Threshold generation of harmonics in the microwave range in bismuth

S. A. Vitkalov, V. F. Gantmakher, and G. I. Leviev
Institute of Solid State Physics, Academy of Sciences of the USSR

(Submitted 23 April 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **39**, No. 12, 540–542 (25 June 1984)

When bismuth is irradiated with an intense electromagnetic wave, it is usually possible to observe the stationary generation of the second harmonic whose power increases quadratically with the pump power. Against the background of this typical generation and in the presence of a magnetic field, we have observed threshold generation, which is sometimes two orders of magnitude more powerful. This generation, whose pulses are several times shorter than the pumping pulses, is delayed relative to the beginning of irradiation by several microseconds.

In this experiment we analyzed the nonlinear optics of bismuth single crystals in a magnetic field $H \approx 10$–80 kOe at temperatures or 2–4.2 K in the microwave range. Under these conditions, magnetoplasma waves with the spectrum $\omega = \alpha H k$ propagate in bismuth. The coefficient $\alpha \sim 10^4$ cm/s Oe, (Ref. 1) so that the velocity of the wave $v = \omega/k \approx 10^8$–$10^9$ cm/s. This indicates that the ratio of the amplitudes of the electric and magnetic fields is $E_\omega/H_\omega = v/c \approx 10^{-2}$, and the field-induced forces eE_ω and $(v_F/c)eH_\omega$ acting on the carrier have the same ratio as $v/v_F \gg 1$ (the Fermi velocity is $v_F \approx 3\times 10^7$ cm/s). The comparatively large electric field distinguishes the conditions of our experiment from experiments in normal metals with a long mean free path, where the magnetic field of the wave is the source of the nonlinearity.[2,3] On the other hand, comparing the conditions of the experiment with those usually realized in a gaseous plasma, it should be emphasized that the spectrum of the carriers is not only degenerate ($\epsilon_F \gg T$), but the ultraquantum limit $\epsilon_F \lesssim \hbar\Omega$ ($\Omega = eH/mc$ is the cyclotron frequency) is attained in these fields for electrons. For holes in the strongest fields with $\mathbf{H} \| C_3$ we would have the ratio $\epsilon_F \hbar\Omega \approx 4$.

Because of the nonlinear processes in bismuth, higher-order harmonics appear in the spectrum of the reflected signal. The experiments consisted of studying the powers of the second and third harmonics $P_{2\omega}$ and $P_{3\omega}$ as a function of the incident power P_ω and as a function of the external field and of the time.

The experiments were performed in the following types of Bi samples: 1) discs with a thickness of $d = 0.2$ mm and 1 mm and a diameter of $\Phi 17/8$ mm with orientation $C_3 \| \mathbf{n}$ to within 3°, and 2) a $11\times 9\times 13$ mm cube, in which the C_1 axis (bisector), C_2 axis (binary), and C_3 axis (trigonal) are oriented perpendicular to the faces to within 3°.

The samples were part of the wall of a rectangular cavity (TE_{101} mode). The magnetic field lay in the plane of this wall and $\mathbf{H} \perp \mathbf{E}_\omega$. Thus the magnetoacoustic-type waves were excited. The experiments were performed with the field $\mathbf{H}$ oriented along all principal directions: C_1, C_2, and C_3. To avoid heating the sample we used the pulsed technique. The microwave radiation with frequency $\omega/2\pi = 9.2$ GHz was introduced in the form of rectangular pulses with a duration of 9 μs and a repetition frequency of

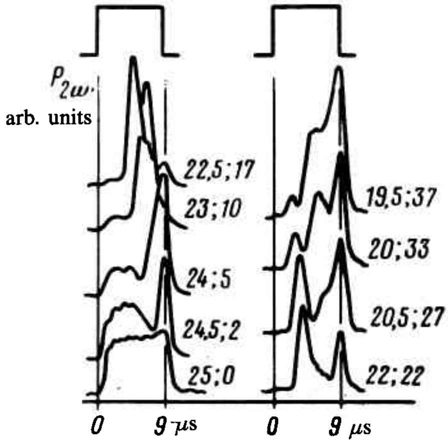

FIG. 1. Time dependence of $P_{2\omega}$ for different values of P_ω. The numbers on the curves indicate the damping in dB in the pumping and receiving channels. $P_\omega \simeq 10^3$W($H_\omega \simeq 100$Oe) corresponds to 0 dB. The Bi sample is a cube with $C_3 \| \mathbf{H} = 55.2$ kOe; $\mathbf{k}_\omega \| C_1$;$T = 4.2$K.

15 Hz. The radiation at frequencies 2ω and 3ω was detected with a heterodyne receiver; the difference frequency was 200 MHz. The shape of the harmonic pulse was studied with the help of a Boxcar-162 stroboscopic analyzer with a time resolution of 0.2 μs.

The generation of the second harmonic under the conditions described was observed previously.[4] The main result of this communication is that when the power of the incident wave attains a threshold value $P_{\rm th}$ distortions appear in the harmonic pulse and its amplitude at the maximum increases sharply. The temporal fine structure of the second-harmonic pulse depends on many factors and can assume diverse forms. We can, however, identify some general properties of its evolution, which are illustrated in Fig. 1. The distortions, which arise at the end of the pulse, move toward the beginning of the pulse as the incident power increases; the generation spikes have the shape of narrow (about 1 μs) pulses and can repeat during the time of the pumping pulse. The appearance of a generation spike of the harmonic at the end of the pumping pulse is sometimes preceded by a drop in generation (see Fig. 2). As the pumping power is increased, the fine structure of the pulse is smeared and the pulse of the second harmonic assumes a triangular shape. The ratio of the generation powers at the end and beginning of the pumping pulse is 10^2–10^3.

The amplitude of the second harmonic at the beginning of the pulse increases quadratically with the power up to fields H_ω greatly exceeding the threshold field. The structure described above and its characteristic times remained essentially unchanged when the thickness of the specimen was varied 50 fold.

The phenomenon described above is observed only in fields $H \gtrsim 10$ kOe, in contrast to the nonstationary effects in weaker fields observed in Ref. 5. An increase in the field in the interval 20–80 kOe has virtually no effect on $H_{\rm th}$ with the exception of a narrow region of fields, in which one of the last electronic Landau levels intersects the Fermi level, where $H_{\rm th}$ decreases by 4–5 dB. It is possible to ajust the power P_ω so that

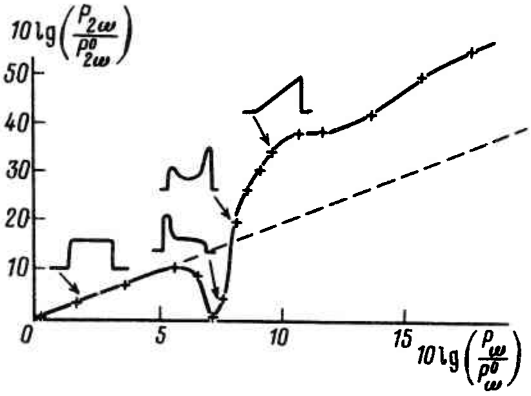

FIG. 2. Dependence of $P_{2\omega}$ on P_{ω}. The Bi sample is a cube with $C_1 \| \mathbf{H} = 25$ kOe; $\mathbf{k}_{\omega} \| C_3$; $T = 4.2$ K; $P_{\omega}^{0} \simeq 16$ W

strong generation of the second harmonic at the end of the pumping pulse will be observed only in this region (see Fig. 3).

Qualitatively analogous distortions were also observed in the pulse of the third harmonic, but these distortions had a higher threshold.

At present, we cannot propose a consistent explanation of these phenomena. Characteristically, however, at threshold powers the amplitude of the electric field, E_{ω}, always attained values for which the transverse drift velocity of the carriers in the magnetic field $v_{\rm dr} = c(E_{\omega}/H)$ was on the order of the sound velocity s. Under such conditions, a break due to the Čerenkov phonon radiation (the Esaki effect) is observed with dc current in the current-voltage characteristic. The time it takes an I-V characteristic to establish in this case is longer than or on the order of 1 μs.[6] This delay is related to the time required to establish a steady-state distribution function in the phonon system. It is possible that the phenomenon which we observed is a high-frequency variant of the Esaki effect. Since the inequalities

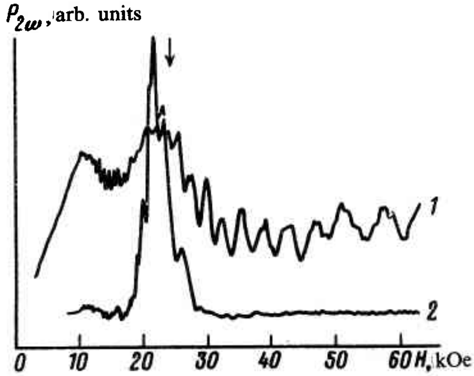

FIG. 3. Dependence of $P_{2\omega}$ on H: 1) $H_{\omega} \simeq 6$ Oe; $P_{2\omega} = 0$ dB. 2) $H_{\omega} = 9$ Oe; $P_{2\omega} = 31$ dB. The Bi sample is a cube; $C_1 \| \mathbf{H}$; $\mathbf{k}_{\omega} \| C_2$; $T = 4.2$ K.

$$\Omega \gg \omega \gg \tau^{-1}$$

(Ω is the cyclotron frequency and τ is the collision time) hold the electronic distribution function follows the instantaneous values of the field E_ω. For this reason, the Čerenkov radiation can easily be the source of the nonlinearity leading to generation of harmonics.

It has not been ruled out that the explanation of these phenomena should be sought in other processes, such as a high-order parametric instability, which is associated with the nonparabolic nature of the electronic spectrum in bismuth.[7] In any case, the model must explain in addition to the experimental characteristics indicated above, the role of the strong magnetic field and the drop in the threshold with an increase in the state density at the Fermi level (Fig. 3).

The authors thank V. E. Zakharov for useful discussions and I. A. Voropanov for growing the bismuth samples.

[1]V. S. Edel'man, Usp. Fiz. Nauk **123**, 257 (1977) [Sov. Phys. Usp. **20**, 849 (1977)].
[2]V. T. Dolgopolov, Usp. Fiz. Nauk **130**, 241 (1980) [Sov. Phys. Usp. **23**, 134 (1980)].
[3]S. A. Vitkalov, A. P. Kopasov, and F. I. Leviev, Zh. Eksp. Teor. Fiz. **85**, 2200 (1983) [Sov. Phys. JETP (to be published)].
[4]F. I. Leviev and É. G. Yashchin, Fiz. Tverd. Tela (Leningrad) **20**, 1779 (1978) [Sov. Phys. Solid State **20**, 1028 (1978)].
[5]É. G. Yashchin, Zh. Eksp. Teor. Fiz. **68**, 1127 (1975) [Sov. Phys. JETP **41**, 558 (1975)].
[6]T. Yamada, J. Phys. Soc. Jpn. **20**, 1647 (1965).
[7]Yu. A. Romanov, Izv. Vyssh. Uchebn. Zaved., Radiofiz. **23**, 617 (1980).

Translated by M. E. Alferieff
Edited by S. J. Amoretty