

## **Better Spectrometers, Beautiful Spectra and Confusion for All**

J. C. Pearson, C. S. Brauer, B. J. Drouin, & S. Yu

*Jet Propulsion Laboratory, California Institute of Technology, 4800 Oak  
Grove Drive., Pasadena, CA 91109*

**Abstract.** The confluence of enormous improvements in submillimeter receivers and the development of powerful large scale observatories is about to force astrophysics and the sciences that support it to develop novel approaches for interpretation of data. The historical method of observing one or two lines and carefully analyzing them in the context of a simple model is now only applicable for distant objects where only a few lines are strong enough to be observable. Modern observatories collect many GHz of high signal-to-noise spectra in a single observation and in many cases, at sufficiently high spatial resolution to start resolving chemically distinct regions. The observatories planned for the near future and the inevitable upgrades of existing facilities will make large spectral data sets the rule rather than the exception in many areas of molecular astrophysics. The methodology and organization required to fully extract the available information and interpret these beautiful spectra represents a challenge to submillimeter astrophysics similar in magnitude to the last few decades of effort in improving receivers. The quality and abundance of spectra effectively prevents line-by-line analysis from being a time efficient proposition, however, global analysis of complex spectra is a science in its infancy. Spectroscopy at several other wavelengths have developed a number of techniques to analyze complex spectra, which can provide a great deal of guidance to the molecular astrophysics community on how to attack the complex spectrum problem. Ultimately, the challenge is one of organization, similar to building observatories, requiring teams of specialists combining their knowledge of dynamical, structural, chemical and radiative models with detailed knowledge in molecular physics and gas and grain surface chemistry to extract and exploit the enormous information content of complex spectra. This paper presents a spectroscopists view of the necessary elements in a tool for complex spectral analysis.

### **1. General Introduction**

The spectral line survey of Sutton et al. (1985) and Blake et al. (1986) provided submillimeter astrophysics with the first demonstration of the power of a fairly complete (215-263 GHz) spectrum. The comparatively large frequency coverage provided the first comprehensive consistent spectroscopic picture of the Orion molecular cloud. The survey, though limited by modern standards in both frequency coverage and sensitivity, demonstrated the power of the spectral survey technique in obtaining a comprehensive picture of the source chemistry, structure, dynamics and energy balance. For the first time, interpreting the spectrum required some understanding of excitation, structure, dynamics, chemistry and spectroscopy. The interpretation of the data required several years of effort and resulted in at least one Ph.D. Thesis.

In the intervening years, there has been an enormously successful effort in improving all aspects of receiver technology culminating in the construction of the large scale observatories Herschel, ALMA and SOFIA. As a result, near quantum limited ( $\sim 3h\nu$ ) receivers are available to 1 THz and slightly worse  $\sim 10h\nu$  to at least 5.4 THz. Equally importantly, the available IF bandwidth and the LO tuning range have expanded enormously as has the analysis bandwidth of backend spectrometers. As a result, 4 GHz of intermediate frequency (IF) bandwidth is routine and IF bandwidths exceeding 25 GHz are possible below 1.5 THz with SIS mixers. The full scientific impact of the new technology is only beginning to be realized, but the application of modern, high sensitivity receivers to line surveys of warm molecular objects have illuminated the need for a re-thinking of the methodology and tools necessary to reduce and interpret spectroscopic data. The most critical aspect of this re-examination is to realize that a very beautiful and nearly complete spectrum can be obtained in a relatively short observation time and the starting point must necessarily be the global structure of the line forest, not the individual lines.

Submillimeter laboratory spectroscopy is in a similar situation with the source technology developed for astrophysics receivers and commercial communications finding its way into laboratory spectrometers (Brown et al. 2008; Drouin, Maiwald, & Pearson 2005; Petkie et al. 1996) making it possible to collect large continuous pieces of spectra without any tuning elements or stitching together of separate measurements. The result, similar to astronomical spectra, is comprehensive data sets with hundreds of thousands of lines from all the populated states limited only by the dynamic range of the spectrometer which can easily exceed  $10^6$ . Historically, the analysis of microwave spectra relied on measuring a few lines very accurately and fitting them to a molecular Hamiltonian to predict the line frequencies and intensities not measured. Even in complex molecules, only a few hundred lines would be measured and fit with the molecular model. As a result, submillimeter laboratory spectroscopists are poorly prepared to analyze complex spectra in a global way and, like astrophysicists, were trained to look at individual lines, not the structure of the line forest. However, any low resolution infrared spectroscopists will immediately tell you it is the structure of the forest that provides the molecular fingerprint and in most cases, enough information to determine the structure and modes of the molecule. In the rotational band there is often enough information to quickly connect the energy levels involved allowing the spectrum to be analyzed quickly. A complete spectrum is particularly valuable in complex molecules featuring large amplitude motions and strong ro-vibrational interactions.

Adding to the urgency of developing improved methods of spectral analysis is the expectations associated with state-of-the art observation facilities. The development costs of modern observatories ( $\sim \$1B$  for Herschel, ALMA and SOFIA and on the order of  $\$100M$  for VLA/EVLA, CARMA, GBT, CCAT) will be directly reflected in an increase in the competitiveness and required quality of proposals for time and the correspondingly higher expectations of time allocation committees and funding agencies for science results. The urgency is further compounded by the competition for time with simple observations of continuum and high Z objects where reduction of data and publication of results can be significantly simpler. As a result, spectroscopists need to develop approaches to simplify analysis and rapidly extract the enormous information content in a

complex spectrum. A number of significant steps towards the goal of speeding and simplifying the spectral analysis of astrophysical line observations have been made by a number of observers; however, no well organized effort exists that seeks to address the full problem or fully exploit the structures of the line forest. The rest of this paper will be devoted to presenting a vision of how to see the forest and extract the information.

## 2. Spectroscopy: The Science of Patterns

The first step in developing an improved line analysis approach is to realize that molecular spectroscopy, at any wavelength, under any conditions, is ultimately a science of pattern recognition. Spectroscopy in the infrared, optical and ultraviolet has historically generated complete spectra and has relied heavily on pattern recognition for nearly 100 years. In general, a molecular spectrum features three types of branches, P ( $\Delta J = -1$ ), Q ( $\Delta J = 0$ ) and R ( $\Delta J = +1$ ). In general, the *P*- and *R*-branches feature well-defined Boltzmann intensity distributions, while the *Q*-branch is often tightly bunched and unresolved at short wavelength. The branches present depend on the symmetry of the states involved with the rotational band of linear molecules featuring only an *R*-branch. Figure 1 is a simulation of the CO  $v = 1 - 0$  band at 300K showing the *P*- and *R*-branches. In this example, the rotational constant is the difference between upper and lower vibrational states. Since CO is relatively rigid, the difference in rotational constants is quite close to the ground state value. A *Q*-branch will appear when there is some angular momentum not aligned with the molecular axis.

The next consideration is the structure of the molecule, which can be linear and therefore only have one moment of inertia ( $I_b$ ), a symmetric top, with two unique moments of inertia ( $I_a, I_b = I_c$  for a prolate rotor or  $I_c, I_b = I_a$  for an oblate rotor) or an asymmetric top with three unique moments of inertia ( $I_a \neq I_b \neq I_c$ ). Linear molecules feature no projection of the overall angular momentum on the axis of rotation, thus  $K = 0$ , and in the absence of unpaired electrons or bending vibrations (l-type doubling) they feature only one series of lines. The rotational band will be an *R*-branch while vibrational bands in the absence of angular momentum perpendicular to the axis will have *P*- and *R*-branches. Symmetric top molecules feature  $K$  projections but due to symmetry have no strongly allowed transitions between the different  $K$ 's resulting in a series of lines for each  $K$ . The rotational band features only *R*-branches while vibrational bands in the absence of angular momentum perpendicular to the axis will have *P*- and *R*-branches. In linear and symmetric top molecules, the spacing between successive  $J$  lines in the *P*- and *R*-branches is twice the  $B$  ( $\propto 1/I_b$ ), rotational constant. In asymmetric top molecules the situation is complicated by the asymmetry, which lifts the degeneracy of some of the  $K$ 's and provides at least a weak out-of-plane component to the angular momentum through mixing the symmetric top wave functions. In spite of asymmetry splitting, the *P*- and *R*-branch spectrum of asymmetric tops is still very close to having a line in each series every  $B + C$ . This does fail in the limit of very rapid asymmetry splitting, but away from that limit it is a surprisingly good approximation.

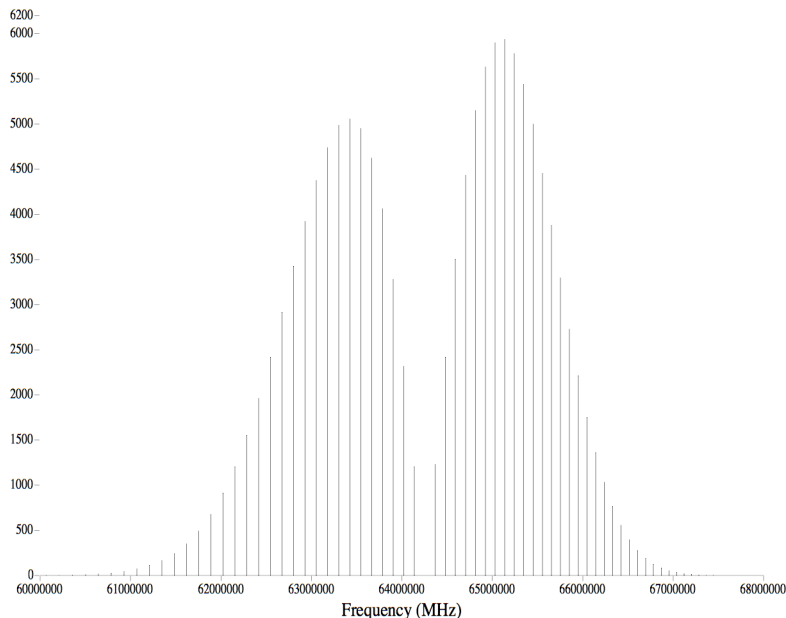


Figure 1. A simulation of the CO  $v=1-0$  band at 300K showing the  $P$ - and  $R$ -branches. Here, the rotational constant is the difference between upper and lower vibrational states.

The next important consideration is the projection of the dipole moment. In linear and symmetric top molecules, the dipole moment is along the axis of rotation and does not change the projection quantum number unless it is between vibrational states with different symmetry. As a result, the spectrum will feature a line every  $2B$ . In asymmetric molecules the spectrum can feature a dipole moment along any axis of the molecule. In near-prolate molecules, which dominate the known interstellar asymmetric tops, a moment along the ‘a’ axis gives rise to a spectrum featuring a line approximately every  $B+C$ . The pure rotation features a strong  $R$ -branch and weak  $Q$ -branches allowed by asymmetry splitting. Vibrational or electronic transitions will have all three branches with the  $P$ - and  $R$ -branches usually being the strongest. Transitions arising from dipole moments along the ‘b’ or ‘c’ axis change the projection so they feature a line every  $B+C$  but with an origin of  $AK^2$ . In near-prolate molecules, the  $P$ -branch is weaker than the  $Q$ -branch, which is weaker than the  $R$ -branch. The  $P$ -branches will generally go through zero frequency. The  $Q$ -branches will be regularly spaced clumps of lines. Figure 2 shows the rotational spectrum of the three different symmetry dipole moments for a molecule using the lowest order constants for ethyl cyanide. The peak line strength has been plotted on the same scale with the same value of the dipole used for all. It is immediately apparent that the ‘b’ and ‘c’ symmetry spectra, dominated by the  $R$ -branches, are at much higher frequency.

Several approaches for visualizing the spectrum have been developed to assist in assignment of the spectrum. The best known is the Loomis-Woods approach (1928) that takes advantage of the fact that  $B+C$  is generally a

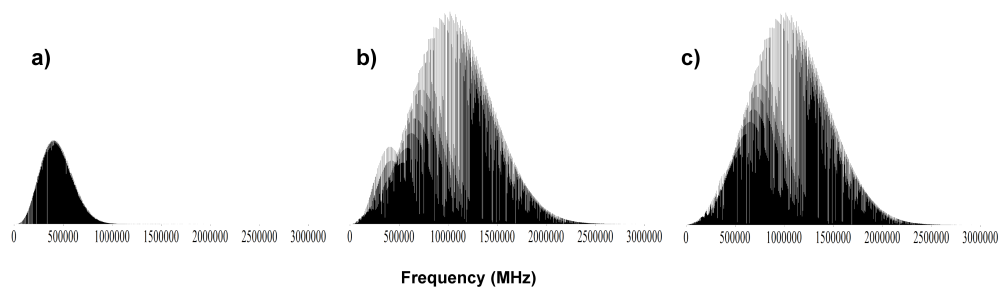


Figure 2. The predicted rotational spectrum using molecular constants of  $\text{CH}_3\text{CH}_2\text{CN}$  and a unit dipole moment. a)  $a$ -type transitions. b)  $b$ -type transitions. c)  $c$ -type transitions. Intensities have been calculated using a unit dipole moment. The striking feature is that the  $b$ - and  $c$ -type spectra are at a much higher frequency than the  $a$ -type spectrum.

good pattern. It is useful to modify the approach to follow a specific branch, with its specific  $B + C$  value, and line up successive  $J$  values on a vertical axis. The obvious astronomical analog is to line up successive transitions of a linear molecule like CO to confirm the assignment and determine the rotational temperature. In this case, one can visually identify satellite lines that slowly curve away from the main branch. This approach is particularly well adapted to identifying vibrational and isotopic species of  $a$ -type spectra in linear, symmetric and asymmetric molecules. Figure 3 shows the oblate paired  $J = K_c$  transitions of ethyl cyanide, the strongest lines in the frequency range, centered on the ground state line from  $J = 71$  to  $J = 76$ . Several series of satellite lines are marked and the strongest three have been identified as the in-plane bend,  $\nu_{13}$ , the first excited torsional state  $\nu_{21}$ , and the out-of-plane bend,  $\nu_{20}$ . Several other lines are marked, some of which are almost certainly the overtone of the torsion and the in-plane bend as well as the combination  $\nu_{21} + \nu_{13}$ . Figure 4 shows the  $aR$ -branch band head for the ground state lined up on  $K = 14$ . Here, a large number of satellite lines can be seen that include the other  $K$ 's of the ground state, band heads of  $\nu_{13}$ ,  $\nu_{21}$  and  $\nu_{20}$  and more satellite lines are observed to high frequency. It must be noted that the rotational quantum number is near 70 and the spread of the vibrational states is still relatively small in spite of strong perturbations and large amplitude internal motions present in the  $\nu_{13}$  and  $\nu_{21}$  spectrum.

### 3. Using Spectral Features

The fundamental molecular constants, even if based purely on low frequency data, provide sufficient information to rapidly assign a spectrum, especially when it is complete. The fundamental constants will provide a good (if not excellent) guess as to the origin of the various branches with low-lying origins. A simple way to assign the spectrum is to start with the lowest  $K$  quantum number of  $P$ - and  $R$ -branches and center a series of spectral windows (as shown in Figures 3 and 4) on the predicted frequencies. Since the lowest  $J$  members of

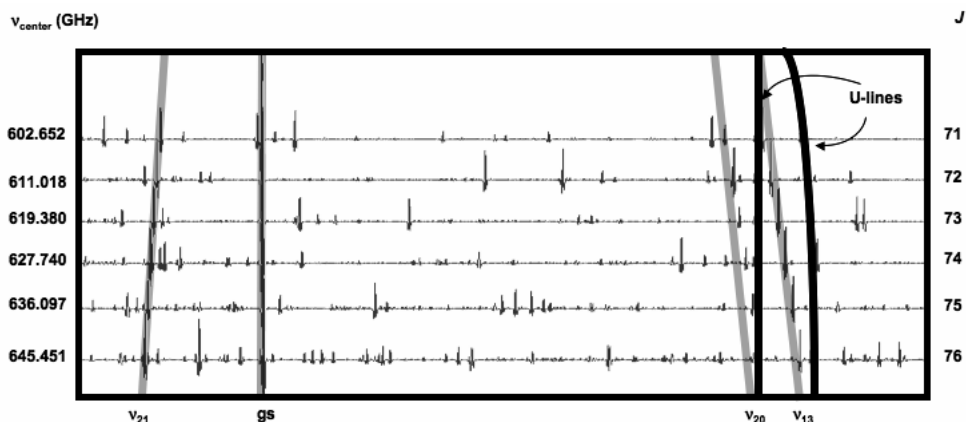


Figure 3. The spectrum of  $\text{CH}_3\text{CH}_2\text{CN}$  in increments of 1.4 GHz, from 603 to 645 GHz, lined up on the oblate paired  $J = K_c$  transitions. The axis on the right indicates the upper rotational level,  $J$ , and the center frequency for each rotational level is shown on the left axis. The grey shaded lines, labeled at the bottom of the figure, indicate from left to right, transitions as follows: The first excited torsional state ( $\nu_{21}$ ), the ground state ( ${}^aR\ K_p = 0$  (gs)), the out-of-plane bend ( $\nu_{20}$ ) and the in-plane bend ( $\nu_{13}$ ). The thick black lines indicate series that are heretofore unidentified. The U-lines are also labeled.

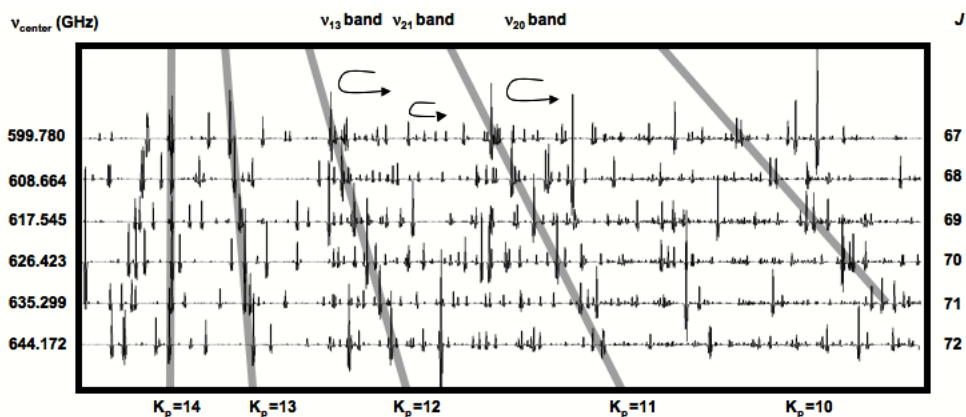


Figure 4. The  ${}^aR$ -branch band head for the ground state of  $\text{CH}_3\text{CH}_2\text{CN}$  in increments of 1.8 GHz, from 599 to 645 GHz, lined up on  $K = 14$ . The axis on the right indicates the upper rotational level,  $J$ , and the center frequency for each rotational level is shown on the left axis. The grey shaded lines, labeled at the bottom of the figure, indicate from left to right,  $K_p = 14, 13, 12, 11$  and  $10$ . The excited state band heads are indicated by curved arrows and are labeled at the top of the figure.

these branches were almost certainly the source of the molecular constants, the agreement at the lowest quantum numbers should be excellent but will likely diverge smoothly with increasing  $J$ . Even when diverging, the series of lines can usually be easily followed as long as there are no perturbations. Once one series is found, the next higher  $K$  should be located and the process repeated. If the molecular constants are what is desired, each series is included in the analysis to improve the predictions of the next. The same process is used in the analysis of laboratory spectra to larger quantum numbers and could be used on astronomical spectra where the laboratory data ends at significantly smaller quantum numbers than observed.

In many cases, a molecular spectrum provides opportunities to connect different energy levels through different paths or combinations. These residual around such a closed loop should be less than the sum of experimental errors or differences to obey conservation of energy. This approach is an extension of the Rydberg-Ritz combination principle applied to molecules (Ritz 1908). The most commonly used experimental technique in rotational spectroscopy is the construction of ‘ladders’ connecting energy levels. The simplest ladder is the  $a$ -type  $R$ -branches and  $Q$ -branches. If the  $Q$ -branches are  $b$ - or  $c$ -type, the entire energy manifold can be connected together, often multiply redundantly. Figure 5 shows an example of an energy ladder for the  $e_0$  substate of  $\text{CH}_2\text{DOH}$ , which features both  $a$ -type and  $b$ -type transitions. In principle, this approach requires no knowledge of quantum mechanics beyond conservation of energy. In practice, it helps to know approximately the molecular constants to reduce the number of possible combinations to try. The infrared spectrum of methanol provides a stunning example of the power of this technique where the torsional band ( $v_t = 1 \leftarrow 0, 2 \leftarrow 1, 3 \leftarrow 2$  and  $2 \leftarrow 0$ ) spectrum was completely and correctly analyzed without any quantum mechanics (Moruzzi et al. 1995). In theory, a complete laboratory spectrum could be reduced to a number of connected sets of energy levels by a computer with no prior knowledge other than a requirement that energy be conserved and that there was at least one set of connected energy levels.

The manual version of combination differences attempts to identify sequences of  $P$ - or  $R$ -branch lines and connect them with the more obvious  $Q$ -branch lines. Frequency differences between successive members of  $Q$ -branches provide the change in frequency relative to two  $P$ - or  $R$ -branches. Once a series with the same residual is located then it can immediately be concluded they are connected. Ladders, like those shown in Figure 5, will immediately confirm the conclusion.

The other useful tool for spectroscopists is a graphical display of residuals in their analysis. A simple method is to plot with a circle with a diameter proportional to the frequency residual on a graph with  $J$  on the  $x$ -axis and  $K$  on the  $y$ -axis for the upper states of the transitions involved. Isolated large circles indicate poor measurements or assignment errors while series of larger circles indicate systematic problems with the model. A similar approach also is used with intensity measurements to find blended lines and other problems. Such an approach would be equally useful in astronomical spectra when comparing a global data set to a model. In this case the larger circles in regions where the agreement was pretty good would be indications of blended or overlapping

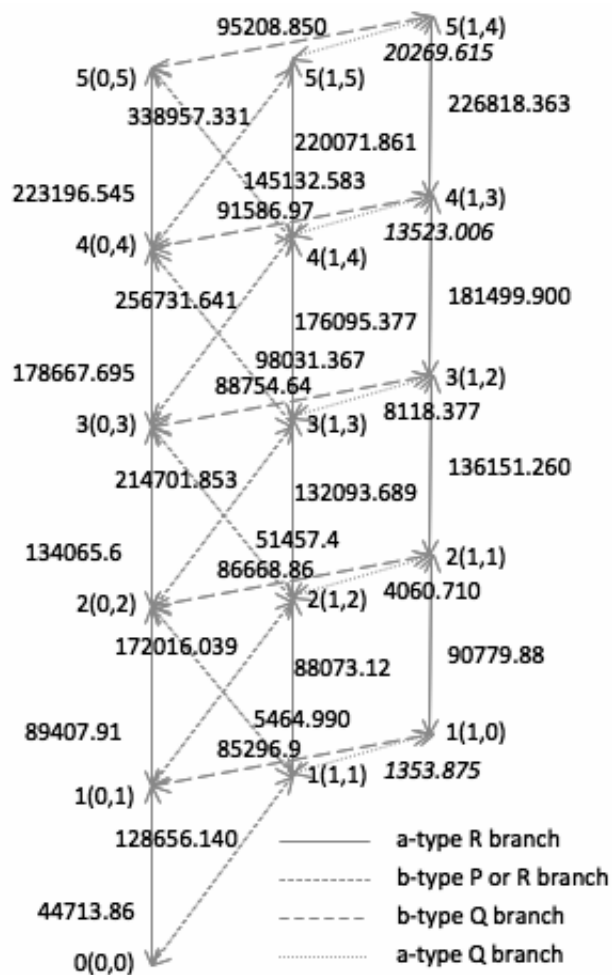


Figure 5. An energy ladder for the  $e_0$  substate of  $\text{CH}_2\text{DOH}$ , which features both  $a$ -type and  $b$ -type transitions. All frequencies are measured, except those in italics, which are calculated.



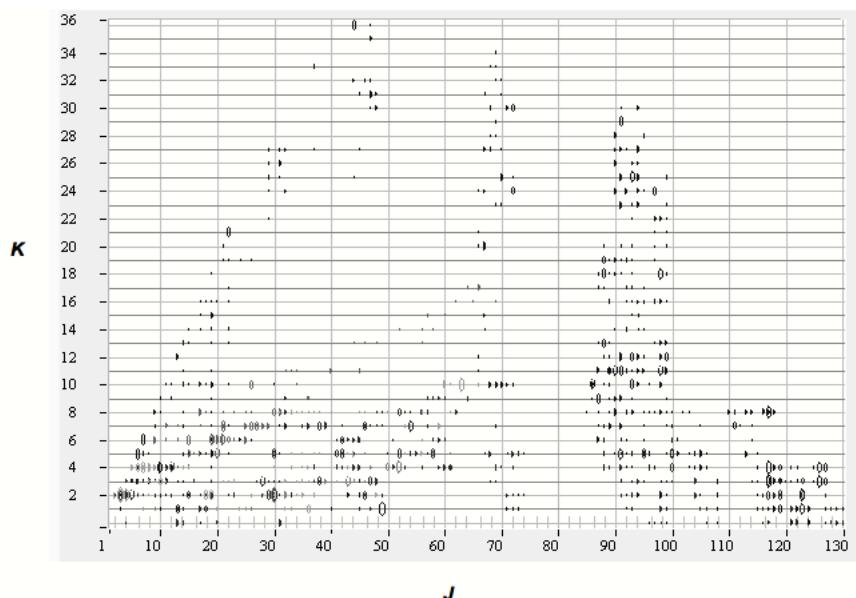


Figure 6. A residual plot of 3880 fitted lines of the ground state of  $\text{CH}_3\text{CH}_2\text{CN}$ . The rotational quantum number,  $J$ , is plotted on the abscissa and  $K$  is on the ordinate. The ratio of the residual:uncertainty is indicated by circles.

lines that probably need to be temporarily excluded from the analysis until all was assigned. Figure 6 is a plot of residuals from a recent analysis of the ground state of  $\text{CH}_3\text{CH}_2\text{CN}$ , using Brian Drouin's FIT program, available on the JPL website, <http://www.spec.jpl.nasa.gov>. With this program, one can plot the  $P$ -,  $Q$ - and  $R$ -branches, as well as  $a$ -,  $b$ - and  $c$ -type transitions individually, allowing easy isolation of problem areas.

Isotopologues, like vibrational states, can often be easily identified visually in a spectrum using the parent species and some intuition about how the molecular constants might change. In general the ground state main isotope lines will have smoothly diverging satellites to lower frequency for heavier isotopes and higher frequency for lighter isotopes. These satellites will have a relative intensity determined by the isotopic abundance and vibrational partition function, if excited. This approach has been very successful in identifying the isotopologues and vibrational states of linear molecules such as  $\text{HC}_3\text{N}$  (Pardo et al. 2004).

#### 4. Astrophysical Spectral Tools

Astrophysical spectra obviously present some rather different challenges than laboratory spectra, however the basic techniques used to handle laboratory spectra still need to be applied. For astrophysical spectra, the underlying goal is to derive as much as possible about the source structure and the underlying physical and chemical conditions. The obvious first step is to begin with the simplest

model possible invoking no or very simple (i.e. spherical) structure and thermal equilibrium. The first requirement for spectral analysis is to be able to directly, visually compare the modeled spectrum to the observed spectrum. In this case, like in making laboratory assignments, two things are extremely valuable. First, it is highly desirable to be able to display all the transitions in a branch simultaneously, exactly as is done for making quantum number assignments. Second it is necessary to display the difference between the observed and calculated spectrum. The series methodology will quickly show systematic residuals in the  $J$  quantum number and therefore energy, as well as systematic deviations in line shapes that will simplify determination of the underlying model limitation. The branch approach is obviously the only way to assess the quality of a spectroscopic or excitation model for a linear molecule, but it can be equally effective on small subsets of complex molecular data. In this case, selecting a few branches in  $K$  with fixed  $J$  to give an exponential ( $\sim AK^2$ ) energy series and  $P$ - or  $R$ -branch series, which give energy increasing with  $(B+C)J(J+1)$ , will provide a stringent test of excitation models. The same technique is used to quickly refine molecular constants. Several tools for laboratory and astronomical spectra allow the spectrum to be compared with a model, however none of the available astrophysical spectral tools allow for entire branches to be parsed for simultaneous display and analysis in this way.

The next step is to determine how to rapidly deal with the rest of the spectrum. Once the presence of a molecule is confirmed in a source or source component, then it must immediately be assumed that all the transitions of that species and all its isopologues are present. At the source component level it should also be assumed that the isotope ratio is the only adjustable parameter. From this point, being able to display statistics on the residuals is necessary, as is the ability to exclude obviously blended outliers from the analysis, preferably with the point and click technique. This makes the main exercise determining the best model for the source or source component on a molecule-by-molecule basis. Once the easily identifiable molecules have been evaluated, a cleanup of the spectrum is then possible. Here a nearly complete simulation can be applied and the difference taken, properly accounting for the blended features. If all is complete the residual will be the true continuum, however it is likely that there are many features not in catalogs or deviating from catalogs. At this point, the spectrum might as well be a laboratory spectrum because all the steps required for making further progress involve determining the carriers of the lines.

The shape of residuals will become a first derivative when predicted frequencies start to deviate from the observed frequency. Once again, displaying residuals from branches will make the molecular model shortcomings very obvious. The true rest frequencies can easily be exported and fit with a molecular model resulting in a better catalog exactly as is done in the laboratory. U-lines from molecules not cataloged or identified will just be emission features subject to all the pattern recognition tricks used by spectroscopists. Patterns like  $Q$ -branches or  $R$ -branch band heads might be identifiable. Additionally the spectrum might be suitable for combination difference methods at this point potentially resulting in lines that are connected.

## 5. Astronomical Tool Requirements

The first requirement of any line assignment tool is to be able to display a spectrum generated by a model and a catalog. The second requirement is to be able to sort a catalog by branch and display multiple windows showing the branch as observed and modeled. The preferred method is vertical with the highest quantum number (and energy) at the top independent of frequency or wavelength. The frequency width of the window should be definable by the user. The third requirement is to be able to take differences between observed and calculated and display the residuals on the same plots as the data and models. The fourth requirement is to associate all transitions of a molecule and its isotopologues with the identified features with the only adjustable parameters being the isotopic abundance. The fifth requirement is to be able to view statistics on the residuals in a global way with the ability to click on a residual to see the spectrum and to be able to exclude/include from the analysis with one click. A filter for energy and intensity would be particularly useful to avoid low signal-to-noise lines in the early phases of analysis. Ideally, two colors would be used to show included and excluded data. The sixth requirement is to be able model blended lines from different components and molecules and generate a global difference while displaying the actual and the model.

A number of other features are nice to have. These include the ability to export new frequencies with quantum numbers associated from a catalog so that the catalog can be updated off line, exporting graphics from the analysis of a series of lines for publications and automatic management of the list of identified lines both included and excluded. Several U-line tools would be nice to have as well. First is a brute force combination difference calculation, preferably with some limits on molecular parameters. Ideally this would produce a ladder display and relative energy levels like Figure 5 so quantum number assignments could be made. Second is a linear series finder for all sequences that can reasonably be modeled by a rotational constant and a distortion constant. The hardest of these features to implement is actually the display capability.

Ultimately it will be desirable to actually fit the source parameters in the source model to the observed spectrum; however, there is a significant prospect that this is not a numerically well conditioned problem making some user intervention in the parameters to fit essential. Regardless the tools described previously will be essential in understanding the quality and problems with such a fit.

## 6. Conclusions

Molecular astrophysics is a spectroscopic science and will need to borrow the best available spectroscopic analysis tools to handle the volume and quality of spectra that will arrive from new observatories coming on line. The information content in a nearly complete spectrum is truly enormous and a complete analysis will provide a previously impossible understanding of the physical and chemical conditions. The current limitations in molecular data will be annoying but the astronomical spectra will probably be of sufficient quality to help improve the molecular data. The key to analyzing a complete spectrum efficiently is a tool

with a simple visual interface that automates sorting and displaying of spectral branches. This, coupled with some automatic book keeping and a point and click interface, will make the problem tractable, making the hard part of the task getting the physics of the source model correct. Since this was ultimately the point of making the observation, the problem is tractable if the tools can be made and optimized to extract the physics of both the interstellar medium and the molecules it contains.

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