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Comment on "Raman scattering with nanosecond resolution during pulsed laser annealing of silicon"

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In a recent letter¹ von der Linde and Wartmann report time-resolved Raman measurements of the optical phonon population in Si during intense excitation with a 10-ns pulse at 532 nm. The results indicated a phonon temperature in excess of the melting temperature for a UV Raman probe ($\lambda = 355$ nm), but a different, much lower temperature for a Raman probe in the visible ($\lambda = 532$ nm). Here we show data confirming the supposition that this difference arises from the temperature dependence of the factors which correct for absorption and resonance Raman effects.

In Ref. 1 the phonon temperature T is obtained from the relation $\exp(h\omega_p/kT) = C(S/A)$, where S/A is the measured Stokes/anti-Stokes ratio and ω_p is the phonon frequency. The two largest contributions to C involve corrections for the frequency dependence of the band-to-band absorption and of the Raman cross section as discussed in detail elsewhere.¹⁻³

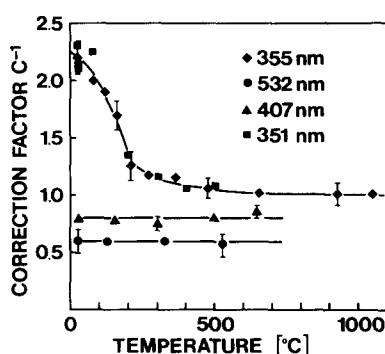


FIG. 1. Correction factor C^{-1} determined from oven measurements. Data at 532 and 355 nm were measured with the system of Ref. 1; data at 407 and 351 nm were measured with a cw Kr laser and do not include the (constant) instrumental correction used in Ref. 3 for data at 405 nm.

Because of considerable uncertainties in calculating the correction factor from available literature data, C has been determined experimentally by Raman measurements on oven-heated silicon wafers. The data of Fig. 1 show that the correction factor C is only weakly temperature dependent for Raman probes at 532 and 407 nm. This is expected because the effect of increasing absorption⁴ tends to balance the effect of increasing Raman cross section⁵ as the direct gap shifts to lower energy with rising temperature. Therefore, the use of room-temperature corrections gave rise to little error in the inferred phonon temperature when probed at 405 nm. At 532 nm, on the other hand, the measured value of $C = 1.7$ differs from the calculated value used in Ref. 1. The difference was shown to be due to an error in correcting for the spectral response of the photomultiplier tube. As shown in Fig. 1 large temperature dependent corrections arise for Raman probes in the UV. Note that the measured room-temperature value of C (355 nm) is in good agreement with the UV C value used in Ref. 1. The rapid increase of C with rising temperature is explained as follows. In the UV near 350 nm the Raman cross section is steeply falling with frequency,⁵ whereas the absorption above the direct gap varies very little with photon energy.⁴ As the conduction band shifts toward the valence band with increase of temperature, the Raman resonance curve also shifts and broadens, leading to a decrease of the Raman cross-section ratio.

If one now applies the measured high-temperature correction factor of $C = 1.05 \pm 0.1$ to the high-temperature points of Ref. 1, the phonon temperature at the transition to the high reflectivity phase is changed from 1700 to about 600 K. Furthermore, agreement of the visible and UV Raman probe experiments is obtained if the spatial averaging over the temperature gradient normal to the surface is taken into account. Thus, the two sets of data of Ref. 1, at both 532 and 355 nm, are internally consistent and also agree well with the phonon temperature obtained using a 405-nm probe.³

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