exchanging indices j and k. Here, $\{\phi_{e_i}\}$ ($\{\phi_{h_j}\}$) are the electron (hole) single-particle wave functions, and $\bar{\epsilon}(\boldsymbol{r}, \boldsymbol{r}')$ is the

(i.e., $\varepsilon_{e_{th+i}} \approx E_{th} = E_g$); (ii) $\varepsilon_{e_{th+i}}$ is well above threshold. Condition (i) corresponds to situations where the excess energy Δ is only enough to excite a valence electron from a state close to the band edge ($h_1 - h_4$) into e_1. In case (ii), instead, Δ

Direct Carrier Multiplication by an Electron in the Presence of a Hole (Case II) (Figure 1b). To allow a comparison with case I above, we consider photogenerated electron−hole pairs where the electron occupies a level e_{th+i} ($i = 1,..., 8$) and the hole occupies the state at the top of the valence band. This configuration can either be generated directly by the absorption of photons with energies $h\nu_i = 2E_g + \delta\varepsilon_i$ (with $\delta\varepsilon_i = 4-60$ meV), in which case all excess energy $h\nu_i - E_g = E_g + \delta\varepsilon_i$ is given to the electron whereas the hole has no kinetic energy (a typical scenario, for example, in $Si_{0.32}Ge_{0.68}$ for $h\nu = 2E_g$[14,15]) or it can be the result of a higher-energy excitation. In the latter case, the excess energy $h\nu_i - E_g$ might be distributed between the electron and the hole. However, as the hole relaxes to the top of the valence band with characteristic times that are much smaller than our calculated DCM lifetimes, we can safely assume it to occupy its ground state in our initial DCM configuration. For DCM calculations, we consider the same states and follow the same procedure as we did in the case of the charged dot. The AC lifetimes are obtained by summing over 30 hole final states $\{h_{mi}\}$ whose energies are centered around $\varepsilon_{h_1} - E_g$. We find the following:

(a) For excess energies $\Delta = \varepsilon_{e_{th+i}} \approx E_{th}$ (Figure 5a), the DCM lifetime calculated in the presence of a hole is about a factor of 2 larger than that computed without it, both on average ($\langle\tau_{DCM}^{(w.h)}\rangle = 122$ ps, $\langle\tau_{DCM}^{(noh)}\rangle = 74$ ps) and at the position of the arrow [$\tau_{DCM}^{(w.h)}(\downarrow) = 76$ ps, $\tau_{DCM}^{(noh)}(\downarrow) = 35$ ps]. When a (photogenerated) hole is in its ground state, in fact, the number of final states $|h_m, e_1\rangle$ available to the e−h pair created via DCM is reduced. This leads to an increase in the lifetime compared to that of the configuration with no hole.

(b) Both DCM (with a hole) and AC lifetimes are of about the same order of magnitude, ∼100 ps, for excess energies $\Delta \approx E_{th}$. The DCM process is, however, slightly faster, with an average lifetime of 122 ps compared to the AC average lifetime of 132 ps. But more importantly, the DCM lifetime with a hole is about 2/3 of τ_{AC} at the arrow, yielding a DCM efficiency of 61% for the value of $\varepsilon_{e_{th+i}} - E_g$ calculated for this specific dot size.

(c) The presence of a hole in h_1 has a much smaller effect on the DCM lifetime for higher excess energies ($\varepsilon_{e_{th+8}}$), as shown in Figure 5b. This occurs because in this case the energy of the electron is (∼1

the VBM, see Figure 2) found[36] in spherical dots between levels h_4 and h_5: in that energy range, there are no energy-conserving transitions $h_n \rightarrow e_i$ available to the DCM process; the next transition $h_5 \rightarrow e_1$ is more than 100 meV higher in energy (Figure 2). Instead, such a gap does not exist within deep hole states (the ones involved in the AC process); therefore, the AC lifetime is almost constant for all energies. As a consequence, τ_{DCM}, which is smaller than τ_{AC} for excess

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