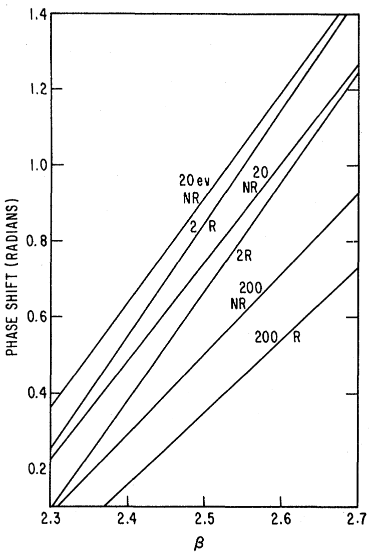

FIG. 4. p-wave phase shifts as a function of β for cesium II. The two relativistic cases $\lambda = 0$ and $\lambda = 1$ are very close; the average is shown on the curves labeled R.

use of Eq. (1) or (2), and are not shown. The cases for $l = 1$ were prepared to show that our argument is not limited to s waves.

A tentative conclusion may be drawn from these numerical experiments: At low energies (~200 eV, or less) it makes little difference whether the relativistic or nonrelativistic description of electron scattering is used, even from heavy atoms. What seems to be important is that whichever potential is used for scattering also yield the correct levels for the bound states.

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MULTIPHOTON PLASMA PRODUCTION AND STIMULATED RECOMBINATION RADIATION IN SEMICONDUCTORS

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We report the observation of stimulated recombination radiation from PbTe at its band gap (~6.5 μ) induced by intense 10.6-μ radiation from a Q-switched CO_2 laser. Since in PbTe no second harmonic can be generated, we believe our experiments provide the first example of multiphoton pair production, where the number of pairs created is sufficient to produce a dense plasma and laser oscillation in the semiconductor. The mechanism responsible for electron-hole pair production in recent experiments on GaAs [1] and CdS [2] is obscured by possible production of the second harmonic of the incident radiation and subsequent pair production by this second harmonic. Further, as numerical estimates given below[3] indicate, the intensity of the 10.6-μ radiation in our experiments is already approaching the limit where Zener breakdown competes with two-photon absorption.

The apparatus is shown in Fig. 1. The Q-switched CO_2 laser was described in detail earlier,[4] and produced ~10 kW of 10.6-μ radiation in a pulse width of ~200 nsec. Samples were mounted on a cold finger (temperature T) in a helium Dewar.

With the 10.6-μ radiation focused to a spot ~200 μ in diameter, the spectrum shown in Fig. 2(a) was obtained from single-crystal n-type PbTe at 15°K. No recombination radia-

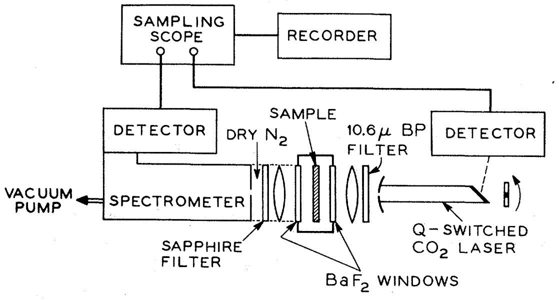

FIG. 1. Apparatus. BaF_2 windows provide optical access to the He Dewar. The optical path for recombination radiation is continuously flushed with N_2 gas to eliminate water-vapor absorption near 6.5 μ.

tion was observed at $T \simeq 80°\text{K}$. Similar results were obtained in p-type samples. The recombination radiation, confined to a forward-backward cone angle of 3° about the 10.6-μ beam direction, was elliptically polarized in the ratio of 7/1. The direction of polarization was independent of the CO_2 laser polarization. The observed peak recombination radiation per pulse was $\sim 3\times10^{-4}$ W having a linewidth of ~0.002 eV [see Fig. 2 (a)]. The relative power at 65 μ varied approximately as the square of the 10.6-μ power, in agreement with the assumption of two-photon excitation. It should be mentioned that we also observed band-gap recombination radiation (~5.3 μ) in single-crystal n-type InSb ($N_0 \simeq 5\times10^{15}\ \text{cm}^{-3}$; $\mu \simeq 6\times10^5\ \text{cm}^2\ \text{V}^{-1}\ \text{sec}^{-1}$ at 77°K) under the experimental conditions described for Fig. 2(a). However, because of the presence of the second harmonic in InSb, the pair-production mechanism cannot be unambiguously identified as two-photon excitation and our results will not be discussed here.

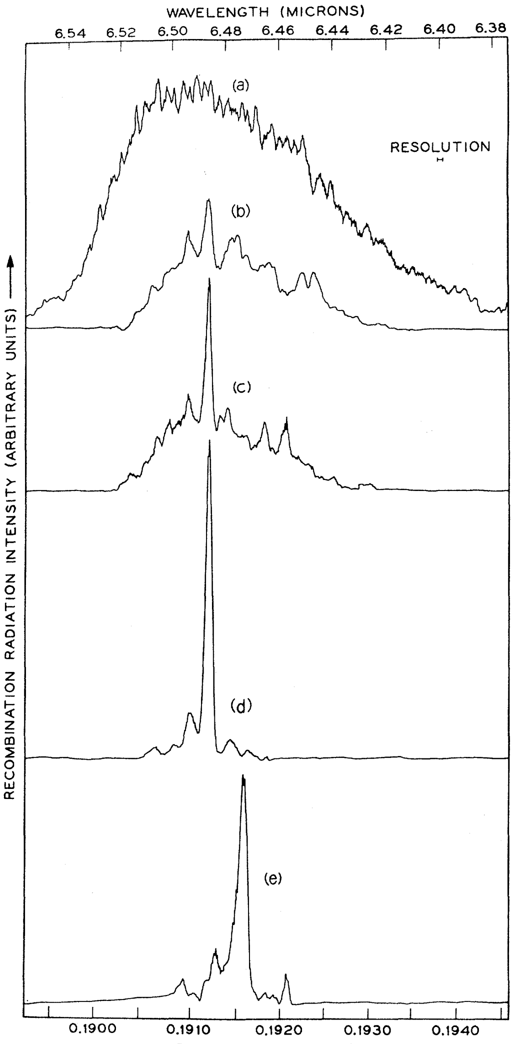

FIG. 2. Spectrum of recombination radiation from single-crystal n-type PbTe ($n_0 \simeq 10^{17}\ \text{cm}^{-3}$; $\mu \simeq 10^6\ \text{cm}^2\ \text{V}^{-1}\ \text{sec}^{-1}$). The sample was a polished slab 1.59 mm thick. (a) ~10-kW CO_2 laser beam focused to a spot ~200 μ in diameter; $T \simeq 15°\text{K}$. (b), (c), (d) ~10-kW CO_2 laser beam focused to ~50 μ; laser intensity increased in successive 10% increments; $T \simeq 15°\text{K}$. (e) Same conditions as in Fig. 2(d), except $T \simeq 20°\text{K}$.

In considering the creation of electron-hole pairs by 10.6-μ radiation in PbTe, we employ Keldysh's finite-frequency generalization[3] of the theory of Zener breakdown.[5] The fundamental parameter is the ratio, γ, of the Zener tunneling time to the period of the electric field:

$$\gamma \equiv \omega/\omega_t = (\omega/eE)(2m_\gamma\epsilon_g)^{1/2}.$$

Here ω is the frequency of the incident radiation; E is the electric field strength; m_γ is the reduced electron-hole mass ($\sim 0.01m_e$); and ϵ_g is the direct forbidden energy gap (~0.2 eV).

For $\gamma \ll 1$, Kane's formula[5] for zero-frequency Zener tunneling is obtained. In the limit $\gamma \gg 1$, Keldysh's formula[3] takes the form

$$W = \frac{2}{9\pi}\omega\left(\frac{m_\gamma\omega}{\hbar}\right)^{3/2} \Phi[(2\langle x+1\rangle - 2x)^{1/2}] \times\left(\frac{e^2E^2}{16m_\gamma\omega^2\epsilon_g}\right)^{\langle x+1\rangle} \times\exp\left\{2\langle x+1\rangle\left(1-\frac{e^2E^2}{4m_\gamma\omega^2\epsilon_g}\right)\right\}, \tag{1}$$

where

$$\Phi(z) = e^{-z^2}\int_0^z e^{y^2}dy, \quad x \equiv \frac{\epsilon_g}{\hbar\omega}\left\{1+\frac{e^2E^2}{4m_\gamma\omega^2\epsilon_g}\right\}.$$

$\langle x\rangle$ means the integer part of x; here $\langle x+1\rangle = 2$. Equation (1) has the form of a two-photon absorption probability. It is interesting to note that Zener breakdown and multiphoton absorption are both limiting cases of the same calculation.

For a spot size 200 μ in diameter, the experimental field strength, $E \approx 10^4$ V/cm, gives $\gamma \approx 20$ for the light holes and electrons. From

Eq. (1) we obtain $W = 10^{25}$ cm^{-3} sec^{-1}. Assuming that each pair recombines radiatively[6] with an active volume $V = 4\times10^{-5}$ cm^3 and a solid angle of emission $d\Omega = 2\times10^{-3}$ sr, we expect a peak power of ~10^{-3} W per pulse. In view of experimental uncertainties, there is acceptable agreement between the calculated and measured (see above) power.[7]

Equation (1) neglects parity conservation which would forbid a two-photon absorption in PbTe at the conduction-band minimum. However, in our case, because $2\hbar\omega - Eg > 0$, the excitation occurs away from the minimum and the selection rule is thus relaxed. Braunstein[8] has calculated two-photon absorption taking into account such selection rules. The application of his theory to an allowed-forbidden two-photon absorption in a two-band model of PbTe gives order-of-magnitude agreement with the above results for W.

Assuming that the radiative recombination time τ_r is short compared to the Q-switched laser-pulse duration (~200 nsec), the steady-state density of pairs created per pulse is given by

$$N_e = W\tau_r. \tag{2}$$

Using the reasonable value of $\tau_r \approx 10$ nsec (Ref. 6) gives $N_e \approx 10^{17}$ cm^{-3}. For this N_e, at 15°K the quasi-Fermi levels of the excess holes and electrons are separated by more than ϵ_g, so that the recombination radiation should be stimulated. However, in order to achieve laser oscillation, bulk scattering losses as well as reflection losses in the PbTe samples must be overcome.

By modifying the focusing arrangement in the above experiment to obtain a spot size ~50 μ in diameter, laser oscillation was obtained in the PbTe. Figures 2(b), 2(c), and 2(d) show the radiation spectra for successive 10% increases in the CO$_2$ laser intensity. It is seen that the mode at ~6.486 μ grows much faster than the rest of the radiation, indicating that laser oscillation is achieved. This mode lies close to the peak of the recombination radiation spectrum for lower CO$_2$ laser intensity shown in Fig. 2(a). Finally, Fig. 2(e) shows the radiation spectrum from PbTe under the same CO$_2$ laser intensity as in Fig. 2(d), but at somewhat higher sample temperature. Here, the mode at 6.473 μ dominates, indicating that, as expected,[9] the peak of the band-gap radiation has shifted towards shorter wavelengths. The laser radiation was linearly polarized (better than 99%) probably as the result of the properties of the optical cavity. Additional confirmation of laser oscillation is given in Fig. 3 which shows the dependence of the recombination radiation on the CO$_2$ laser intensity under these experimental conditions. However, unlike the lower intensity case [Fig. 2(a)], the field dependence of the pair-creation rate cannot be inferred from Fig. 3, because the emission characteristics of the recombination radiation are greatly different in the lasing and nonlasing situations.

We have shown that nonlinear absorption of radiation in a semiconductor can result in quite large electron-hole pair-production rates. Because the incident radiation is only weakly absorbed, such pair production is not limited to surface regions of the semiconductor. In achieving for the first time laser oscillation in bulk PbTe, we have demonstrated the usefulness of multiphoton pair production in homogeneous optical pumping of semiconductor lasers.

The effect described here is of interest in another context. The ability to generate dense plasmas of controllable density and spatial extent inside semiconductors should make fea-

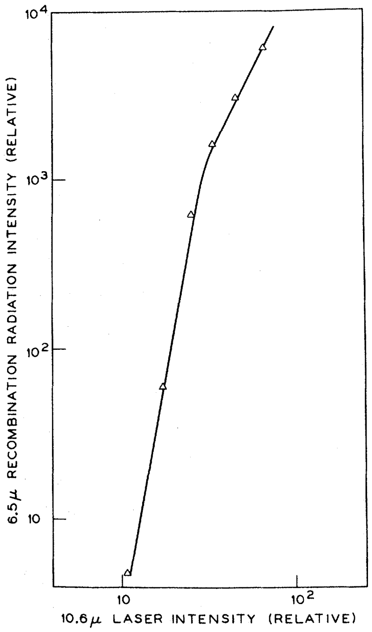

FIG. 3. Variation of PbTe total recombination radiation intensity with CO$_2$ laser intensity; $T \simeq 15$°K; maximum CO$_2$ laser power ~10 kW; focus ~50 μ in diameter.

sibile experiments otherwise prohibitively difficult in solid-state plasmas, such as the observation of light scattering by plasmons. On the other hand, it is important to be aware of this pair-production effect when attempting high-intensity optical experiments in even "transparent" solid-state plasmas.

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DETECTION OF NUCLEAR MAGNETIC RESONANCE IN A 235-nsec NUCLEAR STATE BY PERTURBED ANGULAR CORRELATIONS

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We have observed resonant rf absorption by 10^4 nuclei in the 74.8-keV excited state of Rh^{100} by combining nuclear magnetic resonance in a ferromagnet[1] with perturbed angular correlations (PAC) of the 84.0-74.8-keV γ-ray cascade.[2] The 4-day isotope Pd^{100}, which was diffused into a nickel foil, fed the cascade via electron-capture decay, maintaining an average of 0.006 nuclei in the 74.8-keV state during the experiment. A total of 2.8×10^6 coincidences were recorded by two 3-in.×3-in. NaI(Tl) counters at 180°, while the sample was polarized in a field of 100 G along the counter axis. Perpendicular to the dc field, an oscillating field of 3 G was stepped through the frequency range 289.5-355.5 MHz. Resonant absorption was detected by a drop in the coincidence rate centered about 322.5 MHz, as shown in Fig. 1.

The method of PAC has for sometime provided the only way to measure precession frequencies in short-lived nuclear states, with lifetimes in the range $10^{-11}\ \mathrm{sec} < \tau < 10^{-6}$ sec. Time-differential PAC,[3] and more recently the digital analysis method (DAPAC),[4] have raised the upper limit on lifetimes available to at least 10^{-3} sec and have greatly improved the accuracy possible with PAC. Still it is desirable to develop a method that combines the precision of nmr with the extremely high sensitivity of PAC in order to enjoy the advantages of both methods.

Abragam and Pound discussed the possibility of detecting resonant absorption in a short-lived nuclear state by observing the angular-correlation anisotropy as early as 1953.[5] They formulated the conditions for such an experiment: (1) The splitting of the substates should be large compared to the natural nuclear linewidth; i.e., $\omega_L\tau_N \gg 1$, where ω_L and τ_N are, respectively, the Larmor frequency and the nuclear lifetime. (2) The rf field, H_1, must have sufficient amplitude to induce transitions in each nucleus in times of the order of the nuclear lifetime, $\gamma H_1 \sim \tau_N{}^{-1}$, where γ is the nuclear gyromagnetic ratio. (3) Other time-dependent interactions should be minimal, so that the nuclear spin-correlation time τ_c is of the order of the lifetime, $\tau_c \sim \tau_N$. These conditions, especially (2), turn out to be very stringent when applied to the PAC sources