# Kinetics of the interaction of nonequilibrium carriers in germanium with alternating and constant magnetic fields at low temperatures

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We investigate the time dependence of the absorption of the energy of an alternating magnetic field as a result of excitation of eddy currents in a nonequilibrium electron-hole plasma in germanium at low temperatures. It is shown that absorption by free electrons and holes predominates in undeformed germanium. In samples subjected to inhomogeneous deformation, the main contribution to the absorption is made by "large" electron-hole drops (EHD). From the dependence of the absorption on the intensity of the constant magnetic field we determined the mobility of the free carriers  $(4.7 \times 10^6 \text{ cm}^2/\text{V-sec})$  and the mobility in large EHD  $(1.3 \times 10^6 \text{ cm}^2/\text{V-sec})$  at 2 K. The possible scattering mechanisms that determine the mobility are discussed.

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### **1. INTRODUCTION**

In preceding studies<sup>1,2</sup> the conductivity of electronhole drops (EHD) in germanium was determined by measuring the absorption of the energy of an alternating magnetic field on account of excitation of eddy currents in a nonequilibrium conducting medium. The mobility of the nonequilibrium carriers was determined from the dependence of this absorption on the intensity of a constant magnetic field, in analogy with the usual transverse magnetoresistance effect. To produce nonequilibrium carriers, quasistationary photoexcitation was used in Refs. 1 and 2, because the apparatus was not sensitive enough to investigate the kinetics of the absorption. For a reliable separation of the contributions of the free and condensed carriers to the absorption, however, it is important to determine the absorption time dependence, which is investigated in the present paper.

#### 2. PROCEDURE

To increase the sensitivity to the high-frequency absorption, we used autodyne detectors of known circuitry,<sup>3,4</sup> and placed the investigated germanium samples in the coil of their tank circuits. The samples were excited with an argon laser having a power P up to 0.5W, modulated by a rotating perforated disk, producing light pulses with a rise time ~10  $\mu$ sec. The duration of the light pulse ranged from 50 to 500  $\mu$ sec, depending on the experimental conditions. The signal due to the change of the Q of the autodyne-detector tank circuit as a result of excitation of the samples was amplified with a broad band amplifier, and recorded with a strobe-integrator and with an oscilloscope. The remaining elements of the apparatus did not differ from those used previously.<sup>1,2</sup> The apparatus made it possible to investigate the kinetics of the high-frequency absorption due to excitation of eddy currents in a system of non-equilibrium carriers with a time resolution ~5  $\mu$ sec in a wide interval of photoexcitation levels.

At low modulation of the autodyne tank-circuit Q by the photoexcited samples, we can put<sup>5</sup>

$$\Delta U \sim QW/U.$$

Here U is the amplitude of the high-frequency oscillations of the autodyne tank-circuit, W is the power alternating magnetic-field power  $h = h_0 e^{i\omega t}$  absorbed by a conducting sphere of radius R placed in a tank circuit with quality factor Q (Ref. 6):

(1)

$$W = \frac{1}{2} V \omega \alpha'' h_0^2, \qquad (2)$$

where  $V = \frac{4}{3}\pi R^3$ ,  $\omega = 2\pi f$ , f is the autodyne frequency, and  $\alpha''$  is the imaginary part of the magnetic polarizability:

$$\alpha'' = -\frac{9}{4\pi} \frac{1}{x^2} \left( 1 - \frac{x}{2} \frac{\operatorname{sh} x + \sin x}{\operatorname{ch} x - \cos x} \right),\tag{3}$$

where  $x = 2R/\delta$ ,  $\delta = c/(2\pi\omega\sigma)^{1/2}$  is the depth of the skin layer, and  $\sigma$  is the conductivity.

At a skin-layer depth much larger than the EHD dimensions,  $x \ll 1$ , and

$$\alpha'' = \frac{1}{80\pi} x^2, \quad W = \frac{\pi R^3 \omega^2 h_0^2 \sigma}{15c^2}.$$
 (4)

In the opposite case, at  $x \gg 1$ 

$$\alpha'' = \frac{9}{8\pi} \frac{1}{x}, \quad W = \frac{3cR^2 \omega'' h_0^2}{8(2\pi\sigma)^{\frac{1}{2}}}.$$
 (5)

The effect of a constant magnetic field of intensity H on the absorption W can be taken into account in the expression for the conductivity of the EHD:

$$\sigma = \frac{\sigma_o}{1 + (\gamma \mu H/c)^2},$$
(6)

where  $\sigma_0 = en_0(\mu_e + \mu_h)$ ,  $n_0$  is the concentration and  $\mu_e$  and  $\mu_h$  are the mobilities of the electrons and holes in the EHD. It can be shown that

$$\mu = (\mu_e \mu_h)^{\nu_h}, \quad \gamma = \begin{cases} 1, & \mathbf{H} \parallel \mathbf{h} \\ 2^{-\nu_h}, & \mathbf{H} \perp \mathbf{h} \end{cases}.$$
(7)

At  $\mu_e = \mu_h$  expressions (6) and (7) are valid for all *H*, and at  $\mu_e \neq \mu_h$  they are satisfied at H = 0 and  $\gamma \mu H/c \gg 1$ . From this we have at  $x \ll 1$ 

$$W = \frac{\pi R^{5} \omega^{2} h_{0}^{2}}{15 c^{2}} \frac{\sigma_{0}}{1 + (\gamma \mu H/c)^{2}}.$$
 (8)

At  $x \gg 1$  we have

$$W \sim \sigma^{-\gamma_{2}} = \left(\frac{1 + (\gamma \mu H/c)^{2}}{\sigma_{0}}\right)^{\gamma_{2}}.$$
 (9)

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With increasing intensity of the constant magnetic field H, the conductivity  $\sigma$  decreases as a result of the magnetoresistance effect, and the thickness of the skin layer  $\delta$  increases. At  $\delta \approx 2R$ , the absorption W reaches a maximum in a field

$$H_m = \frac{c^2}{2\gamma R (2\pi\omega\sigma_0)^{\nu_h}}.$$
 (10)

With further increase of H, when the condition  $x \ll 1$  begins to be satisfied, the function W(H) is described by expression (8), i.e.,  $W \sim H^{-2}$ . Thus, depending on the experimental conditions, expressions (8) – (10) can be directly used to determine the mobility  $\mu$  in EHD.

We note in conclusion that it follows from (1) that the response of the autodyne detector  $\Delta U$  increases with decreasing voltage on its tank circuit U. At too small U, however, the oscillations in the autodyne became unstable and the noise at the output of the circuit increased drastically. The feedback was therefore adjusted to set a value U that maximized the signal/noise ratio at the output of the circuit.

#### 3. ABSORPTION IN UNDEFORMED GERMANIUM

When excitons condense in undeformed germanium, an EHD cloud is produced with a characteristic dimension  $10^{-3}-10^{-4}$  cm (Ref. 7) surrounded by excitons and by free carriers. The absorption of energy from the alternating magnetic field is due in this case to the interaction with both the EHD and the free carriers. An investigation of the kinetics of the absorption makes it possible to separate these two absorption components.

Figure 1 shows the time dependence of the absorption  $\Delta U$ . It is seen from the figure that after the excitation is stopped the absorption signal  $\Delta U$  decreases linearly with time in accordance with the law

$$\Delta U \sim (1 - t/t_0), \tag{11}$$

where the cutoff time is  $t_0 \approx 300 \ \mu \text{sec.}$  At the same time we observed in the recombination radiation of germanium under the experimental conditions only the EHD emission, which relaxed exponentially  $I_r \approx \exp(-t/\tau_0)$ , where  $\tau_0 = 40 \ \mu \text{sec}$  is the EHD lifetime. Recognizing<sup>2</sup> that  $\Delta U \sim I_r^{5/3}$ , one should expect the high-frequency absorption to be likewise exponential with a time constant  $\frac{3}{5}\tau_0 = 24 \ \mu \text{sec.}$  It is seen from Fig. 4, however, that  $\Delta U$  relaxes non-exponentially and with a characteristic time larger by an order of magnitude. At the same time, a relation of type (11) describes the relaxation of the concentration of free electrons and holes in the at-



FIG. 1. Time dependence of the absorption signal  $\Delta U$  and of the intensity of the exciting radiation I in undeformed germanium with  $N = 2 \times 10^{10}$  cm<sup>-3</sup> at 2 K and P = 0.2 W, f = 5 MHz.

mosphere surrounding the EHD.<sup>8</sup> This relation was obtained under the assumption that in the absence of excitation the free carriers and produced in the crystal by evaporation from the surface of the EHD, and vanish as a result of binding into excitons. The value of  $t_0$  is then determined by the temperature and by the initial radius of the EHD. Since the evaporation of electrons and holes from an EHD is negligibly small at 2 K, we can assume that the hot carriers are produced inside the EHD by Auger recombination, but leave the drops only from a thin surface layer. A rough estimate shows that the thickness of this layer can be  $\sim 10^{-5}$  cm. In this case the flux of electrons and holes in the space surrounding the drops is also proportional to the surface of the drops and expression (11) describes the time dependence of the concentration of the free carriers of the charge in the atmosphere surrounding the EHD.

Thus, absorption is nondeformed germanium is determined by the presence of free carriers in the atmosphere surrounding the EHD, and not by the eddy currents surrounding the small drops themselves. This conclusion is clearly confirmed by measurements of the absorption in a thin germanium plate differently oriented relative to the direction of the high-frequency magnetic field h in the tank-circuit coil and at fixed geometry and intensity of the photoexcitation. The plate thickness was chosen to be  $7 \times 10^{-3}$  cm, i.e., much larger than the characteristic dimension of the EHD. When the exciting radiation was sharply focused on the large surface of the plate, the EHD cloud assumed the form of a pancake<sup>9</sup> with thickness equal to that of the plate and with radius of the order of 1 mm. When the larger surface of the plate was oriented perpendicular to h, the absorption signal  $\Delta U$ , while somewhat weaker than in thick germanium samples under identical excitation, it nevertheless exceeded by two orders of magnitude the noise level. When the orientation of the plate was changed so that h was parallel to its larger surface, the response of the autodyne detector, at the same sensitivity, did not exceed the noise level. Inasmuch as in both cases the concentration and dimension of the EHD remained unchanged because the excitation conditions were identical, and all that changed was the orientation of the pancake-shape cloud relative to the direction of the alternating magnetic field, the change of the absorption could be due only to the conductivity of the cloud as a whole, i.e., to the conductivity of the atmosphere surrounding the EHD.

Measurement of the absorption of the energy of the alternating magnetic field upon photoexcitation of the germanium samples was used to determine the mobility of the free carriers at low temperatures. In fact, in thick germanium samples, the shape of the cloud of the EHD and of the atmosphere surrounding them is close, in the case of sharp focusing of the exciting radiation, to a hemisphere with almost constant average concentration of the electron-hole pairs.<sup>7</sup> In this case expression (8) should describe sufficiently well the dependence of the absorption and the constant magnetic field *H* for the case when  $\Delta U$  is determined by interaction with the free carriers in the cloud. The experimental effective mobility of the free carriers determined from this relation at 2 K turned out to be  $4.7 \times 10^6$  cm<sup>2</sup>/V-sec in ger-

manium samples having a residual-impurity concentration  $N < 10^2$  cm<sup>-3</sup>. This value of the mobility agrees with the values determined for germanium by other methods and extrapolated into the low-temperature region accordance with the law  $\mu \propto T^{-3/2}$  (see, e.g., Ref. 10), and indicates that scattering by acoustic phonons predominates.

#### 4. ABSORPTION BY LARGE ELECTRON-HOLE DROPS

Large EHD drops were produced by photoexcitation of samples of inhomogeneously deformed germanium with dimensions  $4 \times 3.8 \times 2.5$  mm in the apparatus described in Ref. 2. The deforming force was applied in the (111) direction through a caprone plunger, and a single drop was produced in the samples in the region of minimum deformation.

Figure 2 shows the time dependence of the response  $\Delta U$  of the autodyne detector to the exciting-radiation pulse. It is seen from the figure that the decrease of the absorption, after the end of the excitation, is nearly exponential with a time constant 200  $\mu$ sec. The same figure shows the time dependence of the intensity of the recombination radiation of a large EHD, which relaxes likewise exponentially a time constant  $\tau_0$ =350  $\mu$ sec, corresponding to the lifetime of large EHD.

At a moderate photoexcitation power, not exceeding 300 mW, and at an autodyne-detector frequency 5 MHz, the dependence of the absorption  $\Delta U$  on the magnetic field intensity H was well described by expression (8), which was used to calculate the effective mobility  $\mu$ . Figure 3 shows the dependence of the mobility calculated in this manner on the intensity of the photoexcitation in measurements both during the exciting pulse and after its termination. The same figure shows plots of the intensity of the recombination radiation of a large EHD against the excitation intensity. It is seen from the figure that in the absence of excitation the measured mobility is practically independent of the excitation level (of the EHD size) up to  $P \approx 250$  mW and is equal to 1.3  $\times 10^{6}$  cm<sup>2</sup>/V-sec. In measurements during the time of excitation and at a power  $P \leq 30$  mW, the mobility is  $\mu = 4.7 \times 10^6$  cm<sup>2</sup>/V-sec, after which, with P increased to 50-60 mW, the absorption  $\Delta U$  increases sharply, while  $\mu$  decreases to  $1.5 \times 10^6$  cm<sup>2</sup>/V-sec. In the same region of the excitation power, the intensity  $I_r$  of the



FIG. 2. Time dependence of the intensity of the exciting radiation *I*, of the absorption signal  $\Delta U$ , and of the intensity of the recombination radiation *I*, in an inhomogeneously deformed germanium with  $N = 2 \times 10^{10}$  cm<sup>-3</sup> at 2 K and P = 0.2 W, f = 5MHz.





FIG. 3. Dependence of the carrier mobility  $\mu$  and of the recombination-radiation intensity  $I_r$  on the excitation power P in inhomogeneously deformed germanium with  $N = 2 \times 10^{10}$  cm<sup>-3</sup> at 2 K: 1,3— $\mu$  and  $I_r$  measured with a delay of 150  $\mu$ sec after the end of the excitation; 2,4— $\mu$  and  $I_r$  measured during the time of excitation; f=5 MHz.

EHD radiation increases sharply. With further increase of the power, the mobility remains practically un-changed and the radiation intensity increases linearly.

It can be concluded from the foregoing that the mobility measured during the time of low-intensity excitation pulse corresponds to the mobility of the free carriers outside the EHD. This value agrees well with the value of  $\mu$  determined in undeformed germanium for free carriers. At sufficiently intense excitation, when the EHD dimension increases, absorption inside the drop predominates and the measured value of  $\mu$  corresponds to the mobility of the charge carrier in the liquid phase. The difference between the measurements, both during the time of the excitation pulse and after its termination, is small.

In measurements of the absorption at high frequencies and high intensity of photoexcitation, an increase of  $\Delta U$ is observed with increasing intensity of the constant magnetic field H (Fig. 4) in accordance with expression (9), which is valid at a skin-layer thickness  $\delta$  smaller than the drop dimension 2*R*. The maximum of the absorption is reached at  $H_m = 300$  Oe, which is reasonable in accordance with the estimate (10). With further increase of H, we get  $\Delta U \sim H^{-2}$  in accordance with (8).

Thus, an investigation of the kinetics of the absorption of the energy of the alternating magnetic field by a large



FIG. 4. Dependence of the absorption signal  $\Delta U$  on the intensity of the constant magnetic field H in a germanium sample with  $N = 2 \times 10^{10}$  cm<sup>-3</sup> at 2 K and P = 0.25 W, measured during the excitation time, f = 30 MHz.

EHD in nonuniformly deformed germanium has shown that the determination of the mobility of the carriers in the electron-hole liquid from the  $\Delta U(H)$  dependence is correct even in the presence of excitations, provided that the intensity of the excitation is  $P \ge 100$  mW. It follows from this also that the use of a quasistationary excitation of large EHD (Ref. 1, 2) is perfectly permissable for the measurement of the mobility of condensed carriers. No account was taken in Refs. 1 and 2, however, of the factor  $\gamma = 2^{-1/2}$  for  $h \perp H$ , and therefore the values of  $\mu$  were somewhat underestimated there. The values of the mobility in large EHD, in both pure and doped germanium, determined from the kinetic measurements, are the following:

$N,  \mathrm{cm}^{-3}$ :	2.1010	2.6.1011	1.5.1014	2.1014	8.1014
$\mu$ , cm <sup>2</sup> /W·sec :	1.3.106	1.3-10*	3.1.105	2.6·10 <sup>5</sup>	8.4 104

#### 5. SCATTERING MECHANISMS

The mobility in large EHD begins to decrease substantially at impurity concentration  $N \ge 10^{14}$  cm<sup>-3</sup>, and in this concentration region we have  $\mu \sim N^{-1}$ . Thus, in samples of doped germanium the carrier mobility inside the EHD is determined by the scattering by impurities. Estimates made in Ref. 2 show that the absolute value of  $\mu$ agrees with the assumption of scattering by neutral impurity atoms. This is natural, since the concentration of the electrons and holes in large EHD,  $n_0 \approx 6$  $\times 10^{16}$  cm<sup>-3</sup>, is lower than critical for impurity atoms of groups III and V ( $\sim 2 \times 10^{17}$  cm<sup>-3</sup>, Ref. 11), at which these atoms become ionized.

At lower impurity densities, the mobility is constant and consequently it can be determined by scattering from the latter vibrations, or by electron-hole scattering. To determine the contribution of these scattering mechanisms, we present some estimates.

In both deformed and undeformed germanium, the mobility of the free carriers was the same for both puregermanium samples used in the experiments, and was independent of the photoexcitation intensity; this confirms the lattice character of the scattering. It was noted above that the obtained value  $\mu = 4.7 \times 10^6$  cm<sup>2</sup>/ V-sec corresponds at 2 K approximately to scattering by acoustic phonons. In acoustic scattering<sup>10</sup>

$$\mu = \frac{e}{m^*} \tau_{ac}, \quad \tau_{ac} = a \frac{E^{-/n}}{kT}, \quad (12)$$

where  $E = 9 \pi k T / 16$  for nondegenerate carriers and  $E = F_e$ or  $F_{k}$  for degenerate carriers, e is the charge of the electrons,  $m^*$  is the effective mass, k is the Boltzmann constant, T is the temperature,  $F_e$  and  $F_h$  is the Fermi energy of the electrons and holes, and a is a constant that depends only on the properties of the germanium lattice. For the inhomogeneous deformations used in the present study and corresponding to maximum pressure 250-500 kg/cm<sup>2</sup>, we have  $F_e \approx F_h \approx 3 \text{ meV}.^{12}$  It follows then from (12) that at 2 K the ratio of the mobilities of the free and condensed carriers in acoustic scattering should be equal to 3.1. This is close to the experimental mobility ratio 3.5. Thus, the assumption that the electrons and holes are scattered by acoustic phonons in the large EHD does not contradict the experimental data.

The electron-hole scattering in a degenerate EHD plasma has not yet been calculated with sufficient accuracy. For a rough estimate we can use the relation proposed by Kittel<sup>13</sup> on the basis of intuitive considerations. This relation is based on the assumption that the electron-hole scattering is effective only in an energy band of the order of kT near the Fermi energy. The fractions of the electrons and holes in this band are approximately  $kT/F_e$  and  $kT/F_h$ . Since the average distance between the carriers in the EHD is  $(3/4\pi n_0)^{1/3}$  and their velocities on the Fermi level are  $\nu_F = (2F/m^*)^{1/2}$ , the effective scattering time turns out to be

$$\tau_{eh} = \left(\frac{3}{4\pi n_0}\right)^{\frac{1}{2}} \left(\frac{m_e m_h}{4F_e F_h}\right)^{\frac{1}{2}} \frac{F_e F_h}{(kT)^2}.$$
(13)

Substitution of the values of  $n_0$ ,  $F_e$ , and  $F_h$  given above yields  $\tau_{eh} = 10^{-10}$  sec for large EHD<sup>12</sup> and  $(m_e m_h)^{1/2} = 0.2m$ ; this corresponds to  $\mu = 8 \times 10^5 \text{ cm}^2/\text{V-sec}$ . This value is also close to the experimental  $\mu$  in large EHD.

Thus, our estimates show that the mobility in large EHD can be determined both by scattering from acoustic phonons and by electron-hole scattering. To separate the predominant scattering mechanism, it may be useful to investigate the temperature dependence of  $\mu$ , inasmuch as in acoustic scattering  $\mu \propto T^{-1}$ , and in electronhole scattering  $\mu \propto T^{-2}$ . Preliminary investigations of the temperature dependence of  $\mu$  have shown that it is stronger than  $T^{-1}$  but weaker than  $T^{-2}$ . Account must be taken here also of the results of Gantmakher and Levinson, 14 who have shown that the electron-hole scattering can determine the absolute value of the plasma conductivity, but does not manifest itself in the transverse magnetoresistance. It was noted in Ref. 2 that the absolute value of the energy of the alternating magnetic field absorbed by large EHD agrees within the limits of the accuracy with the value calculated on the basis of measurements of the EHD dimensions at  $n_0$ =  $6 \times 10^{16}$  cm<sup>-3</sup> and the mobility determined from the dependence of the absorption on the H. With the foregoing taken into account, it can be concluded either that scattering by acoustic phonons predominated in large EHD, or that the contribution of the phonon and electron-hole scattering are approximately equal.

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## Instability of plane evaporation front in interaction of laser radiation with a medium

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The stability of the boundary between condensed matter and its vapor is investigated in the case when the evaporation is produced by a laser beam. It is shown that at sufficiently high radiation intensities-more than 10<sup>6</sup> W/cm<sup>2</sup> for most metals—the evaporation front is unstable. The maximum instability growth rate is possessed by perturbations with wavelengths on the order of the depth of penetration of the light in the evaporated matter. The growth time of these perturbations is much shorter than the time of establishment of the quasistationary evaporation regime.

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#### 1. INTRODUCTION

The results of numerous experiments on the evaporation of condensed substances by laser radiation can be satisfactorily explained by using the surface-evaporation model.<sup>1-3</sup> According to this model evaporation comes from a thin (of the order of the interatomic distances) surface layer of condensed phase, to which the energy is transferred from the light-absorption region by heat conduction. Since the depth of penetration of the radiation into the condensed medium is always much larger than the thickness of the layer from which the evaporation takes place, the temperature near the phase boundary increases with increasing distance from the boundary, and reaches a maximum at a certain depth. It is easy to show that in this case the phase separation boundary is unstable. Indeed, when a certain section of the boundary shifts towards the more heated condensed phase, the heat flux to this section increases, and this accelerates the boundary and increases further the initial perturbation. Understandably, the amplitude of the boundary displacement can not exceed the thickness of the surface layer in which the heat flux is directed from the condensed phase toward the evaporation front. This surface layer, as will be shown below, can be destroyed by shortwave perturbations within a time much shorter than the time of establishment of the stationary evaporation regime.

The described evolution of the process presupposes absence of the volume evaporation that could develop in principle in a liquid layer or on the grain boundaries in a polycrystalline solid phase. It is important, however, that in those cases when the vapor is formed in the volume, the result is the same as in the considered instability of the surface evaporation, namely destruction of the superheated surface layer and dispersion

of the liquid phase. The formation of metastable states of the condensed phase is therefore not very likely in experiments on laser evaporation. There is no point in discussing this question.

The considered instability-producing mechanism is, of course, not a feature of the laser-induced (or any other) evaporation process. The necessary condition for the instability is that when the heat is released in the volume (for example, under the influence of a shock wave or an electron beam), the phase transition with energy absorption take place not in the volume but on some surface. This situation is typical of melting and evaporation of solids, where the nucleation of the new phase calls for noticeable additional expenditure of energy.<sup>4,5</sup> An increase in the area of the surface on which the phase transition takes place leads to a decrease of the free energy of the system as a whole. For this reason, a plane phase boundary is unstable. We note that the boundary perturbations with the shortest wavelengths should be damped because of the increase in the surface energy.

Thus, the discussed phenomenon is of quite general character. Manifestations of the instability in question can be expected in a large group of experiments. We deal in the present article with experiments on laser evaporation of solids, since this topic has been well investigated experimentally and theoretically, so that the stability problem can, in particular, be correctly formulated. It seems to us, in addition, that the most suitable conditions for the observation of the evaporation instability are realized in experiments and technological processes in which lasers are used.

We point out that the mechanism of the onset of the thermal instability has been previously considered° in connection with the study of phase transitions accompanying electric explosion of conductors. The an-

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