Widely tunable CW Optical Parametric Oscillators:
Mastering the challenges posed in quantum technology research

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ABSTRACT

Widely tunable continuous wave optical parametric oscillators (cw OPOs) are gaining popularity as novel sources of tunable laser light, not least due to the unprecedented wavelength coverage in the visible and the near infrared spectral range. While the potential and the advantages of tunable cw OPOs are becoming increasingly recognized, in particular within the quantum research community, the experimental requirements are often challenging. In this context, we discuss the characteristics of state-of-the-art tunable cw OPO designs and describe several tuning schemes tailored to meet various experimental needs. In an illustrative fashion, we compare several recently published experimental datasets from photoluminescence excitation experiments, which have been carried out on ensembles as well as on individual quantum emitters under different experimental conditions.

Keywords: Tunable laser, optical parametric oscillator, photoluminescence excitation, color center, single photon emitter

1. INTRODUCTION

Quantum technology research is concerned with creation, control, and detection of single quantum objects such as electrons or photons. Considerable public and private investments nowadays are driven by the vision of a 'second quantum revolution', by which applications like quantum cryptography or quantum computing would become commonplace. Thereby, the envisioned device architectures essentially rely on so-called qubits, which might be considered as manipulable two-level quantum systems. In quantum cryptography, for example, such systems are capable to transmit information in the form of single photons that carry one qubit apiece (so-called single-photon emitters), while in quantum computing, the systems are sought to interact in quantum analogies to classical logic gates.

Though there are controversial views on how soon real-world devices are to be expected, there is common agreement on the physical criteria that need to be met by any underlying quantum system: Major challenges include a sufficiently long decoherence time, scalability, controllability, and quantum state measurability. Among a variety of concepts, the implementation of single-photon emitters in the solid state is widely recognized to offer technologically appealing advantages, like the potentially available arsenal of existing (classical) chip fabrication techniques.

Clearly, in the ongoing intensive research for best suited quantum systems, the experimental and theoretical characterization of their internal energy level structure is of fundamental importance. From the experimental perspective, photoluminescence excitation (PLE) spectroscopy, in which the emission intensity of either an ensemble of quantum emitters or a single quantum emitter is measured across a range of excitation (laser) frequencies, has been well established. Yet, the experimental demands are often challenging: The characterization of quantum systems frequently necessitates tuning the laser light frequency throughout a wide spectral range in a quasi-continuous or truly continuous way, while high finesse spectral resonances, as typically observed at cryogenic temperatures, require sufficiently narrow linewidths. Along the same lines, the requirements in terms of absolute frequency accuracy as well as long term frequency stability are typically demanding. Here, we discuss the performance characteristics of state-of-the-art widely tunable laser light sources based on continuous wave optical parametric oscillator (cw OPO) technology, and show several tuning mechanisms based on internal and/or external frequency selection and stabilization. To illustrate the performance under real-world laboratory conditions, we compare selections from experimental datasets that have been recently published by three different research groups studying single-photon emitters of various types[4,5,6].
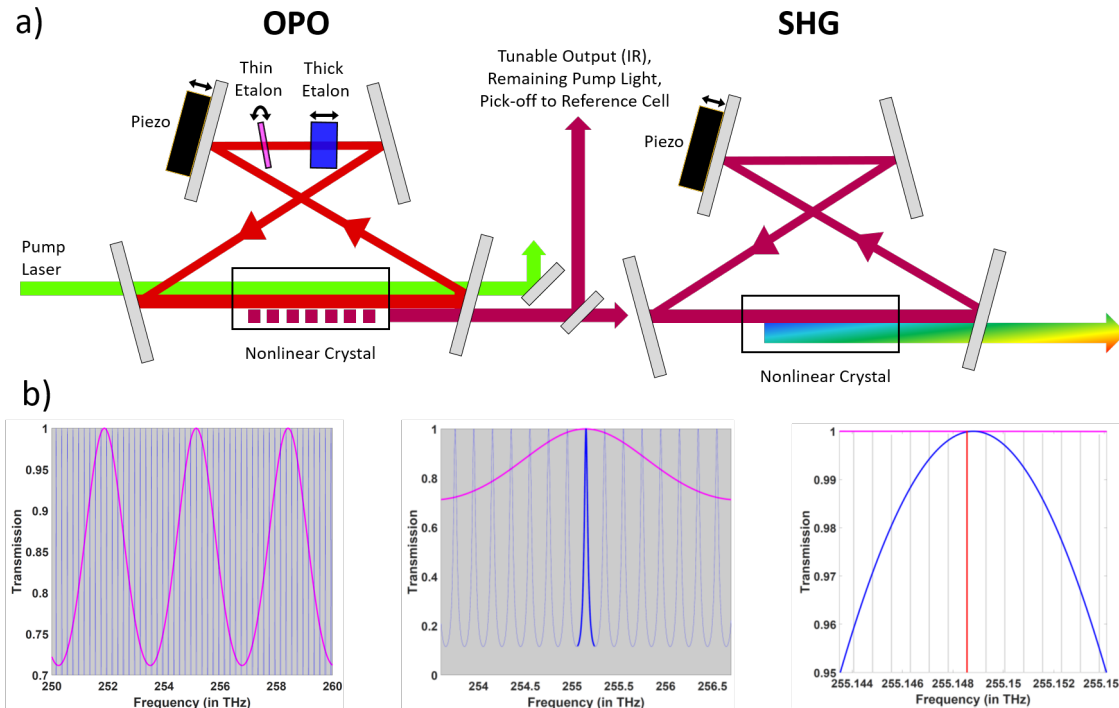
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Figure 1. (a) Schematic beam path inside a cw OPO system. In a first step (OPO), a 532 nm laser pumps a nonlinear crystal to generate signal and idler photons in the near infrared spectral range (900 - 1300 nm). Wavelength selection using a two-stage etalon process and subsequent second harmonic generation (SHG) converts either signal or idler photons into the visible range of the spectrum (450 - 650 nm). The green arrow depicts the pump laser, dark red and light red arrows depict the signal respectively the idler beam (arbitrary assignment). (b) Mode selection within the OPO resonator. From left to right: Transmission spectra of the OPO resonator and the two Etalons presented on different frequency scales. The OPO resonator modes (grey color) undergo a coarse pre-selection by the thin Etalon modes (magenta color), which have a typical linewidth in the range of 10 nm and a free spectral range of the same order of magnitude (i.e. finesse of 1). In combination with the thick Etalon mode (blue color) providing a linewidth in the order of GHz (free spectral range: 100-200 GHz) only a single OPO resonator mode (red color) is supported for resonant oscillation contributing to the laser light output.

2. OPO CHARACTERISTICS AND TUNING PERFORMANCE

The huge potential of OPOs, compared to conventional sources of laser light, derives from their exceptional wavelength versatility, as they are in principle not limited by the wavelength coverage of suitable laser gain media[1]. In principle, the OPO concept has been experimentally demonstrated already more than half a century ago[2], but the subsequent progress in research and development of parametric devices has been stalled by several technical obstacles[1]. This applies, in particular, to OPOs operating in continuous wave mode, so that the generation of widely tunable cw laser light in the visible range had to rely mostly on conventional laser devices for decades. Only relatively recently, there have been exciting advances in cw OPO technology, driven both by the emergence and increasingly sophisticated design of new nonlinear crystals, as well as the increasing availability of suitable highperformance solid-state pump laser sources[1]. These advances have spurred the practical realization of widely tunable cw OPO devices with novel characteristics[3]. Figure 1 shows the optical cavity layout schematics and mode-selection principles in a commercialized cw OPO scheme designed to cover the visible spectral range, and whose performance characteristics shall be discussed in the following.
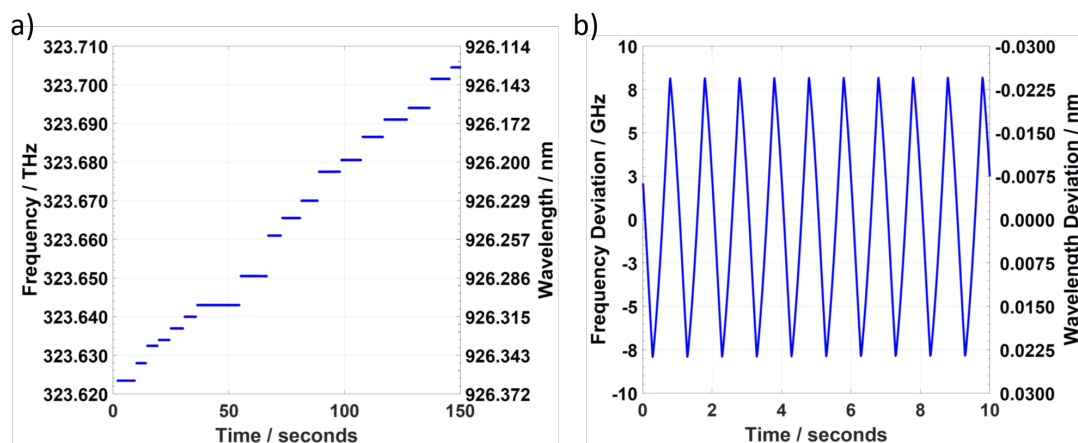
Figure 2. (a) Frequency tuning of the OPO shown in Figure 1 by stepping the thick Etalon. The frequency of the laser light output can be changed in discrete steps down to 2 GHz (in the IR range). (b) Truly continuous (mode-hop free) scan at a central wavelength of 940 nm. The scan range is >10 GHz and can reach up to more than 20 GHz, depending on the central wavelength. After frequency conversion, this results in mode-hop free scans over more than 40 GHz in the visible wavelength range.

Generally, the operation of cw OPOs puts stringent requirements on pump laser sources in terms of preferential single mode operation, noise characteristics, beam quality, and beam pointing stability. Depending on power requirements, we therefore resort to either high performance diode pumped solid state (DPSS) lasers (lower power), or to a high performance fiber laser based solution (higher power) for pumping. In either case, the wavelength of the original pump laser light is centered at 532 nm. Figure 1a illustrates the subsequent beam path and cascaded sequence of nonlinear optical processes in two cavities, referred to as OPO and SHG cavity, respectively. In an intuitive picture, pump laser photons are first split into pairs of photons of lower energy, commonly referred to as signal photons (in the 900 to 1050 nm wavelength range) and idler photons (in the 1080 to 1300 nm wavelength range). This three-wave mixing (of pump, signal, and idler waves) is intimately linked to the second order nonlinearity of the employed nonlinear crystal. In order to achieve sufficient conversion efficiency, the nonlinear medium is operated inside a resonator cavity, to ensure multiple paths that increase the gain at each round trip. The employed scheme is commonly referred to as singly resonant OPO design: For a particular operational wavelength of the entire system, the OPO cavity is operated "on resonance" at either a particular signal wavelength or a particular idler wavelength, which means the effective OPO cavity length is actively stabilized to a multiple integer of that wavelength. While keeping one of the generated waves resonantly circulating inside the OPO cavity, its counterpart can be extracted for wavelength conversion into the visible spectral range by a second nonlinear process. This step is required since the primary OPO process by physical nature generates output at wavelengths longer than those used for pumping. As illustrated in Figure 1a, wavelength conversion is achieved in a second, separate cavity by frequency doubling of the primary OPO cavity output, a process widely known as second harmonic generation (SHG).

The clear key performance characteristic of the cw OPO platform presented here is to provide convenient access to the visible wavelength range of 450 to 650 nm. In addition, for the case the SHG generation module is not employed or switched off, the wavelength range of 900 to 1300 nm can be covered in the near infrared. Except for the so-called degeneracy gap around 532 nm (1064 nm) of 15 nm (30 nm) width, this wavelength coverage is truly continuous and can be fully automated. For illustration, an exemplary quasi-continuous and a truly continuous scan are shown in Figure 2 (see caption of Figure 2 for more detailed description). Depending on the particular choice of pump laser, output powers of up to 800 mW can be achieved. The system delivers high quality cw output with typical linewidths of < 500 kHz corresponding to typical coherence lengths well above 100 m throughout both the visible and the near infrared tuning range. Thereby, a long term frequency stability of < 150 MHz over hours is routinely achieved at typical lab conditions. For applications with highest demands, the performance characteristics can be further improved by operating the system in closed-loop mode, i.e. in conjunction with an external wavelength measurement device (wavemeter). In this operation mode, the achievable long term stability essentially approaches the measurement resolution of the external wavemeter device itself, which can be as low as on the order of a few MHz.
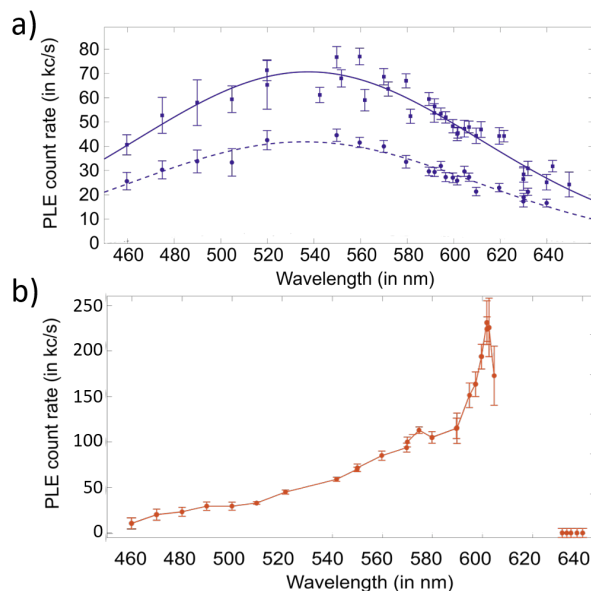
a)

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Figure 3. (a) Photoluminescence excitation spectra of an ensemble of Si-V centers recorded at two excitation powers. Both spectra are recorded at room temperature and for excitation wavelength tuning from 460 nm to 650 nm. The PLE spectra were recorded for 5mW (blue circles) and 10mW (blue squares). Blue dashed and blue solid lines represent Gaussian fits to the data. (b) Photoluminescence excitation spectrum of an ensemble of Ge-V centers recorded at 0.25 mW excitation power. The spectrum is recorded at room temperature for excitation wavelength tuning from 450 nm to 640 nm. Adapted with permission from reference 4.

3. PHOTOLUMINESCENCE EXCITATON STUDIES OF SINGLE-PHOTON EMITTERS

We start with a general remark and note that the different emitters under study here not only do share the potential of single-photon emission (generation of exactly one photon per excitation event), but also can be discussed within similar physical concepts and terminology. A crucial factor governing their physics is the extent of coupling of the emitters electronic transitions to lattice vibrations of the surrounding crystal host. Simply speaking, the strength of this phonon coupling can be inferred by the appearance of phonon sidebands (PSBs) in absorption and/or emission spectra, in addition to the so-called zero-phonon-line (ZPL) of the purely electronic transition. Further, the emitters optically active electronic levels might couple to intermediate dark states (also referred to as shelving states), thereby reducing the efficiency of (resonant) fluorescence emission. Under certain circumstances, and as illustrated below, it is possible to reinstate resonant fluorescence by employing an additional small amount of non-resonant excitation, an effect that can be used to "gate" the fluorescence of the emitter (by switching the non-resonant excitation source on and off).

Among a variety of solid state quantum emitters, the prominent nitrogen-vacancy center (N-V) in diamond arguably is the most extensively studied color center in photonics research. However, the search for systems with potentially even improved properties, like lower susceptibility to external noise, recently has directed the interest of the scientific community towards alternative defects based on group-IV elements. As such, the silicon-vacancy center (Si-V) has drawn attention due to its favorable set optical properties, such as a strong ZPL emission, high spectral stability and narrow inhomogeneous distribution of transition frequencies. Similarly, the germanium-vacancy center (Ge-V) was proposed to offer a very similar structure to that of Si-V and comparably attractive properties. Despite their potential, the photoluminescence excitation spectra of Si-V and Ge-V have been barely studied until recently, not least due to the lack of cost-effective tunable excitation sources that would offer sufficient output power for excitation power dependent measurements.

Figure 3 compares PLE spectra of ensembles of both Si-V and Ge-V color centers recorded at room temperature conditions[4]. The data has been recorded by exploiting the full coarse tuning range of the OPO scheme shown in Figure 1,
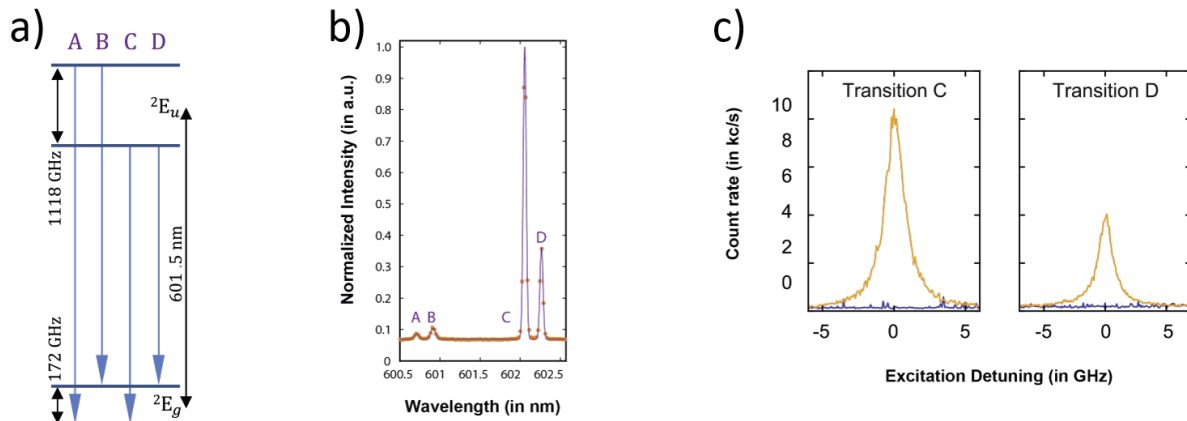
Figure 4. (a) Electronic level structure of the ground and the first excited state of a Ge-V center. (b) Normalized photoluminescence spectrum of a single Ge-V center recorded at a temperature of 5 K, excited at 532 nm with a power of 0.4 mW. The purple line is fit with four Gaussian peaks labeled as A, B, C, and D from high to low energy. (c) Photo-luminescence excitation spectra of transition C (left) and D (right) recorded under tuning resonant excitation. Zero detuning corresponds to 602.2903 nm and 602.4828 nm respectively. Orange lines represent the signal when an additional 532 nm gating laser is switched on, blue lines represent the signal when the gating laser is off. Adapted with permission from reference 5.

with a step-size of approx. 10 nm, straight-forward accessible by automated crystal selection and temperature tuning. Although the spectra are recorded under quite comparable experimental conditions, their qualitative difference is immediately striking. The Si-V ensemble shows a broad very broad PLE signal distributed over the entire measured wavelength range, for both applied excitation powers of 5 mW and 10mW. In turn, the PLE measurement of the Ge-V ensemble, recorded at an excitation power of 0.25 mW, clearly reveals a maximum count rate when resonantly driving the ZPL at approx. 602 nm (cf. Figure 3, bottom). Furthermore, the Ge-V PLE spectrum is in good agreement with the mirror image of the photoluminescence spectrum flipped at the central ZPL emission (not shown here for brevity). While a detailed discussion is beyond the scope here, we point out that theoretical simulations support the interpretation of the Si-V data by a dipole-allowed transition to the second excited electronic state, with one of the involved electronic bands lying deeply inside the diamond valence band (thereby inducing strong broadening by band mixing). Obviously, such effects are not observed for Ge-V, albeit a similar electronic level structure is proposed and classified as ground state ^2E_g, first excited state ^2E_u, and second excited state $^2A_{2u}$.

A further refined electronic level structure of Ge-V is shown in Figure 4a: Because of strong spin-orbit coupling, the ground state (^2E_g) and the first excited state (^2E_u) are split into a pair of energy levels with twofold spin degeneracy at zero magnetic field. In fact, by recording the PLE spectrum of a single Ge-V center at cryogenic temperature conditions, this characteristic four-line fine structure in the ZPL emission can be experimentally revealed, as shown in Figure 4b. Note that the spectral window shown in Figure 4b is a tiny cutout around the ZPL emission at approx. 602 nm (i.e. around the prominent peak in Figure 3, bottom). In Figure 4c, the resonant fluorescence characteristics of the two lower energy features (labeled as C and D, respectively) of the single Ge-V emitter are precisely characterized. For this purpose, the laser excitation wavelengths are tuned within an interval of approx. +/- 5 GHz across the spectral resonances by truly continuous frequency tuning in conjunction with a wavemeter device.

It should be emphasized that for both transitions C and D, the resonant PLE spectra are only detectable at additional non-resonant excitation, i.e. when the Ge-V center is additionally excited with a gating laser (at 532 nm). This gating effect can be immediately perceived from Figure 4c when comparing the PLE spectra with the gating laser on or off (orange lines and blue lines, respectively). We note that the non-resonant excitation alone has been demonstrated to be too weak to induce any detectable fluorescence from the emitter. Rather, the main role played by the light is indeed that of a switch controlling the onset and decrease of resonant fluorescence. The dynamics of the so-called shelving of (resonant) fluorescence has been recently studied in detail also in time domain experiments, e.g. by modulation of the intensity of
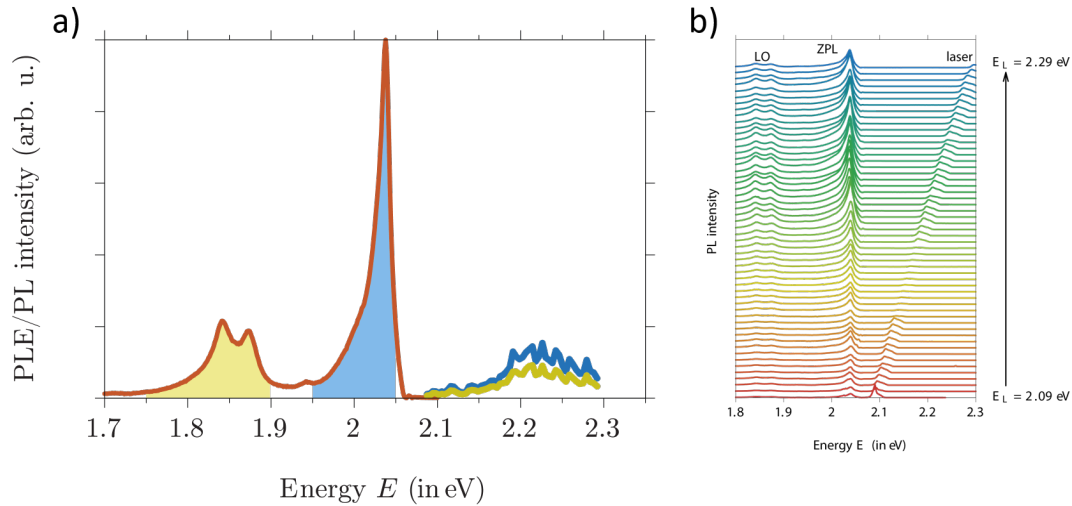
Figure 5. (a) Red line: Photoluminescence spectrum of a single defect center in hBN, blue shaded area: integration interval over ZPL for PLE spectrum, yellow shaded area: integration interval over LO sideband for PLE spectrum, blue line and yellow line: corresponding PLE spectra. (b) Photoluminescence spectra of the single defect center in hBN recorded under laser excitation energy tuning from 2.09 eV (593.30 nm) to 2.29 eV (541.48 nm). The shown data is corrected for background and Raman-lines. Adapted with permission from reference 6.

either the resonant or the non-resonant excitation beam, while keeping its counterpart in constant power mode. The experimental findings are discussed in detail in reference 5, where the authors also quantitatively explain the observed dynamics by the presence of a dark state.

Before closing, we turn to a very recent study[6] of defect centers in hexagonal boron nitride (hBN), which can be classified as single-photon sources in a layered van der Waals material. While potentially offering the ultimate limit of miniaturization (of a two-dimensional structure), their experimental and theoretical characterization is challenging: On the one hand, the emission from individual defect centers in hBN shows a strong variation, and appears within a wide range of transition energies ranging over the entire visible spectrum. On the other hand, they show strong coupling to phonons, with the photoluminescence exhibiting significant side-band emission, well separated from an asymmetrically broadened ZPL. This is exemplified by the photoluminescence spectrum of a single hBN defect recorded at room temperature, shown in Figure 5 (a).

As has been shown recently[6], excellent agreement with the experimental emission spectra can be achieved by employing a theoretical model that accounts for coupling of the defects to bulk longitudinal optical (LO) phonons and longitudinal acoustic phonons, two phonon contributions, as well as a local mode. For the sake of brevity, here, we draw the attention to PLE measurements that demonstrate efficient excitation of the defect centers by LO phonon assisted absorption. In a simple intuitive picture, noting that the prominent LO phonon side-band in the emission spectrum appears approx. between 150 meV and 200 meV below the ZPL line (cf. Figure 5a), one would expect the presence of a corresponding spectral feature also in the PLE spectrum of the emitter - or, in other words, signatures of a symmetry of the emission and absorption spectral lineshape around the ZPL. Indeed, by employing a suitable crystal temperature tuning protocol of the cw OPO that is used for the emitter excitation, and measuring the photoluminescence intensity for over 50 different excitation energies ranging from 2.09 eV (593.30 nm) to 2.29 eV (541.48 nm), such a feature is unambiguously recovered. Figure 5b shows the corresponding dataset which has been corrected for background and Raman contributions. For each excitation energy, the PLE intensity is extracted by integrating the photoluminescence spectrum either over the ZPL or over the LO phonon sideband (cf. blue shaded area and yellow shaded area in Figure 5a, respectively). As shown in Figure 5a, both PLE datasets exhibit a pronounced maximum between 2.2 eV and 2.3 eV, an energy range that nicely agrees with the energy range expected by mirroring the LO phonon sideband in emission around the ZPL.

4. OUTLOOK

As illustrated by a few examples in this contribution, the coming of age of cw OPO technology comes in tandem with remarkable efforts in quantum technology. From the laser technology point-of-view, we note that the OPO concept presented here is quite general, i.e. it can be used to cover different tuning ranges. This will allow for continuous adaption to new experimental requirements respectively novel types of single-photon emitters and systems alike. Overall, we expect cw OPO technology to mature into a recognized choice among laser light sources that advance the rapidly evolving field of quantum research.

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