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Radiative-lifetime and absolute-oscillator-strength studies for some resonance transitions of Si I, II, and III

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Radiative-lifetime studies have been carried out for resonance transitions of silicon I, II, and III in the ultraviolet using an electron-beam phase-shift apparatus. Lifetimes ranging from 0.03 to 220 nsec were determined. Significant discrepancies with earlier results were found for oscillator strengths of certain transitions, e.g., the Si II 1190–1194-Å multiplet, the Si II 1808-Å multiplet, and the Si III 1206-Å multiplet, which are important transitions in the establishment of the interstellar abundance of silicon. In making these measurements, some very short lifetimes were encountered, requiring that we make strenuous efforts to avoid errors in the reference phase. This led to redeterminations of the radiative lifetimes for the cascade-free transitions in Ne II at 462 Å and in N II at 1085 Å, with lifetimes of 0.15 ± 0.03 and 3.0 ± 0.1 nsec, respectively.

I. INTRODUCTION

Although there have been a number of measurements of oscillator strengths for Si I, II, and III transitions, the data are incomplete and quite uncertain in some important cases. In particular, the Si II multiplets that are observed in absorption in the interstellar medium serve to define the interstellar abundance of silicon, since the bulk of the silicon is once-ionized. Recent studies[1] of the absorption by the interstellar medium between us and various bright type *O* and *B* stars have shown absorption due to Si II, III, and IV. Among the transitions observed, one of the most important, the Si II multiplet at 1190 Å, had only a lower limit given experimentally for its *f* value from lifetime measurements. Theoretical and relative *f*-value studies have indicated that the lifetime is considerably shorter than the limit given by Savage and Lawrence.[2] The multiplet of Si II at 1808 Å, on the other hand, is affected by a configuration interaction that reduces its *f* value to a low level. Savage and Lawrence[2] again gave only an approximate lifetime. The transitions at 989 and 1020 Å for Si II did not have experimentally determined *f* values, but have been observed with the Copernicus satellite.[1]

II. EXPERIMENTAL

We have used the electron-beam phase-shift apparatus constructed by Smith[3] in these measurements. The excitation occurs via the dissociative states reached during the electron-molecule collisions within the rf-modulated electron beam. The modulation frequency was varied from 0.171 to 54 MHz during the course of these measurements, so as to cover the range of frequencies giving easily measurable phase shifts for the various lifetimes found. The electron beam was sinusoidally modulated and accelerated through 100 to 200 V during the experiments. We have used the cascade-free Ne II transition at 1908 Å (see Savage and Lawrence[2]), for the phase reference for the Si transitions and the 1808-Å Si II transitions, and have compared with the N II 1085-Å and the Ne II 462-Å transition for the remainder of the measurements. There exist a large number of measurements of the upper-state lifetime for the N II transition at 1085 Å (see Smith, Bromander, Curtis, and Buchta[4]; Berry, Bickel, Bashkin, Désesquelles, and Schectman[5]; and Buchet, Poulizac, and Carré[6]). These measurements are all in excellent agreement, with an average of all the above of 3.0 ± 0.1 nsec. The Ne II transition at 462 Å has not been so well studied, but is known to have a very short lifetime of less than 0.2 nsec (Hesser,[7] Mickey,[8] and Hinnov[9]). The last two studies are *f*-value measurements, and not lifetime determinations. Hesser gave the lifetime of the transition as $0.0 \pm ^{0.2}_{0.0}$ nsec. We felt that a thorough comparison of these two reference lines with our Si transitions could establish a zero of phase with the accuracy required for these measurements, and was necessary since some of the lifetimes were certain to be very short. We have produced the silicon emission lines by electron-molecule dissociative collisions with SiF_4 at pressures from 0.5 to about 2.0 Torr No radiative entrapment effects were found in any of the cases studied.

As will become apparent below in the results, we were faced with measuring some phase shifts near the limit of our present time response; that is, only a few millicycles at 54 MHz. Very small errors due to possible effects, such as variation of transit time in the photomultiplier tube (PMT) due to different illumination of the photocathode when the slit width is changed, etc., required very close checking. We have determined that none of the following affected our measurements within the limits noted: (i) PMT gain, below the double pulse level for the Bendix magnetic multiplier used (~1900 V), (ii) count rates below 10 000 counts per second (cps), (iii) all slit widths used, (iv) electron accleration voltage variations, (v) measurements with mixed and separate gases, (vi) all electronic gain levels, and (vii) receiver bandpass. We were also careful to maintain a very low background pressure of $\sim 1\times10^{-6}$ Torr in the monochromator, and in the electron source. Under these conditions, we could show that the phase shifts were accurate to within a fraction of a millicycle. Our most exhaustive measurements were made at the highest frequency, 54 MHz, where we are presently able to make measurements with greatly improved stability compared to our previous apparatus. The smallest phase shift measured at 54 MHz was 3.5 millicycles (mc) for the Si III resonance transition, compared to 8 mc for the Ne II 462-Å transition, so that we can immediately give an absolute lower limit to the lifetime for the Ne II upper state of 0.09 nsec, assuming the Si III phase shift to correspond to a zero lifetime. An improved estimate is made from the intercomparison below.

III. THEORETICAL FREQUENCY DEPENDENCE OF THE PHASE SHIFT

Since problems with multiple cascading and blending were encountered in this experiment, a detailed analysis of the theoretical frequency dependence of the phase shift in the presence of these effects was made. Although present experimental accuracies permit the application of these results to only a limited degree, the general relationships are presented below.

A. General solution

We consider a system of levels which are excited by a modulated electron beam of current $(Q+M\cos\omega t)e$, and are depopulated only by radiative emission, with negligible collisional de-excitation and radiation trapping. Under such conditions the instantaneous population of each level is describable by an equation,

$$\frac{dN_n}{dt}=\sigma_n(Q+M\cos\omega t)+\sum_{j=n+1}N_j(t)A_{jn}-\frac{N_n(t)}{\tau_n}, \tag{1}$$

where σ_n is the excitation cross section, A_{jn} is the transition probability, and τ_n is the level mean life. The simultaneous solution of these coupled differential equations yields an emitted intensity $I_{nf}(t)$ of the form[10]

$$I_{nf}(t)=\eta_f(t)A_{nf}=\xi Q+\eta M\cos(\omega t-\phi). \tag{2}$$

The phase shift ϕ is expressible in complex notation as[10]

$$\phi=-\arg\left[\sum_{nf}\left(\sigma_n P_{nf}+\sum_j\sigma_j P_{jn}P_{nf}+\sum_k\sum_j\sigma_k P_{kj}P_{jn}P_{nf}+\cdots\right)\right], \tag{3a}$$

where

$$P_{jk}=\tau_j A_{jk}/(1+i\omega\tau_j). \tag{3b}$$

The nested sums are performed over (from left to right): blended levels (transitions from levels n to f which yield unresolvable wavelengths); direct cascades (from levels j into level n); two-step indirect cascades (from levels k via levels j into level n), and, etc., for indirect cascading in arbitrary numbers of steps. Since a primary level has r additional mean lives contributing to its decay through various direct and indirect cascades and/or blends, Eq. (3) contains $2r+1$ independent constants which can be evaluated as fitting parameters for the dependence of the phase-shift data as a function of frequency. These constants correspond physically to the $r+1$ mean lives τ_j and r-branched excitation cross sections $\sigma_j\tau_j A_{jk}$ for the cascades and blends (relative to the corresponding quantity for the primary level, which factors outside the argument).

The basic form of the frequency behavior of Eq. (3) depends only upon the total number of contributing mean lives; so for any combination of r cascades and/or blends, Eq. (3) can be written in the form

$$\phi=\sum_{j=1}^{r+1}\tan^{-1}\omega\tau_j-\tan^{-1}\left(\frac{a_1-a_3\omega^3+\cdots a_k\omega^k}{1-a_2\omega^2+a_4\omega^4-\cdots a_l\omega^l}\right), \tag{4}$$

where $k=r=l+1$ for odd r, and $l=r=k+1$ for even r. As before, there are $2r+1$ (positive, as written above) constants, of which r of them $(a_1,\ldots,a_r)$ are of a different explicit form for each of the specific situations of cascading and blending. It is also possible, through the use of trigonometric reduction formulas, to write Eq. (4) in the equivalent form

$$\frac{\omega}{\tan\phi}=\alpha_0+\sum_{j=1}^{r}\left(\alpha_j\omega^{2j}+\beta_j\frac{\omega^{2j+1}}{\tan\phi}\right). \tag{5}$$

When expressed as Eq. (5) the theoretical relationship is linear in the fitting parameters, and analytic least-squares fitting programs can be used, setting $r = 0, 1, 2$, etc., with goodness of fit thus indicating the exponential content. The $2r+1$ parameters in either Eq. (4) or Eq. (5) can be related to the mean lives and relative excitation cross sections through direct calculation by Eq. (3), but this requires a knowledge of the details of the cascading and blending conditions, and sometimes Eq. (5) can be used to correlate phase shifts among various contributing levels and reduce the number of fitting parameters.

B. Phase-shift dependences for analyzable cases

With the accuracies attainable in this experiment, it is seldom possible to realistically analyze more than one cascade or blend, and then only if the lifetimes are well separated, by at least a factor of 3 or more. The form of the phase shifts in these cases is given below: $r = 0$, no cascades or blends,

$$\phi = \tan^{-1}\omega\tau_1; \tag{6}$$

$r = 1$, one direct cascade or one blend,

$$\phi = \tan^{-1}\omega\tau_1 + \tan^{-1}\omega\tau_2 - \tan^{-1}\omega a_1, \tag{7a}$$

where

$$a_1 = \frac{\sigma_1\tau_1}{\sigma_1 + \sigma_2\tau_2 A_{21}} \quad \text{(if 2 cascades into 1)}. \tag{7b}$$

The "cascade fraction" is often defined as $\beta \equiv \sigma_2\tau_2 A_{21}/\sigma_1 = \tau_1/a_1 - 1$ or

$$a_1 = \frac{(\sigma_1 A_{1m} + \sigma_2 A_{2n})\tau_1\tau_2}{\sigma_1\tau_1 A_{1m} + \sigma_2\tau_2 A_{2n}} \quad \text{(if 2 and 1 are blended)}.$$

C. Linearization techniques for limited cascading or blending

If two independent phase shifts contain one or more of the same mean lives, there will be a cancellation of terms if the two are subtracted. Thus, in some cases, the difference between two phase shifts can assume a simple theoretical form.

If it were possible to make parallel measurements of the phase shifts of a primary level and its dominant cascade (or of a blend in another branch), the subtracted difference would both be independent of the instrumental phase shift, and would also bring about a cancellation of terms in the theoretical frequency dependence. For example, for a singly cascaded three-level system $2 \rightarrow 1 \rightarrow 0$, the difference in phase shifts [from Eqs. (7) and (6)] is

$$\phi_{10} - \phi_{21} = \tan^{-1}\omega\tau_1 - \tan^{-1}\omega a_1. \tag{8}$$

Using trigonometric reduction formulas this can be written

$$\frac{\omega}{\tan(\phi_{10} - \phi_{21})} = \frac{1 + (\tau_1 a_1)\omega^2}{\tau_1 - a_1}. \tag{9}$$

The quantity on the left-hand side could be linearly least-squares fitted to a straight line, and τ_1 and a_1 determined from the slope and intercept. This method would avoid the use of a known mean-life standard, utilizing instead the correlation between the level and its cascade. The requirement that both the primary transition and its dominant cascade fall within the same detection range greatly restricts the applicability of this method. However, Lawrence and Savage[11] have made a similar linearization without specifically measuring the cascade phase shift, but rather by assuming that it has the form $\phi_{21} = \tan^{-1}\omega\tau_2$. Thus by making a search fit on one of the fitting parameters, they force that parameter to correspond physically to one of the mean lives. A linear fit to Eq. (9) is made for each of a series of searched values of τ_3, and that value which yields the best least-squares fit to a straight line determines all three parameters. This simplifies the interpretation of the fitting parameters and can increase the reliability of the fit by excluding nonphysical values for the cascade meanlife.

IV. DETERMINATION OF REFERENCE STANDARDS FOR MEAN LIVES

In order to determine the instrumental phase shift, it is necessary either to measure a known reference standard (preferably one which is neither cascaded nor blended) in parallel with the unknown, or to have prior knowledge of cascading and blending in the unknown. This presented a particular problem in our measurements, since lifetimes of the order of 0.1 nsec were to be measured. Existing standards had lifetimes of the order of 3 nsec with 5% uncertainties, which would introduce error limits larger than the desired mean life. Therefore we sought a precisely known short-lived mean-life standard.

If one could assume that a level were neither cascaded nor blended, then both its mean life and the instrumental phase shift could be determined as parameters for data fitted to Eq. (6). Further, if two separate transitions were analyzed in this manner and both yielded the same instrumental phase shift, then the assumption of no cascading or blending would be confirmed. This was the procedure which we adopted. Through a series of cross measurements and consistency checks we were able to make a 15% measurement of the $2s^2 2p^5\,{}^2P^\circ - 2p^6\,{}^2S$ transition in Ne II at 462 Å, which was neither cascaded nor blended, and had

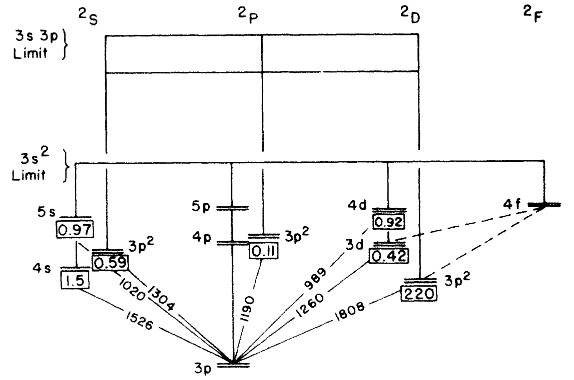

FIG. 1. Energy-level diagram for the low-lying levels of Si II. Levels are labeled by their dominant *ls* configuration and by our measured mean lives (boxed) in nsec, and resonance transition wavelengths are given in Å.

a mean life of 0.15 ± 0.03 nsec. This result is in very good agreement with a recent calculation of Nicolaides and Sinanoğlu,[12] which predicted a mean life of 0.15 nsec, and also agrees with an earlier measurement of Mickey,[8] which yielded a value of 0.14 nsec and also that of Hinnov.[9] These cross measurements also improved the accuracy of a longer-lived standard, the $2p^2\,{}^3P$-$2p^3\,{}^3D^\circ$ transition in N II at 1085 Å, for which we obtained a mean life 3.0 ± 0.1 nsec, in excellent agreement with the average lifetime of the measurements quoted above.[4-6]

V. RESULTS

All lines in the wavelength region 900–2300 Å were identifiable as transitions due to silicon or fluorine except for some weak Lyman radiation probably due to a small hydrofluoric acid contaminant. Under these excitation conditions Si II lines were very intense but only the strongest lines of Si I and Si II were apparent. Lifetime measurements were made for one level in Si I, seven levels in Si II, and one level in Si III.

A. Si I

A measurement of the Si I $3s3p^3\,{}^3D^\circ$ transition at 2208 Å yielded a meanlife of 20.7 ± 3 nsec, with a frequency dependence consistent with a single moderately strong cascade with $\tau_2 = 125$ nsec and $\beta = 0.5$. This result is in reasonable agreement with an earlier measurement of 18 ± 3 nsec by Savage and Lawrence.[2]

B. Si II

Lifetime measurements for the Al I isoelectronic sequence are particularly valuable. Although the spectra are relatively uncomplicated, the states show strong effects of configuration mixing, for example between the normal $3s^2nl$ and the displaced $3s\,3p\,nl$ configurations. These effects were often quite pronounced in our results, and mean lives for the low-lying resonance transitions ranged over three orders of magnitude, from 0.1 to more than 200 nsec. An energy-level diagram of these low-lying transitions, designated according to the dominant configuration, and labeled with our mean-life results is shown in Fig. 1. A comparison of our results with other measurements and with theoretical estimates is given in Table I.

The two lowest-lying ^2D states of this sequence are a particularly interesting case of configuration mixing. They have been studied by Froese Fischer,[18] who has shown them to be describable by two energy-ordered configuration admixtures which vary smoothly as a function of Z, but under-

TABLE I. Lifetimes for Si II upper states.

λ (Å)	Si II Dominant configuration	τ_1 (nsec) This work	τ_1 (nsec) Other expt.	Theory	Absorption f value, this work	Cascade τ_2 (nsec)	Analysis β
989.87	$3s^23p\,{}^2P^\circ$-$3s^24d\,{}^2D$	0.92 ± 0.1		1.05[a]	0.20	20	0.3
1020.70	$3s^23p\,{}^2P^\circ$-$3s^25s\,{}^2S$	0.97 ± 0.2				130	2.0
1190.42 1193.28	$3s^23p\,{}^2P^\circ$-$3s3p^2\,{}^2P$	0.11 ± 0.02	0.4[b]	0.26[a], 0.24[c]	1.94[d]		0
1260.42	$3s^23p\,{}^2P^\circ$-$3s^23d\,{}^2D$	0.42 ± 0.06	0.45[e], 0.40[f], 0.7[h], 0.5[b]	0.33[a], 0.39[g]	0.99	40	0.3
1304.37	$3s^23p\,{}^2P^\circ$-$3s3p^2\,{}^2S$	0.59 ± 0.08	1.4[b]	0.94[a]	0.19	20	0.07
1526.72	$3s^23p\,{}^2P^\circ$-$3s^24s\,{}^2S$	1.5 ± 0.3	0.9[h]	0.88[a]			
1808.01	$3s^23p\,{}^2P^\circ$-$3s3p^2\,{}^2D$	220. ± 22.	490[b], 300[h]	128[a], 60[c], 96[g]	0.004	20	2

[a] Weiss, Ref. 13.
[b] Hofmann, Ref. 14.
[c] Beck and Sinanoğlu, Ref. 15.
[d] Multiplet value.
[e] Berry *et al*., Ref. 16.
[f] Irwin and Livingston, Ref. 17.
[g] Froese Fischer, Ref. 18.
[h] Savage and Lawrence, Ref. 2.

go a crossing of mixing coefficients between Si II and Al I. Thus the dominant configuration for Al I is not dominant for the higher members of either sequence, which can lead to confusion if the sequence is labeled according to its asymptotically dominant configuration. Further, a node in the dipole matrix element for the ground-state transition of the lowest ^2D state (1808 Å in Si II) occurs for a value of Z close to that corresponding to Si II (Froese Fischer reported the node closer to P III, but pointed out that its exact position is very sensitive to the mixing coefficients). Figure 2 shows a plot of absorption f value versus $1/Z$ for the isoelectronic sequence of this lowest ^2D state resonance transition, and compares the theoretical predictions of Froese Fischer with our own and other recent measurements. Our results are clearly consistent with the trend of the measurements, which indicate a slight translation of Froese Fischer's curve toward Al I. Several different theoretical estimates for this transition probability are listed in Table I, and the various calculational techniques yield values differing by 100%. This is not surprising in the context of these strong cancellation effects, and our measurement of a (220 ±22)-nsec mean life is thus in reasonable agreement with the theoretical values, and falls in the range suggested by Savage and Lawrence,[2] from 200 to 400 nsec. Clearly, the 1808-Å transition in Si II has an unusually low f value, particularly so when considering that is a resonance transition. Despite its low transition probability, the 1808-Å transition was very intense in our spectra, due to growing-in cascades from the $4p\,^2P^\circ$ and $4f\,^2F^\circ$ levels (with mean lives of 8.3 and 4.1 nsec, as measured by Berry *et al.*[16]) Here again we see the effects of configuration mixing since, as has been emphasized by Crossley,[24] these cascades would be two electron jumps, and thus forbidden for pure configurations. The next lowest ^2D state corresponds to the other admixture studied by Froese Fischer, and its resonance transition occurs at 1260 Å in Si II. Our measured value for this mean life of 0.42 ± 0.06 nsec is in excellent agreement both with the theoretical predictions of Froese Fischer[18] and of Weiss,[13] and with other recent measurements.[17] Although it is 50% shorter than the earlier measurement of Savage and Lawrence, it is still consistent with their result, since an analysis of the phase-frequency diagram was not attempted by them.

The third-highest ^2D state, with its resonance transition at 989 Å, was also in excellent agreement with a theoretical calculation of Weiss,[13] and had a mean life of 0.92 ± 0.1 nsec. Thus for the three lowest-lying ^2D states there seems to be good agreement between theory and experiment, despite very strong configuration mixing effects. This was not true for the lowest ^2S and ^2P states, with resonance transitions at 1304 and 1190.43 Å, respectively. The lifetimes of 0.11 ± 0.02 nsec for the 1190.43-Å transitions, and 0.59 ± 0.08 nsec for the 1304-Å transition were each a factor of 2 shorter than calculations by Weiss,[13] and factors of 3 to 4 shorter than those derived from the emission measurements of Hofmann.[14] The ^2P transitions were particularly short-lived with an implied f value of 1.94, near the total oscillator strength possible for this transition according to the sum rule.

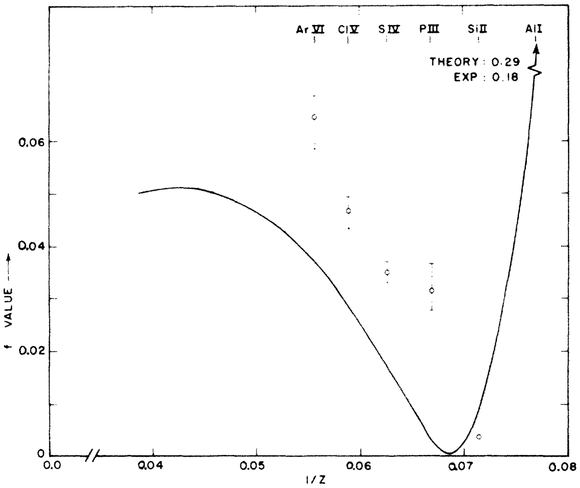

FIG. 2. Isoelectronic sequence for the Si II $3s^23p\,^2P$–$3s3p^2\,^2D$ (see text concerning the ambiguity of this notation) transition. Experimental data sources are: Al I, Ref. 19; Si II, this work; P III, Ref. 20; S IV, Ref. 21; Cl V, Ref. 22; and Ar VI, Ref. 23. The theoretical source is Ref. 18. An extensive set of figures of this type are given in Ref. 13.

C. Si III

The only strong Si III transition observed was at 1206 Å. The interpretation of the observed phase shifts is complicated by an unusual blending of the primary transition with its own cascade. The primary transition is $3s^2\,^1S$–$3s3p\,^1P^\circ$ at 1206.51 Å, a direct cascade from $3s3d\,^1D$ is at 1206.53 Å, and an indirect cascade through $3p^2\,^1D$–$3s3d\,^1D$ is at 1207.52 Å. These transitions are not spectrally resolvable under the present lifetime-measurement conditions. This blending was pointed out by Berry *et al.*[16] in reporting a measured lifetime of 0.4 nsec, assigned to the primary transition. This was recently remeasured by Irwin and Livingston,[17] with similar results. Our results suggested a much shorter lifetime, around 0.03 nsec, but this would imply an f value greater than 2, the number of electrons outside closed shells, and would require substantial negative contributions to satisfy sum rules. The cascading transitions themselves are computed to have mean lives shorter than that for the resonance transition.[25] Thus, the measured phase shifts are a complicated, and presently unresolvable, mixture of the cascading, blending, and the resonance transitions. Our usual two-term cascade analysis, described in some detail above, yields a fit of the data to within experimental errors. The resulting lifetimes, however, represent some weighted average of the lifetimes of the states contributing at the observed wavelength, and their respective cascades. The purpose of this discussion is to point out that only an experiment that spectrally separates these various transitions, or which selectively populates the upper state of the resonance transition can yield a true value of the lifetime. We believe that our present results show conclusively that such an experiment has not yet been conducted. For the present, the theoretical estimate of the f value of this system must be considered the most reliable.

It was pointed out to us that the discrepancy we find for the *relative* f values for the Si II transitions at 1190, 1260, and 1304 Å is puzzling when considered in comparison with the measurements of Hofmann,[14] and that the measurements of Hofmann should be reliable as to the relative f values for these three transitions, and agree with the calculations due to Weiss.[13] We agree that these studies would seem to be of high quality, but still assert that the data here are superior to either, since we directly measure relative lifetimes at high accuracy. If there were an error in our data, we would be inclined to place it in the 1260-Å f value, since, there, a significant correction for cascading was made in the phase-shift data. We believe the absolute scale of our results to be very reliable. This is especially important in view of the sensitive character of the configuration interactions shown to be present in these series.

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