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# Bayesian Optimization of Hubbard U's for Investigating InGaN Superlattices

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## 1. The choice of orbitals for DFT+U

As discussed in the main text, the Hubbard U corrections have been applied to different orbitals in the literature. This is likely due to the fact, that the frontier orbitals are formed by hybridized d- and p-orbitals of Ga/In, as well as p-orbitals of N. Most often, the correction is applied to the d-orbitals of Ga or In [14,23,24]. Terentjevs et al [23,24] applied, in addition, the correction to the 2p-orbital of nitrogen. Yu et al [26] applied the U-terms to the p-orbitals of In and As, when optimizing the Hubbard U's for InAs. Because of that, we also tried different possibilities. In particular, we considered applying the Hubbard correction to the following combinations of the orbitals:

1. Ga or In-d-orbitals;
2. Ga-d or In-d + N-p orbitals;
3. Ga-p or In-p + N-p orbitals.

When two U-parameters were simultaneously optimized, we increased the number of the Bayesian optimization steps to 100 to ensure convergence. The search range for the d-orbitals was set to 1 to 10 eV, while the range for the p-orbitals was wider (−10 to 10 eV). The resulting optimized U's are summarized in Table S1.

**Table S1.** Optimized U-values for different combinations of the orbitals involved into the optimization.

| Material | Involved Orbitals | Optimized U's (eV) | | | Max f(U) (eV²) |
|---|---|---|---|---|---|
| | | Ga-p/In-p | Ga-d/In-d | N-p | |
| GaN | Ga-d | - | 5.61 | - | −0.1538 |
| | Ga-d + N-p | - | 4.50 | −7.70 | −0.1521 |
| | Ga-p + N-p | −6.13 | - | -10* | −0.1726 |
| InN | In-d | - | 7.14 | - | −0.1247 |
| | In-d + N-p | - | 7.32 | 0.98 | −0.1246 |
| | In-p + N-p | −6.36 | - | −10* | −0.1275 |

* the lowest value within the allowed range, likely still not the true maximum.

The combination of Ga/In-p + N-p has proven troublesome, since it soon became clear that even the lowest allowed value of U's (−10 eV) for the p-orbitals of N did not seem to be enough. In addition, the band gap was found to be underestimated more than in other 2 combinations (0.65 eV for InN and 3.27 eV for GaN). Hence, we decided to discard Ga/In-p + N-p. The combination of Ga/In-d + N-p appeared to perform best to optimize the objective function and delivered better band gaps, as compared to the previous case. However, applying these parametrizations is complicated due to expectedly different values of the U-parameters of the nitrogen p-orbitals (−7.70 eV in GaN vs +0.98 eV in InN). Finally, when the correction is applied only to the d-orbitals of either Ga or In, it results in a marginally worse value of the objective function maximum, while retaining virtually the same value of the band gap as in the previous combination. Therefore, we believe that

applying the Hubbard U-term only to the d-orbitals of Ga and In is the best strategy to deal with composite materials made of GaN and InN simultaneously.