Effects of Nonlinear Elastic Surface Pulses in Anisotropic Silicon Crystals

A. Lomonosov and P. Hess

Institute of Physical Chemistry, University of Heidelberg, Im Neuenheimer Feld 253, 69120 Heidelberg, Germany
(Received 31 March 1999)

The propagation of nonlinear surface acoustic wave (SAW) pulses was investigated in anisotropic single-crystal silicon. The effects of frequency-up and frequency-down conversion were found to depend on the plane and direction of SAW propagation, yielding a variety of waveforms. The formation of steep shock fronts that broke the covalent crystal was observed in the (112) direction of the Si(111) plane. Solitary behavior of surface waves was studied by investigating the interaction between nonlinearity and dispersion for silicon covered with a thin oxide layer.

PACS numbers: 68.35.Gy, 41.20.Jb, 43.35.+d, 62.65.+k

The effects of nonlinear elastic properties of anisotropic crystal structures on the propagation of surface acoustic waves (SAWs) have so far not been well investigated experimentally [1]. Anisotropy plays an important role, for example, in understanding the failure of crystalline materials such as silicon [2]. Whereas the longitudinal and transverse modes propagate with different velocities in the bulk material, and mostly the longitudinal mode is affected by nonlinearity in this case, the two modes are coupled in elliptically polarized surface waves, leading to new nonlinear features. Pronounced nonlinear behavior can be expected for surface waves, due to a significant confinement of the energy near the surface. A common feature of all kinds of surface waves is that the energy is localized at the surface within the depth of one wavelength. Hence the generation of higher harmonics, for example, leads to an increase of the energy density within the correspondingly smaller penetration depth.

The generation of harmonics in finite amplitude narrow band wave trains has been widely investigated theoretically [3–5] and experimentally using interdigital transducers [6,7]. In contrast to these periodic surface waves, the nonlinear evolution of short elastic surface pulses of the Rayleigh type, considered here, has specific features, such as frequency-up and down conversion, which cause distinct effects. The generation of higher frequencies, in the time domain, corresponds to shock front formation. Nonlinear generation of lower frequencies appears as pulse lengthening. For isotropic solids this effect has been observed for materials with a negative nonlinear parameter, such as fused silica [8]. Other materials, e.g., copper or aluminum, have positive nonlinear parameters and hence a shortening of the surface elastic pulse is expected.

In crystals the linear and nonlinear elastic properties are more or less anisotropic, opening a new dimension in nonlinear acoustics [9–11], as demonstrated in this Letter. (1) Different types of nonlinear waveforms were observed, such as the shortening or lengthening of the elastic pulse and U- or N-type pulse shapes, since the complex values of the nonlinear matrix elements are either imaginary or real depending on the crystal plane and direction. (2) The stress in the shock fronts of nonlinear SAWs can reach the fracture strength of the solid. In fact, for the first time cracking of a solid by propagating SAWs was observed in silicon single crystals. (3) The interaction of nonlinearity and dispersion was studied in short elastic surface pulses. The results presented indicate that these two effects may, to some extent, compensate each other, leading to solitary behavior, investigated only theoretically before [12].

We consider the straight-crested SAW propagating along the x axis, where v is the surface velocity component along x. The nonlinear evolution of a SAW can be interpreted as an interaction between different spectral components \( v_n \), and thus is described by a system of coupled equations,

\[
\frac{\partial v_n}{\partial t} = - \sum_{k+l=n} B_{kl} v_k v_l - n^2 a v_n,
\]

where the real part of \( a \) is associated with attenuation, its imaginary part with dispersion, and \( B_{kl} \) depends on the second-order and third-order elastic constants. Here we use the simplification for the matrix \( B_{kl} \) suggested in [4],

\[
B_{kl} = \frac{kl}{|k||l|} \frac{B_{11}(k + l)^2}{|k| + |l|}.
\]

This matrix describes both local nonlinearity (existing also in fluids) and nonlocal nonlinearity, which appears only in Rayleigh waves [4]. The parameter \( B_{11} \) characterizes the generation of second harmonics. Real \( B_{11} \) values lead to the development of N-type distortions of the vertical component, whereas imaginary values to U-shape waveforms.

To simulate the evolution of a single pulse it was represented by a discrete Fourier series of harmonics of the fundamental frequency \( f_0 \). In the time domain this means that instead of a single pulse we consider an infinite sequence of pulses with the period \( T = 1/f_0 \). The period \( T \) must be long enough to prevent the interaction between
neighboring pulses during the whole time interval considered. On the other hand, the choice of a large period entails an increase in the number of harmonics to be taken into account and consequently requires a longer computational time. In our simulations we used $T = 120$ ns, whereas the maximal duration of the nonlinearly extended pulses did not exceed 35 ns.

To integrate Eqs. (1) and (2) a predictor-corrector method with an adaptive step was used. Up to 300 spectral components $v_n$ were taken into account. The parameter of $B_{11}$ was determined by fitting the predicted pulse duration to the experimental value.

The nonlinear SAW pulses were launched by absorption of 7 ns Nd:YAG laser pulses (1.06 $\mu$m) in a 50 $\mu$m thick water suspension of carbon particles, deposited on the sample surface. The almost complete absorption of radiation in this layer protected the sample surface from severe thermal damage such as melting and ablation.

The laser radiation was focused into a strip of 30 $\mu$m width and 5 mm length. Laser pulses of 30 mJ (fluence 20 J/cm$^2$) heated the absorbing layer by several hundred degrees. After 30–50 ns delay explosive evaporation occurred due to the recoil momentum, producing a pressure shock of up to 3 GPa at the surface finally limited by the fracture of the crystal. This SAW excitation technique has an efficiency comparable to that of confined plasma methods for bulk wave excitation [13] but does not expose the substrate to high temperatures; it can also be applied to transparent solids such as diamond or quartz [8].

The propagating SAW pulses were detected by a two-point probe beam deflection setup [8]. The two identical cw laser (532 nm) beams were focused to probe spots of 4 $\mu$m diameter to measure the normal component of the surface velocity. The distance between the line source and first probe spot could be varied between 2 and 30 mm. The second probe spot was located at a fixed distance of about 15 mm from the first one. The overall bandwidth of the setup was 500 MHz.

In the (112) direction of the Si(111) plane nonlinearity appears as a lengthening of the original pulse and the formation of two shock fronts at the edges. Figure 1(a) shows the transformation of a bipolar pulse into an N-type shape during propagation. A positive sign of the velocity indicates an outward movement of the surface. The leading positive peak propagates faster and the trailing negative peak slower than the linear signal. Such a behavior points to a negative real $B_{11}$ value for wave propagation in this direction. The corresponding deviations are $+0.16\%$ and $-0.45\%$, respectively for the pulse depicted by the solid line in Fig. 1(a). The dotted line represents the calculated profile. One can conclude that the model does not describe the regime of “strong” nonlinear evolution very well, in which the spikes are well developed at both edges of the pulse.

The horizontal velocity of this pulse, presented in Fig. 1(b), was calculated by performing a Hilbert transform of the measured vertical velocity. Positive values correspond to a longitudinal compression, negative to rarefaction. During propagation the profile of the horizontal velocity pulse transforms into a U shape. This behavior is quite different from nonlinear distortions of bulk waves in fluids or even nonlinear SAWs in isotropic fused silica, where the vertical velocity exhibits a U shape and the horizontal component has an N shape [8]. In terms of Eq. (1), such a difference is determined by the correlation between the imaginary and real parts of $B_{11}$. In isotropic solids (as well as for longitudinal waves in fluids) $B_{11}$ is imaginary. This means that the nonlinear interaction takes place only between longitudinal components of the acoustic field. In contrast to this, the evolution of a SAW pulse propagating in silicon along the (112) direction shows that $B_{11}$ contains a substantial real part, and correspondingly the interaction between shear components provides a major contribution to the nonlinear distortions.

In the frequency domain, shock formation is manifested by the generation of higher frequencies, connected with the appearance of new peaks in the spectrum, whereas frequency-down conversion is connected with a shift of peaks towards lower frequencies, as shown in Fig. 2, which presents the spectra for two different pulse amplitudes.
Increasing the energy of the exciting laser pulse allowed the shock formation distance to be reduced to several hundred micrometers. In this case it was possible to generate stresses that exceeded the mechanical strength of silicon. Above the critical stress, dramatic changes in the surface pulse propagating in the Si(111) plane were observed (Fig. 3). The pulse amplitude dropped to about half of its former value. A negative front now appeared at the beginning of the pulse and correspondingly the horizontal velocity consisted of a single peak, which was negative, in contrast to the situation shown in Fig. 1. Thus we observed a short tensile pulse, which can be related to the growth of a new free surface due to fracture. Inspection of the sample surface by scanning force microscopy confirmed crystal fracture. Cracks were observed at distances 0.3 to 2 mm from the source, extending perpendicular to the propagation direction, i.e., along (110). The length of the cracks was about 30–50 μm. Thus the growth velocity can be estimated from the tensile pulse duration to about 5000 m/s, resulting in a crack tip velocity of 2500 m/s, in close agreement with Ref. [2].

The solid line in Fig. 3(a) shows an N-type pulse recorded under the same conditions, but excited by a weaker laser pulse. Spikes have not yet developed and the evolution could be modeled in this case quite well by Eqs. (1) and (2) (see dotted line).

A very different kind of nonlinear waveform evolution was observed for propagation on the Si(001) plane in the directions 19° from the (110). These are the directions of a pure mode, in which the vectors of energy flow and phase velocity are parallel. In this case a shortening of the initial pulse duration to 3 ns took place, about half the exciting laser pulse duration, as shown in Fig. 4. The solution of Eq. (1) describes this behavior if a positive imaginary value is substituted.

The influence of dispersion on the nonlinear SAW evolution was studied for Si(111) coated with a thermally grown oxide layer of 110 nm thickness. Since the phase velocity of silicon oxide is smaller than that of silicon, such a layered structure has normal dispersion, i.e., the phase velocity decreases with increasing frequency, and the imaginary part of \( a \) in Eq. (1) is negative. The calculated dispersion curve is depicted in Fig. 6 by the solid line and can be approximated by a linear dependence in the frequency range considered.

Two regimes of surface pulse evolution on oxide-covered silicon were studied with different dispersion/ nonlinearity ratios. Figure 5(a) illustrates the case of relatively weak nonlinearity with a dominating dispersion effect. Here the measured and calculated oscillatory pulse profiles agree reasonably well. Similarly the measured and calculated dispersion curves are in good agreement, as shown in Fig. 6.
FIG. 5. (a) Waveforms of the vertical velocity measured in the (112) direction of the oxide-covered Si(111) plane at 4 mm (dashed line) and 19 mm (solid line) and simulation at 19 mm (dotted line). (b) Waveforms measured for stronger laser excitation.

At higher nonlinearity the phase shifts caused by the interaction between different spectral components are comparable to the influence of dispersion, and under certain conditions these two processes may essentially compensate each other, as found for the pulse shown in Fig. 5(b). The gradual frequency change in the oscillatory pulse essentially disappeared and the pulse shape resembles two serial Mexican hats [12]. As expected the dispersion effect disappears in this case, as can be seen in Fig. 6.

In conclusion, a large variety of waveforms was realized in anisotropic silicon by means of pulse lengthening, shortening, and shock front formation. These characteristic features of nonlinear pulses could be rationalized by the model presented. Different degrees of nonlinearity and stress levels—even up to fracture—were realized. Whereas for periodic elastic surface waves solitary behavior has been studied experimentally before [7], first results are reported here for short broad band surface pulses.

The authors gratefully acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG, He 909/27-2) of this work, which was carried out under the auspices of the trinational D-A-CH German, Austrian, and Swiss cooperation on the Synthesis of Superhard Materials.