

SECOND HARMONIC GENERATION IN PIEZOELECTRIC SEMICONDUCTORS

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Abstract—*The second harmonic generations in piezoelectric semiconductors are investigated by assuming a parabolic band structure. The second harmonic generation can be found to be expressed in terms of the fundamental. It is shown that the amplitude of second harmonic generation is proportional to the quadratic of the fundamental and depends upon the sound frequency for parabolic band structure in n-type InSb. Therefore the intensity of the second harmonic generation is proportional to the quadratic of the intensity of the fundamental.*

I. INTRODUCTION

When waves with the sound frequency ω traverse a nonlinear crystal, some new waves will be produced with twice the original frequency ω . This is called the second harmonic generation of waves. The nonlinear properties of solids have been used to generate second harmonics in the microwave region.¹⁻⁸ The second harmonic generation of light can be produced by scattering, reflection, and refraction in solids.⁴⁻⁸ In this paper we are going to study the second harmonic generation of ultrasound in piezoelectric semiconductors. Due to the nonlinearities in the electron-phonon interaction responsible for ultrasonic amplification in photoconductive cadmium sulfide, the ultrasonic second harmonic generation in this solid can be observed.² The physical situation is that the self-consistent field produced by the interaction of the conduction electrons with the travelling ultrasonic wave contains the second harmonic. Therefore, the interaction of the conduction electrons with the travelling ultrasonic wave can be used to study second harmonic generation. The quantum treatment of optical second harmonic generation has been investigated by several authors.⁴⁻⁸ In this paper we investigate second harmonic generation in piezoelectric semiconductors using a quantum treatment which is valid for high frequencies and also for strong magnetic fields. For obtaining the linear and nonlinear conductivity tensors, the current density can be expressed in terms of the linear and nonlinear self-consistent fields.^{3,9,10} It is assumed that the semiconductor is nondegenerate and the effect of electron scattering on the electron-phonon interaction is neglected. We also limit ourselves to the case where the ultrasound propagates parallel to a *dc* magnetic field. It is shown that the amplitude of the

second harmonic can be expressed in terms of the fundamental using the linear and nonlinear conductivity tensors.

In Section II of this paper, the equation of motion in piezoelectric semiconductors is developed. In Section III, we use the coordinate transformation to simplify the equation of motion and expressed the amplitude of second harmonic generation in a piezoelectric semiconductor in terms of the linear and nonlinear conductivities. We give a numerical calculation for *n*-type InSb in Section IV and give a brief discussion about our results.

II. DEVELOPMENT OF THE EQUATION OF MOTION FOR THE ELECTRON-PHONON INTERACTION BETWEEN CONDUCTION ELECTRONS AND THE ULTRASOUND

The nonlinear polarizations or current densities can be expressed in terms of the fields. These quantities serve in turn as sources for the fields. Nonlinear effects in solids can be investigated from the nonlinear current density and Maxwell's equations.^{3-5,11,12} It is assumed here that the conductivity tensors are important factors in second harmonic generation for the interaction between conduction electrons and ultrasound in piezoelectric semiconductors. These conductivity tensors depend upon the ultrasound wave vector \vec{q} and frequency ω . However, the dielectric constant is considered as a scalar quantity in our present work.

The propagation of ultrasound in a piezoelectric semiconductor produces an electromagnetic wave and vice versa. The basic equation of motion for an elastic continuum is^{13,14}

$$\rho \frac{d^2 \xi_i}{dt^2} = \frac{\partial T_{ij}}{\partial x_j}, \quad (1)$$

where ρ is the density of the material, $\vec{\xi} = \vec{\xi}_0 \exp i(\vec{q} \cdot \vec{r} - \omega t)$ is the displacement, and T_{ij} is the stress tensor. When an ultrasonic wave interacts with electrons via piezoelectric coupling, the stress-strain relation contains an additional stress term by the induced electric field from the applied stress and is given by¹³

$$T_{ij} = C_{ijkl} S_{kl} - \beta_{ijk} E_k, \quad (2)$$

where C_{ijkl} are the elastic constants, β_{ijk} are the piezoelectric constants, and S_{ij} is the strain tensor which is defined by the relation

$$S_{ij} = \frac{1}{2} \left(\frac{\partial \xi_i}{\partial x_j} + \frac{\partial \xi_j}{\partial x_i} \right). \quad (3)$$

In a piezoelectric material, the polarization induced by applying a strain can be expressed by¹³

$$D_i = \epsilon_{ij} E_j + 4\pi \beta_{ijk} S_{jk}, \quad (4)$$

where D_i is the electric displacement, and ϵ_{ij} are the components of the dielectric tensor. Since the off-diagonal components of ϵ_{ij} are zero except for triclinic and monoclinic crystal structures,¹⁵ we take ϵ_{ij} to have only diagonal components in

our problem. ϵ_{ij} in this expression arises solely from the lattice contribution to the dielectric tensor, hence it is a scalar quantity.

Maxwell's equations are

$$\nabla \cdot \vec{D} = 4\pi\rho_e, \tag{5}$$

$$\nabla \cdot \vec{B} = 0, \tag{6}$$

$$\nabla \times \vec{E} = -\frac{1}{c} \frac{\partial \vec{B}}{\partial t}, \tag{7}$$

$$\nabla \times \vec{H} = \frac{4\pi}{c} \vec{J} + \frac{1}{c} \frac{\partial \vec{D}}{\partial t}. \tag{8}$$

Furthermore, the modified Ohm's law is^{16,17}

$$J_{1i} = \sigma_{ij}(\vec{q}, \omega) E_{1j} + \tau_{ijk}(\vec{q}, \omega) E_{1j} E_{1k}, \tag{9}$$

where σ_{ij} is the linear conductivity tensor and τ_{ijk} is the non-linear conductivity tensor. The longitudinal component of the linear conductivity tensor plays the dominant role in determining the dispersion and absorption of ultrasonic waves in semiconductors.^{18,19} For second harmonic generation, the longitudinal components of the linear and nonlinear conductivity tensors will play the important factors.²⁰ In a nonmagnetic material, \vec{B} and \vec{H} are related by

$$\vec{B} = \vec{H}. \tag{10}$$

We neglect the nonlinear terms which are higher than the second order in the fields, hence the total current density \vec{J} can be expressed by

$$J_i = J_{1i} + J_{2i} \approx \sigma_{ij}(\vec{q}, \omega) E_{1j} + \sigma_{ij}(2\vec{q}, 2\omega) E_{2j} + \tau_{ijk}(\vec{q}, \omega) E_{1j} E_{1k}. \tag{11}$$

It is assumed that the plane wave solutions for the electromagnetic field and the displacement are of the following forms

$$\vec{E} = \vec{E}_1 + \vec{E}_2 = \vec{E}_{10} \exp i(\vec{q} \cdot \vec{r} - \omega t) + \vec{E}_{20} \exp 2i(\vec{q} \cdot \vec{r} - \omega t), \tag{12}$$

$$\vec{\xi} = \vec{\xi}_1 + \vec{\xi}_2 = \vec{\xi}_{10} \exp i(\vec{q} \cdot \vec{r} - \omega t) + \vec{\xi}_{20} \exp 2i(\vec{q} \cdot \vec{r} - \omega t), \tag{13}$$

where the index 1 indicates the fundamental, and the index 2 indicates the second harmonic. From Eqs. (1) and (2), we have the following relation,

$$\rho \frac{d^2 \xi_i}{dt^2} = C_{ijkl} S_{kl,j} - \beta_{ij,k} E_{k,j}. \tag{14}$$

Using Eqs. (3), (12), (13) together with Eq. (14), we find that

$$\begin{aligned} \omega^2 \rho (\xi_{1i} + 4\xi_{2i}) &= \frac{C_{ijkl}}{2} (q_j q_l \xi_{1k} + q_j q_k \xi_{1l} + 4q_j q_l \xi_{2k} + 4q_j q_k \xi_{2l}) \\ &\quad + i q_j \beta_{ijkl} (E_{1k} + E_{2k}). \end{aligned} \tag{15}$$

From Eqs. (7), (8), and (10), we find the wave equation of the electromagnetic field,

$$\nabla^2 \vec{E} - \nabla(\nabla \cdot \vec{E}) = \frac{4\pi}{c^2} \frac{\partial \vec{J}}{\partial t} + \frac{1}{c^2} \frac{\partial^2 \vec{D}}{\partial t^2}. \tag{16}$$

Combining Eqs. (4), (11), (12), (13), and (16), it follows that

$$\begin{aligned}
 & -q^2(E_{1i} + 4E_{2i}) + q_i(\vec{q} \cdot \vec{E}_1) + 4q_i(\vec{q} \cdot \vec{E}_2) \\
 & = \frac{4\pi}{c^2} [-i\omega\sigma_{ij}(\vec{q}, \omega)E_{1j} - 2i\omega\sigma_{ij}(2\vec{q}, 2\omega)E_{2j} - 2i\omega\tau_{ijk}(\vec{q}, \omega) \\
 & \cdot E_{1j}E_{1k}] + \frac{1}{c^2} [-\omega^2\varepsilon_{ij}(\vec{q}, \omega) - 4\omega^2\varepsilon_{ij}(2\vec{q}, 2\omega)E_{2j} - 2\pi i\omega^2\beta_{ijk} \\
 & \cdot (\xi_{1j}q_k + \xi_{1k}q_j + 8\xi_{2j}q_k + 8\xi_{2k}q_j)]. \tag{17}
 \end{aligned}$$

Eqs. (15) and (17) can be rewritten as the following equations,¹⁷

$$\omega^2\rho\xi_{1io} = \frac{C_{ijkl}}{2}(q_jq_l\xi_{1ko} + q_jq_k\xi_{1lo}) + i\beta_{ijk}q_jE_{1ko}, \tag{18}$$

$$\omega^2\rho\xi_{2io} = \frac{C_{ijkl}}{2}(q_jq_l\xi_{2ko} + q_jq_k\xi_{2lo}) + \frac{i}{2}\beta_{ijk}q_jE_{2ko}, \tag{19}$$

$$\begin{aligned}
 q^2E_{1io} - q_i(\vec{q} \cdot \vec{E}_{1o}) & = \frac{4\pi i\omega}{c^2}\sigma_{ij}(\vec{q}, \omega)E_{1jo} + \frac{\omega^2}{c^2}\varepsilon_{ij}(\vec{q}, \omega)E_{1jo} \\
 & + \frac{2\pi i\omega^2\beta_{ijk}}{c^2}(\xi_{1jo}q_k + \xi_{1ko}q_j), \tag{20}
 \end{aligned}$$

$$\begin{aligned}
 q^2E_{2io} - q_i(\vec{q} \cdot \vec{E}_{2o}) & = \frac{2\pi i\omega}{c^2}\sigma_{ij}(2\vec{q}, 2\omega)E_{2jo} + \frac{2\pi i\omega}{c^2}\tau_{ijk}(\vec{q}, \omega)E_{1jo}E_{1ko} \\
 & + \frac{\omega^2}{c^2}\varepsilon_{ij}(2\vec{q}, 2\omega)E_{2jo} + \frac{4\pi i\omega^2\beta_{ijk}}{c^2}(\xi_{2jo}q_k + \xi_{2ko}q_j). \tag{21}
 \end{aligned}$$

III. COORDINATE TRANSFORMATION AND THE SOLUTION OF EQUATIONS

We are going to discuss our problem by considering two special cases:

- Case 1. A Cartesian frame of reference with the x axis along the [001] direction and the z axis along the [110] direction (see Fig. 1);
- Case 2. A Cartesian frame of reference with the y axis along the (110) direction and the z axis along the [111] direction (see Fig. 2).

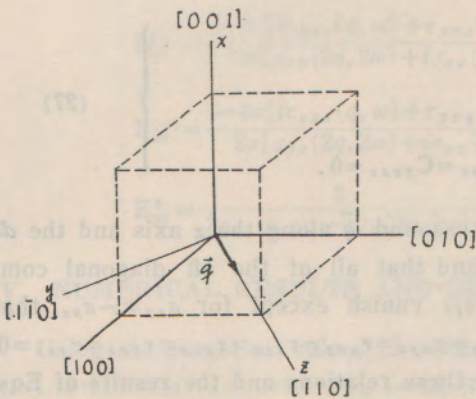


Fig. 1. Frame of reference for ultrasonic waves with wave vector \vec{q} in the [110] direction and the x axis in the [001] direction.

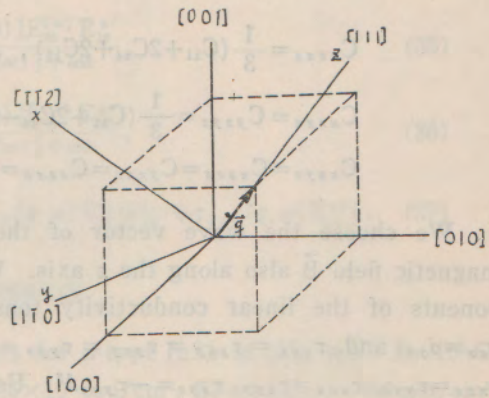


Fig. 2. Frame of reference for ultrasonic waves with wave vector \vec{q} in the [111] direction and the y axis in the [110] direction.

Then the piezoelectric constants β_{ijk} and the elastic constants C_{ijkl} follow the coordinate transformations

$$\beta_{ijk} = a_{i\mu} a_{j\nu} a_{k\lambda} \beta_{\mu\nu\lambda}, \tag{22}$$

and

$$C_{ijkl} = a_{i\mu} a_{j\nu} a_{k\lambda} a_{l\varepsilon} C_{\mu\nu\lambda\varepsilon}, \tag{23}$$

where i, j, k, l are the indices in the x - y - z system, $\mu, \nu, \lambda, \varepsilon$ are the indices in the 1-2-3 system, and the a 's are the direction cosines. After the coordinate transformations, we obtain the following results:

Case 1. For the wave vector of the ultrasound $\vec{q} \parallel [110]$ and using coordinates (1),

$$\left. \begin{aligned} \beta_{zzz} = \beta_{zzx} = \beta_{zxx} = \beta_{123} = \beta_{14}, \\ \beta_{xxx} = \beta_{xxy} = \beta_{yxx} = \beta_{yyz} = \beta_{yzz} = \beta_{zzx} = \beta_{zzy} = \beta_{xzx} = \beta_{yzz} \\ = \beta_{yyz} = \beta_{zyz} = \beta_{xyx} = 0. \end{aligned} \right\} \tag{24}$$

$$\left. \begin{aligned} C_{zzzz} = C_{44}, \\ C_{yyyz} = \frac{1}{2}(C_{11} + C_{44} - C_{12}), \\ C_{zzzz} = \frac{1}{2}(C_{11} + C_{44} + C_{12}), \\ C_{xzyz} = C_{xzzz} = C_{yzzz} = C_{zzxz} = C_{zzyz} = C_{yzzz} = 0. \end{aligned} \right\} \tag{25}$$

Case 2. For the wave vector of the ultrasound $\vec{q} \parallel [111]$ and using coordinates (2),

$$\left. \begin{aligned} \beta_{zzz} = \frac{2}{\sqrt{3}} \beta_{123} = \frac{2}{\sqrt{3}} \beta_{14}, \\ \beta_{xxx} = \beta_{xzz} = \beta_{yyz} = \beta_{yzy} = -\frac{1}{\sqrt{3}} \beta_{14}, \\ \beta_{xzy} = \beta_{xzz} = \beta_{yxx} = \beta_{yzz} = \beta_{zxx} = \beta_{zzy} = \beta_{xyx} = \beta_{yxz} = \beta_{zyz} \\ = \beta_{zxx} = 0. \end{aligned} \right\} \tag{26}$$

$$\left. \begin{aligned} C_{zzzz} = \frac{1}{3}(C_{11} + 2C_{44} + 2C_{12}), \\ C_{xzzz} = C_{yzyz} = \frac{1}{3}(C_{11} + 2C_{44} - C_{12}), \\ C_{xzyz} = C_{xzzz} = C_{yzzz} = C_{zzxz} = C_{zzyz} = C_{yzzz} = 0. \end{aligned} \right\} \tag{27}$$

We choose the wave vector of the ultrasound \vec{q} along the z axis and the $d\vec{c}$ magnetic field \vec{B} also along the z axis. We find that all of the off diagonal components of the linear conductivity tensor σ_{ij} vanish except for $\sigma_{xy} = -\sigma_{yx}$, that $\sigma_{xx} = \sigma_{yy}$ and $\tau_{xxx} = \tau_{xxy} = \tau_{yyx} = \tau_{yyy} = \tau_{zzz} = \tau_{zyz} = \tau_{xyy} = \tau_{xzz} = \tau_{yxx} = \tau_{yzz} = \tau_{zxy} = 0$, $\tau_{xzz} = \tau_{yyz}$, $\tau_{xxx} = \tau_{zyy}$, $\tau_{xyz} = -\tau_{yxz}$.¹⁷ Using these relations and the results of Eqs. (24), (25), (26), and (27), then the solutions of Eqs. (18)-(21) are given as follows:

Case 1. $\vec{q} \parallel [110]$

We define $A^+ = \sqrt{A_x^2 + A_y^2}$, $A^- = A_z$, then the solution is

$$\xi_{20}^{\pm} = \frac{2\pi i q \beta_{14}}{G(q, \omega) [\sigma_{zz}(2q, 2\omega) - 2\sigma_{zz}(q, \omega)]} \left[\left(\frac{\omega^4 \beta_{14}^2}{c^4 q^2} \right) \tau_{zzz}(q, \omega) \xi_{10}^{\pm 2} + \frac{G^2(q, \omega)}{q^2 \beta_{14}^2} \tau_{zzz}(q, \omega) \xi_{10}^{\pm 2} \right], \quad (28)$$

$$\xi_{20}^{\parallel} = -\frac{8\pi^2 G(q, \omega) \tau_{zzz}(q, \omega)}{q \beta_{14} [2\pi i \sigma_{xx}(2q, 2\omega) + \omega \varepsilon]} \xi_{10}^{\pm} \xi_{10}^{\parallel}, \quad (29)$$

with the condition $q^2 \gg \left| \frac{4\pi i \omega \sigma_{ij}}{c^2} \right|$.

Here,

$$G(q, \omega) = \frac{q^2 \beta_{14}^2}{\frac{4\pi i \sigma_{zz}(q, \omega)}{\omega} + \varepsilon} = \frac{1}{4\pi} (\omega^2 \rho - C_{44} q^2).$$

$$E_{20}^{\pm} = -\frac{4\pi i \tau_{zzz}(q, \omega)}{2\pi i \sigma_{xx}(2q, 2\omega) + \omega \varepsilon} E_{10}^{\pm} E_{10}^{\parallel}, \quad (30)$$

$$E_{20}^{\parallel} = \frac{1}{2\sigma_{zz}(q, \omega) - \sigma_{zz}(2q, 2\omega)} [\tau_{zzz}(q, \omega) E_{10}^{\pm 2} + \tau_{zzz}(q, \omega) E_{10}^{\parallel 2}]. \quad (31)$$

Case 2. $\vec{q} \parallel [111]$

The transverse waves are defined in the circularly polarized form, $A^{(\pm)} = A_x \pm iA_y$, $A^{\parallel} = A_z$, then we have

$$\xi_{20}^{(+)} = \frac{16\pi^2 i \omega q \beta_{14} [i\tau_{zzz}(q, \omega) + \tau_{yyz}(q, \omega)] \xi_{10}^{(+)} \xi_{10}^{\parallel}}{\sqrt{3} [4\pi i \sigma_{zz}(q, \omega) + \omega \varepsilon] \{2\pi [\sigma_{xy}(2q, 2\omega) + i\sigma_{xx}(2q, 2\omega)] + \omega \varepsilon\}}, \quad (32)$$

$$\xi_{20}^{(-)} = \frac{16\pi^2 i \omega q \beta_{14} [i\tau_{zzz}(q, \omega) + \tau_{yyz}(q, \omega)] \xi_{10}^{(-)} \xi_{10}^{\parallel}}{\sqrt{3} [4\pi i \sigma_{zz}(q, \omega) + \omega \varepsilon] \{2\pi [\sigma_{yx}(2q, 2\omega) + i\sigma_{xx}(2q, 2\omega)] + \omega \varepsilon\}}, \quad (33)$$

$$\xi_{20}^{\parallel} = \frac{4\pi^2 i \sigma_{zz}(q, \omega) + \pi \omega \varepsilon}{\sqrt{3} i q [2\sigma_{zz}(q, \omega) - \sigma_{zz}(2q, 2\omega)]} \left\{ \left(\frac{\omega^3 \beta_{14}}{c^4 q^2} \right) \tau_{zzz}(q, \omega) \xi_{10}^{(+)} \xi_{10}^{(-)} + \frac{4q^2 \omega \beta_{14} \tau_{zzz}(q, \omega)}{[4\pi i \sigma_{zz}(q, \omega) + \omega \varepsilon]^2} \xi_{10}^{\parallel 2} \right\}; \quad (34)$$

$$E_{20}^{(+)} = \frac{-4\pi [i\tau_{zzz}(q, \omega) + \tau_{yyz}(q, \omega)] E_{10}^{(+)} E_{10}^{\parallel}}{2\pi [\sigma_{xy}(2q, 2\omega) + i\sigma_{xx}(2q, 2\omega)] + \omega \varepsilon}, \quad (35)$$

$$E_{20}^{(-)} = \frac{-4\pi [i\tau_{zzz}(q, \omega) + \tau_{yyz}(q, \omega)] E_{10}^{(-)} E_{10}^{\parallel}}{2\pi [\sigma_{yx}(2q, 2\omega) + i\sigma_{xx}(2q, 2\omega)] + \omega \varepsilon}, \quad (36)$$

$$E_{20}^{\parallel} = \frac{1}{2\sigma_{zz}(q, \omega) - \sigma_{zz}(2q, 2\omega)} [\tau_{zzz}(q, \omega) E_{10}^{(+)} E_{10}^{(-)} + \tau_{zzz}(q, \omega) E_{10}^{\parallel 2}]. \quad (37)$$

IV. NUMERICAL RESULTS AND DISCUSSION

Using the numerical values of parameters for *n*-type InSb at 10°K with $B=10^5$ G, $n_0=1.75 \times 10^{14}$ cm⁻³, $m^*=0.013$ m, $\varepsilon=18$, $\beta_{14}=1.8 \times 10^4$ esu/cm², $E_g=0.2$ eV, $\rho=5.8$ gm/cm³, $v_s=4 \times 10^5$ cm/sec, $q=2.5 \times 10^5$ cm⁻¹, we have the amplitude of second harmonic generation for parabolic band structure as a function of sound frequency ω shown in Fig. 3 for $\vec{q} \parallel [110]$ and Fig. 4 for $\vec{q} \parallel [111]$. The acoustic intensity P_n for the *n*th harmonic generation is defined by²

$$P_n = \frac{1}{2} \left| \frac{\partial \vec{\xi}_{n0}}{\partial t} \right|^2 v_s = \frac{1}{2} \rho \omega^2 n^2 v_s |\xi_{n0}|^2. \quad (38)$$

If we suppose the longitudinal and transverse components of the fundamental are of the same order of magnitude (but not equal to zero), then we have the numerical

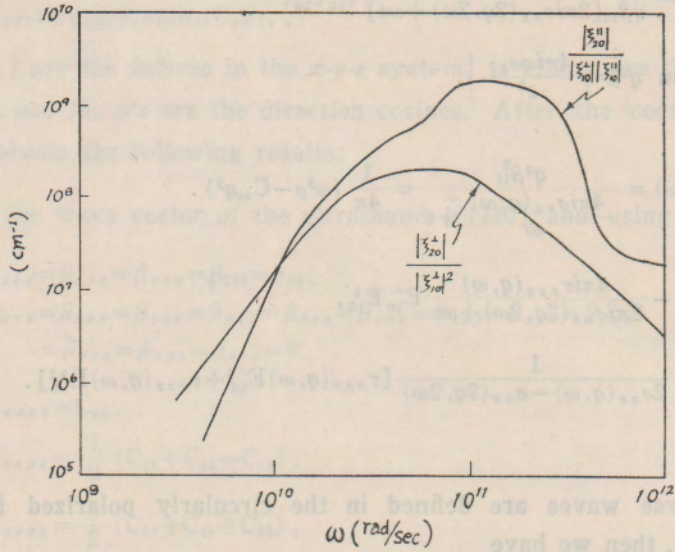


Fig. 3. The amplitude of second harmonic generation in terms of the quadratic of the amplitude of fundamental for parabolic band structure in *n*-type InSb at T=10° K, B=10⁸ G, and $\vec{q}||[110]$.

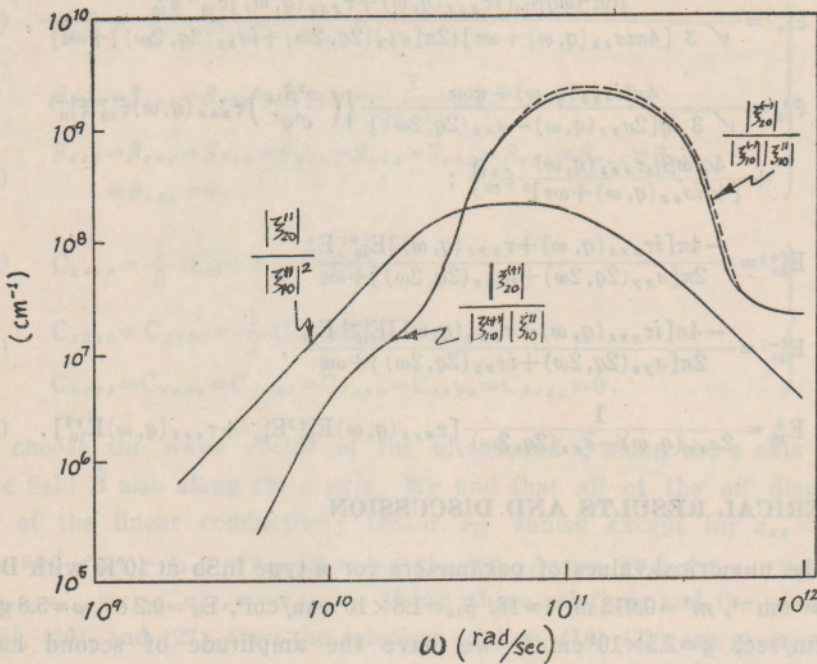


Fig. 4. The amplitude of second harmonic generation in terms of the quadratic of the amplitude of fundamental for parabolic band structure in *n*-type InSb at T=10° K, B=10⁸ G, and $\vec{q}||[111]$.

results for the acoustic intensity of second harmonic generation as a function of sound frequency ω shown in Fig. 5 for $\vec{q}||[110]$ and Fig. 6 for $\vec{q}||[111]$. From these results, it can be seen that the second harmonic generation depends strongly on the sound frequency ω for parabolic band structure. It is also shown that the maximum value of the second harmonic generation for parabolic band structure in *n*-type InSb is in the neighborhood of the sound frequency $\omega=10^{11}$ rad/sec. Since the longitudinal

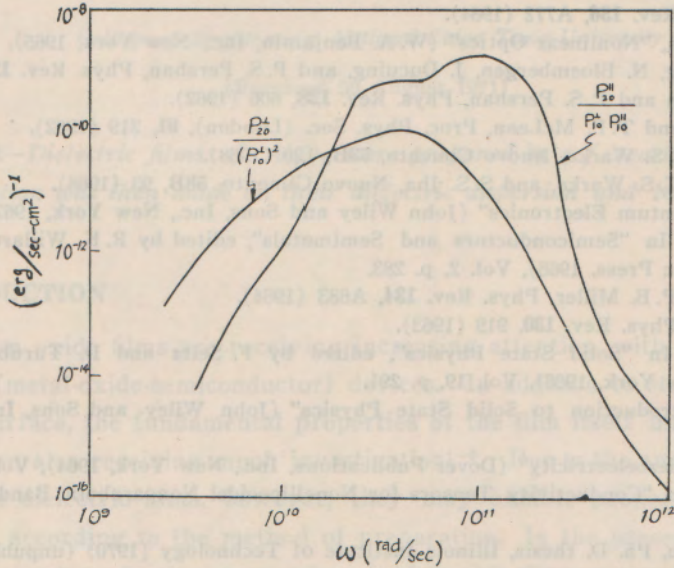


Fig. 5. The acoustic intensity of second harmonic generation in terms of the quadratic of the fundamental for parabolic band structure in *n*-type InSb at $T=10^5$ K, $B=10^5$ G and $\vec{q}||[110]$.

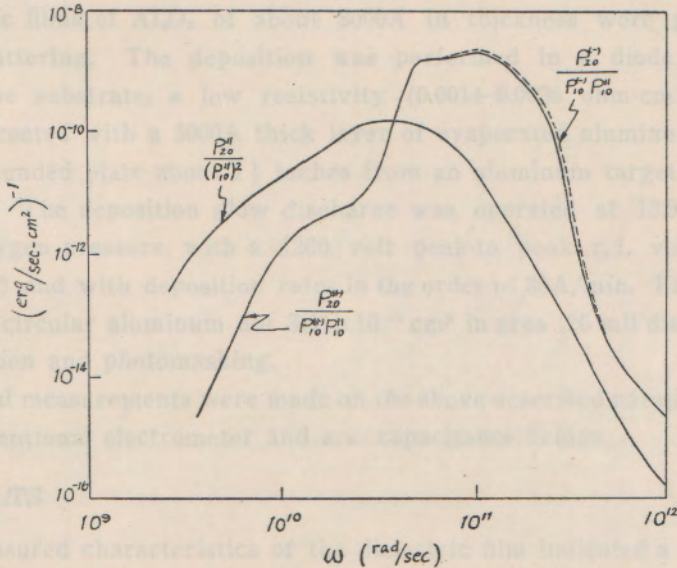


Fig. 6. The acoustic intensity of second harmonic generation in terms of the quadratic of the fundamental for parabolic band structure in *n*-type InSb at $T=10^5$ K, $B=10^5$ G and $\vec{q}||[111]$.

conductivity tensors σ_{zz} and τ_{zzz} are independent of the dc magnetic field for parabolic band structure,²⁰ therefore the second harmonic generation depends very weak on the magnetic field. For the case of the nonparabolic band structure, the second harmonic generation depends strongly on the dc magnetic field.²⁰

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