is −28.9°K. We have performed the calculations for the hcp lattice, assuming 16 sublattices. The lowest eigenvalue was found to be −27.3°K. Unfortunately in this case the lowest eigenvalue does not have an eigenvector that corresponds to a physical quadrupole array. (We tried to guess some configurations and the lowest energy we could obtain is −16.5°K; the corresponding configuration contains only two distinct sublattices with quadrupoles aligned along two different diagonals of the hexagonal unit cell.)

We have shown by classical calculations that the lowest energy of the fcc lattice is −28.9°K and that the lowest energy of the hcc lattice must be greater than −27.3°K. This shows that the molecules in pure ortho-hydrogen at 0°K will prefer to form a fcc lattice with orientations according to Fig. 1 rather than a hcp lattice, in accord with the infrared-absorption measurements of Clouter and Gush.[11]

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EFFICIENT PHASE-MATCHED HARMONIC GENERATION IN TELLURIUM WITH A CO_2 LASER AT 10.6 μ

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Harmonic generation[1,2] and optical parametric phenomena[3] have been, heretofore, limited to near IR frequencies because of lack of suitable pump source at longer wavelengths and also because of lack of known efficient and phase-matchable[4] optically nonlinear materials transparent in this region. In this paper we report extraordinarily large amounts of second-harmonic generation (SHG) in single-crystal tellurium using a focussed, 10.6μ, continuous-wave, CO_2 laser[5] as the fundamental. Thus, Te is the first elemental crystal in which phase-matched harmonic generation with an infrared source is achieved. (Earlier, reflected light harmonics from a Te surface have been reported using a Q-switched ruby-laser output as the fundamental.[6]) Under our experimental conditions, a second-harmonic power (at 5.3 μ) of 10 μW was obtained when a fundamental power of 0.17 W was phase matched over a 9-mm length of the crystal. The measured nonlinear coefficient of Te is about 4000 times d_{36} of potassium dihydrogen phosphate. This optical nonlinear coefficient is the highest of any material reported to date and opens up the possibilities of extending other nonlinear optical effects such as parametric amplification/oscillation into the infrared with cw high-power CO_2 lasers.

Te is a member of Group VI B of the periodic table and is an elemental semiconductor. Crystals of tellurium have 32 (D_3) point-group symmetry and belong to the $P3_{1,2}21$ space group (Hermann-Mauguin notation)[7] assuring an absence of inversion symmetry necessary for SHG and other nonlinear optical phenomena. The crystal in its purest form (intrinsic, p type)[8] is essentially transparent from about 5 μ (band gap at 4 μ) to wavelengths longer than about 25 μ.[9] Single-crystal Te is uniax-

ial and piezoelectric,[10] and has large refractive index. It also exhibits large positive birefringence[9] which allows easy phase matching[4] of the fundamental wave at 10.5915 μ and the second-harmonic wave at 5.2958 μ. (The index-of-refraction data as reported by Caldwell and Fan[9] are shown in Fig. 1.) For investigations of nonlinear optical effects in Te with the 10.6μ CO_2 laser, it is advantageous to work with tellurium crystals having low hole concentrations, since a strong absorption band exists at 11 μ for the extraordinary wave (E parallel to c axis) in samples with high hole concentrations.

For tellurium the components of second-order polarization are given (using Kleinman's representation[11]) by[12]

$$P_x^{2\omega} = d_{11}E_x^2 - d_{11}E_y^2 + 2d_{14}E_yE_z,$$

$$P_y^{2\omega} = -2d_{14}E_zE_x - 2d_{11}E_xE_y, \quad P_z^{2\omega} = 0, \qquad (1)$$

where the E's are the electric field components of the fundamental wave at frequency ω. For piezoelectric crystals having 32 point-group symmetry, d_{14} is expected from Kleinman's symmetry conditions[11] to be very small. (See also Miller's results[13] on quartz which is another piezoelectric crystal of class 32.)

For phase-matched SHG, we have the fundamental beam propagating as an extraordinary wave in the yz plane at the phase-match angle θ_m from the c axis (optic axis) of the Te crystal. The second harmonic, then, will be an ordinary wave polarized along x axis, and the fundamental and the harmonic waves will be phase matched. θ_m is calculated from the data in Fig. 1. It should be pointed out that the data in Fig. 1 are derived from Ref. 9 and were obtained by measurements not in a single crystal but in a polycrystalline sample. Thus the θ_m calculated may not be exactly correct. Using $n_\omega{}^e = 6.240$, $n_\omega{}^o = 4.795$, and $n_{2\omega}{}^o = 4.856$, we obtain $\theta_m = 14°36'$. ($n_\omega{}^e$, $n_\omega{}^o$, etc., are the various refractive indices.)

SHG experiments were carried out with a CO_2-N_2-He laser described in Ref. 5. This laser is capable of generating cw power output in excess of 100 W; however, for the present experiments only a very small fraction of the total power output was utilized. Single crystals of Te (nominal impurity 1:10^6, resistivity ≈ 1 Ω cm) were cut from a boule 7-8 mm in diam and 10 cm long (along c axis). The samples were polished at the required θ_m. With all the samples tested, θ_m was measured to be 14°10' $\pm$ 30' close to the calculated θ_m of 14°36'. Figure 2 shows the SHG output as a function of the external angle deviation from $\theta_m = 19°30'$ for a crystal 0.9 cm long (see Table I for other

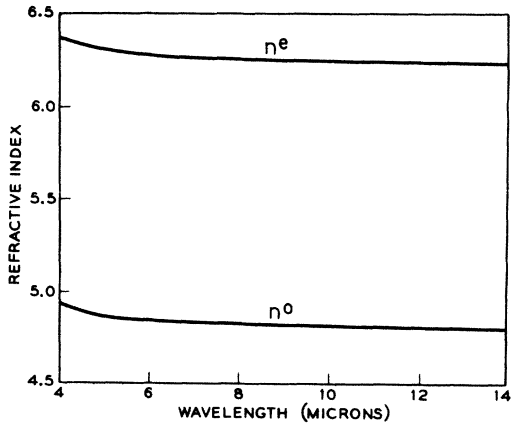

FIG. 1. Refractive index of single-crystal tellurium as a function of wavelength (as reported by Caldwell and Fan[9]).

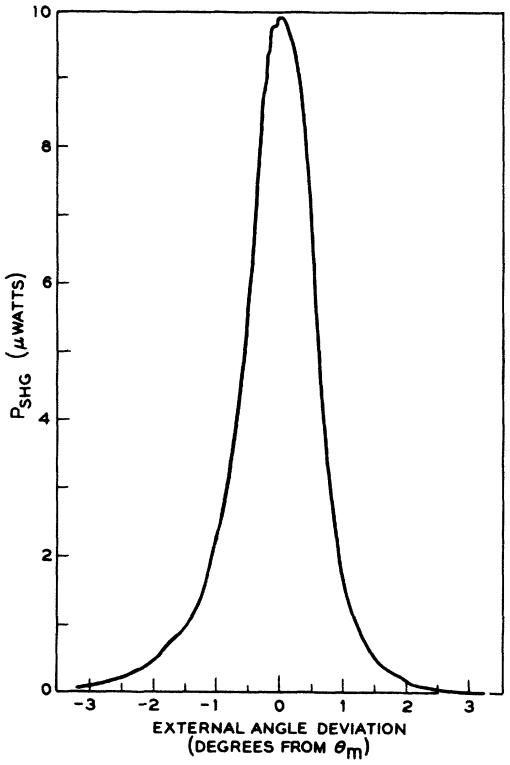

FIG. 2. Second-harmonic power generated in tellurium as a function of external angle deviation from θ_m = 14°10' for the 9-mm-long crystal described in Table I. ($\lambda_{fundamental} = 10.5915$ μ and $\lambda_{SHG} = 5.2958$ μ.)

Table I. Summary of SHG experiments in Te.

Sample length (cm)	No. of samples	$P_{10.6\mu}$ (W)	$P_{5.3\mu}$ (μW)	w_0^2 (mm^2)
0.162	4	0.17	0.5	3.2
0.9	1	0.17	10.0	3.2

experimental data). Phase-matched SHG was obtained in crystals of various lengths from 0.16 to 0.9 cm. Table I gives the results of five of the samples. It can be seen that for the 0.9-cm long crystal, a second-harmonic efficiency of 6×10^{-5} is achieved with a fundamental pump power of 0.17 W. (The fundamental power occurred in a single longitudinal mode of the optical cavity.[5])

To determine the nonlinear coefficient d_{11} of Te we use the data on the thinner sample (Table I) in order to avoid the possible effects of double refraction in the longer crystal.[14] (We assume that d_{14} is negligible compared to d_{11}.) Calculations yield d_{11} (Te) = $(1.27\pm0.2)\times10^{-5}$ esu (see for example Ashkin, Boyd, and Dziedzic[15]). Carrying out similar calculations for the longer sample, we obtain d_{11}(Te) = $(1.02\pm0.2)\times10^{-5}$ esu. This indicates that up to $l\approx1$ cm, the effects of double refraction are still negligible (since $P_{SHG}\propto l^2$). This is consistent with calculated[14] aperture length $l_a\approx2$ cm.

The measured large value of the nonlinear coefficient is in qualitative agreement with Miller's phenomenological theory[16] of SHG in piezoelectric crystals. This theory predicts that piezoelectric crystals with large refractive indices should also have large nonlinear coefficients. Tellurium is the first elemental crystal in which the nonlinear coefficient is measured and reported. Thus, it is instructive to compare the measured d_{11}(Te) with that predicted from the theory. To accomplish this, we first obtain the quantity $\delta_{11}(\mathrm{Te})/(4\pi)^3$. It is shown in Ref. 16 that even though the d's vary by orders of magnitude between crystals of the same class, the corresponding δ's are much more nearly constant. Using the measured value of $d_{11}(\mathrm{Te}) = 1.27\times10^{-5}$ esu, we obtain $\delta_{11}(\mathrm{Te})/(4\pi)^3 = 0.33$ in units of 3×10^{-9} esu. Quartz is another piezoelectric crystal of 32 point-group symmetry (same as Te) for which Miller[16] reports $\delta_{11}(\mathrm{quartz})/(4\pi)^3 = 0.32$ in the same units. Thus, we see that the measured $\delta_{11}(\mathrm{Te})/(4\pi)^3$ is in qualitative agreement with δ's for other crystal of class 32.

It can also be seen that the large nonlinear coefficient of tellurium allows one to reach parametric oscillator threshold at very modest pump powers of a fraction of a watt. And with the CO_2 laser as the pump source, for example, a tunable oscillator in the infrared should be possible because of phase matchability in Te.

In addition, it should be pointed out that single-crystal selenium is another elemental semiconductor belonging to the group VI B of the periodic table. It has the same point-group symmetry (32) and space group ($P3_{1,2}21$) as tellurium. Se has a band gap at about 8000 Å, and the refractive indices are $n^o = 2.78\pm0.02$ and $n^e = 3.58\pm0.02$ in the infrared region as reported by Caldwell and Fan.[9] Thus, phase-matched harmonic generation and optical parametric phenomena should also be observed in selenium using the high-power infrared lasers.

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DECAY OF He^8

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The possible observation of He^8 was first reported in 1961 on the basis of two hammer tracks in emulsions.[1] In 1963 Nefkens[2] tentatively reported observing its decay with a half-life of 30 ± 20 msec on the basis of beta measurements on a boron target irradiated with photons. More, recently, Whetstone and Thomas,[3] while reporting their observation by means of a E-dE/dx counter telescope of He^6 from Cf^{252} fission, reported some events which could have been He^8, although the evidence was not conclusive. Cerny, Cosper, and Gatti,[4] from a similar experiment, report definitely having observed He^8 particles from Cf^{252} fission. It is the purpose of this Letter to report the decay properties of He^8, including our half-life measurement of 122 ± 2 msec, which indicates that the work of Nefkens is erroneous.

From the known energy levels of Li^8 (see Fig. 1), it was expected that He^8 would undergo beta decay to the 0.98-MeV level and to unbound levels at 3.21 MeV and higher energies. Thus, because the ground-state transition is second forbidden, every He^8 decay would be expected to lead to the emission of either a 0.98-MeV gamma ray or a delayed neutron. From logft calculations performed by Kurath[5] and an estimate[6] of the mass of He^8, it was predicted that the half-life would be in the hundred-millisecond range and the delayed neutron branch would be of the order of 10%. Thus the main difficulty[7] in observing He^8 was expected to be the interference from 176-msec Li^9, a delayed neutron emitter which is likely to be produced in greater yield in practically any reaction which produces He^8.

The method adopted[8] involved the diffusion loss[9] of the active He from thin plastic foils or absorbent cotton fibers irradiated by the 2.2-GeV external proton beam of the Brookhaven Cosmotron. In the diffusion-loss method a recoil nucleus comes to the end of its range and then diffuses out of the last foil or fiber.[10] The He activity is then transported to the counters by expanding some carrier helium through the target volume into the evacuated counting chambers. The apparatus is shown schematically in Fig. 2. A plastic target consisted of a roll of 1-mil foil, 2 in. in diameter and 1 in. high, placed on a wire gauze $\frac{1}{2}$ in. above the bottom of an aluminum container. The center hole in the roll of plastic was plugged, and the roll was fitted snugly into the container so that helium carrier gas below the roll would

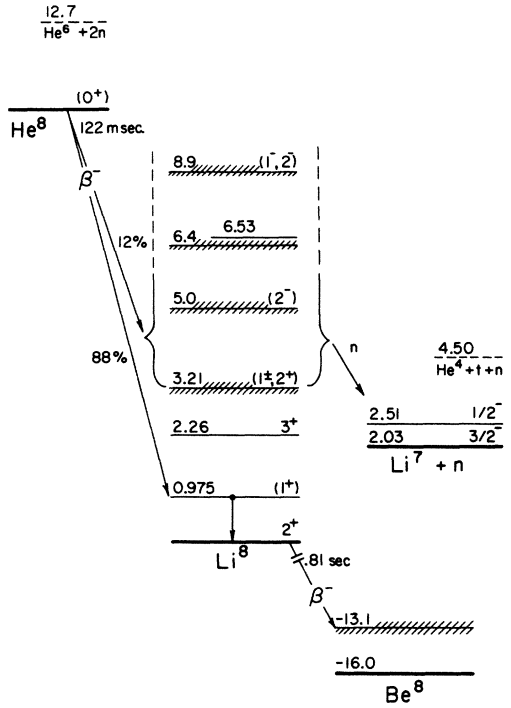

FIG. 1. The decay scheme of He^8, from Ref. 17 and the present work. All energies are relative to the Li^8 ground state.