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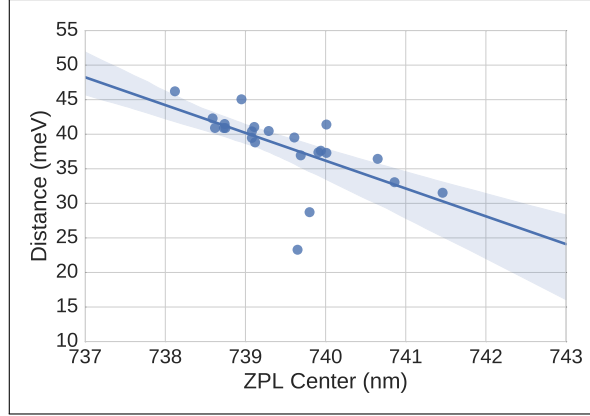


Figure 1: Shift of dominant sideband peak from the ZPL in spectra of SiV centers (group V, samples insitu50, insitu70, insitu100) vs. ZPL center wavelength. The linear fit shows that the shift decreases with increasing ZPL center wavelength. The shaded area is the 95% confidence interval.

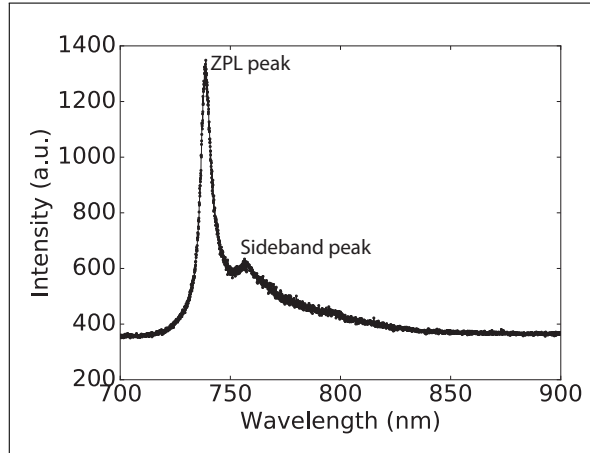


Figure 2: Representative spectrum of an emitter of group V exhibiting a sideband peak.

0.0.1 Sideband

As mentioned before, phonons of the diamond lattice and the color center manifest themselves as peaks in the sideband of the photoluminescence spectrum. Phonons reduce the intensity of the purely electronic transition, the zero-phonon-line. The electron-phonon coupling is quantified by the Debye-Waller factor or the Huang-Rhys. The former is the integrated intensity of the zero-phonon-line I_{ZPL} divided by the integrated intensity of the photoluminescence of the whole spectrum, i.e. ZPL plus sidebands, I_{tot} [?, ?]. The Huang-Rhys factor S is defined as $I_{ZPL}/I_{tot} = \exp\{-S\}$.

From literature it is known, that the SiV center in nanodiamond exhibits a large Debye-Waller of over 70% [?, ?], which is consistent with our measurements of emitter H1 and emitter V1. Nevertheless, sideband peaks are present in many SiV center PL emission spectra. The investigated emitters exhibit two different structures of sideband spectra: The spectra in group V exhibit one strong sideband peak (Figure 2), spectra in group H exhibit several weaker

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sideband peaks.

However, there is no recurring pattern in the sideband of group H. The challenge arises to unequivocally distinguish between peaks stemming from a phonon sideband and peaks stemming from shifted, less intense SiV center ZPLs. The possibility exists, that some peaks identified as phonon sidebands are actually ZPLs stemming from shifted SiV centers. Therefore, we will focus our investigations on the more prominent sideband of group V.

Most of the spectra in group V exhibit a characteristic shape, composed of the ZPL and one strong sideband peak which is mostly shifted 37 meV to 43 meV. In reference [?] the 42 meV sideband peak is attributed to a non-localized (lattice) mode. It is also stated, that the local vibrational mode at 64 meV is much stronger than the 42 meV sideband peak. While the peak attributed to the non-localized mode is very strong in our measurements, we cannot identify the peak attributed to the local vibrational SiV center mode in the spectra of group V. 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 Figure 1 the distance between the center wavelength of the sideband peak and the center wavelength of the ZPL is plotted against the ZPL center wavelength. The distribution is fitted with a linear regression. We attribute the variance in the sideband shift to strain:

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For further investigations, we plotted the ZPL of group V with multiple peaks. We found that two Lorentzian fits fit the peak best. Figure 3 shows histograms of the distribution of the continuous wave and the linewidth of the fitted peaks. Keep in mind that two of the peaks sum up to the peak visible as a ZPL and the third peak is the sideband peak which we attribute to a lattice mode. We found that the linewidth of the sideband peak exhibits values up to 20 nm. This broad width is an indicator, that the local vibrational mode might indeed be outpowered by the more intense lattice mode. However, it is not very easy to find spectra where the sideband peak is pronounced and isolated enough to make proper statistics. The original ZPL is split up in two peaks, one with a median center wavelength of 738 nm and a median linewidth of 4.5 nm and the other with a median center wavelength of 742 nm and a median linewidth of 8 nm. It could be that this is an indication for another sideband peak at 742 nm. This assumption could be verified by cryostatic measurements, where the phonon sideband should vanish and only the ZPL survive and split up into the four-level fine structure.

0.0.2 Cryostatic Measurements

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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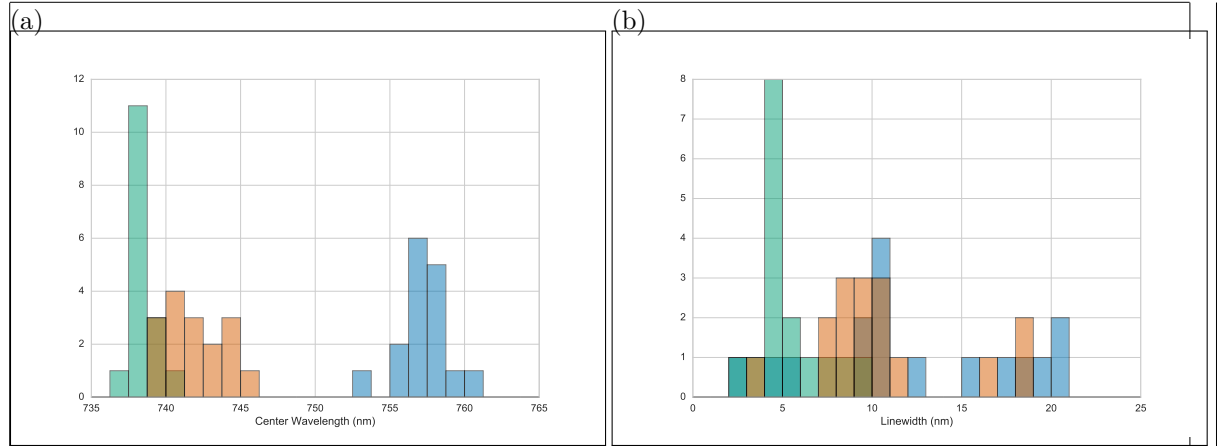


Figure 3

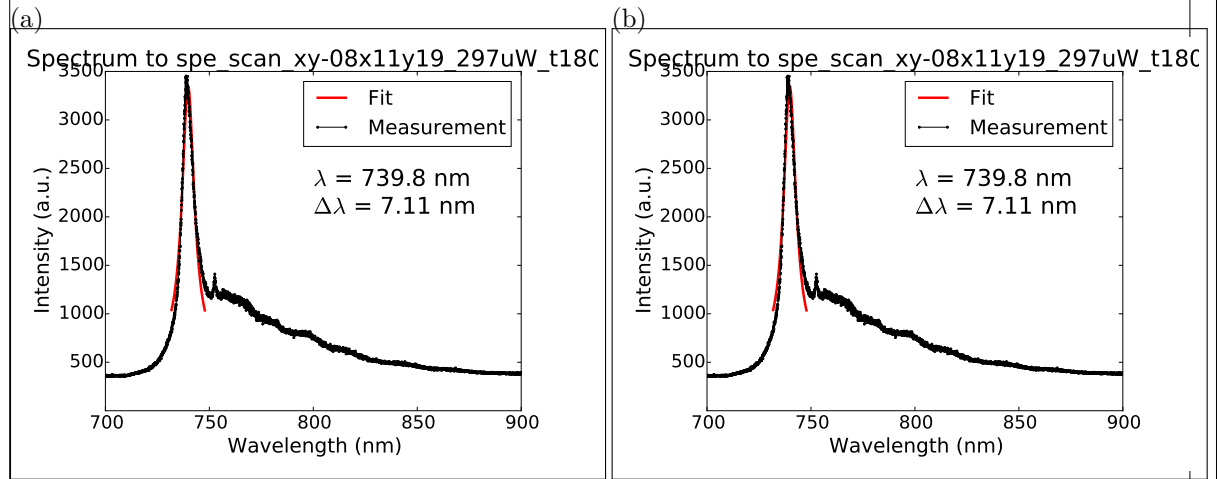


Figure 4

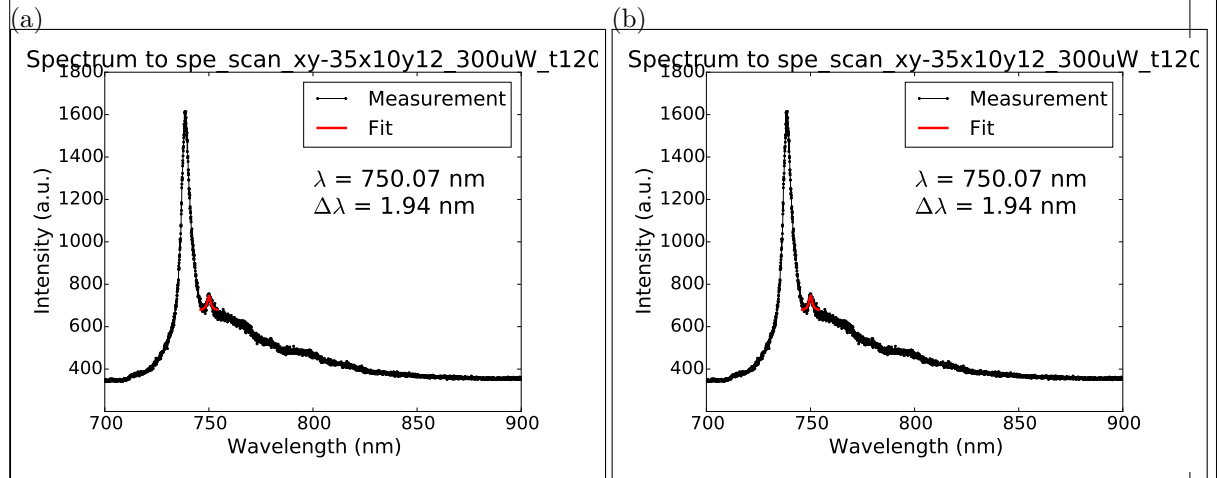


Figure 5