

Possibility of plastic deformation of an ionic crystal due to the nonthermal influence of a high-frequency electric field

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The possibility of macroscopic mass transfer in an ionic crystal placed in a high-frequency electric field has been studied theoretically. The plastic deformation of the crystal is shown to take place due to the vacancy-drift perturbations caused by space-charge formation in the near-surface layer and by surface permeability asymmetry with respect to the direction of the motion of atoms through it. Estimates prove that the effects considered can be essential in technological processes, e.g., in microwave sintering of ceramics.

I. INTRODUCTION

Considerable recent attention has been focused on the development of technologies that use microwave radiation, microwave sintering of ceramics being among them. The analysis of the available data (e.g., Ref. 1) permits one to reveal certain specific features of microwave sintering of powder materials. Many of the observed distinctions from traditional sintering processes cannot be presented as simple consequences of the difference in the temperature fields in the conditions of internal (volumetric) and external heating. In this connection possible mechanisms of the nonthermal influence of the high-frequency (HF) electromagnetic field on the diffusion-controlled viscous processes in crystalline bodies are discussed in the literature (e.g., Refs. 2 and 3). In this paper one of these "hypothetical" mechanisms is considered, dealing with the drift motion of vacancies in the ionic crystal.

According to contemporary notions, plastic deformation caused by external forces (or the so-called creep flow) of crystals is associated with diffusion transfer of atoms in the crystal lattice.^{4,5} Assuming that vacancy mechanism of diffusion predominates in crystals, we shall consider further processes of vacancy transfer. This is more convenient because the ideal gas approach can be used for the vacancies due to their small concentration in the lattice. In this case the crystal surface shift velocity, \mathbf{V} , that we are finally interested in can be connected with the vacancy flux density, \mathbf{J} , on the surface:

$$\mathbf{V} = -\mathbf{n} \cdot (\mathbf{n} \cdot \mathbf{J}_S). \quad (1)$$

Here \mathbf{J}_S is the value of \mathbf{J} on the solid body surface and \mathbf{n} is the outward unit normal to the surface.

In the absence of the electric field the macroscopic vacancy flux is caused by the diffusion:

$$\mathbf{J} = -D\nabla N, \quad (2)$$

where N is concentration, D is the diffusion coefficient of vacancies. In a crystal with no vacancy sources and sinks in its volume the flux is completely defined by the

stress distribution over the solid body surface, where the following relation is valid:⁵

$$\beta(N_S - N_{0S}) = (\mathbf{n} \cdot \mathbf{J}_S), \quad (3)$$

$$N_{0S} = N_0(T) \exp(\mathbf{n} \cdot \hat{\sigma}_s \cdot \mathbf{n} \omega / kT). \quad (4)$$

Here β is kinetic coefficient that determines the surface permeability for vacancies, N_S is vacancy concentration on the surface, N_{0S} is the equilibrium value of the concentration on the surface, $N_0(T)$ is the temperature-dependent bulk equilibrium value of vacancy concentration, $\hat{\sigma}_s$ is the stress tensor on the surface, and ω is the vacancy volume.

Specifically, the values of vacancy oversaturation, $\delta N_S \equiv N_S - N_0$, and flux density, $(\mathbf{n} \cdot \mathbf{J}_S)$, at the surface of a crystalline specimen with the characteristic size Λ , experiencing relatively small shearing stress ($\sigma_s \equiv \mathbf{n} \cdot \hat{\sigma}_s \cdot \mathbf{n}$, $|\sigma_s| \ll kT/\omega$), can be estimated respectively as follows:

$$|\delta N_S| \sim \frac{\beta \Lambda}{\beta \Lambda + D} N_0 \frac{|\sigma_s| \omega}{kT}, \quad (5)$$

$$|(\mathbf{n} \cdot \mathbf{J}_S)| \sim \frac{\beta D}{\beta \Lambda + D} N_0 \frac{|\sigma_s| \omega}{kT}.$$

The distinguishing feature of vacancies in the ionic crystal is that they have effective electric charges, e . Therefore vacancy drift in the electric field, \mathbf{E} , makes an additional contribution into the flux of vacancies of each sort (here the Nernst-Einstein relation for mobility is used):

$$\mathbf{J} = -D\nabla N + DN \frac{e\mathbf{E}}{kT}. \quad (6)$$

The drift part of the vacancy flux exceeds the characteristic diffusion value (5) essentially in comparatively weak fields. For example, if $\Lambda = 1 \mu\text{m}$ and $\sigma = 10^6 \text{ Pa}$, these being typical sizes of grains used in ceramic compositions and value of the capillary stresses existing therein, the second term in (6) equals the first when the electric field is only about 3 V/cm.

The vacancy drift in ionic crystal leads mainly to the occurrence of electric current. But in certain cases the

mass transfer, i.e., the macroscopic motion of matter, is also possible. In the static electric field such mass transfer was observed experimentally.⁶ According to the investigation undertaken in Ref. 7, this effect occurs when diffusion coefficients of positive and negative vacancies are different, and the so-called surface vacancy diffusion in a thin near-surface amorphous layer is essential.

In the first approximation, the vacancy drift in the HF electric field is oscillatory. Therefore a matter flux not vanishing upon averaging can exist only due to various nonlinear effects. Among them there are vacancy drift perturbations caused by space-charge formation in the near-surface layer of crystal and by the dependence of the surface permeability coefficient, β , upon the vacancy flux direction. The analysis of the possible role of these effects is presented below.

It should be mentioned that we have no adequate experimental data permitting to state unambiguously that β depends on the vacancy flux direction. However, the existence of such dependence can be supposed for the reason that the bulk vacancy generation energy differs from its generation energy on the surface; on the other hand, it will be shown that this dependence can influence significantly the mass transfer in a crystal placed in the HF field.

II. FORMULATION OF THE PROBLEM

In the absence of sources and sinks of vacancies in the volume of crystal the variation of their concentration in time is determined by Fick's equation:

$$\frac{\partial N_\alpha}{\partial t} + (\nabla \cdot \mathbf{J}_\alpha) = 0. \quad (7)$$

From here on the quantities with the subscript α will correspond to the vacancies of sort α .

The flux density of vacancies of sort α is determined as in (6) by diffusion and drift processes:

$$\mathbf{J}_\alpha = -D_\alpha \nabla N_\alpha + D_\alpha N_\alpha \frac{e_\alpha \mathbf{E}}{kT}, \quad (8)$$

where $\mathbf{E} = \mathcal{E} - \nabla\varphi$, \mathcal{E} is the HF field vector, and φ is the slowly varying charge separation field potential. Considering the small-sized crystalline body deformation (in particular, the deformation of discrete grains during the ceramic sintering), we shall assume that the HF electric field is quasistatic, i.e.,

$$\begin{aligned} \nabla \times \mathbf{E} &= \mathbf{0}, \\ (\nabla \cdot \mathbf{E}) &= \frac{4\pi}{\omega\epsilon} \sum e_\alpha N_\alpha, \end{aligned} \quad (9)$$

where ϵ is lattice dielectric constant, and the summation is over the sorts of particles. We shall restrict our consideration to a crystal having two sorts of vacancies with the charges $e_\alpha = \pm e$ and with the same equilibrium concentrations N_0 . In this case, for the unambiguous determination of vacancy fluxes it is sufficient to supplement Eqs. (7)–(9) with the boundary conditions⁸ [cf. (3)

and (4)]

$$\mathbf{n} \cdot \sum e_\alpha \mathbf{J}_{\alpha S} = 0, \quad (10)$$

$$\mathbf{n} \cdot \sum \mathbf{J}_{\alpha S} = \beta \sum \left(N_{\alpha S} - N_0 - N_0 \frac{\sigma_s \omega}{kT} \right) \quad (11)$$

[Eq. (10) means that there is no electric current through the solid body surface] and to specify the normal component of the HF electric field, \mathcal{E}_S , and stress, σ_s , at the surface ($|\sigma_s| \ll kT/\omega$).

III. PERTURBATION METHOD

If the electric field is not too strong (the estimations show that the effects of interest to us become essential in the fields that do not perturb the vacancy concentration significantly), the vacancy concentration perturbations caused by it are small, so we can solve Eqs. (7)–(9) with the help of the perturbation method representing the concentration as

$$N_\alpha = N_0 + \nu_\alpha + \tilde{\nu}_\alpha + \eta_\alpha, \quad (12)$$

where ν_α and $\tilde{\nu}_\alpha$ are the HF perturbations of first and second order, $N_0 \gg |\nu_\alpha| \gg |\tilde{\nu}_\alpha|$, and η_α is a slowly varying perturbation, $N_0 \gg |\eta_\alpha|$. In this case we can obtain from (7)–(11) by the first-order perturbation theory that

$$\frac{\partial \nu_\alpha}{\partial t} - D_\alpha \Delta \nu_\alpha + D_\alpha N_0 \frac{4\pi e_\alpha}{\omega \epsilon kT} \sum e_\beta \nu_\beta = 0, \quad (13)$$

$$(\nabla \cdot \mathcal{E}) = \frac{4\pi}{\omega \epsilon} \sum e_\alpha \nu_\alpha. \quad (14)$$

In a quasimonochromatic electric field the concentration perturbations ν_α oscillate with the field frequency, Ω . However, in the second order of the perturbation theory the HF field also influences the quasistationary vacancy fluxes that are of chief interest to us. Averaging Eqs. (7)–(9) over the period of the HF field, we find that the quasistationary vacancy fluxes are described by the following equations:

$$\begin{aligned} (\nabla \cdot \mathbf{j}_\alpha) &= 0, \\ \mathbf{j}_\alpha + \frac{D_\alpha}{kT} \nabla \mu_\alpha &= \frac{D_\alpha e_\alpha}{kT} \langle \nu_\alpha \mathcal{E} \rangle, \end{aligned} \quad (15)$$

where \mathbf{j}_α is a quasistationary vacancy flux density, $\mu_\alpha = kT\eta_\alpha + N_0 e_\alpha \varphi$ is the chemical potential of vacancies of the sort α , and brackets $\langle \rangle$ denote averaging over the period of the HF field. Respectively, the boundary conditions (10) and (11), after averaging, rearrange as

$$\mathbf{n} \cdot \sum e_\alpha \mathbf{j}_{\alpha S} = 0, \quad (16)$$

$$\mathbf{n} \cdot \sum \mathbf{j}_{\alpha S} - \beta_0 \sum \mu_{\alpha S} = -\beta_0 N_0 \frac{\sigma_s \omega}{kT} + \left\langle \beta_1 \sum \nu_{\alpha S} \right\rangle, \quad (17)$$

where $\beta_0 = \langle \beta \rangle$, $\beta_1 = \beta - \beta_0$.

The analysis of Eqs. (13) with the relevant boundary conditions shows that ν_α being harmonically dependent on time are localized within a thin near-surface layer. Its thickness is expressed by some combination of the Debye-Huckel radius $\lambda = \sqrt{\frac{\epsilon\omega kT}{8\pi e^2 N_0}}$ and the characteristic diffusion length $l = \sqrt{D/\Omega}$. (It should be mentioned that in this layer the space charge oscillating with the frequency Ω is localized, and when the values of λ and l are essentially different, the layer thickness is defined by the smallest of them.) Within this layer, the normal components of quasistationary vacancy flux densities can be considered unchanging, whereas the chemical potentials, μ_α , have finite alterations, $\delta\mu_\alpha$, in accordance with Eqs. (15):

$$\delta\mu_\alpha = \mu_{\alpha s} - \tilde{\mu}_{\alpha s} \approx \int e_\alpha \langle \nu_\alpha \mathbf{n} \cdot \mathcal{E} \rangle dx. \quad (18)$$

Here x is the coordinate counted along the outward normal to the surface, the integration is over the HF space-charge layer, and $\tilde{\mu}_{\alpha s}$ denote the values of μ_α close to the surface but beyond this layer. Hence, in the case being considered the quasistationary vacancy fluxes problem can be reduced to the solution of the uniform equations (15) beyond the HF space-charge area, with the boundary conditions (16) and (17) being transformed in an appropriate manner:

$$\mathbf{n} \cdot \sum e_\alpha \tilde{\mathbf{j}}_{\alpha s} = 0, \quad (19)$$

$$\mathbf{n} \cdot \sum \tilde{\mathbf{j}}_{\alpha s} - \beta_0 \sum \tilde{\mu}_{\alpha s} = -\beta_0 N_0 \frac{\sigma_s \omega}{kT} + \beta_0 \frac{p\omega}{kT} + \langle \beta_1 \sum \nu_{\alpha s} \rangle. \quad (20)$$

Here $\tilde{\mathbf{j}}_{\alpha s}$ are the quasistationary vacancy flux densities at the inner boundary of the HF space charge layer and p is the effective pressure of the HF field on the vacancies in this layer:

$$p = \int \frac{1}{\omega} \langle \sum e_\alpha \nu_\alpha \mathbf{n} \cdot \mathcal{E} \rangle dx.$$

Assuming that in a thin layer

$$\sum \nu_{\alpha s} = \left[N_0 \frac{e\mathcal{E}_{S0}\lambda}{kT} \frac{(\kappa_2^2 - \kappa_1^2)e^{i\Omega t}}{(h_1 + h_2)\sqrt{\kappa_1^2\kappa_2^2 - i(\kappa_1^2 + \kappa_2^2)}} + \text{c.c.} \right] / 2, \quad (24)$$

$$\tilde{\mathcal{E}}_S = \left[\mathcal{E}_{S0} \frac{2i\kappa_1^2\kappa_2^2 e^{i\Omega t}}{\kappa_1^2 + \kappa_2^2 + 2i\kappa_1^2\kappa_2^2} + \text{c.c.} \right] / 2, \quad (25)$$

where

$$\kappa_\alpha^2 = \Omega\lambda^2/D_\alpha,$$

$$h_{1,2}^2 = \left[1 + i(\kappa_1^2 + \kappa_2^2) \pm \sqrt{1 - (\kappa_1^2 - \kappa_2^2)^2} \right] / 2, \quad \text{Re}h_{1,2} > 0. \quad (26)$$

$$(\nabla \cdot \mathcal{E}) \approx \frac{\partial}{\partial x} (\mathbf{n} \cdot \mathcal{E}),$$

we can present p in the conventional form of the field pressure:

$$p = \frac{1}{8\pi} (\langle \mathcal{E}_S^2 \rangle - \langle \tilde{\mathcal{E}}_S^2 \rangle), \quad (21)$$

where $\tilde{\mathcal{E}}_S$ is the value of the normal component of the HF electric field at the inner boundary of the HF space-charge layer.

It can be seen from (20) that the quasistationary vacancy fluxes and hence the plastic deformation of the crystal are controlled by three factors: (1) the external stresses [described by the first term in the right part of (20)]; (2) the HF field ponderomotive effect on the vacancies in the thin near-surface layer (the second term); and (3) the surface permeability asymmetry with respect to the vacancy motion direction (the last term). These effects are additive thus permitting one to carry on the analysis of the relative role of each. It is interesting to note that there is no small factor N_0 in the second term on the right part of (20), unlike the first term. It means that the plastic deformations of a crystal in the HF field can be essential even when the field pressure is much smaller than the mechanical stresses.

To estimate the HF field influence on the mass transfer processes in a crystal it is necessary [see (20)] to find the effective field pressure, p , and total vacancy concentration perturbation at the surface, $\sum \nu_{\alpha s}$, using the first-order perturbation theory equations (13) and (14) with the appropriate boundary conditions. If the field frequency is high enough, $D_\alpha\Omega \gg \beta^2$, the HF vacancy fluxes through the crystal surface can be neglected, and the boundary conditions can be written in the following form:

$$D_\alpha \left(\mathbf{n} \cdot \nabla \nu_\alpha \Big|_S - N_0 \frac{e_\alpha \mathcal{E}_S}{kT} \right) = 0. \quad (22)$$

Within the framework of this approach the solution of Eqs. (13) and (14) can be presented in sufficiently simple form, if the HF space-charge layer is considered as a one-dimensional structure. In particular, taking the normal component of the electric field at the surface as

$$\mathcal{E}_S = (\mathcal{E}_{S0} e^{i\Omega t} + \text{c.c.}) / 2, \quad (23)$$

we have

IV. DISCUSSION

As stated above, we distinguish two mechanisms of the HF field influence on the mass transfer processes in a limited crystal. In this section we shall analyze the role of each separately, considering their dependence on such factors as the frequency and amplitude of the external field, concentration, and diffusion coefficients of vacancies, size, and structure of the crystal.

According to (25) and (26), the amplitude of the normal component of the electric field in the space-charge layer decreases with depth, i.e., the effective field pressure on vacancies p (21) is positive. On the other hand, $p \propto |\mathcal{E}_{S0}|^2$. So the vacancy concentration is lower at those parts of the surface where the amplitude of the normal component of the HF field is higher. Therefore, there exists a quasistationary vacancy flux to these areas from those having a smaller amplitude of the normal component of the HF field. Respectively, an oppositely directed macroscopic atom flux takes place, from the areas with the strong field into the areas where the field is weaker. In other words, an ionic crystal of quasispherical shape placed into a uniform external electric field will experience, as a result of the ponderomotive effect, a plastic deformation, flattening in the direction of the external electric field vector. The crystal can be deformed in a similar manner under a monoaxial mechanical stress. This being so, it is convenient to characterize the ponderomotive effect by the value of the equivalent external stress, σ_E , that leads to the same plastic deformation velocity [cf. (20)]:

$$\sigma_E = p/N_0. \quad (27)$$

To analyze the dependence of σ_E upon the crystal properties and the field frequency it is useful to rewrite the expressions (25) and (26) in the following form:

$$\tilde{\mathcal{E}}_S = \left[\frac{\mathcal{E}_{S0} e^{i\Omega t}}{1 + 4\pi G_V / (i\Omega\epsilon)} + \text{c.c.} \right] / 2, \quad (28)$$

where $G_V = N_0 e^2 \sum D_\alpha / \omega kT$ is the crystal conductivity associated with the vacancy motion. This form shows that the spatial distribution of the HF electric field can be calculated by standard methods of continuous media electrodynamics everywhere except the space-charge layer, its thickness being equal to zero in the frame of this description. In particular, it gives the opportunity to account in a comparatively simple way the influence of the amorphized layer that exists at the surface of a real crystal, where vacancy concentration, N_0 , diffusion coefficients, and hence the conductivity associated with them, G_S , are considerably higher than the bulk values.⁹ For instance, if the amorphized layer thickness, a , is great in comparison with the thickness of the space-charge layer, this being usual for dielectrics, and the crystal has a ball-like shape with the radius R , then \mathcal{E}_{S0} varies from $\frac{3}{2+\epsilon}(\mathbf{n} \cdot \mathcal{E}_0)$ at high frequencies [$\Omega \gg 4\pi G_V / \epsilon$, $4\pi(a/R)G_S / \epsilon$] to $\frac{3}{\epsilon}(\mathbf{n} \cdot \mathcal{E}_0)$ at low frequencies ($\Omega \ll \max[4\pi G_V / \epsilon, 4\pi(a/R)G_S / \epsilon]$) [here \mathcal{E}_0 characterizes the external electric field that equals $(\mathcal{E}_0 e^{i\Omega t} + \text{c.c.})/2$]. In this case G_V in (28) should be re-

placed by G_S .

From the above it follows that the effective field pressure rises monotonically as the frequency decreases, reaching at $\Omega \ll 4\pi G_S / \epsilon$ its maximal value (the estimations here and further on imply that $\frac{3}{2+\epsilon} \sim \frac{3}{\epsilon} \sim 1$)

$$p_m = \frac{1}{16\pi} |(\mathbf{n} \cdot \mathcal{E}_0)|^2. \quad (29)$$

It means that at low frequencies (while $\Omega \ll 4\pi G_S / \epsilon$) the velocity of crystal deformation caused by the ponderomotive effect does not depend upon the vacancy concentration and hence upon the temperature. In other words, σ_E increases with decreasing temperature in inverse proportion to N_0 , reaching at $N_0 = 10^{-5}$ and $\mathcal{E}_0 = 1$ kV/cm the values of order 2×10^3 Pa.

At high frequencies, when $\Omega \gg 4\pi G_S / \epsilon$, the effective field pressure is proportional to the conductivity G_S :

$$p \approx \frac{1}{16\pi} \frac{8\pi G_S}{\Omega\epsilon} |(\mathbf{n} \cdot \mathcal{E}_0)|^2. \quad (30)$$

Respectively, the value of the equivalent mechanical compression stress, σ_E , in this case does not depend upon the vacancy concentration:

$$\sigma_E = \frac{e^2 \sum D_{S\alpha}}{\Omega\epsilon\omega kT} |(\mathbf{n} \cdot \mathcal{E}_0)|^2, \quad (31)$$

where $D_{S\alpha}$ are vacancy diffusion coefficients in the amorphized layer.

The crystal surface permeability asymmetry with respect to the vacancy motion direction leads to a macroscopic mass transfer only in the case when the diffusion coefficients of positive- and negative-charged vacancies are different. Another feature distinguishing this effect from the preceding one is that it is essential only if the field frequency is high enough. In fact, at low frequencies ($\Omega \ll 4\pi G_S / \epsilon$) we have $\kappa_{1,2} \ll 1$ and it follows from (24) that the amplitude of oscillations of total vacancy concentration perturbation $\sum \nu_{\alpha s}$ decreases with Ω as a consequence of electric field screening in the crystal:

$$\left| \sum \nu_{\alpha s} \right| = N_0 \frac{e|\mathcal{E}_{S0}|\lambda}{kT} \sqrt{\frac{\Omega\epsilon}{4\pi G_S}} \left| \frac{D_1 - D_2}{\sqrt{D_1 D_2}} \right|. \quad (32)$$

At the same time, at high frequencies ($\Omega \gg 4\pi G_S / \epsilon$) the value of $\sum \nu_{\alpha s}$ decreases with increasing Ω :

$$\left| \sum \nu_{\alpha s} \right| = N_0 \frac{e|\mathcal{E}_{S0}|\lambda}{kT} \sqrt{\frac{4\pi G_S}{\Omega\epsilon}} \left| \frac{\sqrt{2}(\sqrt{D_1} - \sqrt{D_2})}{\sqrt{D_1 + D_2}} \right|. \quad (33)$$

This is caused by the fact that the amount of vacancies coming to the boundary during the half of the field period decreases as the frequency rises.

Hence, there exists an optimal value of the field frequency $\Omega \sim 4\pi G_S / \epsilon$, corresponding to the maximal value of $|\sum \nu_{\alpha s}|$,

$$\nu_m \sim N_0 \frac{e|\mathcal{E}_{S0}|\lambda}{kT}. \quad (34)$$

Here we assumed that D_1 and D_2 are not very different.

If the crystal surface permeability coefficient, β , is greater when the vacancies move toward the surface than when they go in the opposite direction, (i.e., β_1 and $\sum \nu_{\alpha s}$ oscillate in phase), then $\langle \beta_1 \sum \nu_{\alpha s} \rangle > 0$. In this case the vacancy concentration averaged over the HF field period is lower in those areas at the surface where the electric field normal component is higher. Hence, crystal surface permeability asymmetry leads to the flattening of specimen in the direction of the external field vector, similar to the ponderomotive effect. The velocity of the respective plastic deformation can be also characterized by the equivalent mechanical stress

$$\sigma_E = \frac{kT}{N_0 \omega \beta_0} \langle \beta_1 \sum \nu_{\alpha s} \rangle. \quad (35)$$

The quantitative estimation of σ_E is difficult because no data on possible values of β_1 are available. However, if $\beta_1 \sim \beta_0$ and the frequency is of order of its optimal value, then σ_E reaches highly significant values: $\sigma_{Em} \sim e\mathcal{E}_0\lambda/\omega$. In particular, for the typical values $\lambda = 10^{-6}-10^{-7}$ cm, $\omega = 3 \times 10^{-23}$ cm³, and $\mathcal{E}_0 = 1$ kV/cm we obtain $\sigma_{Em} \sim 3 \times 10^5-3 \times 10^6$ Pa. This is comparable with the capillary stresses occurring in a powder compact with the grain size about 1 μm .

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