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Simple model of the dielectric response of disordered substances: an explanation of 'nearly constant loss' and giant dielectric constant

A I Ritus

Institute of General Physics, Russian Academy of Sciences, Vavilov Street 38, 119991 Moscow, Russia

E-mail: ritus@ran.gpi.ru

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Abstract
A new model of dielectric response of disordered substances is proposed. According to this model, the dielectric response is determined by the Drude drift currents localized in minima of the random electrostatic potential. The drift of a charge carrier with mobility μ in local parabolic potential $\varphi(x) = \varphi_{\min} + kx^2/2$ under an external alternating field results in the Debye-type response with 'relaxation frequency' $\omega_r = \mu k$. The use of distribution functions $G(k)$ for values k of local potential wells of a disordered material allows us to describe quantitatively both the spectral domains of the 'nearly constant loss' and the cases of the giant contribution to a low-frequency dielectric constant. In disordered substance the response depends mainly on the width but not the shape of $G(k)$.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In 1977 Jonscher [1] analysed experimental data on the frequency dependences of the conductivity of various disordered substances, such as doped crystals with electronic or ionic conductivity, glasses, polymers, organic acids, and amorphous semiconductors. He has shown that the real part of the conductivity of all these disordered substances is well described by the expression $\sigma'(\omega) = \sigma_{dc}[1+(\omega/\omega_0)^s]$, where σ_{dc} is the dc conductivity, $s \approx 0.7\pm0.2$, and ω_0 is a characteristic frequency. Jonscher has named such a kind of frequency response the 'universal dielectric response' (UDR) [1, 2]. Parameters σ_{dc} and ω_0 exhibit Arrhenius-type temperature dependences with close activation energies. This indicates that the frequency-dependent conductivity originates from migration of charge carriers, just the same as the dc conductivity σ_{dc}. In the majority of papers explaining UDR behaviour the models of conductivity are used, which include jumps of charge carriers over barriers dividing neighbouring vacant possible positions in crystalline or amorphous structure (see, for example [2–8]). In a number of papers the relaxation of an ionic surrounding during a charge motion is also taken into account (see, for example [9, 10]). The averaging of appropriate probabilities over barrier heights, energies of possible positions and distances between them allows one to obtain the term $(\omega/\omega_0)^s$ in the frequency dependence of conductivity.

In 1991 Lee *et al* [11] showed, with the examples of three ionically conducting crystals and two glasses, that the exponent s grows monotonically with decreasing temperature and reaches the limiting value $s = 1$ at relatively low temperatures. The authors of [11] have labelled this limiting regime 'new (second) universality'. Later, in [12–14], it has been shown that the 'new universality' regime is caused by the additional contribution to conductivity, weakly dependent on temperature. Thus, the conductivity of disordered substances is generally described by the expression

$$\sigma'(\omega) = \sigma_{\mathrm{dc}}[1 + (\omega/\omega_0)^s] + A\omega^1, \tag{1}$$

where parameter $A = A(T)$ varies with temperature T weakly as compared to thermally activated σ_{dc} and ω_0. The contribution of the term $A\omega^1$ to the conductivity dominates at low temperatures and high frequencies. As the imaginary part of dielectric constant ε'' (dielectric loss) is related to the conductivity by the expression $\varepsilon'' = \sigma'/\varepsilon_0\omega$ (here ε_0 is the permittivity of free space), the predominance of the 'new universality' $A\omega^1$ corresponds to the predominance of a 'nearly constant loss' (NCL) regime $\varepsilon''(\omega) \approx$ constant. In the majority of papers the NCL behaviour is explained with the help of the conductivity model, which describes the motion of charge carriers in asymmetric double-well potential configurations with a wide distribution of barrier heights and asymmetry energies [4, 15].

There are, however, models of the conductivity of disordered substances with non-power law frequency dependence: 'jump and relaxation' model [10, 16], 'random barriers' model [17], 'ion motion in decaying cage' model [18] and 'effective medium' model [19]. The conductivity spectra obtained in these models are described by an apparent power law with an exponent $s(\omega, T)$, which continuously grows with increase of frequency and decrease of temperature and reaches the limiting value $s = 1$. Nevertheless, the spectra of these models also can be described approximately by using expression (1).

In all the above-mentioned models of conductivity, explaining NCL behaviour, charge carriers overcome barriers between energy states of microscopic systems of atomic scale by hopping or by a tunnel effect.

In this paper, in order to explain the NCL regime, we present a simple model based on classical representations of Drude conductivity in conditions of random electrostatic potential of a disordered substance. We obtain the dielectric response 'from first principles' as the solution of an equation of motion for charge carriers under the action of an external alternating field and internal static field of random potential. Within the framework of our model the NCL regime is accompanied by growth of the real part of dielectric constant $\varepsilon'(\omega)$ with frequency decreasing, and ε' can reach giant values $\sim 10^4$ in the low-frequency range at moderate values of substance conductivity.

2. Dielectric response due to Drude drifts of charge carriers localized in random potential minima of a disordered material

Let us consider a disordered solid-state substance with an average density N of charge carriers characterized by mobility μ and charge q. The various reasons for disorder can be chaotically distributed fixed charged impurities, defects and dislocations in dielectrics and semiconductors, the 'frozen' density fluctuations and composition fluctuations in glasses and ceramics, domains in ferroelectric materials, etc. These inhomogeneities produce a non-uniform random distribution of electrostatic potential $\varphi(\boldsymbol{R})$ in a substance. Here we assume that $\varphi(\boldsymbol{R})$ is a function of the radius vector $\boldsymbol{R}$, slowly varying as compared to the potential bound to the atomic structure of the substance; that is, in the lack of disorder, $\varphi(\boldsymbol{R}) = \text{constant} = \varphi_0$.

According to the definition of mobility, in an electrical field $\boldsymbol{E}$ the drift speed of charge carriers $\boldsymbol{v} = \mu\boldsymbol{E}$. If in a substance in addition to the electrostatic potential $\varphi(\boldsymbol{R})$ there is a uniform alternating electrical field $\boldsymbol{E}(\omega) = \boldsymbol{E}_0 \exp(-\mathrm{i}\omega t)$, then $\boldsymbol{E} = -\operatorname{grad}\varphi(\boldsymbol{R}) + \boldsymbol{E}_0 \exp(-\mathrm{i}\omega t)$ and for the drift velocity of charges $\boldsymbol{v}(\boldsymbol{R}) = \mathrm{d}\boldsymbol{R}/\mathrm{d}t$ we obtain the equation

$$\mathrm{d}\boldsymbol{R}/\mathrm{d}t = \mu(-\operatorname{grad}\varphi(\boldsymbol{R}) + \boldsymbol{E}_0 \exp(-\mathrm{i}\omega t)). \tag{2}$$

Let N_1 of the total number N of charge carriers be localized in minima of random potential, and accordingly $N_0 = N - N_1$ carriers remain free, so the dc conductivity of substance $\sigma_{\mathrm{dc}} = N_0 q\mu$. It is obvious that N_0, N_1 and N depend on the temperature T, and at low enough temperature

$$T \ll q\bar{\varphi}_{\min}/k_{\mathrm{B}}, \tag{3}$$

where k_{B} is the Boltzmann constant and $\bar{\varphi}_{\min}$ the average depth of random potential minima relative to level φ_0, practically all charge carriers will be localized in potential minima; that is, $N_1 \approx N$.

Let the alternating field $\boldsymbol{E}(\omega)$ be directed along the x-axis. As a first approximation, it is possible to consider that near to each local minimum the potential is spherically symmetric and depends quadratically on the distance from a point of a local minimum. Then for the x-axis direction we have

$$\varphi(x) = \varphi_{\min} + kx^2/2, \tag{4}$$

where k is a parameter describing the steepness of the local parabolic potential. Then equation (2) for the charge motion under the action of alternating field $E(\omega)$ in such a local potential takes the form

$$\mathrm{d}x/\mathrm{d}t + \mu kx = \mu E_0 \exp(-\mathrm{i}\omega t). \tag{5}$$

The stationary solution of this equation for the coordinate of a charge is

$$x(t) = \mu E_0 \exp(-\mathrm{i}\omega t)/(\mu k - \mathrm{i}\omega), \tag{6}$$

and for a charge velocity we have accordingly

$$v(t) = -\mathrm{i}\mu E_0\omega \exp(-\mathrm{i}\omega t)/(\mu k - \mathrm{i}\omega). \tag{7}$$

If N_1 charges are localized in potential minima with equal k, the appropriate current density is $j = N_1 q v(t) = \sigma E_0 \exp(-\mathrm{i}\omega t)$, whence we obtain for the dynamic conductivity

$$\sigma(\omega, k) = \sigma'(\omega, k) + \mathrm{i}\sigma''(\omega, k) = -\frac{\mathrm{i}\omega\mu q N_1}{\mu k - \mathrm{i}\omega}, \tag{8}$$

$$\sigma'(\omega, k) = \frac{\mu\omega^2 q N_1}{(\mu k)^2 + \omega^2}, \tag{9}$$

$$\sigma''(\omega, k) = -\frac{k\mu^2\omega q N_1}{(\mu k)^2 + \omega^2}. \tag{10}$$

The complex dielectric constant $\varepsilon(\omega, k)$, related to this dynamic conductivity, is determined by the known expression

$$\varepsilon(\omega, k) = \varepsilon'(\omega, k) + \mathrm{i}\varepsilon''(\omega, k) = 1 + \mathrm{i}\frac{\sigma(\omega, k)}{\varepsilon_0\omega}. \tag{11}$$

Substituting (8) in (11), we obtain

$$\varepsilon(\omega, k) = 1 + \frac{\mu q N_1}{(\mu k - \mathrm{i}\omega)\varepsilon_0}, \tag{12}$$

$$\varepsilon'(\omega, k) = 1 + \frac{k\mu^2 q N_1}{(\mu^2 k^2 + \omega^2)\varepsilon_0}, \tag{13}$$

$$\varepsilon''(\omega, k) = \frac{\mu\omega q N_1}{(\mu^2 k^2 + \omega^2)\varepsilon_0}. \tag{14}$$

Thus, the frequency dependences (12), (13), (14) of the dielectric response, related to the dynamic Drude conductivity of charge carriers localized in the parabolic potential, are similar to Debye frequency dependences with 'frequency of relaxation' $\omega_r = \mu k = 1/\tau_r$. At this frequency there is a maximum of the spectrum $\varepsilon''(\omega, k)$ and an appropriate 'step' in spectrum $\varepsilon'(\omega, k)$. When $\omega \ll \omega_r$ the dielectric constant $\varepsilon'(\omega, k) \approx \varepsilon'(0, k) = 1 + qN_1/k\varepsilon_0$ does not depend on the mobility of charge carriers and is determined only by their concentration and the steepness of the parabolic potential.

Let us evaluate $\varepsilon'(0, k)$ for characteristic parameters encountered in experiments. Let the cyclical relaxation frequency $\nu_r = \omega_r/2\pi = 10^6$ Hz, $q = e = 1.6 \times 10^{-19}$ C, $N_1 = 10^{13}$ cm^{-3} and $\mu = 1000$ cm^2 V^{-1} s^{-1}, which corresponds to electron mobility in silicon at $T = 300$ K. Then $k = \omega_r/\mu \approx 6300$ V cm^{-2} and $\varepsilon'(0, k) = 1 + qN_1/k\varepsilon_0 \approx 2900$. This estimation shows that even the small density of charge carriers (which corresponds to Drude conductivity σ_{dc} of order 10^{-3} Ω^{-1} cm^{-1}) in potential wells of an inhomogeneous solid results in a dielectric response with a giant value of the real part of the low-frequency dielectric constant, which is characteristic, for example, for relaxors.

We have considered the idealized particular case of a non-uniform medium, when N_1 charges are localized in minima of parabolic potentials with equal k. In a disordered non-uniform medium, these N_1 charges will fall into potential minima with various values of the parabola steepness k. Let the distribution of values k in such substance be characterized by a probability density $g(k)$, actually describing an ensemble of the forces returning charge carriers to equilibrium positions. Then for the conductivity and dielectric constant of a disordered material, the following expressions are valid:

$$\sigma'(\omega) = \int_0^\infty g(k)\sigma'(\omega, k)\,\mathrm{d}k + \sigma_{\mathrm{dc}}, \tag{15}$$

$$\sigma''(\omega) = \int_0^\infty g(k)\sigma''(\omega, k)\,\mathrm{d}k, \tag{16}$$

$$\varepsilon'(\omega) = \int_0^\infty g(k)\varepsilon'(\omega, k)\,\mathrm{d}k, \tag{17}$$

$$\varepsilon''(\omega) = \int_0^\infty g(k)\varepsilon''(\omega, k)\,\mathrm{d}k + \frac{\sigma_{\mathrm{dc}}}{\omega\varepsilon_0}, \tag{18}$$

where $\sigma'(\omega, k)$, $\sigma''(\omega, k)$, $\varepsilon'(\omega, k)$, $\varepsilon''(\omega, k)$ are determined accordingly by formulae (9), (10), (13), (14). For generality, we have added in (15) and (18) the terms stipulated by the dc conductivity σ_{dc}. We shall present a probability density in the form of

$$g(k) = \frac{G(k)}{\int_0^\infty G(k)\,\mathrm{d}k}, \tag{19}$$

where $G(k)$ is an arbitrary non-negative constrained function of k. It is natural to guess that in a disordered substance the random potential is described by a Gaussian distribution, for which

$$G(k) = \exp\left[-\frac{(k - k_0)^2}{2\Delta^2}\right], \tag{20}$$

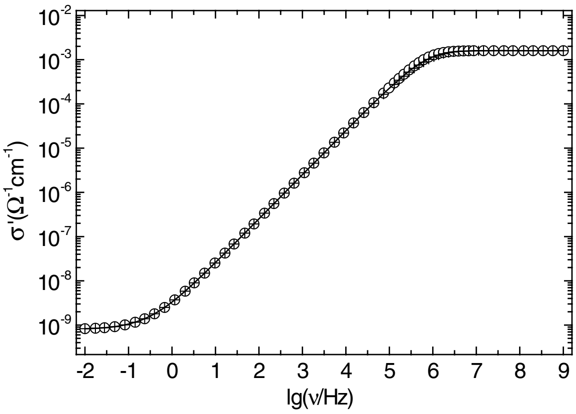

Figure 1. $\sigma'(\omega)$ spectra calculated for the Gaussian distribution (20) with $k_0 = 0$ (solid curve) and for triangular (21) and rectangular (22) distributions with $k_1 = 0$ (crosses and circles, respectively).

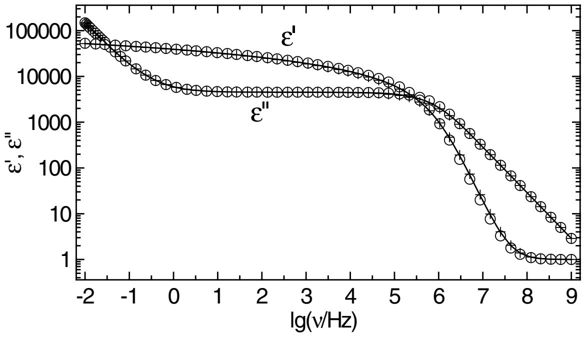

Figure 2. $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra calculated for the Gaussian distribution (20) with $k_0 = 0$ (solid curve) and for triangular (21) and rectangular (22) distributions with $k_1 = 0$ (crosses and circles, respectively).

where Δ is the mean-square deviation of k from k_0. One can consider that the case of $k_0 = 0$, when the spatial fluctuations of the random potential are deviations from the potential of the homogeneous substance $\varphi(x) = \text{constant} = \varphi_0$, is the most probable.

We performed the numerical computations of $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$, using formulae (9)–(20) with $k_0 = 0$ and parameters for the substance from the previous estimation: $\mu = 1000\ \text{cm}^2\ \text{V}^{-1}\ \text{s}^{-1}$, $q = e$, $N_1 = 10^{13}\ \text{cm}^{-3}$ and also $\Delta = 5030\ \text{V cm}^{-2}$ and $\sigma_{\text{dc}} = 8\times10^{-10}\ \Omega^{-1}\ \text{cm}^{-1}$. At such weak dc conductivity $N_0 = \sigma_{\text{dc}}/(q\mu) = 5\times10^6\ \text{cm}^{-3} \ll N_1$; that is, practically all charge carriers are localized in minima of the random potential. The results of calculations are shown in figures 1 and 2 by solid curves. The obtained frequency dependence of the conductivity $\sigma'(\omega)$ (figure 1) corresponds to the first and third terms of the frequency dependence (1): $\sigma'(\omega) = \text{constant} = \sigma_{\text{dc}}$ at low ω and $\sigma'(\omega) \sim A\omega^1$ in the frequency range $1\ \text{Hz} < \omega/2\pi < 10^6\ \text{Hz}$. Thus, here we have obtained the dependence $\sigma'(\omega)$, characteristic for 'new universality' [11] and corresponding to the frequency area of NCL [12] of the dependence $\varepsilon''(\omega)$ (figure 2). The increase of the curve $\varepsilon''(\omega) \sim \omega^{-1}$ at frequencies below 1 Hz is associated with the dc conductivity σ_{dc} (see (18)); if $\sigma_{\text{dc}} = 0$, then the plateau $\varepsilon'' = \text{constant}$ extends up to the lowest frequencies. The frequency spectrum $\varepsilon'(\omega)$ of the dielectric constant has a shape that is characteristic for relaxors: with decreasing frequency the step ε' is observed, which is replaced by a slow monotonic rise near $\nu_r = \omega_r/2\pi = 1.25\mu\Delta/2\pi = 10^6$ Hz, and the dielectric constant reaches the giant value $\varepsilon' \approx 50\,000$ at frequency $\nu = \omega/2\pi = 10^{-2}$ Hz for the parameters chosen. At frequencies above ν_r the conductivity σ' ceases to depend on the frequency and the 'new universality' is replaced by the plateau $\sigma'(\omega) = \text{constant} = \sigma_\infty$. The value of the conductivity at this plateau is numerically equal to $(N_0 + N_1)q\mu$, so all charge carriers of the substance contribute to the conductivity like free carriers at the frequencies $\omega > \omega_r$. The appropriate contribution of these carriers in $\varepsilon'(\omega)$ sharply diminishes at $\omega > \omega_r$ and disappears completely at $\omega/2\pi = 10^8$ Hz. It is notable that the plateau $\sigma'(\omega) = \text{constant} = \sigma_\infty$ is also obtained in some 'hopping' models of conductivity (see, for example [4, 7, 9, 10, 16, 17]).

The features of the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra described above are maintained at any values of the parameters μ, q, N_1, Δ and N_0 (for $N_0 \ll N_1$); only the absolute values of σ', ε', ε'', which are proportional to qN_1, and frequency $\omega_r = 1.25\mu\Delta$ are changed. Therefore, for example, the so-called 'master curve' [2] is obtained for conductivity σ' by plotting $\sigma'(\omega)/(\sigma_\infty - \sigma_{dc})$ versus ω/ω_r. Moreover, the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra do not depend practically on the shape of the distribution functions $G(k)$ at the given parameters μ, q, N_1 and N_0, if the functions $G(k)$ are continuous and distinct from zero starting with $k = k_1$, have close values of half-width $(k_2 - k_1)$ and the same values of $G(k_1)$ and $\int_0^\infty G(k)\,\mathrm{d}k$. For example, figures 1 and 2 shows the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra, calculated for the values μ, q, N_1 and N_0 chosen above for the Gaussian distribution (20) with $k_0 = 0$ (solid curves), for the triangular distribution

$$G(k) = \begin{cases} 1 - k/2k_2, & k_1 \leqslant k < 2k_2, \\ 0, & k < k_1, \quad k > 2k_2, \end{cases} \tag{21}$$

where $k_1 = 0$, $k_2 = 6300$ V cm^{-2} (are shown by crosses), and for the rectangular distribution

$$G(k) = \begin{cases} 1, & k_1 \leqslant k < k_2, \\ 0, & k < k_1, \quad k > k_2, \end{cases} \tag{22}$$

where also $k_1 = 0$, $k_2 = 6300$ V cm^{-2} (shown by circles). For each of these distributions $G(0) = 1$ and $\int_0^\infty G(k)\,\mathrm{d}k = 6300$ V cm^{-2} (this is fulfilled at $k_2 - k_1 = \Delta\sqrt{\pi/2} \approx 1.25\Delta$). The $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra, calculated for all three distributions, practically coincide, and the frequencies ω_r obey the equation

$$\omega_r = \mu k_2 = 1.25\mu\Delta. \tag{23}$$

Thus, the frequency ω_r is determined by the carrier mobility and the value of the greatest steepness of local minima of a random potential. Certainly, these spectra are practically indistinguishable in double logarithmic scale representation, but in linear scale for the y-axes some differences are appreciable. The maximal difference between the spectra obtained for the Gaussian (20) and triangular (21) distributions reaches only ~5%, while for the Gaussian and rectangular (22) ones it reaches ~10–20%.

The practical independence of spectra on the shape of distribution $G(k)$ confirms the adequacy of the considered model of the dielectric response of disordered conducting substance as it is intuitively clear that processes in a random potential are more likely determined by boundaries of distribution, its width and an integral over the distribution, than its shape. Besides, this independence allows using simpler distributions for the spectra modelling, for example, a rectangular one.

By means of (15)–(19), the spectra of the conductivity and dielectric constant of a disordered substance in the case of the Gaussian distribution (20) with $k_0 = 0$ are analytically expressed through the tabulated error function and exponential integral:

$$\sigma'(\omega) = \frac{\sqrt{2\pi}}{2\Delta} N_1 q\omega \exp\left(\frac{\omega^2}{2\mu^2\Delta^2}\right)\left(1 - \operatorname{erf}\left(\frac{\omega\sqrt{2}}{2\mu\Delta}\right)\right) + \sigma_{\mathrm{dc}}, \tag{24}$$

$$\varepsilon''(\omega) = \frac{\sigma'(\omega)}{\omega\varepsilon_0}, \tag{25}$$

$$\varepsilon'(\omega) = 1 + \frac{N_1 q}{\varepsilon_0 \Delta\sqrt{2\pi}} \exp\left(\frac{\omega^2}{2\mu^2\Delta^2}\right) E_1\left(\frac{\omega^2}{2\mu^2\Delta^2}\right). \tag{26}$$

Let us analyse these frequency dependences. At $\omega \ll \mu\Delta$, formula (24) becomes

$$\sigma'(\omega) = N_1 q\mu\left(\sqrt{\frac{\pi}{2}}\frac{\omega}{\mu\Delta} - \frac{\omega^2}{\mu^2\Delta^2}\right) + \mathrm{O}(\omega^3) + \sigma_{\mathrm{dc}}, \tag{27}$$

the conductivity increases linearly with ω and the proportionality factor is

$$A = \frac{N_1 q}{\Delta}\sqrt{\frac{\pi}{2}}. \tag{28}$$

At $\omega \gg \mu\Delta$,

$$\sigma'(\omega) = N_1 q\mu\left(1 - \frac{\mu^2\Delta^2}{\omega^2}\right) + \mathrm{O}(1/\omega^4) + \sigma_{\mathrm{dc}}, \tag{29}$$

and the conductivity levels off to a plateau $\sigma' = N_1 q\mu + N_0 q\mu = Nq\mu$; that is, all charge carriers of the substance contribute to the conductivity like free carriers in this frequency region. According to (25) in the frequency dependence of $\varepsilon''(\omega)$ there is a region of NCL and an area of rise $\varepsilon''(\omega)$ with decreasing frequency at $\omega \ll \mu\Delta$, where

$$\varepsilon''(\omega) = \frac{N_1 q}{\varepsilon_0\Delta}\sqrt{\frac{\pi}{2}} + \frac{\sigma_{\mathrm{dc}}}{\varepsilon_0\omega}, \tag{30}$$

and at $\omega \gg \mu\Delta$ $\varepsilon''(\omega)$ drops in inverse proportion to frequency:

$$\varepsilon''(\omega) = \frac{N_1 q\mu + \sigma_{\mathrm{dc}}}{\varepsilon_0\omega}. \tag{31}$$

Note that the coefficient A and value ε'' in the NCL region are proportional to the density of the localized carriers, and inversely proportional to the width of distribution $G(k)$, and do not depend on carrier mobility. Thus, the NCL regime is entirely stipulated by the steepness distribution of the relief of the random potential and is not related to the character of the charge carriers.

The dielectric constant ε' (26) at $\omega \ll \mu\Delta$ increases logarithmically with decreasing frequency:

$$\varepsilon'(\omega) = 1 + \frac{N_1 q}{\varepsilon_0\Delta\sqrt{2\pi}}\left(\ln\left(\frac{2\mu^2\Delta^2}{\omega^2}\right) - \gamma\right) + \mathrm{O}(\omega^2), \tag{32}$$

(here $\gamma = 0.577\,21\ldots$ is the Euler constant), and tends to unity at $\omega \gg \mu\Delta$:

$$\varepsilon'(\omega) = 1 + \frac{N_1 q\mu^2\Delta\sqrt{2}}{\varepsilon_0\omega^2\sqrt{\pi}} + \mathrm{O}(1/\omega^4). \tag{33}$$

Note that the value of ε' at $\omega \ll \mu\Delta$ is also proportional to N_1 and inversely proportional to the width of the distribution $G(k)$, but weakly depends on the carrier mobility.

In the case of rectangular distribution (22), the following expressions are obtained by means of (15)–(19):

$$\sigma'(\omega) = \frac{N_1 q\omega}{k_2 - k_1}\left[\arctan\left(\frac{\mu k_2}{\omega}\right) - \arctan\left(\frac{\mu k_1}{\omega}\right)\right] + \sigma_{\mathrm{dc}}, \tag{34}$$

$$\varepsilon'(\omega) = 1 + \frac{N_1 q}{2\varepsilon_0(k_2 - k_1)}\ln\left(\frac{\omega^2 + \mu^2 k_2^2}{\omega^2 + \mu^2 k_1^2}\right), \tag{35}$$

and $\varepsilon''(\omega)$ is expressed through (34) according to (25). For the case of $k_1 = 0$ at $\omega \ll \mu k_2$, the conductivity (34) is a linear function of frequency:

$$\sigma'(\omega) \approx \frac{N_1 q\omega}{k_2}\frac{\pi}{2} + \sigma_{\mathrm{dc}}. \tag{36}$$

The proportionality coefficient $\frac{N_1 q}{k_2}\frac{\pi}{2}$ becomes equal to coefficient A (see (28)) for the case of Gaussian distribution, if $k_2 = \Delta\sqrt{\pi/2}$; that is, at the fulfilment of equality of the integrals $\int_0^\infty G(k)\,\mathrm{d}k$ for these distributions. At $\omega \gg \mu k_2$ the conductivity flattens out to the same plateau, $\sigma'(\omega) = N_1 q\mu + \sigma_{\mathrm{dc}}$, as in the case of Gaussian distribution. Appropriate regions of the spectra $\varepsilon''(\omega) = \sigma'(\omega)/(\omega\varepsilon_0)$ for the mentioned distributions also coincide.

The real part of the dielectric constant (35) at $\omega \ll \mu k_2$ and $k_1 = 0$ increases logarithmically with decreasing frequency. If the equation $k_2 = \Delta\sqrt{\pi/2}$ is fulfilled for the width parameters of distributions, (35) takes the form

$$\varepsilon'(\omega) \approx 1 + \frac{N_1 q}{\varepsilon_0\Delta\sqrt{2\pi}}\left(\ln\left(\frac{2\mu^2\Delta^2}{\omega^2}\right) + \ln\left(\frac{\pi}{4}\right)\right), \tag{37}$$

coinciding with (32) for the case of Gaussian distribution. At $\omega \gg \mu k_2$ the dielectric constant (35) tends to unity.

Thus, the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra, calculated for Gaussian (20) and rectangular (22) distributions, coincide in the regions $\omega \ll \omega_{\mathrm{r}}$ and $\omega \gg \omega_{\mathrm{r}}$. At the characteristic frequency $\omega_{\mathrm{r}} = \mu k_2 = \mu\Delta\sqrt{\pi/2}$ they coincide within 8% for $\sigma'(\omega_{\mathrm{r}})$ and within 1% for $\varepsilon'(\omega_{\mathrm{r}})$.

Let us examine now a case when the distribution $G(k)$ begins not with $k = 0$, but with $k = k_1 > 0$, so that $G(k) = 0$ at $k < k_1$ and $G(k) > 0$ at $k > k_1$. Such distributions can originate, for example, in substances in which micrograins or microdomains of the restricted sizes form the random potential. We performed numeric computations of the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra for such distributions, using as examples the distributions (20), (21) and (22) with $k_0 = k_1 = 3\ \mathrm{V\ cm^{-2}}$ and $1.25\Delta = k_2 - k_1 = 6300\ \mathrm{V\ cm^{-2}}$; that is, the former distributions shifted along the k-axis by the value $k_1 = 3\ \mathrm{V\ cm^{-2}}$. In the calculations, the previous values of parameters μ, q, N_1 and N_0 of the substance have been used. The results of the calculations are shown in figures 3 and 4 by solid lines, crosses and circles for Gaussian (20), triangular (21) and rectangular (22) distributions, respectively. In these figures again the $\sigma'(\omega)$, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra, obtained at all three distributions, practically coincide, but the shape of the spectra has radically changed as compared to the spectra in figures 1 and 2. For the frequency dependence of conductivity (figure 3), the region where $\sigma'(\omega) = \text{constant} = \sigma_{\mathrm{dc}}$ now extends up to frequencies of $\sim$10 Hz and passes into the frequency area 15 Hz $< \omega/2\pi <$ 500 Hz, where $\sigma'(\omega) \sim \omega^2$. At frequencies above 500 Hz, as before, the conductivity σ' is proportional to ω up to a frequency of $\sim 10^6$ Hz, and then $\sigma'(\omega)$ ceases to depend on frequency and its value at this plateau is numerically equal to $\sigma_\infty = (N_0 + N_1)q\mu$. Now the NCL region in the frequency dependence of $\varepsilon''(\omega) = \sigma'(\omega)/(\omega\varepsilon_0)$ (figure 4) accordingly occupies frequencies 500–10^6 Hz, and below 500 Hz $\varepsilon''(\omega) \sim \omega^1$ at first, and then below $\sim$10 Hz, the frequency dependence is replaced by $\varepsilon''(\omega) \sim \omega^{-1}$. This increase of the curve $\varepsilon''(\omega)$ is associated with the dc conductivity σ_{dc}, and if $\sigma_{\mathrm{dc}} = 0$, the dependence $\varepsilon''(\omega) \sim \omega^1$ spreads out to the lowest

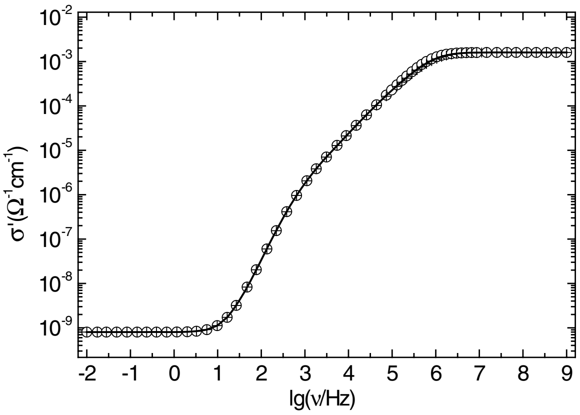

Figure 3. $\sigma'(\omega)$ spectra calculated for the Gaussian distribution (20) with $k_0 = 3$ V cm^{-2} (solid curve) and for triangular (21) and rectangular (22) distributions with $k_1 = 3$ V cm^{-2} (crosses and circles, respectively).

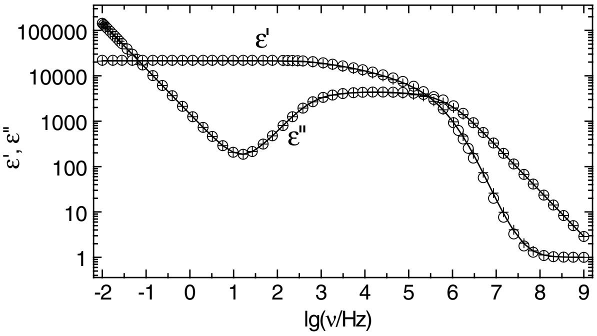

Figure 4. $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra calculated for the Gaussian distribution (20) with $k_0 = 3$ V cm^{-2} (solid curve) and for triangular (21) and rectangular (22) distributions with $k_1 = 3$ V cm^{-2} (crosses and circles, respectively).

frequencies. The frequency dependence of the dielectric constant $\varepsilon'(\omega)$ (figure 4) has, as before, a step ε' near the frequency 10^6 Hz, which is replaced by a slow monotonic increase with decreasing frequency. However, this increase stops at the level $\varepsilon' = \text{constant} \approx 22\,000$, when $\omega/2\pi \leqslant 500$ Hz. Thus, at frequencies below 500 Hz the dielectric response is now similar to the Debye response for frequencies below the Debye relaxation frequency.

Let us analyse the features of the spectra, shown in figures 3 and 4, by means of expressions (34) and (35) for the case of rectangular distribution $G(k)$. We rewrite (34) in the form

$$\sigma'(\omega) = \frac{N_1 q\omega}{k_2 - k_1} \arctan\left[\frac{\frac{\mu k_2}{\omega} - \frac{\mu k_1}{\omega}}{1 + \frac{\mu k_1}{\omega}\frac{\mu k_2}{\omega}}\right] + \sigma_{\text{dc}}. \tag{38}$$

At $\omega \ll \mu k_1 < \mu k_2$ we have the quadratic dependence of $\sigma'(\omega)$:

$$\sigma'(\omega) \approx \frac{N_1 q\omega}{k_2 - k_1} \arctan\left[\frac{\omega(k_2 - k_1)}{\mu k_1 k_2}\right] + \sigma_{\text{dc}} \approx \frac{N_1 q\omega^2}{\mu k_1 k_2} + \sigma_{\text{dc}}. \tag{39}$$

At $\mu k_1 \ll \omega \ll \mu k_2$ the dependence $\sigma'(\omega)$ becomes linear:

$$\sigma'(\omega) \approx \frac{N_1 q \omega}{k_2} \arctan\left[\frac{1-\frac{k_1}{k_2}}{\frac{\mu k_1}{\omega}+\frac{\omega}{\mu k_2}}\right] + \sigma_{\mathrm{dc}} \approx \frac{N_1 q \omega}{k_2}\frac{\pi}{2} + \sigma_{\mathrm{dc}}. \tag{40}$$

Note that the last equality is possible only at $k_1 \ll k_2$ and that the linear function (40) completely coincides with the function (36). Finally, at $\omega \gg \mu k_2$,

$$\sigma'(\omega) \approx \frac{N_1 q \omega}{k_2} \arctan\left(\frac{\mu k_2}{\omega}\right) + \sigma_{\mathrm{dc}} \approx N_1 q \mu + \sigma_{\mathrm{dc}}; \tag{41}$$

that is, σ' flattens out to the same plateau as in the previous spectra.

Thus, in the frequency range above $\omega \approx \mu k_1$ the spectra $\sigma'(\omega)$ and appropriate spectra $\varepsilon''(\omega) = \sigma'(\omega)/(\omega\varepsilon_0)$ for distributions $G(k)$, starting at $k = k_1 > 0$ ($k_1 \ll k_2$), coincide with spectra $\sigma'(\omega)$ and $\varepsilon''(\omega)$ for distributions $G(k)$, starting at $k_1 = 0$. The frequency band of the linear increase of $\sigma'(\omega)$ and the NCL area ranges from $\omega_1 \approx \mu k_1$ up to $\omega_2 \approx \mu k_2$. For the parameters used in the calculations of the spectra in figures 3 and 4, $\omega_1/2\pi \approx 480$ Hz and $\omega_2/2\pi \approx 10^6$ Hz, which coincide with bends of the curves. Below $\omega_1 \approx \mu k_1$ the dependence $\sigma'(\omega)$ in figure 3 is quadratic according to (39) for distributions $G(k)$, starting at $k = k_1 > 0$.

Expression (35) for $\varepsilon'(\omega)$ at $\omega \ll \mu k_1 < \mu k_2$ is reduced to a constant not dependent on mobility:

$$\varepsilon'(\omega) \approx 1 + \frac{N_1 q}{\varepsilon_0(k_2-k_1)} \ln\left(\frac{k_2}{k_1}\right). \tag{42}$$

At $\mu k_1 \ll \omega \ll \mu k_2$ the dependence $\varepsilon'(\omega)$ becomes logarithmically decreasing:

$$\varepsilon'(\omega) \approx 1 + \frac{N_1 q}{2\varepsilon_0(k_2-k_1)} \ln\left(\frac{\mu^2 k_2^2}{\omega^2}\right), \tag{43}$$

and at $\omega \gg \mu k_2$ $\varepsilon'(\omega) = 1$. For the parameters used in calculations of spectrum $\varepsilon'(\omega)$ the value $\varepsilon' = \text{constant} = 21\,970$ is obtained from expression (42); this coincides with the graph in figure 4.

Thus, we have shown both by means of numeric calculations and analytically that the type of the dielectric response spectra of disordered conducting medium weakly depends on the shape of distribution $G(k)$, but to a great extent depends on the positions of the lower k_1 and upper k_2 boundaries of the distribution. If $k_1 = 0$, the conductivity spectrum will be a linear function (36) in the frequency band $0 < \omega < \omega_{\mathrm{r}} = \mu k_2$ and, accordingly, this band will be the NCL region for the spectrum $\varepsilon''(\omega)$ and the logarithmically decreasing region for the spectrum $\varepsilon'(\omega)$. If $k_1 > 0$, and $k_1 \ll k_2$, the frequency dependence of conductivity will be quadratic (39) at first, and then, at frequencies $\mu k_1 < \omega < \mu k_2$, it will become the same linear function (36), and at $\omega > \omega_{\mathrm{r}}$ it will flatten out to the same plateau σ_∞, as in the case of $k_1 = 0$. Accordingly, the NCL area of $\varepsilon''(\omega)$ and the logarithmically decreasing area of $\varepsilon'(\omega)$ now will occupy frequency band $\mu k_1 < \omega < \mu k_2$. Finally, if $k_1 > 0$, and k_1 and k_2 are of the same order, then a linear region in the $\sigma'(\omega)$ spectrum and appropriate areas in the $\varepsilon''(\omega)$ and $\varepsilon'(\omega)$ spectra will disappear. It is obvious that at $k_1 > 0$ and $(k_2 - k_1)/k_1 \ll 1$ the spectra of the dielectric response are described by the expressions (9), (10), (13), (14) with $k = k_2 \approx k_1$, that is by 'Debye' frequency dependences with 'relaxation frequency' $\omega_{\mathrm{r}} = \mu k$.

Thus, our model of Drude drift of the charges localized in minima of the random potential of a disordered substance describes, at various boundary positions of the distribution function $G(k)$, both the purely 'Debye' dielectric response and responses with NCL behaviour and with 'giant' values of a dielectric constant. At the same time the shape of the distribution function of the minima walls' steepness weakly influences the dielectric response; it is sufficient that $G(k)$

is limited. This can be by a natural Gaussian distribution, or even a rectangular distribution, unlike exotic distribution functions of barrier heights between adjacent minima of potential (for example, exponential [20]), used in 'hopping' theories of ac conductivity. Moreover, closely disposed minima of potential are not required at all for our model. The principal peculiarity of the model is the use *of a distribution* $G(k)$ *of force constants* k, describing the ensemble of the forces returning charges to their equilibrium positions (model DFC), unlike the use of the probability distribution of charge carrier hops over barriers or the distribution of relaxation times (model DRT). Within the framework of our model, the NCL regime is entirely stipulated by the parameters of the random potential relief and is almost unrelated to the properties of charge carriers. This circumstance can explain the similarity of parameters of the NCL regime for many disordered substances. For example, the values of ε'' for the NCL regime for a large number of conducting glasses, melts, and crystals at various temperatures lie in a narrow interval $3 \times 10^{-3} < \varepsilon'' < 2$ [21]. Note that practically the same interval $10^{-3} < \varepsilon'' < 10$ is occupied by values of ε'' for various substances in the UDR regime [1, 2].

The temperature dependence of the dielectric response within the framework of the presented model is determined by the temperature dependences $G(k, T)$, $\mu(T)$, $N_0(T)$ and $N_1(T)$. For solids, far from temperatures of phase transitions, the random potential, 'frozen' in the lattice, weakly depends on temperature and accordingly $G(k, T) \approx$ constant. The mobility $\mu(T)$ also usually depends on temperature much more weakly than $N_0(T)$ and $N_1(T)$, so the temperature dependence of the dielectric response in such cases is conditioned mainly by the temperature dependences of conductivities $\sigma_{\mathrm{dc}}(T)$ and $\sigma_\infty(T)$. Close to temperatures of phase transitions, the dielectric response will be strongly influenced by the temperature dependence $G(k, T)$. This is of particular concern for substances with fuzzy phase transitions (relaxors) in which the polar clusters are chaotically distributed. These clusters, originating at various local Curie temperatures, have different compositions, sizes, electrical dipole moments and activation energy, so the width and the shape of distribution of the potential fluctuations, related to them, essentially depend on temperature.

If the average depth of minima of random potential is equal to $\bar{\varphi}_{\min}$, then by using the Boltzmann distribution $N(\varphi) \propto \exp(-q\varphi/k_{\mathrm{B}}T)$ we obtain for the ratio of the density of the charges, situated in potential minima, to the density of free charges

$$\frac{N_1(T)}{N_0(T)} = \frac{\int_0^{\bar{\varphi}_{\min}} \exp(-\frac{q\varphi}{k_{\mathrm{B}}T})\,\mathrm{d}\varphi}{\int_{\bar{\varphi}_{\min}}^{\infty} \exp(-\frac{q\varphi}{k_{\mathrm{B}}T})\,\mathrm{d}\varphi} = \frac{1-\exp(-\frac{q\bar{\varphi}_{\min}}{k_{\mathrm{B}}T})}{\exp(-\frac{q\bar{\varphi}_{\min}}{k_{\mathrm{B}}T})}, \tag{44}$$

or for the ratio of conductivities

$$\frac{\sigma_\infty(T)}{\sigma_{\mathrm{dc}}(T)} = \frac{N_1(T)+N_0(T)}{N_0(T)} = \exp\left(\frac{q\bar{\varphi}_{\min}}{k_{\mathrm{B}}T}\right). \tag{45}$$

This implies that if σ_{dc} follows the activation law $\sigma_{\mathrm{dc}}(T) \propto \exp(-E_d/k_{\mathrm{B}}T)$, the activation energy for $\sigma_\infty(T)$ should be less by the value of $q\bar{\varphi}_{\min}$. This means that the temperature dependences $\sigma_{\mathrm{dc}}(T)$ and $\sigma_\infty(T)$ essentially differ, and $\sigma_\infty(T)$ depends on temperature more weakly. This corresponds to the experimental data of [12, 21–23], according to which in the NCL regime the conductivity $\sigma'(\omega) = A\omega$ has weaker temperature dependence than σ_{dc}. As $A \propto N_1(T)$ (see (28), (36)), and $\sigma_{\mathrm{dc}} \propto N_0(T)$ and usually $N_0 \ll N_1$, then $\sigma_\infty = (N_0 + N_1)q\mu$ is also proportional to N_1. From (45) the average depth of random potential minima is expressed in terms of experimentally determined $\sigma_{\mathrm{dc}}(T)$ and $\sigma_\infty(T)$:

$$\bar{\varphi}_{\min} = \frac{k_{\mathrm{B}}T}{e} \ln\left(\frac{\sigma_\infty(T)}{\sigma_{\mathrm{dc}}(T)}\right). \tag{46}$$

If the mobility μ of the charge carriers in a substance is known, it will be possible to evaluate also the width of distribution of transverse dimensions $D(k)$ of potential fluctuations and an interval of depth values of the parabolic wells, corresponding to the spectral range of NCL $\omega_1 = \mu k_1 < \omega < \mu k_2 = \omega_2$. If we suppose, for example, that all parabolic wells of potential have depth close to $\bar{\varphi}_{\min}$, then $D(k) = 2x_k$, where x_k is obtained from the equation $\bar{\varphi}_{\min} = kx_k^2/2$; that is

$$D(k) = 2\sqrt{\frac{2\bar{\varphi}_{\min}}{k}}, \tag{47}$$

and the transverse dimensions of the potential wells are in the interval

$$2\sqrt{\frac{2\mu\bar{\varphi}_{\min}}{\omega_2}} < D < 2\sqrt{\frac{2\mu\bar{\varphi}_{\min}}{\omega_1}}. \tag{48}$$

On the contrary, if we suppose that the transverse dimensions of all potential wells are approximately equal to $\bar{D}$ (for example, equal to the average size of micrograins of ceramics) it is analogously possible to calculate an interval of values of depth $\varphi_{\min}$ of the parabolic wells, corresponding to the spectral range $\omega_1 < \omega < \omega_2$ of the NCL:

$$\frac{\omega_1\bar{D}^2}{8\mu} < \varphi_{\min} < \frac{\omega_2\bar{D}^2}{8\mu}. \tag{49}$$

3. Comparison of the model relationships with experimental data

The dielectric response within the framework of the presented model is determined by the parameters N_0, N_1, q, μ, and boundaries k_1 and k_2 of the distribution $G(k)$, corresponding to the spectral area of the NCL $\omega_1 = \mu k_1 < \omega < \mu k_2 = \omega_2$. However, for comparison with experimental spectra it is more convenient to express the formulae for $\sigma(\omega)$ and $\varepsilon(\omega)$ in terms of $\sigma_{\mathrm{dc}} = N_0 q\mu$, $\sigma_\infty = (N_0 + N_1)q\mu$, ω_1 and ω_2, especially because the parameters N_0, N_1, q and μ are not always known. Then, for example, (24) and (26) for the case of Gaussian distribution (20) with $k_0 = 0$ take the form

$$\sigma'(\omega) = \pi\frac{\sigma_\infty - \sigma_{\mathrm{dc}}}{2}\frac{\omega}{\omega_2}\exp\left(\frac{\pi}{4}\frac{\omega^2}{\omega_2^2}\right)\left(1 - \operatorname{erf}\left(\frac{\omega}{\omega_2}\sqrt{\frac{\pi}{4}}\right)\right) + \sigma_{\mathrm{dc}}, \tag{50}$$

$$\varepsilon'(\omega) = \varepsilon_\infty + \frac{\sigma_\infty - \sigma_{\mathrm{dc}}}{\varepsilon_0 2\omega_2}\exp\left(\frac{\pi}{4}\frac{\omega^2}{\omega_2^2}\right)E_1\left(\frac{\pi}{4}\frac{\omega^2}{\omega_2^2}\right), \tag{51}$$

and (34) and (35) for the rectangular distribution (22) turn into

$$\sigma'(\omega) = \frac{\sigma_\infty - \sigma_{\mathrm{dc}}}{\omega_2 - \omega_1}\omega\left[\arctan\left(\frac{\omega_2}{\omega}\right) - \arctan\left(\frac{\omega_1}{\omega}\right)\right] + \sigma_{\mathrm{dc}}, \tag{52}$$

$$\varepsilon'(\omega) = \varepsilon_\infty + \frac{\sigma_\infty - \sigma_{\mathrm{dc}}}{2\varepsilon_0(\omega_2 - \omega_1)}\ln\left(\frac{\omega^2 + \omega_2^2}{\omega^2 + \omega_1^2}\right). \tag{53}$$

In (51) and (53), for generality we have substituted unity by the dielectric constant ε_∞ presenting the integral contribution of high-frequency dispersion processes in ε'.

Now we will give several examples of experimental dielectric responses of various substances and their descriptions within the framework of our model.

The work reported in [24] is one of few in which the regime of 'new universality' and the plateau σ_∞ are observed in conductivity spectra $\sigma'(\omega)$ without the superposition of UDR and other phenomena[1]. In this work the electrical properties of amorphous Se, As_2Se_3 and

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[1] In [10, 16], examples of experimental spectra $\sigma'(\omega)$ with levelling off to the plateau σ_∞ for several ionic conductors are given, but procedures of removing vibrational or Debye components of the spectra were applied to reveal the plateau.

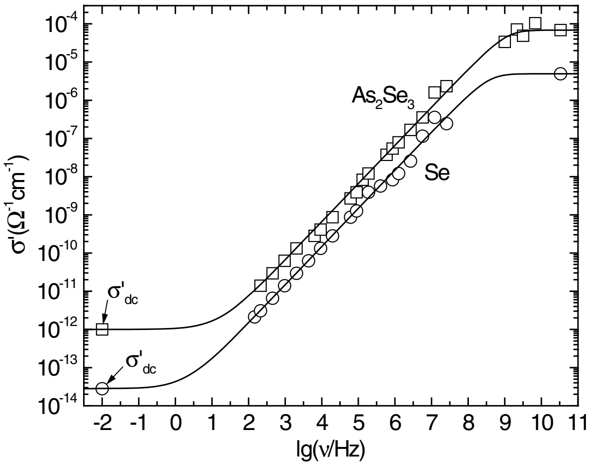

Figure 5. $\sigma'(\omega)$ spectra for Se and As_2Se_3 at $T = 300$ K according to experimental data [24] (circles and squares) and their fits using equation (52) of the proposed model with parameters given in text (solid lines).

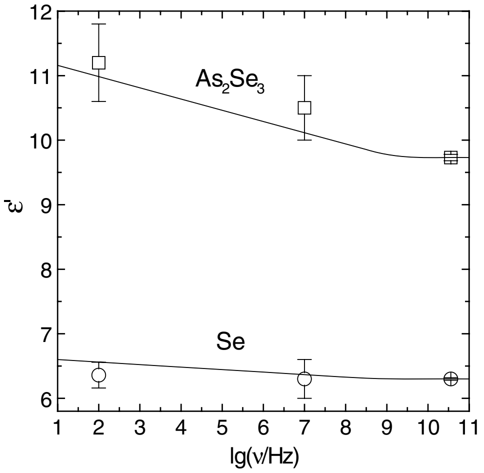

Figure 6. $\varepsilon'(\omega)$ spectra for Se and As_2Se_3 at $T = 300$ K calculated by using equation (53) of the proposed model with the same parameters as in figure 5 (solid lines), and experimental data according to [24] (circles and squares).

As_2S_3 over the 10^2–3.6×10^{10} Hz frequency range were researched, and also the values of σ_{dc} were measured. Figure 5 shows the experimental $\sigma'(\omega)$ spectra for Se and As_2Se_3 at $T = 300$ K according to [24] (circles and squares) and our fitting curves, calculated by formula (52) with $\omega_1 = 0$ (solid lines). Appropriate experimental values were taken for σ_∞ and σ_{dc}, and the frequency ω_2 was the unique fitting parameter ($\omega_2/2\pi = 5.26 \times 10^8$ Hz, $\sigma_\infty = 4.9 \times 10^{-6}\ \Omega^{-1}\ \mathrm{cm}^{-1}$, $\sigma_{dc} = 2.8 \times 10^{-14}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ for Se and $\omega_2/2\pi = 1.63 \times 10^9$ Hz, $\sigma_\infty = 6.9 \times 10^{-5}\ \Omega^{-1}\ \mathrm{cm}^{-1}$, $\sigma_{dc} = 1 \times 10^{-12}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ for As_2Se_3). $\varepsilon'(\omega)$ spectra for Se and As_2Se_3, obtained by means of formula (53) with the above-stated parameters and with the values ε_∞, equated to experimental values of ε' at frequency 3.6×10^{10} Hz are shown in figure 6. Experimental points according to [24] in figure 6 are shown by circles and squares; note that the

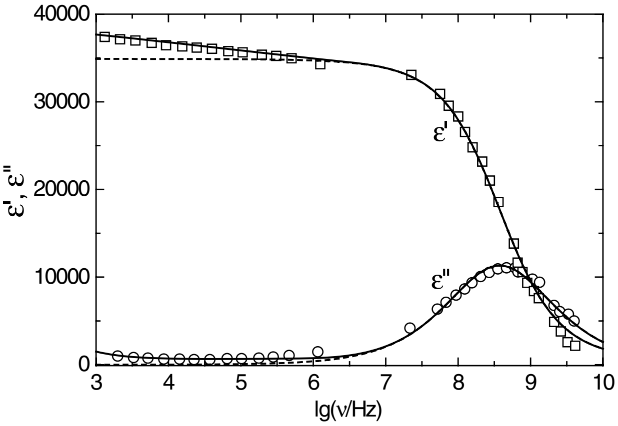

Figure 7. $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra for BNNT at $T = 396$ K according to [27] (circles and squares) and their fits using the Cole–Cole expression (54) together with equations of the proposed model (solid lines). Parameters are given in the text. Dashed lines shows fits using the Cole–Cole expression only.

experimental errors at frequency 3.6×10^{10} Hz are much less than errors at lower frequencies. In this figure the calculated curves within the limits of error coincide with experimental values; that is, the contribution $\Delta\varepsilon'$ to the dispersion ε' at frequencies below 3.6×10^{10} Hz is totally described by our model of the Drude drift currents localized in random potential minima. It is interesting that the contribution $\Delta\varepsilon'$ through higher conductivity σ_∞ in As_2Se_3 is partly compensated by greater frequency ω_2, because in this amorphous substance, unlike Se, besides the 'frozen' density fluctuations there are additional 'frozen' composition fluctuations, so the spectra of potential heterogeneities and, accordingly, of values k are wider.

The hole mobility $\mu_p = 0.34$ cm^2 V^{-1} s^{-1} in Se and $\mu_p = 4.3 \times 10^{-5}$ cm^2 V^{-1} s^{-1} in As_2Se_3 at $T = 300$ K were measured in [25] and [26], respectively. Using these data and values of σ_∞ and σ_{dc}, we calculated by means of (46) and (48) that in Se the average depth of potential minima equals $\bar{\varphi}_{min} \approx 0.49$ eV, and their transverse dimensions D lie in the interval 2000 Å $< D < \infty$, while in As_2Se_3 $\bar{\varphi}_{min} \approx 0.47$ eV and 12.5 Å $< D < \infty$ (certainly, the actual upper bounds are limited by the sizes of the samples).

The dielectric dispersion in $Ba_2NaNb_{5(1-x)}Ta_{5x}O_{15}$, $x = 0.57$ (BNNT) crystal was investigated in [27]. The authors of [27] connected the dispersion ε in this crystal at frequencies below 10^7 Hz to a soft relaxation mode and described it by the Cole–Cole expression

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon(0) - \varepsilon_\infty}{1 + (\mathrm{i}\omega\tau)^{1-\alpha}}, \tag{54}$$

where τ is the average relaxation time, and α a parameter describing the distribution of relaxation times. However, BNNT possesses significant conductivity, which should yield an additional contribution in $\varepsilon(\omega)$. We have described this contribution according to our model by formulae (52), (53) and $\varepsilon''(\omega) = \sigma'(\omega)/(\omega\varepsilon_0)$. Results of calculations of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ by these formulae together with expression (54) for BNNT at 396 K are shown in figure 7 by solid lines, and experimental data according to [27] are shown by circles and squares. Dashed lines show the results of calculations without taking into account ac conductivity. The parameters for the calculations $\varepsilon_\infty = 465$, $\alpha = 0.26$, $1/2\pi\tau = 375$ MHz and $\sigma_{dc} = 5 \times 10^{-7}$ Ω^{-1} cm^{-1} at $T = 396$ K have been taken from [27], and the frequency $\omega_2/2\pi = 1.4$ MHz was determined visually as the frequency where experimental points begin to deviate from the curve $\varepsilon'(\omega)$,

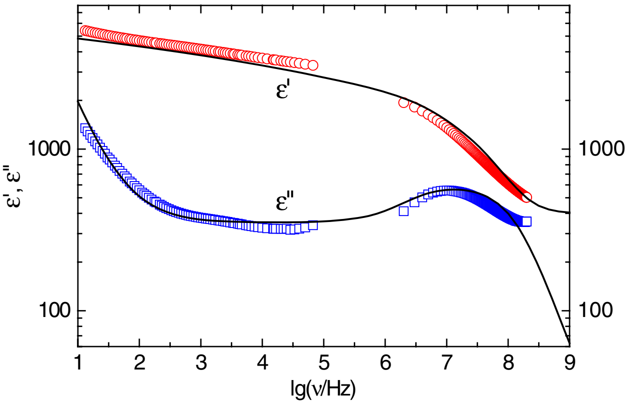

Figure 8. $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra for $(1-x)\mathrm{SrTiO_3}$–$x\mathrm{SrMg_{1/3}Nb_{2/3}O_3}$ with $x = 0.03$ at $T = 300$ K (circles and squares) and their fits using equations of the proposed model (solid lines). Parameters are given in text.

calculated according to (54). The value of conductivity $\sigma_\infty = 3 \times 10^{-4}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ was a unique fitted parameter. Figure 7 demonstrates that our model well describes the NCL regime and additional growth of ε' with decreasing frequency in the 10^3–10^6 Hz range. Note that this increment at frequency 10^3 Hz amounts to the enormous value $\Delta\varepsilon' \approx 2900$ at $T = 396$ K and becomes negligible, according to [27], already at $T = 410$ K. Our explanation of this behaviour is that on coming out of range of the fuzzy phase transition ($T_\mathrm{C} = 382$–402 K) the depth of potential fluctuations sharply decreases and the density of the localized charge carriers, i.e. the value of $\sigma_\infty \sim \Delta\varepsilon'$ (see (53)), also sharply decreases. Using values of σ_∞ and σ_dc, by means of (46) we found that at $T = 396$ K in BNNT crystal, investigated in [27], the average depth of potential wells $\bar{\varphi}_\mathrm{min} \approx 0.22$ eV.

The dielectric properties of $(1-x)\mathrm{SrTiO_3}$–$x\mathrm{SrMg_{1/3}Nb_{2/3}O_3}$ solid solution ceramics with $x = 0.005$–0.15 were studied in [28] in the 10–10^6 Hz frequency range. Giant dielectric relaxation was observed in spectra at temperatures 150–300 K. Since in these substances there are no ferroelectric phase transitions, the authors have suggested that the relaxation is connected with a local charge compensation of $\mathrm{Mg^{2+}}$ and $\mathrm{Nb^{5+}}$ ions by free carriers or ionic lattice vacancies. It is possible to explain the observed giant value of low-frequency permittivity ε' from the estimations of the authors, or by means of the Maxwell–Wagner model (at the certain selection of electrodynamic parameters of ceramics grains), or by means of model of the reorienting dipole centres consisting of $\mathrm{Mg^{2+}}$ ions associated with mobile oxygen vacancies. We emphasize that both models are concerned with mobile charge carriers.

Our model of the Drude drift currents localized in random potential minima completely describes the dielectric ε' and ε'' spectra of this giant relaxation. Figure 8 shows the experimental $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ spectra for $(1-x)\mathrm{SrTiO_3}$–$x\mathrm{SrMg_{1/3}Nb_{2/3}O_3}$ ceramics with $x = 0.03$ obtained by us in the 10 Hz–200 MHz frequency range at $T = 300$ K, and the results for $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ calculated according to our model's formulae (52), (53) and $\varepsilon''(\omega) = \sigma'(\omega)/(\omega\varepsilon_0)$. For the calculations, the spectra have been divided into two adjacent segments, corresponding to the NCL area and the widened 'Debye' area.

The parameters in (52), (53) were the following: $\omega_2/2\pi = 1.26 \times 10^8$ Hz, $\omega_1/2\pi = 2.5 \times 10^6$ Hz, $\sigma_\infty = 0.035\ \Omega^{-1}\ \mathrm{cm}^{-1}$, $\varepsilon_\infty = 400$ for the 'Debye' area and $\omega_2'/2\pi = 2.5 \times 10^6$ Hz, $\omega_1'/2\pi = 0.1$ Hz, $\sigma_\infty' = 3.1 \times 10^{-4}\ \Omega^{-1}\ \mathrm{cm}^{-1}$, $\sigma_\mathrm{dc} = 9 \times 10^{-9}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ for the NCL area. Initial values of fitting parameters σ_∞, σ_∞', and σ_dc were taken from experimental data; that

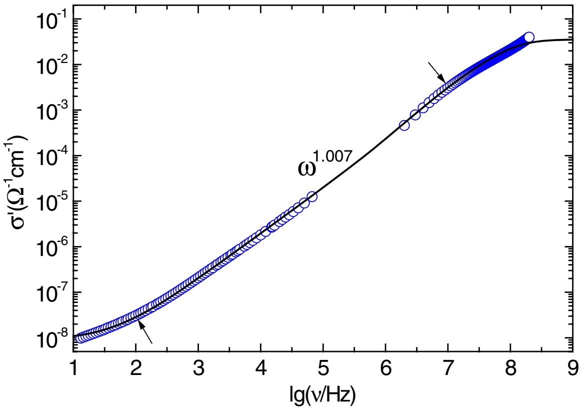

Figure 9. $\sigma'(\omega)$ spectrum for $(1-x)\mathrm{SrTiO_3}$–$x\mathrm{SrMg_{1/3}Nb_{2/3}O_3}$ with $x = 0.03$ at $T = 300$ K (circles) and its fit using equation (52) of the proposed model with parameters given in text (solid line). In the interval indicated by arrows, the curve slope correspond to $\sigma'(\omega) \propto \omega^s$ with $s = 1.007 \pm 0.007$.

is, $\sigma_\infty \approx \sigma(126\ \mathrm{MHz})$, $\sigma'_\infty \approx \sigma(2.5\ \mathrm{MHz})$, $\sigma_{\mathrm{dc}} \approx \sigma(10\ \mathrm{Hz})$. Figure 8 demonstrates good accordance of calculated curves and experimental points. Note that $\varepsilon_\infty = 400$ is close to the value $\varepsilon'(2.5\ \mathrm{cm}^{-1}) \approx 320$ for pure $\mathrm{SrTiO_3}$ obtained in [29] from infrared measurements. Since our measurements were fulfilled with the help of different instruments in the 10–100 000 Hz range (LCR meter Good Will LCR-819) and in the 1–200 MHz range (impedance meter HP 4191A), there is a small discrepancy in the experimental data. Besides, a parasitic resonance of the circuit formed by the high sample capacitance in combination with a self-inductance of the HP 4191A terminals has started to affect the spectra at highest frequencies. Figure 9 shows the experimental $\sigma'(\omega)$ spectrum and the fitting curve calculated by (52) for the ceramics investigated. In the interval indicated by arrows, $\sigma'(\omega) \propto \omega^s$ with $s = 1.007 \pm 0.007$; that is, the curve slope corresponds to the 'new universality' regime.

According to [30], the electron mobility in $\mathrm{SrTiO_3}$ is $\mu_{\mathrm{e}} = 7.5\ \mathrm{cm^2\ V^{-1}\ s^{-1}}$ at $T = 300$ K. Using this value and the above-mentioned values σ_∞ and σ_{dc}, we obtained by means of (46), (48) and (47) that in $(1-x)\mathrm{SrTiO_3}$–$x\mathrm{SrMg_{1/3}Nb_{2/3}O_3}$ ceramics with $x = 0.03$ the average depth of potential wells $\bar{\varphi}_{\min} \approx 0.4$ eV, their minimal transverse dimension $D_{\min} \approx 1.7\ \mu$m and their most probable dimension, corresponding to the 'Debye' maximum, $D_{\mathrm{Deb}} \approx 6\ \mu$m. These values agree with grain dimensions $D \sim 1$–$30\ \mu$m in samples of the $\mathrm{SrTiO_3}$ and $\mathrm{Ba}_x\mathrm{Sr}_{1-x}\mathrm{TiO_3}$ ceramics, manufactured by various technologies [31]. Note that the quite moderate number of charge carriers $N \approx 3 \times 10^{16}\ \mathrm{cm^{-3}}$ corresponds to values of parameters $\sigma_\infty = 0.035\ \Omega^{-1}\ \mathrm{cm^{-1}}$ and $\mu_{\mathrm{e}} = 7.5\ \mathrm{cm^2\ V^{-1}\ s^{-1}}$, nevertheless ensuring, according to our model, a value of giant relaxation $\Delta\varepsilon' = 5000$.

The quoted examples show that experimental data on the dielectric response of various substances—amorphous, crystalline, ceramic solid solutions—are well described by our model of the Drude drift currents localized in minima of the random potential.

4. Conclusion

A new model of the dielectric response of disordered substances is proposed. Within the framework of this model, the dielectric response of disordered substances is stipulated by the Drude drift currents localized in minima of the random electrostatic potential, 'frozen' in a substance. The random potential is caused by a static inhomogeneous charge distribution in the disordered substance. Mobile charge carriers necessary for the model will always be present, for example, owing to various uncontrollable impurities and defects in researched materials. At the same time, because of the localization of charge carriers in deep enough minima of the random potential, the dc conductivity can become extremely small, characteristic for 'good' dielectrics. The solution of the equation of motion for the Drude drift of a charge carrier with mobility μ under the summary action of a field of local parabolic potential $\varphi(x) = \varphi_{\min} + kx^2/2$ and an external alternating field shows that the appropriate dielectric response belongs to the Debye type with 'relaxation frequency' $\omega_r = \mu k$. The use of distribution functions $G(k)$ for values k of local minima of a disordered inhomogeneous material allows us to describe quantitatively both the frequency domains of the NCL ('new universality') and the cases of the giant contribution to a low-frequency dielectric constant. Owing to summation of the Debye-type responses, the model satisfies the Kramers–Kronig relations. The model equations are fulfilled for $\omega \ll 1/\tau_{sc}$, where τ_{sc} is the Drude time between carriers' scattering. The dielectric response within the framework of the model is determined by densities of free and localized charge carriers, their mobility and the distribution $G(k)$ with boundaries k_1 and k_2 linearly linked with the width of the NCL spectral range $\omega_1 = \mu k_1 < \omega < \mu k_2 = \omega_2$. This model, in our opinion, gives universal adequate representation of physical processes in any disordered substances, whose dielectric response is phenomenologically described by the Debye frequency dependence with a distribution of relaxation times (except for the Debye responses related to dipole polarization). The principal feature of the model is the use *of a distribution of force constants* k, $G(k)$, describing the forces returning charge carriers to equilibrium positions (model DFC), unlike the use of distribution of barrier heights, resulting in a distribution of probabilities of charges hops $w \sim 1/\tau$ within the limits of model of distribution of relaxation times τ (model DRT).

The description of experimental dielectric spectra by means of the proposed model allows evaluating such parameters of the random potential of disordered substances as ranges of static field values, the geometrical sizes and depths of minima of potential fluctuations and their average values.

In conclusion, we emphasize that the random potential of a disordered medium is a common phenomenon for the explanation of ac conductivity both in the NCL regime and in the UDR regime. The distribution *of electrical fields* in local minima of a mesoscale potential determines the parameters of the NCL regime, while the distribution *of energies of barriers* between local minima of atomic scale caused by the random potential determines the parameters of the UDR regime.

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