Local characterization of mobile charge carriers by two electrical AFM modes: multi-harmonic EFM versus sMIM

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Abstract
The characterization of mobile charge carriers of semiconductor materials has spurred the development of numerous two dimensional carrier profiling tools. Here, we investigate the mobile charge carriers of several samples by multi-harmonic electrostatic force microscopy (MH-EFM) and scanning microwave impedance microscopy (sMIM). We present the basic principles and experiment setups of these two methods. And then several typical samples, i.e. a standard n-type doped Si sample, mechanical exfoliation and chemical vapor deposition grown molybdenum disulfide (MoS2) layers are systemically investigated by sMIM and MH-EFM. The difference and (dis)advantages of these two modes are discussed. Both modes can provide carrier concentration profiles and have sub-surface sensitivity. They also have advantages in sample preparation in which contact electrodes are not required and insulating or electrically isolated samples can readily be studied. The basic mode, physics quantities extracted, dielectric response form and parasitic charges in scanning environment result in difference in experiment results for these two kinds of methods. The techniques described in this study will effectively promote research on basic science and semiconductor applications.

1. Introduction
Two dimensional (2D) semiconductor materials is the foundation of modern electronics and performance of their devices is influenced by electrical parameters such as carrier type, dopant concentration, defects densities and so on. Hence, the development of semiconductor industry demands for sub 10 nm resolution, combined with sufficient sensitivity (down to the 10¹⁵ atoms cm⁻³ level) and high-quantification accuracy over a dynamic range of 10¹⁵–10²⁰ atoms cm⁻³ [1]. The need for such an extreme spatial resolution as well as the applicability towards standard devices has spurred the development of numerous 2D carrier profiling tools. Until today, various methods have been developed for this purpose, such as secondary ion mass spectrometry, field-effect scanning electron microscopy, etc. One category of these methods is scanning probe microscopy (SPM)-based techniques.

SPM is implemented by position various types of probes in very close proximity with extremely high precision to the sample. These probes can detect electrical current, atomic and molecular forces, electrostatic forces, or other types of interactions with the sample. SPM-based mobile charge carriers detection methods include scanning Kelvin probe force microscopy (SKPM) [2–4], scanning capacitance microscopy (SCM) [5, 6], scanning surface harmonic microscopy (SSHM) [7] and scanning spreading resistance microscopy (SSRM).
sMIM was proposed by Lai et al. [8, 9], etc. The SKPM methods can measure contact potential difference between the probe and sample surface, which is dependent on the carrier concentration-related work function difference, and can qualitatively obtain the local carrier concentration. In the SCM, tip–sample contact forms a metal–insulator–semiconductor (MIS) capacitor, whose local carrier type and concentration can be obtained by capacitance–voltage (C–V) behavior. In the SSHM (consists with a STM with a microwave cavity), the nonlinear tip–sample MIS capacitance $C$ results in higher-order harmonics in the tunneling current. The capacitance is a measure for the local active carrier concentration of the semiconductor in the same way as in the SCM technique. In SSRM method, the local carrier concentration depends inverse proportionally on the spreading resistance around the probe–sample contact. Although the above methods can characterize the charge carrier type and concentration of sample, they are restricted by limited sensitivity, laborious sample preparation, destructive of sample, lower signal-to-noise ratio and others.

The newly-developing powerful method scanning microwave impedance microscopy (sMIM) is a promising tool for 2D mobile charge carriers profiling with nanometer resolution [10–20]. It also has advantages in sub-surface scanning ability and simple in sample preparation [21]. Considering the limitation of fiscal condition, multi-harmonic electrical force microscopy (MH–EFM) can be used in mobile charge carriers characterization of semiconductor 2D materials [22].

Figure 1 illustrates the microwave electronics. Generally speaking, a gigahertz (GHz) signal applied to a scanning probe to detecting the complete complex-valued tip–sample impedance $Z_{\text{tip-sample}}$, which results from the local electronic properties. Because of the large mismatch between tip–sample impedance $Z_{\text{tip-sample}}$ and the transmission line impedance $Z_0 = 50$ Ω, impedance matching section is need to maximizing the microwave power delivered to probe [10, 11]. As shown in figure 1, firstly, microwave transmits into the probe from the microwave source. Then microwave near-field interacts with sample and backtracks. The microwave

Figure 1. A schematic of sMIM setup. The shielded probe connected with microwave source (~GHz frequency band). Microwave transmits into the probe, near-field interacted with the sample, and then reflection microwave backtracks. The reflected signal is suppressed by the common-mode cancelling through a directional coupler (D), amplified by radio frequency (RF) amplifiers (A), and then demodulated by a quadrature mixer (M1). sMIM-Im and sMIM-Re information can be obtained. The signal further modulated by quadrature mixer M2, dC/dV and dR/dV can be obtained. Insert: schematic of lumped element model.
reflection coefficient is $\Gamma = \frac{Z_{\text{tip-sample}} - 50 \Omega}{Z_{\text{tip-sample}} + 50 \Omega}$. A cancellation signal is provided to suppress the background so that small changes can be amplified without saturating the output. And finally amplified signal demodulated by the mixer M1. The effective tip–sample impedance can be divided into two parts—the real and imaginary parts. Standard dielectric sample (Al2O3@SiO2) is needed to calibration the phase of the M1, until the output contrast only occurs in one channel (this is imaginary part of $Z_{\text{tip-sample}}$) sMIM-Im). The other channel is the real part of $Z_{\text{tip-sample}}$ (sMIM-Re). For semiconductors or insulators, the sMIM-Im and sMIM-Re are corresponding to the capacitance ($C$) and resistance ($R$) between tip–sample, respectively, as shown in the insert image lumped element model.

An alternating current (AC) voltage with frequency $\sim$kHz is applied to the tip as a modulation voltage. Using the quadrature mixer M2, $dC/dV$ and $dR/dV$ amplitude (carrier concentration) and phase (carrier type) can be obtained. (See details in supplementary material is available online at stacks.iop.org/JPCO/2/025013/ mmedia.) Furthermore, any stray field contribution from the non-tip part (i.e. cantilever and base) is essentially constant during a single scan. The effect of stray capacitance can be removed by simulations.

In this letter, the AFM, Asylum Research MFP-3D Infinity (under ambient condition) was used. Microwave (3 GHz) imaging was performed with a ScanWave (Prime Nano, Inc.) sMIM add-on unit installed on the AFM. The sMIM microwave output power was set to 100 mW. Fully shielded sMIM cantilever probes had spring constants in a range 1–2 N m$^{-1}$. The C–V curves were obtained in ambient with a home-made system, which combining the Dynamic Signal Analyzer (HF2LI, Zurich Instruments) with an Asylum MFP-3D infinity.

MH-EFM is also a powerful tool to study the carrier concentration of semiconductor 2D materials [22] without adversely affected by stray capacitance, as shown in figure 2. In MH-EFM, dual pass mode was used to detect topography and electrical properties of sample. In first pass, the AFM works the same way as the typical AC mode (tapping model). In second pass (lift mode), short-range repulsive forces and the van der Waals force can be ignored because the tip–sample distance is large enough; meanwhile an AC voltage with a frequency of $f_m$ is applied between the tip and sample, resulting in multi-harmonic electrostatic force. The electrical force is as follows:

$$F = -\frac{1}{4}\left[\frac{\partial C}{\partial z}V_{ac}^2 + \frac{\partial^2 C}{\partial V \partial z}V_{ac}^2 \cos \omega t + \frac{\partial C}{\partial V}V_{ac}^2 \cos 2\omega t + \frac{1}{2}\frac{\partial^2 C}{\partial V \partial z}V_{ac}^2 (\cos 3\omega t + \cos \omega t)\right].$$

Accordingly, the second and third harmonic component given by:

$$F_{2\omega} = -\frac{1}{4}\frac{\partial C}{\partial z}V_{ac}^2 \cos 2\omega t,$$

$$F_{3\omega} = \frac{1}{2}\frac{\partial^2 C}{\partial V \partial z}V_{ac}^2 (\cos 3\omega t + \cos \omega t).$$
The concentration of sample is different: the lower area is highly doped area and the upper light doped one. This sample is standard n-type doped Si sample with different dopant concentration. Here, a traditional semiconductor material grown molybdenum disulfide (MoS2) layers are chosen as the representative of 2D semiconductor materials.

Furthermore, the penetration depth of these two modes has been studied by suspended MoS2 over circular holes — standard n-type doped Si sample with different dopant concentration is, which agrees with the parameters of the given sample.

Therefore, the information corresponding to \( \frac{\partial C}{\partial z} \) and \( \frac{\partial C}{\partial V} \) can be obtained by detecting the 2\( \omega \) and 3\( \omega \) components. In another word, the amplitude of cantilever vibration (\( A_{2\omega} \) and \( A_{3\omega} \)) can reflect the capacitance (or dielectric constant) and carrier concentration of the sample, respectively [36-38]. Furthermore, the \( A_{3\omega} \) has been amplified greatly by a resonance phenomenon. Specifically, resonant frequency of probe \( f_0 \) is about 72 kHz; so we setting the frequency of applied AC voltage \( f_3 = f_0/3 \sim 24 \text{ kHz} \). By setting the frequency in such way, \( f_{3\omega} \) is coincident with the free resonance frequency of cantilever. It should be noted that the theory of MH-EFM is also applicable when \( f_{3\omega} \) located at non-resonance frequency. In this letter, we set \( f_{3\omega} \) at probe resonance frequency in order to get better sensitivity.

In this paper, the MH-EFM imaging were performed in ambient with a home-made system, which combining the Dynamic Signal Analyzer (HF2LI, Zurich Instruments) with an Asylum MFP-3D infinity. Metal coated probe are used in this mode.

The above studies proved that MH-EFM and sMIM can both profile the carrier concentration of semiconductors. In order to exhaustive delineation of the carrier concentration characterization by these two modes, several typical samples have been chosen to investigate here. Firstly, a standard n-type doped Si sample was used as a representative of traditional semiconductor material. Then, mechanical exfoliation and CVD grown molybdenum disulfide (MoS2) layers are chosen as the representative of 2D semiconductor materials. Furthermore, the penetration depth of these two modes has been studied by suspended MoS2 over circular holes arrays. During the experiment, all the samples are grounded. The experimental results are shown in the following parts.

### 3. Results

#### 3.1. Traditional semiconductor material

Here, a traditional semiconductor material — standard n-type doped Si sample with different dopant concentration has been studied systematically. This standard sample is provided by Prime Nano. The dopant concentration of sample is different: the lower area is highly doped area (nominal \( \sim 10^{21} \text{ cm}^{-3} \)), the center of higher strip area is light doped (nominal \( \sim 10^{15} \text{ cm}^{-3} \)). (See details in supplementary material.)

Figure 3(a) shows the topography of doped Si sample. Figure 3(b) shows the C–V curves taken in sMIM mode. The C–V curves acquired at different positions show quite different characteristics, which is attributed to the difference of dopant concentration in sample. In the C–V curves, the depletion region (slope rapidly changed region) can be used to describe the carrier concentration of sample. The higher the slope is, the lower carrier concentration is, which agrees with the parameters of the given sample.

The dC/dV amplitude images with different tip bias \( V_G \) taken in sMIM mode are shown in figures 3(c)–(e). The contrast between lower doped and higher doped regions can be seen clearly when \( V_G \) is 1.2 V, while it is invisible when \( V_G \) is 3 and −2V. Since dC/dV can be seen as the slope of C–V curve, the behavior of dC/dV amplitude can be understood derived from the C–V curves. When \( V_G \) is 1.2 V, the slope of C–V curve is significant different for different doping areas, and this resulting in the strong contrast of light dopant regions, as shown in figure 3(d). However, when \( V_G \) larger than 3 V or smaller than −2 V, the slopes of all the C–V curves are nearly the same. This is why the amplitude images with different tip bias \( V_G \) are almost the same.
are basically same (slopes are all approximately zero), so there is no contrast can be seen between high doped and less doped regions, as show in figures 3(c) and (d).

Figures 3(f)–(h) show amplitude $A_{3\omega}$ images taken in MH-EFM mode with different gate voltages. The contrast of light or heavy doped areas is different when $V_G$ changes from $-2$ to $3$ V. When $V_G$ is $1.2$ V, the contrast of light doped area is brighter than that of heavy doped areas, shown as figure 3(g). However, there is no contrast can be seen when $V_G$ is $3$ and $-2$ V. In MH-EFM, the sample is under low-frequency AC modulation, the carriers are driven in and out of the area underneath the tip. Although the electric field distributions around sample are not strictly same in MH-EFM and sMIM modes, the modulated $A_{3\omega}$ signals are basically consistent with our observation in sMIM mode.

3.2. 2D semiconductor materials

2D MoS$_2$, as a member of transition metal dichalcogenides, has shown promising potential in next generation optoelectronic and electrical devices due to its unique optical and electrical properties. Synthesis of few layers MoS$_2$ has obtained a huge development, such as mechanical exfoliation, solution-based exfoliation, vapor-phase growth [39]. Here both the mechanical exfoliation and CVD grown MoS$_2$ are chosen as our sample. Firstly, the MoS$_2$ flakes obtained by mechanical exfoliation using adhesive tape (Scotch) and transferred to SiO$_2$ /Si substrate has been studied.

Figure 4(a) shows the topography of mechanical exfoliation MoS$_2$ flakes. The topography profiling and Raman spectroscopy [40] (figure S3) show that the thickness of MoS$_2$ changes from two layers (2L) to bulk, marked in figure 4(a).

The sMIM characterize of few layers MoS$_2$ are shown here. The lumped element model [41] between tip and sample is shown in figure 4(b). Considering the following parts are also use MoS$_2$ as sample, the lumped element model is also applicable for figure 5. The $C-V$ curves taken at different positions are demonstrated in figure 4(c).

The sMIM-Im value of substrate (SiO$_2$/Si) does not change with the gate voltages, showing an insulator behavior. The $C-V$ curves taken on MoS$_2$ flakes show that the MoS$_2$ flakes are n-type doped, and the carrier concentration is different for different thickness. It shows that the carrier concentration of 2L and bulk MoS$_2$ are
larger than that of three layers (3L) MoS\textsubscript{2}. Besides the influence of layer thickness, local carrier concentration may be influenced by interfacial impurities, strain, disorders and charge transfer \cite{42, 43}.

Figures 4(d)–(f) are dC/dV amplitude images with different tip bias \(V\text{G}\), which are taken in sMIM mode. Comparing to the substrate, the contrast of 2L, 3L and bulk MoS\textsubscript{2} flakes all can be seen clearly when \(V\text{G} = -5\text{V}\), while 2L and bulk regions became weaker when \(V\text{G} = 0\text{V}\) and disappear when \(V\text{G} = 5\text{V}\). The behavior of dC/dV amplitude can be easily interpreted by the C–V curves. When \(V\text{G} = -5\text{V}\), the slope of C–V curves are similar for different thickness MoS\textsubscript{2} layers and significantly different from substrate. So the contrast between MoS\textsubscript{2} layers is small and the contrast between MoS\textsubscript{2} and substrate is large. When \(V\text{G} = 5\text{V}\) is applied, the slopes of C–V curves at 2L and bulk layers are similar to substrate, so there is no contrast can be seen between substrate, 2L and bulk MoS\textsubscript{2}, as shown in figure 4(f). However, the slope of C–V curve at 3L MoS\textsubscript{2} is larger than other regions at \(V\text{G} = 5\text{V}\), so the contrast of 3L is much brighter than others regions.

Figures 4(g)–(i) show \(A_{2\omega}\) images of MoS\textsubscript{2} flakes obtained in MH-EFM mode, and the gate voltages are marked in it. The contrast of them is different when \(V\text{G}\) changes. However, the change is absolutely different from the data obtained by sMIM mode, and it is hard to understand. The reasons will be discussed in the following Discussions parts.

Expect the mechanical exfoliation samples, the CVD grown MoS\textsubscript{2} flakes \cite{44–46} has been studied by sMIM and MH-EFM. The MoS\textsubscript{2} flakes were grown on SiO\textsubscript{2} (300 nm)/Si substrate, via traditional CVD method.

The MoS\textsubscript{2} flakes have a mixed zigzag/armchair edges because the edges are not straight \cite{47}, as shown in figures 5(a) and (d). These flakes can be divided into two different kinds: the purely uniform monolayer (marked by red arrow) and pyramid-shaped flakes (marked by blue arrow). The formation of different structures may be origin from different defects and dislocation concentration, i.e. screw dislocation can result in pyramid-shaped flakes \cite{48}.

These two kinds of sample have different contrast in sMIM-Im image, as shown in figure 5(b). The monolayers are hard to be distinguished from substrate, while the pyramid-shaped flakes are easy to be seen. The reason is that the thickness of monolayer is too small, so that the reflection microwave mainly reflecting the properties of substrate, other than properties of monolayers MoS\textsubscript{2}. Meanwhile, the pyramid-shaped flakes thick enough, so the electrical properties of pyramid-shaped flakes can be obtained by sMIM.
It is known that the conductivity of pyramid-shaped flakes is larger than monolayer because of the decreased band gap or dislocation [48, 49]. Hence, the difference of conductivity between pyramid-shaped MoS<sub>2</sub> and substrate results in the sMIM contrast [46]. Classically, the sMIM-Im is corresponding to the dielectrics of sample. One may ignore the sMIM-Im can also indicate the conductivity of sample. According to the finite-element analysis (FEA) in [46], we can see the sMIM-Im is relating to electrical conductivity. And the lower the sMIM-Im is, the lower conductivity is. So we believe the sMIM-Im contrast (in figure 5(b)) of different thickness MoS<sub>2</sub> derived from their conductivity.

Figure 5(c) shows the C–V curves taken at substrate, monolayer and pyramid-shaped flakes, respectively. The sMIM-Im value of substrate does not change with the gate voltages, showing an insulator behavior. The C–V curve of monolayer is basically same as the substrate, which confirms the reflection microwave on monolayers mainly reflecting the properties of substrate. However, the C–V curve of pyramid-shaped flakes shows n-type semiconductor behavior.

Figures 5(d) and (e) show the simultaneously obtained topography and A<sub>2ω</sub> images of sample taken in the MH–EFM mode. In the A<sub>2ω</sub> image, the brighter the contrast is, the more quantity mobile charge is. The brighter contrast of the MoS<sub>2</sub> layers represents they have more mobile charges, in contrast with the simple dielectric response of insulating SiO<sub>2</sub> substrate. Furthermore, it can be seen the contrast of pyramid-shaped MoS<sub>2</sub> is brighter than that of monolayer sample. This suggests the conductivity of the pyramid-shaped MoS<sub>2</sub> is larger than monolayer, which is consists with previously works [48, 49].

Figures 5(g)–(i) show A<sub>ω</sub> images of sample with different gate voltages taken in MH–EFM mode. The contrast of uniform monolayer does not change with the gate voltage, while that of pyramid-shaped flakes changes a lot as the gate voltage changing. It means that, in pyramid-shaped MoS<sub>2</sub>, the carrier concentration of sample, and have sub-surface scanning ability. They also have advantages in sample band gap or dislocation [46, 48, 49]. Moreover, the signal of sample is not homogeneous. The sMIM signal near MoS<sub>2</sub> edge is stronger than the central region of sample. One may ignore the sMIM-Im can also indicate the conductivity of sample. According to the previous experimental data taken in these two modes, one can find that the MH–EFM is more sensitive than the sMIM for CVD grown MoS<sub>2</sub> layers. The high sensitivity may be derived from the linear dielectric response form and signal amplified by a resonance phenomenon.

3.3. Sub-surface capability: MoS<sub>2</sub> over SiO<sub>2</sub>/Si with circular holes arrays

In order to study sub-surface capability of these two modes, the MoS<sub>2</sub> flake over SiO<sub>2</sub>/Si with circular holes arrays has also been studied. Briefly, the SiO<sub>2</sub>/Si substrate patterned with different diameter holes defined by e-beam lithography. Then mechanical exfoliation MoS<sub>2</sub> flakes have been transferred onto the substrate to form suspended MoS<sub>2</sub>.

Figure 6(a) shows topography image of locally suspended MoS<sub>2</sub> membranes over holes arrays. Thickness of the MoS<sub>2</sub> layers is uniform ∼30 nm. The buried holes cannot be seen in the topography, however, they can be observed in sMIM-Im image, as shown in figure 6(b). Although the thickness of MoS<sub>2</sub> is uniform, sMIM-Im signal of sample is not homogeneous. The sMIM signal near MoS<sub>2</sub> edge is stronger than the central region of sample.

Figure 6(c) shows the C–V curves taken at different positions, which are marked by the color dots in figure 6(b). The C–V curves indicate that the carrier concentration of MoS<sub>2</sub> flakes is non-uniform. The carrier concentration difference may be resulting from strain, interfacial impurities, disorders and charge transfer [42, 43]. Figures 6(d)–(f) are dC/dV amplitude images with different tip bias V<sub>g</sub> taken in sMIM mode. The dC/dV amplitude contrast of whole MoS<sub>2</sub> is uneven, which confirm the difference of carrier concentration in MoS<sub>2</sub> flakes. The dC/dV images show the same physics as the C–V curve, i.e. when V<sub>g</sub> is 5 V, the same contrast of the central region of MoS<sub>2</sub> and substrate (include exposed holes) means the slope of C–V curves at these areas are same.

Figures 6(g)–(i) show A<sub>ω</sub> images of sample with different gate voltages taken in MH–EFM mode. The buried holes can also be observed in figures 6(g), (h), which indicate the MH–EFM also has sub-surface scanning ability. According to our experimental data, the penetration depth of MH–EFM method at least can reach to 75 nm (see details in supplementary material), which agrees with previously work [50]. The reason is that electrostatic fore is long-range interaction force. The contrast in A<sub>ω</sub> images changes for different V<sub>g</sub>. However, the change is absolutely different from the data obtained by sMIM mode. The reasons will be discussed in the following Discussions parts.

4. Discussions

Theoretically and experiment results of sMIM and MH–EFM modes have been performed. They both can profile the carrier concentration of sample, and have sub-surface scanning ability. They also have advantages in sample...
preparation. Contact electrodes are not required and insulating or electrically isolated samples can readily be studied.

The difference and (dis)advantages of these two modes are further discussed here. Firstly, the basic mode of sMIM and MH-EFM are different. The sMIM can be implemented in both contact mode and AC mode, while the MH-EFM is implemented in AC mode [11, 51]. In our work, all the sMIM data is obtained at contact mode since better signal-to-noise ratio and more physical quantity can be obtained. More importantly, the basic mode may have great influence on the experiment data. The MH-EFM data is likely to be affected by charges in moisture adhered to the sample as well as in the ambient air around the tip, both of which are potential sources of alterations in charge distribution. Conversely, in sMIM mode (contact mode), tip makes direct contact with the sample surface by penetrating through any moisture layer, producing a point of direct contact, which is less likely to be influenced by parasitic charges from the scanning environment.

Secondly, the physics quantities extracted by the two methods are different. The MH-EFM can extract surface potential, capacitance and carrier concentration qualitatively. While the conductivity, permittivity, carrier type and concentration can be extracted quantitatively combining the FEA with spectroscopy study (i.e. $C-V$, $R-V$ and $C-z$ curves) in sMIM mode [11, 46, 52, 53].

Thirdly, detection sensitivity would be different for the determined carrier concentration. The dielectric response form may effect on the detection sensitivity. The dielectric response (the relationship between capacitance and dielectric constant, $C-\varepsilon$) has different form in these two modes. The dielectric response is linear in MH-EFM, while it is nonlinear in sMIM mode. Expect the influence of dielectric response form, signal amplified by a resonance phenomenon improve the detection sensitivity in MH-EFM mode. So the sensitivity is different for the determined carrier concentration. Note that the $A_{3\omega}$ images sometimes even has better sensitivity than sMIM mode, as shown in figure 5.

5. Conclusions

The mobile charge carriers profiling of several samples has been investigated by MH-EFM and sMIM, respectively. These two modes both enable provide carriers concentration imaging and have sub-surface
sensitivity. The dielectric response form and parasitic charges in scanning environment may result in difference of the two kinds of methods. The techniques described in this study will effectively promote research on basic science and semiconductor applications.

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