



The impure hearts of semiconductors

Inspired by the rapidly evolving field of quantum technology, the investigation of photon generation from single-quantum emitters has evolved into a fundamental research area of its own with substantial potential applications. Much of this research is founded on the ability of these single-photon delivery systems to be used for the ultra-secure encoding of information and the creation of qubit circuitry. Semiconductors have become the field's materials of choice in such studies, as single-atom impurities in heterogeneous semiconductors form naturally occurring ultimate quantum dots. Gallium arsenide (GaAs) containing nitrogen pairs have been shown to fall in this class of optoelectronic materials.

Resolving luminescent centres both spectrally and spatially can reveal critical information about the physics of semiconductor impurities – information that would be otherwise masked by ensemble averaging. The measurement of single-molecules embedded in a host by optical spectroscopy requires two fundamentals to be satisfied. First, the density of the embedded molecules must be such that only one luminescent entity is located within the excitation or detection volume. Next, the photon emission rate from a single recombination centre must be fast enough to provide a signal exceeding noise levels.

Photoluminescence Mapping

At the National Renewable Energy Laboratory (Colorado, US) and the Sandia National Laboratory (New Mexico, US), the possibility of measuring luminescence from single impurity centres in a GaAs semiconductor was recently demonstrated using optical spectroscopy with diffraction limited spatial resolution. Samples were grown using molecular beam epitaxy, with structures corresponding to a 25nm GaAs:N layer, clad by a protective 5nm stratum of GaAs on both sides, and surrounded by two $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ barriers. From secondary ion mass spectrometry, the nitrogen concentration was estimated to be $3 \times 10^5 \mu\text{m}^{-3}$, corresponding to a pair surface density of $0.6 \mu\text{m}^{-2}$.

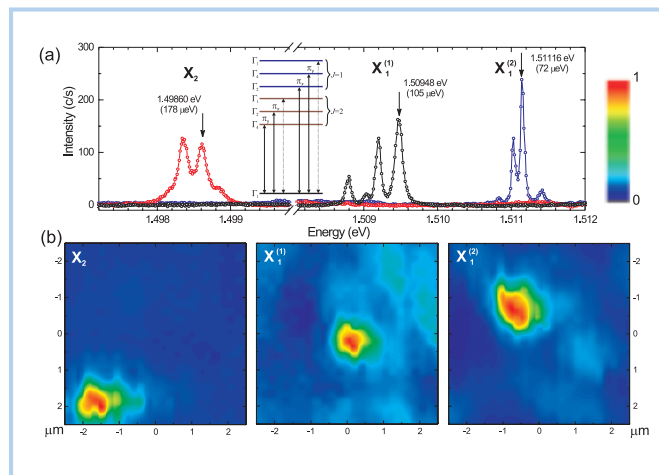


Figure 1 (a) Photoluminescence spectra from three regions of a $25 \mu\text{m}^2$ spatial map. $X_1^{(1)}$, $X_1^{(2)}$, and X_2 correspond to three nitrogen pairs. X_1 and X_2 are pairs with discrete separation. Inset: linearly polarized optical transitions for C_{2v} symmetry. (b) Photoluminescence intensity maps corresponding to $X_1^{(1)}$, $X_1^{(2)}$, and X_2 . Intensity is plotted for the luminescence peaks shown by arrows in (a).

To obtain submicron resolution signal measurements and perform spatial mapping of the semiconductor microstructure, the research team required a cryostat that could maintain exceptional stability and allow for the long integration times necessary for the detection of single quantum emitters. For this, the investigators turned to the MicrostatHiResII from Oxford Instruments. The MicrostatHiResII is a vacuum loading continuous flow cryostat that benefits from extremely low sample vibration and sample drift at both constant and changing temperatures. The system also met the study's cold temperature requirements of 5 K (operating capability of 2.7 K to 500 K) and its design provided the large numerical aperture essential for the use of the high-magnification optics necessary to achieve submicron spatial resolution. Experimental luminescence excitation was performed at 532 nm using an intensity of 90 nW, with a resolution of $0.5 \mu\text{m}^2$.

A photoluminescence map was created, which yielded two types of radiative centres, arbitrarily designated X_1 and X_2 . As shown in Figure 1(a), the higher energy centre, X_1 , can be found under different configurations, here labelled $X_1^{(1)}$ and $X_1^{(2)}$, and like X_2 , they correspond to nitrogen impurity pairs. Figure 1(b) displays spatial intensity maps for each centre, measured at the energy of the luminescence features indicated by the arrows on the spectra in Figure 1(a). These maps demonstrate that high spatial resolution can indeed be used to probe for the distribution of single impurity centres in a semiconductor. Furthermore, by mapping the

photoluminescence intensity of each point as a function of energy and polarisation angle, the four transitions allowed by the symmetry (C_{2v}) of the emitters was also resolved. For each radiative centre, it was found that the orientational degeneracy was lifted, as expected for single-photon emitters.

Resolving the Future

In a later segment of the study, the investigators concluded that by mapping the variations of energy in each point and by associating the experimental results with theoretical models, luminescence energy, exchange interactions, and crystal-field splitting, the presence of strain and electrical fields could be studied. This approach has practical applications for manufacturers of ultra-small transistors and diodes, as it could be used to provide valuable information on dopant distribution statistics in such devices.

Importantly, the approach outlined here also provided the researchers with a unique method of probing semiconductor impurities with a level of resolution and sensitivity previously unattainable with other techniques. In future, it is expected that the technique will be used for highly sensitive impurity distribution mapping and for the detailed analysis of host characteristics.

Reference:

Francoeur S, Klem JF, and Mascarenhas A. *Optical Spectroscopy of Single Impurity Centres in Semiconductors*. *The American Physical Society: Physical Review Letters*. 2004:93(6).

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