Optical properties of isoelectronic centers in crystalline silicon

Vinh, N.Q.

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Chapter 2
Experimental Techniques

This chapter concerns instrumental and experimental details of photoluminescence spectroscopy, time-resolved measurements, photoluminescence excitation, two-color spectroscopy and magneto-optical measurements used in the investigations described in this thesis.

2.1 Photoluminescence and time-resolved experiments

Photoluminescence (PL) is the spontaneous emission of light from a material under optical excitation. The excitation energy and intensity can be chosen to probe different excitation types and also different parts of the sample. PL investigations can be used to characterize a variety of materials parameters. Features of the emission spectrum can be used to identify surface, interface, and impurity levels...

Under pulsed excitation, PL intensity transients yield lifetimes of excited states. In addition, thermally activated processes of non-radiative recombination cause changes of PL and can be investigated in this way.

PL analysis is nondestructive. The technique requires very little sample manipulation or environmental control. When light of sufficient energy is incident on a material, photons are absorbed and (electronic) excitations are created. These excitations relax and the system returns to the ground state. The PL can be collected and analyzed to provide information about the photo-excited states. The PL spectrum reveals transition energies. The PL intensity gives a measure of the relative rates of radiative and non-radiative recombinations. Variation of the PL intensity upon change of external parameters, e.g., temperature, can be used to further characterize electronic states and bands.

The typical PL experimental set-up used in our laboratory is illustrated in Fig.
2.1. The PL experiments were carried out in a variable temperature continuous-flow cryostat accessing the 1.5 – 300 K range (Oxford Instruments Optistat CF). The samples were excited using a continuous-wave (cw) argon-ion laser operating at 514.5/488/351 nm, or a tunable Optical Parametric Oscillator (OPO) laser system, producing pulses of 5 ns duration at 20 Hz repetition rate. The luminescence was resolved with a 1 m F/8 monochromator (Jobin-Yvon THR-1000) equipped with a 900 grooves/mm grating blazed at 1.5 μm and detected by a high sensitivity germanium detector (Edinburgh Instruments) or a Hamamatsu R5509-72 InP/InGaAs nitrogen-cooled photomultiplier tube.

Time-resolved measurements were carried out using a Tektronix TDS 3000 digital oscilloscope in combination with the Hamamatsu detector. The experimental response time of the system was $\tau_{dec} \approx 3$ ns for the Hamamatsu detector at 50 Ω; $\tau_{dec} \approx 350$ ns at 1 kΩ; $\tau_{dec} \approx 35$ μs at 100 kΩ and $\tau_{dec} \approx 400$ μs at 1 MΩ. The PL response could be integrated in time using a digital oscilloscope (TDS 3032, Tektronix). In the cw mode, excitation modulation was achieved by using a pinhole and chopping the laser beam mechanically at 200 Hz. In these cases, the PL spectrum was recorded by using a lock-in amplifier. The time resolution was measured as 15 μs using 1 kΩ setup.

A program called "OptiLab" was developed for remote operation of the experimental setup. The program uses a GPIB card and a serial port (RS-232) to control: the high resolution spectrometer, variable temperature cryostat, lock-in amplifier, ADC and laser. The software was written in C language and CVI program under Windows environment.
2.2 Photoluminescence excitation spectroscopy

In PL spectroscopy, which is performed at a fixed excitation energy, the luminescence properties are generally investigated, while photoluminescence excitation spectroscopy (PLE), which is carried out at a fixed detection energy, provides mainly information about the absorption properties and probes excited states of optical centers. In this experiment, the sample is excited by photons of the energy $h\nu$ tuned to be absorbed by specific optical centers. Therefore, this technique requires a tunable excitation source. In our setup, for this purpose the OPO has been used. Its emission wavelength can be tuned in wavelength range (400 nm to 2200 nm). The high resolution PLE, i.e., the combination of highly resolved PL spectra as a function of the well-defined excitation energy ($\Delta E \approx 1\text{meV}$), provides detailed information about the system under investigation.

2.3 Two-color spectroscopy

Shallow excited states in semiconductor can be conveniently investigated by two-color spectroscopy. In these experiments, one beam with sufficient quantum energy is used to create a large population in the excited state. The second mid-infrared “probe” beam (with the photon energy of the order of 10 to few hundred meV) is used to induce transitions within the excited state of the system.

The idea of the two-color mid-infrared (2C-MIR) experiments is presented in a cartoon in Fig. 2.2. The pump beam from the second harmonic of a Nd:YAG laser (532 nm) with a repetition rate of 5 Hz and a pulse duration of 100 ps provides band-to-band excitation leading to emission from the optical dopant. A mid-infrared (MIR) probe pulse with duration of $\sim 5\mu$s is provided by a free-electron laser (FEL). The FEL can be activated with a selected time delay $\Delta t_F$ with respect to the Nd:YAG laser pulse and its wavelength is tunable. Measurements are carried with the sample fixed on the cold finger of a flow cryostat in a variable temperature range from 4.2 to 300 K. This technique is very useful for tracking energy transfer paths of optically doped semiconductor matrices, for ionization of shallow centers, and for investigation of thermally activated recombinations.

The two-color experiments described in this thesis (Chapter 4) have been performed at the Dutch Free Electron Laser facility for Infrared Xperiments (FELIX) at the FOM Institute for Plasma Physics “Rijnhuizen” in Nieuwegein [8]. The user facility provides continuously tunable radiation in the spectral range of 4 – 250 $\mu$m, at peak powers up to 100 MW in a (sub)picosecond pulse. The infrared beam features short micropulses, which have a nominal duration of 1 ps and are separated by intervals of either 1 ns (1 GHz) or 40 ns (25 MHZ). - see inset to Fig.
Figure 2.2: Experimental configuration for two-color experiments. The band-to-band excitation is provided by a second harmonic of a ND:YAG laser. The MIR beam is provided by FEL.

2.2. The micropulses form a train with a total duration of up to 10 $\mu$s.

2.4 Magneto-optical spectroscopy

This technique comprises a simultaneous application of an optical excitation and a magnetic field, and is generally named Zeeman spectroscopy. It can be very informative in case when narrow spectral lines are observed in optical transitions. The usual requirement is for the sample to be at a low temperature and in a high magnetic field with good optical access, so that it can be excited with laser radiation and luminescence can be collected for analysis with a high resolution spectrometer. Upon application of magnetic field, a splitting of individual PL bands will occur. The details of this splitting, multiplicity and g factor for each particular transition, provide information on orbital angular momentum and spin of the center. The necessary condition is that the magnetic field must be such that the linear Zeeman term $H = \mu_B \cdot g_{\text{eff}} \cdot S_{\text{eff}} \cdot B$ is significantly larger than the natural linewidth observed. In experimental practice, magnetic fields of 5 T or higher from a superconducting magnet are used.

The magneto-optical setup available in our laboratory is shown in Fig. 2.3. The magnet with optical access (Spectromag 8) up to 6 T is immersed in liquid helium. The samples are excited by radiation from an Ar$^+$ ion laser operating at
2.4. Magneto-optical spectroscopy

Figure 2.3: Diagram of a magneto-optical spectrometer which is suitable for magneto emission studies. It is consist of, mainly, a superconducting magnet, a high resolution spectroscopy, a laser, a λ/4 plate and a linear polarizer.

514.5 nm, and the luminescence is analyzed with a 1.0 m or 1.5 m F/8 high resolution monochromator (Jobin-Yvon THR-1000/THR-1500 equipped with a 900 grooves/mm grating blazed at 1.5 μm) and detected by a high sensitivity germanium detector (Edinburgh Instruments). Temperature of the samples was varied from 1.5 – 300 K. The experimental setup is operated by remote control since access to the magnet room is usually restricted when the magnet is in operation.

For experiments, different geometries may be used: with \( \vec{k} \parallel \vec{B} \), Faraday configuration, gives PL of circular polarization \( \sigma^+ \) and \( \sigma^- \), the Voigt configuration, \( \vec{k} \perp \vec{B} \), yields polarization \( \sigma \) and \( \pi \) (Here \( \vec{k} \) is propagation vector of the light). The polarization is very important, because most of the optical transitions are polarized according to the corresponding optical selection rules. The analysis of circular polarized light can be made by using a λ/4 plate and a linear polarizer.