RHK Technology

Application Note

Integration of the RHK R9 Controller for Tip-Enhanced Photoluminescence (TEPL) Spectroscopy and Imaging

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Introduction

This application note describes the use of tip-enhanced photoluminescence (TEPL) spectroscopy and nano-imaging of excitonic properties in semiconducting 2D materials based on RHK's R9 controller in Prof. Markus Raschke's group at the University of Colorado Boulder. This includes the study of structural heterogeneity, edges, and grain boundaries, and their influence on both dark and bright excitonic properties in these materials with nanometer spatial resolution. Specifically, by tilting the tip, the in-plane transition dipole moment of the bright excitons can be accessed spectroscopically. Conversely, with the tip oriented normal with respect to the surface, the out-of-plane transition dipole moments of dark excitons can be probed, with switching, and modulation of the dark exciton emission.

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Connection to R9*plus*

In Prof. Markus Raschke's group at the University of Colorado Boulder, the RHK R9*plus* controller is used to control a custom-built tip-enhanced Raman (TERS) and tip-enhanced photoluminescence (TEPL) atomic force microscope (AFM) system. A schematic is shown in Figure 1. The high voltage outputs of the R9*plus* control the AFM positioning stage (PI NanoCube P-611.30), and shear force feedback is maintained by modulating a dither piezo with one of the R9*plus* low voltage outputs. A ±15 V DC power output is used to power a custom amplifier to detect the oscillation amplitude of the quartz tuning fork.

Optical signal information is sent to the R9*plus* from the Princeton Instruments Lightfield software through a LabView interface developed in Prof. Raschke's group. The optical signal in a user-defined spectral window is integrated and sent to a DAQ card (Measurement Computing DT3016) which transduces the signal to an analog voltage that is read by the R9*plus*. In this way, optical images can be correlated with topographic information in the R9*plus* software with fast acquisition speeds of <50 ms/pixel.

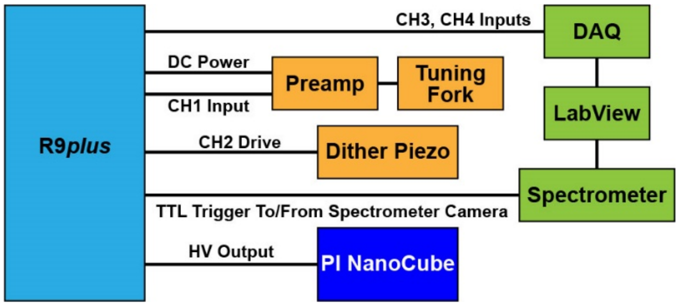

Figure 1. Wiring schematic of positioners, shear force feedback hardware, and optical signal hardware with the R9*plus*.

Experimental Setup

A schematic of the TEPL spectroscopy setup is shown in Figure 2a. For the experiments, the sample was mounted to the PI nanopositioner with sub-nm precision positioning below an electrically driven and controlled quartz tuning fork (32 kHz resonance frequency), which was used to regulate the tip-sample distance using the AFM shear-force amplitude feedback. Electrochemically etched Au tips (5–10 nm apex radius) were attached to the tuning fork and coarsely positioned using a piezoelectric stepper motor. In the TEPL spectroscopy setup, the sample was mounted at a 35° angle to the tip axis to maximize the electric field confinement. Excitation was provided by a helium-neon laser beam (632.8 nm, <1 mW) with a half wave plate for polarization control that was focused onto the tip-sample interface using an objective lens. Photoluminescence signal was collected in a backscattering geometry, passed through a dichroic mirror with a 633 nm cut-off, and sent to a spectrometer (SpectraPro 500i, Princeton Instruments) with a thermoelectrically cooled, electron-multiplied, charge-coupled device (CCD, ProEM+: 1600 eXcelon3, Princeton Instruments). The spectrometer was calibrated using a hydrogen mercury lamp and a 150 g/mm grating blazed for 800 nm was used to provide high bandwidth spectral information.

Radiative Control of Dark Excitons

Excitons (Coulomb-bound electron-hole pairs) are elementary photoexcitations in semiconductors that can couple to light through radiative relaxation. In contrast, dark excitons show anti-parallel spin polarization with generally forbidden radiative emission (Figure 2b). Prof. Raschke's group demonstrated a tip-enhanced nano-optical approach to observing dark excitons with out-of-plane transition dipole orientations at room-temperature in WSe_2 with Purcell factor control through the tip-sample nano-cavity (Figure 2c). With all previous approaches relying on cryogenic temperatures, this approach provides a new and facile way to harness

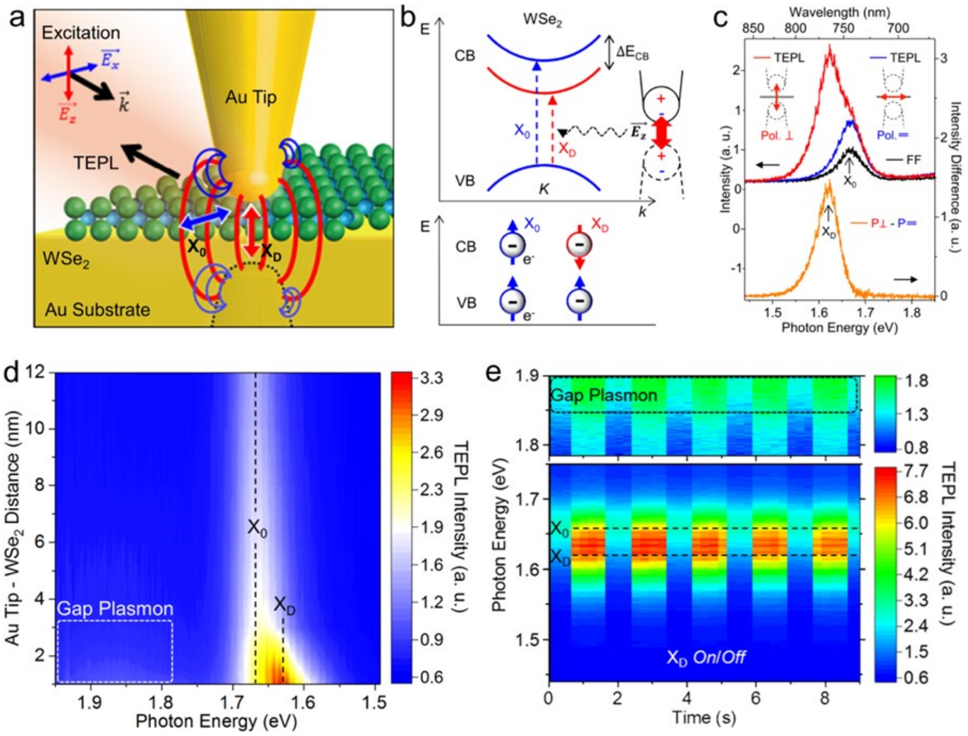

Figure 2. (a) Schematic of the TEPL spectroscopy setup. (b) Physical model of dark excitons. (c) Spectral signatures of dark excitons (X_D) and bright excitons (X_0). (d) Spectral evolution of dark exciton PL as a function of tip-sample distance exhibiting precise control by the R9*plus*. (e) Demonstration of controlled enhancement of the dark exciton signal by modulation of the AFM tip position.

excitonic properties in low-dimensional semiconductors for quantum optoelectronics and other applications.

In addition, this tip-enhanced nano-optical approach allows the scientists to switch and programmatically modulate the dark exciton emission of a monolayer WSe$_2$ at room temperature for the first time. Using the plasmonic nano-cavity, they demonstrated a $\sim 6\times10^5$-fold enhancement in dark exciton photoluminescence quantum yield achieved through coupling of the antenna-tip to the dark exciton out-of-plane optical dipole moment, with large Purcell factor of 2×10^3. They then controlled this Purcell factor by precisely changing the tip-sample distance to decrease the near-field interaction (Figure 2d) and leveraged this technique to programmatically modulate the dark exciton signal by precisely controlling the tip-sample nano-cavity in time and space (Figure 2e).

These experiments demonstrate the ability to measure optical signals that are not accessible through room temperature far-field optical measurements and the ability to control these signals through precise tip-sample positioning.

Why They Use RHK R9*plus*

Accessible, user-defined inputs allow for seamless integration of externally measured signals. This allows the users to pre-characterize the sample by measuring the far-field optical signal to identify regions of interest on the sample before engaging in shear force feedback.

Optical signals can be easily correlated to topographic features due to R9*plus*' ability to obtain real-time imaging of multiple signal channels such as topography, TERS, SHG, TEPL, FWM, etc.

The R9*plus* software has an easy to use GUI interface which enables flexible region of interest selection (user defined XY pixel number) with features like drift correction and image referencing.

Numerous digital inputs and outputs are available to trigger external instruments required for their experiments (e.g. the spectrometer camera). This allows synchronized data collection between AFM signals and spectrometer data channels.

Setting up a Phase-Locked Loop (PLL) is not typically a simple task as there are feedback loops that need to be correctly tuned for proper function and to avoid a tip crash. However, the R9*plus* software not only makes it easy to find the resonance frequency of the tuning fork, but the built-in feedback tests used to properly adjust the feedback parameters of the PLL make the process of PLL setup extremely simple.

The R9*plus* also has a highly advanced "Lock Guard" capability that helps protect the probe from damage by continuously monitoring up to six data channels, retracting the probe within microseconds when detecting any out-of-bounds condition.

The experimental design is constantly evolving. Since everything is controlled digitally, the R9*plus* allows easy reconfiguration of the setup. Often times, only simple modifications to the IHDL® file are required to define a new experimental design which minimizes the need to change external wiring.

Future Implementations of R9*plus* Capabilities

There are many capabilities of the R9*plus* that have not been explored in this Application Note that Prof. Raschke's group plans to implement in the future. One example is the implementation of the dual scan capability. They currently use separate stages for both tip and sample but would like a better way to control these stages in a streamlined manner. With the dual scan capability and the numerous feedback loops available in the R9*plus*, they will be able to choose between scanning the sample stage (for larger range scans) and the tip (for smaller range, higher resolution scans) all within the user interface of the R9*plus* software.

An added benefit to the dual scan feature they foresee is the ability to quickly find the laser "hot spot" on the sample for optimal laser alignment. By scanning the tip position and monitoring optical feedback, the laser "hot spot" can be quickly located and the tip can be moved to a location within that "hot spot".

Another feature they hope to explore further is the Q-control capability for tuning the quality factor (Q) of the tuning fork. Q-control is particularly useful since a large Q factor results in slower response times with amplitude feedback detection, leading to undesirable scanning times. Having Q-control eliminates this issue and allows measurements to be taken in a wide range of temperatures and in ultra-high vacuum conditions.

Additional Information Related to This Work

Park, K.-D. et al. Radiative control of dark excitons at room temperature by nano-optical antenna-tip Purcell effect. Nat. Nanotechnol. 13, 59–63 (2018).

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