Supporting Information

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SI Text

Microcrack Formation. The amount of strain energy stored in a thin film is directly proportional to its thickness (1). Our membranes, bonded to polyimide (PI), contain insufficient strain energy to drive separation or cause interfacial fracture. Under conditions of strong interfacial adhesion and high externally applied stress, some of the strain in the Ge nanomembrane (NM) may be relaxed by dislocations or microcracks, or possibly by phase transformations at the interface. We find microcracks at high stress levels and can observe them in the Raman microscope. Cracks increase in density as the stress increases. Some appear to emanate from the etchant access holes used to facilitate the NM release, whereas others do not. To investigate this issue further, we have made smaller round Ge NMs without etch holes. Each such disk is larger than the separation of the etch holes in the larger membranes used in this work. The disks show the same strain behavior observed in Fig. 2, but the first formation of cracks in these circular NMs occurs at a somewhat higher bulge cell pressure than for the NMs with etch holes. Crack formation in the NMs may therefore potentially be reduced by eliminating the etch holes.

Surface Oxidization. Although the Ge surface may in principle develop a native oxide during the final NM processing step (i.e., the wet etch in H_2O_2), we expect that layer to be very thin because it would mostly dissolve in the aqueous etch solution, and therefore to have negligible impact on the measured results.

Theoretical Bandgap Energies Versus Strain. The key parameters of the deformation-potential theory used to compute the Ge bandgap energies as a function of strain (2, 3) are the hydrostatic deformation potentials a_v , a_c , and $\Xi_d + \frac{1}{3}\Xi_u$ (which describe the strain-induced shifts of, respectively, the average valence-band maximum, the conduction-band minimum at Γ , and the conduction-band minima at L), and the shear deformation potential b(related to the strain-induced splitting of the valence bands at Γ). For the first two parameters, we use the theoretical values of $a_v = 1.24$ eV and $a_c = -8.24$ eV computed by Van de Walle (4), which are in good agreement with the measured pressure dependence of the direct bandgap of Ge (5, 6). For $\Xi_d + \frac{1}{3}\Xi_u$, we use the value of -2.34 eV recommended in ref. 5, based on the measured pressure dependence of the indirect bandgap (7). Regarding the shear deformation potential, a range of theoretical values from several different sources (4, 8, 9) is commonly used in the literature; here we choose b = -2.16 eV as computed by Fischetti and Laux (8), which produces a slightly better agreement with our measurement results. The values of all other simulation para-

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meters (i.e., unstrained-Ge bandgap energies, spin-orbit splitting, and elastic moduli) are well established in the literature.

Theoretical Spontaneous-Emission Spectra. The transverse-electricpolarized direct conduction-to-heavy-hole (cГ-HH) and conduction-to-light-hole (c\Gamma-LH) spontaneous-emission spectra are calculated using textbook formulas (2, 3) involving the joint densities of states, Fermi distribution functions, and oscillator strengths. We assume perfectly parabolic energy bands, so that the required expressions only depend on a handful of well-established materials parameters-i.e., bandgap energies, effective masses, and momentum matrix elements. The bandgap energies are computed as described in the previous paragraph. The momentum matrix elements are calculated from the Bloch functions of electrons, HHs, and LHs as described in refs. 2 and 3; the strain dependence of the Bloch functions is also included in this computation, following ref. 10. The density-of-states effective masses of the Γ and L conduction-band minima and of the HH and LH valence-band maxima are obtained as a function of strain from refs. 8 and 11. To account for the experimentally observed strain variations across the NM area at fixed pressure, the calculated spontaneous-emission spectra are convoluted with a Gaussian function of strain, with mean value and standard deviation based on the Raman measurement results. Finally, the resulting spectra are further convoluted with a Lorentzian function of energy to account for lifetime broadening, with a typical value of 0.1 ps for the intraband scattering time (3). The only fitting parameter, besides a single overall multiplicative factor, is then the quasi-equilibrium carrier density N produced by the pump pulses, which determines the quasi-Fermi levels and therefore the distribution functions.

Because of the assumption of parabolic energy bands, the calculated emission spectra can be expected to be very accurate only at photon energies near the Γ -point bandgap energies, where the c Γ -HH and c Γ -LH contributions are peaked. Away from the Γ point, the valence bands of tensilely strained Ge are actually known to feature large nonparabolicities (11). As a result, at photon energies away from the Γ -point bandgaps, the calculated emission intensities can be expected to underestimate the experimental values. For the same reason, this model also underestimates the total carrier density *N* required to have sufficient hole occupation in the HH valence band for appreciable c Γ -HH light emission. Therefore, the value of *N* obtained by fitting the experimental photoluminescence data using this model should be regarded as a lower bound.

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