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OPERATIONAL FREQUENCY STABILITY OF RUBIDIUM AND CESIUM FREQUENCY STANDARDS

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OPERATIONAL FREQUENCY STABILITY OF RUBIDIUM AND CESIUM FREQUENCY STANDARDS

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INTRODUCTION

In the course of testing various rubidium and cesium frequency standards under operational conditions for use in NASA tracking stations, about 55 unit-years of relative frequency measurements for averaging times from 10 to 10⁷ s have been accumulated at Goddard Space Flight Center (GSFC). Statistics on the behavior of rubidium and cesium standards under controlled laboratory conditions have been published by many institutions (see, for example, ref. 1), but it was not known to what extent the lesser controlled environments of NASA tracking stations affected the performance of the standards. The purpose of this report is to present estimates of the frequency stability of rubidium and cesium frequency standards under operational conditions based on the data accumulated at GSFC.

Table 1.—Atomic Frequency Standards Used in Experiments

Serial no. or designation	Manufacturer
Rb 107	Varian Associates
Rb 136	Varian Associates
Rb 138	Varian Associates
Cs 110	Hewlett-Packard Co.
Cs 136	Hewlett-Packard Co.
Cs 137	Hewlett-Packard Co.
Cs 138	Hewlett-Packard Co.
Cs 139	Hewlett-Packard Co.
Cs 152	Hewlett-Packard Co.
Cs 182	Hewlett-Packard Co.
Cs 185	Hewlett-Packard Co.
Cs 186	Hewlett-Packard Co.
HM:	
H-10 no. 2	Varian Associates
NX-1	(a)

^a An experimental hydrogen maser developed at GSFC. See ref. 2.

DATA DESCRIPTION

The three rubidium gas cells (designated Rb) and nine cesium beam frequency standards (Cs) on which the measurements were made, as well as the two hydrogen masers (HM) used as references for many of the tests, are listed in table 1 along with their serial numbers or designations and their manufacturers. During the tests the standards were kept in a laboratory at GSFC. Except for the shielding built into the standards themselves, there was no special control of the ambient magnetic, electric, vibration, and temperature conditions. The ambient magnetic and electric conditions were typically noisy. The standards were driven by ac power and were in no way isolated by transformers. Vibration from nearby air conditioning equipment and from trucks at a nearby loading platform was not shielded in any way. The ambient temperature was typically between 298 and 303 K. There

were, however, several brief excursions to temperatures as low as 291 K and as high as 313 K due to equipment problems. These conditions are less controlled than those in the NASA tracking stations. Hence the stabilities of the standards when operating in the tracking stations should be at least as good as the stabilities calculated in this paper.

The measurements made on the standards consisted of average relative frequency measurements for varying averaging times. In some of the data sets, average relative frequency measurements were missing or were bad because of ac power failure or recorder failure. All such points were a posteriori linearly interpolated from the nearest earlier (in epoch time) good average relative frequency measurement and the nearest later (in epoch time) good average relative frequency measurement.

The total number of measurements made for all types of data used in this report are given in table 2. Data sets are said to be of the same "type" when the following parameters are the same for

Table 2.-Average Relative Frequency Data Sets

	Type of data			Number of	Number of measurements							
Test unit	Reference unit	Averaging time $ au_0$, s	Dead time d, s	data sets m	Total ^a	Interpolated						
Rb	Rb	3 600	0.0	2	3 090	0						
Rb	Cs (10-s TC)	10	2.3	1	1 076	18						
Rb	Cs (10-s TC)	100	2.2	1	538	1						
Rb	Cs (10-s TC)	1 000	2.7	1	223	0						
Rb	HM	10	2.3	13	8 473	67						
Rb	HM	100	2.2	10	6 405	16						
Rb	HM	1 000	2.7	9	5 126	15						
Rb	HM	3 600	.0	7	13 320	308						
Cs	Cs	3 600	.0	3	8 851	263						
Cs (10-s TC)	HM	10	.2	8	4 841	0						
Cs (10-s TC)	HM	100	.2	8	4 871	11						
Cs (10-s TC)	HM ·	1 000	.2	8	4 787	25						
Cs (60-s TC)	HM	10	.2	8	4 634	0						
Cs (60-s TC)	HM	10	2.3	3	1 904	2						
Cs (60-s TC)	HM	100	.2	8	4 706	0						
Cs (60-s TC)	HM	100	2.2	3	2 496	0						
Cs (60-s TC)	HM	1 000	.2	8	4 804	3						
Cs (60-s TC)	HM	1 000	2.7	1	692	0						
Cs	HM	3 600	.0	13	37 404	1391						
Cs	НМ	604 800	.0	. 1	88	6						

TC = time constant.

^aTotal number of measurements for all m data sets, including the interpolated measurements.

each set: test unit, 1 reference unit, duration or averaging time τ_0 of each average relative frequency measurement, and dead time d between successive measurements (that is, the time during which no measurement was taken). The servo time constants are indicated only for the cesium standards and only when $\tau_0 \leq 1000$ s. The difference in effect of a 10- and 60-s time constant for $\tau_0 \geq 3600$ s can be neglected because the time constants in such cases are too small with respect to τ_0 to have an appreciable effect. The rubidium standards tested all have a fixed servo time constant which is on the order of 1 ms.

Neither temperature effects nor long-term frequency drift was removed from the data before analysis because the object of the tests was to measure the stability of the frequency standards under operational conditions, where both temperature fluctuations and long-term frequency drift are present.

STATISTICAL ANALYSIS

Let there be given a set of m identical test frequency standards and a set of m identical reference frequency standards. Let $\phi_n(t)$, $1 \le n \le m$, denote the instantaneous fluctuations (measured in time units) of the epoch time output of the nth test standard compared to the epoch time output of the nth reference standard. Let $y_n(t)$ be the instantaneous (fractional) frequency fluctuation of the nth test standard compared with the nth reference standard; i.e.,

$$y_n(t) \equiv \frac{d\phi_n(t)}{dt} \tag{1}$$

Let $\overline{y}_n(t)$ be the average relative (fractional) frequency fluctuation of the *n*th test standard compared with the *n*th reference standard:

$$\vec{y}_{n}(t) = -\frac{1}{\tau} \int_{t}^{t+\tau} y_{n}(t) dt = \frac{\phi_{n}(t+\tau) - \phi_{n}(t)}{\tau}$$
 (2)

The constant τ is called the averaging time of $\overline{y}(t)$. The Allan standard deviation $\sigma(2, T, \tau)$ of the frequency fluctuations of the set of test standards compared with the set of reference standards is defined to be (ref. 3)

$$\sigma(2, T, \tau) = \sqrt{\frac{1}{m} \sum_{n=1}^{m} \langle \operatorname{var} \left[\overline{y}_{n}(t+T) - \overline{y}_{n}(t) \right] \rangle}$$
 (3)

where the symbol $\langle \rangle$ denotes infinite epoch time average. The analysis of all data listed in table 2 consisted in the calculation of an estimate, which is denoted by $s(2, T, \tau)$, of $\sigma(2, T, \tau)$ in the following manner.

Taking any type of data from table 2, let the number of average relative frequency measurements in the *n*th data set, $1 \le n \le m$, be m_n . Denote this *n*th set of average relative frequency measurements

¹Although there are sometimes significant differences in the frequency stabilities of various rubidium standards, the three rubidium standards listed in table 1 all had mutually close stabilities. For this reason, these rubidium standards will be considered to be identical. Because the nine cesium standards listed in table 1 all had mutually close stabilities, they too will be considered to be identical.

by $\{\overline{y}_n(i)\}_{i=1}^{m_n}$. For $i=1, 2, \ldots, m_n-1$, denote the variance of the two average relative frequency measurements $\overline{y}_n(i)$ and $\overline{y}_n(i+1)$ by $v_n(i)$:

$$v_n(i) = \frac{[\bar{y}_n(i+1) - \bar{y}_n(i)]^2}{2} \tag{4}$$

The square root of the average over both i ($1 \le i \le m_n - 1$) and n ($1 \le n \le m$) of these $v_n(i)$ is the desired estimate of $\sigma(2, \tau_0 + d, \tau_0)$:

$$s(2, \tau_0 + d, \tau_0) = \sqrt{\frac{\sum_{n=1}^{m} \sum_{i=1}^{m_n-1} v_n(i)}{\sum_{n=1}^{m} (m_n - 1)}}$$
 (5)

From the original data sets $\{\overline{y}_n\}_{i=1}^{m_n}$, $1 \le n \le m$, new data sets with averaging time $\tau_1 = 2\tau_0$ and dead time d (assumed small with respect to τ_0) can be approximated by defining

$$\overline{y}_n(i;1) = \frac{\overline{y}_n(i+1) + \overline{y}_n(i)}{2} \tag{6}$$

 $i=1, 2, \ldots, m_n-1$ and $n=1, 2, \ldots, m$. Denote the variance of $\overline{y}_n(i;1)$ and $\overline{y}_n(i+2;1)$ by $v_n(i;1)$:

$$v_n(i;1) = \frac{[\overline{y}_n(i+2;1) - \overline{y}_n(i;1)]^2}{2}$$
 (7)

 $i = 1, 2, ..., m_n - 3$ and n = 1, 2, ..., m. Estimate $\sigma(2, \tau_1 + d, \tau_1)$ by²

$$s(2, \tau_1 + d, \tau_1) = \sqrt{\frac{\sum_{n=1}^{m} \sum_{i=1}^{m_n - 3} v_n(i; 1)}{\sum_{n=1}^{m} (m_n - 3)}}$$
 (8)

Let k be the exponent of the largest power of 2 contained in any of the m_n , $1 \le n \le m$. For $j=2,3,\ldots,k-1$, the data set $\{\overline{y}_n(i;j)\}_{\substack{i=1\\i=1}}^{m_n-2^j+1}$ with averaging time $\tau_j=2^j\tau_0$ and dead time d is successively calculated from the data set $\{\overline{y}_n(i;j-1)\}_{\substack{i=1\\i=1}}^{m_n-2^{j-1}+1}$ by pairwise averaging:

$$\overline{y}_n(i;j) = \frac{\overline{y}_n(i+2^{j-1};j-1) + \overline{y}_n(i;j-1)}{2}$$
(9)

²Throughout this paper the convention is adopted that whenever a summand, e.g., $m_n - 3$ in $\sum_{n=1}^{m} (m_n - 3)$, is less than zero, it is treated as zero; and whenever a summation, e.g., $\sum_{i=1}^{m} v_n(i; 1)$, has an upper limit that is less than the lower limit, it also is treated as zero.

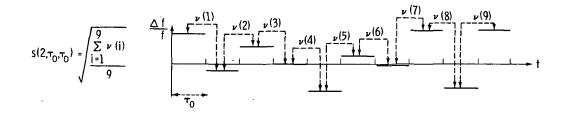
 $i=1,2,\ldots,m_n-2^j+1; n=1,2,\ldots,m; j$ fixed. Denote the variance of $\overline{y}_n(i;j)$ and $\overline{y}_n(i+2^j;j)$ by $v_n(i;j)$:

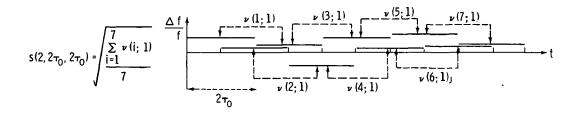
$$v_n(i;j) = \frac{[\overline{y}_n(i+2^j;j) - \overline{y}_n(i;j)]^2}{2}$$
 (10)

 $i = 1, 2, ..., m_n - 2^{j+1} + 1$ and n = 1, 2, ..., m. Estimate $\sigma(2, \tau_j + d, \tau_j)$ by

$$s(2, \tau_j + d, \tau_j) = \sqrt{\frac{\sum_{n=1}^{m} \sum_{i=1}^{m_n - 2^{j+1} + 1} v_n(i; j)}{\sum_{n=1}^{m} (m_n - 2^{j+1} + 1)}}$$
(11)

An example of this procedure for zero dead time is presented in figure 1. The quantity v represents the variance between the ordinates of the two lines to which the dotted line near v points.





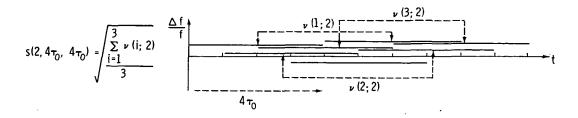


Figure 1.—Calculation of $s(2, \tau, \tau)$.

RESULTS

For each type of data listed in table 2 and for each averaging time $\tau_j = 2^j \tau_0$, $0 \le j \le k-1$ (τ_0 and k change with the type of data), the estimate $s(2, \tau_j + d, \tau_j)$ of $\sigma(2, \tau_j + d, \tau_j)$ was calculated.³ The results are presented in table 3 and figure 2 for all data involving a rubidium standard as either the test or the reference unit and in table 4 and figure 3 for the cesium versus cesium and cesium versus hydrogen maser data.

In order to use the data in tables 3 and 4 to estimate the frequency stability of the rubidium and cesium standards tested, rather than the relative frequency stability of a comparison of two of these standards or of a comparison of one of these standards to a hydrogen maser, the following procedure is used. Denote the Allan standard deviations of the test standard versus a hypothetical perfect standard, the reference standard versus a hypothetical perfect standard, and the test standard versus the reference standard by $\sigma_T(2, \tau + d, \tau)$, $\sigma_R(2, \tau + d, \tau)$, and $\sigma_{T\cdot R}(2, \tau + d, \tau)$, respectively. Because the variances $\sigma_T^2(2, \tau + d, \tau)$ and $\sigma_R^2(2, \tau + d, \tau)$ are linear functions (in fact, weighted integrals) of the respective power spectral densities of the test and reference standards (ref. 3), and because the power spectral density of the comparison of two frequency standards is the sum of the power spectral densities of each of the standards, the following relation occurs:

$$\sigma_{T,R}^{2}(2,\tau+d,\tau) = \sigma_{T}^{2}(2,\tau+d,\tau) + \sigma_{R}^{2}(2,\tau+d,\tau)$$
 (12)

For comparisons of two identical standards (rubidium standard versus rubidium standard and cesium standard versus cesium standard), $\sigma_R(2, \tau + d, \tau) = \sigma_T(2, \tau + d, \tau)$. Hence, from relation (12),

$$\sigma_T(2, \tau + d, \tau) = \frac{\sigma_{T-R}(2, \tau + d, \tau)}{\sqrt{2}}$$
 (13)

For all data for which a hydrogen maser was used as a reference, it is assumed that the instabilities of the maser were sufficiently small so as to have

$$\sigma_T(2, \tau + d, \tau) \approx \sigma_{T-R}(2, \tau + d, \tau) \tag{14}$$

The normalized standard deviation $\sigma_T(2, \tau, \tau)$ can be calculated from $\sigma_T(2, \tau + d, \tau)$ by the relation

$$\sigma_T(2, \tau, \tau) = \frac{\sigma_T(2, \tau + d, \tau)}{\sqrt{B_2(r, \mu)}} \tag{15}$$

where $B_2(r, \mu)$ is a bias function (defined in ref. 4); $r = (\tau + d)/\tau$; and μ , representing the type of noise of the standard for the fixed averaging time τ and fixed dead time d, is determined from

$$\sigma_{\tau}(2, \tau + d, \tau) \propto \tau^{\mu/2} \tag{16}$$

³The analysis was carried out by programs E00016 and E00036 of the GSFC Computer Program Library. Program E00016 is for input relative phase data; program E00036 is for input relative frequency data. Although program E00016 reads relative phase data as input, its output is the Allan standard deviation of relative frequency $s(2, \tau_j + d, \tau_j)$ defined in eqs. (5), (8), and (11). These two programs are based on a program written by David W. Allan of the National Bureau of Standards, Boulder, Colo.

Table 3.—Rubidium Standard Frequency Stability

The Reference with the contract of the contra	$\overline{}$																			_						_	
Type of data Type		$s(2, \tau + d, \tau), \\ \times 10^{-12}$	1.881	1.447	.877	.783	1.057	.917	.782	119.	.721	.829	.717	.857	1.011	2.633	2.783	2.691	2.368	1.867	1.740	1.784	1.629	1.637	1.700	2.788	4.549
Proper of data Proper of data Prope of data Prope of data Prope of data Prope of data			2.2	2.2	2.2	2.5	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.7	0	o.	0.	0.	0.	0.	o.	0.	0.	0.	0.	0.
Type of data Reference T, s d, s standard Standard Type of data Type of data Type of data Type of data Standard T, s d, s X10 ⁻¹² Test Reference T, s d, s X10 ⁻¹² Test Reference T, s d, s X10 ⁻¹² Test Reference T, s d, s Standard Total Test Te	f data	7, 8	3 200	6 400	12 800	25 600	1 000	2 000	4 000	8 000	16 000	32 000	64 000	128 000	256 000	3 600	7 200	14 400	28 800	27 600	115 200	230 400	460 800	921 600	1 843 200	3 686 400	7 372 800
Type of data Reference 7, s d, s (2, r + d, r) Runit (1, s) (2, r + d, r) Rb (3, s) (3, s) (1, s) Rb (3, s) (3, s) (3, s) 17, s (3, s) (3, s) (3, s) 18, s (3, s) (3, s) (3, s) 19, s (3, s) (3, s)	Type o	Reference unit					НМ									НМ		-									
Type of data Reference		Test					Rb									Rb											
Type of data Reference Type of data S(2, 7 + d, 7), and below the following and below the follow the follow the follow the following and below the		$s(2, \tau + 4, \tau), \times 10^{-12}$	2.023	1.405	.708	.387	4.015	2.736	2.092	1.553	1.310	1.466	1.950	22.844	29.558	39.497	31.205	5.432	5.673	2.404	1.945	1.125	12.655	8,568	5.308	3.877	2.587
Type of data Reference 7, s 4, s x 10 ⁻¹² Test Reference unit x 100 0.0 1.128 x 10 ⁻¹² Test Reference unit x 100 0.0 0.991 x 100 0.991 x 100 0.991 x 100			2.2	2.2	2.2	2.2	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.2	2.2	2.2	2.2	2.2
Type of data Reference T, S d, s X10 ⁻¹² Test Ref Munit S S S Test Ref Munit S S S S Test Ref Munit S S S S S S S S S	lata	7,8	3 200	6 400	12 800	25 600	1 000	2 000	4 000	8 000	16 000	32 000	64 000	10	20	40	08	160	320	640			100	200	400	800	1 600
Type of data Reference and the state of th	Type of c	Reference unit					Cs (10-s TC)							HM									HIM				
Type of data Reference T, s	!	Test					Rb				·			Rb									Rb				
Type of data Reference unit Rb		$x(2, 7+4, 7), \\ \times 10^{-12}$	1.128	.874	.991	1.254	1.481	1.542	1.493	1.559	1.893	2.211	32.092	26.099	18.610	12.680	9.782	6.584	4.452	3.902	5.389	10.868	9.972	7.746	5.708	4.177	2.723
Type of di Reference unit Rb Cs (10-s TC)			_																								
Reference of the control of the cont	data	7, S	3 600	7 200	14 400	28 800	57 600	115 200	230 400	460 800	921 600	1 843 200	10	20	40	08	160	320	640	1 280	2 560	5 120	100	200	400	800	1 600
Test unit Rb Rb	Type of	Reference unit	Rb										Cs (10-s TC)										Cs (10-s TC)				
		Test	Rb										Rb										Rb	_			

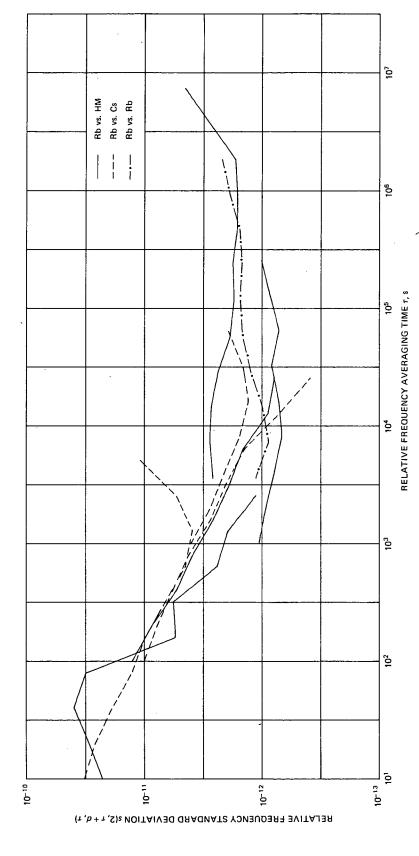


Figure 2.—Rubidium standard relative frequency stability.

Table 4.—Cesium Standard Frequency Stability

(2, 1 + d, 1).	×10-12	2,199	1.523	1.099	.790	290	435	370	000	. 343	.375	3.885	2.963	2.170	1.542	1.042	672	007		707	.340	1.777	1.249.	.923	.732	.615	.572	995.	.570	.556	.555	.590	.612	.464	.337	239	- - - - -		/01:	r i		
	d,s	0.2	7:	7	7	,				7.	-7	2.7	2.7	2.7	2.7	2.7	2.7	; ;	7:7	1:7	2.7	o.	0.	0.	0.	0:	0.	0.	0.	0.	0.	0.	0.	0.	0			•		?	•	1
ta	7,8	1 000	2 000	4 000	8 000	16,000	32 000	000 77	000 50	128 000	256 000	1 000	2 000	4 000	8 000	16 000	32 000	000 75	04 000	1.28 000	256 000	3 600	7 200	14 400	28 800	57 600	115 200	230 400	460 800	921 600	1 843 200	3 686 400	7 372 800	604 800	1 209 600	2 419 200	4 838 400	000 000	009 070 6	19 555 600		
Type of data	Reference unit	НМ										НМ	-									НМ												НМ								
	Test unit	Cs (60-s TC)	•									Cs (60-s TC)										ర												చ								
s(2, r+d, r)	×10 ⁻¹²	190	5.391	4.524	4.510	4 579	4 140	2 7 7	3.149	2.287	1.786	1.201	080'9	5.321	5.582	5 875	5 912	217.5	5.039	3.636	2.217	1.943	4.149	3.687	2.962	2.144	1.535	1.072	.760	.580	.595	5.699	5.535	4.840	3,690	2 750	1 931	1011	1.318	1.003	.883	
	d, s	0.2	?	7	7.	,	; c	; ‹	7. (7:	7:	7.	2.3	2.3	2.3	23		; ;	7.7	7.7	2.3	2.3	7.	7:	7.	7.	.2	7:	7.	7.	7.	2.2	2.2	2.2	2,2	, ,	, ,	7.7	7:7	7.7	7.7	1
	7, S	256 000	10	20	40	80	160	200	320	640	1 280	2 560	10	20	40	08	160	100	320	040	1 280	2 560	100	200	400	800	1 600	3 200	6 400	12 800	25 600	100	200	400	800	1 600	3 300	3 200	6 400	17 800	72 600	
Type of data	Reference unit		НМ										НМ										НМ	-								НМ										
	Test unit		Cs (60-s TC)										Cs (60-s TC)										Cs (60-s TC)									Cs (60-s TC)										
(+ p + + C)	x 10-12	1.825	1.266	.918	.728	200	248	2	6/5.	919.	.440	.333	.352	14.269	11.743	8 941	8089	0.00	4.721	3.732	1.886	1.484	1.740	6.014	4.332	3.090	2.166	1.616	1.218	.936	069	.388	1.934	1.413	1 073	838	900	671.	.6/4	.481	.459	
	d, s	0.0	0.	0.	0		2 0	? •	ب د	0.	0.	0.	0.	.2	.2	,	; c	, ,	7. 0	7:	7.	7	7.	7.	7.	.2	.2	7.	.2	7	7	.2	7	. 2	٠,	; ,	, c	4 (7, 6	7.	?!	
ta	7, S	3 600	7 200	14 400	28 800	57.600	115 200	007 011	230400	460 800	921 600	1 843 200	3 686 400	10	70	40	2 &	3 5	091	320	640	1 280	2 560	100	200	400	800	1 600	3 200	6 400	12 800	25 600	1 000	2 000	4 000	000	900 91	000 01	32 000	000 49	128 000	
Type of data	Reference unit	ی												НМ										НМ									НМ					•				
	Test unit	C)												Cs (10-s TC)	`						•			Cs (10-s TC)									Cs (10-5 TC)	\ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \								

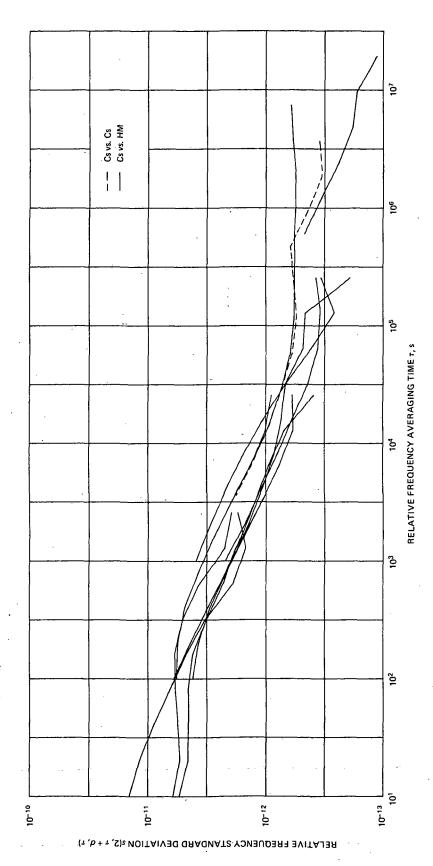


Figure 3.—Cesium standard relative frequency stability.

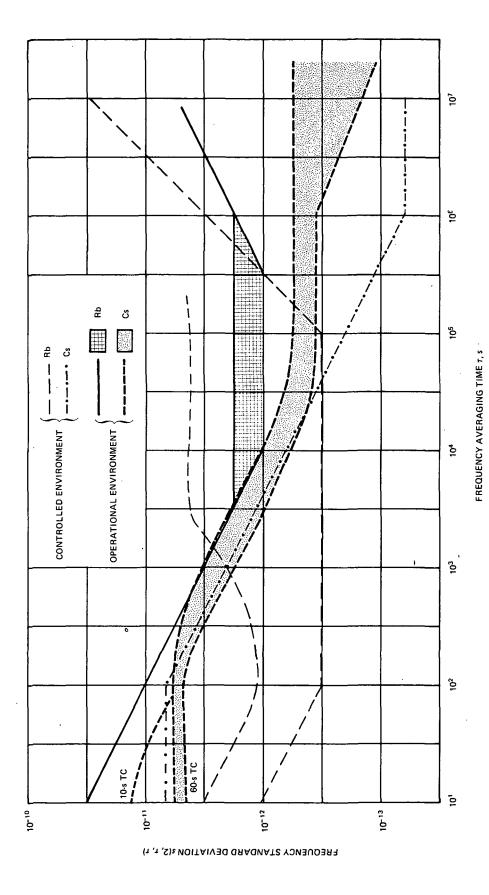


Figure 4.—Rubidium and cesium standard frequency stabilities.

It is of interest to note that for the r and μ of the data analyzed in this report, $B_2(r, \mu)$ differs from unity by less than 0.1 percent and can be ignored. Hence, for the data in this report,

$$\sigma_T(2, \tau, \tau) \approx \sigma_T(2, \tau + d, \tau)$$
 (17)

Of course, relation (17) is an exact equality whenever d = 0.

Using the estimates $s(2, \tau + d, \tau)$ of $\sigma_{T,R}(2, \tau + d, \tau)$ from figures 2 and 3 in relations (13) and (14) and using relation (17), the standard deviations $\sigma_T(2, \tau, \tau)$ of the rubidium and cesium standards tested can be estimated. These estimates of $\sigma_T(2, \tau, \tau)$ are presented in figure 4 as the "operational environment" curves. Also shown in figure 4 are curves taken from references 1 and 5 representing the performance of rubidium and cesium standards in a "controlled environment." By "controlled environment" is meant an experimental environment shielded from magnetic, electric, vibration, and temperature effects much more than the "operational" environment in which the data presented in figures 2 and 3 were taken. The upper curve for rubidium standards under a controlled environment in figure 4 is taken from reference 5 and represents the measured performance of Varian rubidium standards under controlled conditions. The lower curve for rubidium standards under a controlled environment and the curve for cesium standards under a controlled environment in figure 4 are taken from reference 1 and represent the measured performance of Hewlett-Packard rubidium and cesium standards under controlled conditions.

CONCLUSIONS

From figure 4 it is apparent that an operational environment degrades the performance of the rubidium standards (by up to one order of magnitude) for frequency averaging times between 10 and 10^3 s and that it degrades the performance of the cesium standards (by up to one order of magnitude) for frequency averaging times between 3×10^4 and 2×10^7 s. For all other averaging times in the range covered by the data in figure 4, the stabilities of the standards are not degraded by the operational conditions.

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Goddard Space Flight Center
National Aeronautics and Space Administration
Greenbelt, Maryland, December 22, 1971
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⁴That this was the case for the data presented in reference 1 was verified by private communication with G. M. R. Winkler. That the environment in which the data presented in reference 5 was taken was "controlled" is assumed.

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