2.3 Swirl Defects in Si (Investigated in a HVTEM)

2.3.1 Background

- When you grew a large, dislocation-free single crystal of silicon around 1975 you were entitled to expect that the freshly crystallized Si contained a number of intrinsic point defects close to the equilibrium concentration at the melting point, and that these point defects were vacancies. Since the crystal contained no sinks like dislocations or grain boundaries where those vacancies could disappear during cooling down, you were also justified in expecting some vacancy agglomerates at room temperature. Forming agglomerates like dislocation loops or voids, after all, was the only way to reduce the vacancy concentration to the very small equilibrium value at room temperature.
  - It was thus no big surprise that (rather tricky) defect etching techniques or X-ray investigations did indeed show that some small defects could be found in some of the as-grown Si crystals. In the case of the Siemens crystal growers you found on occasion something like what is shown in the first picture of my picture collection.
  - Yes, Siemens (at its Munich location) was still involved in Si crystal growing in 1975, if not for much longer. Today it has been long forgotten that Siemens sort of "invented" Si technology. We still have the "Siemens process" for making "high-purity silicon and that, is mostly due to the efforts of **Eberhard Spenke** (and others) from Siemens Research. Look it up!
- With hindsight, we now know that we should have expected a complicated dance of Si self-interstitials, vacancies, oxygen interstitials and (substitutional) carbon atoms., all present at roughly comparable and very low concentrations (in comparison with metals). With luck your crystal produced a relatively simple defect structure - and that's what we see in the typical swirl pattern shown here.
  Knowing from your etch pattern that you had swirl defects in your Si crystal, you certainly wanted to know what, exactly, you had there. Defects are never good news for making integrated circuits, and knowing details about the nature of these defects might help you to avoid them. The thing to do, clearly, was to look at these bugggers in a transmission electron microscope (TEM). Unfortunately, if you tried to do this, you run into two almost insurmountable problems:
  1. After defect etching you see where the defects has been. Typically defects etching works because Si dissolves more rapidly around a defect, producing a small etch pit on the surface. Well visible in a (optical) microscope - but your defect is actually no longer there, it was etched out. So you must look for the defects in an un-etched part of the crystal. Then you run into the second problem
  2. The defect density is so low that it is very unlikely to find one in a typical TEM specimen. A typical TEM specimen is a disc of 3 mm diameter but only a small part of that disc is thin enough to be transparent to the electron beam.
- The Siemens people around **Bernd Kolbesen** (in particular G. Schuh) overcame both problems by some rather ingenious tricks:
  1. They developed an etching technique that produced a hillock instead of a pit - at least for the bigger "A-swirl" defects. The defect thus was still there, below the hillock. The trick for this was to etch at low temperatures. Why this works the way it does nobody knows. The smaller and more ubiquitous "B-swirls", by the way, still produced etch pits.
  2. The developed a "large area thinning technique" allowing to thin the whole wafer down to around 5 µm. You now could see the hillocks looking through the wafer and that allowed to cut out (by some “simple" lithography) small discs, almost ready to go into a TEM.
- The only remaining problem was that a thickness of 5 µm is too large for a regular TEM - but not fir a high voltage TEM (HVTEM). My Max-Planck-Institut was the only owner of such an (expensive) thing in Germany, and I was the guy supposed to use for some Si research. As a matter of course the head of the institute (Prof Seeger) referred the Siemens people top me. I agreed to look at their samples - in exchange to getting some Si samples for my own research work (the electron irradiation business mentioned before).
  - A win-win situations, as it turned out. With rather big winnings for all (like a prize from the venerable Academy of Science in Göttingen. We also became quite famous all over the world of Si crystal growing science – all of the 50 or so people involved knew what we found!
- What did we find? Simple. A-swirls - the bigger ones producing etch hillocks - were agglomerates of Si self-interstitials; dislocations loops of interstitial character to be precise. This induced us to proclaim that Si self-interstitials must be the dominating intrinsic point defect in thermal, and that was seen as abominable heresy by almost all the established popes of crystal defects. Except for one: My boss, Prof. Alfred Seeger. He (together with K. P. Chik) had reasoned already in 1968 that interstitials should dominate in silicon. As it turned out much later, his reasoning was based on questionable experimental evidence, but he was right in principle. A small war broke out, which was fun (at least for me). From today's point of view, Si interstitials are there, but so are vacancies and everything is far more complicated than anyone could have imagined back in the seventies.
  - Enough said. Read the papers if you want to learn more

2.3.2 Major Publications

- The swirl research produced a number of publications and conference contributions but here I will only list a few:

5 FÖLL, H., WILKENS, M.: **A simple method for the analysis of dislocation loops. Phys. Stat. Sol. (a) 31 (1975) 519**
97 citations
I actually improved upon the existent way of analyzing dislocation loops by the “inside – outside contrast method”. This became my most prominent paper for quite a while

6 FÖLL, H., KOLBESEN, B.O.: **Agglomerate von Zwischengitteratomen (Swirl-Defekte) in Silizium - Ihre Bedeutung für Grundlagenforschung und Technologie. Jahrbuch der Akademie der Wissenschaften in Göttingen (1976), p. 27 (invited paper)**
The paper we had to write because we won a price!, It is interesting in a small way because a drawing I made for the occasion went “viral” as far as that could happen before the Internet. You can learn about that here.

7 FÖLL, H., KOLBESEN, B.O.: **Formation and nature of swirl defects in silicon. Appl. Physics 8 (1975) 319**
All about the TEM of many samples and the detailed analysis of the dislocations loops
257 citations, quite a lot.

9 FÖLL, H., GÖSELE, U., KOLBESEN, B.O.: T**he formation of swirl defects in Si by agglomeration of Si self-interstitials. J. Crystals Growth 40 (1977) 50**
197 citations
An involved theory about the formations of swirl defects. We actually invented nucleation theory and “Time-Temperature-Transformation (TTT) diagrams”. We could have looked that up but that never occurred to us. A lot of work with lots of good stuff but not really “the truth”. Things are far more complex than we could have known by then

- In the last publication the name Ulrich **Gösele** appears for the first time. He was about my age (one year older), doing his Ph.D work at the MPI like me. He was one of my best friends for life. We interacted for many years, scientifically and otherwise, and he made it to the very top of the German scientific establishment. I had the honor to deliver the laudatio at the big party on hs 6oth birthday, and the obituary a year later when he unexpectedly died. Uli Gösele was important to me ans that’s why I include both “Papers” in this archive.i

Prof. Dr. Ulrich Gösele **Laudatio, Pictures, Obituary**

- Looking at this some 45 years later, I'm amazed how innocent we were. Today, a major discovery like the "Si interstitials" would be exploited far more, with press releases, lots of proposals for funding tis and that, ceaseless traveling from conference to conference, publications a plenty, and so on. Not fun for blokes like me.

2.3.3 Pictures

- I prepared a small selection of what we produced, containing all (or at least most) of the pictures in my thesis and the publications in part 1 and 2.
Part 3 contains some random finds in my files.

Swirl pictures Part 1 Part 2 Part 3