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Optimizing Detection Schemes for Broadband Pump-Probe Microscopy

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ABSTRACT

Although similar to more commonly implemented single wavelength approaches, broadband pump-probe or transient absorption microscopy presents unique experimental challenges due to the simultaneous requirements of a broadband probe pulse and a small sample volume. Here we provide an in-depth analysis of broadband detection schemes and their common noise sources to provide strategies for balancing the conflicting needs of high sensitivity and low probe fluence. We show that broadband pump-probe microscopy is atypically sensitive to laser shot noise and therefore, low pump on/off modulation frequencies, on the order of 100s of Hz to a few kHz, are essential to measure small ($\sim 10^{-3} - 10^{-4}$) amplitude transient spectra while remaining in the perturbative limit.

1 | Introduction

Broadband pump-probe microscopy (BPPM) has recently emerged as an essential tool for probing structure–function relationships in nano- and mesoscale materials. The sub-micron spatial resolution and ultrafast temporal resolution of the technique, together with the ability to simultaneously probe kinetics over 100s of nanometers, provide important insight into the electronic structure of emerging bulk and nanoscale materials, particularly in the presence of structural or chemical heterogeneity (Beane et al. 2019; Davydova et al. 2016; Ginsberg and Tisdale 2020; Grumstrup et al. 2015; Jones et al. 2020; Zhu et al. 2019; Zhu and Cheng 2020). While powerful, obtaining sufficient signal to noise (S/N) levels while remaining in the low fluence limit is uniquely challenging for BPPM.

Numerous strategies for maximizing S/N levels in transient spectroscopy have been reported (Adhikari et al. 2024; Dobryakov et al. 2010; Erkang et al. 2020; Kim et al. 2024; Lang 2018; Wilson et al. 2021). Generally, pump-probe (or transient absorption) spectroscopies require resolution of small differential signals (on the order of 10^{-3} – 10^{-4}), typically obtained by alternately measuring sample transmittance (or reflectance) of a probe pulse in the presence and absence of a temporally separated pump pulse. While raising the pump intensity will increase signal size, many-body interactions and sample damage thresholds often limit this approach, and so efforts to reduce noise must be pursued. To measure small signal amplitudes in the presence of typical noise sources, a common strategy is to maximize the spectral acquisition rate and chop the pump at exactly half the acquisition frequency. At sufficiently high acquisition frequencies, shot-noise-limited

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Summary

- Small spot size and broadband spectral coverage result in nonnegligible photon shot noise in broadband pump-probe microscopy.
- As a result, optimization of detector efficiency and low-frequency acquisition is essential to measuring small transient signals.

performance can be achieved, and the minimum detectable signal size, $(I_{pp})_{\min}$, is determined solely by the number of detected photons, N_{ph} , according to Equation (1):

$$(I_{pp})_{\min} \approx (S/N) \sqrt{2/N_{ph}} \quad (1)$$

Here $I_{pp} = (I_{on} - I_{off})/I_{off}$ is the dimensionless pump-probe signal calculated from “pump on” (I_{on}) and “pump off” (I_{off}) probe intensities and (S/N) parameterizes the desired S/N level. Details of Equation (1) can be found in the [Supporting Information](#). For a typical signal magnitude $I_{pp} = 1 \times 10^{-3}$ and desired $S/N = 10$, the minimum number of detected photons, $N_{ph} \geq 2 \times 10^8$. Although $N_{ph} \gtrsim 10^8$ per measurement can be routinely achieved in ensemble (large spot size) transient absorption measurements, broadband transient microscopy necessarily requires a probe with a small (submicron) spot size and broad bandwidth (often 100s of nm). These technical requirements impose effective upper limits on N_{ph} because of the sample damage threshold and/or the requirement that the probe be “weak” relative to the pump. A weak probe is essential to minimize further photoexcitation of the sample, which can mask or distort the pump-induced dynamics. Thus, in contrast to ensemble pump-probe measurements, probe shot noise is often the limiting noise source in BPPM. Note that in general, pump pulse characteristics also contribute to the differential measurement S/N . As noted above, pump intensity is often constrained by measurement requirements, and we have found that pump noise generally has limited impact on the overall measurement noise. We therefore treat the pump intensity parametrically and only consider probe noise below.

To provide a strategy for mitigating these limitations in the design and optimization of a BPPM, we perform a comprehensive noise assessment while measuring transient spectra of a prototypical porphyrin-derived organic semiconductor. We compare spectra collected using two classes of detectors—a high-speed CMOS line camera optimized for high frequency acquisition and a “scientific” CMOS area scan camera optimized for low noise, fluorescence imaging measurements. For both detectors, we show that a judicious choice of experimental parameters can provide near- or shot-noise-limited detection. Surprisingly, we find that in many cases, relatively low acquisition frequencies of ~ 100 s Hz – kHz are best for achieving optimal measurement sensitivity in BPPM. Finally, we outline an approach for practitioners to characterize their own systems so that they may determine operational parameters (integration time, modulation rate, camera type) for optimal data collection.

2 | Materials and Methods

2.1 | Materials

Thin film samples were drop cast from tetrahydrofuran solutions of a novel Zn porphyrin (ZPn) derivative ([Supporting Information](#)) onto glass microscope slides. The solvent was allowed to evaporate under ambient conditions, producing coffee-ring type film morphologies ([Supporting Information](#)). Transient spectra were collected from the edges of these regions to ensure finite probe transmission.

2.2 | Methods

Broadband pump-probe spectra were collected on a home-built microscope that has been described in detail elsewhere (Piland and Grumstrup 2019). Briefly, the pump and probe pulses (~ 300 fs) are derived from the attenuated output of a Coherent Monaco operating at a 1 MHz repetition rate. The pump pulse train is passed through an acousto-optic modulator, frequency doubled in a β -barium borate (BBO) crystal, passed onto a mechanical delay stage, and then coupled into a pair of galvanometer-controlled mirrors and a 4-f lens relay before the objective. The pump energy at the sample is 183 fJ/pulse with a spot size diameter of $\sim 0.5 \mu\text{m}$. The probe is a white light continuum generated by focusing $\sim 1.1 \mu\text{J}$ of the fundamental into a 3 mm thick yttrium aluminum garnet (YAG) crystal. Optimization of the focus and power of the 1030 fundamental beam in the YAG crystal produces a white light continuum with intensity fluctuations of $\sim 0.75\%$ MSD (Calendron et al. 2015; Majus et al. 2011). The probe is coupled collinearly with the pump before the objective with a broadband 50:50 plate beam splitter. Both pump and probe are focused onto the sample with an achromatic 100 $\times$, 0.9 NA objective. The transmitted beams are collected with an NA-matched objective, the remaining pump filtered with a dielectric longpass filter, and the probe is focused into a homebuilt spectrograph built from commercial camera lenses and a transmission volume phase grating. A schematic of the collection optics and spectrograph is provided in the [Supporting Information](#).

3 | Results

To understand the constraints of a BPPM measurement, we first consider how typical operational conditions and noise sources translate to instrument sensitivity. As noted previously, the sensitivity for a finite duration measurement is limited by shot noise via Equation (1). Shot noise derives from the stochastic nature of photons as quantum particles, and although photon number is characterized by a Poisson distribution, in a typical pump-probe measurement the photon number may be approximated as a normal distribution with mean N_{ph} and standard deviation (shot noise) $\sqrt{N_{ph}}$ (Audier et al. 2020). In general, the S/N imposed by shot noise therefore scales as $\sqrt{N_{ph}}$. The primary challenge with BPPM is that in contrast to conventional transient spectroscopies, there are practical limits placed on N_{ph} due to both the sample damage threshold and the requirement that a probe remain “weak” relative to the narrowband pump. As a result,

it is not uncommon for shot noise to be the primary barrier to resolving even routine transient signals, which would be trivial in conventional (nonspatially resolved) transient spectroscopic measurements.

To illustrate this challenge, consider the scenario described above in which a transient signal of $I_{pp} = 1 \times 10^{-3}$ must be resolved with $S/N = 10$. A representative set of operational parameters for a conventional ensemble transient absorption instrument is a (per pulse) probe fluence at the sample of $\sim 40 \mu\text{J}/\text{cm}^2$, a spot size diameter of $\sim 100 \mu\text{m}$, a postsample photon detection efficiency of 10% (accounting for sample absorption as well as losses from collection optics, spectrograph, and detector quantum yield), and dispersion of the broadband probe onto 1000 detector pixels. The average (per pixel) detection rate under these conditions is $\sim 1.3 \times 10^6$ photons/pulse at a wavelength of 640 nm. For this scenario, the minimum N_{ph} needed to overcome shot noise at the specified S/N is reached after integrating only 150 laser pulses for both “pump on” and “pump off” spectra. In contrast, for a typical broadband pump probe microscope, the spot diameter is typically $100\times$ smaller, which implies the per pixel detection rate is 10^4 lower with a corresponding $100\times$ larger shot noise level for the same number of integrated laser pulses/experiment time.

Figure 1 illustrates the substantial role shot noise plays in determining S/N in BPPM, where we compare transient spectra collected from the porphyrinic ZPn thin film for a constant probe fluence of $45 \mu\text{J}/\text{cm}^2$. The broadband probe collected in the absence of the sample is shown in panel A. Panel B of Figure 1 compares transient spectra collected from the drop cast ZPn thin film at 2 and 32 kHz for constant probe fluence. For both spectra, 200 “pump on” and 200 “pump off” spectra were averaged to calculate I_{pp} . The red spectrum, collected at 2 kHz, corresponds to $N_{ph} = 8 \times 10^6$, whereas the teal spectrum, collected at 32 kHz (with proportionally shorter integration time), corresponds to $N_{ph} = 5 \times 10^5$. Both spectra are characterized by a distinct positive peak centered near 660 nm arising from the photoinduced bleaching of the Q-band ground state absorption, superimposed with a broad, low amplitude photoinduced absorption (Gouterman 1961). A quantitative comparison of S/N levels can be calculated by dividing the maximum of the signal amplitude (at 660 nm) by the rms noise level, determined from a unitary Fourier transform of the transient spectrum within ± 10 nm of the peak. Panel D summarizes the experimental (blue squares) S/N values for transient spectra collected between 1 kHz ($N_{ph} = 16 \times 10^6$) and 32 kHz ($N_{ph} = 5 \times 10^5$). Also shown in panel D is the modeled shot-noise-limited S/N (solid blue line). The model closely follows the data but predicts an S/N that is systematically higher than that observed experimentally. This difference stems from two primary causes. First, as discussed in further detail below, the high-speed line camera used to collect data has a noise floor that is 1–3 dB higher than that predicted by shot noise alone. The second is a slight decrease in S/N due to read noise.

Detector read noise is a product of the conversion and amplification of photoelectrons to a digital signal, which for CMOS sensors, is generally limited by thermal and random telegraph noise sources (Chang and Sansen 1991; James et al. 2006; Martin-Gonthier and

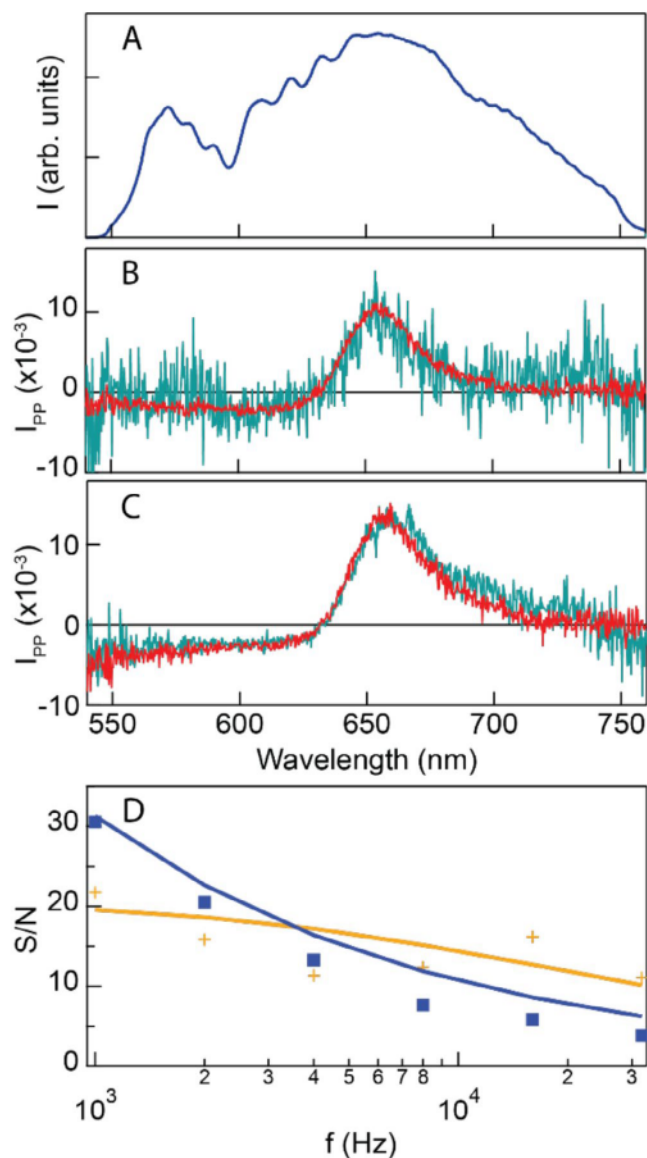


FIGURE 1 | (A) Detected probe spectrum collected with no sample. (B) I_{pp} spectra collected at 2 kHz (red) and 32 kHz (teal). Each spectrum is calculated from 400 acquired spectra, so N_{ph} is inversely proportional to acquisition frequency. (C) A comparison of I_{pp} spectra, again collected at 2 kHz (red) and 32 kHz (teal), but the number of acquisitions is variable so that N_{ph} is held constant for both I_{pp} spectra. (D) S/N of I_{pp} spectra as a function of acquisition frequency. The blue squares show spectral S/N for datasets like those in panel B where N_{ph} decreases with increasing acquisition frequency. The solid line is the modeled shot-noise-limited S/N . Orange crosses show S/N for spectra like in panel C with constant N_{ph} , highlighting the effects of read noise. The solid orange line assumes read noise is added in quadrature.

Magnan 2009). To assess the importance of read noise in BPPM, we collected transient spectra of the ZPn thin film at increasing acquisition rates while keeping the total number of detected photons, N_{ph} , constant. Representative spectra collected at 2 kHz ($\sim 500 \mu\text{s}$ integration time, red) and 32 kHz ($\sim 31 \mu\text{s}$ integration time, teal) are shown in Figure 1C. Although N_{ph} is the same for both spectra, the 2 kHz spectrum is qualitatively less noisy than that collected at 32 kHz. The orange crosses in Figure 1D summarize the

calculated S/N as a function of acquisition frequency, and the solid orange line shows the modeled dependence, which assumes read noise is added in quadrature to shot noise. The orange crosses are again calculated by dividing the maximum of the signal amplitude by the rms noise level within 10 nm of the peak at 660 nm. For the experimental conditions of a signal to read noise ratio of $\sim 550:1$ at 1 kHz, the overall S/N drops by a factor of 2 over the full frequency range, reflecting the (read noise) cost of spectral acquisition at higher frequencies. Note also that read noise becomes increasingly relevant for low pixel count values.

If laser shot noise and read noise were the only factors to consider, integration of many laser pulses (i.e., long integration times) would be sufficient to recover arbitrarily small signals in BPPM. Unfortunately, other sources of noise place limits on integration times. Figure 2 shows the power spectrum of the white light continuum probe (Figure 1A) at $\lambda = 640$ nm. The two spectra shown in Figure 2 are collected with two different detectors: a “scientific” CMOS area-scan camera (sCMOS, Teledyne Prime BSI Express) optimized for low-read noise and high sensitivity detection (green), and a high-speed line camera (HSLC, Teledyne Octopius) optimized for high acquisition rates (blue). Specifications for both cameras can be found in the Supporting Information along with a schematic of the spectrograph used for testing. Plotted with the data are the calculated shot noise floors (horizontal lines at -50.4 dB and -54.6 dB) which are slightly different for the two data sets because of small differences in photon flux to each camera.

For frequencies $f \gtrsim 1$ kHz for the HSLC and $f \gtrsim 300$ Hz for the sCMOS, the power spectra show asymptotic noise floors that reflect the sum of all uncorrelated or “white noise” processes with a flat power spectrum (Boie and Cox 1992). For the sCMOS camera, the noise floor quantitatively matches the calculated shot noise limit. For the HSLC, the average noise floor is 2–3 dB above the calculated shot noise limit and is punctuated by a

series of distinct peaks that rise 20–30 dB above the noise floor. We have found these peaks appear at well-defined frequencies, suggesting they are characteristic of the timing/ADC electronics intrinsic to the HSLC (Piland and Grumstrup 2019).

At low frequency, both power spectra are dominated by “ $1/f$ ” or “pink” noise, which is characteristic of time-dependent nonequilibrium processes of the laser source and the detection electronics (Keshner 1982). Fits to the data show that although the slope in both spectra can be well-modeled by a $1/f^2$ dependence, the sCMOS camera overall has ~ 10 dB lower noise levels at frequencies below the asymptotic limit. As a result, the sCMOS camera performs better at lower acquisition frequencies than the HSLC.

4 | Discussion

Analysis of the power spectra presented in Figure 2 and the results from Figure 1 provide a strategy for optimizing data collection parameters for BPPM. As noted previously, the small spot size and wide bandwidth intrinsic to BPPM ensure that shot noise will generally be of greater importance to BPPM than in conventional ensemble pump probe techniques. For BPPM, the primary objective must be to accumulate enough photons to achieve the desired S/N. Because the accumulation of spectra must be balanced with the fact that differential signals are calculated from sequentially collected “pump on” and “pump off” spectra, the acquisition frequency lower limit is set by the intersection of the $1/f$ and shot noise models for each detector. Once beyond this lower limit, the impacts of read noise can be mitigated by choosing an acquisition frequency that nearly saturates the detector, ensuring the effects of read noise are negligible.

For the laser and detector characteristics presented in Figures 1 and 2, the optimal acquisition frequencies for the HSLC are between 1 and 3 kHz. In this regime, the maximum number of photons are integrated per acquisition cycle (ensuring the contributions from read noise are limited), the probe fluence at the sample can be reduced to its minimum practical level, and characteristic $1/f$ noise from the laser source and detection electronics no longer dominates. Note that higher acquisition frequencies offer no benefit in terms of noise reduction. In fact, acquisition at higher frequencies is likely to reduce S/N due to the contributions from read noise and the narrow peaks characteristic of the HSLC electronics (Figure 2). For the sCMOS, although its maximum acquisition rate is significantly lower than the HSLC, it has overall better noise characteristics. As a result, the shot noise limit can be reached for any acquisition rate $\gtrsim 300$ Hz. Note that noise characteristics of the continuum generation process will vary substantially with experimental parameters, so a power spectrum should be carefully measured under typical operating conditions to determine the minimum acquisition frequency. Ideally the power spectrum is measured with a broadband detector to provide spectrally-resolved noise information, however, we have found detection using a fast photodiode and oscilloscope together with several interference filters provides sufficient data to select an optimal acquisition frequency.

The above analysis shows that optimal acquisition rates for BPPM generally will lie in the 100–1000s of Hz region. These

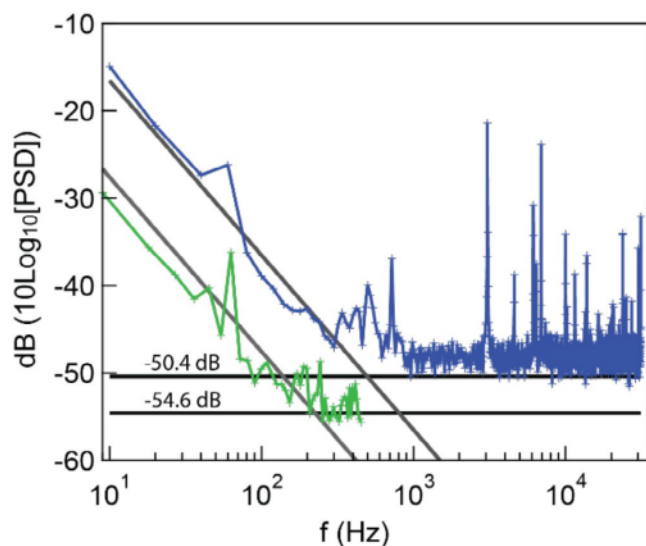


FIGURE 2 | Power spectra of broadband probe collected over 1 total second by the HSLC at 62.5 kHz (blue) and the sCMOS detector at 901.4 Hz (green). Calculated shot noise floors for the two measurements are shown as horizontal solid lines (-50.4 and -54.6 dB, respectively). Low frequency noise in both spectra is well fit with a $1/f^2$ dependence (solid lines with negative slopes).

relatively low acquisition rates suggest that HSLCs offer no practical advantage over slower sCMOS or CCD area scan cameras for BPPM. Indeed, given the importance of maximizing probe photon detection efficiency, area scan detectors like the sCMOS detector used here offer several distinct advantages over line cameras for transient microscopies. First, the larger active area of an area-scan camera ensures that beam distortion (due to diffraction at the sample plane) does not reduce N_{ph} (Piland and Grumstrup 2019). In contrast, if the beam extends beyond the single pixel height of the HSLC, the overall QY of the spectrometer is reduced, effectively increasing the shot noise floor. Second, area scan cameras can be readily obtained with a backside illuminated architecture, raising QY of the overall postsample detection system. Third, the ability to bin multiple pixels in an area-scan camera provides a straightforward route to increasing the effective full well capacity and thus the (shot-noise-limited) S/N. Finally, area scan detectors allow for probe beam referencing, which can allow for greater reductions in low frequency noise (Feng et al. 2019).

It is worth noting that slower acquisition rates necessarily mean longer experimental time. Particularly for imaging measurements that require point-scanning hundreds or thousands of spatial pixels, the longer integration times necessary to overcome shot noise in BPPM may become prohibitively long. Long scan times are especially important given the sensitivity of pump-probe microscopy to objective focus, laser power, and beam pointing stability. For such scenarios, integration of a spectral region of interest provides an effective means to linearly increase photon count rates, albeit at the expense of added read noise and decreased spectral resolution.

In conclusion, we have analyzed noise sources and developed a set of optimization strategies for obtaining maximal sensitivity in broadband pump probe microscopy instruments. For a representative set of experimental parameters, we showed that shot noise is often the limiting noise source in BPPM. Thus, spectral acquisition rates, postsample detector efficiency, and camera choice must be carefully optimized for each system to ensure sufficient sensitivity is achieved. For many samples, low acquisition rates—between 100 and 1000s of Hz—are optimal for BPPM data collection.

Author Contributions

Kendall Benton: investigation, validation, formal analysis, data curation, visualization, methodology, writing – original draft, writing – review and editing. **Emma Orcutt:** investigation, methodology, writing – review and editing. **Skyler Hollinbeck:** investigation, methodology, writing – review and editing. **Erik M. Grumstrup:** conceptualization, methodology, writing – review and editing, project administration, supervision, funding acquisition, resources.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Figure S1:** (A) structure of ZPn. (B) ZPn thin film used as a sample in this work. Transient spectra were collected from the thinner edge regions of the coffee-ring region. **Table S1:** Camera specifications as determined from manufacturer specifications. **Figure S2:** Schematic of BPPM detection scheme. CL, camera lens; CO, collecting objective; DG, diffraction grating; F, 540 nm Long Pass Filter; I, Iris; L, lens; M1, fixed mirror; M2, removable mirror to redirect light to CMOS HSLC; MO, microscope objective; S, sample.