

## *Application of the Adjoint Spectrum to the Frequency Tracking in Spectrometry with Tunable Diode Lasers Suffering from Temperature Drift*

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### SYNOPSIS

A numerical technique called as "adjoint spectrum" has been proposed by the authors in a previous work. Two applications of it are presented here. It is demonstrated that by generalizing the aspect of adjoint spectrum it becomes possible to measure densities of mixed gases simultaneously. An experimental proof of this has been carried on and simultaneous measurement of densities of both methane and dinitrogen oxide was realized. On the other hand, it is also reported that frequency shifting of spectra, which is a cause of error in spectrometric measurements, can be corrected by exploiting the adjoint spectrum technique in connection with the Taylor expansion. Numerical examples showing the success of this method are reported as well.

### 1. INTRODUCTION

The adjoint spectrum technic has been proposed by the authors as an algorithm which eliminate predictable spectral interferences from a measured spectrum<sup>1)</sup>. Its fundamental idea has been translated from the theoretical treatise of modal analysis methods for neutron flux distribution in nuclear reactor cores<sup>2)</sup>. The nomenclature "adjoint" have its origin there. Some basic properties of the adjoint spectrum method have been described in our preceding work<sup>3),4)</sup>.

The fundamental principle of our adjoint method is based on the well-known least square fitting technique which is employed to determine several parameters of an experimental formula to the data obtained experimentally with significant noise. Uncertainty in the parameters of the formula is reduced by virtue of the limited autocorrelation between different data items. The number of the data items, namely, exceeds far more than the number of the parameters to be determined.

Recent progress in continuously tunable lasers allows to measure infrared spectrum of gaseous material of multiple constituents<sup>5),6)</sup>. A good linearity holds between their mixing ratio and the absorption spectra, and the least square method can exploit its power to its maximum.

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A differential absorption method has been employed in the field of laser radar to avoid an spectral interference of coexisting interfering species<sup>7),8)</sup>. This method is also convinced as a simplest case of our adjoint spectrum method.

In this paper<sup>7),8)</sup>, is demonstrated another application against the compensation of the laser frequency shift. In the application of a tunable diode laser spectrometry, the domain of the laser frequency scanning would move slightly, less than the width of an absorption line, due to inherent temperature fluctuation of the laser temperature less than 10 mK at around 77K. This fluctuation results in the span error of the spectrometry system, and this effect can be weakened by this method.

## 2. RECALL OF THE ADJOINT SPECTRUM METHOD<sup>4)</sup>

Let's  $S_x(\nu)$  be a spectrum of an atmospheric gas as measured with a spectrometer, and  $S_0(\nu)$  a spectrum of the same pure gas cell in a reference measured with the same system. In the absence of interferences,

$$S_x(\nu) = k_0 S_0(\nu) , \quad (1)$$

should hold, where  $k_0$  is proportional to the density of the gas measured.

If  $m$  species of foreign absorbers exist within the laser frequency region being scanned, the above expression becomes :

$$S_x(\nu) = k_0 S_0(\nu) + \sum_{i=1}^m k_i S_i(\nu) , \quad (2)$$

where  $S_i(\nu)$  is the spectrum corresponding to the  $i$ 'th interference absorber.

Since there exists an isomorphism between the measured spectra and vectors, eq.(2) can be written in a vectorial form as,

$$\mathbf{S}_x = k_0 \mathbf{S}_0 + \sum_{i=1}^m k_i \mathbf{S}_i . \quad (3)$$

Using the vector space theory, it was demonstrated that the coefficient  $k_0$ , which knowledge yields the target gas density, is obtained as follows :

$$k_0 = \frac{\langle \mathbf{S}_x, \mathbf{S}_0^* \rangle}{\langle \mathbf{S}_0, \mathbf{S}_0^* \rangle} \quad (4)$$

where,  $\mathbf{S}_0^*$  is the adjoint spectrum with respect to the reference spectrum  $\mathbf{S}_0$ , determined by,

$$\mathbf{S}_0^* = \mathbf{S}_0 - \sum_{i=1}^{\mu} \langle \mathbf{S}_0, \mathbf{u}_i \rangle \mathbf{u}_i , \quad (5)$$

where  $\{\mathbf{u}_i, i = 1, 2, \dots, \mu\}$  is an orthonormal vector space deduced from the sequence of interference vectors  $\mathbf{S}_i, i = 1, 2, \dots, m$  according to the Gram-Schmidt orthogonalization algorithm.

## 3. APPLICATION OF THE ADJOINT SPECTRUM TO MULTIDETECTION IN SPECTROMETRY

Suppose that  $n$  species of gaseous absorbers are to be measured simultaneously, which exist together with  $m$  different interferences in the same frequency region being scanned. In this case eq.(3) becomes,

$$\mathbf{S}_x = k_0 \mathbf{S}_0 + \dots + k_n \mathbf{S}_n + \dots + k_{n+\mu} \mathbf{S}_{n+\mu} \quad (6)$$

where the first  $n$  terms are for gases to be measured and the last  $\mu$  terms are for interferences.

To determine the coefficient  $k_i$  corresponding to the  $i$ 'th gas spectrum  $S_i$ , it is required only to find the adjoint spectrum  $S_i^*$  related to that spectrum using eq.(5) as,

$$S_i^* = S_i - \sum_{j=1}^{n+\mu} \langle S_i, u_{ij} \rangle u_{ij} \quad \text{with } i \neq j \quad (7)$$

where only  $S_i$  is considered as a reference spectrum and all other spectra including  $S_0$  as interferences. The coefficient  $k_i$  is then solved for using eq.(4), as

$$k_i = \frac{\langle S_x, S_i^* \rangle}{\langle S_i, S_i^* \rangle} \quad (8)$$

This is repeated until all coefficients  $k_i$  for  $i = 0, 1, \dots, n$  are computed, which yield the desired densities of the  $n$  species of gases. An experimental demonstration of this is given in section 4.

## 4. APPLICATION OF THE ADJOINT SPECTRUM METHOD TO FREQUENCY TRACKING PROBLEM

### 4.1 Principle

A serious problem which occurs in a tunable diode laser absorption spectrometry system is the shift of the laser frequency due to the slight drift of heat sink temperature as small as 10 mK at around 77K. This induces an error in the density of the measured gas especially in single-beam measuring systems. This phenomenon takes very long time to settle down and an external disturbance such as direct sunlight or a breeze of different temperature will again stimulates the temperature transients through the cryostat window or through the housing.

Now, if by some means the frequency shift,  $\Delta\nu$ , of a spectrum  $S(\nu)$  is detected and converted to an electrical current, this latter can then be fed back to control the laser driving current in such a way that the spectrum remain still within the prescribed spectral window.

Let's  $S(\nu)$  and  $S(\nu + \Delta\nu)$  be two spectra measured independently, with  $\Delta\nu$  the frequency shifting between them. Since the speed of the shifting is usually very slow as compared to the duration of measurement of one spectrum, the frequency variation  $\Delta\nu$  is sufficiently small and allows the Taylor expansion to be written in the following form :

$$S(\nu + \Delta\nu) \cong S(\nu) + \Delta\nu \frac{dS(\nu)}{d\nu} \quad (9)$$

or in a vectorial form as,

$$S_{\Delta\nu} \cong S + \Delta\nu S' \quad (10)$$

where  $S'$  is the vector corresponding to the first derivative spectrum  $dS(\nu)/d\nu$ .

Thus,  $\Delta\nu$  can be solved for by applying the adjoint spectrum as in eq.(4) :

$$\Delta\nu = \frac{\langle S_{\Delta\nu}, S'^* \rangle}{\langle S', S'^* \rangle} \quad (11)$$

where  $S'^*$ , the adjoint spectrum with respect to  $S'$  with  $S$  as interference, is given by :

$$S'^* = S' - \left\langle S', \frac{S}{\|S\|} \right\rangle \frac{S}{\|S\|} \quad (12)$$

It can be therefore concluded that the frequency shift interval,  $\delta\nu$ , can be determined by application of the adjoint spectrum in connection with the Taylor expansion for small variations. Hereafter, investigations on the limit of shift correction and precision of this method are demonstrated through a numerical example shown in the next.

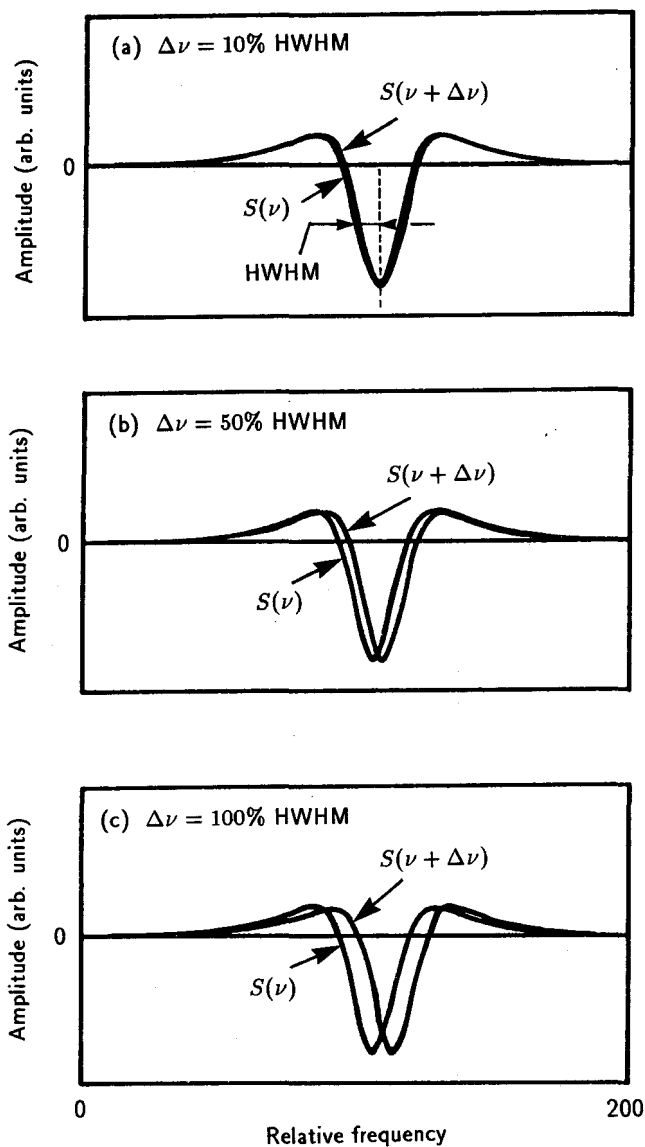


Fig.1 An example of absorption spectrum (a) and one which is obtained by a laser whose emission frequency has slightly shifted. Two spectra are generated numerically assuming a Lorentzian line profile and the second harmonic detection system.

## 4.2 Numerical Examples

Figure 1 is an illustration of the frequency shift of a second harmonic spectrum. The shift is expressed in terms of the half width at half maximum (HWHM) of the spectrum. From (a) through (c) the shift  $\Delta\nu$  is taken as, 10%, 50% and 100% of the HWHM respectively. To verify the results of section 4.1, the shift  $\Delta\nu$  is calculated as in eq.(11) and used in eq.(9) to obtain a shift-corrected spectrum  $S_{sc}(\nu)$  as,

$$S_{sc}(\nu) = S(\nu + \Delta\nu) - \Delta\nu \frac{dS(\nu)}{d\nu} \quad (13)$$

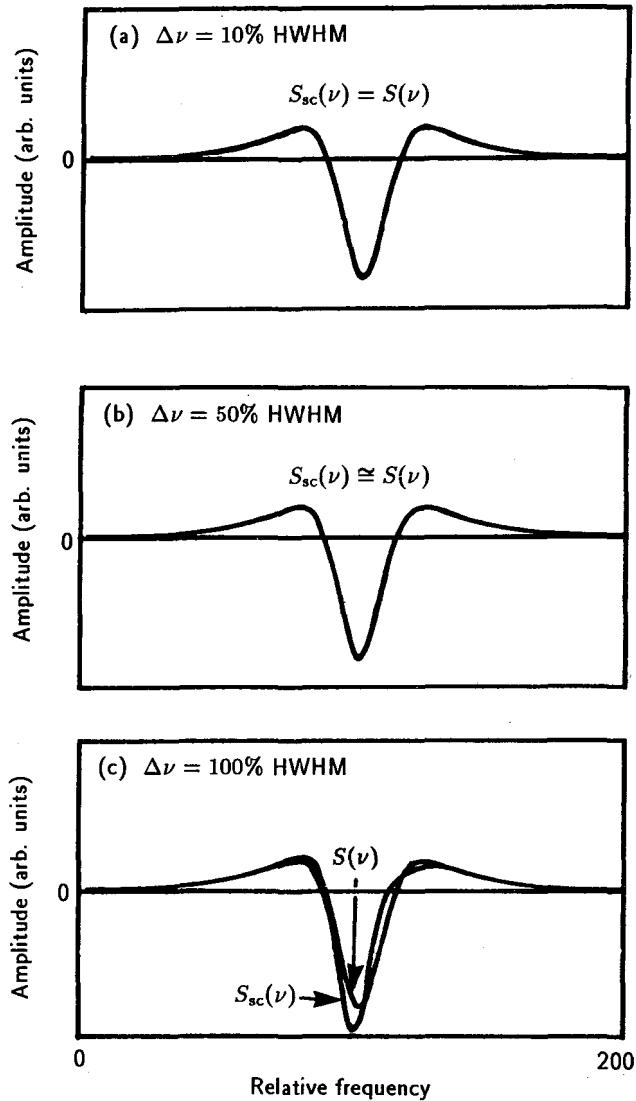


Fig.2 The shifted spectrum in Fig.1 and that reconstructed with the adjoint spectrum method. Only little difference is found between them, which demonstrate the validity of the algorithm.

This is displayed in Fig.2 together with the original spectrum  $S(\nu)$  for comparison. It can be verified that for values of the shift until about 50% HWHM the calculated spectrum and the original spectrum almost overlap which means that the shift has been corrected successfully with the proposed method. For larger shifts, a notable difference between both spectra arises and therefore the approximation given by eq.(9) is no more adequate and more terms of the Taylor expansion should be consider. In such a case, a direct search method should be employed though is beyond the present scope.

To investigate the accuracy of this method, an error  $E_r$  is defined as,

$$E_r = \frac{\|S(\nu) - S_{sc}(\nu)\|}{\|S(\nu)\|} \quad (14)$$

This is shown in Fig.3 as a function of the shift  $\Delta\nu$ . It can be concluded from this figure that for shifts under 30% HWHM the error resulting from the correction of the shift by the proposed method is as small as than 0.25%. This proves the extent of success of this method in correcting a spectrum shift.

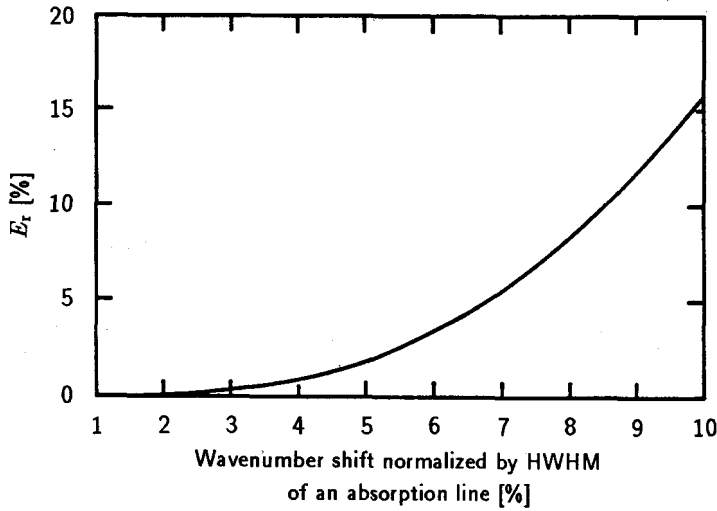


Fig.3 Relative error in reconstructing spectrum.

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