# Optical Preprocessing in Spectroscopy

*Miro Taphanel*

Vision and Fusion Laboratory
Institute for Anthropomatics
Karlsruhe Institute of Technology (KIT), Germany
miro.taphanel@ies.uni-karlsruhe.de

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**Abstract:** This technical report reviews the state of the art in optical preprocessing for spectroscopic applications. In particular, unconventional spectral techniques are reviewed that do not use a spectrometer, or make no use of it in the classical manner. In a theoretical part of this technical report, a vector space representation is derived to describe spectral processing and necessary assumptions are outlined. Based on this mathematical concept, optimal optical filters for spectroscopic applications can be designed. The second part of the technical report is about the optical hardware of these unconventional spectral technics.

# 1 Introduction

The near-infrared NIR spectral range $800 - 2500$nm is very interesting for technical applications. In contrast to the visible range, a transmission or reflectance spectrum is related to stretch-and-bend vibrations of covalent bonds in molecules. In detail, NIR reports on $1^{\text{st}}$, $2^{\text{nd}}$ and $3^{\text{rd}}$ vibrational overtones, combinations, and echoes of those that occur in the mid-IR [GL10]. These mechanisms cause broaden, often overlapping, peaks. On the other hand, the mid-IR range has higher technology cost.

In addition to the classical spectroscopy, a lot of efforts were done to develop unconventional methods, which do not use a spectrometer, or not in the classical way [Bia86b, BBWB08a, MSK$^+$02a, PSB99, DUDL07, FH95a, FH95b, MSL$^+$01a, HM04a, NAD$^+$98a, CUL05a, SGB11]. The reasons for these developments are higher signal-to-noise ratio [BBWB08a], easier instrumentation [Bia86b] and faster data acquisition [MSK$^+$02a]. Every time, when an application is limited by noise, costs, or speed, these technics can be an option.

Over the years a lot of keywords were used for merchandising purposes. This is an overview for the most important acronyms:

- OSP Optical Signal Processing [Bia86b]
- OC Optical Computing [ML05]
- OR Optical Regression [PSB99]
- MOE Multivariate Optical Element [MSK$^+$02a]
- MFC Molecular Factor Computing [DUDL07]
- CP Computational Photography [HKW]
- MOC Multivariate Optical Computation [BBWB08a]
- ISP Integrated Sensing and Processing [ML05]
- HICI Hyperspectral Integrated Computational Imaging [CUL05a]
- PAT Process Analytical Technology [DUDL07] (general topic)

# 2 Vector Space Representation for Continuous Light Spectra

The natural character of light spectra is continuous. The natural character of measured light spectrum is discrete. In between there is a sampling process, which is referred to as measurement process. The reduction of continuous spectra into discrete values allows to arrange these as a vector. The vector $\mathbf{t} = (t_1, t_2, \cdots, t_n)^\top$ describes a transmission spectrum sampled at $n$ discrete wavelengths.

In the following, it is discussed how this vector representation can be used to describe an optical filtering process and a filtering process in combination with an additional camera sensor. A filter $\mathbf{f} = (f_1, \cdots, f_n)^\top$, regardless of its optical realization, describes a wavelength dependent percental transmission $f_i \in [0, 1]$. This transmission describes the percental throughput of intensity, when light passes this element. The spectrum of the light intensity $\mathbf{I}^0 = (I_1^0, \cdots, I_n^0)^\top$ is modulated according an element-wise multiplication:

$$\mathbf{I}^1 = \mathbf{f} \circ \mathbf{I}^0 = (f_1 I_1^0, \cdots, f_n I_n^0)^\top,$$

when passing the filter $\mathbf{f}$. In the case of an additional photon sensitive sensor, which is typically sensitive for multiple wavelengths. The sensor signal $s$ becomes:

$$s = \alpha \mathbf{f}^\top \mathbf{I}^0 \sim \mathbf{f}^\top \mathbf{I}^0, \tag{2.1}$$

and can be described as vector dot product. The factor $\alpha$ is necessary for physical unit consistency, because the unit of $s$ is not specified here. In literature, $s$ is often defined as gray value. In this case $\alpha$ is a factor that maps light intensity to grey value. The target of most of the unconventional spectroscopic technics is to connect the sensor signal $s$ with a quantity of interest. So far no assumptions were necessary.

## 2.1 Vector Space Representation and Concentration Changes

Spectroscopy is well suited to measure concentrations of molecules in mixtures. In this article, the convention is used to name a specimen *analyt* if concentrations are of interest and *material* if the chemical composition is static. Furthermore, unimportant analyts are named as *interferents*. The following section discusses the problem of changing concentrations in spectroscopy [Mor77c] and the use of a vector representation. The impact of concentration changes to the analyt spectrum can be modeled by the Beer-Lambert law:

$$t_A(\lambda) = \frac{I^1(\lambda)}{I^0(\lambda)} = \mathrm{e}^{-\epsilon_A(\lambda) c_A l}. \tag{2.2}$$

The continuous transmission spectrum $t_A(\lambda)$ is defined as the ratio of the output intensity $I_1(\lambda)$ behind the analyt $A$ and input intensity $I_0(\lambda)$, respectively. The transmission $t_A(\lambda)$ is an exponential function of the absorption coefficient $\epsilon_A(\lambda)$, the concentration $c_A$, and the path length $l$ of the light travelling through the volume. In general, there is no limitation to transmission spectra $t_A(\lambda)$, however, the path length $l$ of a reflectance spectrum $r_A(\lambda)$ is unknown and must be determined empirically. In vector space representation, equation (2.2) can be rewritten as:

$$\mathbf{t}_A = \frac{\mathbf{I}^1}{\mathbf{I}^0} = \mathrm{e}^{-\boldsymbol{\epsilon}_A c_A l},$$

where $\mathbf{t}_A$ and $\boldsymbol{\epsilon}_A$ are vectors. According to the Beer-Lambert law, there is no linear relationship between a transmission spectrum $t_A(\lambda)$ and a concentration $c_A$. To use this law in a linear vector space it needs to be linearized. For example, if the signal $s$ of equation (2.1) shall be proportional to a concentration $c$ and the task is to determine an adequate filter vector $\mathbf{f}$, this undertaking will fail due to the nonlinearity of the Beer-Lambert law. However, under the assumption of small

concentrations $c \approx 0$ the Beer-Lambert law can be suitable linearized using a Taylor approximation of degree 1:

$$\begin{aligned} t_A(-\epsilon_A(\lambda)c_A l) &= \mathrm{e}^{-\epsilon_A(\lambda)c_A l} \\ &\approx \mathrm{e}^{-\epsilon_A(\lambda)c_A l}|_{c_A=0} + (c_A - 0)^1 \frac{\partial \mathrm{e}^{-\epsilon_A(\lambda)c_A l}}{\partial c_A}|_{c_A=0} \\ &= 1 - \epsilon_A(\lambda)c_A l \end{aligned}$$

In this form the transmission spectrum $t_A(\lambda)$ scales linear with changes in concentration $c_A$. Hence, it is possible to use equation (2.1) to find a linear relationship of signal $s_A$ and a concentration $c_A$. This is an approximation and will fail when concentrations become to high.

Normally, applications do not just deal with one analyt, but a mixture of them. The Beer-Lambert (2.2) law can be extended for this case. If a second absorber $B$ is introduced, the overall transmission is the entry-wise product of the transmission $\mathbf{t}_A$ and $\mathbf{t}_B$.

$$\mathbf{t} = \mathbf{t}_A \circ \mathbf{t}_B = \mathrm{e}^{-\boldsymbol{\epsilon}_A c_A l} \circ \mathrm{e}^{-\boldsymbol{\epsilon}_B c_B l} = \mathrm{e}^{-\boldsymbol{\epsilon}_A c_A l - \boldsymbol{\epsilon}_B c_B l}$$

For small concentrations this can be approximated to:

$$\mathbf{t} = \mathbf{1} - \boldsymbol{\epsilon}_A c_A l - \boldsymbol{\epsilon}_B c_B l.$$

and an useful extension is to use the difference from a reference level:

$$\boldsymbol{x} = \mathbf{1} - \boldsymbol{t} = \boldsymbol{\epsilon}_A c_A l + \boldsymbol{\epsilon}_B c_B l \tag{2.3}$$

In this form $\boldsymbol{x}$ is the absorbance vector as the result of the vector addition of the absorbance components $\boldsymbol{\epsilon}_A c_A l$ and $\boldsymbol{\epsilon}_B c_B l$.

The assumption of small concentrations is quite strict. The same formula (2.3) can be derived by the use of negative log transmissions:

$$\boldsymbol{x} = -\ln(\mathbf{t}) = \boldsymbol{\epsilon}_A c_A l + \boldsymbol{\epsilon}_B c_B l$$

Although this is exactly the same result as (2.3), it is not an approximation anymore. The negative logarithm transformation converts the exponential Beer-Lambert law into a linear form. However, in practice every channel of a spectrometer must be transformed according to the negative logarithm separately. Furthermore, you one to ensure that the spectral width of the bandpass filters is small enough, so only slightly changes of $\epsilon(\lambda)$ can be guaranteed. In this case $\epsilon(\lambda)$ can be approximated by a single constant $\bar{\epsilon}(\lambda_k)$ for the spectral interval of the bandpass filter [Mor77c].

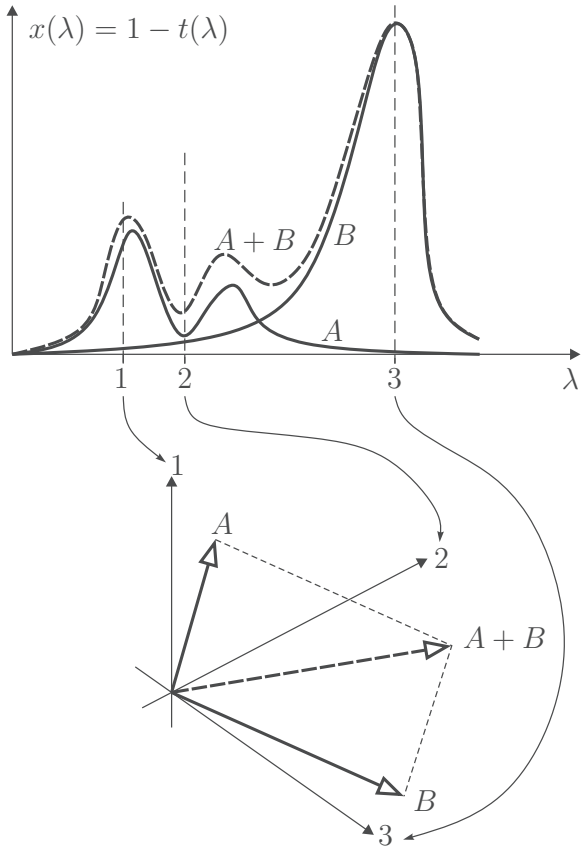


**Figure 2.1**: Vector space representation of sampled light spetra of analyt $A$ and $B$. Each sampling wavelength defines one dimension of the vector space. Extracted from [Mor77c]

In summary, with an approximation for low concentrations it is possible to derive a linear relationship of the concentration $c$ and the absorbance spectrum $\mathbf{x}$. Arbitrary concentrations can be handled by a linear model, if each channel is transformed according to the negative logarithm. The mixture of two absorbance spectra can be modeled by an addition of the absorbance vectors. These two properties proportionality and additivity satisfy the requirements of a linear vector space.

### 2.2 Vector Space Representation and Constant Concentrations

Applications using materials with a static chemical composition are quite common. For Example in remote sensing one task is to perform a spectral unmixing while the potential components are known. The problem in remote sensing is that each pixel of a hyperspectral imaging system images more than a single material. With the linear mixture model[KM02]:

$$\mathbf{r} = a\mathbf{r}_A + b\mathbf{r}_B + \cdots + \mathbf{n}$$

a measured reflectance spectrum $\mathbf{r}$ can be explained by a linear combination of known spectra $\{\mathbf{r}_A, \mathbf{r}_B, \cdots\}$, with linear coefficients $\{a, b, \cdots\}$ and a noise vector $\mathbf{n}$. The linear modell can be imagined as checkerboard distribution of multiple pure materials. In contrast to applications where the concentration of analyts can change, the chemical composition is fixed. For this reason the spectra of the known materials can be treated as ground truth, also called *endmembers*. The reflectance vector $\mathbf{r}$ is a linear combination of other vectors $\{\mathbf{r}_A, \mathbf{r}_B, \cdots\}$. The two properties proportionality and additivity are satisfied and the requirements for a linear vector space are fulfilled. With the vector dot product [Mor77c] it is possible, e.g., to design a filter $\mathbf{f}$, so that the signal $s_A = \mathbf{f}^\top \mathbf{r}$ is proportional to $a$, describing the spatial proportion of material $A$.

## 3 Optimal Filter Design Rules

The linear vector space theory is used in this section to derive optimal filter designs. The way how spectra can be represented in a linear vector space dependents on the application. If concentrations are of interest an analyt vector represents an absorbance vector $\mathbf{x}_A = -\ln(\mathbf{t}_A)$ or the approximation $\mathbf{x}_A = \boldsymbol{\epsilon}_A c_A l$ of analyt $A$. If reflectance spectra are treated as ground truth, the target vector $\mathbf{r}_A$ describes a reflectance vector.

In this section it is not necessary to distinguish between different applications any longer, because an uniform vector space representation is possible. In certain circumstances, a filter vector $\mathbf{f}$, however, cannot be directly interpreted as a filter transmission characteristic. Fore example if the vector $\mathbf{x}_A = -\ln(\mathbf{t}_A)$ is used instead of the transmission vector $\mathbf{t}_A$, the resulting optimal filter vector $\mathbf{f}$ should be transformed inversely $\mathbf{f}' = \mathrm{e}^{-\mathbf{f}}$, to receive the transmission characteristic for the optical filter.

The task of optimal filter design is to find a filter $\mathbf{f}$, so that a concentration or proportion of a target vector $\mathbf{a}$ can be determined. The relationship between a

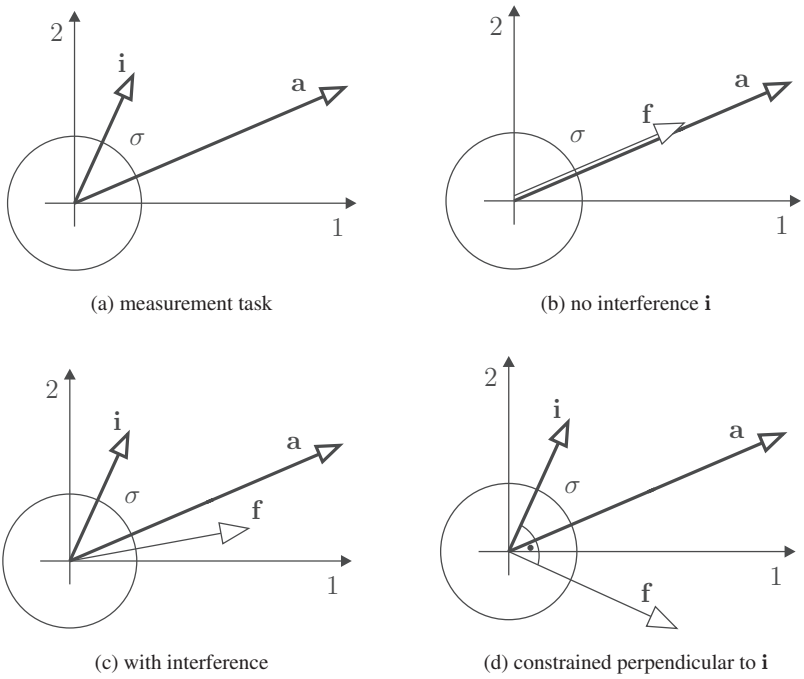


**Figure 3.1**: Optimal filter vectors $\mathbf{f}$ for different optimum criteria. A measurement vector $\mathbf{m} = \mathbf{a} + \mathbf{i} + \mathbf{n}$ is composed of a target vector $\mathbf{a}$, an interference vector $\mathbf{i}$ and a noise vector $\mathbf{n}$ depicted as the standard deviation $\sigma$. Vectors shown in a two dimensional vector space, with each dimension corresponding to a sample wavelength. Extracted from [Mor77c].

concentration and a target vector can be defined by the vector dot product.

$$s \sim \mathbf{f}^\top \mathbf{m},$$

in which $\mathbf{m} = \mathbf{a} + \mathbf{i}^1 + \mathbf{i}^2 + \cdots + \mathbf{n}$ is the vector addition of the target vector $\mathbf{a}$, the other interference vectors $\{\mathbf{i}^1, \mathbf{i}^2, \mathbf{i}^3, \cdots\}$, and a system noise vector $\mathbf{n}$. The filter $\mathbf{f}$ is designed in such a way, that the signal $s$ is proportional to a concentration or a proportion respectively. The system noise vector $\mathbf{n}$ is a random variable which describes the sum of all involved noise sources, with $\mathrm{E}\{\mathbf{n}\} = \mathbf{0}$ and $\mathrm{Cov}\{\mathbf{n}\} = \sigma^2\mathbf{I}$.

The optimal criteria can be defined heuristically, with or without constrains, and by a signal-to-noise ratio. In the following, different optimal filter vectors are designed using these optimal criteria. The first case is an optimal filter for a single target $\mathbf{a}$. The measurement vector is defined as $\mathbf{m} := \mathbf{a} + \mathbf{n}$ (depicted in Fig. 3.1b). Using the signal-to-noise ratio [Mor77c],

$$\begin{aligned} \mathrm{SNR} &= \frac{\mathrm{E}\{\mathbf{f}^\top \mathbf{m}\}}{(\mathrm{Var}\{\mathbf{f}^\top \mathbf{m}\})^{\frac{1}{2}}} \\ &= \frac{\mathbf{f}^\top \mathbf{a}}{\mathbf{f}^\top (\mathrm{Cov}\{\mathbf{n}\}\mathbf{f})^{\frac{1}{2}}}, \end{aligned} \tag{3.1}$$

as optimum criterion, an optimal filter $\mathbf{f}$ is found if both vectors $\mathbf{f} = \mathbf{a}$ point in the same direction, while the noise vector $\mathbf{n}$ is uniform and uncorrelated. The author[Mor77c] calls this filter the *matched filter*, because $\mathbf{f} = \mathbf{a}$.

If, in addition, an interferent vector $\mathbf{i}$ with random length disturbs the target vector, the optimal filter vector tends away from this interferent (depicted in Fig. 3.1c). The optimal filter vector then again can be derived by solving the SNR (3.1) for this case. According to [Mor77c] the solution is

$$\mathbf{f} = (\sigma^2\mathbf{I} + \sigma_\mathbf{i}^2 \mathbf{i}\mathbf{i}^\top)^{-1}\mathbf{a},$$

with the assumption of white noise and $\mathbf{I}$ as identity matrix.

To describe the distribution of the concentration of a known interferent $\mathbf{i}$ as white noise can be problematic. An option to get rid of this problem is to design the filter vector perpendicularly to the interferents. This is depicted in 3.1d for a single interferent. In this case, a change of the interferent analyt has no effect to the resulting signal $s$. If the filter vector $\mathbf{f}$ should be perpendicular to multiple interferents and the number of interferents is less than the dimension of the vector space, the target vector $\mathbf{a}$ can be projected onto the subspace of these interferents. Then, $\mathbf{f}$ is optained by the difference of $\mathbf{a}$ and the projection of $\mathbf{a}$ onto this subspace

[Mor77c]. In detail

$$\begin{aligned}\mathbf{f} =&(\mathbf{I} - \mathbf{\Phi}(\mathbf{\Phi}^\top\mathbf{\Phi})^{-1}\mathbf{\Phi}^\top)\mathbf{a} \text{ and}\\ \mathbf{\Phi} =& [\mathbf{i}^1, \mathbf{i}^2, \cdots],\end{aligned}$$

with the matrix $\mathbf{\Phi}$ that organizes the interferents vectors colom wise. Another way how to obtain a filter vector that is orthogonal to the interferents was introduced by [Bia86b]. By the Gram−Schmidt process a new orthogonal basis can be constructed with one direction orthogonal to all interferents. Again, the interferents vectors are arranged colom wise, but this time the target vector $\mathbf{a}$ is added at last

$$\begin{aligned}\mathbf{M} &= [\mathbf{i}^1, \mathbf{i}^2, \cdots, \mathbf{i}^p, \mathbf{a}]\\ \mathbf{M} &= \mathbf{QR} \text{ (QR-factorization)}\\ \mathbf{f} &= \mathbf{q}_l, \text{ with } \mathbf{Q} = [q_1, \cdots, q_l].\end{aligned}$$

This matrix $\mathbf{M}$ can be decomposed into an orthogonal matrix $\mathbf{Q}$ and an upper triangular matrix $\mathbf{R}$ using the Gram-Schmidt process. The optimal filter is then the last colom vector $\mathbf{q}_n$ of the matrix $\mathbf{Q}$.

Beside these analytical motivated methods, also heuristically methods are very common. In Section 2.1 it was already discussed, that changes in concentration cause nonlinear changes in the spectrum due to the Beer-Lambert law. In the majority of articles [BBWB08a][MSK$^+$02a][HM04a][NAD$^+$98a][DUDL07][PSB99], a linear regression approach is chosen, that do not pay attention to this problem. Often linear regression is combined with a principle component analysis. The linear regression model [HM04a]

$$s = \mathbf{f}^\top\mathbf{a} = \sum_{j=1}^{l} f_j a_j$$

can be reduced to a vector dot product with regression coefficients $f_j$ equal to the filter vector entries. $l$ is the maximal number of wavelengths and equivalent to the dimensionality of the vector space.

# 4 Filter Technology

This section gives an overview of different possibilities how optical filters can be realized. Only filter design methods are of interest, that allow to produce custom

filter transmission characteristics. In general, a filter can be placed before or behind the analyt, filtering the illumination part or the image formation part, respectively. If the illumination part is filtered, the filter need not fulfill image formation quality and this enables some methods that are prohibited in the image formation part. Optical filter technologies:

- Partial glass filter according to Dresler[Ric81]. Multiple standard glas filter are spatial assembled side by side and in series. The single glass filter fragments are chosen in such a way, that the resulting filter approximates the target filter characteristics. This filter is placed in the plane of the aperture.

- Chromatic light dispersion with filter mask [Bia86b]. This kind of filter works only in the illumination part. Polychromatic light is split up and projected onto a mask. The intransparent part of the mask absorbs wavelength dependent light proportions. After remixing the illumination, the light features the target spectrum. State of the art are programmable filter mask for prototyping issues.

- Narrowband laser line illumination [Mor77c]. Laser lines are monochromatic and can replace single channels of spectrometers.

- Liquid analyt mixture [DUDL07]. A cuvette is filled by multiple analyts of known spectral characteristics. The mixture can approximate a target filter characteristic.

- Interference filters [LM08]. A sequence of thin films is applied onto a substrate. In theory arbitrary target filter characteristics can be design.

- Nano structured plasmons filters [KTE+99]. Structured metal films with holes and other geometries in nano scale show are wavelength dependent transparency. The transmission characteristic can be influenced by the dimensions and the layout of the metal structure.

- Material as Screen for intermediate images [SGB11]. Similar to an analyt mixture that is used as transmission filter, intransparent materials can be used in reflectance mode as filter. Either as reflectance mirror in the illumination part, or as screen for intermediate images in the image formation part.

- Spectrometer with weighted integration time for each channel [BBWB08a]. This is a virtual filter and a spectrometer is still required. According to the target filter design the integration time is weighted for each channel.

These methods suffer from the problem of negative coefficients of the target filter design. This problem can only be solved by the use of two filters, one for the positive coefficients and one for the negatives. An elegant solution was proposed by [MSK+02a] who used an interference filter as beam splitter. The transmission characteristic of this interference filter approximates the positive target filter coefficients and the reflected the negative ones.

# 5 Conclusion

Unconventional spectroscopic technics can be described in an uniform way, using a vector space representation. Instead of a spectrometer, only one or a few sensors are used, together with complex filter transmission characteristics. According to the application an optimal criteria can be formulated to determine the corresponding optimal filter. On the hardware side, a lot of technics were developed to realize arbitrary filter transmission characteristics.

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