Low Surface Recombination in Hexagonal SiGe Alloy Nanowires

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Low Surface Recombination in Hexagonal SiGe Alloy Nanowires: Implications for SiGe-Based Nanolasers


ABSTRACT: Monolithic integration of silicon-based electronics and photonics could open the door toward many opportunities including on-chip optical data communication and large-scale application of light-based sensing devices in healthcare and automotive; by some, it is considered the Holy Grail of silicon photonics. The monolithic integration is, however, severely hampered by the inability of Si to efficiently emit light. Recently, important progress has been made by the demonstration of efficient light emission from direct-bandgap hexagonal SiGe (hex-SiGe) alloy nanowires. For this promising material, realized by employing a nanowire structure, many challenges and open questions remain before a large-scale application can be realized. Considering that for other direct-bandgap materials like GaAs, surface recombination can be a true bottleneck, one of the open questions is the importance of surface recombination for the photoluminescence efficiency of this new material. In this work, temperature-dependent photoluminescence measurements were performed on both hex-Ge and hex-SiGe nanowires with and without surface passivation schemes that have been well documented and proven effective on cubic silicon and germanium to elucidate whether and to what extent the internal quantum efficiency (IQE) of the wires can be improved. Additionally, time-resolved photoluminescence (TRPL) measurements were performed on unpassivated hex-SiGe nanowires as a function of their diameter. The dependence of the surface recombination on the SiGe composition could, however, not be yet addressed given the sample-to-sample variations of the state-of-the-art hex-SiGe nanowires. With the aforementioned experiments, we demonstrate that at room temperature, under high excitation conditions (a few kW cm$^{-2}$), the hex-(Si)Ge surface is most likely not a bottleneck for efficient radiative emission under relatively high excitation conditions. This is an important asset for future hex(Si)Ge optoelectronic devices, specifically for nanolasers.

KEYWORDS: nanowires, hexagonal SiGe, surface passivation, SiGe, silicon photonics, surface recombination velocity, atomic layer deposition, direct bandgap

1. INTRODUCTION

The continuous pursuit of technological progress has raised increasing interest in optical communication (e.g., optical interconnects$^{1–3}$) and light processing devices (e.g., lab-on-a-chip$^4$), which both require efficient light sources and detectors compatible with current silicon electronics. Unfortunately, one of silicon’s main drawbacks is its incapability to efficiently emit and absorb light. This has sparked interest in implementing other semiconductors on silicon substrate. Among the candidates are III–V materials, such as InP and GaAs. These materials have a direct bandgap, but they are rare and expensive$^5$ and have a relatively large lattice mismatch with the Si substrate,$^6$ which make them very difficult to integrate.

A new and very promising approach is based on germanium. This is a semiconductor that is chemically well matched to silicon and readily used in the semiconductor industry.$^7$ Although the natural crystal structure of Ge (diamond cubic) does not exhibit a direct bandgap, it was predicted by Joannopoulos et al.$^8$ that the hexagonal crystal structure does. More recent and much more accurate theoretical works have confirmed this prediction for hexagonal Ge$^9$ and for hexagonal SiGe nanowires.$^{10}$ Fadaly et al.$^{11}$ finally showed with experimental evidence that the hexagonal crystal structure of Ge shows indeed efficient direct-bandgap emission. The use of GaAs/Ge core/shell nanowires (NWs) proved key to realizing hexagonal germanium (hex-Ge). Besides this, Fadaly et al.$^{11}$ demonstrated that the direct bandgap is tunable from 0.3 to...
0.7 eV by alloying the hex-Ge with 0–35% Si to create hexagonal silicon germanium (hex-SiGe). This makes hex-SiGe alloys a very promising candidate as silicon-compatible light emitters and absorbers, such as LEDs, lasers, and photodetectors compatible with Si-photonics circuitry. The scientific impact of hexagonal polytypes of Si and Ge has also been discussed in a recent review article. Optical characterization of the hexagonal GaAs/SiGe core/shell nanowires by Fadaly et al. revealed that the photoluminescence (PL) efficiency is significantly lower at room temperature (~300 K) than at a few Kelvin (~6 K). The latter suggests the presence of nonradiative recombination pathways. For nanostructures such as nanowires, surface-to-volume ratios are relatively large and surface defects can consequently play a much more pronounced role than in planar structures. Surface recombination via defects at the surface of the hex(Si)Ge crystals is therefore one of the primary suspects.

In this work, it is investigated whether and to what extent surface recombination is limiting the PL of hex-Ge and hex-SiGe alloys with the objective to clarify how the radiative emission of hex-SiGe at room temperature can be enhanced. It is however beyond the scope of this paper to investigate the detailed compositional dependence of the surface recombination mechanism since sample-to-sample variations of these state-of-the-art nanowires are presently too large to clearly observe compositional trends. For this purpose, temperature-dependent photoluminescence measurements were performed on nanowires with and without effectively proven surface passivation schemes. Case studies were performed for ultrathin (~25 nm) passivation layers and stacks that were earlier successfully studied on planar cub-Ge and cub-Si substrates: aluminum oxide (Al₂O₃), a stack of amorphous silicon and aluminum oxide (a-Si:H/Al₂O₃), and a stack of phosphorus oxide and aluminum oxide (POₓ/Al₂O₃). These passivation schemes were selected as they have demonstrated very good passivation of cub-Ge and cub-Si surfaces with effective surface recombination velocities (S_eff) between S_eff ≈ 300 cm/s and S_eff ≈ 2 cm s⁻¹, which is very low compared to typical recombination velocities of, for example, InP surfaces (10⁻¹⁰–10⁻⁹ cm s⁻¹) or GaAs surfaces (4 × 10⁻¹⁰–10⁻⁹ cm s⁻¹). Besides this, these layers were selected because they are well documented and can be deposited by atomic layer deposition (ALD). This is a CMOS-compatible method that can provide excellent conformity on high-aspect-ratio structures such as nanowires. The latter makes these passivation layers preferred over, for example, an epitaxially grown Si passivation layer for which the required monolayer thickness control is challenging on 3D nanostructures such as these nanowires. The authors have also omitted higher-bandgap III-V semiconductors as a passivation layer since such materials are not compatible with the idea of using hex-SiGe for a Si-based laser. In addition to the investigation of these passivation schemes, time-resolved photoluminescence (TRPL) measurements were performed on unpassivated hex-SiGe nanowires as a function of their diameter. With these experiments, we demonstrate that at room temperature, under high excitation conditions (a few kW cm⁻²), no large improvements or degradations of the PL occur after applying the passivation schemes. Moreover, no increase in lifetime was found with an increasing nanowire diameter. These findings suggest that in this regime the surface of the hex-(Si)Ge is most likely not a bottleneck for efficient radiative emission, which is an important advantage for future hex-(Si)Ge optoelectronic devices such as nanolasers.

2. EXPERIMENTAL DETAILS

2.1. Sample Characteristics. The hexagonal GaAs/SiGe core/shell nanowires studied in this work were grown using an identical...
Both the nanowire characteristics and the passivation schemes applied to them are listed. The listed pre-deposition surface treatments and the post-deposition anneal temperatures ($T_{\text{pDA}}$) were optimized for the surface passivation of each passivation scheme (see earlier work). However, surface recombination velocities of planar cubic SiGe are still very rarely reported in the literature and are therefore not given in the table. Note that for cubic-Si and cubic-Ge the optimum growth conditions for a-Si:H regarding surface passivation are slightly different. The native oxide is removed in situ by the PO$_2$/Al$_2$O$_3$ deposition process through a self-cleaning effect.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Sample Name</th>
<th>Nanowire Composition</th>
<th>Diameter ($\mu$m)</th>
<th>Length ($\mu$m)</th>
<th>Passivation Scheme</th>
<th>Surface Treatment</th>
<th>$T_{\text{pDA}}$ ($^\circ$C)</th>
<th>$S_{\text{PL}}$ (cm$^2$ s$^{-1}$) on cub-Ge</th>
<th>$S_{\text{nl}}$ (cm$^2$ s$^{-1}$) on cub-Si</th>
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</thead>
<tbody>
<tr>
<td>Passivation</td>
<td>H05916</td>
<td>Si$_2$Ge$_3$</td>
<td>0.43 ± 0.07</td>
<td>1.6 ± 0.4</td>
<td>Al$_2$O$_3$ (22 nm)</td>
<td>1% HF$_{\text{wa}}$</td>
<td>425</td>
<td>$\sim$300$^{13}$</td>
<td>$&lt;6^{4,15}$</td>
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<tr>
<td>Passivation</td>
<td>H07771</td>
<td>Si$_2$Ge$_3$</td>
<td>2.08 ± 0.03</td>
<td>8.3 ± 0.3</td>
<td>a-Si:H/Al$_2$O$_3$ (2/11 nm)</td>
<td>20% HBF$_{3}$</td>
<td>325</td>
<td>$\sim$2.7$^{16}$</td>
<td>$\sim$1.8$^{6}$</td>
</tr>
<tr>
<td>Passivation</td>
<td>H06950</td>
<td>Si$_2$Ge$_3$</td>
<td>0.617 ± 0.02</td>
<td>5.62 ± 0.02</td>
<td>PO$_2$/Al$_2$O$_3$ (4/10 nm)</td>
<td>none$^c$</td>
<td>250</td>
<td>$\sim$8.9$^{17}$</td>
<td>$&lt;6^{4,19}$</td>
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<tr>
<td>Diameter Series</td>
<td>H06950-2</td>
<td>Si$_2$Ge$_3$</td>
<td>0.617 ± 0.02</td>
<td>5.62 ± 0.02</td>
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<td>Diameter Series</td>
<td>H06948</td>
<td>Si$_2$Ge$_3$</td>
<td>1.0 ± 0.1</td>
<td>5.2 ± 0.9</td>
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<tr>
<td>Diameter Series</td>
<td>H06988</td>
<td>Si$_2$Ge$_3$</td>
<td>1.2 ± 0.1</td>
<td>5.86 ± 0.07</td>
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<tr>
<td>TEM</td>
<td>H05895</td>
<td>Si$_2$Ge$_3$</td>
<td>0.06 ± 0.005</td>
<td>1.7 ± 0.1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

“Both the nanowire characteristics and the passivation schemes applied to them are listed. The listed pre-deposition surface treatments and the post-deposition anneal temperatures ($T_{\text{pDA}}$) were optimized for the surface passivation of each passivation scheme (see earlier work). However, surface recombination velocities of planar cubic SiGe are still very rarely reported in the literature and are therefore not given in the table. Note that for cubic-Si and cubic-Ge the optimum growth conditions for a-Si:H regarding surface passivation are slightly different. The native oxide is removed in situ by the PO$_2$/Al$_2$O$_3$ deposition process through a self-cleaning effect.”
thick Al₂O₃ film. As expected, the ALD process allows thus for good conformality on these nanostructures. The film measures about 22 nm, which is in line with the nominal growth per cycle (GPC) of this process (GPC ≈ 1.1 Å/cycle). Earlier work by Fadaly et al. (extended data Figure 6b,d) shows no signs of clustering or segregation of either Si or Ge in the hexagonal SiGe nanowires. Energy-dispersive X-ray spectroscopy (EDX) performed in this work on similar SiGe nanowires reveals that after deposition of ALD Al₂O₃, no clear signs of clustering or segregation appear (Section 1 in the Supporting Information).

The integrated photoluminescence of hex-Si₀.₂₃Ge₀.₇₇ nanowires with and without an Al₂O₃ passivation film is shown in Figure 2d. Typical spectra can be found in Section 3 in the Supporting Information. Figure 1d shows the integrated PL as a function of inverse temperature for SiGe NWs with and without Al₂O₃ passivation film. The PL intensities of the as-grown wires (black) and the passivated wires (red) were normalized at 4 K. The latter implies that the vertical axis also represents the internal quantum efficiency. As expected, the internal quantum efficiency. As expected, the internal quantum efficiency. As expected, the internal quantum efficiency. As expected, the internal quantum efficiency.
quantum efficiency decreases with increasing temperature due to the activation of nonradiative recombination mechanisms. For both the passivated and unpassivated wires, the quenching of the PL with temperature seems to be characterized by two knees. This implies the presence of two separate nonradiative recombination channels. Consequently, the data is fitted with the Arrhenius eq (eq 2, see Section 2) using two terms, yielding activation energies of $E_{a,1} \approx 3.2–3.8$ meV and $E_{a,2} \approx 25–27$ meV (details of fit parameters such as these activation energies can be found in Section 2 in the Supporting Information). The fits are represented by solid lines in Figure 2d. When the passivated and as-grown wires are compared, it also becomes clear that the PL of the Al$_2$O$_3$ passivated wires shows less quenching with increasing temperature. This is also reflected by the slightly lower quenching rate for the passivated wires ($C_{2,pass} \approx 100$ au vs $C_{1,unpass} \approx 183$ au) obtained by the Arrhenius fit, and the 66% higher absolute PL intensity at the highest measurable temperature (90 K). If the nonradiative recombination can be (partially) attributed to the surface, then there is a slight reduction of the surface recombination.

3.2. Effect of a-Si:H/Al$_2$O$_3$ Passivation. Figure 3a–d shows TEM images of a single hexagonal SiGe nanowire after it has been subjected to 40 s Plasma-Enhanced Chemical Vapor Deposition (PECVD) a-Si:H and subsequently 100 cycles of plasma-enhanced ALD Al$_2$O$_3$. The properties of the nanowire are identical to those described in Figure 1a–c. The Al$_2$O$_3$ capping layer has an equal thickness of 10.5 $\pm$ 0.5 nm along the whole length of the nanowire, which indicates a very conformal coating. The PECVD a-Si:H interlayer is present along the whole nanowire, as well, which is important. When considering the film thickness, it is observed that the a-Si:H is substantially thicker on the top (3.0 $\pm$ 0.5 nm) than on the side walls (1.5 $\pm$ 0.5 nm). The lower thickness on the sides of the nanowire can be attributed to a lower flux of reactive plasma species during PECVD of a-Si:H.

The effect on the integrated PL of the a-Si:H/Al$_2$O$_3$ stacks, which can suppress recombination velocities on cub-Ge down to $S_{a,2} \approx 3$ cm/s (Table 1), is shown in Figure 3e. The integrated PL intensities of the as-grown hex-SiGe wires (black) and the passivated wires (red) have been normalized at 7 K (typical spectra are shown in Section 3 in the Supporting Information). The PL quenching with temperature in Figure 3e seems to be characterized by two knees for both the passivated and unpassivated wires, similar to that in Figure 2d. The Arrhenius fits of the data (solid lines) yield for the activation energies ($E_{a,1}, E_{a,2}$) of the as-grown and passivated wires similar values of 2.69 and 2.58 meV for $E_{a,1}$ and 22 and 21 meV for $E_{a,2}$. When comparing the passivated and as-grown wires, it becomes evident that they show very similar behavior. The PL intensity of the passivated wires decreases a little bit less with increasing temperature ($C_{2,pass} \approx 20$ au vs $C_{1,unpass} \approx 30$ au). This hints at a reduction in nonradiative recombination pathways, presumably related to the surface. The reduction is, however, slight. Also note that for even higher excitation densities, nonradiative recombination becomes relatively less important. The latter implies that for higher excitation densities, the differences between the quenching rates of the passivated and as-grown wires will be reduced to an even smaller difference. Another experiment performed with wires of pure hexagonal germanium also shows this experimentally (Section 3 in the Supporting Information).

3.3. Effect of PO$_3$/Al$_2$O$_3$ Passivation. The conformality of the PO$_3$/Al$_2$O$_3$ deposition process used in this work has already been demonstrated on nanowires in earlier work by Black et al. and Theeuwes et al. For this stack, we therefore focus only on the changes in photoluminescence as a consequence of applying this stack on the surface of hex-SiGe nanowires. Figure 4 shows the integrated PL intensity as a function of the inverse temperature for hex-Si$_{0.12}$Ge$_{0.88}$ nanowires without and with a PO$_3$/Al$_2$O$_3$ passivation stack. The integrated PL intensities are obtained for an excitation density of 4.1 kW/cm$^2$ and normalized to their respective intensity at 4 K. The data is fitted with the Arrhenius equation (solid lines through the data, parameters listed in Section 2 in the Supporting Information).
Figure 5. Overview of the integrated photoluminescence of various hex-Ge (closed symbols) and hex-SiGe (open symbols) nanowire ensembles passivated with various passivation schemes. The integrated PL is normalized with respect to the integrated photoluminescence of the same nanowires without a passivation scheme. The nanowires are measured at 293 K with the exception of the SiGe nanowires with the AlOx, a-Si:H/AlOx and POx/AlOx passivation for which the highest measurable temperatures were 90, 149, and 160 K, respectively. Excitation densities used for the measurement vary slightly per sample but lie between 1 and 7 kW cm\(^{-2}\). The exact dimensions and composition of the nanowires and the measurement conditions associated with each data point are listed in Section 4 in the Supporting Information.

provided in Section 4 in the Supporting Information. Figure 5 shows that in most cases the passivation film slightly enhances the PL except for the thermal ALD AlOx film and the POx/AlOx stack. Second and more generally, the presented passivation schemes show each a relatively mild impact on the integrated photoluminescence of the hex-Ge and hex-SiGe nanowires. To be more precise, the deviation from the as-grown nanowires (dashed line) is mostly within a factor of 2. The latter indicates a relatively small influence of the surface on the photoluminescence.

3.5. Lifetime vs Nanowire Diameter. To further elucidate the importance of the surface, TRPL measurements were performed on individual unpassivated nanowires to determine the carrier recombination lifetime as a function of their diameter. The results of the experiments are presented in Figure 6. Figure 6a–c shows SEM images of the nanowire ensembles from which individual nanowires were taken and studied within this experiment (Section 2). In Figure 6d, the weighted average lifetime (\(\tau\)) as a function of nanowire diameter at room temperature is displayed. For each data point, about 5–7 nanowires were averaged. The composition of SiGe nanowires is chosen to be Si\(_{0.23}\)Ge\(_{0.77}\), which is approximately in the middle of the composition range that yields a direct bandgap (Si\(_{0.15}\)Ge\(_{0.85}\)–Si\(_{0.35}\)Ge\(_{0.65}\)). From Figure 6d, a clear correlation becomes visible between the diameter and lifetime: the thicker the nanowires, the lower the lifetime. This is exactly the opposite of what one expects if the lifetime is surface-limited. This data suggests therefore that at room temperature the native hex-SiGe surface is not the limiting factor but rather nonradiative recombination centers in the bulk. These bulk recombination centers may not be distributed homogeneously throughout the nanowire and may increase with diameter, leading to the diameter dependence of the lifetime as observed in Figure 6d. This was for example the case for the so-called I\(_3\) basal stacking fault,\(^{41}\) a structural defect in hex-SiGe that does however not act as a recombination center.\(^{41}\) Since the As concentration is rather homogeneous after the first 20 nm from the GaAs core,\(^{11}\) the diameter dependence of the lifetime seems unlikely to be related with relatively high and unintentional As content. Note finally that for temperatures substantially lower than room temperature, it was shown by Fadaly et al.,\(^{11}\) that recombination becomes purely radiative; i.e., nonradiative recombination is no longer dominant. According to their work, typical temperatures below which recombination becomes purely radiative are <40 K or even <200 K depending on the sample.\(^{41}\) In this work, the transition temperature for the wires presented in Figures 2, 3, and 4 seems to be around 50 K.

4. DISCUSSION

When considering the nanowires that were studied in more detail (Figures 2–4), we observe some improvements by the AlOx and a-Si:H/AlOx passivation schemes. The effect of the...
PO$_4$/Al$_2$O$_3$ passivation scheme remains somewhat less clear. Despite some influence of the passivation schemes, the quenching of the PL from 4 K to room temperature remains fairly similar for the nanowires with and without passivation: between one and 2 orders of magnitude. The latter indicates that the nonradiative recombination channels are still very active after applying the passivation films. This could either mean that the passivation is not very effective or that the surface is not the main cause for nonradiative losses in these nanowires. Considering that these passivation schemes have demonstrated very low surface recombination velocities on cubic germanium and cubic silicon, the former seems unlikely. Moreover, the quick screening of several other passivation schemes (Figure 5) shows that all tested films and pretreatments have only mild impact on the PL, which also hints at a small influence of the surface on the PL rather than a strongly limiting surface. These two reasons make it most likely that the surface is not one of the main nonradiative recombination channels in these nanowires. This statement is further reinforced by time-resolved photoluminescence measurements of SiGe nanowires with various diameters. In this experiment, no increase of the lifetime was found for an increasing diameter (Figure 6). This result also implies a surface that hardly influences the PL of the SiGe.

Since the lifetime of the nanowires seems not surface-limited, the measured lifetimes in Figure 6 represent merely a lower limit for the surface lifetime ($\tau_s$). From the latter, an upper limit for the surface recombination velocity of the native hex-SiGe surface can be estimated ($S_{\text{eff,max}}$). Using the data of the nanowire with the smallest diameter, this results in

$$\tau_s = \frac{1}{S_{\text{eff,max}} \frac{\text{NW volume}}{\text{NW outer surf area}}} = \frac{1}{\frac{S_{\text{eff,max}}}{2\pi r^2 + 2\pi r h}} \approx 3.8 \times 10^4 \text{cm s}^{-1},$$

where $h$ represents the length of the nanowire, $r$ is the radius, and $r_{\text{core}}$ is the radius of the GaAs core. This upper limit indicates a surface recombination velocity of the native hex-SiGe surface that is relatively low compared to typical surface recombination velocities of, for example, GaAs ($S_s \approx 4 \times 10^1$–10$^7$ cm s$^{-1}$) and InGaAs ($S_s \approx 0.5 \times 10^4$ cm s$^{-1}$). Compared to InP ($S_s \approx 10^2$–10$^4$ cm s$^{-1}$), these passivation schemes lead to only relatively small changes in the integrated photoluminescence compared to the nanowires with a native surface. Additionally, the lifetime of the nanowires with a native surface was not found to decrease with decreasing diameter. Considering these observations, we conclude that the hex-SiGe surface is most likely not strongly influencing, hence not limiting, the PL in the high excitation regime. An upper limit for the surface recombination of $S_{\text{eff,max}} < 3.8 \times 10^4$ cm s$^{-1}$ was estimated. The strong decrease in internal quantum efficiency with increasing temperature may consequently stem predominantly from nonradiative recombination in the bulk of the hex-SiGe. The results of this research imply that for hex-SiGe-based devices with micrometer dimensions operating at relatively high excitation densities, like nanolasers, surface recombination will most likely not be a bottleneck. Despite not being strongly limiting, direct contact between the SiGe surface and layers that facilitate (very) high carrier recombination rates (e.g., metals, unannealed plasma-processed dielectrics, etc.) should still be avoided. The result of this research is considered useful for the realization of a hex-SiGe-based laser, which is an important milestone on the roadmap of silicon photonics.

5. CONCLUSIONS

Several ultrathin (<25 nm) passivation layers that have demonstrated low surface recombination velocities on cubic germanium and silicon have been applied to hex-Ge and hex-SiGe nanowires. At room temperature and relatively high excitation densities (1–8 kW cm$^{-2}$), these passivation schemes lead to only relatively small changes in the integrated photoluminescence compared to the nanowires with a native surface. Additionally, the lifetime of the nanowires with a native surface was not found to decrease with decreasing diameter. Considering these observations, we conclude that the hex-(Si)Ge surface is most likely not strongly influencing, hence not limiting, the PL in the high excitation regime.
**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsanm.3c05770.

Energy-dispersive X-ray spectroscopy (EDX) of SiGe nanowires coated with ALD Al2O3: parameters Arrhenius fits; photoluminescence spectra and excitation dependence of PL; details about the nanowires and the different passivation schemes used in Figure 5; time-resolved photoluminescence data; estimation of the band offset between hex-SiGe and wurzite GaAs; deterioration of the PL after deposition of plasma-enhanced ALD Al2O3; and degradation of the PL over time (PDF)

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**Notes**

The authors declare no competing financial interest.

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