump-probe experimental setup to measure EFISH generated by laser wakefields.

EFISH microscopy can also be applied to biological system, for example, to study the electrical properties of cell membranes. The cell membrane serves as a barrier and passage of most polar molecules [42]. In order to transfer molecules, a membrane potential is created, as shown in Fig.4.3. A change in the voltage can open and close the ion channel. EFISH microscopy provides a new tool to map and measure the potential across the membrane, and enable

us to understand the mechanism of transfer of ions better.

Figure 4.3: Ion-channels for cell membrane.

There are evidently a variety of different directions in which EFISH microscopy could go in the future, and it will be interesting to see what kinds of applications this instrument will find.

Appendix

Appendix 1

Medici simulation for electric field distribution in MOS space-charge region

Medici input file for MOSCAP

Gate length=5um tox=90A

Chromium Gate metal thickness=1000A

Author: Sagnik Dey

Date: Apr 2004

COMMENT Specify a rectangular mesh

MESH SMOOTH=1

X.MESH WIDTH=6.0 H1=0.125

Y.MESH Y.MIN=-0.109 DEPTH=0.1 H1=0.025

Y.MESH DEPTH=0.009 H1=0.003

Y.MESH DEPTH=0.25 H1=0.03125

Y.MESH DEPTH=0.25 H1=0.0625

COMMENT Eliminate some unnecessary substrate nodes

ELIMIN COLUMNS Y.MIN=0.375

COMMENT Specify oxide and silicon regions

REGION SILICON

REGION OXIDE Y.MAX=0 Y.MIN=-0.009

```

REGION    OXIDE      Y.MAX=-0.009 Y.MIN=-0.109 X.MAX=0.5 X.MIN=0
REGION    OXIDE      Y.MAX=-0.009 Y.MIN=-0.109 X.MIN=5.5 X.MAX=6.0
COMMENT    Electrode definition
ELECTR    NAME=Gate    X.MIN=0.5  X.MAX=5.5    Y.MIN=-0.109 Y.MAX=-0.009
ELECTR    NAME=Substrate  BOTTOM
COMMENT    Specify impurity profiles and fixed charge
PROFILE    P-TYPE  N.PEAK=1E15  UNIFORM      OUT.FILE=MOSCAP1DS
INTERFAC   QF=1E10
PLOT.2D    GRID  TITLE="Example 1 - Initial Grid"  FILL
COMMENT    Regrid on doping
REGRID     DOPING  LOG  IGNORE=OXIDE  RATIO=2  SMOOTH=1+
IN.FILE=MOSCAP1DS
PLOT.2D    GRID  TITLE=" Doping Regrid"  FILL
COMMENT    Specify contact parameters for Chromium
CONTACT    NAME=Gate  workfunc=4.5
COMMENT    Specify physical models to use
MODELS     CONMOB  FLDMOB  fermidirac
CONSRH     AUGER
COMMENT    Symbolic factorization, solve, regrid on potential
SYMB       CARRIERS=0
METHOD     ICCG  DAMPED
SOLVE      Solving Poisson and Continuity
LOOP STEPS=9

```

```

ASSIGN NAME=vg N.VAL=( 0,2,4,6,8,-2,-4,-6,-8)

SYMBOL      CARR=0

METHOD      ICCG  DAMPED

SOLVE       V(Gate)=0 V(Substrate)=0 OUT.FILE=TEMPSOL

MODELS      fermidirac  PRPMOB  CONSRH  AUGER

SYMBOL      CARR=0 newton

SOLVE       V(Gate)=@vg

PLOT CONTROLS

PLOT.2D      BOUND  TITLE="ELECTRIC FIELD CONTOURS at Vg"=@vg  FILL
+ dev=cl/post plot.out="moscap_contours.ps"

Uncomment the above line to redirect output to the ps file

CONTOUR E.FIELD ABS MIN=0 MAX=5E+4  LINE=1  FILL

L.END

```

Figure 1.1: Electric field contour at $V=0$

ELECTRIC FIELD CONTOURS

Figure 1.2: Electric field contour at $V=8$

Figure 1.3: z component of electric field at $V=8$, $x=5$, horizontal axis is the depth from Si-Oxide interface.

Figure 1.4: x component of electric field at $V=8$, $x=5$, horizontal axis is the depth from Si-Oxide interface.

Figure 1.5: z component of electric field at V=8, x=5.5, horizontal axis is the depth from Si-Oxide interface.

Figure 1.6: x component of electric field at $V=8$, $x=5.5$, horizontal axis is the depth from Si-Oxide interface.

Figure 1.7: z component of electric field at $V=8$, $x=4.5$, horizontal axis is the depth from Si-Oxide interface.

Figure 1.8: x component of electric field at $V=8$, $x=4.5$, horizontal axis is the depth from Si-Oxide interface.

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