$\omega(^{12}CO) = 2143$ and $\omega(^{13}CO) = 2095$ cm^{-1} $\therefore$ $\Delta H = 24$ cm^{-1}. The equilibrium constant for this process at 80 K has been determined by Smith and Adams (1980) and it fits in beautifully with this simple theory and neatly explains the observed $^{12}CO/^{13}CO$ abundances.

The explanation of anomalous isotopic abundances by fractionation is a very strong argument in favour of ion–molecule reactions. These fit nicely as they are fast at the low temperatures as they have negligible activation energies in general. For higher temperature reactions isotope fractionation should be negligible.

Smith and Adams (1977) have determined many of the rate constants required for a detailed analysis of ion–molecule chemistry using the SIFT technique (*see* review in this issue). Huntress (1977) had discussed complementary data obtained using the ICR (ion cyclotron resonance) technique. The effective temperatures of the reactants in the ICR method may be quite high and thus the data may not apply directly to the low temperature ISM.

Prasad and Huntress (1980) have recently presented the results of a very large computer study of the chemistry of the ISM. It is still not at all obvious how molecules such as HC_9N can be produced in cold clouds by a sequence of two-body coupling reactions. Schiff and Bohme (1980) have suggested a possible sequence of steps. It may be that radiative association and neutral–neutral processes may become significantly more important for larger molecules.

The third process which must be taken very seriously is that of molecule formation in high temperature, high density, circumstellar envelopes followed by their subsequent ejection into the general ISM. This mechanism is further discussed in the last section.

DISCUSSION

It would be remarkable if radio spectroscopy were to continue to spring as many exciting surprises in the next decade as it has in the last, but of course this is possible. There are a few areas which can be identified as requiring major efforts in development. New laboratory techniques for producing astrophysically important microwave and infrared spectra are needed. The spectra of free radicals, molecular ions (positive and negative), unstable molecules with multiple bonds and second row atoms are particularly important. More work on high temperature species is also necessary. In addition collisional and radiative energy transfer processes are important and as higher temperature regions are probed, vibrational satellites are becoming detectable. Certainly a great deal of effort is being put into the solution of some of these problems as discussed below.

Obviously one must always strive for higher sensitivity, speed of data collection, spatial resolution, etc. and as successive advances are made some of the goals will hopefully be achieved and new, unexpected discoveries will be made. The most recent high resolution surveys are indicating that the interstellar radio spectrum appears to be rather cluttered up at least at mm wave frequencies. It is now becoming clear that we have only identified the sparse long grass and there may well be a rather dense undergrowth of weak lines which will be difficult to disentangle. Much greater sensitivity may just uncover myriads of weak lines offering few prospects for certain identification. There may thus be a limit to the number of molecules that can be identified by radio techniques ($\sim 150?$).

In the infrared range the problem may be more serious as each vibrational band is accompanied by an entourage of complicated rotational branch structure which will limit the value of the technique for the study of larger molecules.

The study of interstellar molecules is an expanding field, expanding in many directions and the rest of this section is devoted to a review of some of the important questions which

need to be answered and also an attempt to uncover some questions that still need to be asked.

Mapping

In the next phase of this subject a great deal of mapping will probably be carried out and from these some coherent view of interstellar molecule ecology may develop. Certainly this arduous task should form a basis for the determination of the physical conditions which exist in the ISM. The next steps are, of course, to strive for higher and higher spatial resolution by studying higher frequencies such as those in the infrared which in general require higher excitation temperatures or by use of aperture synthesis techniques (Welch *et al.*, 1981). Such developments are extremely important in indicating just how granular the clouds may be. Homogeneity is not an obviously ubiquitous property of the Universe and each successive breakthrough in spatial resolution seems to yield new and unexpected information which requires successive overhauls of the models of interstellar clouds.

There is also room for lower resolution surveys. Just what can be achieved is indicated by the Columbia CO survey using the small 4 ft dish in New York (Cohen *et al.*, 1980; Blitz, 1979; Fig. 26). This survey has shown that molecular filaments link the various clouds (Morris, Montani and Thaddeus, 1980). The survey has also yielded data on the distribution of GMCs in the Galaxy—perhaps some of the most important results as far as astrophysics is concerned—and shown just how extensive the GMCs are with important implications with regard to the structure mass and evolutionary track of the Galaxy.

These studies should yield information about the longevity of the clouds; whether they are 10^7–10^8 years old and tied to the spiral arms (Cohen *et al.*, 1980) or even older and concentrated in a doughnut-shaped ring showing no spiral structure (Solomon and Sanders, 1979). As far as astrophysics is concerned a knowledge of molecular distributions will assist the understanding of the energy balance mechanism which controls cloud collapse to star and planet formation. The discovery of molecules showed that clouds could keep cool by losing energy at very low temperatures by rotational emission. The cloud gravitational energy thus converts via kinetic energy to rotational energy and then to microwave or infrared photons which leave the cloud at the velocity of light. In this way a better understanding of the cloud collapse scenario has evolved.

The spatial abundance relationships between molecules should assist in deciding which, if any, of the various chemistries is most important. Searches probably by the new infrared techniques for stars, hot or cold, new or old, embedded in the clouds are also important to this end. It is certainly now clear that maps such as those of Beckwith *et al.* (1978) (Fig. 30) must be the first shots for the baby star scrap book. The more we can learn about the birth of stars the more we should learn about planet formation—something which seems to be only vaguely understood (Hartman, 1978). Indeed this situation should cause us to stop and reflect that even though we have much more detailed information about the solar system than any other bit of the Universe we appear to understand its origin less well than the rest!

Laboratory experiments, theoretical calculations and future radio detections

As the sensitivity of the radio technique improves there will undoubtedly be new molecular identifications and it is perhaps instructive to conjecture what possible new results may turn up.

The recent detections of HNCS (Frerking, Linke and Thaddeus, 1979), CH_3SH (Linke, Frerking and Thaddeus, 1979) the sulphur analogues of hydrogen isocyanate (HNCO) and methanol (CH_3OH) indicate that the good old Mendeleevian principle is still

a valuable guide. Thus, we can draw up a list of a few possible contenders—second row analogues of first row interstellar species.

The species HCN and HNC are, as we have seen, well characterized species and there is every reason to suspect that the phosphorus analogue $HC\equiv P$ studied by Tyler (1964) and the silicon analogues HNSi and HSiN whose microwave spectra are not known may be detectable. HCP will be difficult to detect because its dipole moment is only 0.39 Debye as compared with 2.985 Debye for HCN. In addition the cosmic abundance ratio P : N is $\sim 1:270$, implying an intrinsic factor of $\sim 1/16\,000$ between the HCN and HCP intensities. The spectra of the symmetric top molecules PH_3 (Helminger and Gordy, 1969) and $CH_3C\equiv P$ (Kroto, Nixon and Simmons, 1979) are more likely to be detectable contenders as interstellar analogues of NH_3 and $CH_3C\equiv N$ respectively.

Perhaps the most likely phosphorus containing species is cyanophosphaethyne $N\equiv C-C\equiv P$ which was made and studied for the first time with interstellar observation as a priority (Cooper et al., 1978) (Fig. 11). It is a linear molecule and in general these are more readily detected than other species as discussed previously. It is a very close relative of $HC\equiv C-C\equiv N$ which is quite abundant in some clouds. It also has a fairly large dipole moment of 3.5 Debye. These factors suggest that its intensity may only be about 1/300 of that of $HC\equiv C-C\equiv N$ for which ^{13}C isotopes with only slightly higher abundance have been detected. Initial search attempts have proved negative. There is some evidence that phosphorus is depleted in the ISM relative to the cosmic abundance. It is however possible that in sources such as IRC + 10216 the depletion is less than in the cold clouds.

Phosphaethyne the phosphorus analogue of $CH_2=NH$ (Johnson and Lovas, 1972) was first detected by Hopkinson et al. (1976), (Kroto et al., 1980). The species $CH_2=CHC\equiv P$ (Ohno, Kroto and Nixon, 1981) and $HC\equiv C-C\equiv P$ (Kroto, Ohno and Nixon, 1981) are also known. They are the analogues of $CH_2=CHC\equiv N$ detected by Gardner and Winnewisser (1975) and $HC\equiv C-C\equiv N$ detected by Turner (1971).

Several sulphur analogues of oxygen compounds have already been detected. The observation of OCS (Jefferts et al., 1971) is a good indication that CO_2 is quite abundant and it can only be a matter of time before it is detected by one of the new infrared techniques. CS is well studied and a radio line from the Orion molecular cloud is shown in Fig. 29 (Scoville, 1979). The widespread abundance of H_2CO (Chaisson, 1978), the detection of acetaldehyde $CH_3CH=O$ (Gottlieb, 1973) and ketene $H_2C=C=O$ (Turner, 1977) together with the detection of H_2CS (Sinclair et al., 1973) suggest that thioacetaldehyde, $CH_3CH=S$, and thioketene, $CH_2=C=S$, whose microwave spectra are known (Kroto and Landsberg, 1975; Georgiou, Kroto and Landsberg, 1979), should be detectable.

The detection of formic acid, HCOOH by Zuckerman, Ball and Gottlieb (1971) suggests that its thio analogues might be detectable. Monothioformic acid HCOSH has been studied by Hocking and Winnewisser (1977). The recent possible detection of $HOCO^+$ (Thaddeus, Guélin and Linke, 1981) indicates that it might be worthwhile searching for $HOCS^+$ and $HSCS^+$, etc. Liszt (1978) has discussed searches for magnetic dipole transitions of S_2.

Silicon analogues are of great interest to chemists as well as astronomers. A significant point about Si is highlighted by Fig. 29 where the spectrum of SiO from Orion is compared with that of CS from the same source. The line widths imply that SiO is only detected in the high temperature expanding cloud in the region of the KLBN source and not in the surrounding cloud presumably because it has condensed out in grains. Some very likely candidates for detection are the species HSiN (Lovas, 1974) and HNSi, analogues of HCN and HNC. Ab initio calculations aimed at estimating the B_0 values of these two species (Kroto et al., 1978) revealed the interesting prediction that the iso species HNSi should be much the more thermodynamically stable, the reverse situation exists in the case

of HCN and HNC. These calculations seem to yield B_0 values to ~ 1 per cent. In this case for instance $\Delta E(J = 1 \rightarrow 0) = 2B = 79\,070 \pm 130$ MHz for HNSi. These calculations indicated a similar preference for Si to form $CH_2=Si$ rather than $HC{\equiv}SiH$ the acetylenic analogue (Murrell, Kroto and Guest, 1977). The former extremely interesting species was detected by Dubois and Leclercq (1980) by its electronic spectrum using a flash discharge technique during the study of C/Si species of astrophysical interest. This species, which will have a rather similar microwave spectrum to CH_2S, should be detectable in similar regions to those where SiO is observed. Some other interesting Si species are known, at least by their optical spectra such as SiC_2 (Kleman, 1956) and SiH_2 (Dubois, 1968). SiC_2 is one of the Si analogues of C_3 identified by Douglas (1951) as the carrier of the 4050 Å comet bands.

It can only be a matter of time before even longer cyanopolyynes are discovered. The most recent results indicate that the ratio of HC_9N/HC_7N in TMC1 is $\sim 1/4$ (Broten *et al.*, 1978). This seems at first to be an unusually slow decrease in polyyne abundance with increasing chain length. Interestingly and perhaps importantly $CH_3C{\equiv}C-C{\equiv}N$ does not appear to be so readily detected as the linear cyanopolyynes. One possible reason is of course the line dilution effect, caused by the third degree of freedom which distributes the available molecules among the various $|K|$ states for a given value of J as discussed previously. The rotational frequencies of the longer systems $CH_3(C{\equiv}C)_2C{\equiv}N$ and $CH_3(C{\equiv}C)_3H$ are known (Alexander *et al.*, 1978) but so far searches have been unsuccessful. The molecule $CH_2=CHC{\equiv}N$ has been detected by Winnewisser and Gardner (1975) but its spectrum does not appear to be that strong. This species differs from $HC{\equiv}C-C{\equiv}N$ by the addition of two extra H atoms and its low abundance may be significant as far as the origins of these molecules are concerned. The species $CH_2=CH-C{\equiv}C-C{\equiv}N$ which is related to HC_5N in a similar way has also been made and its microwave spectrum studied (Kroto, Phillips and Walton, 1982). Similarly the molecule $HC{\equiv}CNCO$ related to HNCO (Buhl, Snyder and Edrich, 1972) and the polyynes has also been made and studied (Cooper, Kroto and Walton, 1982).

Herzberg (1980) has discussed the interplay of molecular spectroscopy and astronomy and highlights ways in which future advances may be forthcoming with particular reference to the species HeH^+, HNe^+, H_3, H_3^+, HC_nN etc.

There a large number of free radicals and unstable molecules whose electronic spectra are known but whose microwave and infrared spectra are only now being studied. Many of these such as C_3, C_2N, CNC, NCN, CN_2, N_3, HNCN, NCO, etc. are quite feasible interstellar species and their known properties are listed by Herzberg (1966).

A wide range of techniques is being developed for the study of the high resolution spectra of molecules which are temperamental in the laboratory (Carrington, 1974, 1980; Morino *et al.*, 1979).

The microwave techniques developed by Woods and coworkers (Woods, 1973; Gudeman *et al.*, 1981) for studying species in discharges have proven extremely valuable in confirming the U line assignments particularly in the case of the molecular ions (*see* page 345). An improvement in the sensitivity of this method would yield some extremely valuable laboratory frequencies for new molecular ions and so guide radio searches. Such an advance would also assist in the assignment of some of the 120 known U-line frequencies (Turner, 1979) some of which are quite likely to belong to slightly more complex and perhaps also more stable molecular ions than the very simple linear ones so far assigned.

Particularly interesting advances in the methods for obtaining spectroscopic data from molecular ion beams using lasers have been developed by various groups including Wing *et al.* (1976) and Carrington and Sarre (1977) to work in the optical and infrared regions. The data obtained at infrared frequencies should pay astrophysical dividends as infrared

astronomy improves. At present there are no sufficiently sensitive microwave techniques to carry out general laboratory searches for new rotational frequencies of ions. A most important breakthrough in ion spectroscopy has been made by Oka (1980) who has observed H_3^+ using a very sensitive tunable infrared laser to detect the very weak absorption directly through a hydrogen gas discharge. The molecule is the first equilateral triangle species detected. The interaction between laboratory infrared spectroscopy and infrared astronomy promises to provide more exciting information in the future especially about protostellar objects.

One of the most valuable advances in high resolution spectroscopy of free radicals is that developed by Evenson and coworkers (*see*, for instance, Evenson *et al.*, 1980). This is the technique of laser magnetic resonance which recently was used to detect C atoms in the laboratory (Saykally and Evenson, 1980). The detection of interstellar C (Phillips *et al.*, 1980) was facilitated by this measurement. As far-infrared astronomical sensitivity improves this particular technique should yield as valuable interstellar information as the molecular information already produced.

Theoretical calculations have played a useful rôle in this field, especially in adding to the circumstantial evidence used to identify some of the U lines. In general they have been ab initio calculations on rather simple systems such as HCO^+, HNC, etc. and it seems that the rotational constants for these systems can be calculated to about 1 per cent. This is really not good enough for certain identification, however it is often useful to rule out possibilities (Kroto *et al.*, 1978). In only one case has a calculation been truly successful and that is the special case (Oka, 1978) in which HC_9N was identified by numerical extrapolation (Broten *et al.*, 1978). There are few cases where this procedure is likely to be as successful as in the case of the polyynes (perhaps C_nH and C_nN).

The general value of theoretical calculation has been reviewed by Wilson (1980) who has given references to calculation on such species as: CSiH, SiCH, $HSiO^+$, $HOSi^+$, HSiN and HNSi, which are likely to give rise to detectable interstellar radio lines.

Grains and chains

It is clear that interstellar molecules are tied to grains and it is unlikely that we shall be entirely sure about molecule formation and the associated cloud ↔ star transition without a much more concrete understanding of what grains are. Grains have been identified by a process of elimination in that it is not obvious what other medium could give rise to the observed wavelength dependence and polarization characteristics of the interstellar scattering. The lemma to Holmes' Law may apply here in that we have to be certain that all other alternative possibilities have been eliminated before we can be sure the last one that remains is correct. It seems now worth considering how molecules may affect the picture. The long chain molecules may hold a key to some important properties. The detection of cyanopolyynes and their probable coexistence with grains implies that a whole *range* of C_n chains, where n may become very large, may exist in the ISM. Douglas (1977) has suggested that C_n chains may be resonsible for the diffuse interstellar lines. Whether or not they are it seems certain that these chains may be ubiquitous. The interventions of H and N atoms will chop the chains giving them neat ends (i.e. either $\cdots C \equiv C - H$ or $\cdots C \equiv C - C \equiv N$) and large dipole moments which allow them to be detected by radio techniques. The results suggest very strongly that the species: C_n, C_nH, C_nN, HC_nH and NC_nN (and perhaps C_nO, etc.) must be very abundant and indeed the detection of the cyanopolyynes may only be the tip of the iceberg as they are the only ones which we can at present detect readily. Species such as HC_nN will be rather more difficult to detect in warmer clouds due to the line dilution caused by the excitation of the vibrational degrees of freedom (Fig. 22). Hot molecules flex so much that effectively they are no longer linear

and thus lose the linear advantage by populating the bending vibrational (third rotational) degree of freedom.

Such large molecules may not be so readily photodissociated as the energy of electronic excitation will tend to distribute evenly along the chain rather than concentrate in a particular part of the molecule. At low pressures these chains may be kinetically stable as entropy related geometric factors stabilize them against chemical reaction.

The existence of such species in the ISM may have optical repercussions. Simple free electron theory indicates that they should have large electronic transition coefficients. The bands should leak down to longer and longer wavelengths as the chains increase in length. The effect on the optical transmission of the ISM may well be significant especially if it is realised that resonant and non-resonant (Rayleigh) scattering may be important. The wavelength dependence of the scattering will be a complex convolution due to an ISM filled with a whole range of molecules including a significant proportion of chains.

The polarization of light by passage through the ISM has been explained by scattering due to needle shaped grains aligned by interstellar magnetic fields. It is possible that if significant numbers of even-n C_n chains exist in space they may align because they may be paramagnetic. Although the ground state of C_2 is diamagnetic the $a^3\Pi$ excited state is only 716.24 cm^{-1} higher (Ballik and Ramsay, 1963). The separation between the states is likely to be smaller or even inverted in longer chains and there is thus a possibility of a simple strong mechanism for alignment by interstellar magnetic fields of an interstellar species with optical properties that may significantly affect the interstellar extinction.

There are a few experimental clues that indicate that there is something special about the chemistry of chain molecule formation. For instance it is well known that C_n molecules exist in the vapour over carbon at high temperature and that the proportion of longer chains increases with temperature above 2000 K (Chupka and Inghram, 1955). It would also appear that there is an alternation in the stability as the chains with odd n are more stable than adjacent chains with n even (Pitzer and Clementi, 1959). An observation which at the time was unique was made by Schüler and Reinbeck (1951) who observed the so-called T spectrum emitted from discharges in hydrocarbons. The bands were particularly strong from diacetylene and acetylene discharges. The carrier was positively identified as the ion HC≡C—C≡CH$^+$ by Callomon (1956). Recently cyanopolyynes have been produced in discharges by Winnewisser *et al.* (1980). It is certainly very likely that such molecules are among the *primary* gaseous species produced in Urey-Miller type experiments and do not survive the secondary reactions that occur, particularly in the condensed phase.

The properties of interstellar grains have been reviewed by Aanestad and Purcell (1973).

The diffuse interstellar lines

As far as the diffuse interstellar lines are concerned it is not at all clear what the right mode of attack might be, if indeed there is one. Certainly even better spectra are needed and efforts in this direction are continually being made.

Thus accurate relative intensities, line widths and frequencies together with higher resolution of the detailed line shapes will be invaluable. With the best data available it may be possible for someone to pick up the crucial lead and solve this 50 year old problem. It is not clear whether laboratory experiments are worthwhile. If the lines belong to absorbers trapped in grain matrices then the problem depends not only on the trapped species but also on the grain material which causes matrix line shifts. It is of course possible that the lines belong to some extremely weak optical resonances of grains or free molecules which only become detectable at colossal path length which would be impossible to emulate in a terrestrial laboratory.

Chemistry

The significance of the clear evidence that objects such as IRC + 10216 are pumping molecules, and probably grains as well, into the ISM needs to be clarified. Such objects have only become detectable with the improvement of infrared techniques and it is important to determine how widespread they are and whether they can account for the huge quantities of molecules observed.

There are several arguments to suggest that circumstellar shell formation is not of primary importance (Dalgarno and Black, 1976). At present there do not seem to be enough stars around pumping out enough material. In addition the lifetime of an average unshielded molecule is ~ 1000 years and this is not long enough for molecules to survive the journey to the cold regions where they are seen.

The exciting discovery of a wide range of typical interstellar molecules in cool stars such as IRC + 10216 suggests that these arguments should not go unquestioned.

The significant amounts of carbon, oxygen and nitrogen must have been formed in stars and so there must have been enough of them around at some time to form the amount observed. One farily well accepted mechanism is that grains are formed in circumstellar shells and it now appears that molecules form at roughly the same time. Having been formed simultaneously they may take part in a symbiotic protection-formation cycle during various stages of molecular cloud evolution. The scenario that follows from this is one in which the interplay of radiation induced gas-phase reactions and grain-surface catalysis are secondary processes rather than the primary formation route.

A curious inconsistency in the presently accepted chemical schemes is the fact that the only viable mechanism for the formation of H_2, *the* most important molecule of all, is via grain-surface catalysis (McCrea and McNally, 1960; Hollenbach and Salpeter, 1971) and yet for *all* other molecules, gas-phase processes are invoked. It is possible that in some regions, such as a secondary halo outside the HII region, a process such as

$$H^* \xrightarrow{\quad H \quad} H_2^+ \xrightarrow{\quad H \quad} H_2$$
$$+e^- \qquad\qquad +H^+$$
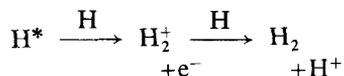
might occur as all excited H atoms with $n \geqslant 3$ have enough energy to form H_2^+. Indeed the detection of atoms with $n \sim 100\text{–}300$ indicates atom diameters of $\sim 1/100$ mm† implying colossal cross-sections and perhaps not insignificant lifetimes for the H* atoms to filter down below the $n = 3$ level.

Radiative association processes are at present being invoked as possible mechanisms for the formation of large molecules. It is likely that the more complex conditions that occur during collisions in larger molecules should allow the collision complexes longer time to dissipate excess kinetic energy, initially throughout the molecule, and then via the many more accessible molecular emission routes. It may in the future be possible to study these types of process directly. Smith and Adams (1981) have obtained circumstantial data on these types of processes.

Other molecule formation scenarios

One question of importance to interstellar chemistry relates to the efficiency with which stars and planets form. These processes are thought to be very inefficient and if so, one really needs to know just how inefficient they are and in what form the waste material is

† $a_n = 0.529n^2$ Å $\therefore \sim a_{300} \sim 10^5$ Å $\sim 10^{-2}$ mm where a_n is the nth Bohr orbit.

left. There most certainly will be molecules formed by a sort of *Damp Squib Mechanism* of interstellar molecule formation.

There are many other ways in which the ISM may be coerced into forming molecules. An interesting possibility is that they may form along accretion wakes caused by the passage of stars or star clusters through the ISM. Stars may be able to create the required density/temperature conditions for these reactions to proceed efficiently in interstellar clouds and leave a trail of molecules in their wake.

At the end of this review one might ask 'what, if anything, have all these recent discoveries got to do with the question of Life, the Universe and Everything that the mice wished to understand' (Adams, 1979)? Well, although we are still having to think about it, there is at least one curious observation that can be made about bio-emotive molecules such as glycine. It now seems certain that such molecules are formed in circumstellar shells and ejected into the ISM. A presently accepted view is that these molecules are then destroyed and gas phase or grain surface processes produce them a *second* time in the centres of cold, dense clouds. The planetary specialists suggest that they are then destroyed yet again during planet formation where they are synthesized for a *third* time in the earth's prebiotic secondary atmosphere via Urey-Miller type processes (Keosian, 1968). Why should these molecules be made three times? Can we be quite sure that the molecules are destroyed and in particular is it possible that the molecules can indeed survive either one or both the processes of star cloud migration and planet formation?

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