Second Harmonic Generation in Bismuth During the Anomalous Skin Effect

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The frequency doubling of a UHF wave reflected from bismuth is investigated between 4.2 and 300°K. At low temperatures, a plateau is observed in the generation power vs. temperature curve. The possible relation between this dependence and the anomalous nature of the skin effect is discussed.

THE response of a system of electrons becomes nonlinear in a strong electromagnetic field. This leads to a dependence of the conductivity of the system on the external field. Similar effects are easily observed in semiconductors and in a gaseous plasma. In metals and even in semimetals, it is more difficult to observe nonlinear effects because of the large conductivity, which does not permit us to create a strong electric field. In metals and semimetals, nonlinear effects are more readily noted that are connected with the action exerted on the electron system by the magnetic field of the wave that penetrates into the conductor.

Recently, a number of theoretical papers devoted to nonlinearities in metals have appeared.[1-3] Nonlinear effects have been observed experimentally, so far as we know, only in Bi. Bate and Wisseman[4] observed second harmonic generation (SHG) in the propagation of magnetoplasma waves at a frequency of 15 MHz; Yashchin and the author[5] investigated SHG in the case of the reflection of a UHF wave at room temperature in the absence of a magnetic field; Khaĭkin and Yakubovskiĭ[6] observed the onset of a constant difference in the potentials under the action of magnetoplasma waves. Some of the experimental results[4,5] can be understood if we take into account the effect of the Lorentz force on the electron. For an explanation of the effect observed in[6], such an account is insufficient. As the authors wrote, the effect is two orders of magnitude larger than follows from the estimate which takes the Lorentz force into consideration. Possibly the appearance of a dc potential difference is connected with the warming up of the electron gas, such as takes place in constant fields, where the heating leads to a deviation from Ohm's law.[7]

In the present work, we have investigated SHG in Bi in the UHF range and the temperature interval from 4.2°K to room. Here the skin effect undergoes transition from the normal effect to the limiting anomalous effect, which also determines the temperature dependence of the generation curve.

EXPERIMENTAL SETUP

We used a bimodal cylindrical resonator with a noncontact piston (Fig. 1) operating in the modes E_{010} (fundamental frequency) and H_{111} (at the frequency of the harmonic). The resonator was placed in a helium dewar and had mechanical leads which permitted us to rotate the piston and to move it independently along the axis of the resonator. A depression is made in the piston in the shape of a disk for the sample. The sample is a single crystal with a diameter of 18 mm and height 2 mm, grown in a polished quartz mold[8], with its trigonal axis directed at an angle of 45° to the surface. A thin teflon film is packed between the piston and the sample, so that there is no galvanic contact. Such a fastening of the sample is advantageous for two reasons: first, the gap between the piston and the sample allows the sample to be deformed freely upon change in temperature; second, the break in the current lines along the perimeter of the sample, which significantly deteriorates the Q of the resonator in both modes, makes it almost independent of the temperature. Temperature regulation is maintained by two heaters, one of which was lowered to the bottom of the helium dewar, and the second was attached to the resonator and is located 30 cm above the bottom. An UHF wave was generated by a magnetron with a power of the order of several kilowatts, operating in a regime of microsecond pulses. The pulse repetition frequency was chosen to be low (1–40 Hz) so that the sample was not heated on the average, as monitored directly by a thermocouple. The heating in the helium during the time of the pulse did not exceed several degrees, as revealed by the impedance jump at the time of the pulse, which was measured by E. G. Yashchin and the author.

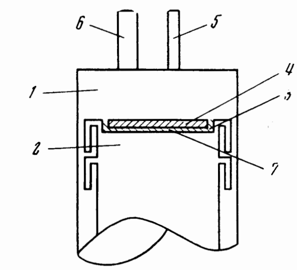

FIG. 1. Schematic diagram of apparatus. 1—resonance chamber; 2—contactless piston with mechanical leads (not shown); 3—teflon film, 4—sample; 5—11 × 5.5 waveguide; 6—23 × 10 waveguide; 7—thermocouple.

Figure 2 shows the temperature dependence of the power P_2 of second harmonic generation. When the temperature is changed from 265 to 70°K, the power increases by two orders of magnitude; the power changes very little upon further cooling. The dependence of the generated power on the incident power at 4.2°K is shown in Fig. 3, and is quadratic dependence within the limits of experimental error. Figure 4 shows the dependence of the generated power at room temperature (a) and at helium temperature (b) on the angle of rotation of the sample about the resonator

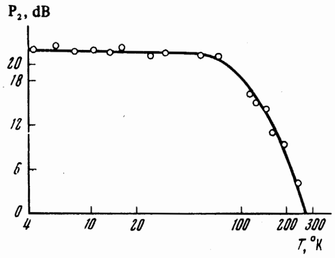

FIG. 2. Temperature dependence of the second harmonic generation.

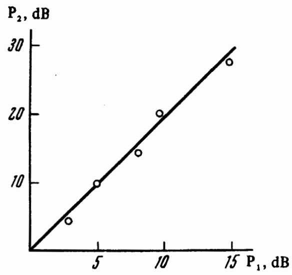

FIG. 3. Dependence of the second harmonic generated power on the incident power.

axis. The curves are functions of $\sin^2\varphi$, shifted relative to one another in phase.

DISCUSSION OF RESULTS

In order to regard the influence of the constant magnetic field on the impedance of the metal under the conditions of the normal skin effect as a perturbation, it is necessary to satisfy the inequality $r > l$, where r is the Larmor radius and l the mean free path of the electron. Under conditions of a strongly anomalous skin effect, the corresponding inequality $r > l \cdot l/\delta$ is rigorously established,[9] where δ is the skin depth. The amplitude of the high-frequency magnetic field reached several Oersteds in our experiment, i.e., it did not exceed the criterion of a weak field in the conditions of the anomalous skin effect. But, on the other hand, the criterion of weakness is insufficiently clear in relation to a high-frequency and spatially inhomogeneous field. It is quite possible that the average of the effect of the field at different points in space and at different moments of time expands the range of weak fields. Experiment shows (Fig. 3) that at all temperatures, and throughout the entire interval of fields studied, the second harmonc power P_2 is proportional to the square of the power at the fundamental frequency P_1^2. Having this fact in mind, we shall assume everywhere below that one can restrict oneself to terms of second order in the expansion of the response of the system of electrons in terms of the field.

Let an electromagnetic wave of frequency ω be incident on a metal occupying the half-space $z > 0$. The wave is normal to the surface. The amplitude of the current $j^{2\omega}$ induced in the metal at the frequency 2ω, with account of spatial dispersion, can be connected with the fields in the following way:

$$j^{2\omega}(z)=\int_0^\infty \hat{\sigma}(z-z')E^{2\omega}(z')\,dz' + \int_0^\infty\int_0^\infty \hat{\sigma}^{NL}(z-z'\ z-z'')E^{\omega}(z') \times H^{\omega}(z'')\,dz'\,dz''. \quad (1)$$

Here $E^{2\omega}$ is the amplitude of the electric field at the frequency 2ω; E^{ω} and H^{ω} are the amplitudes of the fields at the frequency ω. For simplicity, we assume

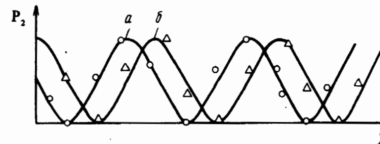

FIG. 4. Dependence of the second harmonic generated power (in arbitrary units) on the angle φ of rotation of the sample about the axis of the resonator; a—at room temperature, b—at liquid helium temperature.

that all the fields and currents are linearly polarized in the plane parallel to the surface of the metal. Writing down the kernels $\hat{\sigma}$ and $\hat{\sigma}^{NL}$ in such form presumes diffuse reflection of the electrons from the boundary. The field $E^{2\omega}$ is determined from Maxwell's equations with the current (1). Here the fields of the fundamental frequency are assumed to be given:

$$\frac{\alpha^2E^{2\omega}}{dz^2}+\frac{4\omega^2}{c^2}E^{2\omega}=-\frac{8\pi i\omega}{c^2}\int_0^\infty\hat{\sigma}E^{2\omega}\,dz'-\frac{8\pi i\omega}{c^2}\int_0^\infty\int_0^\infty\hat{\sigma}^{NL}E^{\omega}H^{\omega}\,dz'\,dz'', \quad (2)$$

c is the speed of sound in vacuum.

For a qualitative analysis of this equation, we shall make use of an approximate method developed by Bolotovskiĭ[10] in the solution of the linear problem of the anomalous skin effect. As in the linear theory, we need to know the solution of this equation only in the neighborhood of $z = 0$. The nonlocal couplings in (1) cover a region λ of the order of the mean free path of the electrons. We replace the kernels $\hat{\sigma}(z - z')$ and $\sigma^{NL}(z - z', z - z'')$ for small z by functions equal to some constants σ_0 and σ_0^{NL}, respectively, in the range from zero to λ, and equal to zero outside of this interval. Equation (2) is then written in the form (for small z)

$$\frac{d^2E^{2\omega}}{dz^2}+\frac{4\omega^2}{c^2}E^{2\omega}=-\frac{8\pi i\omega}{c^2}\sigma_0\int_0^\lambda E^{2\omega}(z')\,dz'-\frac{8\pi i\omega}{c^2}\sigma_0^{NL}\int_0^\lambda\int_0^\lambda E^{\omega}H^{\omega}\,dz'\,dz''. \quad (3)$$

This is an inhomogeneous equation and its solution is made up as usual of the general solution of the homogeneous and the particular solution of the inhomogeneous equations. The homogeneous solution has the form $E_{\text{hom}}e^{ikz}$ where the wave vector k is determined from the equation

$$k^3=(8\pi\sigma_0\omega\,/\,c^2)\,(e^{ik\lambda}-1).$$

For $k\lambda \ll 1$, this should be identical with the expression for the wave vector, which is known from the theory of the normal skin effect; we then get for σ_0 the relation $\sigma_0 = \sigma/\lambda$, where σ is the conductivity of the metal. In the case of the extremely anomalous skin effect, $k\lambda \gg 1$, and we obtain another equation for k:[10]

$$k^3=-8\pi\omega\sigma_0\,/\,c^2.$$

Let the fields at the fundamental frequency have the form $E^{\omega}(z) = E_0e^{iqz}$, $H^{\omega}(z) = H_0e^{iqz}$. We then get for the inhomogeneity in (3)

$$(8\pi i\omega\sigma_0^{NL}\,/\,c^2q^2)\,(e^{iq\lambda}-1)^2E_0H_0.$$

Hence $\sigma_0^{NL} = \alpha/\lambda^2$, where α is a quantity that determines the nonlinear current for the normal skin effect and is connected with the conductivity σ and the Hall coefficient R by the relation $\alpha = \sigma^2R$. The inhomogeneous solution of Eq. (3) has the form $E_{\text{inhom}}e^{i2qz}$, where

$$E_{\text{inhom}} = \frac{8\pi i\omega\sigma_0^{NL}\lambda^2}{c^2}\frac{H_0^\omega E_0^\omega}{4q^2(\omega)-k^2(2\omega)} \quad (4)$$

for the normal skin effect and

$$E_{\text{inhom}} = \frac{16\pi i\omega\sigma_0^{NL}}{c^2 q}\frac{H_0^\omega E_0^\omega}{k^3(2\omega)-8q^3(\omega)} \quad (5)$$

for the extremely anomalous skin effect. Matching fields on the boundary in the metal and in the vacuum,[11] we get the following for the amplitude of the wave reflected from the metal at the double frequency, $E_R^{2\omega}$:

$$E_R^{2\omega} = E_{\text{inhom}}\,[k(2\omega)-2q(\omega)]/\,k(2\omega). \quad (6)$$

We can conclude from Eqs. (4)–(6) that (a) $(E_R^{2\omega})^2 \sim \sigma R^2$ for the normal skin effect and (b) that E_R^2 does not depend on the path length in the case of the limiting anomalous skin effect. These conclusions are in excellent agreement with the temperature dependence of the generated power, which is represented in Fig. 2. Actually, the ratio of the values of σR^2 at the 80 and 273°K amounts to 70;[12] in the experiment, the ratio of the squares of the fields at the same temperatures is equal to 75. The absence of the path length in case b) for the formulas for the reflected power leads to the vanishing of the temperature dependence of the generated power at low temperatures, which is also seen from Fig. 2.

We shall now discuss briefly the results shown in Fig. 4. For this purpose, we need to consider the real geometry of the experiment. The amplitude of the oscillation at the frequency 2ω established in the resonator is proportional to the integral I:[13]

$$I = \int \mathbf{j}^{2\omega}(z)\mathbf{E}^{2\omega}(z)\,dV.$$

Here the integration is carried out over the volume of the metal, $\mathbf{E}^{2\omega}$ is the field of the H_{111} mode in the metal. We can establish (for example, by means of the reciprocity theorem) that this formula is valid even in the case of the anomalous skin effect. We expand the field and the current in the metal in Fourier series (here we shall assume that the fields are equal to zero outside the metal):

$$\mathbf{E}^\omega(z) = \mathbf{E}_0^\omega \int e^{ikz}e^\omega(k)\,dk, \quad H^\omega(z) = H_0^\omega \int e^{ikz}h^\omega(k)\,dk, \quad (7)$$

$$\mathbf{E}^{2\omega}(z) = \mathbf{E}_0^{2\omega} \int e^{ikz}\,e^{2\omega}(k)\,dk,$$

$$j_i^{2\omega}(z) = \int \alpha_{ijk}(k_1k_2)E_j^\omega(k_1)H_k^\omega(k_2)\,e^{i(k_1+k_2)z}\,dk_1\,dk_2.$$

We then obtain for the integral I

$$I \sim \int \alpha_{ijk}(k_1k_2)\,e^\omega(k_1)\,h^\omega(k_2)\,e^{2\omega}(k_1+k_2)\,dk_1\,dk_2 \int E_{0j}^\omega H_{0k}^\omega E_{0i}^{2\omega}\,ds.$$

Here ds is the surface element of the sample. If we substitute the fields of the oscillations E_{010} and H_{111} in this expression, we can then establish the fact that the dependence of the generated power on the angle of rotation of the sample about the resonator axis always has the form $\sin^2(\varphi + \Delta)$. The phase Δ is determined in the general case by the form of the impedance tensor and the tensor α_{ijk}. In our experiment, the phase difference at room and liquid helium temperatures was apparently determined by the additional anisotropic impedance that arises in the anomalous skin effect. The fact is that in the integration over k_1 and k_2 the principal contribution to Eq. (7) at room and liquid helium temperatures is made by the region of wave vectors from zero to l^{-1}, since the components of the tensor α_{ijk} behave like k^{-3} when $kl \gg 1$[14] (we note that the last formula of[14] is incorrect). Therefore, the form of the tensor α_{ijk} varies little on going from room temperature to that of liquid helium.

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