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The largest number not of the form $ax + by$ with $x, y > 0$, $gcd(a, b) = 1$ is $ab - a - b$.

Proof:

First, we will show that $ax + by = ab - a - b$ is not possible if $x, y > 0$.
Then we will show that $ax + by$ can be any value higher than $ab - a - b$

Assume to the contrary that there exist an (x, y) such that $ax + by = ab - a - b$
Then, taking both sides mod a, we get that:

$by \equiv -b\ (mod\ a) \Rightarrow y \equiv -1\ (mod\ a)$, since $(a, b) = 1$

Since $y > 0$, we have $y \geq a-1$. But $ax+b(a-1) = ax+ba-b \geq ba-b > ab-a-b$, and we have a contradiction.

Now, to show that $ax + by$ can be any value greater than $ab - a - b$

WLOG let $a > b$
Let $ax + by = ab - a - b + k$, $k > 0$

Then, mod a, we have $by \equiv k - b\ (mod\ a) \Leftrightarrow b(y + 1) \equiv k\ (mod\ a) \Leftrightarrow y \equiv k * b^{-1} - 1\ (mod\ a)$

Where b^{-1} is the inverse of b mod a

Then $x = \frac{ab-a-b-by+k}{a}$.

We just need to show that $x \geq 0$

This is equivalent to showing that $ab - a - b - by + k > 0$

If $y \leq a - 2$ then $ab - a - b - b(a - 2) + k = a - b + k > 0$, and (x, y) exists.

If $y = a-1$, then $k \equiv 0\ (mod\ a) \Rightarrow k = za, z \geq 1$, and $ab-a-b-b(a-1)+za = (z - 1)a \geq 0$

So all the cases have been covered, and $ax + by$ can be any number greater than $ab - a - b$.

$\square$