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Vibrational relaxation of adsorbates at semiconductor surfaces: H on Ge(100)

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Abstract
Infrared pump-sum frequency generation (IR pump-SFG) probe spectroscopy is used to study the vibrational energy relaxation of hydrogen adsorbed on a Ge(100) surface. We found that the symmetric Ge–H stretch mode at 1992 cm^{-1} ($\upsilon_s = 1$) for the Ge(100)-(2 × 1):H surface has a population relaxation time (T_1) of 0.70 ± 0.07 ns at a substrate temperature of 323 K which is of the same order as Si(100)-(2 × 1):H. The population of the stretch mode decays exponentially in the temperature range tested from 273 to 400 K. The observed decay constants decrease from a value of the order of one nanosecond to a value of the order of 100 picoseconds, when the substrate temperature is increased from 273 to 400 K. This strong temperature dependence is consistent with a decay via a multiphonon process, which may be facilitated by the rapid increase of the density of states associated with the low-frequency TA mode. From the evaluation of the temperature dependence we suggest that the Ge–H stretch mode decays by simultaneously exciting three bending mode quanta at 530 cm^{-1} and more than two bulk Ge phonons.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Hydrogen adsorption on Si and Ge has been studied for more than 20 years because of its importance in device fabrication for computer and photovoltaic applications as well as to the fundamental understanding of chemisorption on semiconductor surfaces [1]. In particular, the system H/Si attracted much research interest in the energy transfer from adsorbate vibrations towards surface excitations [2, 3].

The population relaxation of vibrationally excited states of adsorbates, with a characteristic time constant T_1,[1] is the result of various dynamical couplings. The coupling mechanisms include emission of electron–hole pairs, excitation of other vibrational modes of adsorbates, and creation or annihilation of substrate phonons. On metal surfaces, the relaxation of vibrationally excited adsorbates, such as CO on Pt(111) [4] and Cu(100) [5], is due to the coupling to electron–hole pairs. A rather short vibrational lifetime of the order of 2 ps was observed for these systems. In contrast to metal surfaces, on insulator surfaces the vibrational lifetime of adsorbates is much longer, e.g. 4.3 ms for CO/NaCl(100) [6, 7], which is close to that (33 ms) of gas phase CO [10]. Chang *et al* found that the vibrational energy relaxation of the C–O stretch mode can be attributed primarily to phonon excitations, with additional contributions from fluorescence accompanied by energy pooling (excitation of vibrational overtones) [11]. On semiconductor surfaces, the vibrational relaxation occurs via other adsorbate modes as well as different substrate phonons. In the case of H on Si(100), the impossibility of resonant electronic excitation and the large energy difference between the stretch quantum at 2100 cm^{-1} and the other vibrational quanta (e.g. bending at 630 cm^{-1}), as well as the substrate phonons, implies that a long lifetime is expected. Guyot-Sionnest *et al* reported population relaxation times for H/Si(100) and H/Si(111) in the ns regime [2, 3]. The authors suggested a population relaxation process involving three bending quanta and a bulk Si phonon. On the other hand, from theoretical studies, with the multi-configurational time-dependent Hartree method, Saalfrank *et al* inferred that the relaxation proceeds predominantly via the excitation of two bending quanta and two low-frequency substrate phonons [12]. We have studied the population relaxation of

[1] Vibrational relaxation can be distinguished as population (energy) relaxation and phase relaxation (pure dephasing, characterized by T_2^*). T_2 characterizes the total relaxation process, which is composed of two parts, i.e. $1/T_2 = 1/(2T_1) + 1/T_2^*$.

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CO on Si(100) [8, 9]. IR pump-SFG probe measurements on the C–O stretch mode at 2081 cm^{-1} of CO/Si(100)-(2 × 1) reveal a relatively long, single-exponential relaxation with a decay constant of 2.3 ± 0.5 ns at a substrate temperature of 100 K. Vibration-phonon coupling is expected to dominate the vibrational decay process of CO/Si(100). Since the calculated C–O bending and Si–CO shift quanta at 441 cm^{-1} (in the plane (010)), 429 cm^{-1} (in the plane (001)) and 508 cm^{-1},[2] respectively [13], are smaller than the Si–H bending quantum at 630 cm^{-1}, the relaxation would require the excitation of more low-energy vibrational quanta or phonons than in the system H/Si. However, the underlying microscopic mechanisms for such processes are still not fully understood.

Germanium provides some interesting experimental opportunities, when compared to Si for such energy relaxation studies, because of its relatively low Debye temperature (290 K). This implies that the maximum phonon energies are smaller than the Si ones, namely the highest bulk LO phonons in Ge have an energy of 310 cm^{-1} [14, 15], about half of that for Si (Si phonon edge at 520 cm^{-1}). The Ge–H stretch mode at 1992 cm^{-1} and the bending mode at 530 cm^{-1} have nearly the same energies as the corresponding modes of the Si–H bond [16, 17]. Thus, studies on the energy relaxation on Ge surfaces could shed more light on the energy decay mechanisms. In this report we present first results to understand the relationship between the relaxation rate and its pathway, i.e. how the stretch mode decays into bending quanta with the aid of substrate phonons.

As an experimental method, time-resolved sum frequency generation (TR-SFG) spectroscopy provides a way to study the temporal evolution of adsorbate vibrations on surfaces. SFG is a second order nonlinear optical process that only occurs at the interface between two centrosymmetric media, where the symmetry is broken. In the case of the infrared–visible (IR–vis) SFG, as used in our experiment, the sum frequency photons are generated by the interaction of two laser pulses, one tunable infrared pulse and another visible pulse. The measured signal is enhanced when the energy of the photons of one of the three waves (infrared, visible and sum frequency) involved in the process is in resonance with a vibrational or an electronic transition at the interface. Such a vibrational transition has to be IR and Raman active in order to be SFG active. By using an additional IR laser pulse (pump pulse) prior to the probe pulse pair, a fraction of the adsorbed molecules is vibrationally excited, thus resulting in a reduced difference between the ground- and excited-state populations, such that the intensity of the SFG signal decreases. By varying the delay between the IR pump and the probing pulse pair the vibrational lifetime can be directly measured.

2. Experimental details

The experimental apparatus and procedures have been described in detail elsewhere [8, 9]. Briefly, the UHV chamber, containing LEED/AES facilities and a quadrupole mass spectrometer (QMS), had a base pressure of 10^{-11} mbar. The sample could be directly heated by applying a voltage across the crystal and cooled to around 100 K by liquid nitrogen. The sample temperature was monitored via a thermocouple fixed on the sample using a Ta clip.

The samples were cut from Ge(100) wafers (p-type, 1–10 Ω cm, $22 \times 6 \times 0.25$ mm^3, supplied by CrysTec) and cleaned by wet chemical treatment instead of ion bombardment and annealing. The sequence of chemical treatment is as follows: (1) repetitive chemical oxide formation in H_2O_2 (aq. 3%) for 60 s and oxide etching by HCl (aq. 9%) for 30 s; (2) protective oxide formation in NH_3 (aq. 28%)/H_2O_2 (30%)/H_2O (1:2:20 volume parts). The compounds used for the procedure were analytical grade reagents. The cleaned sample was mounted in the UHV chamber. The *in situ* preparation consisted of outgassing the sample at 573 K for several hours until the chamber pressure was in the low 10^{-10} mbar range, followed by flash annealing to 1000 K to remove the oxide, while maintaining a chamber pressure of 10^{-10} mbar, and then cooling down at a rate of 0.5 K s^{-1}. The cooling rate is very important in obtaining well reproducible data. The outgassing is used to remove the carbon species, as Chan *et al* have observed a large decrease in the carbon signal using x-ray photoelectron spectroscopy [18]. Alternatively, cleaning of the Ge surface was also performed by repeated argon ion bombardment (1 keV, 3 μA mm^{-2}, 30 min: the sample was held at 600 K for the first 5 min and at room temperature afterwards) followed by annealing at 773 K for 5 min to drive out all the Ar gas trapped inside. Afterwards it was necessary to anneal the sample at 923 K for 5 min and 823 K for half an hour before it was allowed to cool down with a rate slower than 1 K s^{-1}. This procedure was chosen because the reconstruction rate of Ge is approximately 0.2 nm min^{-1} at 823 K after argon ion sputtering [18]. A series of annealing cycles at 923 K was carried out until a sharp c(4 × 2) LEED pattern was obtained at 100 K.

The surface structure was checked by means of LEED. At room temperature the clean Ge(100) surface exhibits a (2 × 1) reconstruction, although hints of quarter order spots are sometimes present. At 100 K the quarter order features become well defined and are as sharp as other integer and half integer order spots in the pattern, which indicates a c(4 × 2) reconstruction. It is expected that the long range order becomes more established at low temperature. Kevan reported that at 220 K the 'antiferromagnetic-like' c(4 × 2) is observed to reversibly transform into a 'paramagnetic-like' (2 × 1) reconstruction of the surface [30]. The existence of sharp c(4 × 2)-spots with a low background in the LEED pattern is a strong indication of a clean surface with low defect density. In addition, we checked that all AES signals of impurities were below the noise level. Peak-to-peak noise in the differential Auger spectrum is approximately 100 times smaller than the peak-to-peak signal of the Ge feature at 1147 eV.

A Ge(100)-(2 × 1):H surface was prepared by adsorption of atomic hydrogen on a clean surface in ultrahigh vacuum. Atomic hydrogen was generated by a hot capillary source facing the sample with a distance of 1 cm. H_2 temperature programmed desorption (TPD) spectra were recorded from the Ge(100) surface covered with various amounts of H at a

[2] The experimentally measured frequency for the shift mode is 411 cm^{-1} [29]. Since the back-donation into the CO 2π∗ orbital is overestimated in DFT calculations, the shift frequency is larger than the experimental result.

heating rate of 3 K s^{-1}. Exposure to atomic H at a substrate temperature of 473 K results in a saturated adlayer dominated by monohydrides. The TPD peak for H_2 desorption is located at 594 ± 5 K. This peak is attributed to desorption from the monohydride phase. The other peak, attributed to desorption from the dihydride phase, was not observed after using our preparation procedure.

The SFG experiments were performed using an actively-passively mode-locked Nd:YAG laser system (Ekspla), generating 22 ps pulses with a repetition rate of 20 Hz. A part of its second harmonic light (532 nm, ∼10 μJ) was used as the visible upconversion pulse, while the tunable IR light (linewidth 9 cm^{-1}) was produced by an optical parametric generator (OPG) system. The IR beam was split into a pump (∼80%) and a probe (∼20%) beam by a dielectric beam splitter, with typical energies at the sample of 120 μJ and 30 μJ for pump and probe, respectively. The delay of the visible and IR probe pulses with respect to the IR pump pulse could be adjusted by means of motorized delay lines. The usable range of the IR pump delay line was overall 4 ns. Both IR beams were focused by the same CaF_2 lens to a spot of ∼1 × 2 mm^2. Vibrational lifetime measurements were done by alternatively blocking and unblocking the IR pump beam allowing a direct comparison between the SFG signal intensities with and without pump beam (e.g. 20 pulses each). Typically 40 sequences of this type were recorded for every single delay line step, which results in an overall number of typically 1600 pulses per delay line step.

3. Results and discussion

3.1. SFG spectra

Figure 1 shows a set of SFG spectra obtained from the clean Ge(100)-(2 × 1) and Ge(100)-(2 × 1):H surfaces at various sample temperatures. For the clean Ge(100) surface, a strong non-resonant background in the 1800–2100 cm^{-1} region was observed. In the spectra shown in figure 1 the background signal seems to fade out on both sides of the centre frequency. This is due to a limitation of the experimental setup regarding signal registration and does not represent a real decay. The background SFG signal notably decreases with rising surface temperature.

At substrate temperatures around 400 K the non-resonant background signal was notably reduced, and we observed an intense peak centred at 1992 cm^{-1}, which is attributed to the fundamental frequency of the symmetric stretch of the Ge–H bonds, in agreement with earlier IR spectroscopy experiments [17]. The half width of this resonance in the spectra taken at 420 K appears to be 13 cm^{-1}, This width must not conflict with earlier reports from Chabal [17], who found a linewidth of about 3 cm^{-1} for a saturated surface at room temperature using IR spectroscopy. In our experiment several factors contribute to the total linewidth: (i) the coupling to the intense non-resonant background, (ii) the finite spectral resolution in our experimental setup (governed by the IR bandwidth 9 cm^{-1}) and (iii) the larger pure dephasing at the relatively high sample temperatures. The pure dephasing was determined for the system Si(100):H by Tsai *et al* [31] to result

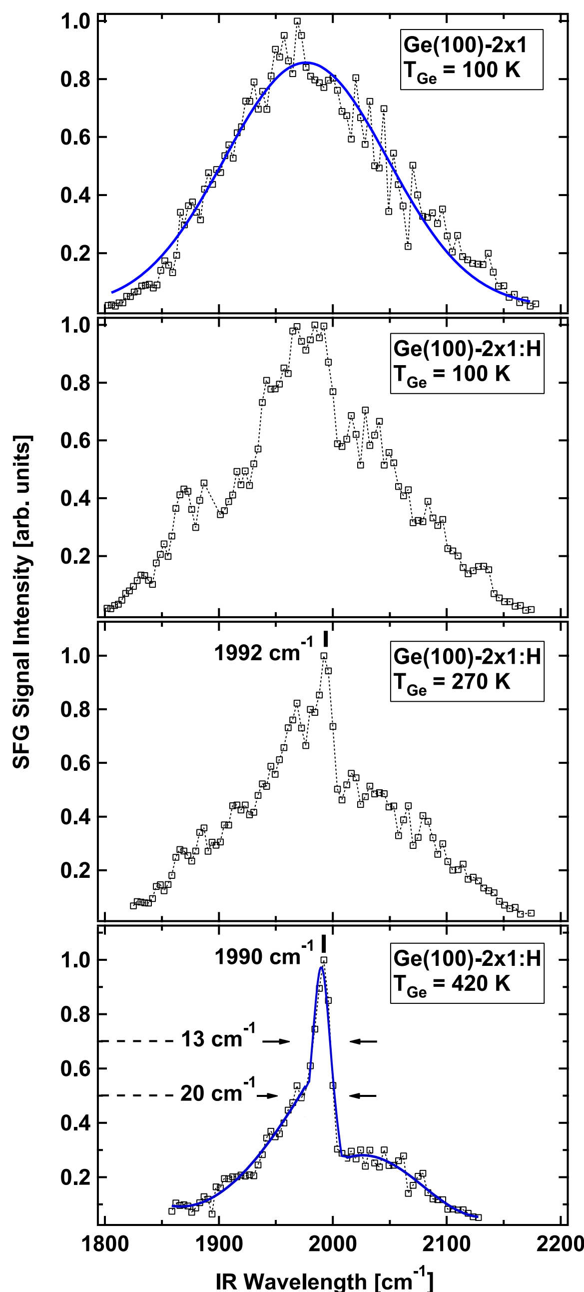

Figure 1. SFG spectra obtained with ppp polarization from clean Ge(100)-(2 × 1) at 100 K and Ge(100)-(2 × 1):H monohydride phase, showing the $\nu = 0 \rightarrow 1$ transition of the Ge–H stretch mode at 100 K, 270 K and 420 K, respectively. The signal intensities have been normalized independently to the intensity maximum observed. The solid lines represent fits to the experimental data.

in a linewidth broadening of about 3 cm^{-1} in the temperature range from 300 to 430 K.

Different from the intensity of the non-resonant SFG signal, the signal attributable to the Ge–H stretch mode changes only slightly (approx. 15%) in the temperature range between 300 and 430 K. As shown in figure 2, the SFG intensity was recorded at different temperatures and the intensity of the non-resonant background recorded with the clean surface was subtracted from that associated with the H-covered surface for each of the temperatures. The logarithm

Table 1. The vibrational lifetimes of the Ge–H and Si–H stretch mode for different hydrogenated surfaces.

System	Ge(100)-(2 × 1):H[a]	a-Ge:H[b]	Si(100)-(2 × 1):H[c]	a-Si:H[d]
Lifetime (ps) @ temperature (K)	1200 @ 273 K 300 @ 400 K	306 @ 10 K 100 @ 220 K	6000 @ 100 K 1000 @ 300 K	690 @ 10 K 460 @ 100 K
Decay channels	3 ω_B + 4 TA	2 ω_B + 3 phonons	3 ω_B + 1 phonon	3 ω_B + 1 TA

[a] This work.
[b] Reference [23].
[c] Reference [3].
[d] Reference [24].

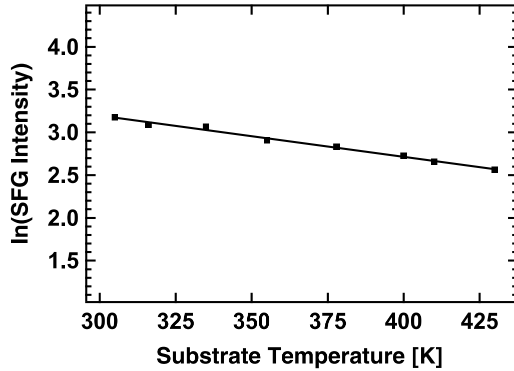

Figure 2. SFG intensity of Ge–H stretch (logarithmic scale) as a function of the substrate temperature. Squares represent the experimental data.

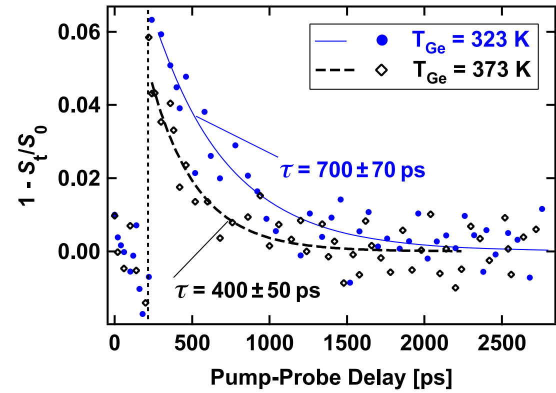

Figure 3. Population decay of the excited Ge–H symmetric stretch as a function of pump–probe delay at 323 and 373 K for the same sample. The lines represent a single-exponential fit to the decay with time constants of 700 ps and 400 ps, respectively. A bleach of ∼6% is observed. Thus the population difference between the ground and excited state is reduced to ∼94% and the population density is ∼3%.

of the SFG intensity after the background subtraction at each sample temperature is then plotted, assuming that the phase relation between the two signals is about 0°.

In addition, we determined the dependence of the SFG signal intensities upon the IR pulse energy where a linear relationship was found between them.

It can be ruled out that the observed decrease of the SFG intensity of the Ge–H stretch vibration arises from thermal desorption or photodesorption for the following reasons. The vibrational quantum is smaller than the indirect band gap of Ge (0.66 eV at 300 K) and the typical onset of optical absorption at 0.90 eV. We consider it to be very unlikely that carriers generated by our visible upconversion pulse (2 eV) cause photodesorption, as in the analogous system Si(100)-(2 × 1):H the $\pi*$ orbital of the Si–H bond needs to be excited resonantly in order to induce photodesorption, requiring a photon of 8 eV [19]. On Ge the same transition would require a photon of about 7 eV [20]. Secondly, we calculated the transient temperature rise induced by the incident visible laser pulse (method: [21]; Ge material parameters: [22]). It was smaller than 15 K at the intensities used. Our SFG measurements were taken at temperatures lower than 430 K, which is far from the H_2 desorption temperature of 600 K. We also compared the TPD spectra before and after SFG measurements. Within the limits permitted by the accuracy of our measurements, laser induced H_2 desorption from a Ge surface was not observed.

3.2. Lifetime and energy transfer

To perform the pump–probe measurements, we resonantly excited the stretch mode at 1992 cm^{-1} with IR pump pulses. The quantity S_t/S_0 was plotted against the pump–probe delay, where S_t and S_0 are the SFG intensities with IR pump pulses on and off, respectively. The motivation for this way of extracting the decay constant will follow in section 3.3.

Figure 3 displays the decay observed at the sample temperatures of 323 and 373 K. The measurements at different pump excitation densities and substrate temperatures between 270 and 400 K exhibit an exponential decay, which could be well fitted by a single-exponential function of the form

$$S_t = S_0 \exp(-t/\tau), \quad (1)$$

where τ is the characteristic decay constant, i.e. the population relaxation time T_1. The population relaxation time was 0.70 ± 0.07 ns and 0.40 ± 0.05 ns at 323 K and 373 K, respectively, as shown in figure 3. The decay rate increases by a factor of about five from 273 to 400 K. Until now we could not perform successful measurements of the population relaxation time at temperatures below 273 K because of the strong non-resonant background. The population excitation density of $<3\%$ on the Ge–H stretch mode is too small to collect any reliable results under our experimental conditions.

In contrast, non-exponential decays are observed in analogous experiments for a-Ge:H and a-Si:H [23, 24], which are indicative of vibrations at different sites with different lifetimes. Table 1 lists some results for adsorbate (Ge–H and Si–H) vibrations on crystalline Si and Ge surfaces and in a-Si:H as well as in a-Ge:H [2, 3, 23, 24]. Notably, in amorphous

environments the vibrational decay is faster than that on single crystals. For the system a-Ge:H, the Ge–H stretch vibration decays into two bending quanta with the energy mismatch bridged by three bulk phonons. This is in contrast to H/Si, in which case for either crystalline or amorphous substrate, the vibrational relaxation involves three bending quanta and one TA-like mode.

Interestingly, the observed vibrational decay rate is very similar to the one observed in experiments on the stretch vibrations of the system Si(100)-(2 × 1):H [3]. This is surprising, as the latter process has been discussed to take place via three bending quanta and one substrate phonon, or alternatively two bending mode quanta and two phonons. For Ge(100)-(2 × 1):H at least two phonons must be excited to make up the energy difference between stretch and three bending quanta. Alternatively, also two bending quanta and three phonons could be excited. Intuitively, one would expect that the necessity to decay into five low-frequency modes would result in a longer lifetime when compared to H/Si(100). However, that is not observed.

3.3. Data analysis

3.3.1. Population decay. The SFG spectra in the Ge–H stretch region, as shown in figure 1, are composed of a resonant contribution from the adsorbate mode and a non-resonant contribution from the Ge surface. The spectra for the fundamental excitation can be analysed with the conventional expression for the total second order susceptibility [32],

$$\chi^{(2)}_{\mathrm{tot}} = \chi^{(2)}_{\mathrm{NR}} + \chi^{(2)}_{\mathrm{RES}} = A_{\mathrm{NR}} \exp(\mathrm{i}\phi) + \frac{A_{\mathrm{RES}}(N_0 - N_1)}{\omega_{\mathrm{IR}} - \omega_{\mathrm{RES}} + \mathrm{i}\Gamma_{\mathrm{RES}}}. \tag{2}$$

Here, A_{NR} and ϕ are the magnitude and the relative phase between the resonant and the non-resonant surface contribution, and A_{RES}, ω_{RES} and Γ_{RES} are the amplitude, frequency and half width of the resonant signal of the fundamental excitation, respectively. A_{RES} depends on the population difference between the ground and the excited state $\Delta n = N_0 - N_1$. The probe IR field is sufficiently weak that Δn remains unchanged. The SFG signal is proportional to $|\chi^{(2)}_{\mathrm{tot}}|^2$.

The effect of the pump pulse is to transfer population from the vibrational ground state to the first excited state, thereby reducing the population difference and, hence, the intensity of the probe signal. However, the population transfer is small, such that the change in $\chi^{(2)}_{\mathrm{RES}}$ is small. The dependence of the resonant part on the delay time is represented by:

$$\chi^{(2)}_{\mathrm{tot}} = \chi^{(2)}_{\mathrm{NR}} + \chi^{(2)}_{\mathrm{RES}} \left[1 - \Delta n \exp\left(-\frac{t}{T_1}\right)\right]. \tag{3}$$

As long as the population transfer is small, then $\Delta n \ll 1$. If one makes a square upon the equation, it becomes evident that the terms linear in Δ will dominant the signal and necessarily decay with T_1. Thus, the change in signal scales linearly with the change in population. The unpumped-probe signal is recovered with the population relaxation time T_1. To extract the decay constant of the vibrational population from experimental data, we plotted the bleach $1 - S_t/S_0$, which is proportional to Δ, against the pump–probe delay. In the limit of a small modulation, S_t/S_0 will always recover with T_1, irrespective of the presence of a non-resonant background.

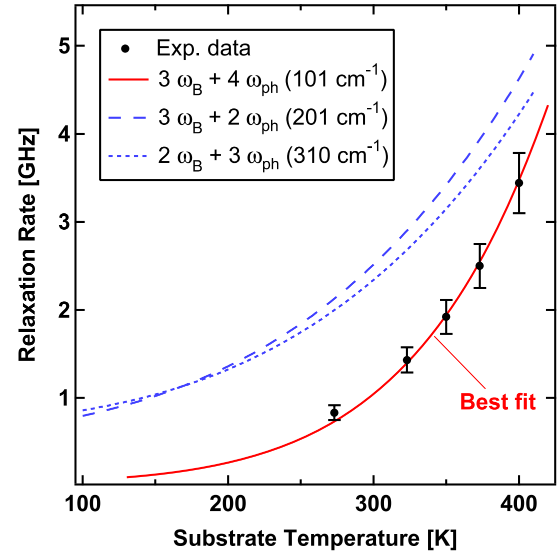

Figure 4. The temperature dependence of the vibrational relaxation rate of the Ge–H symmetric stretch for the Ge(100)-(2 × 1):H surface. Dots represent experimental data; lines represent theoretical curves calculated using equation (4). The selected decay channels are also shown (ω_{B} and ω_{ph} refer to the Ge–H bending mode at 530 cm^{-1} and Ge phonon modes, respectively).

3.3.2. Evaluation of the temperature dependence. The temperature dependence of the decay constant sheds further light on the decay dynamics. It can be approximated using the established expression [25, 26]:

$$\frac{1}{\tau} = \frac{1}{\tau_0} \left\{ \frac{\exp(\hbar\omega/k_{\mathrm{B}}T) - 1}{\Pi_i[\exp(\hbar\omega_i/k_{\mathrm{B}}T) - 1]} \right\}, \tag{4}$$

which describes multiphonon decay rates for a vibrational mode having the frequency ω into a combination of accepting modes with frequency ω_i due to anharmonic coupling. The Bose–Einstein factors $\langle n_i \rangle = 1/[\exp(\eta\omega_i/k_{\mathrm{B}}\,T) - 1]$ give the average number of quanta in each phonon or bending mode. The coupling process is governed by a temperature-dependent population relaxation rate $1/\tau$, of which the zero temperature value is $1/\tau_0$, and the condition $\Sigma_i \hbar\omega_i = \hbar\omega$ that must hold in order to satisfy energy conservation. In figure 4 we present calculated curves using equation (4) and the experimental data. We tried to reproduce the data using different combinations of low-frequency modes. However, no satisfactory fit can be found for the combination of either two or three bending quanta with three or two phonons with energies of 310 cm^{-1} and 201 cm^{-1} [27, 28], respectively. In this case a zero temperature value τ_0 of about 1 ns has to be assumed. The temperature dependence in the temperature range from 273 to 400 K is better reproduced assuming decay into three bending mode quanta and four quanta of the transverse acoustic mode

at 101 cm^{-1}. However, in this case the zero temperature value τ_0 must be chosen as 40 ns. Thus, the '3 ω_B + 4 TA' model suggests a decay rate at low temperature which is surprisingly small. The predicted decay rate for relaxation into any other combination of a smaller number of accepting modes significantly underestimates the increase of the decay rate with increasing temperature. This would imply that despite the larger number of quanta involved in the decay for H/Ge(100) the lifetime remains comparable to the one reported for H/Si(100). This would be indicative of a larger matrix element for phonon excitation at the Ge surface. This process may be facilitated in the temperature range between 273 and 400 K by the small energy of the low-frequency TA mode and the high density of states associated with it [27, 28].

However, our results on the decay channels for the system H/Ge(100) suggest a mechanism only over a limited temperature range. Below room temperature the decay could follow different relaxation pathways, since in this temperature range, the contribution of small energy phonons is restricted when compared to the large energy phonons. To obtain a better microscopic picture of the energy relaxation in the system H/Ge(100), we have, besides experiments, also started DFT calculations. We aim at identifying the dissipation processes that determine the lifetime of the stretch mode of the Ge–H bond. The energy transfer to other system vibrations and to substrate phonons will also be theoretically studied. The results will be presented in a forthcoming paper.

4. Summary

The vibrational population relaxation of hydrogen adsorbed on a Ge(100) surface was studied using IR pump-SFG probe spectroscopy. T_1 is of the order of one nanosecond for the Ge–H symmetric stretch for the Ge(100)-(2 × 1):H surface at room temperature. Because of a strong non-resonant background signal from the Ge substrate, the pump–probe measurements were performed at sample temperatures above 273 K. In the temperature range tested up to 400 K the vibrational energy relaxes following a single-exponential decay. We attribute this to coupling to lower-frequency adsorbate modes and phonons. The measured decay constants decrease from a value of the order of one nanosecond to a value of the order of 100 picoseconds, when the substrate temperature increases from 273 to 400 K. This strong temperature dependence, which is expected to be stronger with increasing order of the multiphonon processes, may be facilitated by the rapid increase of the density of states associated with the low-frequency TA mode. These results reveal an important aspect of vibrational relaxation in the system H/Ge(100): from the evaluation of the temperature dependence it is suggested that relaxation of the Ge–H stretch mode initiates simultaneous excitation of three Ge–H bending quanta and four bulk Ge phonons at substrate temperatures above 273 K. However, the lifetime of the vibrational excitation is similar to that observed for the system H/Si(100), in which case four low-frequency modes suffice to match the energy of the stretch mode.

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