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List of Tables

0.0.1 Sideband

It is known that the photoluminescence spectra of SiV centers in nanodiamond are dominated by the zero-phonon-line. As a result phonon side band contributions remain small, a fact expressed in large Debye-Waller factors of over 70 % established previously [1, 2]. Our own measurements are consistent with these of emitter H1 and emitter V1 results. We also find distinct sideband peaks in many SiV center photoluminescence emission spectra. The investigated emitters exhibit two different structures of sideband spectra: The spectra in group V exhibit one strong sideband peak, spectra in group H exhibit several weaker sideband peaks. Figure 1a and Figure 1b illustrate the respective observations.

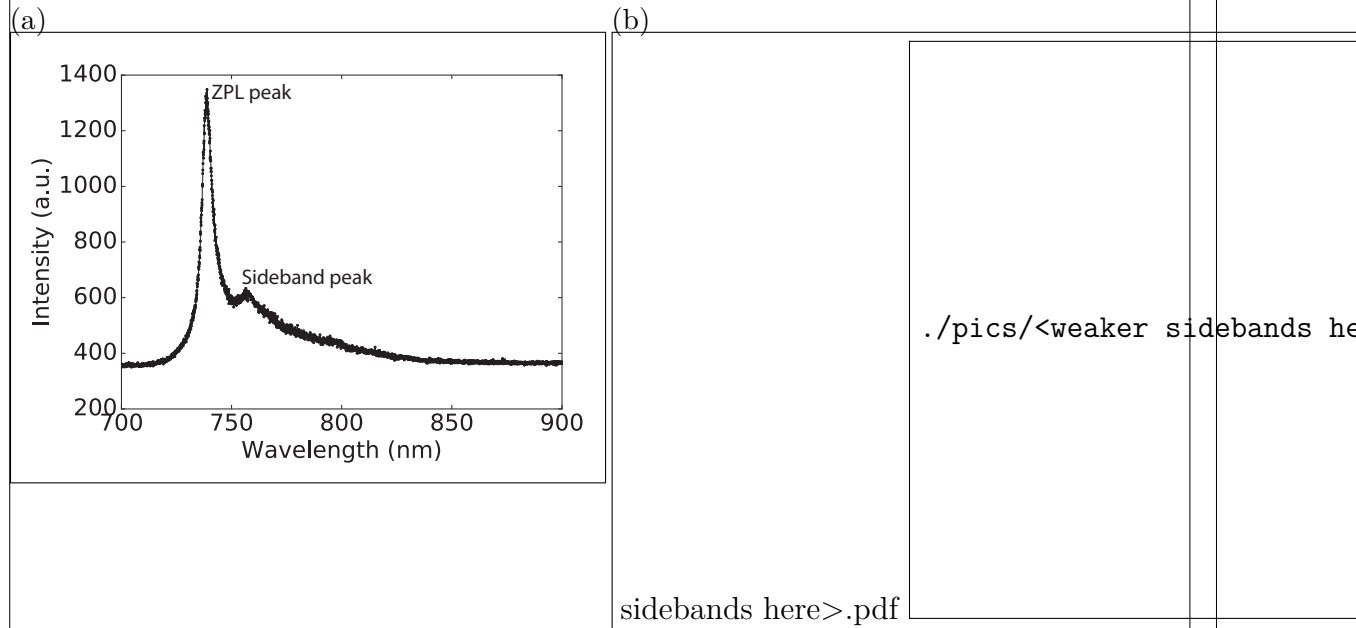


Figure 1: Representative spectra of emitters showing single (a) and multiple (b) sideband peaks. The former belong to group V, while the latter are members of group H.

Most of the spectra in group V exhibit a characteristic shape, composed of the ZPL and one strong sideband peak. 70 % of the photoluminescence spectra with one distinct sideband peak exhibit a shift of the sideband peak from the ZPL between 37 meV to 43 meV. The range of line shifts for the prominent sideband peak coincides with a well-known feature at 42 meV, associated with SiV centers [3, 4], but also to a larger number of optically active defects [4]. The occurrence of this 42 meV sideband feature for a large number of defects and the absence of isotopic variations [5], favors an assignment as non-localized lattice vibration. We furthermore observe that the dominant sideband peak shifts towards smaller distance from the ZPL for increasing ZPL center wavelength, i.e. increasing strain. Figure 2 presents a linear fit to data for emitters in group V.. The low phonon energy of the sideband feature and its shift with strain might arise from a local “softening” of the crystal lattice in the vicinity of a defect [4].

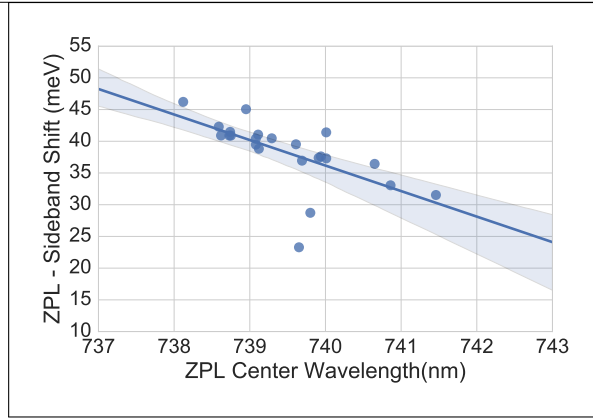


Figure 2: Shift of dominant sideband peak from the ZPL in spectra of SiV centers (group V, samples insitu50, insitu70, insitu100) vs. ZPL center wavelength. The linear fit shows that the shift decreases with increasing ZPL center wavelength, i.e. with increasing strain and exhibits a slope of $(-4 \pm 1) \text{ meV nm}^{-1}$. The shaded area is the 95 % confidence region.

A recent study [6] suggests that the 42 meV mode, similar to other broad phonon side band features, originates from a resonance attributed to phonons causing the dynamical Jahn-Teller effect with SiV centers [7]. As the Jahn-Teller coupling varies with strain it is also expected that the resonance shifts accordingly.

In the spectra of group V, we do not observe a typical SiV center sideband feature at 64 meV, attributed to a local vibration of the silicon atom, frequently much stronger than the 42 meV sideband peak. A possible explanation is, that the lattice mode at 37 meV to 43 meV is so strong that the local vibrational mode at 64 meV cannot be separated from the tail of the lattice mode.

In group H we observe many spectra which exhibit several peaks within the spectral range of our detection range between 710 nm to 900 nm. The challenge arises to unequivocally distinguish between peaks stemming from a phonon sideband and peaks stemming from shifted, less intense SiV center ZPLs.

Interestingly, we assert a tendency for peaks to accumulate at a shift of around 43 meV, 64 meV, 150 meV and 175 meV. This pattern in the phonon side band of group H is consistent with side band shifts reported in [4, 8, 1].

The possibility exists, that some these peaks believed to be phonon side bands are actually shifted ZPLs stemming from other SiV centers. To address this question, we perform photoluminescence measurements at cryogenic temperatures.

0.0.2 Cryostatic Measurements

Remark:

den teil mit den cryogenic measurments musst du nochmal ordentlich aufschreiben.

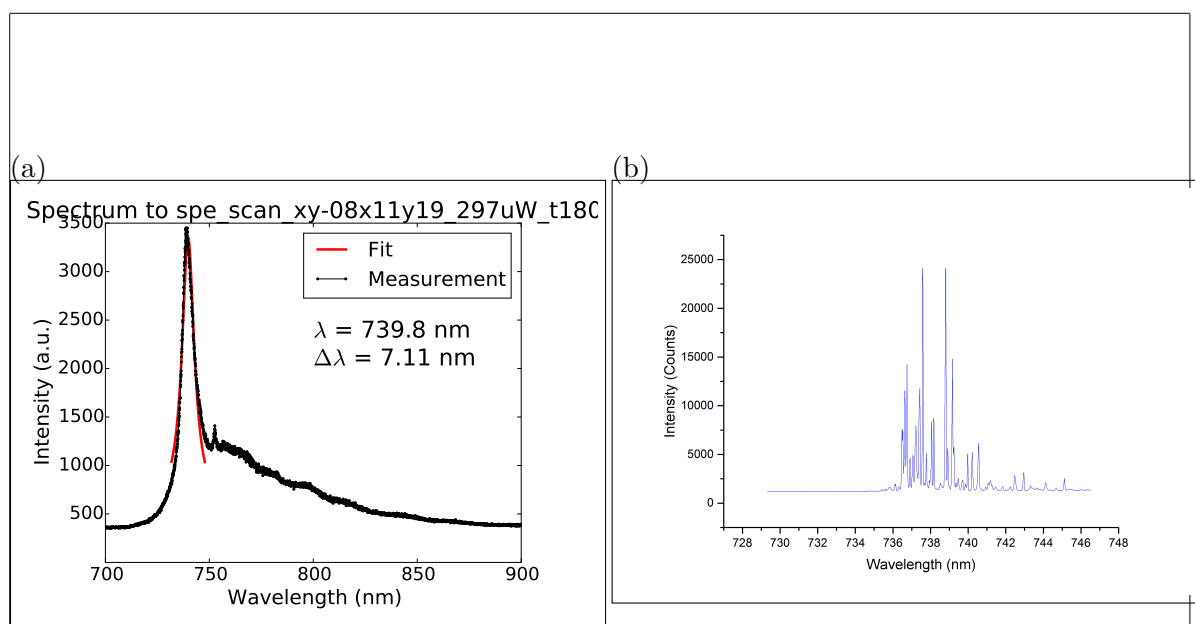


Figure 3

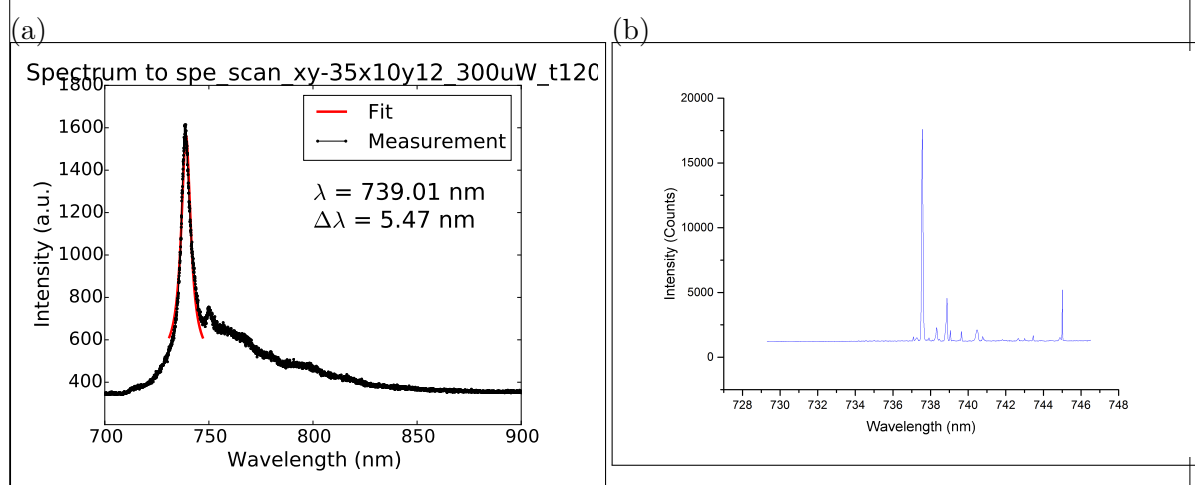


Figure 4

Hier fehlen noch dinge.

- Wie hast du messungen ueberhaupt machen koennen? Das setup in ?? spricht ja nicht von tieftemperaturen. Hast du den aufbau von jemand anderem (janine?) benutzt? Wenn ja dann unbedingt erwaehnen. Ansonsten erklaren wie sich der genutzte aufbau vom normalen unterscheidet.
- die bilder die du da bereits eingefuegt hast, die versteh ich nicht. Sind das nur platzhalter und da kommt noch was anderes hin? Ich seh den zusammenhang die geschichte mit der feinstruktur nicht. Das sollte noch geklaert werden.
- wenn man die 4-fach aufspaltung nicht sieht und stattdessen mehrere linien hat, was bedeutet das? Klar ist, man hat kein einzelnes SiV center am start. Abgesehen davon, laesst sich noch was anderes sagen? Wenn man den wood vor lauter lines nicht sieht, dann heisst das auch, dass man nicht sagen kann ob manche der phonon side band peaks von anderen SiV centers kommen oder nicht. Wenn hier keine konklusive aussage moeglich ist, dann sollte das diskutiert werden.
- die sache mit den multiple lines that were strained is mir nicht klar. Soll das heissen die waren verschoben und der grund dafuer war strain? wenn ja, verschoben gegenueber welchem wert, man erwartet ja 4 linien fuer SiV centers.

We performed some cryostatic measurements to to pursue two goals: First, if we see the four-level splitting of the ZPL this is further proof that the dominant peaks of our spectra are indeed all due to SiV centers and do not stem from other impurities. Second, if the sideband peaks vanish at cold temperatures, this is evidence that they are caused by phonons. So we cooled down a sample, but all we saw was a wood of lines. Apparently, there were multiple SiV centers in the nanodiamonds and they were all strained, messing up the four-level line structure.

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