Evolution of low-frequency dispersion in solids with change of temperature

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Characteristics of Low Frequency Dispersion or QDC (quasi-DC)

Main universal features of Low Frequency Dispersion (LFD) or QDC (quasi-DC) dielectric spectra with n > 0:

1. Unlimited increase of both dielectric constant and dielectric loss towards low frequencies leading to storage of huge amount of electrical charge.
2. Weak temporal dependence of discharging current of LFD in the time domain, obeying fractional power dependence on time.
3. Thermally activated temperature dependence as from temperature dependence on certain frequency, so from frequency shift on log-log scale.
4. Stable shape of spectra on log-log scale allowing to obtaine master curve spectra on wider frequency range shifting spectra along logaryphmic frequency axis.
5. Characterised by activation energy E_a and small value of exponent n.
6. Transition to weak frequency dependent region on higher frequencies.
7. Transition from LFD with small value of n to LFD with n = 0.5-0.6 has been observed.

Main universal features of the dielectric or admittance spectra with n ≈ 0.8:

1. Weak super-linear increase of dielectric constant and dielectric loss with increasing temperature that may be fit by inverse Arrhenius law $\exp(T/T_0)$.
2. At lower frequencies temperature dependence of dielectric constant and loss are stronger showing lower value of parameter T_0.
3. Often the exponent *n* decreases with increasing temperature according to a linear law.
4. The temperature dependesncies of dielectric constant and loss is weaker than for an usual thermally activated Arrhenius processes.
5. Very weak temperature dependencies of dielectric constant and loss are observed for spectra with *n* approaching 1.
6. Temperature dependencies of dielectric loss or conductivity are frequency dependent.
7. Limitation of range of the values of complex dielectric constants and complex ac conductivity measured experimentally for dielectric response with *n* ≈ 0.8.

To explain behavior of LFD with change of temperature it is supposed, that dipoles create some structures, relaxation times of which are distributed according to power law on frequency with fractional exponent *n* less than one. Discrete function of distribution *g*(*f*) of effective dipoles on frequency *f* can be written in form

$$g(f)=\frac{\Delta \mathrm{N f}}{\Delta \mathrm{f}}=\frac{g_0}{f^{1-n}}=\frac{g_0}{10^{(1-n)i}} \qquad (1)$$

where *i* – integer, value of $0<n<1$ in a case of dominating of LFD is close to zero.

To consider character of behavior of dielectric spectra with change of temperature let take imaginary component of complex capacity C_2. Contradictory, on spectra with small value of *n*, considering spectra dominating by LFD for calculation of C_2 on certain frequency must be taken into account distribution function and for lower values of *f*.

$$C_2(f_i)=\sum_{\mathrm{k=i}}^{-\infty} g(f_k)\frac{f/f_k}{1+(f/f_k)^2} \qquad (2)$$

Some experimental LFD spectra with parameters and prediction of the model

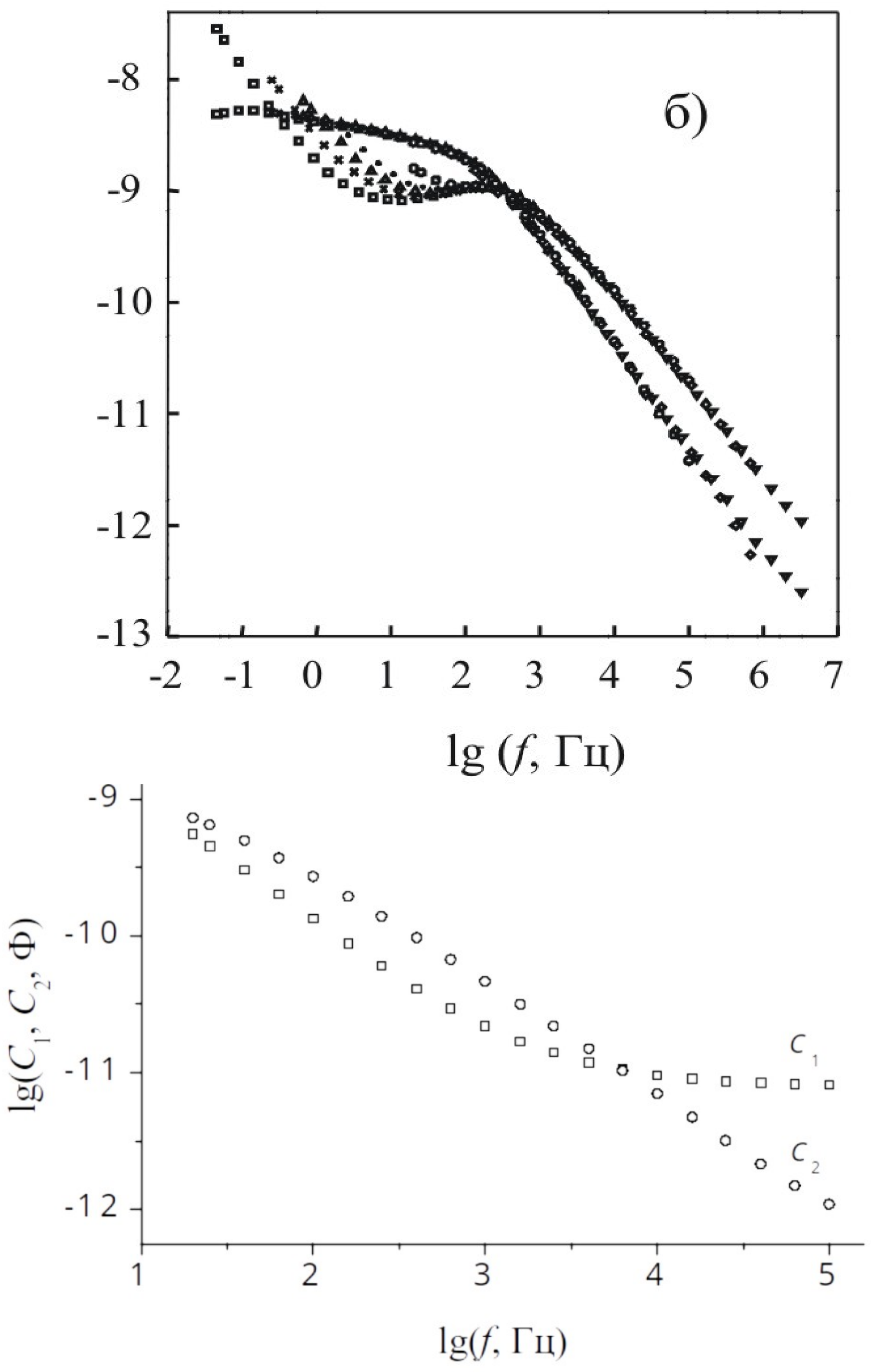

Fig. 1. Low-frequency dielectric spectra of low-resistivity GaSe layered crystal at temperature 90 K (bottom) and normalized LFD spectra to temperature 128 K (top)

Table 1. Parameters of LFD in different samples depending on temperature range

Зразок	рисунок	*T*, K	E_a	*n*
GaAs:Cr (Cr_B)	[Jon86], fig. 5	125–240	0,07–0,08	0,36
	[Jon86], fig. 12	250–335	~0,6	
GaAs (Cominco 160S)	[Jon86], fig. 6	183–263	0,21–0,23	~0,1
	[Jon86], fig. 17	295–353	0,6–0,7	0,14
GaAs (R21 F1)	[Jon86], fig. 7	90–250	0,1–0,37	0,5
	[Jon86], fig. 8	250–373	0,8	–
GaAs (A333)	[Jon86], fig. 9	300–360	0,53–0,57	0,1
GaAs:Cr (Cr-C)	[Jon86], fig. 13	215–325		temp. dependent
GaAs (Cominco 126 S73)	[Jon86], fig. 15	210–373	0,58	0,074
GaAs (R34 9F)	[Jon86], fig. 18	300–383	0,6–0,69	–
GaSe	[Fln98], fig.1-3	90–150	0,19	0,19
AsSe thin film	[Bord06], fig. 2	293–303	0,57	0,13
$Cd_{0,75}PS_3K_{0,5}(H_2O)$	[Jee97], fig.6-7	300–330	~0,45	0,1–0,13

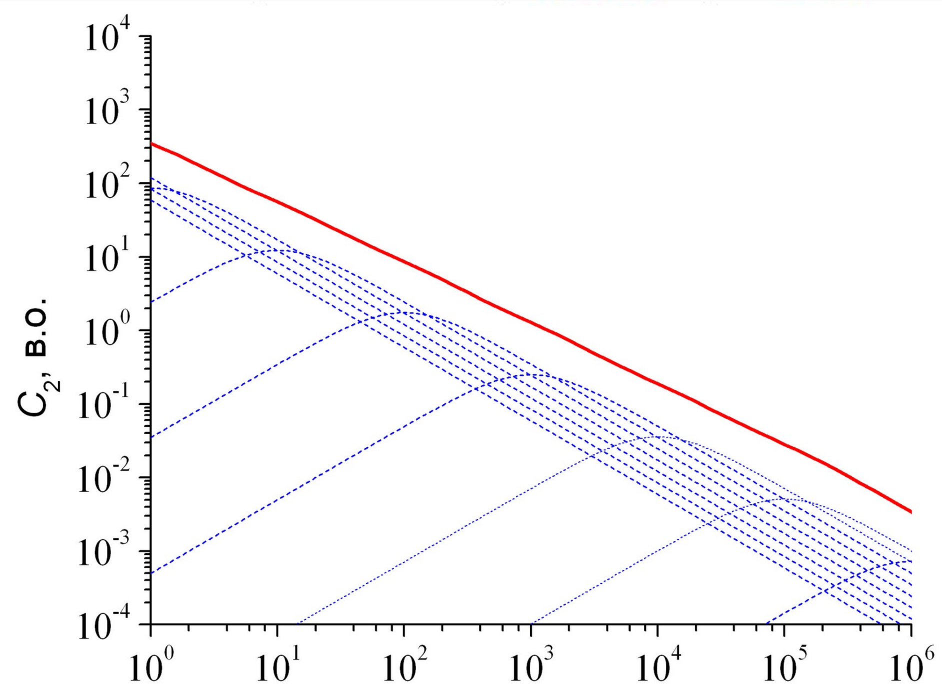

Fig. 2. Form of frequency dependence of imaginary component of capacity C_2 (solid line) created in a result of summation biased by phase on π/2 component of response of effective dipoles with power discrete distribution of concentration on relaxation times according to power law with *n*= 0.2 (dash lines)

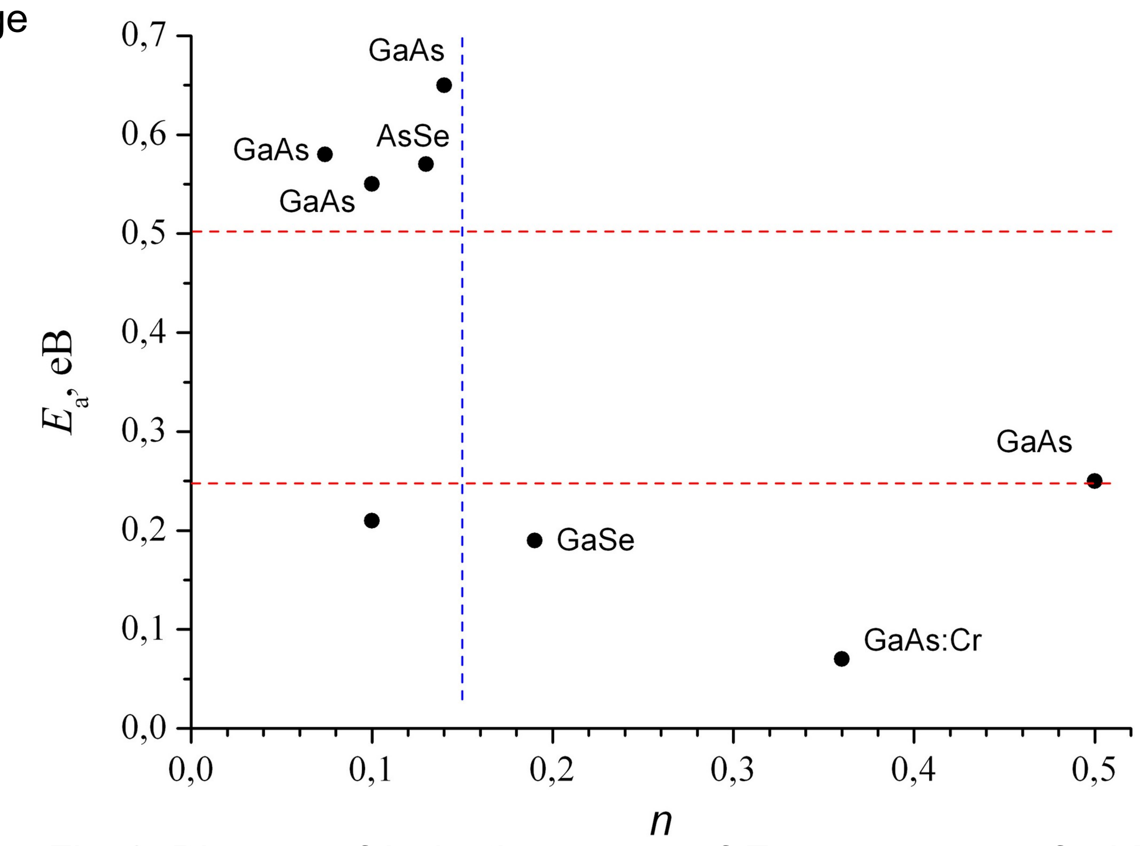

Fig. 3. Diagram of Activation energy of E_a vs exponent *n* for LFD in different semiconducting samples

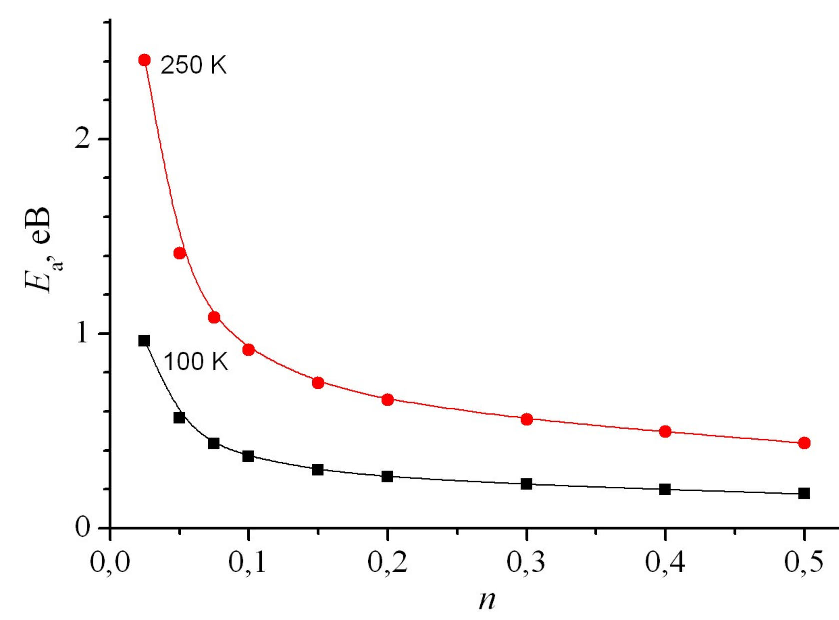

Fig. 4. Dependence of activation energy E_a on exponent *n* according to the model

Formulation of problem of thermally activated dipoles and its solution

Further let compare the contribution of component on certain frequency with contribution of effective dipoles on lower frequencies with change of temperature. Increase of imaginary part of capacity under the condition of increase of temperature on magnitude Δ*T*, caused by effective dipoles with relaxation times, that corresponds certain frequency f_i could be given in a form

$$\frac{\Delta C_{2,i}}{g(f_i)}=0{,}5(1-n)\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}\ , \qquad (3)$$

but dipoles with characteristic frequencies f_{i-k}

$$\frac{\Delta C_{2,i-k}}{g(f_i)}=(1-n)^k\ln\left(\frac{f_0}{f_{i-k}}\right)\frac{\Delta T}{T}=(1-n)^k\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}+\ln(10)(1-n)^k k\frac{\Delta T}{T} \qquad (4)$$

If take into account a contribution, caused by all components with characteristic frequencies less than change will take a form:

$$\frac{\Delta C_2}{g(f_i)}=0{,}5(1-n)\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}+\sum_{\mathrm{k=1}}^{\infty}(1-n)^k\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}+\ln(10)\sum_{\mathrm{k=1}}^{\infty}(1-n)^k k\frac{\Delta T}{T}$$

After calculation of sum Eq. 5 will take a form:

$$\frac{\Delta C_2}{g(f_i)}=0{,}5(1-n)\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}+\frac{1-n}{n}\ln\left(\frac{f_0}{f_i}\right)\frac{\Delta T}{T}+\ln(10)\frac{1-n}{n^2}\frac{\Delta T}{T} \qquad (6)$$

where $g(f_i)$ is describe by Eq. 1. To describe the behavior of LFD with change of temperature it is important to explain independence of exponent *n* on temperature. First two terms in Eq. 6 are frequency-independent, that in result would lead to increase of (1-*n*) with increase of temperature. However, third frequency independent significant term weakens this dependence. If sum of rows in Eq. 6 to limit by finite number of terms, then third term will gives contribution, that is lower for every more lower frequency (Eq. 6) and thus more strongly weakens dependence of *n* on temperature.

Conclusion

The proposed model of distribution of thermally activated effective dipoles of media with distribution of relaxation times according to power fractional dependence can explain behavior of LFD with change of temperature, giving possibility to estimate characteristic activation energy E_a and to explain the tendency of dependence of activation energy on temperature range, where LFD dominates.

The model explains temperature independence of exponent *n* leading to keeping the shape of LFD dielectric spectra on log-log scale with change of temperature.