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SPATIAL ORDERING OF DEFECTS BY LASER IRRADIATION

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A closed mathematical model is constructed for the kinetics of processes in laser-irradiated crystals, including a system of nonlinear equations for the temperature and strain fields and for the point-defect density. Two types of instability are considered: diffusion–strain and recombination–strain. Criteria for their onset are obtained. Formation of a periodic structure of point defects in a thin film is analyzed.

1. INTRODUCTION

Laser irradiation is a very effective method of endowing various materials with new properties of importance for many applications in microtechnology and optoelectronics. Thus, lasers can be used to produce films equipped with a superstructure and having electronic and optical properties that are not observed in homogeneous materials. Most timely in this connection is the development of a consistent description of the kinetic processes induced by laser action.

We construct in the present paper a closed mathematical model for processes in crystals at temperatures below the melting point. The model includes a system of nonlinear equations for the temperature field, for the strain field, and for the density of the point defects (vacancies, interstices, substitutional atoms). Such a system of equations is quite complicated. A significant amount of information can be obtained, however, by merely investigating the stability of this system. Onset, from a uniform distribution of point defects in a solid, of spatially inhomogeneous states is the result of the development of various instabilities produced by laser irradiation. We consider here two types of instability: diffusion-strain and recombination-strain. We discuss their mechanism and criteria for their realization.

The diffusion mechanism is used to investigate the formation of a periodic structure of point defects in thin films. A specific analysis is carried out of an experiment in which the film is produced by deposition from the gas phase while a focused cw CO_2 laser acts on the substrate. When the defect density exceeds a certain critical value, the spatially uniform distribution becomes unstable and goes over into an inhomogeneous state comprising an ordered array of point defects in the form of concentric rings with parameters that depend on the film properties and on the laser-radiation intensity. We have investigated the nonlinear

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stationary regime of diffusion–strain instability, which becomes stabilized by allowance for the anharmonicity of the elastic continuum.

2. MATHEMATICAL MODEL OF DEFECT KINETICS UNDER LASER IRRADIATION

Many experimental and theoretical papers have been published by now on the defect kinetics under laser irradiation of solids. A large number of interesting results have been obtained (see, e.g., [1]). At the same time, it is possible to formulate a closed system of equations that make up a mathematical model for the processes accompanying laser irradiation. It is known that defects generated by laser irradiation produce in a crystal a noticeable strain due to the difference between the values of the covalent radii of the defect and of the matrix atoms. In the presence of strain, the defects are subject to additional drift flows that influence substantially the kinetics of the formation of the spatial distribution of point defects. Assume that at the initial instant t = 0 there are produced in the crystal mobile point defects, such as vacancies and interstitial atoms. The basic processes that control the variation of the density of the point defects are diffusion, drift motion, mutual annihilation, and finally absorption by the drains. With these processes taken into account, the equations for the point-defect density can be written in the form

$$\frac{\partial n_a}{\partial t} = -\operatorname{div} j_a - \gamma n_i n_v - \beta_a n_a, \tag{1}$$

$$j_a = -D_a \operatorname{grad} n_a + v n_a, \tag{2}$$

where n_a (a = i or v, with i and v corresponding to interstices and vacancies, respectively) is the density of the point defects, β_a are the reciprocal times for trapping of defects of type a by drains, and γ is the recombination rate. Expression (2) determines the defect flux; its first term describes diffusion with a coefficient $D_a = D_{0a}\exp(-\varepsilon_{ma}/T)$ (D_{0a} is pre-exponential factor, ε_{ma} is the defect migration energy, and T is the temperature in energy units), the second term takes into account the defect drift ($v = (D/T)F$ is drift velocity) due to application of a force $F = -\operatorname{grad} U_{int}$ by the inhomogeneous strain field, U_{int} is the energy of interaction of one defect with the strain field: $U_{int} = K\Omega_a \operatorname{div} U$, K is the modulus of isotropic compression, Ω_a is the activation volume for the formation of a defect of type a, and U is the displacement vector. According to [2] we have $\Omega_a = \Delta\omega_a/\omega N_a$ where ω is the volume of the matrix atom, N_a is the density of the matrix atoms, and $\Delta\omega_a$ is the change of the crystal volume due to formation of one defect in it [2]. For example, for a vacancy or a small-radius impurity we have $\Omega_v < 0$, while for interstices or large-radius impurities we have $\Omega_v > 0$. It is taken into account in the derivation of (1) that the escape of defects to the surface can be neglected compared with the escape to internal drains.

The equation for the material-displacement vector is [3]

$$\frac{\partial^2 U}{\partial t^2} = C_t^2 \Delta U + (C_l^2 - C_t^2)\operatorname{grad}(\operatorname{div} U) + \frac{K}{\rho}\sum_{a=i,v}\Omega_a \operatorname{grad} n_a - \frac{K}{\rho}\operatorname{grad} T \tag{3}$$

where C_l and C_t are the longitudinal and transverse speeds of sound and ρ is the density of the medium. The third term in the right-hand side of this equation describes concentration stresses introduced into the medium by the defects, and the last term describe the thermal stress due to the inhomogeneous temperature field (a_T is the volume expansion coefficient).

The equation for the temperature field in the medium can be written in the form

$$C\frac{\partial T}{\partial t} + a_T K T \frac{\partial}{\partial t}(\operatorname{div} U) + \sum_{a=i,v}\Theta_a \frac{\partial n_a}{\partial t} = \kappa \Delta T + Q_T \tag{4}$$

where κ is the thermal conductivity, C is the heat capacity per unit volume, and Θ_a are kinetic coefficients representing the total heat transferred by the diffusion of the point defects and released from a unit volume that captures a point defect. In order of magnitude we have $\Theta_a \sim \varepsilon_{fa}$ [2], where ε_{fa} is the energy to produce a defect of type a. In the derivation of (4) we neglect the heat released upon recombination. Q_T describes the source of the heat released upon absorption of the laser energy: $Q_T = (1-R)K_0 I(r,t)$, where R is the reflectivity of the crystal, K the coefficient of optical absorption, and I(r, t) the radiation intensity.

Equations (1)-(4) constitute a closed system and describe completely the kinetics of point defects when a solid is heated by a laser. To make practical use of the foregoing mathematical model we must specify the characteristics of the external irradiation, as well as the properties and the parameters of the sample subject to the laser action.

3. INSTABILITIES IN IRRADIATED CRYSTALS

The physical mechanism that produces the instabilities is the onset in the medium, by fluctuation, of inhomogeneous strain or temperature fields. For example, elastic interaction of the strain field with the defects produces a force that causes thermal motion of the defects and heat transport, and accordingly leads to additional modulations of the defect density and of the temperature. The resultant inhomogeneous distributions of the defects and of the temperature serve in turn as sources of a force that deforms the elastic continuum and enhances the initial strain fluctuations. The resultant positive feedback leads to diffusion instability in the system.

Diffusion–strain instability was analyzed in [4] on the basis of a simplified mathematical model that takes no account of the heat-conduction equation and of the thermoelastic stresses. It turns out that when the vacancy density exceeds a certain critical value $n_* = T/K\Omega^2$ the sign of the effective diffusion of the point defects is reversed, and directed flows of vacancies and of interstitial atoms are produced in the compression and tension regions, respectively. The range of values of the wave vectors of the critical fluctuations was determined. Thus, in contrast to the case of low density of the point defects, when diffusion takes place from their higher-density region to the lower-density one, after n_* is reached a channel is opened for the formation of seeds of pores which is connected with the rising diffusion of the point defects in the inhomogeneous strain field.

We consider below the feasibility, in principle, of realizing, in the bulk of an irradiated massive crystal, an instability due to the influence of the strain field on the recombination and defect-drain processes. The elastic field of stresses alters the probabilities of these processes. If E is the energy barrier for recombination of one defect, then if a strain field $\varepsilon = \operatorname{div} U$ is produced in the medium via fluctuations, the barrier is lowered, at the proper sign of the amplitude by an amount $K\Omega_{am} \operatorname{div} U$, where Ω_{am} is the activation volume of the recombination process. A change of E causes a change of the recombination rates (γ and β_a). This leads to an additional change of the point-defect density. The result is the appearance of forces $F_a \sim \operatorname{grad} n_a$ that enhance the initial strain fluctuations, and positive feedback is produced. This instability mechanism takes place at low-temperature irradiation, when the processes that lead to diffusion of point defects are insignificant, while the dynamics of the change of the point-defect density is determined mainly by recombination and absorption by drains, the role of which can be assumed by grain boundaries, impurities, etc. To facilitate the analysis we neglect formation of point defect + impurity complexes and the change of the impurity density by irradiation.

The corresponding kinetic equations are

$$\begin{aligned} \frac{dn_v}{dt} &= g - \gamma n_i n_v - \beta_v n_v, \\ \frac{dn_i}{dt} &= g - \gamma n_i n_v - \beta_i n_i \end{aligned} \tag{5}$$

where g is the rate of generation of point defects by irradiation and is assumed constant, β_v and β_i are the rates of absorption by the drains, and γ is the rate of mutual recombination. The kinetic coefficients γ, $\beta_{v,i}$ are given by

$$\gamma = 4\pi R_0 D_v, \qquad \beta_{v,i} = \eta_{v,i} \rho_s D_{v,i} \tag{6}$$

where $D_{v,i} = D^0_{v,i} \exp\left(-\frac{E_{v,i}}{T}\right)$ are the diffusion coefficients, $E_{v,i}$ are the migration energies, ρ_s is the drain density, $\eta_{v,i}$ are the preference factors ($\eta_i > 1$, $\eta_i - 1 \ll 1$, $\eta_v = 1$), and R_0 is the radius of the mutual recombination of the point defects.

Mutual recombination of point defects and their absorption by drains are accompanied by a lattice deformation whose size is given by

$$\operatorname{div} U = n_i \Omega_{im} + n_v \Omega_{vm} + n_i n_v \Omega_{im} \Omega_{vm} \tag{7}$$

Here Ω_{vm}, Ω_{im} are the activation recombination volumes for vacancies and interstitial atoms, respectively ($\Omega_{vm} < 0$, $\Omega_{im} > 0$) [2].

The system (5)-(7) is nonlinear. The nonlinearity is due to mutual recombination γ of the defects and to absorption $\beta_{v,i}$ by drains, processes that depend on the strain in the medium. These are in fact the terms that lead to instability of the stationary state in the system.

The stationary nonequilibrium densities (n_v^0, n_i^0) satisfy the following relations:

$$g = \gamma(\epsilon_0) n_v^0 n_i^0 + \beta_v(\epsilon_0) n_v^0,$$

$$g = \gamma(\epsilon_0) n_v^0 n_i^0 + \beta_i(\epsilon_0) n_i^0,$$

$$\epsilon_0 = n_v^0 \Omega_{vm} + n_i^0 \Omega_{im} + n_i^0 n_v^0 \Omega_{vm} \Omega_{im}$$

where

$$\gamma_0 = \gamma(\epsilon_0) = 4\pi R_0 D_{0v} \exp\left(-\frac{E_v - K|\Omega_{vm}|\epsilon_0}{T}\right), \quad \beta_{v,i}^0 = \beta_{v,i}(\epsilon_0)$$

$$= \eta_{v,i} \rho_s D_{0v,i} \exp\left(-\frac{E_{v,i} \mp K|\Omega_{v,im}|\epsilon_0}{T}\right)$$

Let us show that in the system described by Eqs. (5)-(7) the stationary state $(n_{v,i}^0, \epsilon_0)$ is unstable. Assume that local fluctuations take place in the system and lead to perturbations $\delta n_{v,i}$ and $\delta\varepsilon$ in a certain region

$$n_{v,i} = n_{v,i}^0 + \delta n_{v,i}, \quad \epsilon = \epsilon_0 + \delta\epsilon, \quad |\delta n_{v,i}| \ll n_{v,i}^0, \quad |\delta\epsilon| \ll \epsilon_0,$$

$$\gamma(\epsilon) = \gamma_0 + \Gamma\delta\epsilon, \quad \beta_{v,i}(\epsilon) = \beta_{v,i}^0 + B_{v,i}\delta\epsilon, \quad \Gamma = \partial\gamma/\partial\epsilon_0,$$

$$B_{v,i} = \partial\beta_{v,i}/\partial\epsilon_0$$

Substituting these expressions in (5)-(7) and linearizing with respect to small perturbations of the form $\delta n_{v,i} \sim \exp \lambda t$, we obtain the following equation for the instability growth rate

$$\lambda^2 + a\lambda + b = 0,$$

$$a = \gamma_0 (n_v^0 + n_i^0) + \beta_v^0 + \beta_i^0 + \Omega_2 (\Gamma n_v^0 + B_i) n_i^0 - \Omega_1 (\Gamma n_i^0 + B_v) n_v^0,$$

$$b = [\gamma_0 n_i^0 + \beta_v^0 - \Omega_1 n_v^0 (\Gamma n_i^0 + B_v)] [\gamma_0 n_v^0 + \beta_i^0 - \Omega_2 n_i^0 (\Gamma n_v^0 + B_i)] -$$

$$- [\gamma_0 n_v^0 + \Omega_2 n_v^0 (\Gamma n_i^0 + B_v)] [\gamma_0 n_i^0 - \Omega_1 n_i^0 (\Gamma n_v^0 + B_i)],$$

$$\Omega_1 = \Omega_{vm} + n_i^0 \Omega_{im} \Omega_{vm}, \quad \Omega_2 = \Omega_{im} - n_v^0 \Omega_{im} \Omega_{vm}$$

The system becomes unstable if $a < 0$. It follows hence that the necessary condition for instability is a sufficiently rapid growth of the coefficients γ and β_v as ε increases. In fact, the onset of a strain field in the medium can stimulate the recombination of point defects by lowering the energy barrier. This change causes an exponential growth [see Eqs. (6)] of the recombination coefficients and hence an additional growth of the stress field in the medium. The result is a positive-feedback mechanism that leads to instability in the system.

Consider a system in which the density of the interstitial atoms and the lattice deformation that they produce is small. Then, neglecting in Eqs. (5)-(7) the mutual recombination of the defects ($\gamma \equiv 0$) and the strain associated with the interstices ($\Omega_{im} = 0$), we have:

$$\lambda = \beta_v^0 \left(\frac{K\Omega_{vm}^2 n_v^0}{T} - 1 \right) \exp\left(- \frac{K\Omega_{vm}^2 n_v^0}{T} \right)$$

Therefore $\lambda > 0$, if $n_v^0 > n_* = T/K\Omega_{vm}^2$. At typical values of the constants $K = 5 \cdot 10^{11}$ dyn/cm^2, $\Omega_{vm} \approx 10^{-23}$ cm^3, and $T = 300°K$ we obtain for the critical vacancy density the estimate $n_* \sim 10^{19}$ cm^{-3}, which is smaller by several orders than the atom density in the matrix; this attests to feasibility of realizing this instability mechanism.

The recombination–strain instability phenomenon must be taken into account in the analysis of the mechanism and in the estimate of the criterion for material failures due to increase of elastic stresses in a medium, with an aim at a more reliable prediction of the behavior of various construction materials and of tools on their basis, operating when exposed to laser emission and to other high-intensity radiation.

4. FORMATION OF PERIODIC ANNULAR DEFECT STRUCTURES IN FILMS

We investigate in this section the mechanism and the conditions for the onset of periodic structures of defects in thin films. Consider a crystal film of thickness h, deposited on a disk-shaped dielectric substrate. The substrate is heated by a focused cw CO_2 laser (Fig. 1). We assume that the absorption coefficient is so high that the substrate becomes optically transparent. The temperature field produced on the substrate surface by the laser emission is assumed to be stationary and bell-shaped. Let the film contain defects of density n(r, t). Since the film is weakly bound to the substrate, it can undergo flexural deformation. The initial homogeneous distribution of the defects is then disturbed, diffusion and drift fluxes are produced, and some of the defects are absorbed by drains. Equation (1) takes in this case the form

$$\frac{\partial n_a}{\partial t} = \operatorname{div} \left[D_a \operatorname{grad} n_a + \frac{D_a \Omega_a K h}{2T} \operatorname{grad} (\Delta_r \epsilon) \right] - \beta_a n_a \qquad (8)$$

where the second term in the square brackets describes the defect drift due to the influence of the field of the inhomogeneous flexural strain $\xi(r, t)$, $r = (x, y)$ is the radius vector $\Delta_r = \partial^2/\partial x^2 + \partial^2/\partial y^2$, and ξ is the flexural deformation of the film and is indicative of the shift, along the z axis, of points located on the neutral plane ($z = 0$). Assuming that the surface energy of the film bordering on the vacuum is much higher than the surface energy of the substrate bordering on the vacuum, the film can be regarded as free, and to describe dynamics of the field of the flexural deformation in the film we can use in place of (3) the following equation [3]:

$$h\rho \frac{\partial^2 \xi}{\partial t^2} = - D_\xi \Delta_r^2 \xi - D_\xi \beta_a \Delta_r^2 \xi^3 + h\sigma_T \Delta_r \xi + \sigma_t - \sigma_n \qquad (9)$$

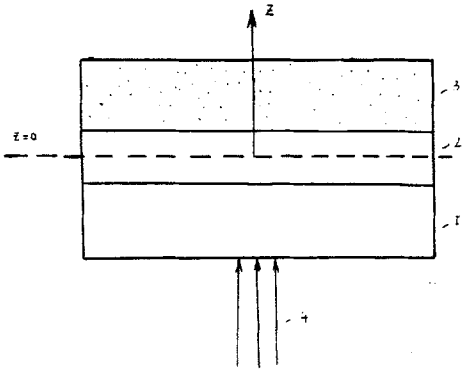

Fig. 1. Experimental setup: 1) substrate, 2) film, 3) vacuum, 4) laser beam.

where ρ is the film density, $D_\xi = E_0 h^3/12(1-\sigma^2)$ is the flexural rigidity, E is Young's modulus, σ is the Poisson coefficient, and σ_T is the stress along the film and is due to the axisymmetric temperature gradient produced on the substrate surface by the laser radiation: $\sigma_T \approx (1-\sigma)\cdot Ka_T(T-T_\infty)$ [3], a_T is the film's thermal-expansion coefficient, T_∞ is the surface temperature as $r \to \infty$, and σ_t takes into account the stress that clamps the film to the substrate. It is due to the differences between the film and substrate lattice parameters. The last term of (9) determines the concentration stresses due to the difference between the covalent radii of the defects and of the matrix atoms. Accordingly to [5] we have for σ_n the expression

$$\sigma_n = K \sum_{a=i,v} \Omega_a n_a$$

The summation here is over all the point defects that produce the strain. The foregoing set of equations is closed and nonlinear. The nonlinearity is due to the terms that describe the interaction of the defects with the flexural-deformation field and depend in turn on the defect density. Note that it is precisely this term which leads to the onset of instability of the uniform distribution of the defects.

a) Investigation of Stability in a Film with One Type of Defect. If only one type of defect is present (small-radius impurities or vacancies), Eqs. (8) and (9) take the form:

$$\frac{\partial n}{\partial t} = \operatorname{div}\left(D \operatorname{grad} n - \frac{DK|\Omega_v|hn}{2T}\Delta\xi\right) - \beta n + g,$$
$$h\rho \frac{\partial^2 \xi}{\partial t^2} = -D_\xi \Delta_r^2 \xi - D_\xi \beta_a \Delta_r^2 \xi^2 + h\sigma_T \Delta\xi + \sigma_t - K|\Omega_v|n \tag{10}$$

To investigate the stability of the system (10) to small perturbations, we put

$$n(r,t) = n_0 + \delta n(r,t), \quad \xi(r,t) = \xi_0 + \delta\xi(r,t)$$
$$|\delta n(r,t)| \ll n_0, \quad |\delta\epsilon(r,t)| \ll \xi_0 \tag{11}$$

where n_0 and ξ_0 are the average homogeneous values of n and ξ while δn and $\delta\xi$ are small increments. After substituting (11) in the system (10) and linearlizing the latter we arrive at the system of equations:

$$\frac{\partial}{\partial t}(\delta n) = D\Delta(\delta n) - \frac{DK|\Omega|hn_0}{2T}\Delta^2\delta\xi - \beta_v \delta n,$$
$$h\rho \frac{\partial^2}{\partial t^2}(\delta\xi) = -D_\xi \Delta^2(\delta\xi) + h\sigma_T \Delta(\delta\xi) - K|\Omega|\delta n \tag{12}$$

The coefficients D, D_ξ, σ_T, ρ are in general variable, since they depend on the inhomogeneous temperature field on the substrate surface. If, however, it is assumed that the characteristic spatial scale over which the temperature changes substantially is much larger than the characteristic dimension of the diffusion zone, the coordinate dependence of the coefficients of the system (12) can be neglected and we can put in (12) D, D_ξ, σT, $\rho = \text{const}$. Under these conditions the solution of the set (12) can be sought in the form:

$$\delta n(r,t) = N \exp(\lambda t) J_0(qr), \quad \delta\xi(r,t) = M \exp(\lambda t) J_0(qr) \tag{13}$$

where $J_0(qr)$ is a Bessel function of zero order, q is the wave vector of the perturbations, N and M are constants, and λ is the instability growth rate. The form of the small deviations (13) suggests that we are investigating the stability of the homogeneous solution of the system (10) against formation of a periodic structure of the density of defects arranged in the form of concentric rings. Substituting (13) in (12) and equating to zero the determinant of the resultant set of equations we obtain, taking into account the adiabaticity of the instability growth rate, the expression

$$\lambda(q^2) = -Dq^2\left(1 - \frac{m}{l^2q^2+1}\right) - \beta,\ m = \frac{n_0K^2\Omega^2}{T\sigma_T},\ l^2 = \frac{D_\xi}{h\sigma_T} \tag{14}$$

It follows from (13) that when the condition $m < 1$ is met, i.e.,

$$n_0 > n_* = \frac{T\sigma_T}{K^2\Omega^2} \tag{15}$$

there exists an interval of wave-vector values $q^2_{c(+)} < q^2 < q^2_{c(-)}$, for which the homogeneous state loses stability. This gives rise to a periodic structure of defects, in the form of concentric rings. The wave vectors of the critical fluctuations are given by

$$q^2_{c(\pm)} = \frac{\left(m - 1 - \frac{\beta l^2}{D}\right) \pm \sqrt{\left(m - 1 - \frac{\beta l^2}{D}\right)^2 - \frac{4\beta l^2}{D}}}{2l^2}$$

It is easily seen that $\lambda(q^2)$ increases in the interval $q^2_{c(+)} < q^2 < q^2_{c(-)}$ and that $\lambda(q^2_{c(\pm)}) = 0$. The dependence has therefore a maximum. At the maximum point

$$q^2_{max} = \frac{1}{l^2}(\sqrt{m} - 1),$$

$$\lambda_{max} = \frac{D}{l^2}(m + 1 - 2\sqrt{m}) - \beta \tag{16}$$

Analysis of expressions (14)-(16) shows that the cause of the periodic structure of the defects is their drift caused by the flexural deformation, which causes $\lambda(q^2)$ to take the form of a curve with a sharp maximum (Fig. 2). Inhomogeneous fluctuations are then produced against the background of the homogeneous fluctuations ($q \equiv 0$) having wave vectors close to $\sim q_{max}$ and increasing at a rate $\sim\lambda_{max}$ exceeding by several orders the growth rate of the homogeneous fluctuations. Under these conditions there is realized in the point-defect system a periodic annular structure with a spatial characteristic determined by q_{max}: $d_0 \approx 2\pi/|q|_{max}$. To analyze the suitability of the considered mechanism we make some numerical estimates. Assuming $\Omega \sim 10^{-23}$ cm^3, $K = 5\cdot10^{11}$ dyn/cm^2, $a_T = 10^{-5}$ deg^{-1}, and $T = 500$°C, we get $n_* = 8\cdot10^{18}$ cm^{-3}. This density is much lower than atomic, attesting to the practical realizability of this approach. For the period of the resultant structure we obtain the estimate $d_0 \sim 4\cdot10^{-3}$ cm.

b) Investigation of Stability in a System with Defects of Two Types. It is of interest to consider a situation in which the system contains two types of defect (impurity atoms of large and small radius, or Frenkel' pairs) with densities n_j ($j = 1, 2$). The corresponding system of equations is

$$\frac{\partial n_j}{\partial t} = D_j\Delta n_j - \frac{D_j\Omega_j n_j K h}{2T}\ \Delta^2\xi - \beta_j n_j - \gamma n_1 n_2 + g,$$
$$\rho h\frac{\partial^2\xi}{\partial t^2} = -D_\xi\Delta^2\xi - D_\xi\beta_a\Delta_r^2\xi^3 + h\sigma_T\Delta_r\xi - K|\Omega_1|n_1 + K|\Omega_2|n_2 \tag{17}$$

where γ is the rate of recombination of the vacancies and interstitial atoms. In the case of impurities we have $\gamma = 0$, $\beta_j = 0$, $g = 0$.

To investigate the stability of this system, we use the solutions in the form (13), and obtain after simple transformations

$$\lambda_{1,2}(q^2) = -\frac{a_q}{2} \pm \sqrt{\frac{a_q^2}{4} - b_q},$$

$$a_q = (n_1^0 + n_2^0)\gamma + \beta_1^0 + \beta_2^0 + D_1q^2\left(1 - \frac{m_1n_1^0}{l^2q^2+1}\right) + D_2q^2\left(1 - \frac{m_2n_2^0}{l^2q^2+1}\right), \tag{18}$$

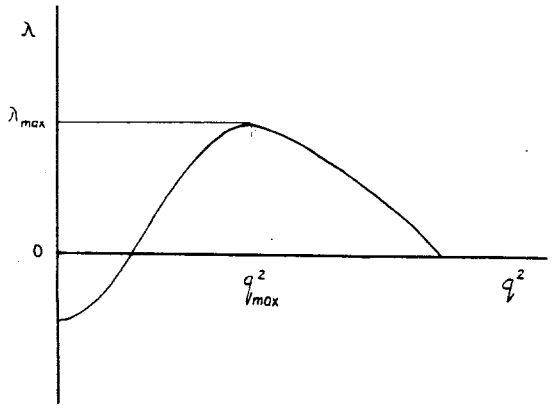

Fig. 2. Qualitative dependence of the instability growth rate on the square of the wave vector.

$$b_q = [D_\perp q^2 (1 - \frac{m_1 n_1^0}{l^2 q^2 + 1}) + \beta_1 + \gamma n_1^0][D_2 q^2 (1 - \frac{m_2 n_2^0}{l^2 q^2 + 1}) + \beta_2 + \gamma n_2^0] - (\gamma n_1^0 + D_1 q^2 \frac{m_{12} n_1^0}{l^2 q^2 + 1})(\gamma n_2^0 + D_2 q^2 \frac{m_{12} n_2^0}{l^2 q^2 + 1}),$$

$$m_{12} = \frac{K|\Omega_1 \Omega_2|}{T\sigma_T}, \quad m_j = \frac{K^2 \Omega_j^2}{T\sigma_T}, \quad j = 1, 2.$$

where n_1^0, n_2^0 are the densities satisfying the homogeneous and stationary equations

$$g = \gamma n_1^0 n_2^0 + \beta_1 n_1^0, \quad g = \gamma n_1^0 n_2^0 + \beta_2 n_2^0.$$

Let us consider a case when the parameters of both components coincide: $D_1 = D_2 = D$, $\beta_1 = \beta_2 = \beta$, $n_1^0 = n_2^0 = n_0$. We obtain then from (13):

$$\lambda_{1,2}(q^2) = \begin{cases} -q^2 D(1 - \frac{2mn_0}{l^2 q^2 + 1}) - \beta \\ \\ -q^2 D - \beta - 2\gamma n_0 \end{cases} \tag{19}$$

It follows from (19) that instability sets in when

$$n_0 > n_* = \frac{T\sigma_T}{2K^2\Omega^2}$$

The wave vectors of the critical fluctuations are determined in this case from the condition $b_{q^2} = 0$.

c) Elimination of Instability in a System of Defects. The ensuing instability of a homogeneous distribution of a system of point defects is eliminated by the anharmonicity of the elastic continuum. We confine ourselves for simplicity to a system with one type of defect, say, vacancies. The solution of the nonlinear system of equations (10) can be represented, in the approximation in which the vacancy density and the strain field are stationary, by a superposition of plane waves:

$$n(r) = n_0 + \sum_q n_q \exp(iqr)$$

$$\xi(r) = \xi_0 + \sum_q \xi_q \exp(iqr) \tag{20}$$

where n_q, ξ_q are the amplitudes of the unstable modes. Substituting these solutions in the system (10) and solving the latter, we obtain the following expressions for the stationary Fourier amplitudes:

$$n_q = \frac{K|\Omega|hn_0 q^2}{T\sqrt{\beta_a}}\sqrt{\frac{q^2}{q_0^2} - 1}\,, \qquad (21)$$

$$\xi_q = \pm\frac{1}{\sqrt{\beta_a}}\sqrt{\frac{q^2}{q_0^2} - 1}$$

where $q_0^2 = \frac{h\sigma_T}{D_\xi}\left(\frac{K^2\Omega^2 n_0}{2T\sigma_T} - 1\right)$. Note that in the derivation of (21) we have neglected, for simplicity, the recombination terms in (10) ($\beta \equiv 0$). Substituting (21) in (20) and summing over q, we obtain for n(r) and ξ(r)

$$n(r) \approx AJ_1(q_0 r),\ \xi(r) \approx BJ_1(q_0 r) \qquad (22)$$

where A and B are certain constants. It follows from (22) that the concentric spatial distribution of the defects is the cause of the concentric distribution of the strain field. The impurity atoms themselves, piling up in the compression region, deform the film and maintain thus the initial inhomogeneous fluctuations of the strain field.

Note that the theory developed above differs from the model [6] used to analyze the produced periodic structures, since it takes into account in a unified manner the dynamics of the temperature, strain, and density fields.

It explains qualitatively a number of experimental facts. Thus, for example, annular concentric flaking of the film was observed [7] in the course of laser deposition of polycrystalline molybdenum films from the gas phase. According to the theory set forth here, this phenomenon can be interpreted in the following manner. When fluctuating flexural deformation sets in, there appear in the film periodically repeating tension and compression regions. The inhomogeneous field of the flexural strain, interacting with the vacancies produced as the film grows, produces under certain conditions a vacancy drift directed counter to the diffusion in the vacancy system. In this case the compression regions attract the vacancies, while the tension regions repel them. Becoming localized in the compression region, the vacancies themselves deform the film, enhancing thereby the initial fluctuations of the strain. The ensuing instability leads to formation, in the vacancy localization regions, of a high supersaturation sufficient for pore nuclei to appear and grow. The result is a periodic annular pore structure in the film. Concentric annular flaking of the film takes place at the locations of the pore accumulations. Calculation using Eq. (15) shows that the critical density of the vacancies is usually reached in the temperature interval 200-400°C, in accord with the experimental data.

The appearance of a vacancy drift flux in the compression region of the film produces a drift of site atoms in the opposite direction, and a flux, in the same direction, of interstitial atoms in the tension region (if the diffusion mechanism constitutes crowding an atom out of a site into an interstice). Assume that the film contains substitutional impurities of two types, with different diffusion activation energies satisfying the inequality $E_{a1} < E_a < E_{a2}$, where E_a is the migration energy of the impurities of the lattice proper. It is then obvious that in the course of the diffusion the more mobile impurity atoms (with activation energy E_{a2}) will be gathered in compression regions, and the less mobile ones in tension regions. As a result, laser irradiation will rid the center of the film of impurities. It is important to emphasize here that the characteristic times needed for observation of the laser "cleanup" effect is only a few seconds, and sufficiently intense laser action can shorten this time by three or more orders. The relatively low content of impurities (mainly carbon and oxygen, whose concentration is not higher than 0.01%) in molybdenum films obtained by laser deposition from the gas phase [7], can apparently be explained within the framework of the mechanism described here. It must also be emphasized that purification by the mechanism proposed in the present paper produces no local damage in the sample in a wide range of irradiation conditions, so that this method is quite valuable for practical use when pure atomic samples are needed.

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CORRELATED STATES IN QUANTUM ELECTRONICS (RESONANT CIRCUIT)

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Coherent and correlated states of a Josephson junction are constructed. Quantum current and voltage noises are calculated. The influence of an external current and of parametric buildup on the Josephson junction is discussed. The feasibility in principle of exciting correlated and coherent states in a Josephson junction is suggested.

The feasibility of using a Josephson junction (simulated by a quantum resonant circuit) to generate squeezed electromagnetic radiation was considered in [1].

Our purpose here is, by using the analogy between a Josephson junction and a quantum resonant circuit, to demonstrate the possible existence of a new Josephson-junction state – a correlated state – and to suggest the theoretical feasibility of exciting a correlated state by a parametric action that can lead effectively to a temporal variation of, say, the critical current of the junction.

We carry out the analysis within the context of the Hamiltonian [2]

$$\hat{H} = \frac{\hat{Q}^2}{2C} + \frac{\hbar I_C}{2e}(1-\cos\varphi) - \frac{\hbar}{2e} I(t)\hat{\varphi}. \tag{1}$$

Here C is the capacitance and I_C the critical current of the junction, e the electron charge, $\hbar$ Planck's constant, $\hat{Q}$ the charge operator, $\hat{\varphi}$ the phase operator, and I(t) the external current fed to the junction. The problem is investigated here in the region of small values of the phase φ, when the term $(1-\cos\hat{\varphi})$ in the Hamiltonian (1) can be replaced by the quadratic expression $\frac{\hat{\varphi}^2}{2}$; conditions for this have been discussed in [3]. The Hamiltonian (1) is thus reduced to the Hamiltonian of a quantum resonant circuit

$$\hat{H} = \frac{\hat{Q}^2}{2C} + \frac{\hbar I_C}{2e}\,\frac{\hat{\varphi}^2}{2} - \frac{\hbar}{2e} I(t)\hat{\varphi}. \tag{2}$$

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