standard formula for rovibrational levels of a linear molecule (18). Three molecular parameters, the vibrational band origin (ν_4) and the lower and upper state rotational constants (B" and B'), were determined by least-squares analysis, and the uncertainties were evaluated from the resulting covariance matrix. Statistical analysis indicated that the two rotational constants are highly correlated (0.99).

 C_3 is thought to be slightly quasi-linear (13, 14). Quasi-linearity is a result of a small barrier to linearity in the molecular bending potential (11). Molecules exhibiting this phenomenon are characterized by highly anharmonic low-frequency bending vibrations and strong stretch-bend interactions. Quasi-linearity manifests itself in the antisymmetric mode of the C₃ cluster in the average molecular structure becoming more compact (the rotational constant increases by 1.2%) upon excitation of the antisymmetric stretch vibration. The rotational constant of a normal linear molecule would be expected to decrease upon similar vibrational excitation. C5 and C7 have low-energy bending vibrations and are also potentially quasilinear. Recent spectroscopic measurements on C₅ indicate that its bending vibrations are reasonably harmonic, and no evidence for quasi-linearity is observed (15). The results presented here similarly do not evidence quasi-linearity in the case of C7. Upon excitation of the v_4 vibration the rotational constant is found to decrease by about 0.35%, and thus the average length of the molecule increases. While this is not suggestive of quasi-linearity in C7, it does not rule it out either.

The detection of the C₅ and C₃ clusters in the carbon star IRC+10216 by Bernath and co-workers (1, 19) with infrared astronomy highlights the importance of carbon clusters in astrophysical processes. The following ion-molecule reaction scheme has been proposed by Suzuki (20) for astrophysical production of carbon chains:

$$C^{+} + C_{n} \rightarrow C_{n+1}^{+} + h\nu C_{n+1}^{+} + H_{2} \rightarrow C_{n+1}H^{+} + H C_{n+1}H^{+} + e \rightarrow C_{n+1} + H$$
(1)

Calculations done by Freed, Oka, and Suzuki (21) compared unimolecular dissociation rates versus rates for infrared emission. Cluster growth was shown to proceed at the Langevin rate for $n \ge 4$. This may explain the high C5 abundance observed in IRC+10216 (1). It follows from the model that C_7 should also be abundant. The C_7 absorption spectrum reported here was a factor of 3 to 5 less intense than the ν_3 spectrum observed for C5. This is expected if the oscillator strength of the v_4 transition of C_7 is similar to that of the ν_3 transition of C_5 .

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The observed decrease of absorption intensity comes from an increased ground-state partition function for the larger C7 cluster and an expected slight decrease in the relative density of C_7 in the molecular beam (6, 22). If the interstellar C_7/C_5 abundance ratio is not too small, then the frequencies reported here will make astronomical observation of this molecule possible. This, in turn, will constitute an important step in understanding the nature and distribution of carbon in the interstellar medium.

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Tunable Far-IR Laser Spectroscopy of Jet-Cooled Carbon Clusters: The ν_2 Bending Vibration of C₃

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Seven rovibrational transitions of the $(01^{1}0) \leftarrow (00^{0}0)$ fundamental bending band of C3 have been measured with high precision with the use of a tunable far-infrared laser spectrometer. The C3 molecules were produced by laser vaporization of a graphite rod and cooled in a supersonic expansion. The astrophysically important v_2 fundamental frequency is determined to be 63.416529(40) cm⁻¹. These measurements provide the basis for studies of C₃ in the interstellar medium with far-infrared astronomy.

HE STUDY OF CARBON CLUSTERS BY both theory and experiment has received a great deal of recent attention (1-3). These species have proven to be remarkably ubiquitous. They are observed, for example, in flames and electrical discharges, in carbon stars, and in laser-produced plasmas of various polymers. They are implicated in the process of soot formation and in the aggregation of interstellar dust grains. Many investigations have evidenced new closed structures of carbon composed of five and six membered rings.

The elucidation of the roles of carbon clusters in these various contexts has been seriously impeded by the absence of spectral and structural data for them. In fact, detailed

laboratory spectra have been measured only for C_2, C_2^-, C_2^+, C_3 , and C_5 , although both infrared and electron spin resonance data have been obtained for several other species from studies in cryogenic matrices (4). We are attempting to improve this situation. In a previous report (5) we described a general new technique for measuring carbon clusters with mid-infrared laser absorption spectroscopy, and infrared (IR) spectra of the C5 cluster were presented as an initial demonstration of the method. The detection of C₅ in the carbon star IRC+10216 by infrared astronomy was presented at the same time by Bernath et al. (6). In this report, we describe a new experiment for measuring far-infrared (FIR) (10 to 350 cm^{-1}) spectra of carbon clusters with high precision (1×10^{-6}) and high sensitivity, and present the first direct measurement of the astrophysically important 63 cm⁻¹ bending mode of

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 C_3 as an initial demonstration of the method.

Ab initio quantum chemistry calculations have shown (4) that for C_n with n < 10, the clusters with odd n have linear ground states of ${}^{1}\Sigma_{g}^{+}$ symmetry, whereas those with even n have two roughly equi-energetic lowest states—a linear ${}^{3}\Sigma_{g}^{-}$ state and a cyclic singlet state. All of the linear clusters are expected to exhibit bending vibrations (7) in the range 40 to 150 cm⁻¹, which is accessible with this experiment. Thus, a systematic study of the structures, properties, and dynamics of these species is now possible. In addition to the intrinsic value of such measurements for characterizing the general nature of carbon-carbon bonding, they will also provide the basis for a systematic study of carbon clusters in the interstellar medium, with the use of recently developed (8) FIR laser heterodyne receivers.

While C₃ has been studied in detail by electronic emission (9) and absorption (10) spectroscopy, mid-infrared laser spectroscopy (11, 12), stimulated emission pumping (13, 14), and matrix isolation IR spectroscopy (4), the bending fundamental has not been accurately characterized. High level ab initio calculations (15) indicate that the ground state of C₃ is weakly quasi-linear, with an equilibrium bond angle of 162° and a barrier to linearity of 21 cm^{-1} . As a result, the bending motion is quite floppy, and the fundamental frequency is predicted to be very low—near 63 cm^{-1} . Until recently, tunable coherent sources of radiation did not exist in this frequency region; consequently, high-resolution FIR spectroscopy of the isolated cluster was not possible. We have recently developed a tunable FIR laser transient absorption spectrometer incorporating supersonic beam production of clusters, and have employed it to make this measurement.

The ¹²C nucleus has spin I = 0, which allows only symmetric spin states for C3, and causes all rotational levels of a given vibronic state to have the same parity. Therefore, the selection rules $s \leftrightarrow s$ and $+ \leftrightarrow -$ allow the $\nu_2 = 1 \leftarrow 0$ energy to be determined only by direct measurement of this FIR transition, not through combination differences of infrared and optical transitions. This precise determination of the v_2 band origin is of considerable astrophysical interest because it will facilitate the detection of C_3 in the interstellar medium at FIR wavelengths. As C3 has no permanent dipole moment, and hence no pure rotational spectrum, this bending vibration will be the lowest frequency emitted or absorbed by C₃. Although this species has been detected in the carbon star IRC+10216 by mid-IR astronomy (16), dense molecular clouds will prob-

Table 1. Measured FIR rovibrational transitions to the ν_2 fundamental. Uncertainities in parentheses are 1σ . Calculated frequencies are from the weighted least-squares fit given in Table 2.

J'	J″	Frequency (cm ⁻¹)	${ \nu_{obs} - \nu_{calc} \over (10^{-3} \ cm^{-1}) }$
5	6	58.07760(23)	-0.28
3	4	59.63761(23)	-0.15
1	2	61.26976(23)	-0.10
4	4	63.26734(3)	0.03
6	6	63.58860(3)	-0.01
5	4	67.54845(3)	-0.02
7	6	69.50230(3)	0.01

ably be too cold for C_3 to exist in the excited states necessary for detection in the mid-IR. Therefore, rovibrational lines in the fundamental bending vibration will be the most promising transitions to search for in these cold sources.

The tunable FIR laser system itself has been described previously (17) and only a brief review and the particular modifications incorporated for the study of carbon clusters will be discussed here. Fixed frequency FIR radiation is generated by continuous wave optical pumping of a 2.4 m far-infrared molecular gas laser with a commercial (Apollo Lasers) 150 W line-tunable CO₂ laser. Far infrared radiation in the range 13 to 150 cm⁻¹ has been generated with this system. The fixed frequency FIR radiation is mixed with tunable microwave radiation in a Schottky barrier diode (Univ. Va. 117) to generate sum and difference frequencies at $v_{\text{FIR}} \pm nv_{\text{MW}}$, where $n = 1, 2, 3, \ldots$ Microwave power from 2 to 110 GHz is available, to give tunability of more than 7 cm⁻¹ about a given fixed frequency far infrared laser line. The tunable sideband power (10 to 100 μ W) is separated from the much stronger (10 to 50 mW) carrier frequency (v_{FIR}) with a polarizing Michelson interferometer, and the laser is detected by a stressed Ge:Ga photoconductor, which responds from 55 to 85 cm^{-1} .

The FIR laser frequencies used in this study were 1891.2743 GHz (CH₂F₂), 1757.5263 GHz (CH₃OH), 1838.8393 GHz (CH₃OH), and 2058.1418 GHz (CH₃OD). Care had to be taken to ensure that the detector would not be saturated by residual laser carrier radiation transmitted through the polarizing Michelson interferometer. The tunable sidebands were typically about as strong as the residual fixed frequency laser power that leaked through the interferometer, as measured with a low finesse (~ 10) Fabry-Perot etalon. At times it was necessary to use the Fabry-Perot to reject the residual carrier frequency. Laser power-to-noise ratios on the order of 10⁵ to 10⁶ were achieved with a 1 Hz lock-in



Fig. 1. The Q(6) transition in the ν_2 bending fundamental of C₃ measured in direct absorption near 63.4 cm⁻¹ with the tunable FIR laser spectrometer. The clusters produced by 248-mm laser vaporization of a graphite rod are cooled in a supersonic expansion.

Table 2. Results of the least squares analysis. Uncertainties in parentheses are 1σ .

State	Constant	Value (cm ⁻¹)
	ν ₀	63.416529(40)
$(00^{0}0)$	В	0.4305723(56)
` '	D	$0.1472(13) \times 10^{-5}$
	H	$0.1333(59) \times 10^{-9}$
$(01^{1}0)$	В	0.4424068(52)
	<i>q</i> e	0.0056939(21)
	\tilde{D}	$0.2361(16) \times 10^{-5}$
	9D	$-0.0869(27) \times 10^{-5}$
	\overline{H}	$0.267(12) \times 10^{-9}$
	<i>q</i> _H	$0.027(23) imes 10^{-9}$

bandwidth at 100 kHz.

A fivefold improvement in sensitivity of the system was obtained by use of a multipass cell of the type presented by Perry and co-workers (18). The cell consists of a nearconcentric cavity in which the laser travels non-axially. Unlike the configuration described by Perry, in which the input and output beams enter and exit through opposite sides, we force the input and output beams to enter and exit from the same side. This is done because the wide FIR beam waist (~ 0.5 cm) necessitates large clearances between the beam and the mirrors. We typically employ 10 to 16 passes, with an insertion loss of \sim 85%. Since the laser travels non-axially in the cell, we observe Doppler broadening of the transitions due to the components of the laser parallel and anti-parallel with the expansion. At 63 cm^{-1} , this broadening is about 3 to 8 MHz.

The C₃ molecules were generated in a supersonic expansion by excimer laser vaporization of a graphite rod (National Arc Carbons) as in our study (5) of C₅. The 248 nm KrF line, with a typical power of 100 mJ per pulse was used at a repetition rate of 120 Hz. The excimer laser was focused on the rod with a 50-cm focal length CaF₂ lens. We found that decreasing the excimer laser power by a factor of 2 did not affect the signal strength. Argon carrier gas was used, with

pressures of 8 to 11 atm behind a pulsed valve and a 1 ms gas pulse length. No signal was observed when with helium as carrier gas:

Similar studies (19) of NH₂ generated by photolysis of NH3 established new methods of detecting absorptions of transient species. There are some differences in our current configuration as compared to that of Cohen et al. (19). Previously, we frequency modulated the laser and demodulated the detector output before sending it to the boxcar integrator. In the present configuration, we no longer frequency modulate the laser, but instead filter the detector output with a 10 kHz to 30 kHz bandpass filter. This filter allows the majority of the frequency components of the signal to pass, but rejects the majority of the 1/f noise. Second, the absorptions were detected by using two boxcar integrators (Stanford Research SR250) and an analog processor (Stanford Research SR235) instead of a single boxcar. The first boxcar opened $\sim 60 \ \mu s$ after the excimer laser pulse and had a gate width of $4 \mu s$. The background subtraction gate opened ~25 μ s after the first, and had a width of 3 μ s. The amount of delay relative to the excimer shot depends on the distance from the source to the FIR laser beam, which was about 3.5 cm. The precise values of the boxcar delays were optimized by simultaneously displaying the output from the detector and the boxcar gates on an oscilloscope. By holding the laser frequency at the absorption maximum of a strong transition the gates could be positioned easily. The outputs of the two boxcars were then subtracted by the analog processor. This approach has the advantage over a single box-

car using background subtraction in that: (i) two different gate widths can be used to optimize the signal-to-noise ratio, and (ii) very small temporal separations between the two gates are possible. A scan of the Q(6)line is shown in Fig. 1. The best signal-tonoise ratio observed for C3 was near 200, obtained when averaging 12 excimer shots per data point and collecting 1000 data points per 150 MHz scan.

The measured transition frequencies of the 7 lines observed in this study are presented in Table 1. The rotational temperature is estimated to be near 10 K. A weighted least squares fit was performed with these 7 lines and 34 combination differences from the $(00^{0}1) \leftarrow (00^{0}0)$, and $(01^{1}1) \leftarrow (01^{1}0)$ infrared bands measured by Hirota and coworkers (11, 12). The energy expressions used were: for the $(00^{0}0)$ state

$$E = BJ(J + 1) - D[J(J + 1)]^{2} + H[J(J + 1)]^{3}$$
(1)

and for the $(01^{1}0)$ state

$$E_{\pm} = \nu_0 + B[J(J+1) - \ell^2] - D[J(J+1) - \ell^2]^2 + H[J(J+1) - \ell^3]^3 \pm 1/2 \{q_\ell J(J+1) + q_D[J(J+1)]^2 + q_H[J(J+1)]^3\}$$
(2)

where v_0 is the band origin, B, D, and H are the rotational constants, centrifugal distortion constants, and third-order distortion constants, respectively, and q_{ℓ} , q_D , and q_H are their associated ℓ -type doubling constants. Here the +(-) signs refer to states with even (odd) J. The results of the fit are given in Table 2.

The long-term frequency drift of the FIR laser leads to uncertainties of about 7 MHz



The intensity of the ν_2 fundamental studied in this work was expected to be quite low, given that the theoretical prediction $(15)^{\circ}$ for the transition dipole was 0.19 debye, compared with 0.44 for the ν_3 band. Hence, the absorption coefficient of the ν_3 band is expected (12) to be 100 times stronger than that of the ν_2 fundamental. The most intense rovibrational lines of the v_3 fundamental observed in a similar supersonic beam source with our diode laser spectrometer exhibited a fractional absorption near 20%, the same result observed for the strongest lines of the ν_2 band. Work is presently under way to better quantify the fractional absorption of the ν_2 and ν_3 bands. Given these results, one can conclude that the ab initio estimates described by Kraemer et al. (15) must be in error. This is an important issue to resolve, since the extraction of column densities from remote observations depends explicitly on the transition moment.

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used in this work. 24 AUGUST 1990

Diplexer

- **REPORTS** 899
- CO, spectrum ánalyzer Π 3 FIR laser (50-1000 µm) h 1-Tunable sidebands Rooftop mixer (GaAs) Detector Microwave source Excimer laser Vaporization -> source Multipass cell

Fig. 2. Schematic diagram of the Berkeley tunable far infrared laser transient absorption spectrometer

CO, laser (10 µm)

(1989).

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Ungrouped Iron Meteorites in Antarctica: Origin of Anomalously High Abundance

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Eighty-five percent of the iron meteorites collected outside Antarctica are assigned to 13 compositionally and structurally defined groups; the remaining 15 percent are ungrouped. Of the 31 iron meteorites recovered from Antarctica, 39 percent are ungrouped. This major difference in the two sets is almost certainly not a stochastic variation, a latitudinal effect, or an effect associated with differences in terrestrial ages. It seems to be related to the median mass of Antarctic irons, which is about 1/100 that of non-Antarctic irons. During impacts on asteroids, smaller fragments tend to be ejected into space at higher velocities than larger fragments, and, on average, small meteoroids have undergone more changes in orbital velocity than large ones. As a result, the set of asteroids that contributes small meteoroids to Earth-crossing orbits is larger than the set that contributes large meteoroids. Most small iron meteorites may escape from the asteroid belt as a result of impact-induced changes in velocity that reduce their perihelia to values less than the aphelion of Mars.

RON METEORITES ARE CLASSIFIED ON the basis of their structures and detailed compositions (1). In all, 13 groups of five or more meteorites have been identified, but 15% of the iron meteorites cannot be so classified and are called "ungrouped" (the choice of five as the minimum number required to form a group is arbitrary). Meteorites in 10 of the 13 groups seem to have formed by the fractional crystallization of large metallic magmas on separate parent bodies. The origin of the meteorites in the remaining 3 groups is not settled; they may have formed as individual pools of shock melt (2). Strong interelement correlations are observed in each group; slopes on element-Ni trends are generally higher in the magmatic than in the nonmagmatic groups. For some elements (Co, Cu, and especially Ga and Ge), the total compositional variation in most groups is much smaller than that between groups; thus, these elements are useful taxonomic parameters.

The ungrouped irons have compositions that place them distinctly outside the group fields on most element-Ni diagrams. Generally the structures (for example, kamacite bandwidths and the nature and content of nonmetallic phases) are also inconsistent with membership in the groups that have the most similar contents of the taxonomic elements. It is convenient to designate sets of one to four compositionally related irons as "grouplets." Each of these grouplets also appears to have formed in a separate parent body. Roughly 40 asteroidal parent bodies are required to account for the 100 ungrouped irons; thus, 50 to 55 asteroids are needed to account for the 605 characterized iron meteorites. Stony meteorites show less diversity; the characterized set requires about half as many parent bodies as the irons.

Clarke (3) recognized that the fraction of ungrouped irons in the set from Antarctica was much larger than that of irons from the remainder of the world. Wasson et al. (1) confirmed this observation and expanded the characterized set (8 ungrouped in a total of 24 Antarctic irons) and discussed mechanisms that could account for the enhanced abundances. I report data for seven additional Antarctic irons (including four ungrouped); thus, the ungrouped fraction is now 12/31 or 0.39. Because each of the 12 Antarctic ungrouped irons has a different composition, the large fraction of ungrouped irons cannot have resulted from atmospheric breakup, a process that could be responsible for the anomalously high fraction of H chondrites in Victoria Land, Antarctica (4).

Two of the seven new Antarctic irons (Table 1) are typical group members: IIIAB GRO85201 and IIICD LEW86540. The



Fig. 1. The quantity of numbered asteroids increases rapidly with increasing deviation of the semimajor axis from that (2.501 AU) corresponding to a 1:3 period resonance with Jupiter. Semimajor axes are from the compilation by Bender (18).

composition of another one, EET87506, places it within IAB fields on most element-Ni diagrams, but its Ir content is about three times as high as that expected, and its structure is anomalous. The remaining four irons are ungrouped; their compositions differ in numerous ways from those observed in groups having the most similar Ga and Ni contents. Compared to mesosiderite metal nodules having a similar Ni content, Ir in ALH84233 is low by a factor of 100, and Co, As, and Au are about 1.2 times as high. Meteorite EET87516 has a structure and Ga content similar to those of group IVA irons, but its Ir content is too high by a factor of 10, its Co content is high by a factor of 1.2, and its Ga content is low by a factor of 1.2 for an IVA iron with a Ni content of 92 mg/g.

One of the most interesting of the new irons is LEW85369, which contains Si dissolved in the Fe-Ni metal; in this and most other compositional respects, LEW85369 resembles the ungrouped Horse Creek iron meteorite and the ungrouped metal nodules from the Mount Egerton stony-iron meteorite. The LEW86211 iron contains $\sim 62\%$ FeS by volume (5), after Soroti the second highest percentage in an iron meteorite. It is compositionally most similar to the low-Ni extreme of group IIE but is deviant on most element-Ni diagrams; for example, Ir and W are 34 and 1.5 times as high, respectively, as the highest IIE values.

In all, 88 of the 574 non-Antarctic irons are ungrouped (1), a fraction of 0.153. If the probability that any randomly chosen iron is ungrouped is 0.153, the probability of finding ≥ 12 ungrouped irons in a set of 31 is 0.0013. It is thus very unlikely that the Antarctic irons are sampling the same population as the non-Antarctic irons.

It is not possible to account for the high

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