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NO_2 Heterodyne Frequency Measurements With a Tunable Diode Laser, a CO Laser Transfer Oscillator, and CO_2 Laser Standards

L.R. Zink
M. Vanek
J.S. Wells

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L.R. Zink
M. Vanek
J.S. Wells

Time and Frequency Division
Center for Basic Standards
National Measurement Laboratory
National Bureau of Standards
Boulder, Colorado 80303-3328

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NO_2 HETERODYNE FREQUENCY MEASUREMENTS WITH A TUNABLE DIODE LASER, A CO LASER TRANSFER OSCILLATOR, AND CO_2 LASER STANDARDS

L. R. Zink, M. Vanek, and J. S. Wells
Time and Frequency Division
National Bureau of Standards
Boulder, Colorado 80303

Heterodyne frequency measurements have been made on selected groups of nitrogen dioxide rovibronic transitions between 1580.8 and 1650.7 cm^{-1}. The groups are separated by 7 to 10 cm^{-1}, and the intent is to provide a limited (and interim) calibration table for the region. In addition to a table of measured frequencies in this region, we have included figures of spectra in the vicinity of the measured lines to provide a map for identifying the transitions measured.

Key words: calibration frequencies at 6.2 μm, heterodyne frequency measurements, nitrogen dioxide spectra.

INTRODUCTION

A program was established several years ago to provide reference frequencies for the use of spectroscopists for the calibration of spectra. The objectives at the initiation of the program (with particular emphasis on the use of tunable diode lasers) were to select molecules meeting certain criterion such as

1. Absorption type standards
2. A marker every 0.5 cm^{-1} (to match TDL mode tunability)
3. A convenient molecule (one easily handled)
4. A simple Hamiltonian
5. Line positions should be good to $\pm$ 3 MHz.

The last criterion requires heterodyne frequency measurements and that has been a major task along with the selection and fitting of the best available measurements from the literature. Some of the better candidates were carbonyl sulfide (OCS) and nitrous oxide (N_2O). A large body of work has been completed on OCS [1-5]. OCS has a fairly high abundance of isotopic species which help provide a definite signature (spectroscopic pattern) in the vicinity of a particular calibration feature. A 6600 entry OCS atlas [6], which gives the transition wavenumber, the transition frequency, the isotopic species, the transition band, the J-value for the transition, the lower state energy level and the intensity in a normal mixture for each line is available. The coverage afforded by OCS shown in figure 1 below.

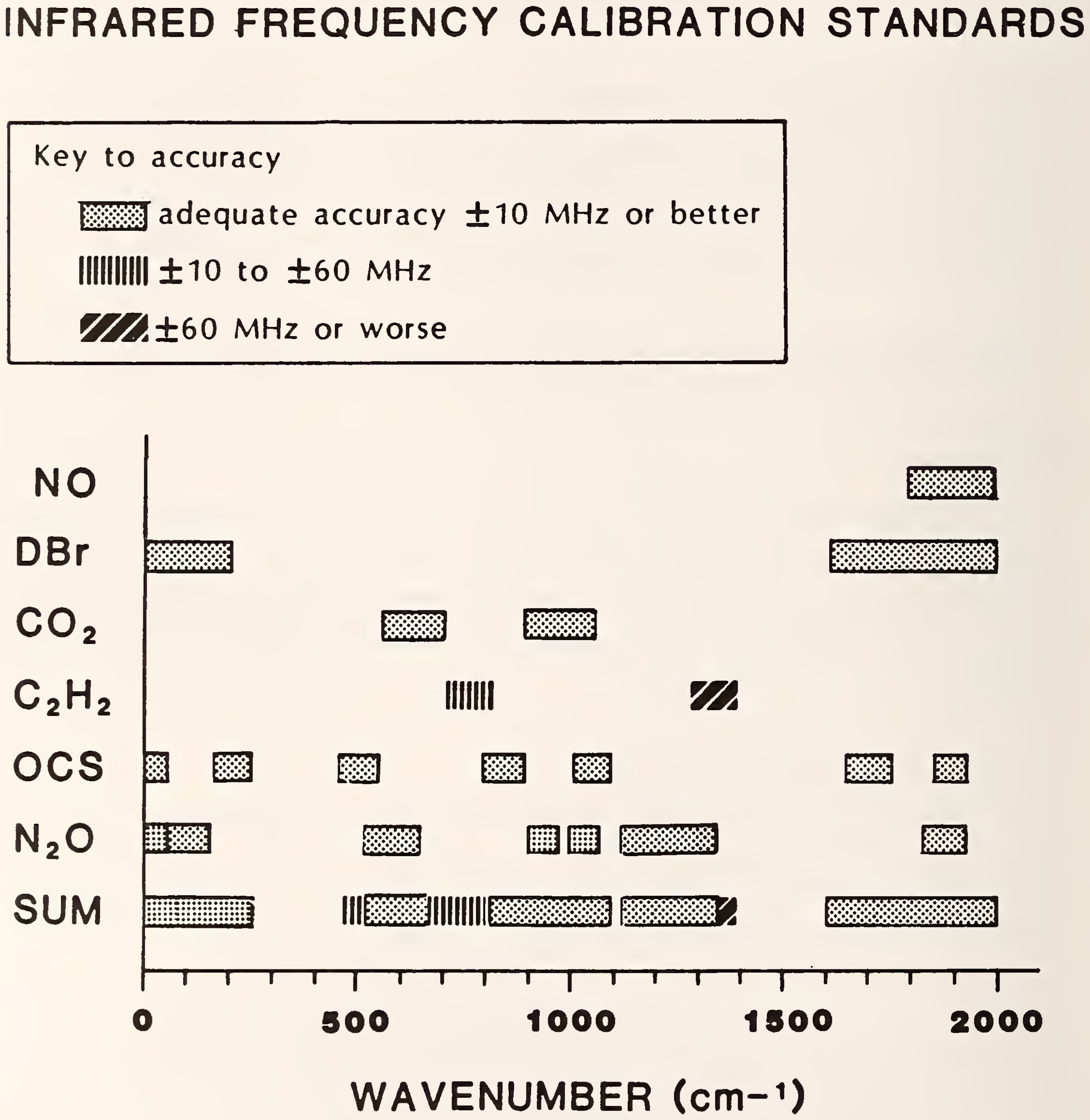

Fig. 1. Accuracy of frequency calibration tables for selected molecules in the 0-2000 cm^{-1} region.

Another prominent molecule in figure 1 is nitrous oxide and a large amount of work has been completed on this molecule [7-9]. An interim calibration table exists for the 8 μm region but more work is needed and has been planned for the future.

While these two molecules satisfy the five criteria above, there are regions of the spectrum where these two molecules leave gaps in the coverage. For example DBr covers some of the gap at 1800 cm^{-1} [10], but it does not meet criterion #2. In DBr the spacing between calibration points is about 10 cm^{-1} and then only two markers appear (separated by about 0.2 cm^{-1}) before another 10 cm^{-1} gap. Nonetheless, users have reported this to be quite useful.

The emphasis of the current work is to address the gap at 1500 cm^{-1} in figure 1 and to provide references similar to that afforded by DBr. In particular, we chose to provide markers every 7 to 10 cm^{-1} and also provide a tunable diode laser (TDL) spectrum of the other lines in the vicinity to serve as a signature.

Nitrogen dioxide does not meet the fourth criterion in that it is an asymmetric rotor with spin split states and its Hamiltonian requires about 25 constants. Further, more than 3000 lines exist in the current Air Force Geophysical Labs (AFGL) tables, and only one or two percent of these are candidates for a heterodyne frequency measurement. Thus, our approach will serve two purposes. First, it will provide frequency measurement based calibration points with strongly absorbing lines in the gap region and serve as an interim calibration table. Second, our past experience with data from different techniques indicates that while Fourier transform spectroscopic (FTS) data are quite satisfactory for some requirements, there can sometimes be a small calibration error. We suggest that the results from our study either will be useful for the calibration of NO_2 in a FTS study, or serve as an independent check of that work. We think that a measurement every 10 cm^{-1} or so will be satisfactory for this purpose. Figure 2 on the next page shows a compressed FTS spectrum of nitrogen dioxide (the intense lines outside the familiar P and R branch structure are H_2O lines) and the position of CO laser lines which were used to make the heterodyne frequency measurements.

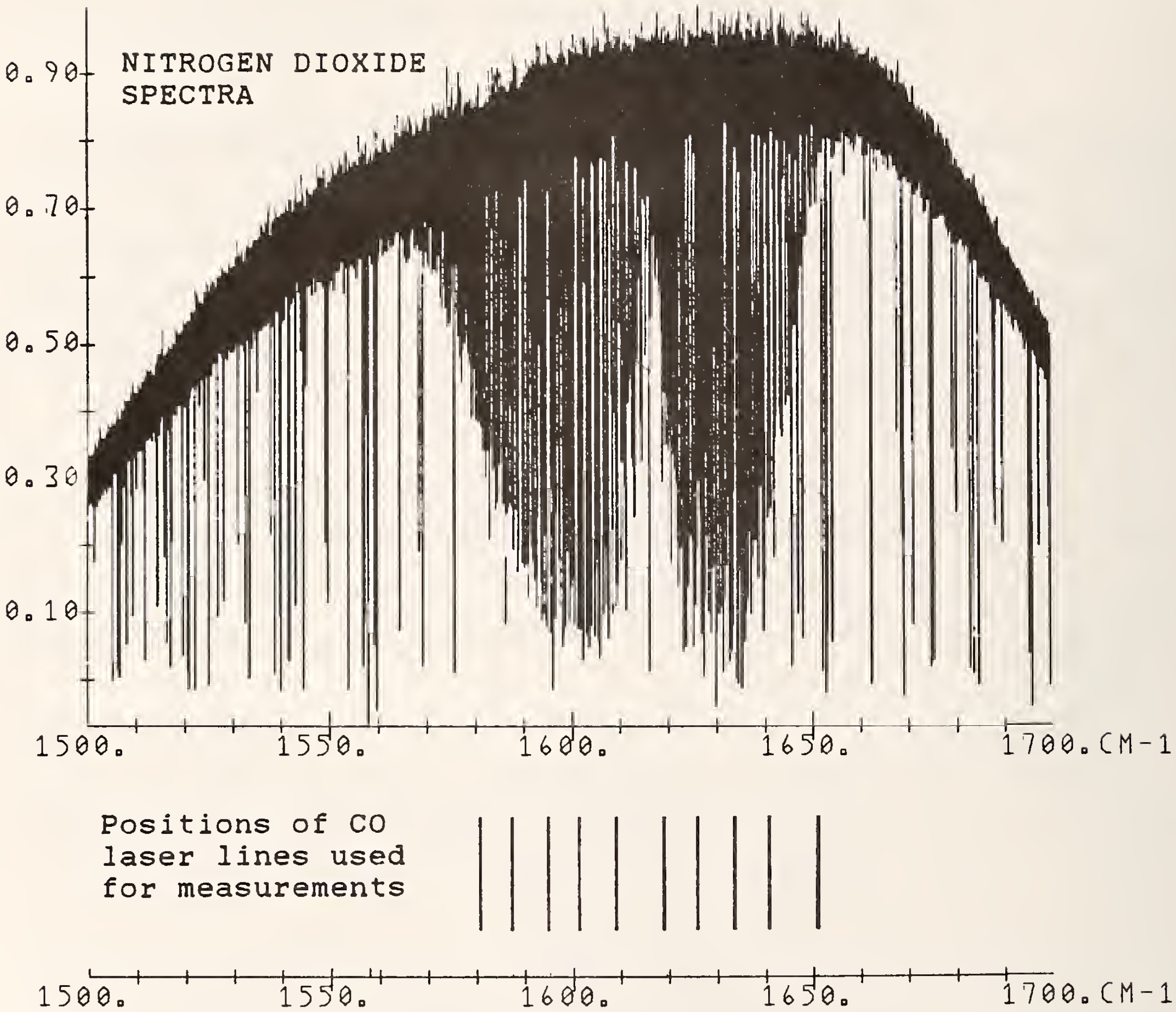

Fig. 2. Overlap between nitrogen dioxide spectra and selected CO laser lines.

EXPERIMENTAL TECHNIQUES

The measurement technique used for these measurements requires the use of a CO laser to act as a transfer oscillator. Its role is to transfer the frequency synthesized from CO_2 laser frequency standards to the frequency of the molecular feature of interest. Each of the frequency measurements of a nitrogen dioxide transition listed in Table I is a combination of two nearly simultaneous measurements. The first one is a determination of the frequency of a CO laser transition relative to the CO_2 laser standards. By the use of two stabilized CO_2 lasers, a metal-insulator-metal diode, and a microwave oscillator, we generate a frequency, ν_s, given by

$$\nu_s = \ell\nu_1 + m\nu_2 + n\nu_M, \tag{1}$$

where ν_1 and ν_2 are the frequencies of the CO_2 laser standards, and ν_M is a microwave frequency. The quantities ℓ, m, and n are integers which are allowed both positive and negative values. For these experiments, the value ℓ was always 3, and m was -1. The microwave frequencies are listed in Table I, and the n value may be inferred since an X-band klystron was used. An rf beat note ν_{B1} (the difference between the frequency ν_s and the CO laser frequency ν_{CO}) was taken from the MIM diode, amplified and displayed on an rf spectrum analyzer where its center frequency was marked by an rf synthesizer. The frequency of the transfer oscillator was then

$$\nu_{CO} = \nu_{xfer} = \nu_s \pm \nu_{B1}. \tag{2}$$

The second part of the measurement was a difference frequency measurement between the CO laser transfer oscillator and a tunable diode laser whose frequency was locked to the NO_2 absorption feature of interest. This difference frequency beat note ν_{B2} had its center frequency marked by a marker oscillator whose frequency was counted and the frequency of the NO_2 transition was then

$$\nu_{NO_2} = \nu_{xfer} + \nu_{B2}. \tag{3}$$

The measurements are given in Table I, which gives the CO_2 transitions and the microwave frequency (in some cases a second harmonic is the value shown) involved in each measurement, the frequency of the NO_2 transition along with our estimated uncertainty and finally the quantum numbers assigned are those taken from the AFGL tapes.

Tunable diode laser spectra in the vicinity of the each of the CO laser lines used for the measurements are given in the appendix. These were observed at a pressure of 113 Pa and a path length of 1.7 m. Differences between the calculated frequency values in the AFGL tables and our heterodyne measurements range from a few megahertz up to hundreds of megahertz for some transitions.

For the heterodyne frequency measurements in Table I, the path length was also 1.7 m. The pressure was reduced so that for each transition measured the absorption was in the linear regime. Typically, these pressures were in the 50 to 100 Pa range.

CONCLUSION

Since NO_2 is an asymmetric rotor with spin split states and the majority of the more than 3000 lines given in the AFGL tables are not close enough to CO laser lines to permit heterodyne frequency measurements on them, we believe the better approach is to measure only enough lines for the calibration and leave the detailed spectra for a FTS study. This will permit us to use our resources to look at some N_2O hot band features in the 1600 cm^{-1} region. This represents something of a trade-off; a long path cell will be required for the N_2O spectra, but a marker every 0.5 cm^{-1} should be available. The greater absorption afforded by NO_2 will permit its use by all, but until a more detailed study with a high resolution FTS instrument such as that accessible to the JPL is complete, only a limited number of reference points will be available.

ACKNOWLEDGMENT

We would like to thank Dr. J. Burkholder for taking some preliminary FTS data on the local Bomen instrument of NOAA at our request. We also thank Dr. D. A. Jennings for the use of his frequency synthesis programs. We are grateful to Dr. A. Maki for his continuing cooperation and advice regarding our frequency standards program. This work was supported in part by the Upper Atmospheric Research Office of NASA.

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APPENDIX

This appendix contains ten figures of nitrogen dioxide spectra in the vicinity of the CO laser lines which were used in the heterodyne frequency measurements. We have included them because we found the lower resolution AFGL data to be somewhat difficult to reconcile with the higher resolution TDL spectra. In some cases, this may have been due to less accurate line position data (calculated values in error due to poor constants) for example. In other cases, perhaps it was simply a resolution problem. In any case, we have found it preferable to have a signature at the resolution attainable by the instrument at hand and, given the current status of the data (numerical data not available at accuracy reported here), we feel spectral samples are the interim answer.

Most of the spectra were recorded at a pressure of 113 Pa (0.85 torr) in an absorption cell 1.7 m long. For the more intense regions of the spectra, the pressure was reduced for the duration of a particular heterodyne frequency measurement. The pressure was in general reduced to the linear absorption regime and this afforded lines to be resolved that do not appear so in the spectra shown (see Figs. A-4 and A-8 for example).

Three sets of numbers (given in cm^{-1}) appear in the figures. The heterodyne frequency measurements are given in square brackets with the uncertainty in the last digits for the measurement given in parentheses within the bracket, i.e., [1580.73450(33)]. These numbers are positioned on the page so that the decimal point lines up above the center of the line that was measured, and in most cases they are placed above the spectra. A second set of numbers comes from newly taken FTS data; in this case only the numbers to the right of the decimal point are listed, except for a few obvious instances. The FTS numbers are meant to supplement the frequency measurements and are listed in parentheses. In some instances, these have been corrected where a disagreement with heterodyne measurement seems to warrant it. Uncertainties are not given for these measurements, but we believe that the values we list are good to within 0.0005 cm^{-1}. The third set of numbers was taken from the AFGL tables; these are positioned near the NO_2 spectra baseline and consist of only the three significant digits listed in the tables.

Fringes from a 7.5 cm germanium etalon were recorded below the NO_2 spectra. Since the etalon was not temperature stabilized, these should not be relied upon for interpolation. We have used them only for line identification purposes.

Table I. Heterodyne Frequency Measurements on Nitrogen Dioxide with the Aid of a CO Laser Transfer Oscillator.

Synthesis Scheme			CO laser	NO_2 Transition							
CO_2 trans. for $3v_1$	CO_2 trans. for $-v_2$	MW freq. MHz	trans. $P_{v"}(J")$	Frequency[a] MHz	Quantum Numbers J'	K_a'	Kc'	J"	Ka"	Kc"	S-St
I636[b] P 22	II626 P 32	24 200	$P_{17}(15)$	49488134.4(90)	49	0	49	48	0	48	±
I636 P 18	II626 P 10	11 600	$P_{18}(11)$	49190236.8(70)	37	4	33	36	4	32	−
				49190087.6(70)	37	4	33	36	4	32	+
I636 P 24	II626 P 20	7 500	$P_{18}(13)$	48973387.0(70)	22	1	22	21	1	21	−
				48971356.7(90)	24	3	22	23	3	21	−
				48971185.8(90)	24	3	22	23	3	21	+
				48967881.8(70)	26	4	23	25	4	22	−
				48967623.7(70)	26	4	23	25	4	22	+
I636 P 24	II626 P 12	0	$P_{18}(15)$	48758099.8(70)	19	5	14	18	5	13	−
				48752991.2(70)	22	6	17	21	6	16	−
				48749412.1(70)	14	3	12	13	3	11	−
				48748404.3(70)	16	4	13	15	4	12	+
I636 P 18	II626 R 16	−11 000	$P_{19}(10)$	48555023.3(70)	21	8	13	20	8	12	+
				48551625.6(70)	10	5	6	9	5	5	−
				48550704.5(100)	13	6	7	12	6	6	−
I636 P 30	II626 P 10	22 000	$P_{19}(13)$	48231914.6(50)	6	6	1	6	6	0	−
				48231205.7(50)	6	3	4	7	3	5	+
				48229667.9(50)	7	6	1	7	6	2	+
				48229360.7(50)	8	6	3	8	6	2	−
				48228499.0(100)	8	6	3	8	6	2	+
				48227145.8(100)	9	6	3	9	6	4	+
				48226313.4(50)	10	6	5	10	6	4	−
				48225637.6(50)	10	6	5	10	6	4	+
I636 P 22	II626 R 28	−10 000	$P_{20}(8)$	48023593.6(70)	12	4	9	13	4	10	+
				48023320.2(70)	12	4	9	13	4	10	−
				48020088.1(70)	15	2	13	16	2	14	±
				48019774.1(70)	16	1	16	17	1	17	±
				48018936.9(70)	10	5	6	11	5	7	+
				48018294.6(70)	10	5	6	11	5	7	−
I636 P 36	II626 P 14	10 000	$P_{20}(10)$	47817726.7(100)	21	3	18	22	3	19	±
I636 P 28	II626 R 24	− 8 800	$P_{20}(12)$	47605993.0(100)	16	8	9	17	8	10	+
				47605568.1(90)	16	8	9	17	8	10	−
I636 P 30	II626 R 26	−20 200	$P_{20}(14)$	47394158.7(100)	not assigned						
				47389778.9(100)[c]	32	5	28	33	5	29	±
				47389452.2(100)	20	9	12	21	9	13	+
				47389228.2(100)	20	9	12	21	9	13	−

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a) The uncertainty in the last digits is given in parentheses.

b) The notation 626 and 636 refers to the $^{12}C^{16}O_2$ and $^{13}C^{16}O^{2}$ isotopic lasers respectively.

c) We estimate that we locate the maximum point of this line to within 10 MHz, however the line is blended, possibly with a water vapor line.

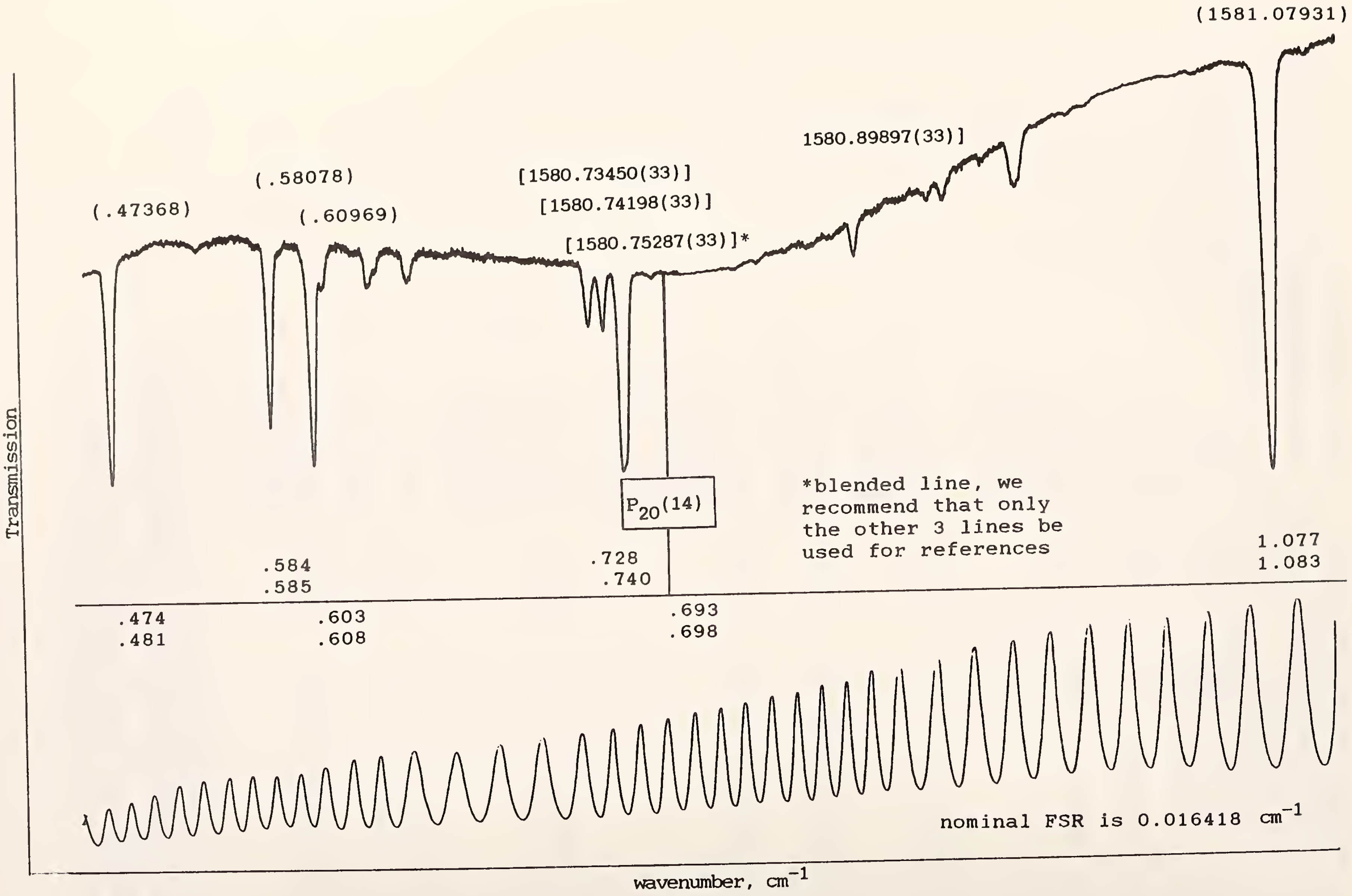

Fig. A-1. Nitrogen dioxide spectra in the vicintiy of the $P_{20}(14)$ CO laser line at 1580.77335 cm^{-1}.

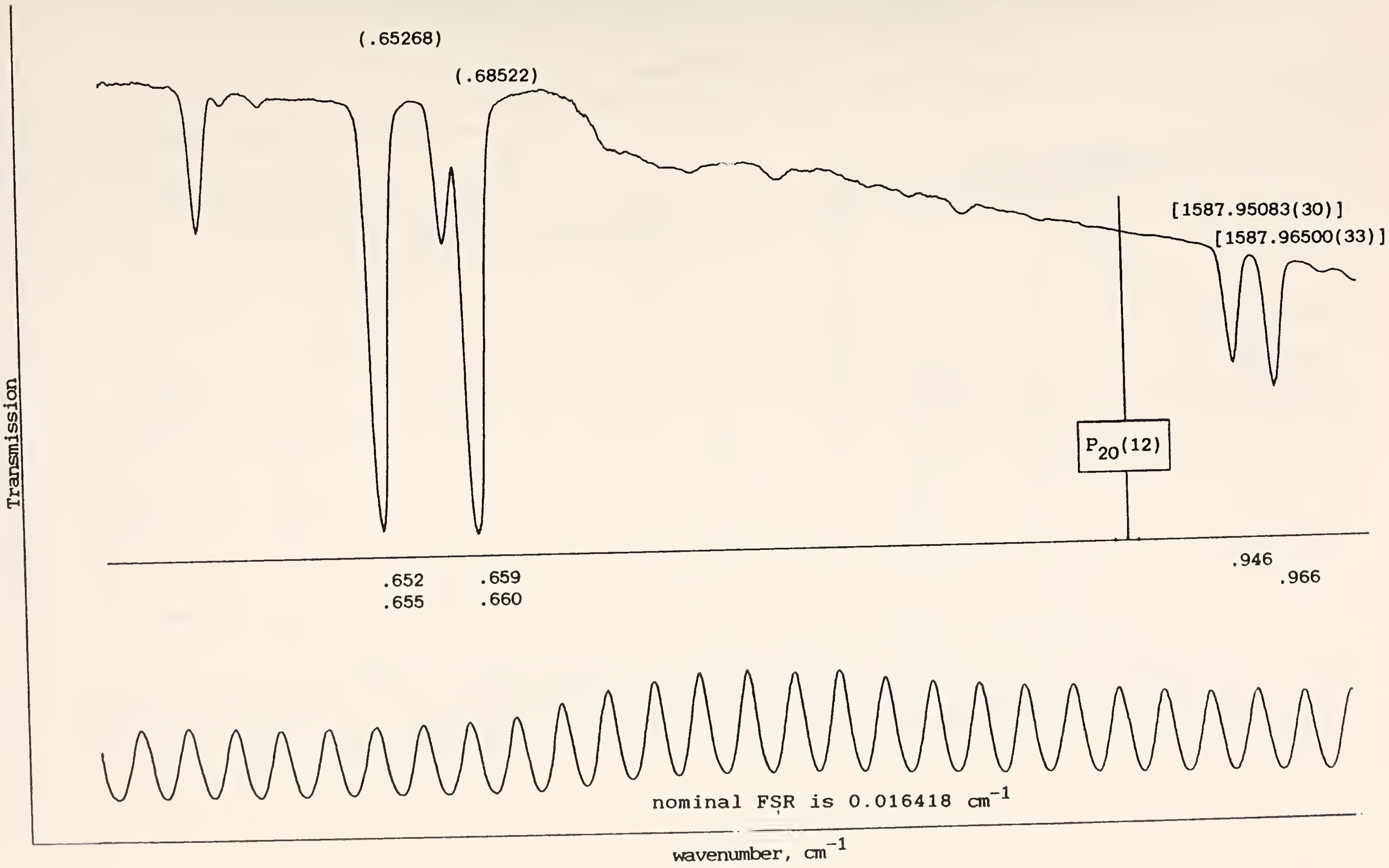

Fig. A-2. Nitrogen dioxide spectra in the vicinity of the $P_{20}(12)$ CO laser line at 1587.91265 cm^{-1}.

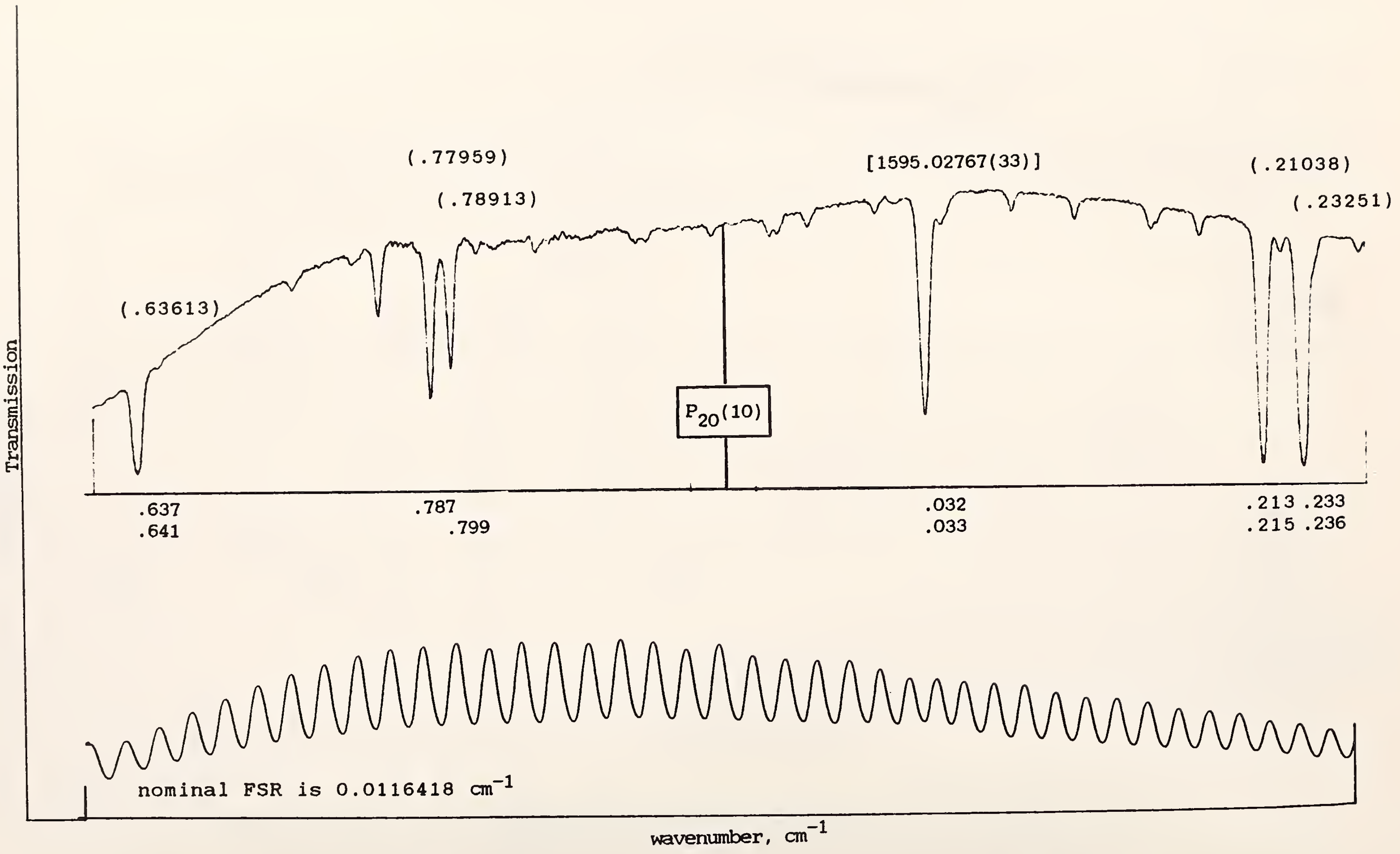

Fig. A-3. Nitrogen dioxide spectra in the vicinity of the $P_{20}(10)$ CO laser line at 1594.91939 cm^{-1}.

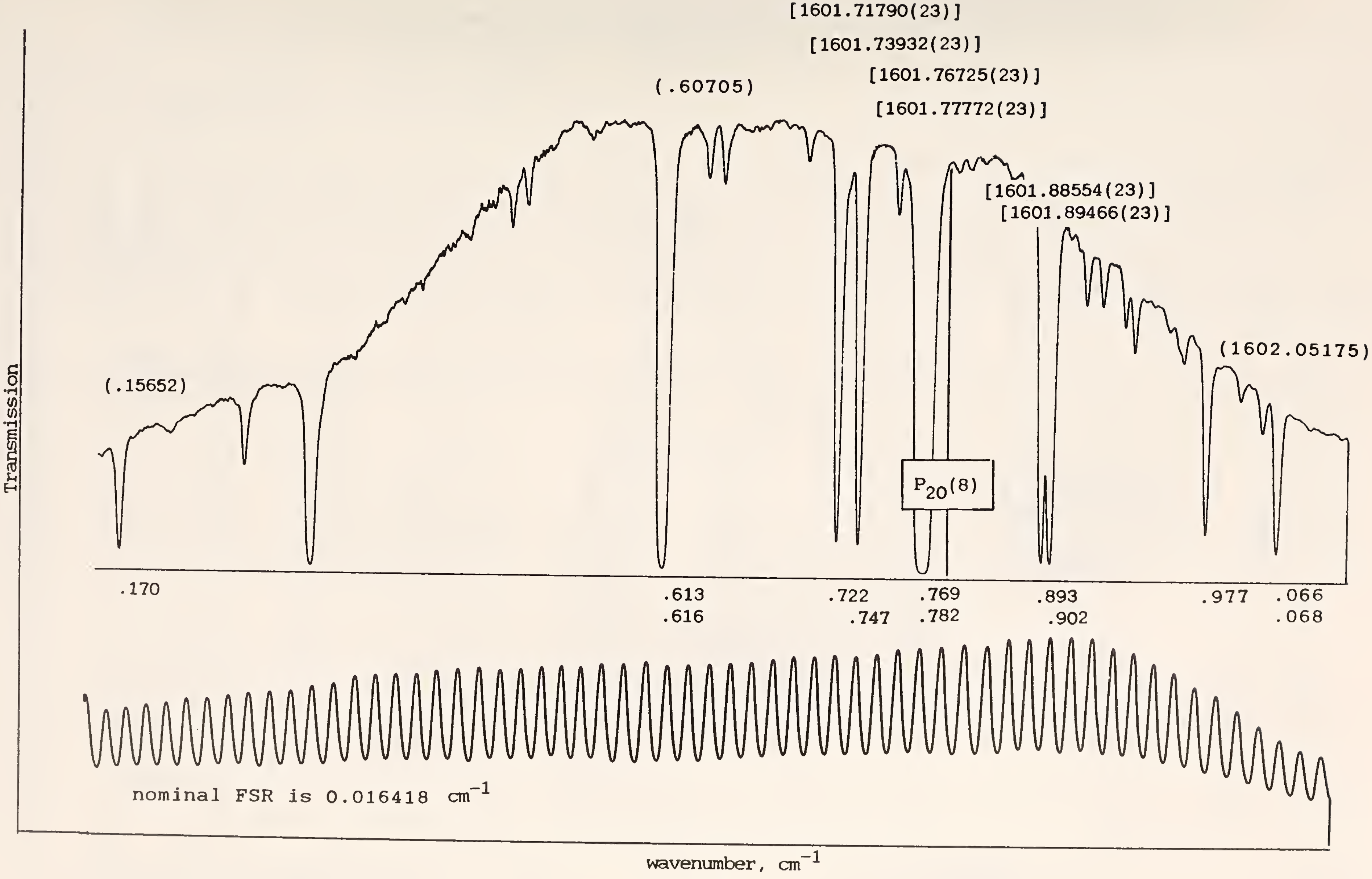

Fig. A-4. Nitrogen dioxide spectra in the vicinity of the $P_{20}(8)$ CO laser line at 1601.79240 cm^{-1}.

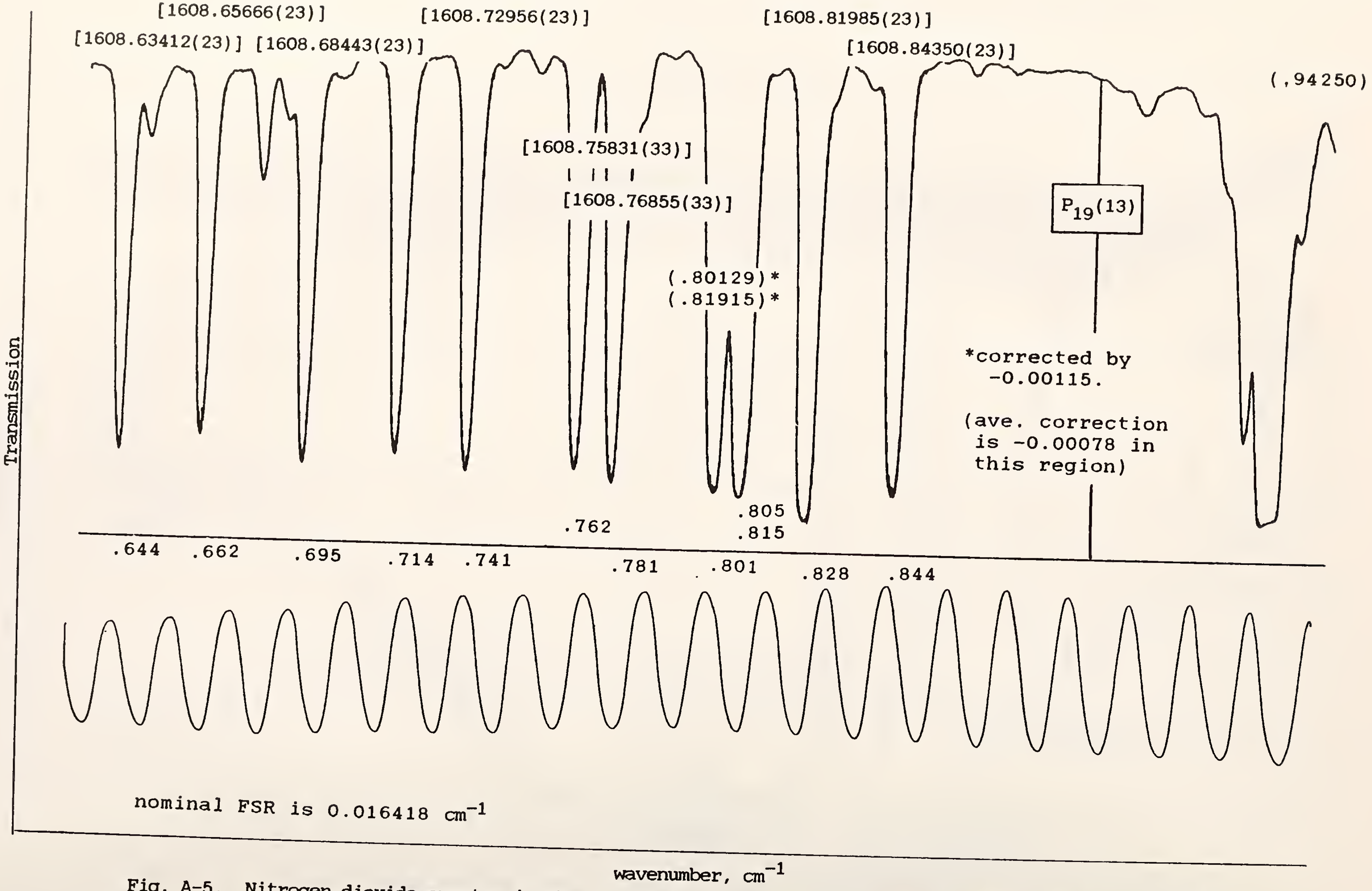

Fig. A-5. Nitrogen dioxide spectra in the vicinity of the $P_{19}(13)$ CO laser line at 1608.89765 cm^{-1}.

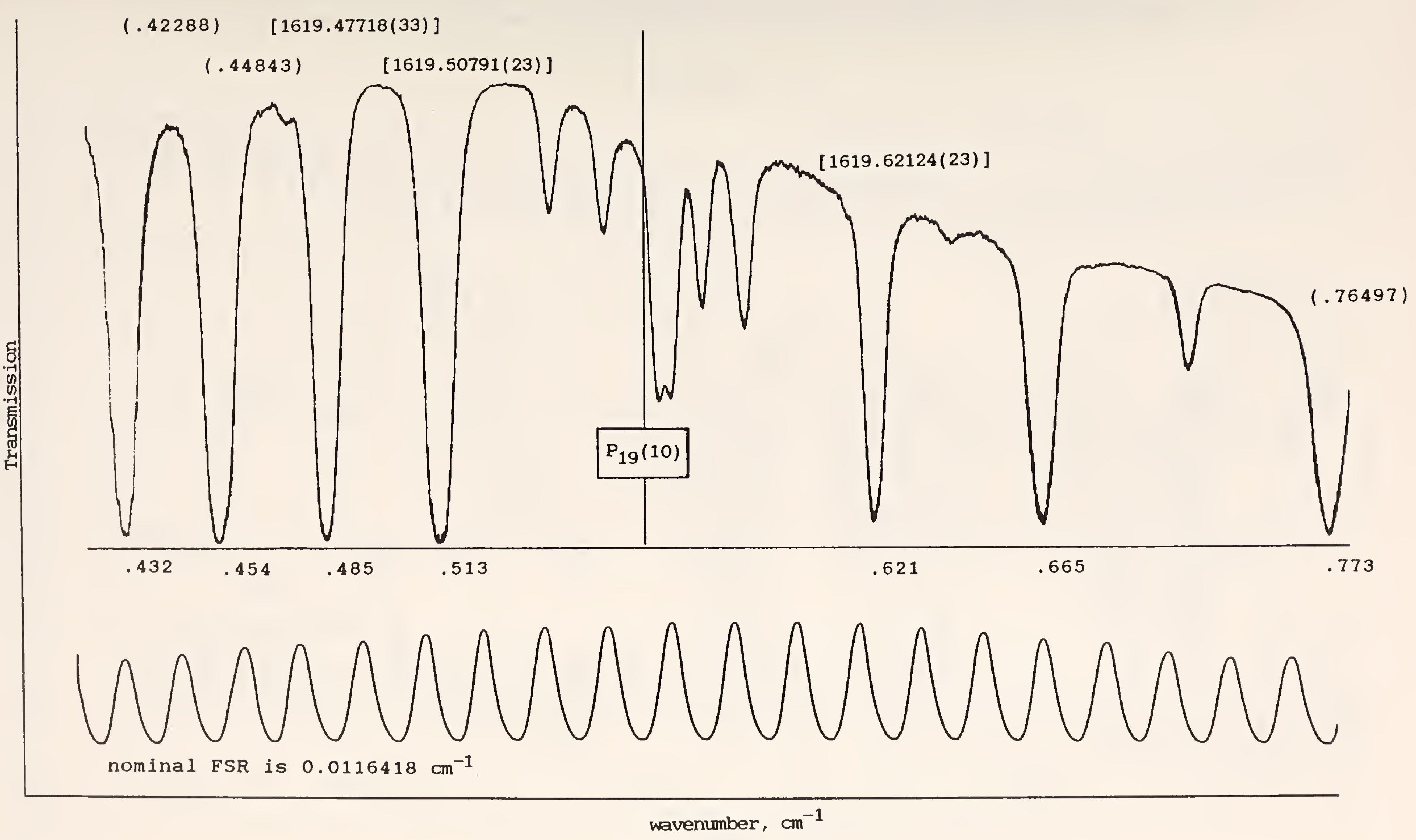

Fig. A-6. Nitrogen dioxide spectra in the vicinity of the $P_{19}(10)$ CO laser line at 1619.56237 cm^{-1}.

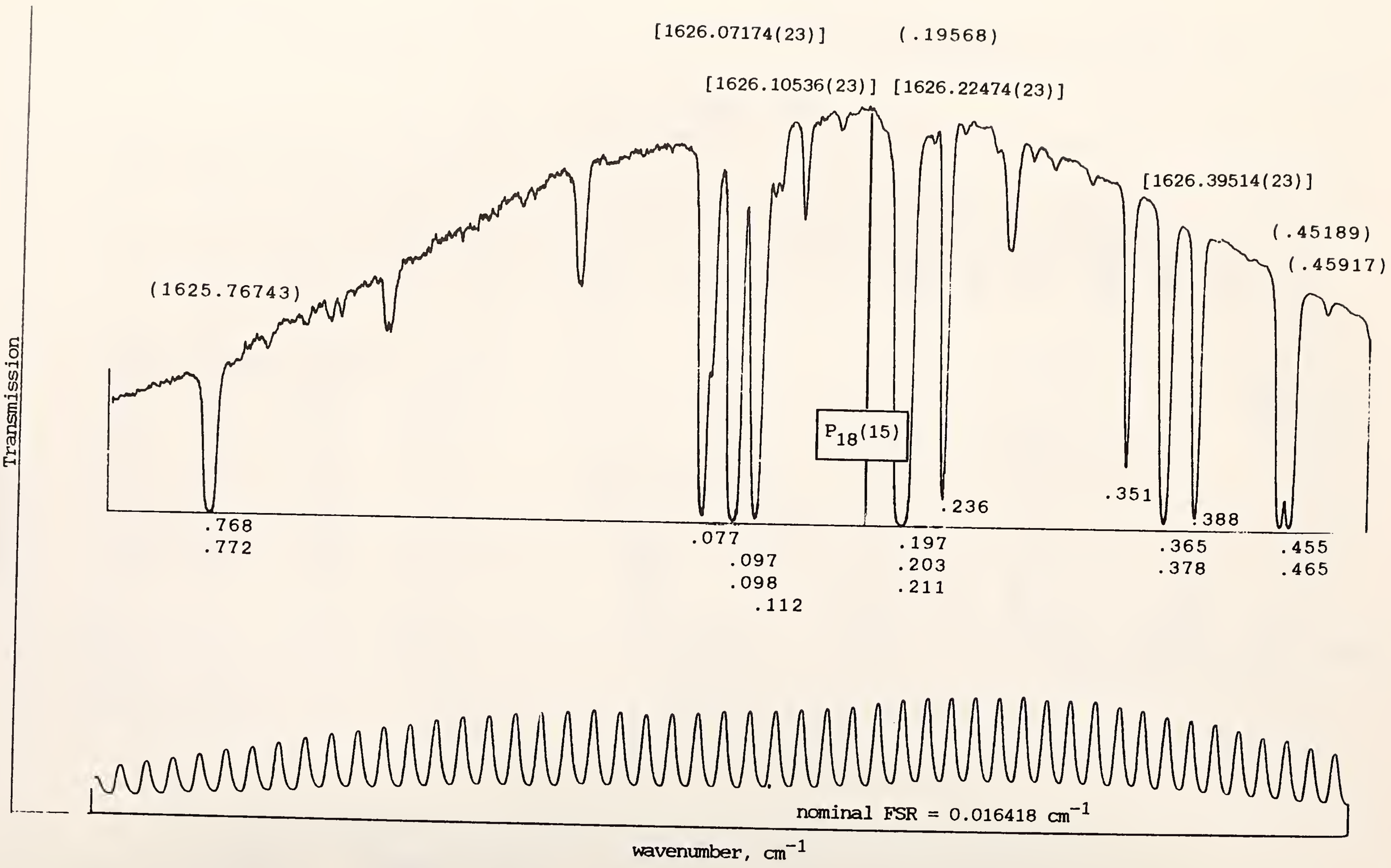

Fig. A-7. Nitrogen dioxide spectrum in the vicinity of the $P_{18}(15)$ CO laser line at 1626.17454 cm^{-1}.

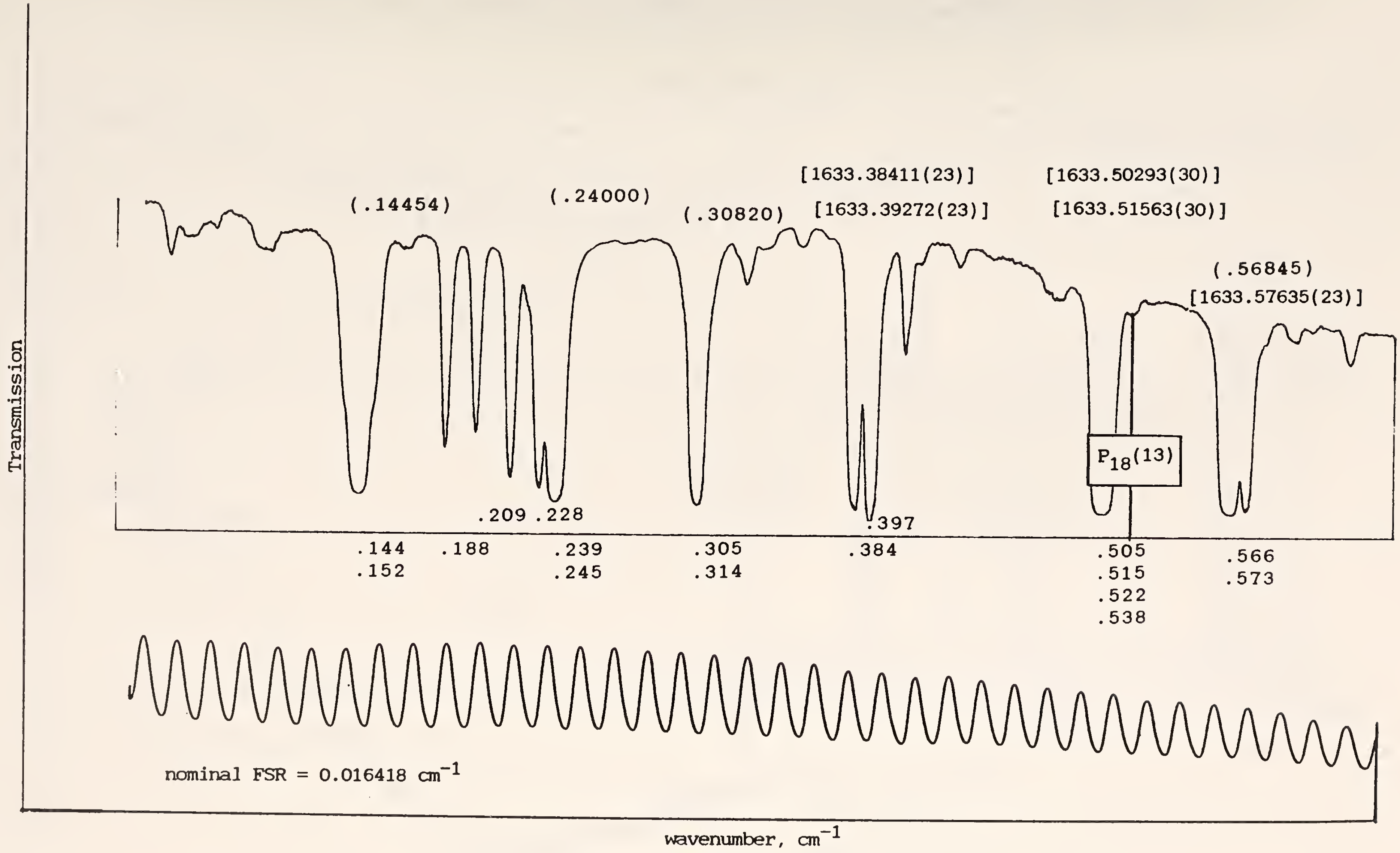

Fig. A-8. Nitrogen dioxide spectra in the vicinity of the $P_{18}(13)$ CO laser line at 1633.51953 cm^{-1}.

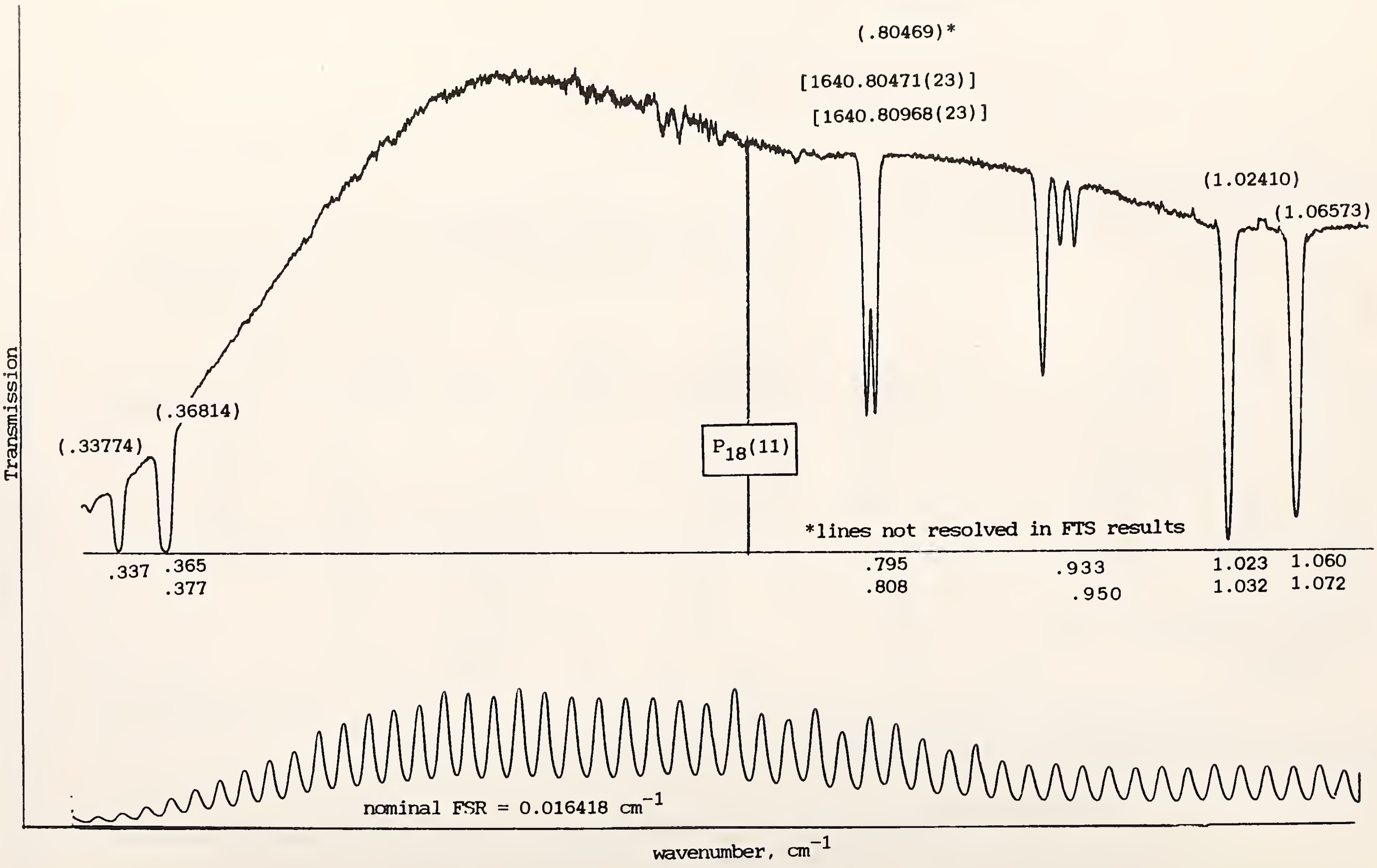

Fig. A-9. Nitrogen dioxide spectra in the vicinity of the $P_{18}(11)$ CO laser line at 1640.73251 cm^{-1}.

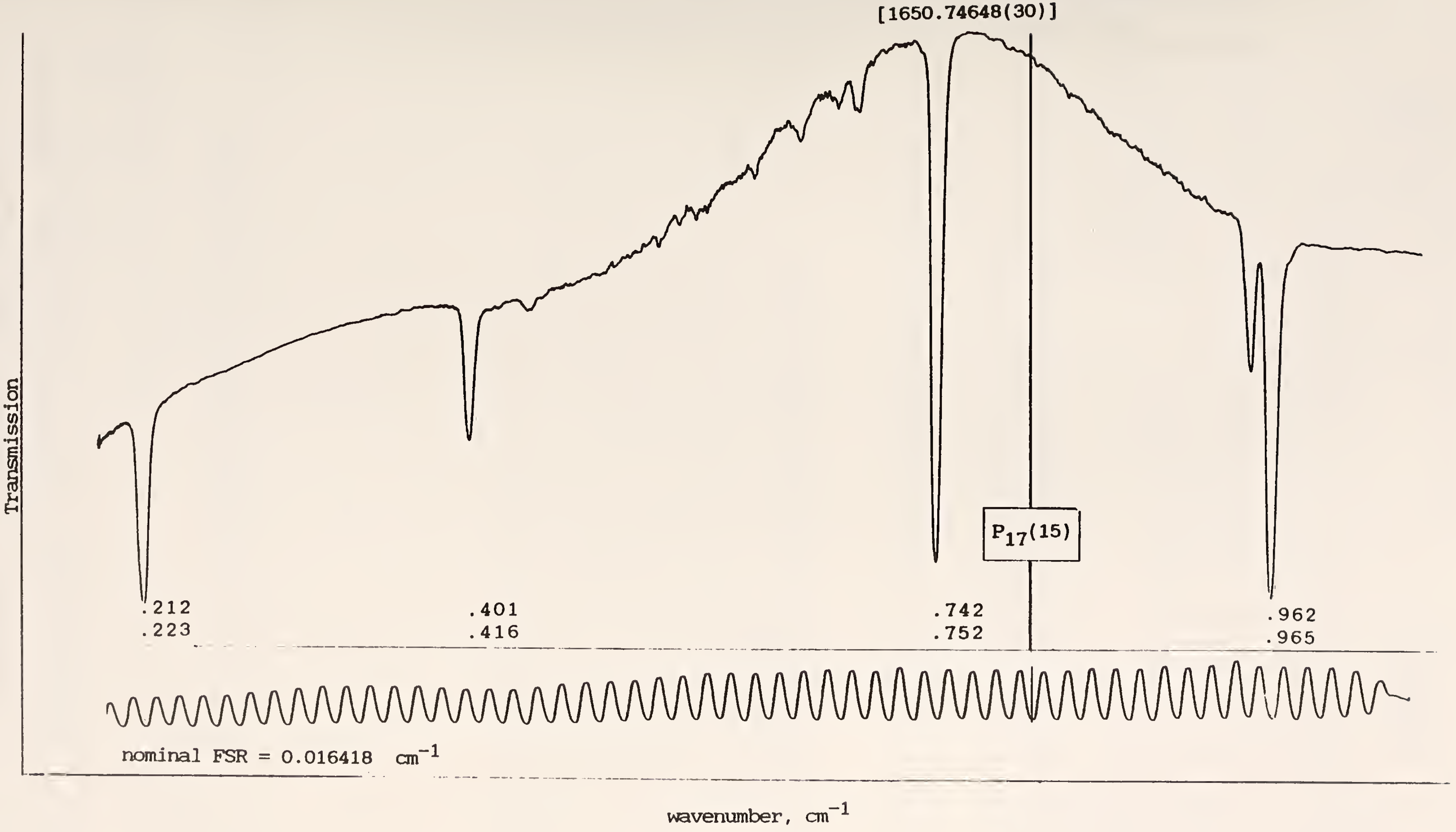

Fig. A-10. Nitrogen dioxide spectra in the vicinity of the $P_{17}(15)$ CO laser line at 1650.81080 cm^{-1}.

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11. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)*

Heterodyne frequency measurements have been made on selected groups of nitrogen dioxide rovibronic transitions between 1580.8 and 1650.7 cm^{-1}. The groups are separated by 7 to 10 cm^{-1}, and the intent is to provide a limited (and interim) calibration table for the region. In addition to a table of measured frequencies in this region, we have included figures of spectra in the vicinity of the measured lines to provide a map for identifying the transitions measured.

12. KEY WORDS *(Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)*

calibration frequencies at 6.2 μm; heterodyne frequency measurements; nitrogen dioxide spectra

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