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Saiful Alam, Suresh Sundaram, Miryam Elouneg-Jamroz, Xin Li, Youssef El Gmili, et al.. InGaN/InGaN multiple-quantum-well grown on InGaN/GaN semi-bulk buffer for blue to cyan emission with improved optical emission and efficiency droop. Superlattices and Microstructures, 2017, 104, pp.291-297. 10.1016/j.spmi.2017.02.036 . hal-01830907

HAL Id: hal-01830907
https://hal.science/hal-01830907

Submitted on 2 Mar 2022

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InGaN/InGaN multiple-quantum-well grown on InGaN/GaN semi-bulk buffer for blue to cyan emission with improved optical emission and efficiency droop

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Abstract

$In_{0.16}Ga_{0.84}N/In_{0.05}Ga_{0.95}N$ Multiple Quantum Well (MQW) structure grown on a 70 nm thick high quality semi-bulk InGaN buffer layer is reported. Temperature dependent photoluminescence (PL) reveals 67.5% of room temperature Internal Quantum Efficiency (IQE) at an emission peak of ~455 nm with FWHM of 20 nm. Low temperature PL study shows clear improvement in emission intensity when conventional GaN buffer and GaN barrier are replaced by semi-bulk InGaN buffer in addition with InGaN barrier. Simulation confirms improved IQE and reduced efficiency droop when using semi-bulk as buffer which is attributed to the improved overlapping of electron-hole wave functions due to the reduced internal electric field from counteraction by surface polarization field. This efficiency improvement is very beneficial for high In content green LEDs where the efficiency is limited by polarization induced Quantum Confined Stark Effect (QCSE) for excess indium content.

Keywords: InGaN multi-quantum-well; InGaN buffer; LED; MOVPE

1. Introduction

GaN is one of the most promising materials for fabrication of efficient light emitting devices in the complete visible spectral range [1]. InGaN/GaN based blue light emitting diodes (LEDs) have a fairly good performance now-a-days [2]. However, to compete with conventional lighting, the efficiency is yet to be improved, particularly in the "green-gap" range [3]. For longer wavelengths such as green emission, the

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efficiency of devices grown in the commonly used +c (0001) direction of GaN decreases with increasing indium (In) content in the active region [4]. This phenomenon is attributed to the built-in piezoelectric field in the biaxially compressively strained InGaN MQWs. Such a piezoelectric field leads to a local spatial separation of electrons and holes within the quantum wells (QWs), which is the so called Quantum Confined Stark Effect (QCSE), and consequently leads to a decreased emission efficiency due to a lower recombination probability between electrons and holes [1,4]. Furthermore, polarization field is considered to be one major possible reason of efficiency droop at high current density [5]. Several groups have proposed reduction of polarization field and improvement of optical performance by using semi- or non-polar direction growth [6–8], asymmetric QWs using AlGaN, InGaN, GaAsN [9–11], staggered QW [12–16], triangular QW [17–19], and using AlInGaN barriers [20]. Moreover, reduction or balance of strain has been suggested by using InGaN barrier [21–23], InGaN pre-layer [24–28] and also using $In_xGa_{1-x}N/Al_yGa_{1-y}N$/GaN MQW [29,30]; however, internal electric field might need to be sacrificed for improved material quality by strain compensation [29] and realization of a high quality InGaN buffer layer is also very critical, which has been addressed [26,31]. Nevertheless, good crystal quality, requirement of less In content to get same wavelength emission compared to that for semi- or non-polar direction growth [29] and mask free fewer processing steps make +c-direction growth still commercially promising and combined theoretical and experimental study to alleviate polarization field and enhance optical emission regarding this direction growth by optimized structure design are still important. In this article, we report on a new approach of growing high IQE InGaN/InGaN MQWs using a high quality semi-bulk (SB) InGaN buffer (4 periods of 15 nm InGaN and 2 nm GaN layers in between two successive InGaN layers) that shows improvement of optical emission and reduction of efficiency droop at an emission wavelength of ~455 nm and we explain this improvement theoretically by simulation. This semi-bulk approach for InGaN growth allows to have a high quality thick InGaN buffer compared to bulk InGaN [32], where inserted thin GaN layer between two successive InGaN layers absorbs excess In and helps to avoid phase separation and quality degradation of the final thick InGaN buffer [32].

2. Experimental section

2.1. Growth

The samples were grown in a T-shape horizontal flow low-pressure metal organic vapor phase epitaxy (MOVPE) reactor [33] on 3.5 µm standard n-doped commercial GaN templates (STN) on (0001) direction sapphire substrate. The growth temperature was 795 ^oC for InGaN growth and 1000 ^oC for GaN growth. The reactor pressure was 100 Torr. Nitrogen (N_2) and Hydrogen (H_2) were used as carrier gas. Trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia (NH_3) were employed as precursor sources for Ga, In and elemental N, respectively. The V/III ratio and TMIn precursor flow were calibrated to achieve desired In percentage in InGaN layers. Three sets of MQW structures were grown, named as sample A, B and C. Sample A had 1 µm n-GaN on GaN template, followed by 70 nm nid-GaN buffer and then 3 periods of 5 nm GaN barrier and 3 nm $In_{0.16}Ga_{0.84}N$ well and finally capped by 2 nm GaN. Sample B had similar structure, except that the GaN barriers were replaced by $In_{0.05}Ga_{0.95}N$. And in sample C both the GaN buffer and GaN barriers were replaced by $In_{0.05}Ga_{0.95}N$ semi-bulk (SB) buffer and $In_{0.05}Ga_{0.95}N$ barriers respectively. The structures are shown in Fig. 1.

2.2. Characterization

High-resolution X-ray diffraction (HR-XRD) measurements were performed in a Panalytical X'pert Pro MRD system with Cu-Kα radiation. XRD reciprocal space maps (RSMs) of the asymmetric (11.4) plane were used to determine degree of relaxation of the InGaN layers. The thickness was measured by growth rate calculated from *in-situ* reflectrometry and confirmed by transmission electron microscopy (TEM).

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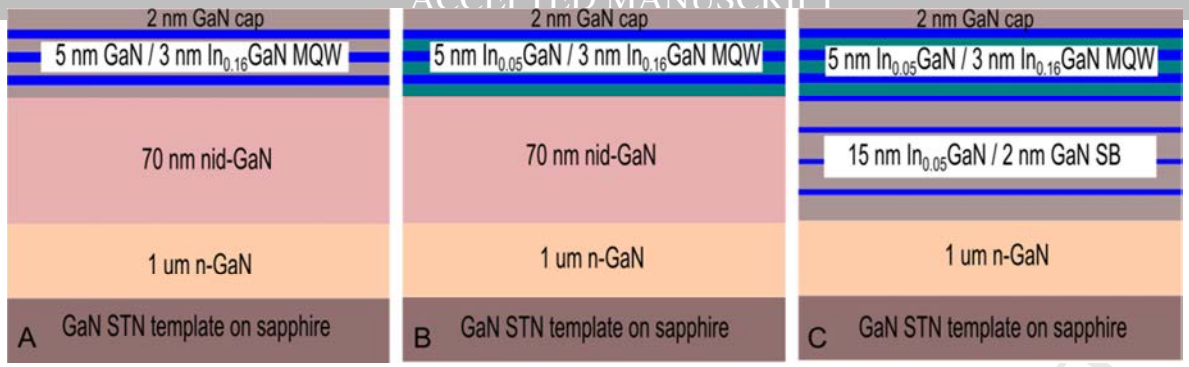


Fig. 1. Structure of the three samples under this study: reference sample A with GaN buffer and GaN barrier, reference sample B with GaN buffer and $In_{0.05}Ga_{0.95}N$ barrier and final structure of sample C with semi-bulk $In_{0.05}Ga_{0.95}N$ buffer and $In_{0.05}Ga_{0.95}N$ barrier.

The surface morphology was studied by scanning electron microscopy (SEM) and atomic force microscopy (AFM). Optical emission properties of InGaN structures were investigated by both cathodoluminescence (CL) and photoluminescence (PL) measurements. Room-temperature and low temperature CL investigations were performed in a digital scanning electron microscope [34]. Temperature dependent PL spectra were measured in a helium cryostat from 15 K to 290 K using a 375 nm laser as excitation source. The excitation power was varied from 0 to 48 mW with a beam diameter of roughly 100 μm. To avoid wafer non-uniformity, similar areas at the center region on the wafers were measured in the PL study carefully at the same time with similar optical conditions to ensure that the results were comparable.

2.3. Simulation

The structures were simulated by Silvaco TCAD software, where self-consistent band structures and wave functions were obtained by solving the Schrödinger equation for electrons, Hamiltonian for holes and Poisson's equation iteratively [35]. The drift-diffusion *k.p* model was used, interface and surface charge and career screening effect were considered and material parameters were taken after Bernardini and Fiorentini et al. [36]. The simulation was carried out for a bias of 50 A and upto 3.4 V.

3. Results and discussion

The growth was optimized to get a ~70 nm thick good crystal quality SB InGaN buffer. The material and morphological quality of this buffer layer were examined by HR-XRD, SEM and AFM. Fitting the XRD experimental data by simulation gave 5±1% In incorporation and reciprocal space mapping (RSM) showed fully strained situation. SEM image (Fig. 2a) showed good surface morphology with V-pit density of 8×10^6 cm^{-2}. Surface roughness of 1.3 nm for a 5 μm × 5 μm scan area was obtained by AFM for this SB InGaN buffer, much better than that of the bulk InGaN [32]. To ensure the SB quality, room temperature CL was carried out that showed a sharp emission peak at ~400 nm with a FWHM of 22 nm (Fig. 2b) confirming single phase In, whereas bulk InGaN has multiple emission peaks corresponding to different separated phases [32]. The MQWs with 16±1% were grown on top. Fig. 2c shows XRD experimental data for sample A, B and C which fits well with the simulation of the designed structure. Sample C had ~15% relaxation (Fig. 2c, inset) and a surface morphology with V-pit density of 8×10^7 cm^{-2}, whereas sample A and B were fully strained with V-pit density of 8.5×10^8 cm^{-2} and 7.1×10^8 cm^{-2} respectively. Using of SB buffer

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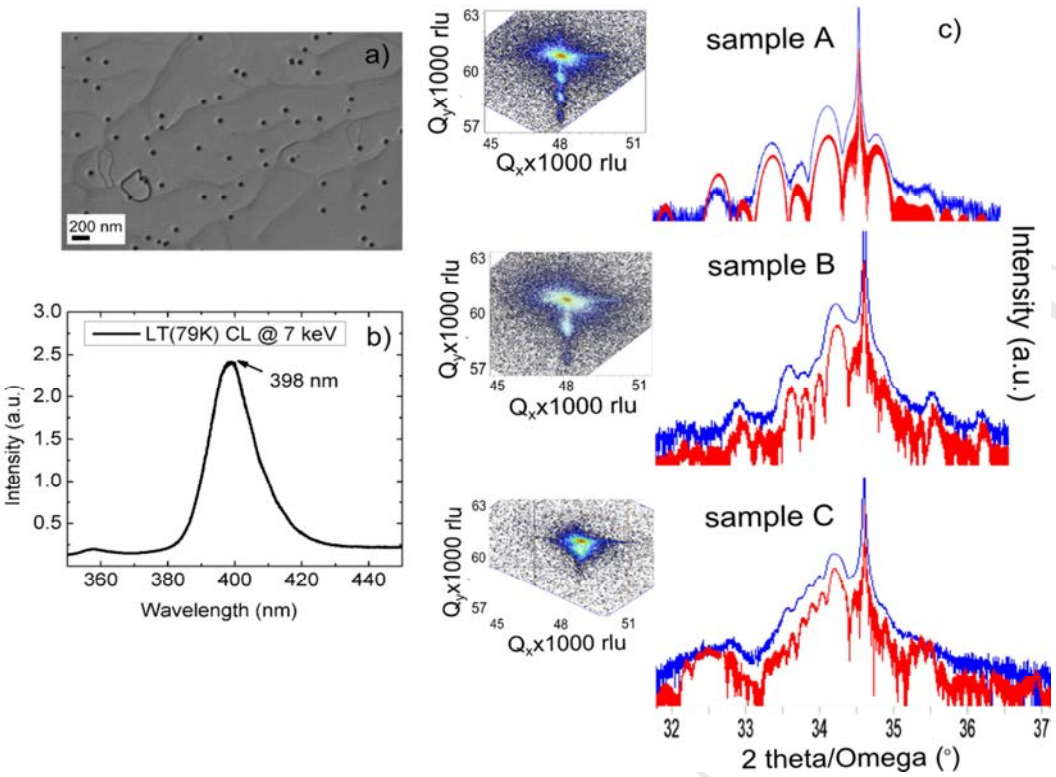


Fig. 2. SEM image of semi-bulk (SB) buffer sample (a), low temperature CL emission of SB buffer (b) and XRD 2θ-ω scan of sample A, B and C (c) (inset shows the RSMs).

The rms roughness of sample C was 1.50 nm and that of sample A and B were 2.0 nm and 1.8 nm respectively from AFM for a 5 μm × 5 μm scan area. Though using SB buffer in sample C increased the size of the V-pits, the less pit density and absence of inclusion defects due to the partial reduction of localized variation of strain energy [37] made the surface morphology comparable. In the low temperature (15 K) PL experiment, sample A showed an emission peak at 490 nm and sample B showed an emission at ~438 nm (Fig. 3a). More than three-fold improvement in emission intensity for sample B is attributed to the reduction in potential barrier height and more homogeneous repartition of carriers in the wells for putting indium in the barrier. LTPL for sample C (Fig. 3b) yielded an emission at ~454 nm with a FWHM of 20 nm. The semi-bulk (SB) peak is also visible at ~405 nm. Even though the internal electric field was reduced in MQW of sample C compared to that in MQW of sample B, as it will be shown in simulation result later, the ~15 nm red shift in emission wavelength for sample C is attributed to the partial strain relaxation related In content variation and slight In pulling induced increase of well thickness. The PL emission intensity, when using the SB InGaN as buffer, got doubled compared to sample B and shows six-fold increment compared to sample A. This improvement was definitely due to the insertion of the SB buffer layer as both sample B and sample C has the same MQW structure except that sample C has InGaN SB buffer beneath the MQWs. From the measured integrated intensity at different laser excitation power and from 15 K to 290 K temperature range and using power normalized integrated intensity, the room temperature IQE was calculated to be 67.5% by considering the radiative recombination efficiency is 100% at 15 K [38]. Fig. 3b (inset) shows the integrated PL intensity vs *1/T* curve for sample A, B and C. It can be seen that the thermal quenching of non-radiative recombination

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process is worse for sample A and B compared to that of sample C and the shape of these curves suggest that the intensity decrease is more prominent at high temperature range for sample A and B than that of sample C revealing the reduced effect of non-radiative recombination channel for sample C [39].

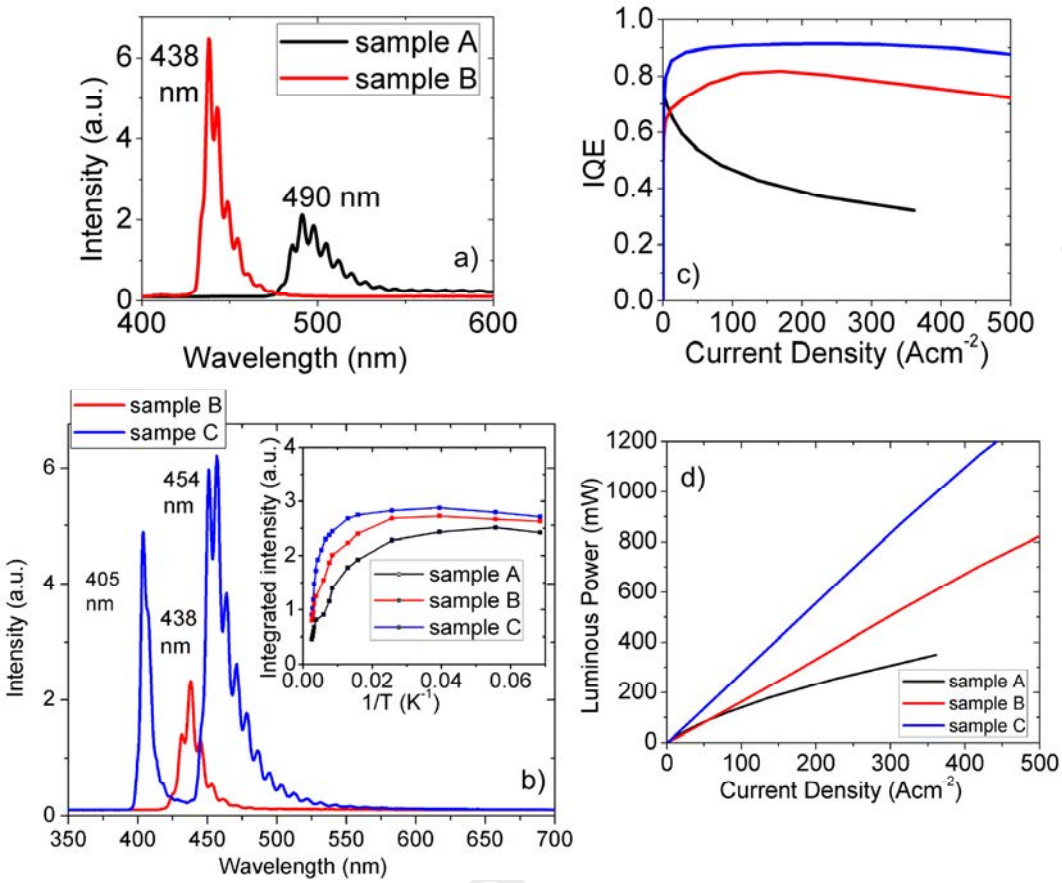


Fig. 3. Low temperature (15 K) PL spectra comparison of sample A and B (a) and sample B and C (b) (inset shows the integral PL intensity versus *1/T* for sample C). Calculated IQE (c) and luminous power (d) comparison of sample A, sample B and sample C.

To understand the improved luminescence performance of our final MQW structure sample C, simulation was carried out [35]. At a current density of 200 A/cm^2, a 130% improvement of room temperature IQE compared to that of sample A and 15% compared to that of sample B have been obtained (Fig. 3c). Moreover, there was no efficiency droop for sample C at this current density compared to sample A and B. The calculated luminous power for sample C was doubled compared to sample A and ~60% improved compared to sample B at this current density (Fig. 3d). This improvement is attributed to the reduction of net electric field in sample C. Using InGaN as a barrier reduced the built-in electric field [40]. Further reduction in the average electric field in the three wells was mainly due to the use of SB InGaN as buffer because the oppositely directed surface charge related field reduced the net built-in electric field [23]. For sample C, 1.5 times less electric filed was calculated compared to sample A and the average electric field in the wells was 0.1 MV/μm less compared to that of sample B (Fig. 4b). The polarization charge concentration was half in the structure with InGaN barrier (sample B) as shown in Fig. 4a compared to that of the reference structure (sample A). The reduction in net electric field when using SB InGaN as buffer resulted in reduced band bending (Fig. 4c). As a

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result, the QCSE was reduced and there was more overlap of electron and hole wave functions (Fig. 5) for the final structure sample C. In fact, 38.7% more overlap compared to sample A and 9% more overlap compared to sample B was obtained.

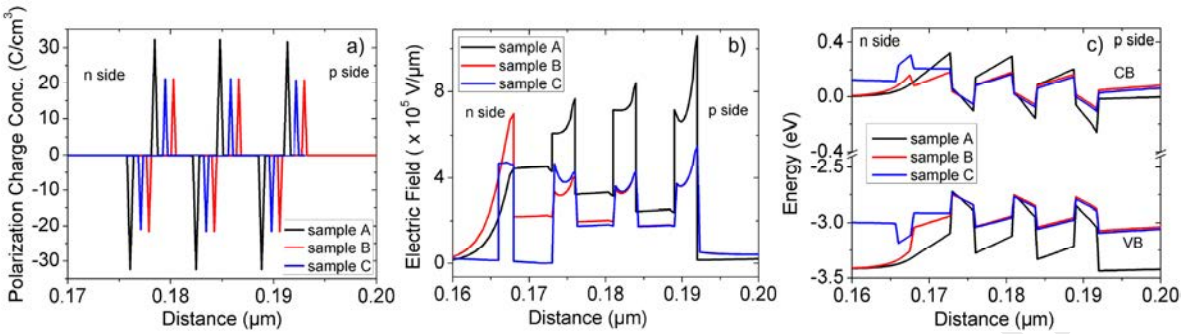


Fig. 4. Polarization charge concentration (a), internal electric field (b) and valence and conduction band structure (c) for the samples under study. Please note that, in (a), the distance scale has been shifted for sample A and B for better visibility.

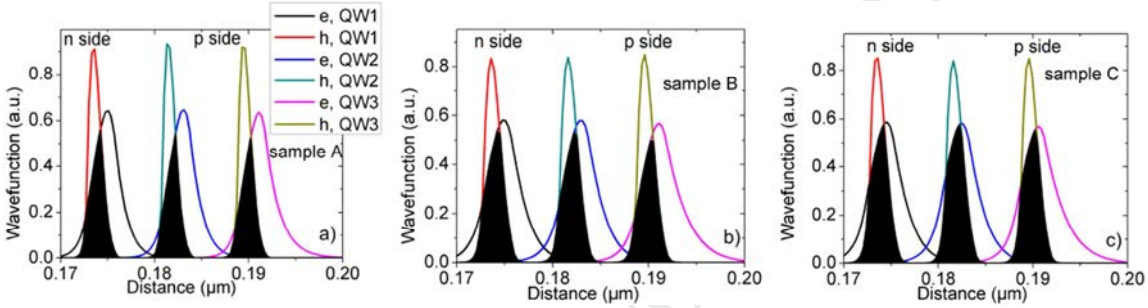


Fig. 5. Electron and hole wavefunction overlap for (a) reference sample A (b) reference sample B and c) our final structure sample C.

This improved overlapping led to more radiative recombination and hence was consistent with the experimental increase of PL emission intensity [25]. For sample C, the spontaneous emission rate and the total radiative recombination rate (not shown here) were also calculated to be higher than that of the reference structures A and B, and was attributed to the reduced electric field [22,27]. This led to more homogenous distribution of carriers [20] in the MQWs grown on SB InGaN buffer and improved tunneling of electrons from the buffer that was un-intentionally n-doped and acted as an electron reservoir. While the design and experiment suggest 5±1% In in the SB buffer and in the barrier for the best optical performance from 450 nm to 480 nm wavelength range, simulations for longer wavelength suggests scaled up In in the barriers and in the SB InGaN buffer for the best room temperature IQE performance (summarized in table 1).

Optimization of growth parameters to get SB InGaN buffer with higher In content maintaining good crystal quality and realization of MQWs on top for longer wavelength (~520 nm) emission is the subject of current study.

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Table 1. Simulated emission wavelength and IQE of MQWs with different In content in well and SB buffer.

In (%) in well	In (%) in SB buffer and InGaN barrier	Wavelength (nm)	IQE (%) @ 200 Acm^{-2}
30	15	510	68%
35	20	535	55%
40	25	560	52%

4. Conclusion

In summary, the InGaN MQW with InGaN barrier grown on a ~70 nm thick fully strained semi-bulk InGaN buffer gave almost double optical emission compared to conventional MQW structure with GaN barrier and GaN buffer. This improvement is due to the screening of built in field by surface polarization field that eventually reduced the net built-in electric field in the wells and also due to more homogenous carrier distribution among the MQWs, when grown on semi-bulk InGaN buffer. These effects resulted in reduced band bending that increased the emission efficiency by more overlapping of electron and hole wave functions. 67.5% internal quantum efficiency was experimentally obtained for our structure with semi-bulk InGaN buffer and InGaN barrier in MQW. This improvement in IQE is the motivation for future work to realize LEDs with emission wavelength in the "green-gap" range using the semi-bulk buffer technique.

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