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General theoretical treatment of spectral modulation light-labeling spectroscopy

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Abstract

We theoretically derive the analytic relationship between experimental parameters and the measured incident (or illumination) optical power spectrum for a new form of spectroscopy, entitled light labeling spectroscopy. The light labeling signals are shown to arise from the interference between fields diffracted from a grating with time varying ruling density. A Gaussian model is used to illustrate the bounds of the method for recovering power spectra without artificial spectral apodization. Finally, several example systems are tabulated to give numerical insight into the possible system performances across a range of wavelength regions.

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1. INTRODUCTION

Light labeling spectroscopy is a multiplexed form of wavelength swept illumination, benefiting from Fellgett's advantage of increased signal to noise ratios over sequential spectral acquisition. Fellgett's advantage states that a multiplexed spectral measurement benefits from an increase in the integration time of each spectral component over a sequential spectral measurement by M , where M is the number of measured spectral points [1]. Directly encoding wavelength dependent intensity modulations onto the power spectrum at modulation frequencies far below the optical carrier "labels" the wavelengths within the optical power spectrum with unique modulation frequencies determined by the system. Much like Fourier transform spectroscopy, light labeled signals are captured in the time domain, and a Fourier transform recovers the power spectrum on an optical frequency axis scaled to laboratory and system parameters.

With light labeling however, the optical frequency axis is not linked to the scanning delay of an interferometer arm (generally the physical motion of a carriage and retro-reflector), but

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the spectral dispersion introduced by a grating with a time varying ruling density. This leads to a critical difference: light labeling is sensitive to the zero-centered angular frequency, Ω , rather than the angular frequency, ω of the power spectrum, where $\Omega = \omega - \omega_0$ and ω_0 is the central angular frequency of the system. The dependence on Ω allows for the implementation of light labeling systems from the visible through the mid-infrared wavelength regions with very similar optical architectures.

Recently, a light labeling system was shown to permit wide-field, epi-detected (and therefore background free), absorption spectra imaging [2]. The unique intensity modulation frequency label for each wavelength within the excitation power spectrum was transferred to absorbing molecules and retained in the emitted fluorescent intensity. The absorbed spectrum was thus retrieved from the temporal dynamics of the emission intensity, which was captured in the shot-noise limit for small fluorescent signals.

There are a diversity of additional applications directly enabled or enhanced by light labeling: high speed hyperspectral imaging; spectroscopy in the mid-infrared, THz, and mm-wave; Fourier transform spectroscopy; high speed multidimensional spectroscopy, and pump-probe spectroscopy. As an example, consider the combination of light labeling spectroscopy with a high speed single-element detector imaging system. Leveraging the large bandwidth of single-element detectors, such imaging systems can produce two-dimensional frame rates beyond 2 kHz [3] by temporally encoding spatial information [3–6]. The hybrid, hyperspectral imaging system could increase the acquisition rate of three-dimensional so-called hypercubes beyond the ceiling of state-of-the-art high speed cameras.

To generate the light labeling modulations in the incident (or illumination) power spectrum, we use a pulse shaper composed of a 4-*F* Martinez [7] with a time varying transmission mask located in the joint Fourier plane, an example of which is shown in Fig. 1. Interestingly, the premise for this method of spectroscopy was first considered around 1950 but abandoned in favor of interferometers such as the Michelson [8]. The current field of single-element detector imaging technologies led us to re-invent the light labeling method to decouple parallel spectral acquisition from an array detector, enabling high acquisition bandwidths. Separately, researchers have employed optical architectures similar to our light labeling system to rapidly modulate nonlinear signals [9] and perform high speed Fourier transform spectroscopy [10].

The object of this article is to model the propagation dynamics of the electric field through the dispersive and imaging elements, where exaggerated beam trajectories and spatial distributions are shown in Fig. 1(b)-(e). The end result of the analysis is an analytic representation of the expected light labeling signals in both the time (measured) and frequency (recovered) domains. We also use a Gaussian model of spatial and spectral distributions to define contours of operation delineating systems with and without significant spectral apodization. Lastly, we provide several example light labeling system to illustrate possible system performances across a range of wavelength regions.

While the analysis presented here assumes a fully coherent pulse, the formalism and many of the analytical results can be extended to incoherent light, provided it is spatially coherent.

Fiber coupling the incoherent optical source, such as a super-luminescent diode, satisfies this caveat. The extension of the light labeling spectroscopic method to incoherent sources will bridge the gap between ultrafast pulse shaping spectroscopy (e.g. Ref. [9, 10]) and a more general type of pulse shaping spectroscopy.

2. PROPAGATION THROUGH THE MARTINEZ

To model the propagation dynamics of the electric field through the light labeling Martinez, we make use of a consequence of the paraxial approximation: the electric field at the Fourier plane of an imaging element is a spatial Fourier transform of the input field parameters at the conjugate plane (so called 2- F propagation) [11]. Inspection of Fig. 1(a) shows we can propagate entirely through the system using a series of 2- F propagation transforms, provided we apply the time variant spatial modulation at plane (x_2, y_2) . For brevity, we will refer to the focusing elements as lenses, but they could as well be concave mirrors, off-axis parabolic mirrors, or some other focal element.

A. A spectrally dispersed field and 2- F propagation

We assume the incident electric field is a coherent, broad bandwidth pulse of light given by

$$E_{\text{in}}(x, y; \Omega) = E_0 a_{\Omega}(\Omega) a_s(x, y). \quad (1)$$

Here, E_0 is the peak field amplitude, a_{Ω} is the complex spectral field envelope, and a_s is the spatial field distribution. The field impinges on the first dispersive element of the Martinez with an incidence angle of γ , which transforms the field by the application of a complex term $t_d(\Omega)$:

$$E_d(x, y; \Omega) = E_0 a_{\Omega}(\Omega) a_d(x, y) e^{i2\pi \frac{\sin(\gamma)}{\lambda} x} t_d(\Omega), \quad (2)$$

where λ is the wavelength of light and c is the speed of light. The geometric projection of the field onto the dispersive element changes the spatial profile of the incident field, but where the dispersive element is either a diffraction grating aligned near the Littrow condition or a prism in the minimum deviation configuration $a_d \approx a_s$, otherwise $a_d(x, y) \approx a_s(x \cos(|\gamma| - |\theta_d|), y)$ where θ_d is the diffraction angle of the central wavelength.

The complex term from the dispersive element has the effect $e^{i2\pi f_{\text{irr}}(\Omega)x} t_d(\Omega) = \eta_d(\Omega) e^{i2\pi \mu \Omega x}$, where $\eta_d(\Omega)$ accounts for the diffraction efficiency of the dispersive element and the complex phase term accounts for the angular dispersion. As the diffraction efficiency can be bundled into the spectral envelope term, we will simply set $\eta_d = 1$ as it does not add to the propagation dynamics. We derive in the Appendix the critical component μ for the two most common dispersive elements (diffraction gratings and prisms) where the Martinez and diffraction angles are oriented such that the diffracted central optical frequency, ω_0 , lies on the optical axis of the lenses.

Under this construction, the angular dispersion only acts in the x - z plane. The field leaving the dispersive element is therefore

$$E_d(x_0, y_0; \Omega) = E_0 a_\Omega(\Omega) a_d(x_0, y_0) e^{i2\pi\mu\Omega x_0}, \quad (3)$$

where the subscripts label the particular (x, y) plane. With this expression, we can readily compute the field distribution in the Fourier plane of the Martinez, plane (x_2, y_2) , after the first 2- F imaging system through a scaled two-dimensional (2D) spatial Fourier transform of the field in plane (x_0, y_0) [11, 12]:

$$E'_2(x_2, y_2; \Omega) = E_0 \frac{e^{i4\pi f / \lambda}}{i\lambda f} a_\Omega(\Omega) A_d\left(\frac{x_2 - \lambda f \mu \Omega}{\lambda f}, \frac{y_2}{\lambda f}\right), \quad (4)$$

where f is the focal length of the lens in the Martinez, and

$A_d(f_x, f_y) = \iint a_d(x, y) e^{-i2\pi(f_x x + f_y y)} dx dy$ is the 2D spatial Fourier transform of a_d . The key feature of the Martinez is the approximate linear mapping of Ω onto the transverse x_2 spatial dimension.

B. Modulation by the mask

We will use a mask composed of an amplitude grating with a time varying groove density and a 50% duty cycle:

$$m(x_2, y_2; t) = w(t) \left(\frac{1}{2} + \frac{1}{2} \cos(2\pi \Delta k v_r t (x_2 + x_c) + \Delta k y_2) \right) \quad (5)$$

which is the mask for a spinning-disk, frequency modulated reticle [13], where v_r is the rotation frequency of the disk, t is the laboratory time frame, and k is the groove density chirp parameter with units of cycles/unit-length (i.e. for a $k = 1 \text{ mm}^{-1}$ the reticle will impart 1 period of modulation at a radius of 1 mm). An example of such a reticle is shown in Fig. 1(c). The reticle is oriented such that the x_2 -axis lies along the radius. The modulation from the mask is symmetric about $t = 0$ (for clarity, the polar angle 0° of the reticle coincides with $t = 0$), where the maximum rotation occurs for a reticle angle of $\pm\pi$.

$w(t) = \text{rect}\left(|t| < \frac{1}{2v_r}\right)$ is a window function that enforces this: the $-\pi \rightarrow \pi$ polar angle

extent of the physical reticle. This window however is subject to variability, e.g. it can be truncated to effect a null in the spatial modulation pattern to create a synchronization signal. If a spatial-light modulator (or another modulator capable of projecting a Ronchi ruling onto the field at the joint Fourier pane of the Martinez) were used in place of reticle, then the y dependence of $m(x_2, y_2; t)$ would be dropped.

The reticle is spatially offset from the origin of x_2 by x_c , where x_c is the radial location ω_0 impinges on the mask. This produces a carrier frequency of $\nu_c = k\nu_r x_c$, i.e. the modulation frequency of the central optical frequency, λ_0 .

We are only considering three fields diffracted by the modulator: a zero-order (undiffracted) field and the ± 1 -order fields. These three fields would be present for most modulator gratings, but the scaling coefficients would need to be adjusted appropriately to incorporate the possibility of higher-diffracted orders. The analysis that follows from here however, for the dynamics resulting from the superposition of these three fields remains accurate aside from a change in scaling coefficients.

Upon passage through the modulator, the field in Eqn. 4 becomes

$$E_2(x_2, y_2; \Omega, t) = E'_2(x_2, y_2; \Omega)m(t) = w(t)(\nu'_0 + \nu'_1 + \nu'_{-1}), \quad (6)$$

where an Euler expansion of the mask modulation function results in the following field terms [14]:

$$\begin{aligned} \nu_0^1(x_2, y_2; \Omega) &= \frac{1}{2}E_0 \frac{e^{i4\pi f / \lambda}}{i\lambda f} a_\Omega(\Omega) A_d\left(\frac{x_2 - \lambda f \mu \Omega}{\lambda f}, \frac{y_2}{\lambda f}\right) \\ \nu'_{\pm 1}(x_2, y_2; \Omega, t) &= \frac{1}{4}E_0 \frac{e^{i4\pi f / \lambda}}{i\lambda f} a_\Omega(\Omega) A_d\left(\frac{x_2 - \lambda f \mu \Omega}{\lambda f}, \frac{y_2}{\lambda f}\right) \quad (7) \\ &\quad e^{\pm i2\pi \nu_c t} e^{\pm i2\pi \Delta k \nu_r t x_2} e^{\pm i\Delta k y_2} \end{aligned}$$

C. 2-F propagation to the second dispersive element

To propagate the modulated field from the joint Fourier plane of the Martinez, plane (x_2, y_2) , to the second dispersive element, plane (x_4, y_4) , we use another scaled 2D spatial Fourier transform. The field incident on the second dispersive element is

$$E'_4(x_4, y_4; \Omega, t) = w(t)(\nu_0 + \nu_1 + \nu_{-1})e^{-i2\pi \mu \Omega x_4}, \text{ with}$$

$$\begin{aligned} \nu_0(x_4, y_4; \Omega) &= -\frac{1}{2}E_0 a_\Omega(\Omega) a_s(-x_4, -y_4) \\ \nu_{\pm 1}(x_4, y_4; \Omega, t) &= -\frac{1}{4}E_0 a_\Omega(\Omega) e^{\pm i2\pi \nu_c t} e^{\pm i2\pi \lambda f \Delta k \nu_r t \mu \Omega} \quad (8) \\ &\quad a_s\left(-(x_4 \mp \lambda f \Delta k \nu_r t), -(y_4 \mp \lambda f \frac{\Delta k}{2\pi})\right), \end{aligned}$$

and where the common phase pre-factor has been dropped.

The second dispersive element applies the same complex phase term as before but in the (x_4, y_4) plane: $\eta_d(\Omega)e^{i2\pi \mu \Omega x_4}$. As it should for a symmetric Martinez, the angular dispersion from the second dispersive element cancels the angular dispersion phase ramp of the exiting

field. Again neglecting the diffraction efficiency component as it is readily included in the spectral envelope $a_0(\Omega)$, the field leaving the light labeling Martinez is $E_4(x_4, y_4; \Omega, t) = \omega(t) (v_0 + v_1 + v_{-1})$. In Eqn. 8 we explicitly show the dependent variables of the v_j 's for completeness, but will now suppress them for brevity.

The v_0 field is a zero-order term from the field undiffracted by the modulator. The $v_{\pm 1}$ fields are the modulator diffracted fields, which contain a time varying spatial chirp in x and a constant spatial chirp in y . It is the interference between the v_j fields and their conjugates which gives rise to the light labeled signals.

3. LIGHT LABELED SIGNALS

Single-point, or bulk, spectral measurements are not insensitive to the spatial chirp on the $v_{\pm 1}$ fields. Wide-field imaging applications however, would not illuminate the sample or target with a spatially independent power spectrum. Therefore, we seek to eliminate the spatial chirp from the $v_{\pm 1}$ fields to increase the utility of the light labeling system beyond singlepoint power spectral measurements.

A. Spatial Filtering

To eliminate the spatial chirp, we will apply a spatial filter by coupling the v_j fields into a single mode optical fiber, where $j = 0, \pm 1$. Here the single mode fiber acts as both a critical part of the light labeling system as well as adding the benefits of a fiber delivery system. For simplicity, we assume the collimation lens for the fiber is situated a focal length away from the second dispersive element in the Martinez, allowing us to apply again use the 2- F propagator to compute the field distributions at the entrance face of the optical fiber (i.e. the single mode fiber entrance face is in a plane conjugate to the joint Fourier plane of the Martinez). Setting the collimator focal length to f_c , the fields at the fiber face are:

$$v_{\text{SMF}, j} = \frac{1}{\lambda f_c} a_j E_0 a_0(\Omega) e^{ji2\pi v_c t} e^{ji2\pi \lambda f \Delta k v_r t \mu \Omega} e^{-ji2\pi \left(x \frac{f \Delta k v_r t}{f_c} + y \frac{f \Delta k}{f_c 2\pi} \right)} A_s \left(-\frac{x}{\lambda f_c}, -\frac{y}{\lambda f_c} \right), \quad (9)$$

where $a_0 = \frac{1}{2}$, $a_{\pm 1} = \frac{1}{4}$, and we have suppressed the common phase pre-factor (of form $\frac{e^{i4\pi f / \lambda}}{i}$).

For convenience, we will assume an idealized case where the spatial mode of the optical fiber, a_{SMF} , is equivalent to the focused zero-order spatial distribution:

$a_{\text{SMF}} = \frac{1}{\lambda f_c} A_s \left(\frac{x}{\lambda f_c}, \frac{y}{\lambda f_c} \right)$ The transmission efficiency of the field terms are given by a normalized overlap integral [15]:

$$\eta_{\text{SMF},j}(\Omega, t) = \frac{\int \int_{-\infty}^{\infty} v_{\text{SMF},j} a_{\text{SMF}}^* dx dy}{\sqrt{\int \int_{-\infty}^{\infty} |v_{\text{SMF},j}|^2 dx dy \int \int_{-\infty}^{\infty} |a_{\text{SMF}}|^2 dx dy}}, \quad (10)$$

where the superscript * denotes the complex conjugate.

By design, $\eta_{\text{SMF},0} = 1$, and while this neglects any Fresnel reflections from the incident and exit faces, these quantities are both small and approximately equivalent across the j terms. The field transmission efficiency for the $v_{\pm 1}$ fields is dependent on both angular optical frequency and time, as the modulator diffraction changes as the the reticle rotates. Canceling the remaining common pre-factors results in a transmission function of

$$\eta_{\text{SMF},\pm 1}(\Omega, t) = \int \int_{-\infty}^{\infty} e^{\mp i 2\pi \left(x \frac{f \Delta k v_r t}{f c} + y \frac{f \Delta k}{f c 2\pi} \right)} \left(\frac{1}{\lambda f_c} \right)^2 A_s^2 \left(\frac{x}{\lambda f_c}, \frac{y}{\lambda f_c} \right) dx dy. \quad (11)$$

The simplification in Eqn. 11, assumes normalized spatial distributions otherwise Eqn. 10 is needed.

For convenience, we will also assume 2- F propagation from the exit fiber face utilizing an identical collimation lens as for the fiber entrance, f_c . The net effect of the single mode fiber is the elimination of all spatial chirp from the $v_{\pm 1}$ fields:

$$\begin{aligned} v_0 &= -\frac{1}{2} E_0 a_{\Omega}(\Omega) a_s(x, y) \\ v_{\pm 1} &= -\frac{1}{4} E_0 a_{\Omega}(\Omega) \eta_{\text{SMF},\pm 1}(\Omega, t) a_s(x, y) \quad (12) \\ &\quad e^{\pm i 2\pi v_c t} e^{\pm i 2\pi \lambda f \Delta k v_r t \mu \Omega} \end{aligned}$$

This reduction is both computationally convenient, and more critically, eliminates any spatial dependence in the light labeled spectral illumination. The fiber coupling term, $\eta_{\text{SMF},\pm 1}$, accounts for two critical consequences of diffraction from the modulator: reductions in the coupling efficiency and a spectral modulation temporal window both of which are functions of wavelength.

B. Light Labeled Signals

The zero-order field term is neither diffracted nor modulated whereas the ± 1 -order field terms are diffracted and contain a carrier oscillator. The total intensity contains interference between these terms,

$$I(x, y, \Omega, t) = |w(t)|^2 (v_0 + v_1 + v_{-1})(u_0 + u_1 + u_{-1}), \quad (13)$$

which can be separated into a DC (time-invariant) signal term and AC (time-varying), positive frequency signal terms:

$$\begin{aligned} I_0(x, y, \Omega, t) &= |w(t)|^2 (v_0 u_0 + v_1 u_1 + v_{-1} u_{-1}) \\ I_1(x, y, \Omega, t) &= |w(t)|^2 (u_0 v_1 + v_0 u_{-1}) \\ I_2(x, y, \Omega, t) &= |w(t)|^2 (v_1 u_{-1}), \end{aligned} \quad (14)$$

where u_j is the complex conjugate of v_j . Notice too, $|w(t)|^2 \propto w(t)$ as it is defined as a rectangular gate function.

A square-law detector captures the total intensity, integrating both the spatial and spectra dependencies of the intensity components. The acquired signal then, is $s(t) = s_0(t) + s_1(t) + s_2(t)$, where $s_k(t) = \iint_{-\infty}^{\infty} I_j(x, y, \Omega, t) dx dy d\Omega$ and $k = 0, 1$, and 2 . As we will show, we can extract the incident optical power spectrum from the complex analytic AC signal terms. The DC light labeled term, $s_0(t)$, is

$$s_0(t) = w(t) \cdot \frac{1}{4} I' \int_{-\infty}^{\infty} \left(1 + \frac{1}{2} \eta_{\text{SMF}, \pm 1}(\Omega, t) \right) |a_{\Omega}(\Omega)|^2 d\Omega, \quad (15)$$

where the integrated spatial intensity is $I' = E_0^2 \int_{-\infty}^{\infty} a_s^2(x, y) dx dy$. As there is no carrier oscillator, the only information encoded into $s_0(t)$ is the time-varying integrated effect of $\eta_{\text{SMF}, \pm 1}$. As with many DC terms, s_0 yields no distinct spectral information about the system.

The first-order, positive frequency light labeled signal term centered at v_c is

$$\begin{aligned} s_1(t) &= w(t) \frac{1}{4} I' e^{i2\pi v_c t} \\ &\int_{-\infty}^{\infty} \eta_{\text{SMF}, \pm 1}(\Omega, t) |a_{\Omega}(\Omega)|^2 e^{i2\pi \lambda f \Delta k v_r t \mu \Omega} d\Omega. \end{aligned} \quad (16)$$

To interpret $s_1(t)$, we first re-cast the integral into a more familiar form assuming $\eta_{\text{SMF}, \pm 1}(\Omega, t) = 1$ (which we will show later is a reasonable approximation for many experimental configurations),

$$\Gamma(\tau) = \int_{-\infty}^{\infty} |a_{\Omega}|^2(\Omega) e^{i\tau\Omega} d\Omega. \quad (17)$$

Having recast the integral as an inverse Fourier transform of the power spectrum onto a temporal delay axis, τ , we find that the measured signal is directly proportional to the field autocorrelation. The temporal delay axis on which we acquire the field autocorrelation is $\tau = \alpha' t$, where $\alpha' = 2\pi\lambda f / kv_t\mu$. The first-order light labeled signal, in the absence of any spectral apodization from fiber coupling, is the field autocorrelation offset in the frequency domain by the light labeling carrier frequency: $s_1(t) \approx w(t) \frac{1}{4} I' e^{i2\pi v_c t} \Gamma(\alpha' t)$. Meaning, we can recover the power spectrum by capturing $s_1(t)$ at modulation carrier frequencies, v , far below the optical carrier frequencies and taking a Fourier transform:

$$\begin{aligned} S'_1(v) &= \frac{1}{4} I' \int w(t) \Gamma(\alpha' t) e^{-i2\pi v t} dt \\ S'_1(v) &= \frac{1}{4} I' \left(W'(2\pi v - 2\pi v_c - \alpha' \Omega) * |a_{\Omega}|^2(\Omega) \right), \end{aligned} \quad (18)$$

where $W'(\Omega)$ is the Fourier transform of the gate function $w(t)$ and $*$ denotes a convolution. It is the central modulation carrier frequency, v_c , which allows us to disentangle the recovered power spectrum, S'_1 , from the DC and higher-order light labeled signals.

In the limit that $W(\Omega)$ is approximately a Dirac delta function, the convolution indicates that the Fourier transform of the measured light labeled signal is the incident power spectrum, where the optical angular frequencies are scaled down to the measured frequency domain:

$$S'_1(v) \approx \left| a_{\Omega} \left(\frac{2\pi(v - v_c)}{\alpha'} \right) \right|^2 \cdot S'_1 \left(\Omega = \frac{2\pi(v - v_c)}{\alpha'} \right)$$

then, is the power spectrum recovered from the light labeled $s_1(t)$ signal within the $\eta_{\text{SMF},\pm 1} = 1$ approximation, where for a broad enough gate function it is identical to the incident power spectrum.

Within the $\eta_{\text{SMF},\pm 1} = 1$ approximation, the second-order, positive frequency, light-labeled signal is $s_2(t) \approx w(t) \frac{1}{16} I' e^{i4\pi v_c t} \Gamma(2\alpha' t)$. $s_2(t)$ contains less energy than $s_1(t)$ but encodes $2 \times$ the spectral resolution as $s_1(t)$. And again, it is the central modulation carrier frequency of $2v_c$ which separates the power spectrum recovered from the second-order light labeled signal from the DC and first-order signals.

Relaxing the $\eta_{\text{SMF},\pm 1} = 1$ approximation and more rigorously analyzing the transform of the field autocorrelation, we can derive a more general form of the power spectrum recovered by light labeling. The two factors which perturb light labeling from the field autocorrelation interpretation discussed above are: $\eta_{\text{SMF},\pm 1}(\Omega, t) \neq 1$ and τ is dependent on λ in addition to t .

We will address the simpler perturbation first, that the coupling into single mode fiber is not unity. For convenience of discussion, we will assume that $\eta_{\text{SMF},\pm 1}(\Omega, t)$ is separable into two components, $\eta_{\text{SMF},\pm 1}(\Omega, t) = \eta_1(\Omega)\eta_2(\Omega, t)$, which we will show in the next section is exact within a Gaussian model. η_1 imparts a constant spectral apodization onto the power spectrum recovered from light labeling and is due to the constant spatial chirp in y . η_2 modifies the gate function, $w(t)$, of the system due to the changing spatial chirp in x , limiting the maximum delay τ at which the field autocorrelation is recorded. In an identical manner to Fourier transform spectroscopy, reducing the total delay window over which the field autocorrelation is measured correspondingly reduces the spectral resolution of the system.

The second perturbation, $\tau = \tau(\lambda, t)$, acts as a chirp in the frequency axis of the power spectrum recovered by light labeling. Expanding the $\lambda\Omega$ within the kernel of the inverse Fourier transform in Eqn. 17 in a Taylor series, $\lambda(\Omega)\Omega = \lambda_0 f(\Omega)$ where

$$f(\Omega) = \sum_{n=1}^{\infty} \frac{(-1)^{(n-1)}\Omega^n}{\omega_0^{n-1}}, \text{ we find that this perturbation is a consequence of large fractional bandwidth } \left(\frac{\Omega}{\omega_0}\right)$$

Using $\alpha = 2\pi\lambda_0 f k v_r \mu$, we get a general form of the power spectrum recovered from light labeling:

$$S_1(\nu) = W\left(\frac{2\pi(\nu - \nu_c)}{\alpha} - f(\Omega)\right) * \left(\eta_1(\Omega) |a_{\Omega}(\Omega)|^2\right), \quad (19)$$

where $W(\Omega)$ is the Fourier transform of $w(t)\eta_2(\Omega, t)$.

$W(\Omega)$ determines the spectral resolution of the light labeling system. For systems where $\eta_2(\Omega, t) \approx 1$, $W(\Omega)$ is a sinc function with a width in the measured frequency domain of $2\nu_r$.

The spectral resolution then is $\delta\Omega = \frac{4\pi\nu_r}{\alpha} = \frac{2}{\lambda_0 f \Delta k \mu}$. Physically, $\delta\Omega$ in the limit of $\eta_2 \approx 1$ is the angular frequency bandwidth needed to subtend one period of the densest grating on the reticle, which is a period of $\frac{2}{\Delta k}$ at the $\pm\pi$ reticle polar angle.

For spectral bandwidths where the maximum $\frac{\Omega}{\omega_0} \ll 1$, we may use only the $n=1$ term, or $f(\Omega) \approx \Omega$. In the limit that $W(\Omega)$ is approximately a Dirac delta function and $\eta_1(\Omega) = 1$, the measured light labeled power spectrum is identical to the incident power spectrum on the axis $\Omega = \frac{2\pi(\nu + \nu_c)}{\alpha}$ (this is the precise field autocorrelation case we derived previously).

For cases where there the power spectrum has appreciable fractional bandwidth, the higher order terms in $f(\Omega)$ need to be retained. The measured angular optical frequency axis for broad bandwidth incident power spectra no longer has a linear mapping to the angular optical frequency axis of the incident power spectrum, $f(\Omega) = \frac{2\pi(\nu - \nu_c)}{\alpha}$. This is wholly

correctable by interpolating the recovered optical frequency axis back onto a linear domain and applying the Jacobian of $f(\Omega)$. We will show a demonstration of this axial de-chirping and spectral reshaping in the next section.

Finally we would like to point a more general consequence of the way we constructed the overlap coupling coefficient, Eqn. 11: it is also a spatial intensity integral. Meaning, the $\eta_{\text{SMF},\pm 1}$ overlap coupling coefficient is common to both systems where the output of the light labeling Martinez is focused onto the face of a single mode fiber **or** directly onto a sample. Therefore, the $\eta_{\text{SMF},\pm 1}$ coefficient is a more general overlap coefficient for the light labeling system than purely an apodizing term imparted by the single mode fiber spatial filter.

4. LIGHT LABELED SIGNALS FOR A GAUSSIAN MODEL

We will now examine the particular case of Gaussian spatial and Gaussian spectral distributions in order to highlight the salient features of light labeling for several wavelength regions of interest: visible, near-infrared, and mid-infrared. We will derive an expression for the overlap coupling coefficient, $\eta_{\text{SMF},\pm 1}$, and analyze its effects on the light labeled signal. We will also demonstrate the effects of large fractional bandwidths on the captured frequency axis caused by the higher-order terms in $f(\Omega)$, and how to correct the captured power spectrum.

A. Overlap Coupling Coefficient

First we must determine the general single mode fiber coupling coefficients $\eta_{\text{SMF},\pm 1}$ in Eqn.

11. Using a normalized Gaussian spatial frequency distribution, $A_s = \sqrt{2\pi}we^{-\pi^2w^2(f_x^2 + f_y^2)}$, where w is the e^{-1} waist of the electric field spatial distribution incident on the light labeling Martinez (and the waist of the single mode fiber spatial distribution is considered to be $\frac{\lambda f_c}{w}$). The coupling coefficient then, is

$$\eta_{\text{SMF}}(\lambda, t) = e^{-\left(\frac{f\Delta k}{\sqrt{2}w} \cdot \frac{\lambda}{2\pi}\right)^2} e^{-\left(\frac{f\Delta k}{\sqrt{2}w} \cdot \lambda v_r t\right)^2}, \quad (20)$$

where the two exponentials are the η_1 and η_2 terms, respectively, discussed previously in Section 3B. The ± 1 subscript is dropped as the coefficient is common to both v_1 and v_{-1} . The coupling coefficient is comprised of two terms: one independent of time and one dependent of time. The first term in Eqn. 20 accounts for a reduction in coupling into the single mode fiber due to the phase tilt imparted by the spatial chirp in y , which is independent of modulator orientation and therefore independent of time. It is however, dependent on λ since it is due to diffraction from an effective spatial grating in the y -dimension, causing a systematic apodization of the power spectrum, as seen in Fig. 2(a).

To capture the many possible experimental configurations of a light labeling system, we use as a single system variable $\frac{f\Delta k}{\sqrt{2}w}$ which we will refer to as the design variable. As an example

of the magnitude of the design variable, consider the following light labeling system: a Martinez containing a focusing element with a focal length $f = 150$ mm, a reticle with a $k = 10 \text{ mm}^{-1}$, and a spatial field waist of $w = 2.5$ mm equates to a design parameter of $0.4 \mu\text{m}^{-1}$. The design variable accounts for the coupling between the monochromatic wavelet focal spots incident on the reticle and the diffractive power of the modulator. Physically, the design variable multiplied by the wavelength, $\frac{\lambda f \Delta k}{\sqrt{2} w}$, is directly proportional to the ratio between the monochromatic wavelet focal spot size and the pitch of the densest grating on the reticle, $\frac{2}{\Delta k}$.

The white line in Fig. 2(a) is the design variable contour for a 10% reduction in power spectral density. The region to the left of the white line then, represents the range of optimal design variables resulting in minimal spectral apodization.

The second coupling coefficient term in Eqn. 20, η_2 , accounts for the spatial chirp in the x -dimension and is time dependent, since the diffraction from the modulator in x changes with the the modulator orientation. This coupling coefficient term has the effect of reducing the temporal window of the encoded spectral modulations, as it adds a Gaussian transmission filter to the reticle dictated window function $w(t)$. The reduction in the temporal window of spectral modulations reduces the system spectral resolution as it limits the maximal delays at which the field autocorrelation is recorded. This term too, is dependent on λ , so the system spectral resolution is also dependent on wavelength.

Fig. 2(b) shows the relative e^{-1} half-width of the Gaussian transmission filter to the half-width of $w(t)$ ($1/2 v_r$). We have artificially set all relative widths greater than 1 equal to 1 in Fig. 2(b), to increase the sensitivity of the color map for values less than 1. The spectral resolution will scale linearly with reductions in the width of the transmission filter.

The white contour line in Fig. 2(b) is the design variable contour for transmission filter widths equivalent to the width of $w(t)$. The white contour lines in Fig. 2(a) and (b) are very nearly identical, and would be identical if plotted at 90.4% instead of 90% in Fig. 2(a). A design variable that results in appreciable spectral apodization will also then, result in a reduced spectral resolution. The white contour line at $\frac{f \Delta k \lambda}{w} = 2\sqrt{2}$ therefore, represents a reasonable boundary for experimental setups where the approximation $\eta_{\text{SMF}} = 1$ is valid.

Fig. 2(b) also shows sample spectral bandwidths of 25 fs full-width at half-maximum pulses at 530, 1060, 2000, and 3500 nm (e^{-5} widths of Gaussian power spectra). Visible and near-infrared systems will suffer from relatively little spectral apodization or wavelength dependent spectral resolution allowing for an increase in the system performance at larger design variables. The mid-infrared spectral region has a more limited range of design variables supporting minimum spectral apodization and $w(t)$ limited spectral resolutions, a consequence of the greater diffraction from the modulator.

B. Frequency Axis Chirp at Large Fractional Bandwidths

At large fractional angular frequencies ($\frac{\Omega}{\omega_0}$), the higher-order terms in $f(\Omega)$ cause a chirp in the frequency mapping of the light labeling system (as opposed to the linear mapping for $f(\Omega) \approx \Omega$). For a particular pulse duration, the maximum fractional bandwidth scales linearly with the central wavelength. As a reference aide, we show in Fig. 3(a) how the fractional bandwidth of a 25 fs pulse scales with central wavelength. Generally, the effect on the recovered power spectra for systems in the visible and near-infrared wavelength regions is small, as the maximum fractional bandwidth is small ($\left| \frac{\Omega_{\max}}{\omega_0} \right| < 0.1$). In the mid-infrared wavelength region however, the uncorrected axis chirp leads to highly inaccurate recovered power spectra.

To demonstrate the axis chirp we show in Fig. 3 the recovered power spectrum, $S_1(\Omega)$ (black curve), when the incident power spectrum, $|a_\Omega(\Omega)|^2$, is a Gaussian comb uniformly spaced in angular frequency and spanning a fractional bandwidth of ± 0.3 (red curve). For purposes of clarity, we have set $\eta_{\text{SMF}} = 1$, which is reasonable for systems with $\frac{f \Delta k \lambda}{w} \ll 2\sqrt{2}$, and assumed $W(\Omega)$ is a Dirac delta function. Notice, the model shown in Fig. 3(b) is for arbitrary ω_0 , so the inaccuracies in the recovered power spectra are dependent purely on the relative angular frequency.

The axial mis-sampling of the recovered power spectrum evident in Fig. 3(b) is fully correctable using the analytic relationship between the frequency axis of the incident power spectrum and the measured frequency axis (recall $\alpha\Omega' = 2\pi\nu$ after re-centering at $2\pi\nu_c$). Interpolating $S_1(\Omega')$ onto the axis $\Omega' = f(\Omega)$ will re-locate the recovered power spectra onto the true angular frequency axis, blue curve in Fig. 3(c). The amplitude of the recovered power spectra is corrected by scaling the interpolated recovered power spectrum by the Jacobian of $f(\Omega)$, blue-dashed curve in Fig. 3(c).

5. EXAMPLE EXPERIMENTAL CASES

In this section, we tabulate several example light labeling systems in the visible, near-infrared, and mid-infrared wavelength regions. The spectral range and resolution of these systems is highly tailorable to the intended application. The total number of spectral points, M , recorded in each light labeling system is dependent on the total spatial extent of the spectral line focus impinging on the reticle, R . It is however, readily calculated by $M = \frac{\Delta\lambda}{\delta\lambda}$, which assumes that care has been taken to ensure the reticle radius is larger than the R .

System 1, in Table 1, is the system we recently used to perform wide-field imaging of background free absorption spectra [2]. The maximum frame rate of the high-speed camera (an Andor Neo operating at 735 Hz with a pixel area of 240×1000) constrained the k of the reticle, since the Nyquist limit set a hard ceiling on the highest measured frequency within the target power spectrum.

The other light labeling systems in Table 1 are theoretical, with the $\frac{\lambda_0 f \Delta k}{w}$ set to $2\sqrt{2}$ to demonstrate maximum system performances. For example, system 2 a reasonable high performance limit of the system demonstrated in Ref [2] (system 1 here). The only difference between this theoretical system and the demonstrated light labeling system is a 23% larger mode field diameter and a $14 \times$ larger reticle k (a reticle which we already use in spatial frequency-modulated imaging systems [6]).

The near-infrared system is a theoretical system designed for an angular optical frequency bandwidth supporting an 8 fs pulse centered at 800 nm. This system is designed to produce a light labeled signal at a carrier frequency above 50 kHz for capturing either hyperspectral images or hyperspectral photon migration signals without an array detector or camera.

The theoretical mid-infrared system is for the power spectral bandwidth of a 25 fs Gaussian pulse centered at $3.5 \mu\text{m}$. The mid-infrared wavelength region lacks inexpensive array detectors, making rapid parallel spectroscopic measurements quite difficult (or exceedingly expensive). A light labeling spectrometer in this wavelength region can attain similar spectroscopic performances as are readily attainable in the visible and near infrared wavelength regions with a Czerny-Turner spectrometer and an array detector, without the interferometric stability and high temporal delay precision requirements of a traditional Fourier transform spectrometer or slow update rate of a scanning monochromator.

The spectral update rate for these example systems can be as high as a few 100 Hz with a spinning disk reticle and over a 1 kHz with a polygonal mirror or resonant galvo mirror. Combining a light labeling spectrometer with spatial frequency modulated imaging, would enable a high-speed two-dimensional imaging system captured on a single element detector with possible hypercube acquisition rates beyond video rate.

The benefit of the spinning disk reticle is the simplicity of the optical system and the economic price point. For example, our current system uses a brushless DC motors (Faulhaber, ~ \$400), reticles printed on a projector *GOBO* (~ \$200), diffraction gratings (~ \$400), and achromatic lenses (~ \$500) for a net cost of \$1500.

We think the inherent interferometric stability of the light labeling system, along with the scalable spectral acquisition rate makes for a unique and adaptable means of encoding spectroscopic information. It opens up up avenues of hyperspectral imaging by eliminating the back-end spectrometer from an experimental setup and instead utilizing the high bandwidth of single-element photo-detectors. The formalism presented here will enable the design and implementation of light labeling systems which maximizes the performance of future devices while minimizing systemic spectral apodization.

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APPENDIX

We now need to determine the effect of the dispersive elements on the incident spectral field. We will analyze the two primary cases used in the laboratory: diffraction gratings and prisms. We begin by noting that the incident field is oriented relative to the surface normal of the entrance face of the dispersive element. The incidence angle γ is pseudo-arbitrary, where we will re-orient the outgoing field such that the z axis collinear with the central wavelength. Of course, γ will almost always be set to either the Littrow angle or the angle of minimum deviation of the central wavelength. The task is to determine the functional form of μ , for the transformations imposed by either a grating or prism.

A. Model for Diffraction Gratings

The grating applies a phase term of $t_d(\Omega) = \eta_d(\Omega)e^{-2\pi N x}$ for the -1 diffracted order. The cumulative phase term governing the outgoing diffracted spectral field is

$e^{i2\pi \frac{\sin(\theta')}{\lambda} x} = e^{i2\pi \left(\frac{\sin(\gamma)}{\lambda} - N \right) x}$, where equating arguments in the exponential we see that this is equivalent to the grating equation. Representing the diffraction angle as a Taylor expansion in Ω , we have $\theta'(\Omega) = \theta_0 + \frac{2\pi c N}{\omega_0^2 \sin(\theta_0)} \Omega$, where $\theta_0 = \arcsin(\sin(\gamma) - N\lambda_0)$ is the diffraction

angle of the central wavelength. For the particular case of $\sin(\gamma) = \frac{N\lambda_0}{2}$ (the Littrow angle), $\theta_0 = -\gamma$.

The outgoing spatial phase now needs to be oriented such that the z axis coincides with the propagation axis of the central wavelength: $\theta(\Omega) = \theta'(\Omega) - \theta_0$. Using a small angle

approximation, the spatial phase front is $e^{i2\pi \left(\frac{2\pi c N \Omega}{\lambda \omega_0^2 \cos(\theta_0)} \right) x}$. Using the fractional bandwidth relation $\omega_0 \gg \Omega^2$, we can approximate the spatial phase term as $e^{i2\pi \mu_g \Omega x}$, where $\mu_g = \frac{N}{\omega_0 \cos(\theta_0)}$.

The spectral field impinging on the second diffraction grating has the phase term $e^{-i2\pi \mu_g \Omega x}$, which is equivalent (within the same approximations made above) to

$e^{i2\pi \frac{-\sin(\theta(\Omega))}{\lambda} x}$. Rotating this phase term back to the grating orientation and applying the

grating phase, we have $e^{i2\pi \frac{\sin(\gamma'(\Omega))}{\lambda} x} = e^{i2\pi \left(\frac{\sin(-\theta'(\Omega))}{\lambda} - N \right) x}$. From inspection, the outgoing, common field angle is $\gamma'(\Omega) = \gamma$, which is what we expect for a 4- F configuration, where the second dispersive element removes all spectral dependence to the propagation orientation.

B. Model for Prisms

The net diffraction of a prism does not lend itself a priori to the simplicity of a single transformation term, such as the $e^{-i2\pi N x}$ term of a diffraction grating. Instead, we will employ geometric optics to determine the spatial phase exiting the prism.

Collapsing the effects of the prism onto a plane (i.e. a thin prism approximation), the exit angle of the spectral field relative to the surface normal of the exit face of the prism is $\sigma'(\Omega) = \arcsin(\sin(\phi)\sqrt{n^2(\Omega) - \sin^2(\phi)} - \sin(\gamma)\cos(\alpha))$, where ϕ is the apex angle of the prism and $n(\Omega)$ is the index of refraction of the prism. As with the diffraction grating case, we now seek to orient the outgoing spectral field such that the z axis lies along the propagation axis of the central wavelength: $\sigma(\Omega) = \sigma'(\Omega) - \sigma_0$, where $\sigma_0 = \sigma'(0)$ is the diffraction angle of the central wavelength.

Expanding $\sin(\sigma(\Omega))$ in a Taylor series, we get an approximation of the outgoing spatial

$$\text{phase: } e^{i2\pi \frac{\sin(\sigma(\Omega))}{\lambda} x} \approx e^{i2\pi \frac{\mu'_p \Omega}{\lambda} x} \text{ where}$$

$$\mu'_p = \frac{\sin(\phi)n(\omega_0) \frac{dn}{d\Omega} |_{\omega_0}}{(n^2(\omega_0) - \sin^2(\gamma))\sqrt{1 - (\sin(\sigma'(\omega_0)))^2}}.$$

Applying the same fractional bandwidth approximation as before, and the outgoing spatial phase term is $e^{i2\pi\mu_p\Omega x}$, where $\mu_p = \frac{\mu'_p}{\lambda}$. And again, the field incident on the second prism has a spatial phase term of $e^{-i2\pi\mu_p\Omega x}$: the exact inverse of the field leaving the first prism.

Defining a prism transfer function $e^{-i2\pi Px}$ by the relationship $e^{-2\pi \frac{\sin(\gamma)}{\lambda} x} e^{-i2\pi Px} = e^{i2\pi\mu_p\Omega x}$, it is evident that for an impinging field with the spatial phase $e^{-i2\pi\mu_p\Omega x}$, the common exit angle is $-\gamma$.

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Fig. 1.

(a) Schematic of a light labeling 4- F Martinez and single mode fiber (SMF) filter. The spatial distributions at (b) the first dispersive element (DE) in plane (x_0, y_0) , incident on the frequency modulated reticle at plane (x_2, y_2) , (d) at plane x_3, y_3 , and (e) at the second DE in plane (x_4, y_4) for the example power spectrum in (b). f and f_c denote the focal lengths of the lenses.

Fig. 2.

(a) The time invariant, spectral apodization of the recovered power spectrum, η_1 , as a function of wavelength and $\frac{f\Delta k}{\sqrt{2}w}$ (b) The e^{-1} temporal half-width of the η_2 Gaussian window relative to the half-width of $w(t)$ as a function of wavelength and $\frac{f\Delta k}{\sqrt{2}w}$, where the color map has a log scaling. The vertical white lines indicate the spectral bandwidth of 25 fs full-width at half-maximum pulses centered at 0.53, 1.06, 2, and 3.5 μm.

Fig. 3.

(a) The central wavelength as a function of the e^{-5} fractional bandwidth of 25 fs full-width at half-maximum Gaussian pulses. (b) The incident power spectra (red curve), the recovered power spectra (black curve), the axially corrected recovered power spectra (blue curve), and the Jacobian of $f(\Omega)$ (blue-dashed curve) as functions of fractional angular frequency.

Table 1.

Example Light Labeling Systems

System #	λ_0 [μm]	λ [μm]	N [1/mm]	f [mm]	k [mm ⁻¹]
1 [2]	0.53	0.07	1200	200	5
2	0.53	0.07	1200	200	70
3	0.80	0.3	1200	66	35
4	3.50	1.8	150	150	20

System #	$\frac{\lambda_0 f \Delta k}{w}$ [-]	r [mm]	$\delta\lambda$ [nm]	M #
1	0.25	18	1.6	44
2	$2\sqrt{2}$	18	0.1	620
3	$2\sqrt{2}$	29	0.6	473
4	$2\sqrt{2}$	44	4.3	430