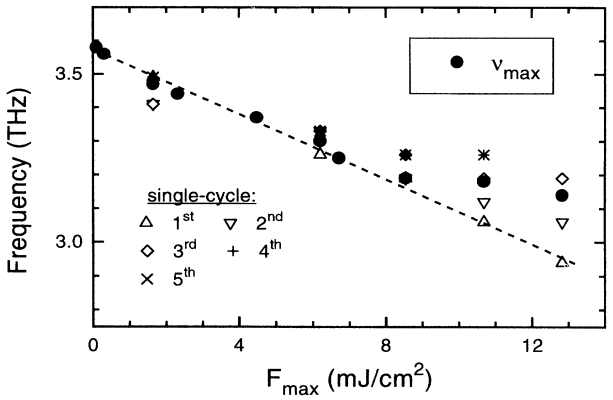

FIG. 3. Excitation-density dependence of the phonon frequency. Solid circles show the maximum of the Fourier transforms, open symbols the "instantameous" frequencies determined by fitting different cycles of the oscillating signal.

fast relaxation of the electronic background signal at high pump intensities.

Taking into account the inhomogeneity of the pump-created carrier density (see below), the data indicate a linear decrease of the phonon frequency with carrier density. The estimation of the photoexcited carrier density achieved in our experiments shows that several percent of the valence electrons are excited at the highest pump intensities, so that a considerable weakening of the crystal is expected [11]. Therefore the observed reduction of the restoring force for the A_1 lattice displacement appears as a natural consequence of the excitation of a large number of electrons from bonding into antibonding states. This effect may be described as a purely electronic softening of the crystal lattice.

In a recent report, Cheng *et al.* have observed a transient redshift of the phonon frequency in Sb and assigned this to ionic screening by the photoexcited carriers [10]. However, screening effects can be excluded for the case of tellurium, since the crystal bonds are predominantly covalent rather than ionic. Also, there is no plasmon-phonon coupling [12] for the A_1 phonon, since this is not infrared active. An additional softening due to lattice heating or emission of nonequlibrium phonons by the photoexcited carriers—which is expected to take place on a ps time scale—appears to be negligible in our experiments, since the frequency shifts occur "instantaneously" with the excitation. In addition, seperate experiments showed that the influence of lattice heating on the coherent phonon frequency is much smaller than that of the photoexcitation [14].

In order to understand the shape of the phonon response and the partial relaxation of the oscillation frequency, the strong inhomogeneity of the initial carrier density has to be taken into account. The probe pulse "sees" signal contributions from different depths below the sample surface, corresponding to different carrier densities. Increasing the excitation fluence corresponds to an increasing range of densities, so that a density-dependent redshift will lead to an asymmetric broadening of the phonon response, as observed in Fig. 2, and to an apparent decrease of the coherent phonon dephasing time. Therefore the distinct low-frequency tail of the phonon response at the highest pump intensities suggests a substantial mode softening in the region of highest carrier density that would be underestimated if only the shift of the maximum were taken into account.

On the other hand, the large density gradient associated with the short absorption length will lead to a very fast carrier diffusion at high excitation power and therefore to a rapid decrease of the carrier density at the surface. Assuming a monotonous decrease of the frequency with carrier density provides a consistent explanation for the temporal shift of the instantaneous oscillation frequency, which corresponds directly to the rapid decay of the electronic background signal.

To check the consistency of our interpretation and to prove the electronic origin of the frequency shifts, we have performed additional experiments with double-pulse excitation of the sample: We use a prepump with variable intensity to create a certain carrier density and observe the influence of these carriers on the dynamics of coherent phonons generated by a second pump pulse. To avoid interference effects of the coherent phonons [15], we chose a time delay of 6 ps between the pump pulses, so that the oscillations created by the prepump have completely vanished when the second pulse hits the sample.

In Fig. 4 we compare the phonon frequency shift under single- and double-pulse excitation. The experimental situation is illustrated in the inset, which shows the transient reflectivity changes induced by the double-pulse excitation. In the double-pump experiment, the fluence of the second pump is kept constant at $F = 0.5$ mJ/cm^2, while the fluence of the prepump is varied from 0.3 to 1.5 mJ/cm^2. For this density range, we did not find a delay dependence of the frequency. Therefore in Fig. 4 we show a single frequency for each measurement, which is obtained by fitting the oscillating signal contribution for positive time delays. In the time interval between the two excitation pulses there is a significant decrease of the electronic background signal created by the prepump, due to carrier diffusion into the sample. To allow a direct comparison of the two types of experiments, we show the data as a function of the maximum electronic background at time delay zero, which is taken as a direct measure of the effective total carrier density created by both pumps. The data clearly show an identical dependence of the oscillation frequency in both types of experiments. As the phonons created by the second pump are affected only by varying the carrier density excited by the prepump, this is an unambiguous proof for a predominantly electronic origin of the observed

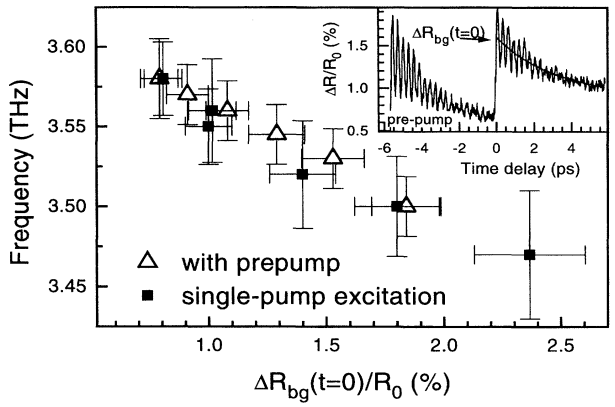

FIG. 4. Phonon frequency shift under single-pulse and double-pulse excitation. The electronic background signal at $t = 0$ is used as a measure of the effective carrier density. The inset illustrates the double-pulse experiment.

mode softening. A possible influence of anharmonicity of the phonon itself [9,16] can be safely excluded, since the intensity of the second pump, and thus the phonon amplitude, was kept constant.

Figure 5 shows the values of the coherent phonon dephasing time, as determined from fitting the data with a damped cosine, for single- and double-pulse excitation. In contrast with the frequency, the dephasing time remains quite unaffected by changing the intensity of the prepump. This indicates that there is no significant direct coupling between the phonon and the free carriers. Therefore the decrease of the dephasing times observed in the single-pump experiments can only be assigned to the increasing inhomogeneity, as already concluded from the shape of the Fourier spectra. In the double-pulse experiment the delay between the first and second pump is sufficiently large to allow a significant reduction of the spatial inhomogeneity by carrier diffusion. The slight offset of the dephasing times in the double-pulse experiment may be attributed to an increase of the lattice temperature due to the prepulse

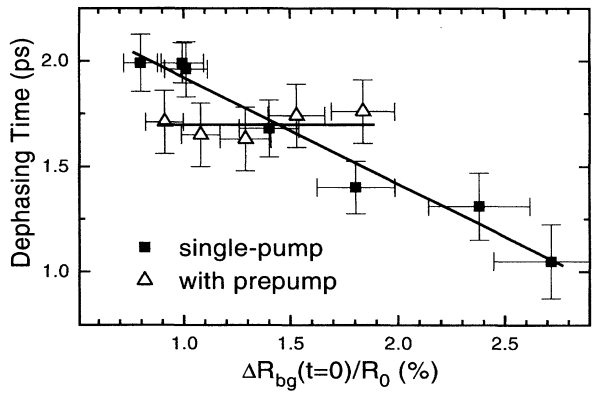

FIG. 5. Density dependence of the coherent phonon dephasing time under single-pulse and double-pulse excitation.

[14], however, this explanation is not crucial for the interpretation of the main results of this paper.

In conclusion, we present an experimental investigation of the coherent phonon dynamics in tellurium as a function of excitation density. All experimental results can be consistently explained in terms of a monotonous decrease of the A_1-phonon frequency with increasing carrier density, i.e., an electronic weakening of the crystal lattice. For the highest pump powers studied, the photoexcited carrier densities can be estimated to be several percent of all valence electrons. At these densities the phonon response contains low-frequency contributions well below 1 THz, suggesting a substantial destabilization of the crystal by the optical excitation. Therefore our experiments provide microscopic information that cannot be obtained from experimental methods that are only sensitive to changes of the crystal structure.

We are grateful to Professor P. Grosse for supplying the samples. This work has been supported by the Deutsche Forschungsgemeinschaft.

*Present address: FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

[1] J. A. Van Vechten, R. Tsu, and F. W. Saris, Phys. Lett. **74A**, 422 (1979).
[2] C. V. Shank, R. Yen, and C. Hirlimann, Phys. Rev. Lett. **51**, 900 (1983); H. W. K. Tom, G. D. Aumiller, and C. H. Brito-Cruz, *ibid.* **60**, 1438 (1988).
[3] P. Saeta *et al.,* Phys. Rev. Lett. **67**, 1023 (1991); S. V. Govorkov *et al.,* Optics Lett. **16**, 1013 (1991); K. Sokolowski-Tinten *et al.,* Appl. Phys. A **53**, 227 (1991); Phys. Rev. B **51**, 14 186 (1995).
[4] S. DeSilvestri *et al.,* Chem. Phys. Lett. **116**, 146 (1985).
[5] T. K. Cheng *et al.,* Appl. Phys. Lett. **57**, 1004 (1990); T. K. Cheng *et al., ibid.* **59**, 1923 (1991).
[6] G. C. Cho, W. Kütt, and H. Kurz, Phys. Rev. Lett. **65**, 764 (1990); W. Kütt, W. Albrecht, and H. Kurz, IEEE J. Quantum. Electron. **28**, 2434 (1992);
[7] T. Dekorsy *et al.,* Phys. Rev. Lett. **74**, 738 (1995).
[8] H. J. Zeiger *et al.,* Phys. Rev. B **45**, 768 (1992).
[9] T. K. Cheng, M. S. Dresselhaus, and E. P. Ippen, Appl. Phys. Lett. **62**, 1901 (1993).
[10] T. K. Cheng *et al., Ultrafast Phenomena IX,* edited by P. F. Barbara and W. H. Knox, Springer Series in Chemical Physics, Vol. 60 (Springer, Berlin, 1994), p. 301.
[11] P. Stampfli and K. H. Bennemann, Phys. Rev. B **42**, 7163 (1990); **46**, 10 686 (1992); **49**, 7299 (1994).
[12] A. V. Kutznetzov and C. J. Stanton, Phys. Rev. B **51**, 7555 (1995).
[13] P. Grosse and W. Richter, in *Landolt-Börnstein, New Series,* edited by O. Madelung (Springer, Berlin, 1983), Vol. 17.
[14] K. Wienecke (unpublished).
[15] T. Dekorsy *et al.,* Europhys. Lett. **23**, 223 (1993).
[16] C. M. Foster *et al.,* Phys. Rev. B **48**, 10 160 (1993); H. J. Bakker, S. Hunsche, and H. Kurz, *ibid.* **48**, 13 524 (1993); **50**, 914 (1994).