

Analysis of low-temperature relaxation resonances in materials containing defects

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A theory has been advanced providing an adequate description of thermal-activated relaxation resonances of various physical origins in defect-containing materials. The algorithm has been proposed for processing the temperature spectra of absorption and susceptibility defect to determine the relaxation process activation parameters.

Предложена теория, позволяющая адекватно описывать низкотемпературные термически активируемые релаксационные резонансы различной физической природы в материалах с дефектами. Описан алгоритм определения активационных параметров соответствующего процесса, основанный на анализе температурных спектров поглощения и дефекта восприимчивости.

When a solid is excited by external fields, the distorted equilibrium is restored spontaneously; the relaxation process exhibits a time lag from the change of parameter characterizing the external action. The process duration is characterized in the simplest cases by a certain relaxation time, τ , while in more complex ones, by a set of relaxation times. The excitation can be due to a mechanical stress, magnetic or electric field, so the relaxation is referred to as the mechanical, magnetic, or dielectric one, respectively. To obtain data on the relaxation physical mechanisms in solids, it is convenient to apply dynamic spectroscopy methods making use of low-amplitude harmonic actions with a period of the order of the relaxation time τ . In such experiments, the body to be studied is subjected to a perturbation $\alpha = \alpha_0 \exp(i\omega t)$ with the amplitude α_0 and circular frequency ω and its response $\beta = \beta_0 \exp(i(\omega t - \varphi))$ is registered; here $\beta_0 = \beta_0(\omega)$ is the response amplitude and $\varphi = \varphi(\omega)$, the phase shift of the response with respect to the external action. The response linearity and periodicity at the frequency ω are provided by the small amplitude α_0 . The ratio $\beta/\alpha = M^*(\omega) =$

$M_1(\omega) - iM_2(\omega)$ is referred to as the material complex susceptibility. For vibrations of a specific physical type, $M_1(\omega)$ has the sense of dynamical elasticity module, magnetic susceptibility, or dielectric one. The ratio $M_2(\omega)/M_1(\omega) = \operatorname{tg}\varphi(\omega)$ characterizes the energy dissipation of the respective vibration process due to relaxation.

An acceptable model to describe the dynamic excitation of most real materials is based on a standard linear body for which the susceptibility (module) defect $\Delta M(\omega)/M_\infty$ and reverse quality $Q^{-1}(\omega)$ are used as the relaxation macroscopic characteristics:

$$\frac{\Delta M(\omega)}{M_\infty} = \frac{M_\infty - M_1(\omega)}{M_\infty} = \frac{\Delta_r}{1 + \omega^2\tau^2}, \quad (1)$$

$$Q^{-1}(\omega) = \frac{M_2(\omega)}{M_\infty} = \Delta_r \frac{\omega\tau}{1 + \omega^2\tau^2},$$

where M_∞ is the unrelaxed susceptibility; $\Delta_r = C_r\Delta_0$, the relaxation force; Δ_0 , the characteristic dimensionless elementary contribution to the material dynamic response; C_r , the relative dimensionless volume concentration of elementary relaxation oscilla-

tors interacting with the sample vibration mode being considered.

In many cases, the relaxation processes at the microscopic level are due to elementary thermal-activated structure rearrangements and the relaxation time $\tau(T)$ is described by the Arrhenius law:

$$\tau(T) = \tau_0 \exp(U_0/kT), \quad (2)$$

where τ_0 and U_0 are the try period and activation energy, respectively, of the elementary relaxators; k , the Boltzmann constant; T , temperature. When such processes are studied using the dynamic methods, the Debye relaxation peak and a blurred "step" corresponding thereto are observed in temperature dependences of Q^{-1} and $\Delta M/M_\infty$ obtained at a fixed frequency $\omega = const$ or in frequency dependences of Q^{-1} and $\Delta M/M_\infty$ obtained at a fixed temperature $T = const$. The resonance condition is defined by the relationship $\omega\tau = 1$, therefore, in temperature spectra of relaxation at a fixed frequency, the resonance is localized near the temperature $T_p^{(0)} = -U_0/k \ln \omega\tau_0$ and the corresponding absorption peak has a characteristic width at the half-maximum amplitude

$$T_h^{(0)} = \frac{2U_0}{k} \cdot \frac{\ln(2 + \sqrt{3})}{(\ln \omega\tau_0)^2 - (\ln(2 + \sqrt{3}))^2}.$$

In frequency spectra of relaxation at a fixed temperature, the resonance is localized near the frequency $\omega_p^{(0)} = (\tau_0 \exp(U_0/kT))^{-1}$ and the corresponding absorption peak has a characteristic width at the half-maximum amplitude $\omega_h^{(0)} = 2\sqrt{3} \cdot (\tau_0 \exp(U_0/kT))^{-1}$. The main task to be solved using the dynamic spectroscopy methods is to reveal such resonances and to consider those in order to determine the parameters of elementary relaxators, τ_0 and U_0 .

Real materials contain randomly distributed structure distortions and imperfections arising both during the manufacturing process and at subsequent treatments. In this case, the elementary relaxators parameters U_0 , τ_0 , and Δ_0 take random additions in different regions of a crystal, so, instead of those parameters, random quantities and distribution functions corresponding thereto are to be considered. The response functions of a macroscopic sample $\Delta M(T, \omega)/M_\infty$ and $Q^{-1}(T, \omega)$

are transformed into complicated functions $\Delta \bar{M}(T, \omega)/M_\infty$ and $\bar{Q}^{-1}(T, \omega)$ obtained by statistical averaging of (1) [1].

A change in the material defect structure may change both the localization temperature and width ($T_p \neq T_p^{(0)}$ and $T_h \neq T_h^{(0)}$) of the relaxation resonances. The interpretation of such changes is among important problems in the dynamic relaxation theory.

It is obvious that the temperature and frequency dependences $\Delta \bar{M}(T, \omega)/M_\infty$ and $\bar{Q}^{-1}(T, \omega)$ must be influenced at the highest extent by the averaging over the distribution of the activation energy and try period. In the case of low temperatures, $kT \ll U_0$, only the statistical character of activation energy can be taken into account while the dispersion of Δ_0 , and τ_0 can be neglected at an exponential accuracy, that is, the influence of the structure defects can be described taking into account the scatter of activation energy U local values.

A change in the material defect structure may result in a shift of the resonance temperature T_p both towards higher and lower temperatures. That is, the structure changes increasing the activation energy scatter with respect to the initial value U_0 may enrich both high-energy ($\bar{U} > U_0$) and low-energy ($\bar{U} < U_0$) states of relaxation oscillators. Both these cases can be described using the quasi-Gaussian distribution function $P(U)$ at a small dispersion $D \ll U_0$, the parameter ν being selected in a suitable manner.

$$P(U, U_0, D, \nu) = \quad (3)$$

$$= \frac{N_\nu}{\sqrt{2\pi D}} \frac{2U_0^{-1}U}{U_0^\nu + U^\nu} \exp\left\{-\left[\frac{U - U_0}{\sqrt{2D}}\right]^2\right\}$$

$$N_\nu = \frac{4U_0^2 + \nu D^2}{4U_0^2},$$

where N_ν is the normalization coefficient meeting the normalization condition

$$\int_0^\infty P(U, U_0, D, \nu) dU = 1.$$

At a statistical distribution of the activation energy, the response functions averaged over (3) should be considered instead of the Debye relaxation spectrum (1):

$$\frac{\Delta \bar{M}}{M_\infty} = \Delta_r \cdot \mu^{(1)}(T, \omega, \tau_0, U_0, D, \nu) \quad (4)$$

$$\bar{Q}^{-1} = \Delta_r \cdot \mu^{(2)}(T, \omega, \tau_0, U_0, D, \nu),$$

$$\mu^{(1)} = \int_0^\infty \frac{1}{1 + \omega^2 \tau_0^2 \exp(2U/kT)} P(U, U_0, D, \nu) dU \text{ and}$$

$$\mu^{(2)} = \int_0^\infty \frac{\omega \tau_0 \exp(U/kT)}{1 + \omega^2 \tau_0^2 \exp(2U/kT)} P(U, U_0, D, \nu) dU.$$

In what follows, the frequency and temperature relaxation spectra are considered separately.

Frequency relaxation spectrum. Let new problem parameters be introduced: $\theta_\omega = [\exp(U_0/kT)]^{-1}$, dimensionless temperature; $\Omega_\omega = \omega/\omega_p^{(0)} = \omega\tau_0 \cdot \exp(U_0/kT) = \omega\tau_0/\theta_\omega$, dimensionless frequency; $d_\omega = \sqrt{2}D/U_0$, dimensionless dispersion characteristic; $x = \exp(U/kT)$, the new integration variable. Then,

$$\mu_\omega^{(1)} = N_\omega^{(\nu)} \cdot \chi_\omega \cdot \int_1^\infty \frac{dx}{x} \cdot f_\omega(x, \theta_\omega, \Omega_\omega, d_\omega, \nu), \quad (5)$$

$$\mu_\omega^{(2)} = N_\omega^{(\nu)} \cdot \chi_\omega \cdot \theta_\omega \Omega_\omega \cdot \int_1^\infty dx \cdot f_\omega(x, \theta_\omega, \Omega_\omega, d_\omega, \nu),$$

$$f_\omega(x, \theta_\omega, \Omega_\omega, d_\omega, \nu) = \frac{1}{(-\ln\theta_\omega)^\nu + (\ln x)^\nu} \cdot \frac{\ln x}{\theta_\omega^2 \Omega_\omega^2 x^2 + 1} \times \exp\left(-\left[\frac{\ln x + \ln\theta_\omega}{d \cdot \ln\theta_\omega}\right]^2\right)$$

$$\chi_\omega = \frac{2(-\ln\theta_\omega)^{\nu-2}}{\sqrt{\pi}d_\omega},$$

$$N_\omega^{(\nu)} = \frac{8 + \nu d_\omega^2}{8}.$$

The frequency dependences of normalized dynamic response function $\mu_\omega^{(1)}$ and $\mu_\omega^{(2)}$ are presented in Fig. 1.

The following characteristics of a relaxation resonance are measurable in experiment: ω_p or $\Omega_p(\theta_\omega, d_\omega, \nu) = \omega_p/\omega_p^{(0)}$, the positions of absorption peak and inflection point of "step" of the susceptibility defect step at the frequency axis determined from

the condition $\frac{\partial \mu_\omega^{(2)}}{\partial \omega} = 0$ or $\frac{\partial^2 \mu_\omega^{(1)}}{\partial \omega^2} = 0$ and $\omega_h = \omega_h^{(+)} - \omega_h^{(-)}$ or $\Omega_h(\theta_\omega, d_\omega, \nu) = \omega_h/\omega_p^{(0)}$, the absorption peak width where $\omega_h^{(+,-)}$ means the frequency values where

$$\mu_\omega^{(2)}(T, \omega_h^{(+,-)}, \tau_0, U_0, D, \nu) = (1/2) \cdot \max \mu_\omega^{(2)}(T, \omega, \tau_0, U_0, D, \nu) = (1/2) \cdot \mu_\omega^{(2)}(T, \omega_p, \tau_0, U_0, D, \nu).$$

At $d_\omega = 0$ to 1 and $\theta_\omega = 1 \cdot 10^{-8}$ to $1 \cdot 10^{-3}$, the relaxation resonance parameters are related to those of elementary relaxators by the following relations:

$$\Omega_p = 1 + C_\omega^{(\nu)} \cdot (d_\omega \ln \theta_\omega)^2, \quad (6)$$

$$C_\omega^{(\nu)} = \frac{\nu - 2}{50} \text{ or } \omega_p = \omega_p^{(0)} \cdot \left[1 + C_\omega^{(\nu)} \cdot \frac{2D^2}{k^2 T^2}\right],$$

$$\Omega_h = \Omega_h^{(0)} \cdot \left(1 + B_\omega^{(\nu)} \cdot (d_\omega \ln \theta_\omega)^2\right), \quad (7)$$

$$B_\omega^{(\nu)} = \frac{5 + \nu}{15} \text{ or } \omega_h = \omega_h^{(0)} \cdot \left(1 + B_\omega^{(\nu)} \cdot \frac{2D^2}{k^2 T^2}\right),$$

where $\Omega_h^{(0)} = 2\sqrt{3}$ is the Debye absorption peak width.

The values $\nu < 2$ answer to the resonance shift towards lower frequencies ($\omega_p < \omega_p^{(0)}$) while $\nu > 2$, to that towards higher frequencies ($\omega_p > \omega_p^{(0)}$).

The activation energy U_0 and its dispersion D can be estimated proceeding from the known temperature value T and experimental values of the resonance localization frequency ω_p and the absorption peak width ω_h :

$$U_0 = kT \cdot \ln \left[\frac{2\sqrt{3}(B_\omega^{(\nu)} - C_\omega^{(\nu)})}{\tau_0(2\sqrt{3} \cdot B_\omega^{(\nu)} \cdot \omega_p - \omega_h)} \right], \quad (8)$$

$$D = kT \cdot \sqrt{\frac{2\sqrt{3} \cdot \omega_p - \omega_h}{2(C_\omega^{(\nu)} \cdot \omega_h - 2\sqrt{3} \cdot B_\omega^{(\nu)} \cdot \omega_p)}}.$$

The relaxation force Δ_r can be found from the condition

$$\left. \frac{\Delta \bar{M}(\omega)}{M_\infty} \right|_{\omega \ll \omega_p}^{\text{experiment}} = \mu_\omega^{(1)} \Big|_{\omega \ll \omega_p} \cdot \Delta_r = \Delta_r.$$

Temperature relaxation spectrum. Let new problem parameters be introduced [2-6]: $\theta_T = T/T_p^{(0)} = -kT \ln(\omega\tau_0)/U_0$, dimensionless temperature; $\Omega_T = \omega\tau_0$, dimensionless frequency; $d_T = \sqrt{2}D/kT_p^{(0)} = -\sqrt{2}D \ln(\omega\tau_0)/U_0$,

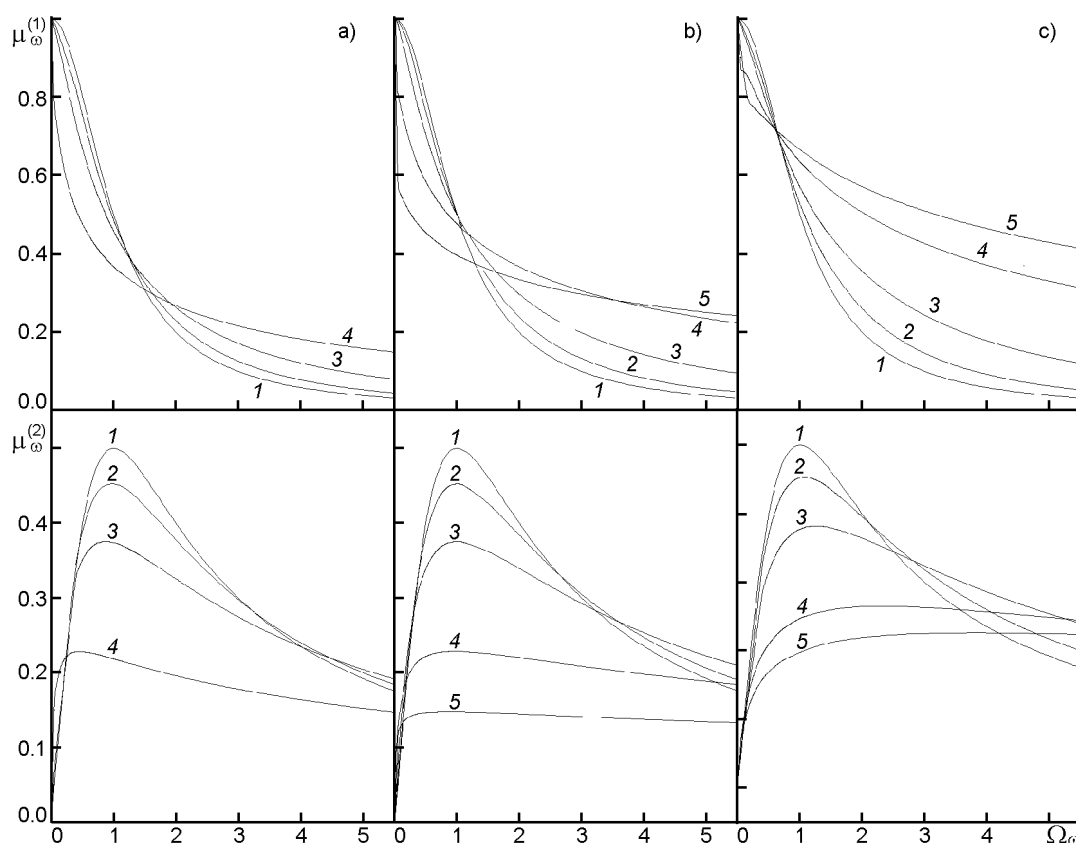


Fig. 1. Transformation of frequency dependences of normalized dynamic response functions $\mu_{\omega}^{(1)}$ and $\mu_{\omega}^{(2)}$ at fixed parameter value $\theta_{\omega} = 1 \cdot 10^{-3}$ and different values of parameter v : $v = 0$ (a), $v = 2$ (b), $v = 6$ (c); $d_{\omega} = 0$ (1), 0.1 (2), 0.2 (3), 0.5 (4), 1 (5).

dimensionless dispersion characteristic; $x = \exp(U/kT)$, the new integration variable. Then,

$$\mu_T^{(1)} = N_T^{(v)} \cdot \chi_T \cdot \int_1^{\infty} \frac{dx}{x} \cdot f_T(x, \theta_T, \Omega_T, d_T, v), \tag{9}$$

$$\mu_T^{(2)} = N_T^{(v)} \cdot \chi_T \cdot \Omega_T \cdot \int_1^{\infty} dx \cdot f_T(x, \theta_T, \Omega_T, d_T, v),$$

$$f_T(x, \theta_T, \Omega_T, d_T, v) = \frac{1}{(-\ln \Omega_T)^v + (\theta_T \ln x)^v} \times$$

$$\times \frac{\ln x}{\Omega_T^2 x^2 + 1} \cdot \exp \left(- \left[\frac{\theta_T \ln x + \ln \Omega_T}{d_T} \right]^2 \right),$$

$$\chi_T = \frac{2\theta_T^2}{\sqrt{\pi} d_T} (-\ln \Omega_T)^{v-1},$$

$$N_{\omega}^{(v)} = \frac{8(\ln \Omega_T)^2 + v d_T^2}{8 \ln \Omega_T^2}.$$

The temperature dependences of normalized dynamic response function $\mu_T^{(1)}$ and $\mu_T^{(2)}$ are presented in Fig. 2.

The following characteristics of a resonance are measurable in experiment: T_p or $\theta_p(\Omega_T, d_T, v) = T_p/T_p^{(0)}$, the positions of the peak and the "step" inflection point at the temperature axis determined from the condition $\partial \mu_T^{(2)}/\partial T = 0$ or $\partial^2 \mu_T^{(1)}/\partial T^2 = 0$, their width and amplitude.

As the absorption peak width characteristic, the parameter $T_h = T_h^{(+)} - T_h^{(-)}$ or $\theta_h(\Omega_T, d_T, v) = T_h/T_p^{(0)}$ is used as a rule ($T_h^{(+,-)}$ means the temperature values where

$$\mu_T^{(2)}(T_h^{(+,-)}, \omega, \tau_0, U_0, D, v) =$$

$$= (1/2) \cdot \max \mu_T^{(2)}(T, \omega, \tau_0, U_0, D, v) =$$

$$= (1/2) \cdot \mu_T^{(2)}(T_p, \omega, \tau_0, U_0, D, v).$$

When experimental data are processed, one has to do with relaxation peaks observed against the background absorption

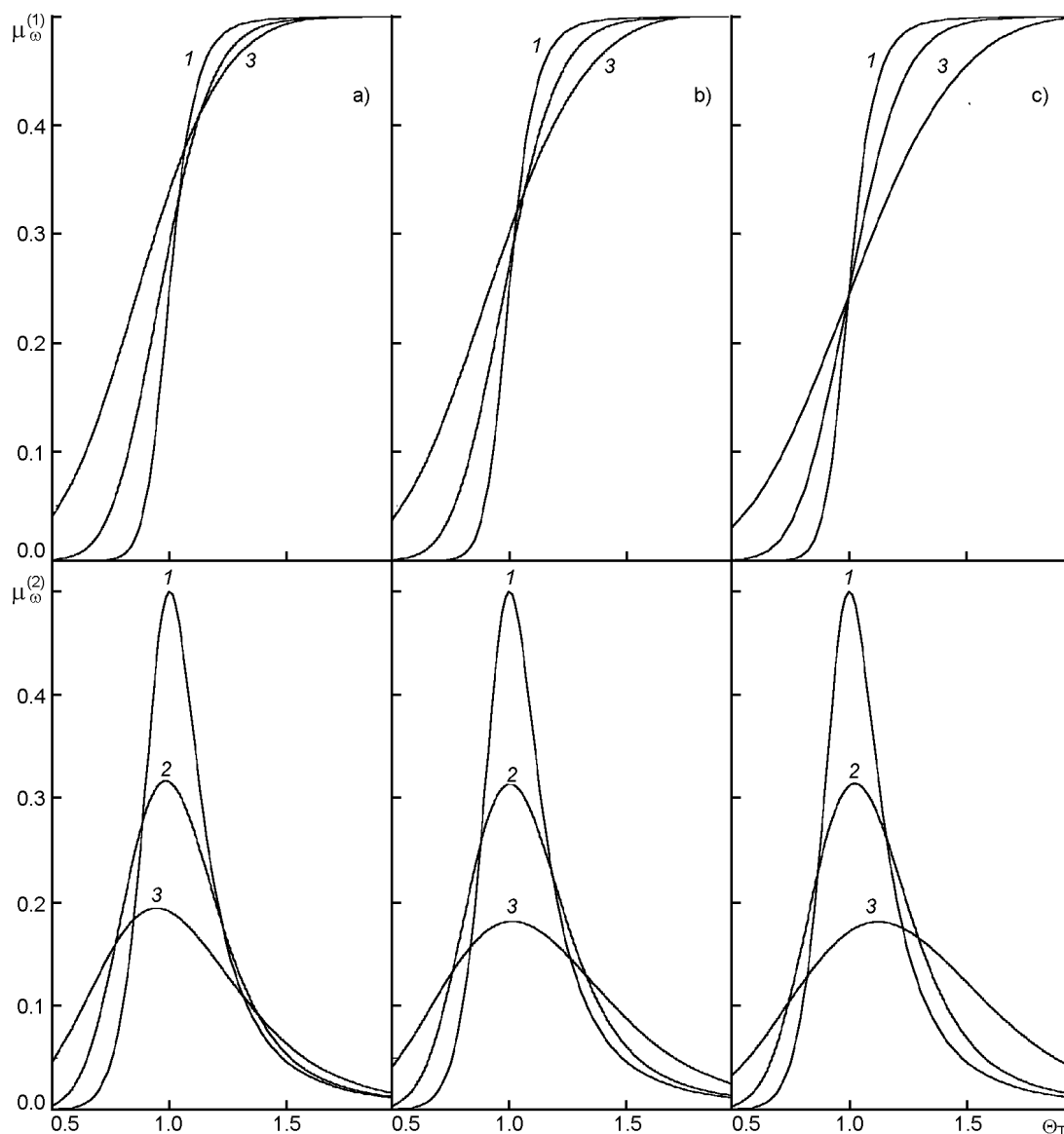


Fig. 2. Transformation of temperature dependences of normalized dynamic response functions $\mu_T^{(1)}$ and $\mu_T^{(2)}$ at fixed parameter value $\Omega_T = 1 \cdot 10^{-4}$ and different values of parameter ν : $\nu = 6$ (a), $\nu = 4$ (b), $\nu = 2$ (c); $d_T = 0$ (1), 1 (2), 3 (3).

noise, so the use of T_h requires the correct background subtraction. If the background depends (even slightly) on the temperature at the peak width, it is difficult to be subtracted. More convenient is to use the parameter $T_k = T_k^{(+)} - T_k^{(-)}$ or $\theta_k(d, \Omega, \nu) = T_k/T_p^{(0)}$ as the peak width characteristic almost independent of the background absorption ($T_k^{(+,-)}$ means the inflection points at the temperature dependence of absorption where $\partial^2 \mu_T^{(2)} / \partial T^2 = 0$ at $T = T_k$).

To characterize the width of "step" of the susceptibility defect step width, let the parameter

$$T_s = T_s^{(+)} - T_s^{(-)} = \left(\frac{\partial \mu_T^{(1)}(T, \omega, \tau_0, U_0, D, \nu)}{\partial T} \Big|_{T=T_p} \right)^{-1} \text{ or } \theta_s(\Omega_T, d_T, \nu) = T_s/T_p^{(0)}, \quad (10)$$

be introduced where $T_s^{(+,-)}$ are temperature values at which tangents to the "step" in the inflection point T_p intersect the high-

temperature and low-temperature asymptotes of the "step" (experimental dependences of the susceptibility defect are to be normalized to $C_p \Delta_0$ so that the high-temperature asymptote of the experimental dependence should be equal to 1).

The relaxation resonance parameters are related to those of elementary relaxators by the following relations:

$$\theta_p = 1 + C_T^{(\nu)} \cdot \frac{d_T^2}{(\ln \Omega_T)^2}, \quad (11)$$

$$C_T^{(\nu)} = \frac{4 - \nu}{8} \text{ or}$$

$$T_p = T_p^{(0)} \cdot \left[1 + C_T^{(\nu)} \cdot \frac{2D^2}{U_0^2} \right].$$

The values $\nu < 4$ answer to the resonance shift towards higher temperatures ($T_p > T_p^{(0)}$) while $\nu > 4$, to that towards lower ones ($T_p < T_p^{(0)}$).

$$\theta_i = \theta_i^{(0)} \cdot (1 + B_i^{(\nu)} \cdot d_T) \text{ or} \quad (12)$$

$$T_i = T_i^{(0)} \cdot (1 - B_i^{(\nu)} \cdot \frac{\sqrt{2}D}{U_0} \ln \omega \tau_0),$$

where $\theta_i = \theta_h, \theta_k, \theta_s$ and $\theta_i^{(0)} = \theta_h^{(0)}, \theta_k^{(0)}, \theta_s^{(0)}$ are the parameter values corresponding to the Debye resonance:

$$\theta_i^{(0)} = \zeta_i \cdot \ln \Omega, \\ \zeta_h = \frac{2 \cdot \ln(2 + \sqrt{3})}{(\ln \Omega)^2 - (\ln(2 + \sqrt{3}))^2} \approx \frac{2.63}{(\ln \Omega)^2}$$

within the range being of importance for experiments

$$\Omega = 1 \cdot 10^{-8} - 1 \cdot 10^{-2}, \quad \zeta_k = \sqrt{3} / (\ln(\Omega))^2,$$

$$\zeta_s = 2 / (\ln(\Omega))^2; \quad B_h^{(\nu)} = (25 - \nu) / 60,$$

$$B_k^{(\nu)} = (25 - \nu) / 40, \quad B_h^{(\nu)} = (25 - \nu) / 40.$$

It follows from (11) that τ_0 can be estimated from the activation plot:

$$\ln \omega = \ln \tau_0^{-1} - \frac{U_0^2 + 2C_T^{(\nu)} D^2}{U_0} \cdot \frac{1}{kT_p}.$$

The formulas providing determination of U_0 and D proceeding from the known $\omega \tau_0$ value and experimental values of the resonance localization temperature T_p and its width characteristic T_i have the following form:

$$U_0 = k(\ln \omega \tau_0)^3 / 2\zeta_i \left(C_T^{(\nu)} + (B_i^{(\nu)} \cdot (\ln \omega \tau_0))^2 \right) \times \\ \times \left(-\zeta_i (B_i^{(\nu)})^2 T_p + \frac{2C_T^{(\nu)} T_h}{(\ln \omega \tau_0)^3} - \right. \\ \left. \left(\frac{\zeta_i^2 (B_i^{(\nu)})^4 T_p^2 - 4\zeta_i (B_i^{(\nu)})^2 C_T^{(\nu)} T_p T_h}{(\ln \omega \tau_0)^3} - \right. \right. \\ \left. \left. - \frac{4(B_i^{(\nu)})^2 C_T^{(\nu)} T_h^2}{(\ln \omega \tau_0)^4} \right)^{1/2} \right), \quad (13)$$

$$D = \sqrt{2} k (\ln \omega \tau_0)^2 / 4\zeta_i \left(C_T^{(\nu)} + (B_i^{(\nu)} \cdot (\ln \omega \tau_0))^2 \right) \times \\ \times \left(-\zeta_i B_i^{(\nu)} T_p - \frac{2B_i^{(\nu)} T_h}{\ln \omega \tau_0} - \left(\zeta_i B_i^{(\nu)} T_p \right) - \right. \\ \left. - \frac{4\zeta_i C_T^{(\nu)} T_p T_h}{(\ln \omega \tau_0)^3} - \frac{4\zeta_i C_T^{(\nu)} T_h^2}{(\ln \omega \tau_0)^4} \right)^{1/2}. \quad (14)$$

The relaxation force Δ_r can be found from the condition

$$\frac{\Delta M(T)}{M_\infty} \Big|_{T \gg T_p}^{(\text{experiment})} = \mu_T^{(1)} \Big|_{T \gg T_p} \cdot \Delta_r = \Delta_r.$$

At $\nu = 0$, the ratio of derivatives with respect to temperature in the inflection points of functions $\mu_T^{(2)}(\theta_T, \Omega_T, d_T, \nu) \Big|_{\nu=2}$ has the same value

$$K = -\max[\partial \mu_T^{(2)} / \partial \theta_T] / \min[\partial \mu_T^{(2)} / \partial \theta_T],$$

that depends on the parameter Ω_T but is independent of the dispersion parameter d_T and makes it possible to approximate it analytically as $K(\Omega_T) = 1 + 10 / (-\ln \Omega_T)^{\sqrt{2}}$ that provides the determination of τ_0 by considering only the peak shape without resort to experiments with varying frequency ω .

In conclusion, the effect of a material defect structure on the parameters of frequency and temperature spectra of dynamic relaxation has been considered theoretically. At low temperatures, the structure defect influence on parameters of a thermally activated relaxation resonance can be explained proceeding from the supposed statistical scatter of the process activation energy and the dependence of that statistical distribution on the material structure perfection. The increase of

the activation energy dispersion has been found to result in broadening of both frequency and temperature relaxation spectra. It has been established also that the resonance position at the frequency and temperature axes depends on the activation energy dispersion, the character of that dependence being defined uniquely by the form of the activation energy distribution function. The theory proposed made it possible to describe adequately the features of acoustic relaxation spectra in niobium and iron [3, 4, 6, 7].

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Аналіз низькотемпературних релаксаційних резонансів у матеріалах з дефектами

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Запропоновано теорію, що дозволяє адекватно описувати релаксаційні резонанси різної фізичної природи у матеріалах з дефектами. Описано метод визначення акти-
ваційних параметрів відповідного процесу, що базується на аналізі температурних спектрів поглинання та сприйнятливості.