

though the Henderson ore is relatively poor in Re, we expect that the Re-Tc fraction obtained from our standard ore sample will contain 100 g of this metal. Thus, if  $10^7$  atoms of  $\text{RuX}^-$  can be detected in the pure "Tc" sample prepared from this fraction, relative abundances  $\text{RuX}^-/\text{Re} = 10^{-17}$  can be measured. This implies a sensitivity to an average  $\text{RuX}^-$  concentration in the earth's crust at 1 part in  $10^{26}$ .

*Note added in proof:* Measurements of the uranium content of samples of Henderson ore and ore concentrate give  $U = 11.8$  ppm and  $\tilde{U} = 1.8$  ppm. Our background estimates in this report should be reduced accordingly.

G. A. COWAN

Los Alamos National Laboratory,  
Los Alamos, New Mexico 87545

W. C. HAXTON

Department of Physics,  
Purdue University,  
West Lafayette, Indiana 47907,  
and Los Alamos National Laboratory

#### References and Notes

- R. Davis, Jr., D. S. Harmer, K. C. Hoffman, *Phys. Rev. Lett.* **20**, 1205 (1968); R. Davis, Jr., *Proc. Int. Conf. Neutrino Phys. Astrophys. Moscow* **2**, 99 (1969); \_\_\_\_\_ and J. M. Evans, *Proc. 13th Int. Cosmic Ray Conf.* **3**, 2001 (1973); R. Davis, Jr., in *Proceedings of an Informal Conference on the Status and Future of Solar Neutrino Research*, G. Friedlander, Ed. (BNL Report 50879, Brookhaven National Laboratory, Upton, N.Y., 1978), p. 1.
- J. N. Bahcall *et al.*, *Phys. Rev. Lett.* **45**, 945 (1980).
- R. T. Rood, in *Proceedings of an Informal Conference on the Status and Future of Solar Neutrino Research*, G. Friedlander, Ed. (BNL Report 50879, Brookhaven National Laboratory, Upton, N.Y., 1978), p. 175; J. N. Bahcall and R. L. Sears, *Annu. Rev. Astron. Astrophys.* **10**, 25 (1972).
- V. A. Lubimov *et al.*, *Phys. Lett. B* **94**, 266 (1980).
- F. Reines, H. W. Sobel, E. Pasierb, *Phys. Rev. Lett.* **45**, 1307 (1980).
- V. A. Kuzmin, *Sov. Phys. JETP* **22**, 1051 (1966); J. N. Bahcall *et al.*, *Phys. Rev. Lett.* **40**, 1351 (1978).
- R. D. Scott, *Nature (London)* **264**, 729 (1976); R. Davis, Jr., in *Proceedings of a Conference on the Ancient Sun*, J. A. Eddy, Ed. (Pergamon, New York, in press); G. S. Hurst *et al.*, *Phys. Today* **33** (No. 9), 24 (1980).
- R. S. Raghavan, *Phys. Rev. Lett.* **37**, 259 (1976).
- M. S. Freedman *et al.*, *Science* **193**, 1117 (1976).
- W. C. Haxton and G. A. Cowan, *ibid.* **210**, 897 (1980).
- J. N. Bahcall, *Rev. Mod. Phys.* **50**, 881 (1978).
- J. K. Rowley, B. T. Cleveland, R. Davis, Jr., W. Hampel, T. Kirsten, *Geochim. Cosmochim. Acta Suppl.* **13**, 45 (1980).
- However, resonance fluorescence techniques appear promising (T. W. Hansch and W. M. Fairbank, Jr., private communications).
- H. J. Kim, R. L. Robinson, C. H. Johnson, *Nucl. Phys. A* **167**, 65 (1971); C. F. Moore *et al.*, *Phys. Rev.* **141**, 1166 (1966).
- C. A. Goulding and C. C. Foster, private communications; C. D. Goodman *et al.*, *Phys. Rev. Lett.* **44**, 1755 (1980). These techniques could also be of great value in eliminating existing uncertainties in the capture rates for  $^{71}\text{Ga}$  and  $^{81}\text{Br}$ .
- G. A. Cowan and W. C. Haxton, in preparation.
- We stress that these results are not inconsistent with anything known about neighboring nuclei. The  $5/2^+ \rightarrow 7/2^+$  and  $5/2^+ \rightarrow 3/2^+$  transitions from  $^{97}\text{Mo}$  can be viewed naively as the conversion of a  $g_{7/2}$  neutron, coupled to an inert even-even core probably not too different from the  $2^+$  state in  $^{96}\text{Mo}$ , to a  $g_{3/2}$  proton. The strong transitions in  $^{95}\text{Ru}$  and  $^{99}\text{Ru}$  are fundamentally different: they include holes, not particles, coupled to even-even cores. One analogous transition, however, is the  $5/2^+ \rightarrow 7/2^+$  decay of  $^{95}\text{Zr}$ , with  $\log(ft) = 7.0$  also large.
- W. C. Haxton, *Nucl. Phys. A* **367**, 517 (1981).
- B. Mason, *Principles of Geochemistry* (Wiley, New York, ed. 3, 1966).
- A. Gunow and G. Kullerud, private communications.
- We have also considered the backgrounds produced in type II supernovae of neighboring stars [see reference 12 in (10)].
- S. R. Wallace, W. B. MacKenzie, R. G. Blair, N. K. Muncaster, *Econ. Geol.* **73**, 325 (1978).
- D. E. Ranta, W. H. White, A. D. Ward, R. E. Graichen, M. W. Ganster, D. R. Stewart, in *Studies in Field Geology*, R. C. Epis and R. J. Weimer, Eds. (Colorado School of Mines, Golden, 1976), p. 477.
- This represents less than 10 percent of the mine's daily production.
- K. B. Lebedev, *The Chemistry of Rhenium* (Butterworths, London, 1962), p. 36.
- One might make this argument stronger: as the uranium content is expected to vary throughout the ore body, the slope of the (linear) graph of the  $^{96}\text{Tc}$  concentration versus  $^{99}\text{Tc}$  will measure all uranium-induced backgrounds. This may require a demonstration that the molybdenite crystal size is not also correlated with the uranium content.
- E. J. Öpik, *Contrib. Armagh Obs. No. 9* (1953); W. A. Fowler, *Nature (London)* **238**, 24 (1972).
- F. W. W. Dilke and D. O. Gough, *Nature (London)* **240**, 262 (1972).
- R. N. Cahn and S. L. Glashow, *Science* **213**, 607 (1981).
- We thank T. Bowles, R. Davis, Jr., A. Gunow, R. Kamilli, L. McHugh, D. Ranta, G. J. Stephenson, Jr., A. Turkevich, A. Wahl, and W. White for helpful discussions. We are particularly indebted to the Amax Corporation for its help and to G. Kullerud for his advice on the geochemistry of molybdenite. This work was partially supported by the Department of Energy and by the National Science Foundation (grant PHY-8021272).

9 September 1981

## Search for Interstellar Superheavy Hydrogen

**Abstract.** Models for fundamental physical interactions allow for the existence of stable or nearly stable elementary particles much heavier than the proton. Stellar spectra were searched for a positively charged superheavy particle,  $X^+$ , which, with a bound electron, should appear as apparently superheavy neutral hydrogen in the interstellar medium. An upper limit for the abundance of  $X$  relative to normal hydrogen in the line of sight toward the bright star  $\gamma$  Cassiopeiae is  $2 \times 10^{-8}$ .

Cahn and Glashow (1) described how models for unified fundamental interactions might allow for the existence of essentially stable superheavy elementary particles. If a positively charged superheavy,  $X^+$ , exists, it will appear chemically as very heavy hydrogen when an electron is bound to it. Smith and Bennett (2) placed an upper limit in terrestrial water samples for the fractional abundance of  $X$  relative to H of  $n(X)/n(H) < 10^{-21}$  if the mass of  $X$  lies between 6 and 350 proton masses; apparently this limit can be reduced by orders of magnitude in the near future. Here we report a search for  $X$  in the interstellar medium. Our upper limit is much greater than the terrestrial bound, but our search may nevertheless be useful since we examine a different portion of the universe and our result is valid for any particle mass greater than about four times that of the proton.

Our search procedure is similar to that used to discover interstellar deuterium (3, 4); that is, we searched for resonance absorption lines from the ground electronic state of hydrogen-like  $X$  atoms in the spectrum of background stars. Because both hyperfine splitting (5) and the energy shifts which result from the finite size of the nucleus are likely to be small (6), the electronic isotope shift of  $X$  is expected to be dominated by the difference in the reduced mass between  $X$  and H. Therefore, we expect the electronic spectrum of superheavy hydrogen to be similar to that of hydrogen only with an

isotope shift that corresponds to an apparent Doppler motion of  $-160 \text{ km sec}^{-1}$  (for an infinitely heavy nucleus) instead of  $-80 \text{ km sec}^{-1}$  for deuterium.

Interstellar Lyman  $\alpha$  absorption is generally so broad that it is usually impossible to detect lines only  $160 \text{ km sec}^{-1}$  from the line center, and it is necessary to use higher order Lyman lines. Consequently, the only suitable data are from the Copernicus satellite, which was sensitive to wavelengths shortward of  $1200 \text{ \AA}$ . Since this satellite is no longer operational, we must use data from previous sensitive searches for interstellar deuterium. We require observations of stars (i) that are rapidly rotating so that blends with stellar lines are not important, (ii) that are sufficiently bright that a high signal-to-noise ratio was obtained, (iii) that are sufficiently nearby that there is no high-velocity normal hydrogen in the line of sight, and (iv) yet are sufficiently far that the amount of hydrogen in the line of sight to the star is appreciable. From the results given in (7), the most suitable star for our search is  $\gamma$  Cassiopeiae, where the interstellar medium in the line of sight is reasonably well understood (8).

From the figure published in the paper of Vidal-Madjar *et al.* (9) and the standard procedure for placing upper limits from Copernicus data (10), we estimate a 3 standard deviation upper limit to the equivalent width of the Lyman  $\beta$  line of superheavy hydrogen toward  $\gamma$  Cassiopeiae of  $1.5 \text{ m\AA}$ . Using the same oscillation

tor strength for this electronic transition as for normal hydrogen and assuming that the line is optically thin, we apply standard procedures (11) to derive an upper limit to the column density of X atoms of  $2.0 \times 10^{12} \text{ cm}^{-2}$ . Since the column density of H is  $1.0 \times 10^{20} \text{ cm}^{-2}$  (9), this implies that  $n(X)/n(H) < 2 \times 10^{-8}$ . This limit obtains for all hypothetical singly charged superheavies with isotope shifts greater than  $120 \text{ km sec}^{-1}$  or to particles with masses greater than about four times that of the proton.

M. JURA

Department of Astronomy, University of California, Los Angeles 90024

D. G. YORK

Princeton University Observatory, Princeton, New Jersey 08544

#### References and Notes

1. R. N. Cahn and S. L. Glashow, *Science* **213**, 607 (1981).
2. P. F. Smith and J. R. J. Bennett, *Nucl. Phys. B* **149**, 525 (1979).
3. J. B. Rogerson and D. G. York, *Astrophys. J. Lett.* **186**, L95 (1973).
4. D. G. York and J. B. Rogerson, *Astrophys. J.* **203**, 378 (1976).
5. M. Ford, F. S. Tomkins, J. K. Brody, M. Hamermesh, *Phys. Rev.* **82**, 406 (1951).
6. L. Wilets, D. L. Hill, K. W. Ford, *ibid.* **91**, 1488 (1953).
7. C. Laurent, A. Vidal-Madjar, D. G. York, *Astrophys. J.* **229**, 923 (1979).
8. R. Ferlet *et al.*, *ibid.* **242**, 576 (1980).
9. A. Vidal-Madjar, C. Laurent, R. M. Bonnet, D. G. York, *ibid.* **211**, 911 (1977).
10. E. B. Jenkins, J. F. Drake, J. B. Rogerson, L. Spitzer, D. G. York, *Astrophys. J. Lett.* **181**, L122 (1973).
11. L. Spitzer, *Physical Processes in the Interstellar Medium* (Wiley, New York, 1978).
12. Partly supported by the National Aeronautics and Space Administration and the National Science Foundation. M.J. is an Alfred P. Sloan Foundation fellow.

23 November 1981

## Queuine, a Modified Base Incorporated Posttranscriptionally into Eukaryotic Transfer RNA: Wide Distribution in Nature

**Abstract.** *Queuine, a modified base found in transfer RNA, appears to be a new dietary factor because (i) previous studies have shown that mice require it for the expression of queuine-containing transfer RNA's, but apparently do not synthesize it, and (ii) significant amounts of free queuine are present in common plant and animal food products.*

Unlike other transfer RNA (tRNA) modifications, queuine is synthesized first as a base which then is incorporated irreversibly (in an exchange reaction in which guanine is removed) into mature tRNA by the enzyme guanine: queuine tRNA transglycosylase (1-3). Queuine is found exclusively in the first position of the anticodon in tyrosine tRNA, histidine tRNA, asparagine tRNA, and aspartic acid tRNA (4). Queuine appears to be the immediate precursor of queuine-containing tRNA ([Q+]tRNA) in mammals (1, 2, 5-7) and has been identified as the free base in animal serum, amniotic fluid, and extracts of *Drosophila melanogaster* (8, 9). However, animals apparently do not synthesize queuine de novo, since germ-free mice on a defined diet do not synthesize [Q+]tRNA unless enabled to do so by any one of the following: loss of germ-free state, consumption of a normal Laboratory Chow diet, parental injection of queuine, or addition of queuine or [Q+]tRNA to the defined diet (6). While both gut flora and diet enable mice to synthesize [Q+]tRNA, it is not known whether this results from the feeding of queuine or its precursor. We report here that free queuine is widely distributed among eukaryotes, with significant levels present in plant and animal products common to the human diet (Table 1).

The data in Table 1 were obtained by means of a whole cell assay; when cultured in serum-free medium, the synthesis of [Q+]tRNA by L-M cells depends on queuine addition to the medium (1, 9, 10). Only queuine has been demonstrated to give this response (9, 10); the nucleoside of queuine, queuosine, is not active in the assay (11). The L-M cell

assay, however, is tedious, unsuitable for more than a few samples at a time, and subject to nonspecific inhibition (9). Therefore, an additional chemically based assay was developed, on the basis of gas chromatography-mass spectrometry with selected ion monitoring (12). The abundant ions of  $m/z$  379, 380, which are highly characteristic of the 7-deaza-guanine nucleus (8) were monitored (Fig. 1). This method provides rigorous chemical evidence for the presence of queuine in human amniotic fluid, extracts of *D. melanogaster*, and coconut water. Previously, queuine had been positively identified only in bovine amniotic fluid (8). The isolates from *D. melanogaster* and coconut water were sufficiently pure to permit acquisition of full-scan mass spectra ( $M = m/z$  709;  $M-CH_3 = 694$ ) (8). Seven amniotic fluids from normal human pregnancies (16 to 28 weeks gestation) were estimated to contain queuine concentrations ranging from 2 to 84 ng/ml (mean = 29 ng/ml), on the basis of selected ion recording peak areas, referenced to standard queuine samples. There was no apparent relation of queuine concentration to gestation time (13).

Our data appear to be sufficient to explain the contribution of diet to [Q+]tRNA formation. However, diet must provide queuine both directly and indirectly (after salvage of free base from [Q+]tRNA), because the ability of germ-free mice to use dietary [Q+]tRNA for endogenous [Q+]tRNA synthesis implies a salvage mechanism. Salvage also would explain the contribution of gut flora to [Q+]tRNA formation in mice,

Table 1. Queuine content of natural products. Queuine was estimated from the appearance of [Q+]tRNA<sup>ASP</sup> in the L-M cell line, with authentic queuine as a standard (1, 9, 10), and a millimolar extinction coefficient for queuine of 10.5 at 260 nm in H<sub>2</sub>O (22). Most solid materials were blended as a 10 percent (weight to volume) aqueous slurry (tomato was blended whole) and centrifuged and then the supernatant was assayed. *Drosophila melanogaster* was extracted as described in (9). Milk was centrifuged and the skim portion was assayed. For multiple samples (number in parentheses) the range of values is given. Many products were negative for queuine by the L-M cell assay (23).

Source	Amount
Bovine amniotic fluid (third trimester) (3)	2300 to 3600 ng/ml
<i>Drosophila melanogaster</i> * (wild type and mutants) (15)	0 to 1100 ng/g
Coconut water (ripe) (5)	87 to 530 ng/ml
Bovine pineal body	300 ng/g
Wheat germ	190 ng/g
Bovine seminal vesicle (adult)	110 ng/g
Bovine testicle (adult)	58 ng/g
Bovine serum (fetal) (2)	33 to 54 ng/ml
Tomato (fresh, ripe)	21 ng/g
Bovine milk (whole and skim) (2)	16 to 17 ng/ml
Bovine serum (calf)	14 ng/ml
Bovine milk (evaporated skim, canned)	12 ng/ml
Yogurt (commercial and homemade) (2)	4 to 6 mg/g
Goat milk (fresh)	3 ng/ml
Goat milk (evaporated, canned)	1 ng/ml
Human milk	1 ng/ml

\*The values for *D. melanogaster* are derived from previously published data [figure 1 in (9)].