

Model of thermal excitation in the Ge crystal

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A model is constructed for the thermal excitation arising as a result of the cooling of the photoexcited charge carriers in a germanium crystal cooled to liquid-helium temperature. If the power of the laser pulses is sufficiently high, the thermal excitation region will be a hot spot with a temperature higher than the Debye temperature of the crystal. As a result of the cooling, the expansion of the hot spot due to heat conduction gives way to a stage governed by the propagation of nondecaying t phonons. The spatially expanding thermal excitation region is a source of ballistic phonons. The duration of the ballistic signal, which is extracted from the total detected phonon signal by a method based on the focusing effect for ballistic phonons, is calculated as a function of the energy of the light pulse.

INTRODUCTION

The propagation of phonons excited as a result of the cooling of the photoexcited plasma of a semiconductor is a topic of current research interest. A crystalline sample, cooled by liquid helium to low temperatures, is subjected to pulses of laser light. The phonons which propagate from the excitation region are detected by a bolometer. The duration and character of the detector signal depend on the power of the laser pulse. If the power of the laser pulses is sufficiently high, the duration of the detector signal will be substantially greater than that of the light pulses, and in this case the narrow peak characteristics of ballistic phonons is accompanied by a long tail ($\sim 10 \mu\text{sec}$) which corresponds to the arrival at the detector of phonons scattered en route in the interior of the sample.^{1,2}

A method exists for extracting the ballistic phonon signal from the total detector signal. This is the so-called spatial filtering method, which is based on the focusing effect for ballistic phonons.³ An experiment performed on a highly pure Ge crystal with the aid of the spatial filtering method showed that the duration of the ballistic phonon signal increased from 0.1 to 1.5 μsec with increasing power for laser pulses 0.1 μsec long.⁴ Such long-lived phonon pulses are explained by the presence in the excitation region of a hot spot which operates for a certain time after the laser pulse has ended and acts as a source of ballistic phonons.

The hot spot is a region in which most of the phonons are concentrated, with an energy density sufficient for the establishment of a Planckian equilibrium at a temperature higher than the temperature of the crystal. A criterion for the establishment of such an equilibrium is that the mean free path of the thermal phonons in respect to the phonon-phonon interaction be small compared to the size of the hot spot. In this case processes involving the coalescence of thermal phonons occur as efficiently as decay processes.

If the energy of the laser pump exceeds a certain value E_{\ominus} , the phonon temperature established in the hot spot at the end of the laser pulse will be higher than the Debye temperature of the crystal. Then the rate of anharmonic interactions of thermal phonons in a pure crystal will be higher than the rate of their scattering by defects during the entire evolu-

tion of the hot spot. The spatial expansion of the spot will be determined by the thermal conductivity of the crystal. However, as the spot expands, the density of heat energy falls off, and a stage commences in which the existence criterion for temperature is not fulfilled. The spontaneous decay of phonons becomes the only anharmonic process. The decay of phonons of the decaying modes will be dominant over isotopic scattering, and the phonons will accumulate on the non-decaying branches of the phonon spectrum. The expansion of such a phononic inhomogeneity was described in Refs. 5 and 6.

If the energy of the laser pump is below E_{\ominus} , the opposite situation obtains during the formation and evolution of the hot spot: at the actual phonon frequencies, scattering by defects is dominant over the anharmonic interaction. This case was considered in Ref. 7. The spatial expansion of the hot spot is governed by nonlocal heat conduction (the various regimes of nonlocal heat conduction were considered in Ref. 8). Then the Planck distribution breaks down, and the nonlocal heat conduction gives way to quasidiffusion. Quasidiffusion is described in Refs. 9 and 10.

In the present paper we construct a model for the development of the hot spot whereby the latter gives way to an expansion of nondecaying phonons. For comparison with the experimental results of Ref. 4, we calculate the excitation-energy dependence of the duration of the ballistic phonon pulse, which is extracted from the detector signal by the method of spatial filtering.

1. THE HOT SPOT

Laser irradiation of the surface of a semiconductor gives rise to an electron-hole plasma which, on recombining and cooling, creates a thermal excitation. If the phonon energy density in the thermal excitation region is sufficiently high, the phonon-phonon interactions rapidly bring the phonons into a Planckian equilibrium at a temperature higher than the temperature of the crystal, which is cooled by liquid helium.

We shall take the initial time to be at the end of the laser pulse. It is assumed that the hot spot has formed prior to this time and is in the shape of a cylinder whose base is the cross

section of the laser beam on the surface of the crystal. The radius of the base is much greater than the height of the cylinder, $r_0 \gg d_0$.

In calculating the energy of the spot we assume that the phonon temperature is the same throughout the volume of the cylinder. We also assume that the height d_0 of the cylinder is independent of the pump energy E (for $E < E_\ominus$ the dependence of d_0 on E can be substantial). Under these assumptions the initial energy of the spot is given by

$$E_0 = \pi r_0^2 d_0 C(T_0) T_0, \quad (1)$$

where T_0 is the initial temperature of the hot spot and C is the specific heat of the crystal.

The energy of the light pulse is related to the initial energy of the spot as $E = E_0/0.6$ (Ref. 4). We shall consider energies $E > E_\ominus$, where

$$E_\ominus = \pi r_0^2 d_0 C_0 \Theta / 0.6 = 3.96 \mu\text{J},$$

which for Ge corresponds to $T_0 > \Theta$ for $r_0 = 30 \mu\text{m}$, $d_0 = 1 \mu\text{m}$ (Ref. 4), Debye temperature $\Theta/k = 374 \text{ K}$ (k is Boltzmann's constant), and $C_0 = 1.63 \cdot 10^{23} \text{ cm}^{-3}$.

We approximate the specific heat by

$$C(T) = \begin{cases} C_0, & T \geq T_c, \\ C_0(T/T_c), & T \leq T_c, \end{cases} \quad (2)$$

where T_c is determined by joining the expressions for the specific heat for $T \ll \Theta$ and $T > \Theta$. From the expression

$$2\pi^2 T_c^3 / 5 (\hbar v)^3 = C_0,$$

where $v = 3.5 \cdot 10^5 \text{ cm/sec}$ is the average sound velocity in Ge, we get

$$T_c = (5C_0/2\pi^2)^{1/3} \hbar v. \quad (3)$$

For further study it is necessary to determine the phonon-phonon collision frequency for subthermal phonons. By continuing the known expressions for the frequency of collisions between transverse (t) phonons at $T \ll \Theta$ and $T > \Theta$ (Ref. 11) to the temperature at which the two expressions for $1/\tau_t(\omega)$ join, we get

$$\frac{1}{\tau_t(\omega)} = \begin{cases} \frac{1}{\tau_t'(T)} \frac{\hbar\omega}{\Theta}, & T \geq T_c', \\ \frac{1}{\tau_t(T)} \frac{\hbar\omega}{T}, & T \leq T_c', \end{cases} \quad (4)$$

where

$$T_c' = \Theta (5/4\pi^4)^{1/3}.$$

Analogously, for the longitudinal (l) phonons in the Ge crystal, which has cubic symmetry, we get

$$\frac{1}{\tau_l(\omega)} = \begin{cases} \frac{1}{\tau_l'(T)} \left(\frac{\hbar\omega}{\Theta}\right)^2, & T \geq T_c', \\ \frac{1}{\tau_l(T)} \left(\frac{\hbar\omega}{T}\right)^2, & T \leq T_c', \end{cases} \quad (5)$$

where

$$T_c' = \Theta [12\zeta(3)]^{-1/2} (v_l/v)^{3/2},$$

$v_l = 3.25 \cdot 10^5 \text{ cm/sec}$ is the average velocity of transverse sound, and ζ is the Riemann zeta function. Expressions for

$1/\tau_{t,l}' \propto \Theta^4 T$ and $1/\tau_{t,l} \propto T^5$ can be found in Ref. 11. The temperatures obey

$$T_c/k \approx T_c'/k \approx T_c''/k \approx 88 \text{ K}.$$

We can thus distinguish two stages in the development of the hot spot: the decrease of the temperature of the spot from T_0 to T_c , and then the decrease of the temperature from T_c to T_K , where T_K is the temperature at the time when the existence criterion for temperature fails in the hot spot.

Let us denote by $\bar{\omega}_{t,l}$ the frequencies at which

$$v_{t,l} \tau_{t,l}(\bar{\omega}_{t,l}) = d. \quad (6)$$

Transverse and longitudinal phonons with frequencies below $\bar{\omega}_{t,l}$ have a mean free path larger than the cylinder height d .

As a result of the spatial expansion, the height d of the cylinder becomes comparable to the radius r of its base. Then the frequencies $\bar{\omega}_{t,l}$ will be determined by the equation.

$$v_{t,l} \tau_{t,l}(\bar{\omega}_{t,l}) = r. \quad (7)$$

Phonons with frequencies $\omega < \bar{\omega}_{t,l}$ leave the hot spot, decreasing its energy. If $\hbar\bar{\omega}_{t,l} \ll \Theta, T$, then the energy remaining in the hot spot is given approximately by

$$E_p = \int_0^{\omega_D} d\omega \rho(\omega) \hbar\omega n(\omega) - \int_0^{\bar{\omega}_t} d\omega \rho_t(\omega) \hbar\omega n(\omega) - \int_0^{\bar{\omega}_l} d\omega \rho_l(\omega) \hbar\omega n(\omega), \quad (8)$$

where $\rho_{t,l}$ is the density of states for the transverse and longitudinal phonons, ρ is the polarization-averaged density of phonon states, and n is the Planck distribution function. Equation (8) yields

$$E_p = VCT [1 - A_t(\hbar\bar{\omega}_t/T) - A_l(\hbar\bar{\omega}_l/T)], \quad (9)$$

where V is the volume of the hot spot,

$$A_t(x) = \frac{2}{3} \left(\frac{v}{v_t}\right)^3 f(x), \quad A_l(x) = \frac{1}{3} \left(\frac{v}{v_l}\right)^3 f(x),$$

$$f(x) = \frac{5}{\pi^4} x^4 D_E(x),$$

and D_E is the Debye function. If $\hbar\bar{\omega}_{t,l} < 2.8T$, then the quantities $A_{t,l}$ in (9) are small compared to unity. The leakage of energy from the hot spot can thus be neglected until the time when the existence criterion for temperature fails:

$$\hbar\bar{\omega}_{t,l} = 2.8 T_K. \quad (10)$$

Allowance for the leakage of energy makes sense only near the temperature T_K , when Eq. (10) holds. However, as a calculation shows, the energy leakage does not have an important effect on the results. We assume for simplicity that the energy of the spot is conserved: $E_p = E_0$.

2. ONE-DIMENSIONAL EXPANSION

As we have said, the hot spot is initially (at time zero) a cylinder of height $d_0 \ll r_0$. The rate of decrease of the energy density as a result of the spatial expansion of the spot is determined by the growth of the height d . We can assume for

simplicity that the radius of the cylinder remains constant: $r = r_0$, i.e., the expansion of the hot spot is one-dimensional up until the height of the cylinder becomes comparable to the initial radius r_0 . For $d \sim r_0$ the shape of the hot spot can be represented as a hemisphere with initial radius r_0 .

To join the final volume $\pi r_0^2 d$ of the cylinder with the initial volume $2\pi r_0^3/3$ of the hemisphere, we set

$$d_0 \leq d \leq 2r_0/3. \quad (11)$$

Introducing the dimensionless variables $y_1 = T/T_0$ and $z = d/d_0$ and taking into account the relation $E_p = E_0$ and Eqs. (1) and (2), we write the expression for the hot-spot energy $E_p = VCT$ for $T > T_c$ in the form

$$zy_1 = 1. \quad (12)$$

It follows from (11) that

$$1 \leq z \leq 2r_0/3d_0 = 20. \quad (13)$$

Using (11) and (12), we can write the inequality $T > T_c$ as

$$1 \leq z \leq T_0/T_c = E/E_c, \quad (14)$$

where

$$E_c = \pi r_0^2 d_0 C_v T_c / 0.6 = 0.93 \mu\text{J}.$$

The initial value of the temperature $T_0 > \Theta$ corresponds to

$$E_\Theta < E < E_m, \quad (15)$$

where $E_m = 10 \mu\text{J}$ is the energy that must be added in order to melt the germanium. Because $E/E_c < 2r_0/3d_0$ for energy interval (15), the temperature goes through the point T_c before the expansion of the hot spot enters the three-dimensional regime, and inequality (14) holds.

For $T < T_c$ we get, with allowance for (1) and (2),

$$(E/E_c)^3 zy_1^4 = 1, \quad (16)$$

where

$$E/E_c \leq z \leq 2r_0/3d_0.$$

For t phonons Eqs. (4)–(6) yield

$$\hbar\bar{\omega}_t/\Theta = \bar{E}_t'/E, \quad T > T_c, \quad (17)$$

$$\hbar\bar{\omega}_t/T = (\bar{E}_t'/E)^{3/4} z^{1/4}, \quad T < T_c,$$

while for l phonons we get

$$\hbar\bar{\omega}_l/\Theta = (\bar{E}_l'/E)^{1/2}, \quad T > T_c, \quad (18)$$

$$\hbar\bar{\omega}_l/T = (\bar{E}_l'/E)^{3/4} z^{1/2}, \quad T < T_c,$$

where the energies (in μJ)

$$\bar{E}_t' = 4.97 \cdot 10^{-2}, \quad \bar{E}_l' = 0.285, \quad \bar{E}_t' = 0.24, \quad \bar{E}_l' = 3.2$$

are evaluated with the following values substituted into the expressions for $\tau_{t,l}$ and $\tau_{l,t}$: Grüneisen parameter $\gamma = 0.63$, density $\rho = 5.32 \text{ g/cm}^3$, average longitudinal sound velocity $v_l = 5.4 \cdot 10^5 \text{ cm/sec}$.

The maximum value of $\hbar\bar{\omega}_{t,l}/T$ corresponds to $z = 2r_0/3d_0$. For energies in range (15) we have $\hbar\bar{\omega}_{t,l}/T < 2.8$. Thus

the existence criterion for temperature is fulfilled over the entire course of the one-dimensional expansion.

The expansion is governed by heat conduction. The continuity equation is of the form

$$\frac{\partial}{\partial t} \left(\frac{E}{V} \right) + \text{div } \mathbf{j} = 0, \quad (19)$$

where the heat flux $\mathbf{j} = -\kappa \nabla T$. Over a wide range of temperatures extending to 20 K, the thermal conductivity κ in crystalline Ge conforms approximately to the relation $\kappa T = B$, where $B = 1.5 \cdot 10^9 \text{ erg/cm} \cdot \text{sec}$ (Ref. 12).

After integrating (19) over the volume of the hot spot, we get

$$\frac{E}{V} \frac{\partial V}{\partial t} = \int \mathbf{j} \cdot d\mathbf{s}. \quad (20)$$

The integration in (20) is over the surface of the cylinder. We shall neglect the contribution to the integral from the lateral surface of the cylinder and assume that the temperature on the surface of the cylinder is half as large as in the interior and that $\nabla T = -T/d$ on the surface. We then find from (20) that

$$d \partial d / \partial t = 2\kappa / C. \quad (21)$$

Converting in (21) to the dimensionless variables z and $\eta = t(v_l/v)^{-3}(r_0/v_l)^{-1}$, we get

$$\partial z / \partial \eta = E_1/E, \quad (22)$$

where

$$E_1 = 6.67\pi (v_l/v)^3 (Br_0^3/v_l d_0) = 2.09 \mu\text{J}.$$

Integrating (22) with respect to z over the limits (13), we find that the time of the one-dimensional expansion, measured in microseconds, is given by $t_1 = 0.134E$. Energy interval (15) corresponds to the time interval

$$0.53 \mu\text{sec} < t_1 < 1.34 \mu\text{sec}. \quad (23)$$

3. THREE-DIMENSIONAL EXPANSION

After time t_1 the hot spot is represented as a hemisphere of radius r . The initial temperature of the three-dimensional expansion regime is found from (16) for $z = 2r_0/3d_0$:

$$T_{r_0} = \Theta (E/E_{r_0})^{1/4}, \quad E_{r_0} = 2r_0 E_\Theta^4 / 3d_0 E_c^3 = 6.1 \cdot 10^3 \mu\text{J}. \quad (24)$$

From (24), with allowance for (15), we find

$$59.7 \text{ K} < T_{r_0}/k < 75.3 \text{ K}. \quad (25)$$

Introducing the dimensionless variables $y_2 = T/T_{r_0}$ and $\xi = r/r_0$, we get, in analogy with (12) and (16),

$$\xi^3 y_2^4 = 1. \quad (26)$$

From (4), (5), (7), and (26) we have

$$\hbar\bar{\omega}_t/T = (\bar{E}_t'/E)^{3/4} \xi^{1/4}, \quad \hbar\bar{\omega}_l/T = (\bar{E}_l'/E)^{3/4} \xi^{1/2}, \quad (27)$$

where $\bar{E}_t' = 0.37 \mu\text{J}$ and $\bar{E}_l' = 4.2 \mu\text{J}$.

The equation describing the three-dimensional expansion is obtained in analogy with (2), by assuming that $\partial T / \partial r = -T/r$ on the surface of the hemisphere:

$$r \partial r / \partial t = 2\kappa / C. \quad (28)$$

In the variables ξ and μ , Eq. (28) becomes

$$\partial \xi / \partial \eta = (E_2 / E) \xi^2, \quad (29)$$

where

$$E_2 = E_1 \cdot 2d_0 / 3r_0 = 4.64 \cdot 10^{-2} \mu\text{J}.$$

The three-dimensional-expansion time t_2 , which is determined by the thermal conductivity, is obtained by integrating (29) over the limits $1 < \xi < \xi_K$, where ξ_K is determined by (10) and (27):

$$t_2 [\mu\text{sec}] = 0.32E(1 - \xi_K^{-1}).$$

The rate of anharmonic interactions of the longitudinal low-frequency phonons is lower than for the transverse phonons. Equation (10) comes to hold sooner for the longitudinal phonons. From Eq. (10) and the second equation in (27) we get

$$\xi_K = (E/E_K)^{1/11}, \quad E_K = 0.8 \mu\text{J}. \quad (30)$$

Interval (15) corresponds to the intervals

$$2.07 < \xi_K < 3.15, \quad 0.66 \mu\text{sec} < t_2 < 2.18 \mu\text{sec} \quad (31)$$

Unquestionably, the frequency dependence given by (5) is incorrect in the region of thermal frequencies (10), and Eqs. (27) and (30) are only good for estimates.

4. EXPANSION OF NONDECAYING PHONONS

At time $t_1 + t_2$, when the dimension of the hot spot reaches $r_K = r_0 \xi_K$, the temperature criterion fails, and the spatial expansion of the phononic inhomogeneity ceases to be of the nature of heat conduction. For interval (15) we get, with allowance for (23) and (31),

$$1.19 \mu\text{sec} < t_1 + t_2 < 3.52 \mu\text{sec}, \quad 62.1 \mu\text{m} < r_K < 94.5 \mu\text{m}. \quad (32)$$

Because of the low density of thermal energy the only phonon-phonon process is phonon decay. The bulk of the phonons have an energy $\hbar\omega_K = 2.8T_K$, where T_K , the "final" temperature of the three-dimensional heat-conduction expansion, is determined from (24), (26), and (30):

$$T_K = T_0 \xi_K^{-14}.$$

Energy interval (15) corresponds to

$$31.8 \text{ K} < T_K/k < 32.8 \text{ K} \quad \text{or} \quad 1.23 \cdot 10^{13} \text{ Hz} < \omega_K < 1.27 \cdot 10^{13} \text{ Hz}. \quad (33)$$

In what follows we assume that the longitudinal phonons are the decaying ones, while the transverse phonons are nondecaying. The decay rate¹³ is given by

$$1/\tau_{l \rightarrow t+l}(\omega) = \gamma_{l \rightarrow t+l} \omega_D (\omega/\omega_D)^5, \quad \gamma_{l \rightarrow t+l} = 2 \cdot 10^{-2} \quad (34)$$

(we are ignoring the decay $l \rightarrow t + t$, which occurs much less often).

The rate of elastic isotopic scattering with mode conversion¹⁴ is

$$1/\tau_{l \rightarrow t}(\omega) = \eta_{l \rightarrow t} \omega_D (\omega/\omega_D)^4, \quad (35)$$

where

$$\eta_{l \rightarrow t} = (a_0^3 \delta / 12\pi) (\omega_D / v_l)^3,$$

$a_0^3 = 4.53 \cdot 10^{-23} \text{ cm}^3$ is the volume of the unit cell of Ge, and $\delta = 5.9 \cdot 10^{-4}$.

It follows from (32) and (33) that for phonons with a frequency

$$\omega > \omega_D \eta_{l \rightarrow t} / \gamma_{l \rightarrow t+l} = 1.6 \cdot 10^{12} \text{ Hz} \quad (36)$$

decay occurs faster than elastic scattering with mode conversion. We see that phonons with frequency (33) satisfy inequality (36). The phonons will accumulate on the nondecaying t branch of the phonon spectrum. As was shown in Refs. 5 and 6, the spatial expansion of the phononic inhomogeneity under these conditions occurs linearly in time, with a velocity

$$s = v_l (v_l^3 / 6v_t^3)^{1/2} = 6.2 \cdot 10^4 \text{ cm/sec}.$$

For comparison of the values of t_1 , T_0 , r_K , $t_1 + t_2$, and T_K in (23), (25), (32), and (33) at pump energies (15), let us give the values calculated in Ref. 7 for $E = 1 \mu\text{J}$ under the experimental conditions of Ref. 4. The initial temperature of the hot spot is $T_0/k = 103 \text{ K}$, and $t_1 = 1.4 \mu\text{sec}$, $T_0/k = 38 \text{ K}$, $t_1 + t_2 = 2.8 \mu\text{sec}$, and $T_K/k = 28 \text{ K}$.

5. DURATION OF THE BALLISTIC PULSE

As we mentioned in the Introduction, the pulses of ballistic phonons arriving at the detector were extracted by the spatial filtering method, which is based on the focusing effect for ballistic phonons. Specifically, the signal arriving at point B from the point of excitation is subtracted from the signal arriving at point A (see Fig. 1). Points A and B lie at angles of α and β to the $\langle 100 \rangle$ direction in the crystal. At propagation angles $\alpha < \varphi < \beta$ the intensity of the ballistic t phonons reaching the opposite face of the crystal from the excitation region falls off sharply. At angles larger than β , ballistic phonons do not propagate (the l phonons do not propagate ballistically in the $\langle 100 \rangle$ direction).

Thus the spatial filtering method can be used to find the pulse of the ballistic t phonons by subtracting the signal arriving at point B from the signal arriving at point A as long as the dimensions of the phononic inhomogeneity are smaller than the distance R between points A and B . When the radius of the hemisphere reaches the value R , the low-frequency ballistic phonons produced in the decay will arrive at A and B in an identical manner. The value of R is given by

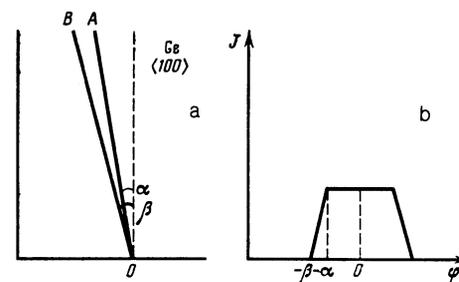


FIG. 1. a) Phonon excitation region (O) and the trajectory of ballistic phonons at angles α (OA) and β (OB) to the $\langle 100 \rangle$ direction in Ge. b) Ballistic signal J near the $\langle 100 \rangle$ direction in Ge versus the propagation angle φ of the ballistic phonons.

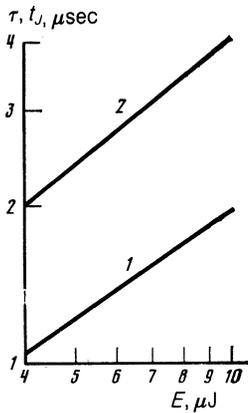


FIG. 2. Exponential damping constant τ (1) and the duration t_J (2) of the ballistic signal versus the energy E of the light pulse. The slopes of the curves $\tau(E)$ and $t_J(E)$ are 0.7 and 0.8, respectively.

$R = L(\tan \beta - \tan \alpha) = 500 \mu\text{m}$, where $L = 1 \text{ cm}$ is the dimension of the sample, and $\tan \beta - \tan \alpha = 5 \cdot 10^{-2}$ (Ref. 4). Because $R \gg r_K$, the time required for the nondecaying t phonons to expand to a hemisphere of radius R is $t_R \approx R/s = 0.81 \mu\text{sec}$.

The time $t_J = t_1 + t_2 + t_R$ is thus the duration of the ballistic phonon signal that is extracted from the detector signal by the spatial filtering method.

Up to time t_J the bulk of the phonons have the frequency⁶

$$\omega_R = \omega_K [s\tau_{l \rightarrow l}(\omega_K)/R]^{1/4} = 4.07 \cdot 10^{12} \text{ Hz.}$$

Consequently, $\omega_K > \omega_R > 1.6 \cdot 10^{12} \text{ Hz}$, and condition (36) holds. It has been shown experimentally⁴ that the intensity

of the ballistic pulse falls off exponentially in time, with a time constant τ . The calculated $t_J(E)$ and $\tau(E)$ curves are shown in Fig. 2.

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