A Study of the Inclusion of Prelayers in InGaN/GaN Single and Multiple Quantum Well Structures

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We report on the effects on the optical properties of blue-light emitting InGaN/GaN single and multiple quantum well structures including a variety of prelayers. For each single quantum well structure containing a Si-doped prelayer we measured a large blue shift of the photoluminescence peak energy and a significant increase in radiative recombination rate at 10 K. Calculations of the conduction and valence band energies show a strong reduction in the built-in electric field across the quantum well occurs when including Si-doped prelayers, due to enhancement of the surface polarisation field which opposes the built-in field. The reduction in built-in field across

the quantum well results in an increase in the electron-hole wavefunction overlap, increasing the radiative recombination rate, and a reduction in the strength of the quantum confined Stark effect, leading to the observed blue shift of the emission peak. The largest reduction of the built-in field occurred for an InGaN:Si prelayer, in which the additional InGaN/GaN interface of the prelayer, in close proximity to the quantum well, was shown to further reduce the built-in field. Study of multiple quantum well structures with and without an InGaN:Si prelayer showed the same mechanisms identified in the equivalent single quantum well structure.

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1 Introduction Light emitting diodes (LEDs) incorporating InGaN/GaN quantum wells (QWs) are now widely used as high efficiency blue-light emitters, with reported room temperature external quantum efficiencies greater than 50 % [1-3]. The high efficiencies of InGaN/GaN light emitters occur despite the large built-in electric field across the QWs [4-6]. This electric field occurs due to the combined effect of discontinuities in spontaneous polarisation at the GaN/InGaN interfaces, and the piezoelectric polarisation due to the strain of the InGaN QWs to the GaN layers [6]. In *c*-plane oriented InGaN/GaN structures the built-in electric field aligns with the growth axis. This strong electric field results in the spatial separation of electron and hole wavefunctions leading to a reduction in the radiative recombination rate and an associated quantum confined Stark effect (QCSE). Slower radiative recombination rates can result in less effective competition with non-radiative recombination processes, in turn leading to a reduction in internal quantum efficiency (IQE).

In order to further improve the IQE of LEDs either the radiative recombination rate must be increased or the non-radiative recombination rate decreased. Strain engineering of the QWs could provide a means to increase the radiative recombination rate by reducing the piezoelectric strain of the InGaN layers, therefore reducing the strength of the built-in electric field and the QCSE. There are several reports that this can be achieved by the inclusion of an InGaN layer prior to growth of the 1st QW, so-called “prelayers”. Typical prelayer structures consist of 20-30 nm of intentionally *n*-type doped (In)GaN, positioned a few nanometers beneath the 1st QW. Alternatively InGaN/GaN superlattice prelayers have been used [7]. In a variety of structures and devices the main reported effects of including (In)GaN prelayer structures prior to the QWs are increases of the luminescence intensity and efficiency [8-12]. Nanhui *et al.* [13, 14] reported on the inclusion of an unintentionally doped 20 nm-thick In_{0.08}Ga_{0.92}N layer in an InGaN/GaN multiple QW structure and found an increase

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in photoluminescence (PL) intensity at room temperature, which the authors attributed to strain relaxation of the QWs. Otsuji *et al.* [8] and Takahashi *et al.* [10] reported an increase in electroluminescence (EL) intensity, for a range of temperatures, when including a 15 nm-thick Si-doped $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ prelayer in a blue emitting three period $\text{In}_{0.3}\text{Ga}_{0.7}\text{N}/\text{GaN}$ multiple QW device. The increase in luminescence intensity was ascribed to enhanced electron capture efficiency, due to the prelayer acting as an *electron reservoir*. A similar explanation for a measured increase in cathodoluminescence (CL) intensity was reported by Xia *et al.* [15] who concluded from a depth resolved CL investigation that some charge transfer occurs between the InGaN prelayer and QWs. Akasaka *et al.* [11, 12] and Törma *et al.* [9] reported increases in the PL efficiency, and the PL and EL intensities at room temperature; this behaviour was ascribed to a reduction in the density of defects in the vicinity of the QWs. Takahashi *et al.* [16] reported that the inclusion of low temperature grown GaN and InGaN prelayers resulted in an increased formation probability of “V”-defects, which are believed to create potential barriers around threading dislocations [17].

Figure 1 PL spectrum of MQW structure, containing a InGaN:Si prelayer.

In our previous work [18] we reported an increase in room temperature internal quantum efficiency (IQE) for a 10 period $\text{In}_{0.16}\text{Ga}_{0.84}\text{N}/\text{GaN}$ multiple QW structure containing a 24 nm thick Si-doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$ prelayer. An additional high energy emission process was identified occurring ~ 70 meV above the PL peak emission energy, as shown in Figure 1. This feature was responsible for $\sim 39\%$ of the 10 K PL emission intensity. The additional emission process was measured to have a faster decay rate at 10 K than the primary PL peak and found to be more stable with increasing temperature, thus the increase in room tempera-

ture IQE was attributed to the contribution of this additional recombination process. High-resolution X-ray diffraction results allowed us to dismiss strain relaxation of the QWs as a cause of this change in optical properties. Thus the high energy emission was attributed to either the recombination of carriers in QWs formed on the semi-polar facets of “V”-defects or to the 1st QW in the stack whose properties were suggested to be somehow modified by the presence of the InGaN prelayer.

In summary, despite a variety of reports on the improvements to the optical properties due to the inclusion of prelayers in InGaN/GaN structures and devices, there is, as yet, no consensus as to how these improvements are achieved. As a logical extension to our previous work, we report in this paper on the optical properties of single QW InGaN/GaN structures containing a variety of prelayers. We have modelled our results using a commercial band profile package and then apply the same model to explain the optical properties of 10-period InGaN/GaN MQW structures containing prelayers.

Figure 2 Schematic diagram showing the structural details of samples A-D and the different prelayers incorporated into each structure.

2 Experimental Details The single QW structures studied here were deposited by MOVPE onto *c*-plane sapphire substrates (with miscut of $0.25 \pm 0.1^\circ$ along *a*) in a 6×2 Thomas Swan close-coupled showerhead reactor. The “quasi-two temperature growth method” [19] was used for deposition of nominally 2.4 nm thick $\text{In}_{0.16}\text{Ga}_{0.84}\text{N}/\text{GaN}$ single QWs onto non-intentionally doped (nid, $n < 10^{17} \text{ cm}^{-3}$) GaN templates with a threading dislocation density of $\sim 4 \times 10^8 \text{ cm}^{-2}$. The GaN templates contained a variety of prelayer structures, shown in Figure 2, including a nid GaN layer (for reference), Si-doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$, and Si-doped GaN layers grown at different temperatures. The prelayers in samples B, C and D were intentionally silicon doped at a level of $5 \times 10^{18} \text{ cm}^{-3}$. The characteristics of the prelayers that may influence the properties of the single QWs are the presence of doping in the prelayer, the pres-

Table 1 Summary of the structural and optical properties of the single QW structures on various prelayers.

Sample	Prelayer	QW Thickness (nm)	In Content (%)	PL Peak Energy (eV)	τ_{10} PL decay time (ns)	Electric Field (meVnm ⁻¹)
A	No prelayer	2.3 ± 0.1	15.7 ± 1	2.635	120 ± 4	254
B	InGa _{0.95} N:Si	2.3 ± 0.1	16.2 ± 1	2.784	10 ± 1	111
C	LT- GaN:Si	2.4 ± 0.1	16.0 ± 1	2.749	19 ± 1	149
D	HT- GaN:Si	2.3 ± 0.1	16.2 ± 1	2.756	19 ± 1	149

ence of indium (and indeed the indium content) and the growth temperature. A reduced growth temperature might, for example, enhance the size of “V”-defects in the material underlying the QW, therefore providing a mechanism by which the prelayer can alter the properties of the QW. Thus samples C and D were grown at 770 °C (similar to the InGa_{0.95}N growth temperature) and 1020 °C (a typical GaN growth temperature) respectively.

Si-doping levels were determined using secondary ion mass spectrometry analyses of calibration structures grown under similar conditions. The thickness and composition of the single QWs were determined using a combination of X-ray diffraction (XRD) and X-ray reflectivity (XRR) following the methodology reported by Smeeton *et al.* [20]. X-ray measurements were performed on a Philips X-pert Gen6 diffractometer. For XRD, a four-bounce asymmetric Ge (220) monochromator and a three-bounce Ge (220) analyser were employed. The combined QW and barrier thicknesses and the average indium compositions (across the InGa_{0.95}N and GaN layer) were determined using a GaN (002) ω -2 θ scan. For XRR, a 4 mm mask was coupled with a 1/32 ° programmable divergence slit and a 0.2 mm programmable receiving slit. The measured specular reflection of the X-ray beam contains a convolution of two Fabry-Pérot interference functions, occurring due to multiple reflections at the InGa_{0.95}N/GaN interfaces. The interplay between the two interference functions was used to determine the QW thickness following the method of Smeeton *et al.* [20].

The optical properties of the single QW structures were studied at 10 K by measuring the photoluminescence and PL decay times. The structures were mounted on the cold finger of a temperature controlled closed-cycle helium cryostat. The structures were inclined at Brewster’s angle with respect to the collection axis in order to minimise the effects of Fabry-Pérot interference oscillations on the PL spectra [21]. A frequency tripled mode-locked, Ti:sapphire laser system, emitting photons with energy of 4.881 eV, was used as the excitation source for the PL spectra and PL decay time measurements. The PL decay transients we measured using a time-correlated single photon counting system. The optical properties of the single QW structures were then analysed and compared with the calculated band profiles using a commercially available device simulation package (nextnano³) [22].

3 Results and Discussion The XRD and XRR measurements performed on each sample found that the QWs in samples A and B to be indistinguishable, with thicknesses of 2.3 ± 0.1 nm and indium compositions of 15.7 ± 1 % and 16.2 ± 1 %. Therefore we can conclude the presence of the InGa_{0.95}N prelayer does not perturb the growth of the subsequent QW. A summary of the thickness and indium composition of the QWs in each of the samples is shown in Table 1.

Strain relaxation caused by the presence/inclusion of InGa_{0.95}N prelayers has been reported as a potential cause for the modification of the optical properties [13, 14]. In single QW structures accurate determination of the strain state by XRD is difficult due to the weak intensity of the XRD peaks. In our previous work [18] reciprocal space maps were recorded around the GaN (204) reflection for 10-period multiple QW structures containing 2.5 nm In_{0.156}Ga_{0.844}N QWs with 7.3 nm GaN barriers, with and without a 24 nm thick Si-doped In_{0.05}Ga_{0.95}N prelayer. The multiple QW structures were both found to be fully strained to the underlying GaN pseudo-substrate [18], thus indicating no strain relaxation had occurred due to the inclusion of the InGa_{0.95}N prelayer. Furthermore previous investigations of the strain state in InGa_{0.95}N epilayers have revealed no evidence of strain relaxation in either 160 nm thick In_{0.05}Ga_{0.95}N [23] or 70 nm thick In_{0.12}Ga_{0.88}N [24] epilayers. As these structures reported in previous studies [23, 24] are considerably thicker and of higher indium composition than the prelayer and have a higher total indium content than the entire prelayer plus QW structure, we conclude that the single QWs studied here are fully strained to the GaN pseudo-substrates, and that the inclusion of 24 nm thick In_{0.05}Ga_{0.95}N prelayers within the structure does not cause strain relaxation.

The low temperature (10 K) PL spectra of samples A to D, when measured with an energy density per pulse of 1.54 μJcm^{-2} , are shown in Figure 3. Sample A was measured to have a PL peak emission energy of 2.635 eV, this structure contained no prelayer and is used as a reference. The PL peak emission energies of samples B, C and D were measured to be 2.784 eV, 2.749 eV and 2.756 eV respectively. We attribute the PL peak emission energy to the recombination of independently localised electrons and holes within the QW [25, 26]. Lower energy emission features, separated from the PL peak by 91 meV, were observed in each sample and are attributed to longitudinal-

optical phonon accompanied carrier recombination, within the QW [21]. The additional emission peak measured at 2.921 eV in sample B was also observed in a structure containing only a 24 nm thick Si-doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$ layer with 7.5 nm GaN cap. We attribute the 2.921 eV emission band to either recombination involving acceptors in the prelayer or free electron to a confined hole state produced by band bending at the GaN/InGaN prelayer interface; however the identification of the precise nature of the emission is the subject of further study. The PL emission energy in InGaN/GaN QWs is strongly dependent on the QW thickness, indium composition and the strength of the QCSE [4, 27, 28]. The QW thickness and indium compositions were found to be the same for all the samples, indicating the change in PL peak emission energy may be due to a change in the strength of the QCSE. If the mechanism responsible for the systematic blue shift of the PL peak energies were a reduction in the strength of the QCSE, a significant modification of the radiative recombination rate would be expected for each QW deposited on a prelayer structure.

Figure 3 Low temperature 10 K PL spectra of single QW structures A to D containing no prelayer (A), a Si-doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$ prelayer (B) and Si-doped GaN prelayers grown at 770 °C (C) and 1020 °C (D).

The low temperature PL decay transients measured, with an energy density per pulse of $1.54 \mu\text{Jcm}^{-2}$, at the main peak of the PL spectrum of samples A to D are shown in Figure 4. The characteristic non-exponential na-

ture of the PL decay transients measured for InGaN/GaN QWs is believed to reflect the variation in in-plane electron and hole wave function overlap [25] which occurs as a consequence of the independent localisation of electrons and holes. Owing to the non-exponential nature of the decay transients a single decay constant cannot be used, thus we characterise the PL decay time scale by the time required for the PL intensity to drop by a factor of ten (τ_{10}). The measured τ_{10} decay times for sample A (reference) was $120 \text{ ns} \pm 4 \text{ ns}$, while the τ_{10} times for samples B, C and D were $9 \pm 1 \text{ ns}$, $19 \pm 1 \text{ ns}$ and $19 \pm 1 \text{ ns}$ respectively. The time scale (τ_{10}) over which carrier recombination occurs is strongly dependent on the strength of the built-in electric field [21] and the competition between radiative and non-radiative recombination processes. As the PL integrated intensity measured at 10 K for each QW was approximately the same we attribute the variation in PL decay time to a modification of the built-in electric field, and in turn the radiative recombination rate. A reduction in PL decay time has been measured for every QW deposited on a Si-doped prelayer, irrespective of whether the prelayer is InGaN or GaN. At this point we note the optical properties of the QWs deposited on low temperature and high temperature grown GaN prelayers, samples C and D, are essentially the same, and thus we conclude the growth temperature of the prelayer does not affect the optical properties of the QW.

Figure 4 Low temperature 10 K PL decay transients of samples A to D containing no prelayer (A), a Si doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$ pre-

layer (B) and Si-doped GaN prelayers grown at 770 °C (C) and 1020 °C (D).

AFM measurements on equivalent 5-period $\text{In}_{0.16}\text{Ga}_{0.84}\text{N}/\text{GaN}$ multiple QW structures (not shown) showed that the morphology of “V”-defects at the sample surface were unchanged between a reference structure (equivalent to sample A) and one containing an InGaN:Si prelayer (equivalent to sample B). Therefore we conclude that the inclusion of an InGaN:Si prelayer does not influence the morphology of the “V”-defects in these structures. We note an increase in “V”-defect diameter does occur when including a low temperature grown GaN prelayer, compared to one grown at high temperature. However as shown above, samples C and D (in which the prelayer growth temperature is varied) exhibit identical optical properties despite any variation in “V”-defect size. Therefore we conclude that “V”-defects are not responsible for the modification of the optical properties reported here.

The systematic blue shift of the PL peak emission energy and reduction in PL decay time measured for each sample containing a prelayer structure is consistent with a reduction in the strength of the electric field across the QW, and hence a reduction in the QCSE. To understand the mechanism(s) responsible for the reduction in electric field the nextnano³ simulation package was used to calculate the conduction and valence band profiles in each structure, along with the wave functions of the lowest energy electron and hole confined states in the QWs.

Figure 5 Simulated profiles of the conduction (CB) and heavy

hole valence (VB) bands for samples A, B, C and D, shown as a function of depth from the sample surface. The profiles for samples C and D are indistinguishable. The y-axis of this figure has been defined with respect to a constant Fermi level at 0 eV.

The band profile of semiconductor heterostructures is largely dependent on the position of the Fermi energy with respect to the conduction and valence bands. Mayrock et al. [6] reported that due to a high density of unpaired bonds in Ga-terminated GaN surface a large polarisation charge exists at the sample surface, sufficient to pin the Fermi level at the valence band edge. However in *n*-type material the Fermi level is pinned at the donor energy below the conduction band edge, thus resulting in significant bending of the conduction and valence bands between the GaN/air surface and the *n*-type regions. The calculated conduction and valence bands for samples A to D are shown in Figure 5. In sample A (no prelayer) the electric field across the QW is determined by the interplay between the discontinuities in the spontaneous and piezoelectric polarisations at the InGaN/GaN interfaces. As the piezoelectric polarisation is stronger than the spontaneous polarisation in InGaN/GaN QWs the resultant built-in field is oriented in the $-c$ direction (for Ga-terminated GaN). The inclusion of *n*-type (In)GaN prelayers in samples B, C and D significantly modifies the band profile in the top 40 nm of the structure, by bending of the conduction and valence bands towards the surface. This bending of the conduction and valence bands produces an additional electric field across the QW oriented in the $+c$ direction. The additional contribution of this surface polarisation field therefore reduces the total electric field across the QW in each structure containing a Si-doped (In)GaN prelayer. The electric fields calculated for samples B, C and D were 111 meVnm^{-1} , 149 meVnm^{-1} and 149 meVnm^{-1} respectively, much less than in sample A (254 meVnm^{-1}). The reduction in electric field is consistent with the reduction in the QCSE and the increase in radiative recombination rate, as shown in Table 1.

Figure 6 The measured τ_{10} PL decay times, for each structure A

to D, against the squared overlap of the wave functions calculated for the lowest energy electron and hole subbands within the QW. Dashed line illustrates the Fermi Golden Rule ($\gamma \propto x^{-1}$).

In Figure 6 is shown the variation in calculated electron and hole wave function overlap (squared) and the measured PL decay times, taken from Table 1. The correlation shown in Figure 6 is interpreted as further evidence that the variation in optical properties measured for the single QW structures containing Si-doped (In)GaN prelayers is due to the reduction in the electric field across the QWs.

From Table 1 we also note the largest change in PL peak energy, τ_{10} PL decay time and electric field across the QW, compared to the reference structure A, occurs for the structure containing the Si-doped InGa_N prelayer (sample B). The larger reduction of the electric field suggests the inclusion of indium within the prelayer may have an additional effect on the band profiles of these single QW structures. To investigate the effect on the QW of including indium within the prelayer, a series of band profiles were calculated with incremental variations in indium composition. Each band profile shown in Figure 7 contains the same Si-doping level as used in sample B, but varies from 0 % (equivalent to sample C) to 5 % (sample B) and up to a maximum of 10 % indium. The inclusion of indium within the prelayer was found to further reduce the total electric field across the QW as a function of increasing In fraction as shown in the inset of Figure 7.

Figure 7 Simulated conduction band profile of a single QW structure containing a 24 nm thick Si-doped ($5 \times 10^{18} \text{ cm}^{-3}$) $\text{In}_x\text{Ga}_{(1-x)}\text{N}$ prelayer (equivalent to structure B) shown for incremental indium composition between 0 and 10 %. Inset shows the electric field across the QW as a function of indium composition of the prelayer.

The presence of In in the prelayer of sample B forms an additional charged interface, of opposing sense to the bottom QW interface, due to the large polarisation discontinuity between GaN and InGa_N [24]. The magnitude of the surface charge at this interface is dependent of the indium composition of the InGa_N layer. The close proximity of this additional charged interface to the QW (3 nm) leads to a reduction in the total electric field across the QW. This additional second mechanism accurately describes the further reduction of the electric field across the QW structure containing an InGa_N prelayer and is consistent with the trend shown in Figure 6.

We have successfully shown that the two mechanisms induced by prelayers that are responsible for the reduction in total electric field across a single QW, and subsequent reduction in the QCSE, can be described by the nextnano package. We now go on to apply the nextnano package in order to understand the effect of prelayers on the optical properties of the MQWs studied previously [18]. The MQW structures contained 10-periods of 2.5 nm thick $\text{In}_{0.16}\text{Ga}_{0.84}\text{N}$ QWs separated by 7.5 nm GaN barriers. One sample (MQW-A) contained no prelayer structure, while MQW-B contained the same prelayer as sample B (24 nm Si-doped $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$) separated from the MQW stack by 3 nm of unintentionally doped GaN. The calculated conduction band profiles of these multiple QW structures are shown in Figure 8.

Figure 8 Simulated conduction band profiles of 10-period $\text{In}_{0.16}\text{Ga}_{0.84}\text{N}/\text{GaN}$ multiple QW structures containing either no prelayer (MQW-A) or a 24 nm thick Si-doped ($5 \times 10^{18} \text{ cm}^{-3}$) $\text{In}_{0.05}\text{Ga}_{0.95}\text{N}$ prelayer (MQW-B). Inset shows electric field calculated for each QW in the MQW stack.

An increase in the surface polarisation field was calculated for MQW-B, compared to MQW-A, due to the inclusion of the Si-doped prelayer. The enhanced surface polarisation field in MQW-B leads to approximately a 10 % reduction in the electric field across each QW in the MQW

stack, when compared to MQW-A, shown in the inset of Figure 8. We note however the surface polarisation field in MQW-B is weaker than in the equivalent single QW structure due to the increased separation of the *n*-type prelayer and the GaN/air interface, ~ 100 nm compared to ~ 10 nm, thus leading to a smaller modification of the QW electric field across the QWs.

A further reduction in the electric field of approximately 15 % was calculated for the 1st QW in the stack, when compared to the other QWs in MQW-B, shown in inset of Figure 8. This further modification of only the 1st QW in the stack occurs due to the same additional mechanism as discussed for sample B, the presence of an additional InGa_N/GaN interface in close proximity to the QW. The interaction between the surface charges at this interface, and those interfaces bounding the 1st QW leads to a partial cancellation of the electric field across the QW. This additional effect was not only calculated to depend on the indium composition of the InGa_N prelayer (shown in Figure 7) but also the separation between prelayer and the 1st QW (not shown). This additional mechanism leads to a total reduction of approximately 25 % in the electric field calculated for the 1st QW in the stack compared to MQW-A. The reduction in electric field of the 1st QW leads to modified emission properties of that QW including an increased radiative recombination rate and a reduction in the QCSE. Thus we attribute the additional high energy emission band identified in Figure 1, to the modified emission properties of the 1st QW in the MQW stack [18]. Clearly prelayers can have major consequences for the optical properties of InGa_N/GaN single QW and MQW structures. If prelayers are to be of any benefit to device performance a similar study of LEDs with and without prelayers is important. This has to be performed separately as the band profiles are further complicated by the presence of a p/n-junction.

4 Summary A systematic modification of the optical properties of InGa_N/GaN single QW structures was identified as a consequence of including a range of Si-doped (In)Ga_N prelayers prior to growth of the QWs. A blueshift of the PL peak emission energy and a significant increase in radiative recombination rate was measured for all single QW structures containing prelayers. The inclusion of Si-doped prelayers was found to strongly enhance the surface polarisation field, which in turn reduced the total electric field across the QW, leading to the measured reduction in the QCSE and the increase in the radiative recombination rate. An additional mechanism responsible for a further reduction in the total electric field across the QW was identified when incorporating indium within the Si-doped prelayer. The additional surface charge at the top prelayer interface, in close proximity to the bottom QW interface, was calculated to further reduce the electric field across the QW due to the partial cancellation of the constituent charges. Calculations of the conduction and valence band profiles of 10-period MQW structures indicated the pres-

ence of both of the modification mechanisms identified in the single QW structures. The calculated band profiles of the MQWs indicate a reduction in electric field across each QW in the stack, along with a further reduction in electric field for the 1st QW only, in a structure containing a Si-doped InGa_N prelayer. Thus we attribute the additional emission band identified in our previous report [18] to the modified emission properties of the 1st QW within the multiple QW stack.

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