doi:10.1017/S1431927616002336
Microsc. Microanal. 22 (Suppl 3), 2016

Temperature Dependence of the Volume Plasmon in Silicon Nanoparticles

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Silicon is the foundational material of the microelectronics industry, and within this industry transmission electron microscopy (TEM) is a widely-used tool. Inelastic scatting of TEM-beam electrons in silicon is most commonly the result of volume plasmon production. Because the volume plasmon energy depends on the probed electron density, measurements of the electron energy loss spectra (EELS) can, in principle, determine dopant profiles. We demonstrate parts-per-thousand sensitivity to silicon volume plasmon energy shifts through curve fitting. This sensitivity has enabled us to measure, for the first time, the temperature dependence of silicon's volume plasmon. The observed decrease in plasmon energy with increasing temperature is induced by thermal expansion of the silicon lattice. This result is a necessary step toward using silicon as the thermometric material in plasmon energy expansion thermometry (PEET) [1].

We measured the silicon volume plasmon using a JEM JEOL 2100F equipped with a Gatan Quantum SE GIF. The TEM high tension was 80 kV and the spectrometer energy dispersion was 25 meV/channel. The spectrum at each pixel in the scanning TEM (STEM) spectrum image was fit twice: a Gaussian fit to the zero-loss peak (ZLP) (from -0.45 eV to 0.45 eV) and a Lorentzian fit to the silicon volume plasmon peak (14.45 eV to 19.00 eV). At each point measured plasmon energy is the difference between the centers of the Gaussian fit and the Lorentzian fit. Individual plasmon energy maps were acquired at temperatures between 25°C and 1000°C. The data collection run consisted of two temperature ramps: one from 1000°C down to 100°C in 100°C steps, then one from 950°C to 50°C in 100°C steps. This two-ramp procedure would reveal hysteresis in the silicon nanoparticles' plasmon energy; none was found.

The plasmon energy maps reveal an unexpected feature: the plasmon energies are larger near the edge of the nanoparticle than they are in the interior. We fit the plasmon energy histograms with two Gaussians to quantify this dichotomy. (Example data from two temperatures, 25°C and 1000°C, is given in Figure 1.) As a function of temperature, both plasmons decrease as temperature at a rate of ~-0.08 meV/K (Figure 2, left). This rate, smaller than aluminum's ~-0.58 meV/K [2,3], is expected because silicon's thermal expansion coefficient (~2.5×10^{-6}/K [4], near 300K) is nearly an order-of-magnitude smaller than aluminum's (~23×10^{-6}/K [5], near 300K). Compared to the mean plasmon energies, the plasmon widths are nearly temperature-independent (Figure 2, right).

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[6] This work was supported by FAME, one of six centers of STARnet, a Semiconductor Research Corporation program sponsored by MARCO and DARPA. Data presented were acquired at the Center for Electron Microscopy and Microanalysis at the University of Southern California.

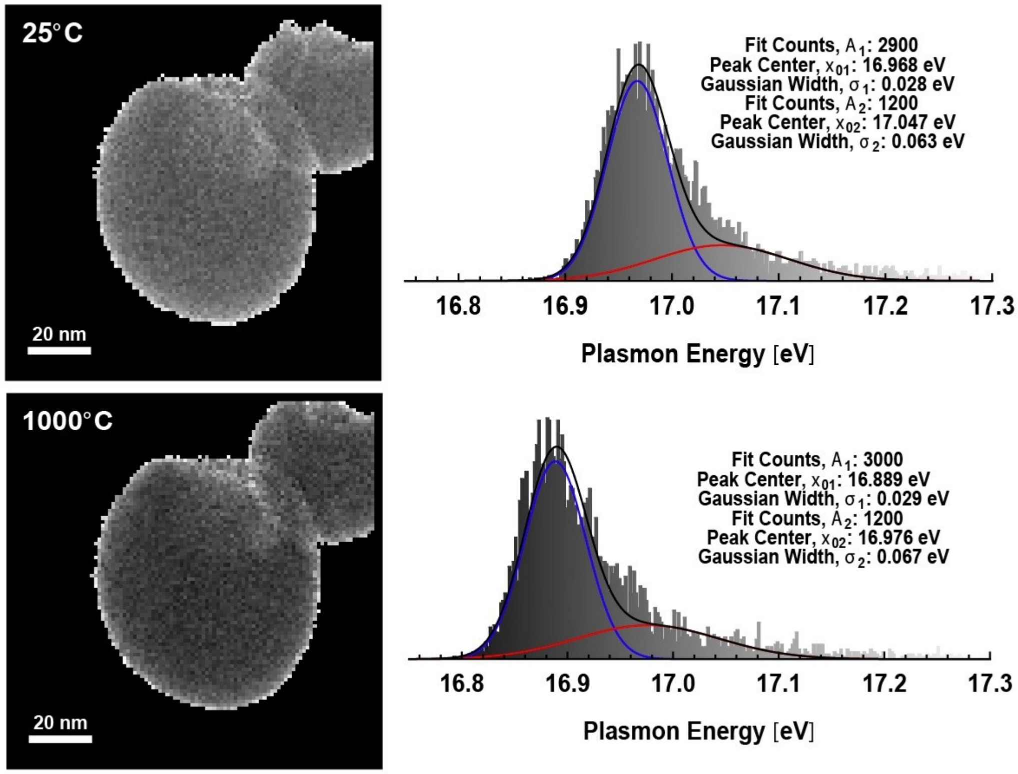

Figure 1. (*top, left*) Plasmon energy map, 100 pixel by 100 pixel with 13 nm pixel size, acquired at 25°C, next to a histogram (*top, right*) of the associated plasmon energies. (*bottom, left*) and *(bottom, right*) Corresponding data acquired at 1000°C. The blue and red curves indicate the Gaussian fits to plasmons we associate with the interior and the edge of the silicon nanoparticles, respectively.

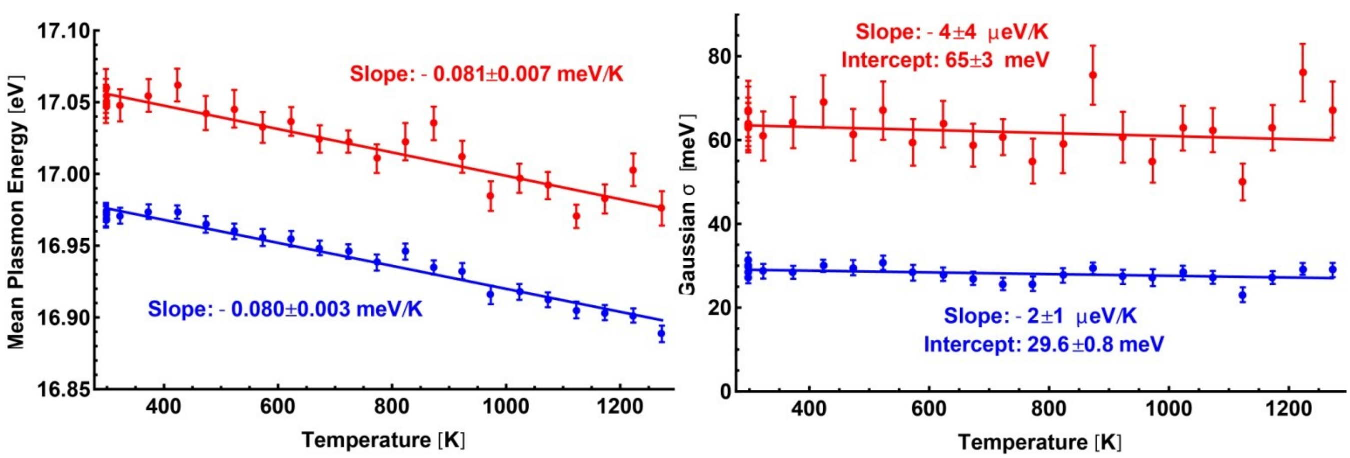

Figure 2. The mean plasmon energy in each volume plasmon group at each temperature (left) and standard deviation in each volume plasmon group at each temperature (right) are plotted. The colors in each plot are associated with the corresponding Gaussian fits in the histograms of Figure 1.

https://doi.org/10.1017/S1431927616002336 Published online by Cambridge University Press