Resolved Fine Structure of the Boron Bound Exciton in Highly Enriched ^{28}Si Using Single Frequency Laser Spectroscopy

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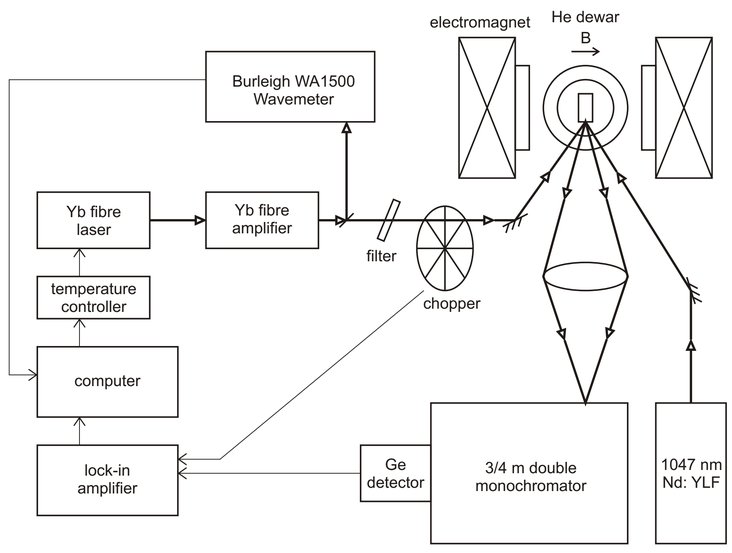
Introduction

While the first comparison of shallow bound exciton photoluminescence (PL) between natural Si and highly enriched ^{28}Si dramatically demonstrated the importance of inhomogeneous isotope broadening, the transitions in ^{28}Si were in fact too narrow to be resolved with the then available instrumental resolution of 1.7 μeV (0.014 cm^{-1}) [1]. We report new results for the boron bound exciton (BE) transition in highly enriched ^{28}Si using a novel apparatus for photoluminescence excitation spectroscopy (PLE) based on a tuneable single frequency laser source with sub-MHz resolution.

Experiment

In order to overcome the resolution limit imposed by commercial spectrometers and the weak luminescence signals characteristic of BE in Si, the no-phonon transitions of the BE are studied in absorption rather than in emission, using a single-frequency laser source with a linewidth of less than 0.3 neV (more than adequate to resolve BE linewidths at their fundamental limit – shallow BE lifetimes in Si are in the 100 ns to 1 μs range [2]). The weak absorption and resulting luminescence signal are detected using the trasverse optical (TO) wavevector conserving phonon replica, which is well separated in energy from the no-phonon transition (~ 58 meV lower in energy). The experimental apparatus used for the present study is shown below.



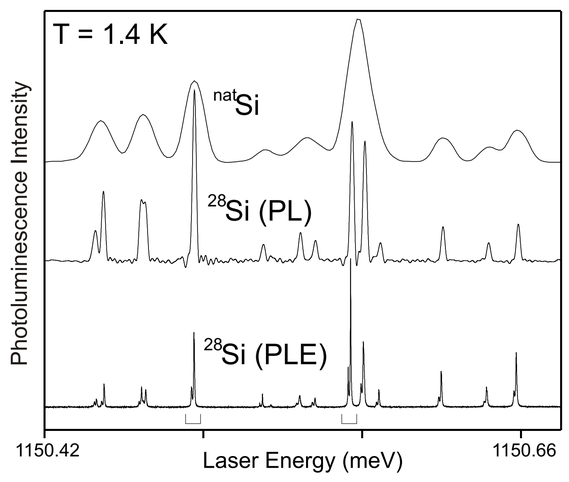
PLE apparatua based on a tunable single frequency laser source

An improved sample of ^{28}Si with an isotopic enrichment of 99.991% and much higher chemical purity (~2×10^{12} cm^{-3} phosphorus, ~5×10^{14} cm^{-3} boron, and <5×10^{14} cm^{-3}) was made available for the present study.

A distributed feedback Yb-doped fibre laser was temperature tuned over the region of interest and the laser frequency was monitored (to one part in 10^{7}) using a wavemeter. A Yb-doped fibre amplifier was used to amplify the laser output to 500 mW, which was then mechanically chopped to allow for lock-in detection of the signal, and focussed onto the edge of the sample. The samples were immersed in a liquid He bath and mounted loosely in a reflecting cavity. A 3/4 m double monochromator separated the luminescence signal from the scattered excitation radiation, and the signal was then detected using a liquid nitrogen cooled Ge photoconductive detector.

Comparison of ^{nat}Si and ^{28}Si

The no-phonon region of the boron BE PL spectrum for natural Si and ^{28}Si, along with that of the PLE spectrum for ^{28}Si, are shown in the figure below.

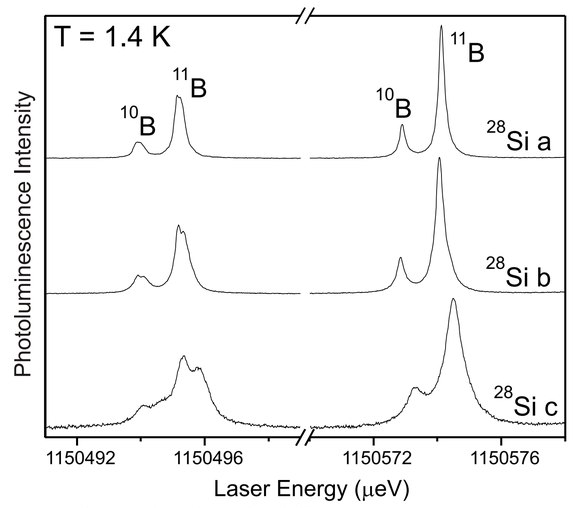


Comparison of boron BE PL spectra of ^{nat}Si and ^{28}Si in the no-phonon region

The ground state of the boron BE has been observed to split into nine components in natural Si (top spectrum), with an additional five components becoming resolved in the earlier photoluminescence spectra of ^{28}Si (middle spectrum), while seventeen distinct ground state components can be observed in the new spectra (bottom spectrum). The narrowest transitions observed using the new apparatus have a FWHM of only 0.22 μeV (0.0018 cm^{-1}) (ΔE/E = 5×10^{6}). In addition, each of these BE components is observed to consist of a doublet with an identical splitting of 0.011 cm^{-1}, and an identical intensity ratio of 80/20. We conclude that these doublets arise from a 1.36 μeV (0.011 cm^{-1}) difference in the binding energy of excitons localized on ^{11}B and ^{10}B acceptors, since the natural abundances mirror the 80/20 intensity ratio, and the difference in BE binding energies is in agreement with the expectation from Haynes' Rule in Si [3] based on the 18.6 μeV (0.15 cm^{-1}) difference in acceptor ionization energy previously observed between ^{11}B and ^{10}B in ^{28}Si [4]. While isotope splittings have previously been observed for much deeper BE in other semiconductors [5], this is the first observation of an isotope splitting for such a shallow BE transition.

Isotope broadening in ^{28}Si

Selected boron bound exciton components of three samples of ^{28}Si with different isotopic enrichments are compared in the figure below.



PLE spectra of two of the B BE no-phonon components for three samples of different isotopic enrichment

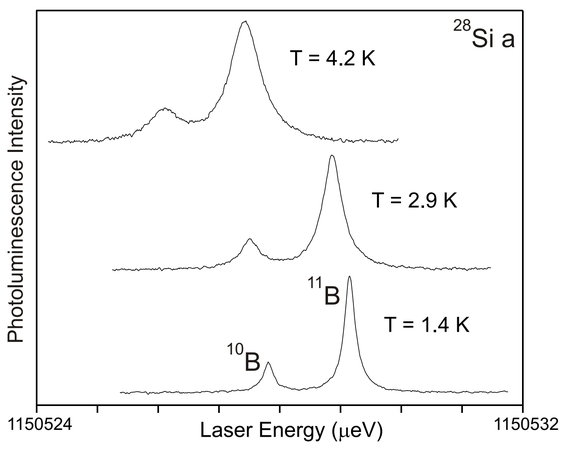
Note that the components become sharper as the isotopic enrichment increases, and in the lower energy doublet, the splitting of the ^{11}B component also becomes smaller, in addition to the observed sharpening. This was observed for Al, Ga, and In BE [6] in comparisons between ^{nat}Si and ^{28}Si and was attributed to the isotopic randomness in ^{nat}Si. Ignoring the small ^{29}Si and ^{30}Si compositions in the ^{28}Si samples, the inhomogeneous isotope broadening will vary as the square root of 1 minus the enrichment. Comparing this with the splitting of the lower energy ^{11}B component and the linewidth of the higher energy ^{11}B component results in remarkably good agreement. Thus, the observed splitting of the ^{11}B component in the most highly enriched (99.991% ^{28}Si) sample indicates that the fundamental limit has not yet been reached and the linewidth is still limited by inhomogeneous isotope broadening.

^{28}Si enrichment	low-E ^{11}B splitting	high-E ^{11}B FWHM	(1-enrichment)$^{1/2}$
0.99991	0.15 μeV (1)	0.24 μeV (1)	0.00949 (1)
0.99983	0.2 μeV (1.33)	0.35 μeV (1.46)	0.01304 (1.37)
0.9992	0.43 μeV (2.87)	0.72 μeV (3)	0.02828 (2.98)

Comparison of splitting of low-energy ^{11}B component and linewidth of high-energy ^{11}B component for samples of different isotopic enrichment

Variation of linewidth with temperature

A comparison of one of the doublets in the boron BE spectrum at various liquid He bath temperatures (measured by monitoring the He vapor pressure) reveals a narrowing of the transitions with decreasing temperature as shown in the figure below. The observed energy shifts are in agreement with the low-T behaviour determined by Cardona, Meyer, et al. [7].



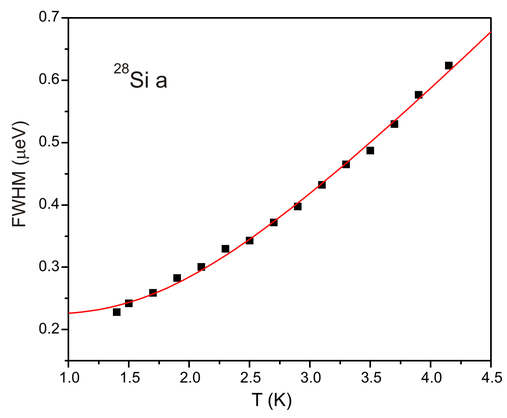
Comparison of one of the components of the boron BE PL spectrum at various liquid He bath temperatures

A plot of the linewidth of the ^{11}B component against temperature is shown below. Assuming two contributions to the total linewidth of this transition: a temperature independent component plus a temperature dependent component, which is proportional to the Bose-Einstein factor, the linewidth versus temperature data can be fit to an expression with the following functional form:

$$FWHM = (A^2 + (B/(\exp(C/kT)-1))^2)^{1/2}$$

where A and B are the linewidths of the temperature independent and dependent components, respectively, and C is an activation energy.

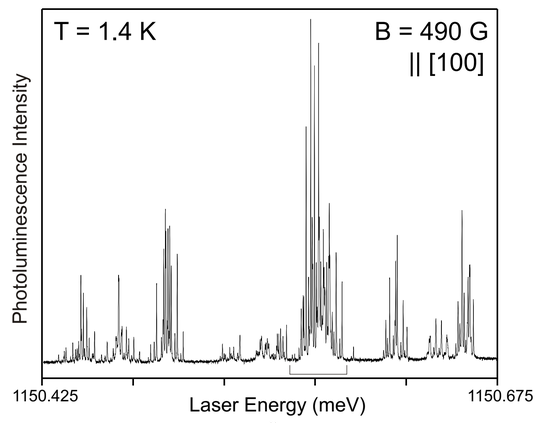
The fit (shown as a red curve in the figure below) yields a value for C of 0.26 meV, which is on the order of the width of the energy range spanned by the boron BE ground state components. This would seem to suggest that the lifetimes of the boron BE components are determined by thermally induced transitions between different boron BE states.



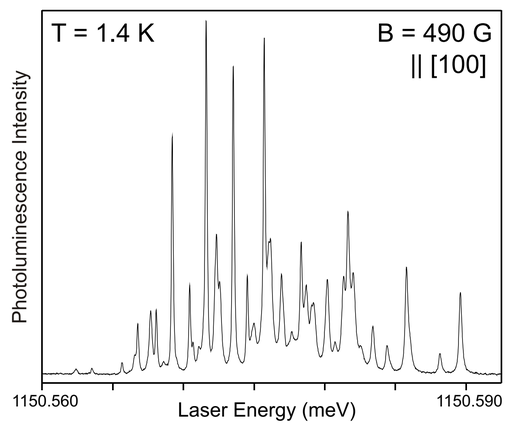
Plot of linewidth of the ^{11}B component against temperature along with fit (shown as red curve)

Boron BE under magnetic field

A PL spectrum of the boron BE in ^{28}Si under an applied magnetic field of 490 G, in which approximately 76 components are observed, is shown in the top figure below. A magnification of the central region of the spectrum is shown in the bottom figure illustrating the density of the boron BE components. The splitting of the boron BE ground state transitions has previously been studied in detail in natural Si under uniaxial stress [8] and magnetic fields [9]. In the absence of all perturbations, 144 components of the boron BE are expected.



PL spectrum of boron BE in ^{28}Si in 490 G magnetic field



Magnification of central region

Acknowledgments

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