An approach for high-frequency transport imaging, referred to as scanning frequency mixing microscopy (SFMM), is developed. Application of two high-frequency bias signals across an electroactive interface results in a low-frequency component due to interface nonlinearity. The frequency of a mixed signal is chosen within the bandwidth of the optical detector and can be tuned to the cantilever resonances. The SFMM signal is comprised of an intrinsic device contribution and a capacitive mixing contribution, and an approach to distinguish the two is suggested. This technique is illustrated on a model metal-semiconductor interface. The imaging mechanism and surface-tip contrast transfer are discussed. SFMM allows scanning probe microscopy based transport measurements to be extended to higher, ultimately gigahertz, frequency regimes, providing information on voltage derivatives of interface resistance and capacitance, from which device characteristics such as Schottky barrier height, etc., can be estimated. © 2006 American Institute of Physics. [DOI: 10.1063/1.2192977]
is used to determine the magnitude and phase of the cantilever response at \( \omega \) or \( \delta \omega \). The output amplitude and phase shift signals are stored in an external control computer as a function of frequency, dc bias applied across the device, and position on device surface (slow scan axis disabled), producing two dimensional (2D) spectroscopic maps that illustrate frequency/bias, coordinate/bias, or coordinate/frequency dependences of response signal.

In SFMM, the modulation signal, \( V_{\text{int}} = V_{\text{dc}} + V_1 \cos(\omega t) + V_2 \cos(\omega t + \delta \omega) t \), is applied across the experimental circuit as shown in Fig. 1(a), where \( \omega \) is chosen in the 1 kHz–40 MHz range (limited by the function generator), and \( \delta \omega \) is typically 10 kHz. Correspondingly, the surface potential has a dc component \( V_{\text{surf}}(x) \) and components at the frequencies of lateral bias, \( V_{\text{surf}}(x) \) and \( V_{\text{sur},\delta \omega}(x) \). The detailed analysis of the dc and first harmonic responses is reported elsewhere. In nonlinear systems, frequency mixing at electroactive interfaces results in additional terms at higher-order mixed harmonics of the modulation signal. Particularly of interest is the low-frequency component \( V_{\text{surf}} \cos(\delta \omega t) \) that can be tuned to be within the bandwidth of optical detection for arbitrarily high \( \omega \).

The oscillating bias results in capacitive force acting on the dc biased tip,

\[
2F_{\text{cap}}(z) = C'_{t}(V_{\text{tip}} - V_{\text{surf}})^2,
\]

where \( V_{\text{tip}} \) is the tip bias, and \( C'_{t} \) is the tip-surface capacitance gradient, resulting in the transfer between the voltage oscillations of the surface and cantilever amplitude. However, this quadratic dependence of the tip-surface force also results in an additional frequency mixing between modulation signals. Thus, the SFMM signal is a sum of an intrinsic signal generated in the device, \( F_{\text{surf}} \), and an extrinsic signal generated in the tip-surface junction, \( F'_{\text{surf}} \):

\[
F_{\delta \omega} = F'_{\delta \omega} + F''_{\delta \omega} \sim V_{\text{surf}}(V_{\text{tip}} - V_{\text{surf}}) + V_1 V_2.
\]

Notice that the intrinsic term is linear in tip bias, while the extrinsic term is tip bias independent. Hence, the intrinsic contribution can be selected by acquisition of data at several (e.g., 3) different tip biases and detecting the slope of local response-bias curve, or by using an additional lock-in and periodically varying the tip bias \( V_{\text{tip}} \) to determine the linear component.

Similarly to linear and nonlinear SIMs, the amplitude of the tip vibration is proportional to the corresponding harmonic of the bias, while the phase is shifted by a position-independent term. Thus, measuring the phase and amplitude of the tip oscillation allows the phase and amplitude of the surface voltage oscillations to be mapped.

Device mapping using SFMM is illustrated in Fig. 2. The spatial distribution of the first harmonic amplitude and phase signal (SIM) as a function of lateral bias is illustrated in Figs. 2(a) and 2(b). As previously reported, the amplitude and phase shifts develop across the interface under reverse bias conditions, while under forward bias, there is only a small signal variation due to the work function difference between metal and semiconductor. The maps of amplitude and frequency of the mixed signal (SFMM) are shown in Figs. 2(c) and 2(d). Note that the amplitude variation is step-like for large reverse biases, exactly zero for forward biases, and contains a sharp feature at the zero bias, i.e., in the region of maximum nonlinearity of the \( I-V \) curve. This agrees with the predictions of Eq. (2), where at large negative biases, the extrinsic contribution dominates, while at zero bias, the intrinsic term dominates.

To distinguish and separate these contributions, we have acquired the response maps for several tip biases and numerically determined the slope and intercept of the corresponding best linear fit at each point in the \( (V_{\text{surf}}, \delta \omega) \) phase space. The resulting slope and intercept maps are shown in Figs. 2(e) and 2(f), providing the decomposition of intrinsic and extrinsic signals. This behavior is further elucidated in Figs. 3(a) and 3(b) where we examine the line profiles along the voltage axis. Note that the extrinsic signal is independent of the surface work function, a feature that constitutes a clear advantage compared to standard SIM. The linear component can also be selected using a second lock-in amplifier and by periodically modulating tip bias.

To analyze SFMM dynamics, the responses at \( \omega \) (SIM), \( 2\omega \) and \( 3\omega \) (NL-SIM), and \( \delta \omega \) (SFMM) were measured simultaneously as a function of frequency (\( \omega \) varied, \( \delta \omega \) fixed).
For a symmetric circuit, the amplitudes of the intrinsic SFMM signals are equal on both sides of the interface, whereas, the phase changes by 180°, in agreement with the experimental observations.

While Eq. (3) is difficult to apply analytically, we can compare the model and experimental results by estimating numerically the response of the experimental system using predetermined device parameters (saturation current and Schottky barrier height). The numerically calculated potential drop across the interface and SFMM signal as a function of circuit termination resistance and lateral bias are illustrated in Figs. 4(a) and 4(b). Note that SFMM signal decreases with the resistance $R$, and the signal maximum shifts to negative biases. Corresponding experimental behavior is shown in Figs. 4(c) and 4(d), in agreement with the model calculations.

To summarize, the nonlinear high-frequency transport behavior across electroactive interfaces can be accessed using a mixed frequency transport imaging technique, referred to here as SFMM. Both intrinsic frequency mixing in the device and electrostatic frequency mixing in the tip-surface junction contribute to the measured signal. These contributions can be separated using approaches based on a secondary tip bias modulation. Compared to SIM and NL-SIM signals, the SFMM signal has a much weaker frequency dependence, and therefore allows for the decoupling of material and probe dynamics. The extension of SPM-based transport measurements to the high-frequency regime will provide information on device behavior under conditions of operation, and allow in situ characterization of device operation on the nanoscale.

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