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MATERIAL STUDY IN 7.8 - 12.4 GHZ FREQUENCY DOMAIN OF A DIVALENT LIQUID CRYSTAL

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Abstract. *An effective permittivity study was performed, in microwave range, for a substance with thermotropic liquid crystal properties. This substance, called shortly HAB, can be isotropic or presents anisotropy, corresponding to nematic or smectic type local order, depending on the exploitation temperature range. The HAB samples were simulated using a HF and microwave computer program: HFSS (Ansoft Technologies) and the frequency evolution of the effective permittivity was determined, in 7.8 - 12.4 GHz domain. The theoretical Rayleigh prediction of the effective permittivity values, valuable for a single frequency, was completed with an extended semi-empirical formula, in order to illustrate the frequency evolution of this quantity and also the permittivity resonances.*

Keywords: *effective permittivity, microwave, simulation method, theoretical prediction, permittivity resonances.*

Introduction

A category of materials of large interest in electronics are the liquid crystals. More used then the liotropic ones, the thermotropic liquid crystals present different local order, depending on substances nature. Cases are to be considered, when the same substance present more then one type of local order, specific for a liquid crystal internal configuration. This is the case which we have considered here, analysing the shortly called HAB substance.

Electrical properties (relative effective permittivity) of the HAB substance are to be determined, using a simulation method, in a frequency range where measurements are difficult to be accurately done. The simulation results are confirmed punctually by those obtained at single frequencies by measurements or are given by the accepted theories in the field.

The Rayleigh bi-dimensional theory offers the most fair results for structures like those analysed here, but doesn't explains the frequency dependence of the results. Based on results obtained from simulations for the effective permittivity resonances, the Rayleigh formula for permittivity was developed in a semi-empirical way, to include these resonances.

Effective permittivity frequency evolution

We have considered the substance with chemical formula:

C_7H_{15}-O-⌬- N=N-⌬-O-C_7H_{15},

presenting two identical flexible terminations (C_7H_{15}-O-) at the ends of a rigid core with two benzene cycles.

This substance, also called HAB, present long molecules with long axes chaotic oriented in the electric isotropic material, in the temperature range below 74.4 ^{0}C and above 124 ^{0}C. Between 74.4 and 95.9 ^{0}C, HAB presents local order of smectic liquid crystal, with parallel long molecules, grouped in parallel equidistant planes. In the temperature range of 95.9 - 124 ^{0}C long molecules remain all parallel but randomly placed in volume and HAB became a nematic liquid crystal [3], [5].

It is interesting to observe the electric anisotropy evolution when the material changes its internal order. The HAB can be considered as a composed structure, with components consisting of long molecules occupying a vacuum continuum. This structure was simulated using the High Frequency Structure Simulator (HFSS) program and exposed to a microwave testing field, inside a rectangular waveguide.

The HAB samples were considered parallelepiped shape, with macroscopic dimensions of 22.86x10.16x10 mm, filling the cross-section of the rectangular waveguide. Every sample consists of 10^6 microscopic basic cells, with local order specific for each temperature domain, having the same orientation in respect with referential axes. The three types of basic cells, for the isotropic, smectic, respectively nematic HAB, having dimensions of Å order [5], are given in figures 1-3.

The exposure conditions were: oscillation mode TE_{10} inside the rectangular waveguide and excitation level of 1 W incident normalized power at each port, defined on samples opposite faces.

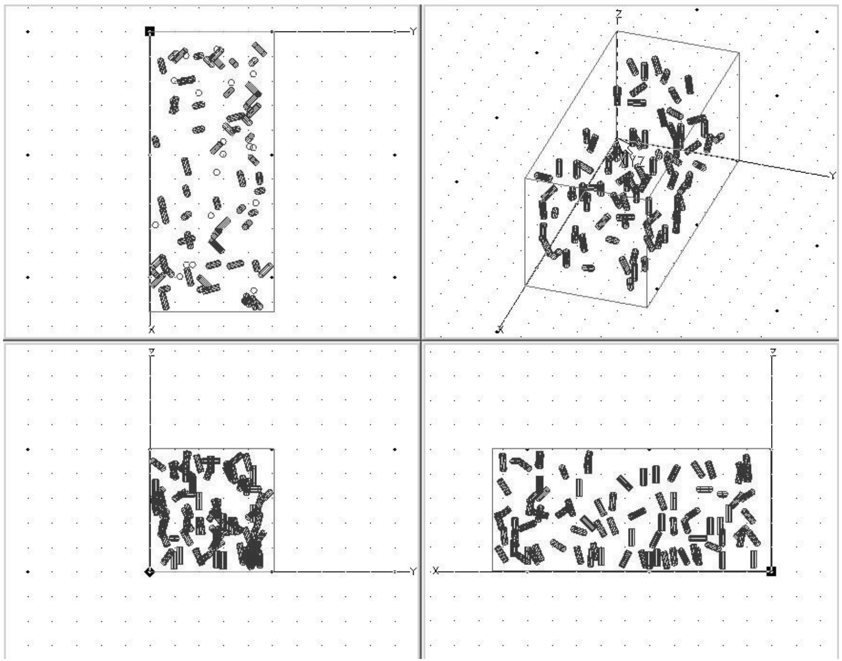

Figure 1. The basic cell for HAB isotropic, at temperatures below 74.4 ^{0}C and above 124 ^{0}C.

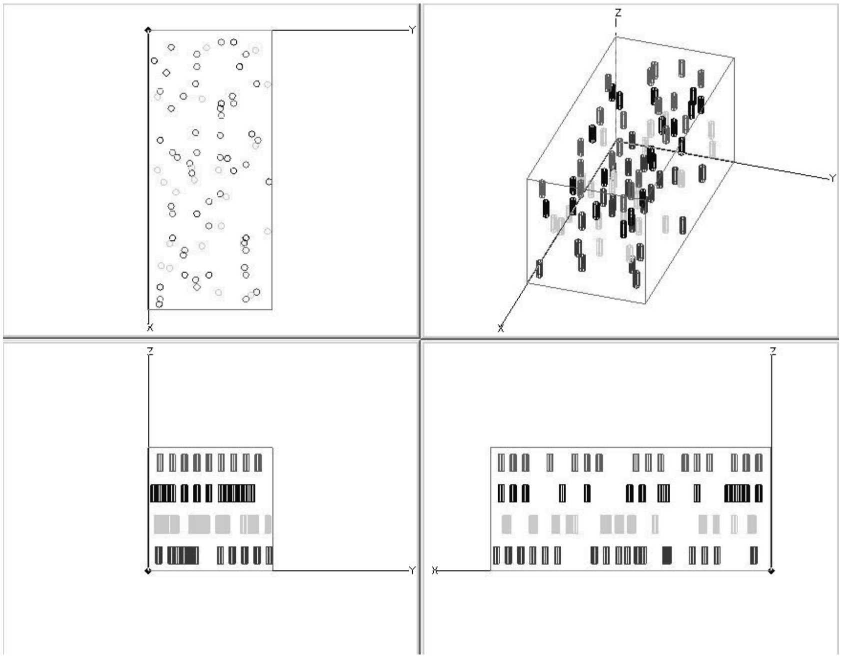

Figure 2. The basic cell for HAB smectic, in the temperature range of 74.4 - 95.9 ^{0}C.

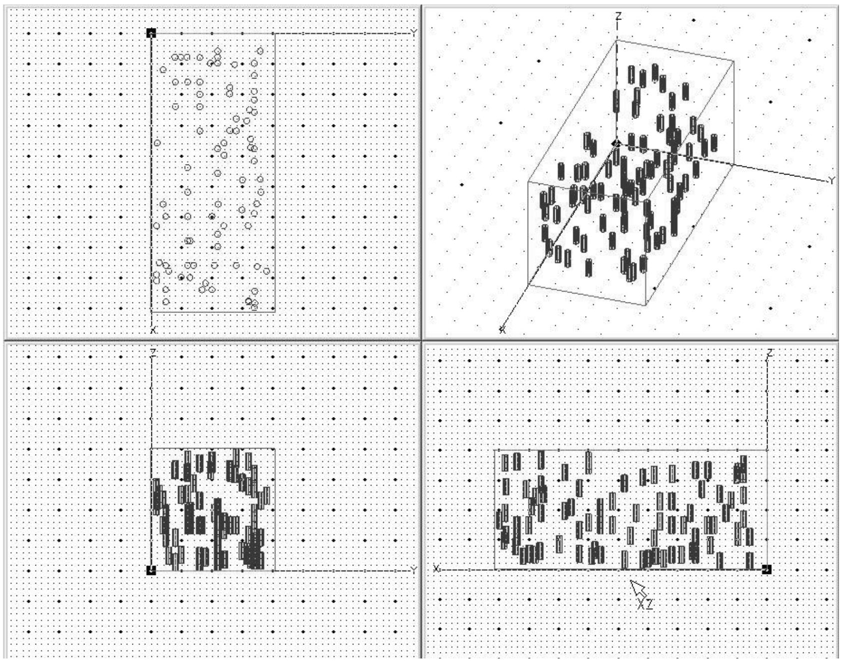

Figure 3. The basic cell for HAB nematic, in the temperature range of 95.9 - 124 ^{0}C.

The "shortcut-open end" method [4] was used to determine the *S* parameters for every field exposed sample. Then, using a proper algorithm, based on physical considerations [6], the relative effective permittivity was computed as:

$$\boldsymbol{\varepsilon}_{r,ef} = \frac{\beta^2 + \left(\frac{\pi}{a}\right)^2}{\varepsilon_0 \mu_0 \cdot \left(2\pi f\right)^2}, \qquad (1)$$

where we have denoted: β - the propagation constant of the testing field inside the material; f – frequency of the applied testing field; a – the length of the waveguide cross-section; ε_0 and μ_0 – electric permittivity, respectively magnetic permeability of the free space.

Using a simulation method with frequency sweeping, the frequency evolution of the effective permittivity was obtained for samples with different local order. Changing the testing field propagation direction, effective permittivity takes different values in the anisotropic material and thus, the electric anisotropy can be computed and its evolution with frequency monitored.

Results obtained for the HAB substance, simulated in all the temperature ranges where its local order is different, are given in figures 4 - 6.

For the cases in which HAB present liquid crystal local order and is electric anisotropic, graphs were presented for the cases of parallel, respectively transverse effective permittivity (denoted $\varepsilon_{\parallel}$, respectively $\varepsilon_{\perp}$). These correspond to the cases when the testing field $\vec{E}$ vector is parallel, respectively transverse on long molecular axes.

Electric anisotropy was also represented (figures 7, 8), calculated as:

$$\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}. \qquad (2)$$

Curves on graphs were realized in 46 points, with a new effective permittivity value for every 0.1 GHz in plus on frequency scale.

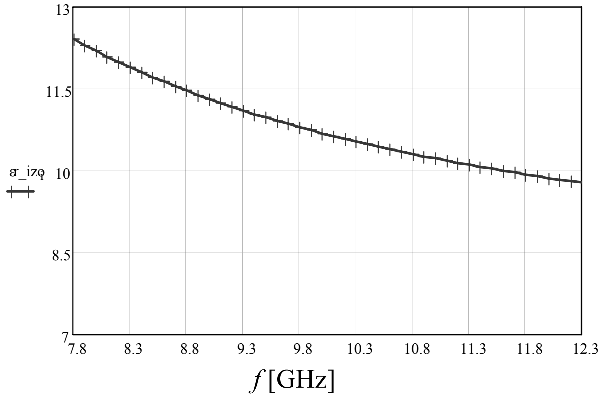

Figure 4. Evolution of $\boldsymbol{\varepsilon}_{r,ef}$ with frequency, for the isotropic HAB substance

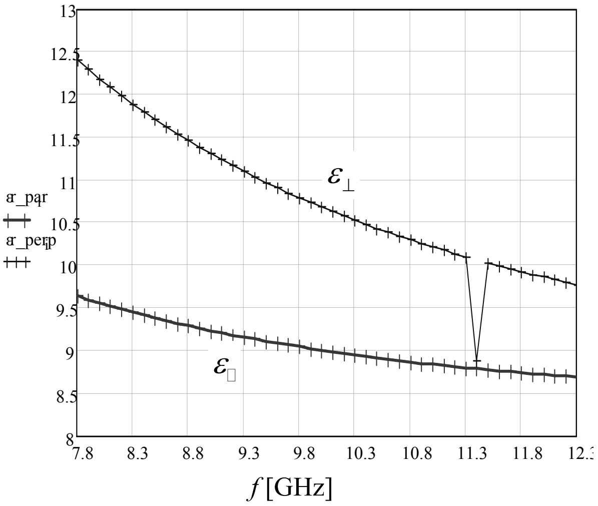

Figure 5. Evolution of $\varepsilon_{\parallel}$ (below) and $\varepsilon_{\perp}$ with frequency, for the smectic HAB liquid crystal.

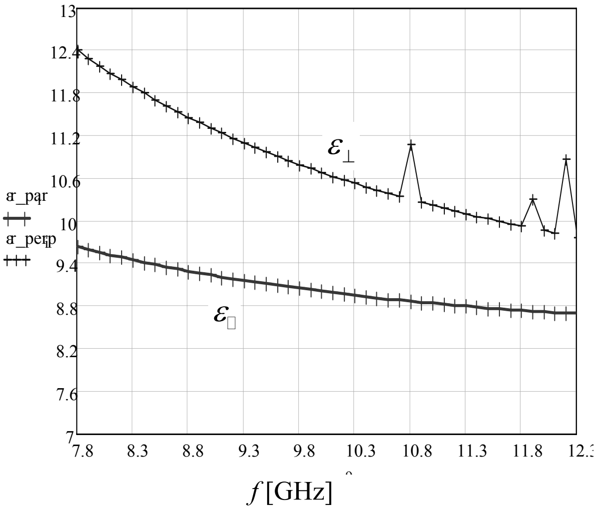

Figure 6. Evolution of $\varepsilon_{\parallel}$ (below) and $\varepsilon_{\perp}$ with frequency, for the nematic HAB liquid crystal.

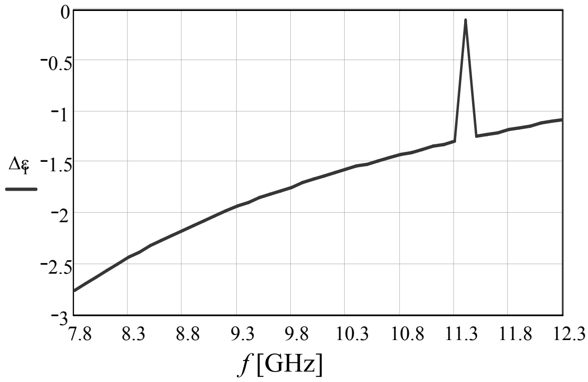

Figure 7. Electric anisotropy evolution with frequency, for the smectic HAB liquid crystal.

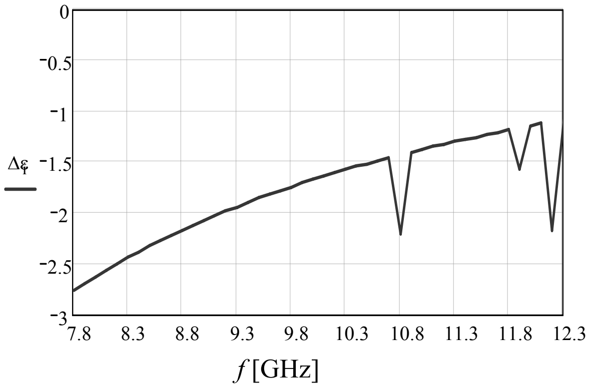

Figure 8. Electric anisotropy evolution with frequency, for the nematic HAB liquid crystal.

The relative effective permittivity takes the value of 14.2 in direct current, for the isotropic HAB (experimental determined [3]). Considering that we are working in the microwave field, the normal decreasing of the effective permittivity, with values corresponding to this frequency domain, is indicated by the graphs.

For the states of liquid crystals of the HAB, one observes that the parallel effective permittivity has lower values then the transverse one. This correspond to an optical negative liquid crystal, smectic or nematic (in both cases the $\Delta\varepsilon$ anisotropy is negative, as shown in figures 7, 8).

The smectic HAB liquid crystal presents a resonant valley, at the frequency of 11.431 GHz, which means that at this frequency the structure is more difficult to be polarized by the external field.

In comparison, the nematic HAB structure presents three resonant peaks in the considered frequency domain, at the frequencies of 10.831, 11.962 and 12.223 GHz. The physical significance of these peaks is that the structure is easier to be polarized in the vicinity of these frequencies.

For the isotropic HAB structure and for the liquid crystals HAB structures but with testing field having the $\vec{E}$ vector parallel with long molecules, resonances in microwave field are present at higher frequencies, above the 12.4 GHz considered here, as we will show in the followings.

All the microwave resonances are geometrical resonances, corresponding to a geometrical parameter of each structure, or to a combination of these parameters [2], [8], [9].

Theoretical prediction of resonances

The previous curves for effective permittivity illustrate the permittivity resonances only in the frequency domain of 7.8 - 12.4 GHz, where the precision of the method is the best. Out of this range the resonances are to be obtained using another module of the program, which gives us the resonances up to 30 GHz.

The first ten resonance frequencies in microwave field are given in table 1, separately for every internal order of the HAB substance and for every orientation of the testing field in respect with the long molecules.

Table 1. The first ten resonances frequencies in microwave range for HAB.

reson. freq.	***f***				

reson. freq.	**f_i for HAB isotropic** [GHz]	**f_i for HAB smectic** [GHz]		**f_i for HAB nematic** [GHz]	
		paral.	transv	paral.	transv
f_0	15.683	16.148	11.431	15.555	10.831
f_1	16.114	19.002	13.862	16.129	11.962
f_2	19.123	19.640	14.323	18.893	12.223
f_3	19.525	20.725	15.648	19.584	14.648
f_4	20.633	21.402	16.831	20.682	15.831
f_5	21.550	21.707	17.796	21.471	16.796
f_6	21.881	23.535	19.581	21.671	16.981
f_7	23.941	24.010	20.286	21.782	17.286
f_8	24.336	24.317	20.875	21.852	17.875
f_9	28.486	27.734	24.106	23.529	21.106

A proper theory that gives fair results for the effective relative permittivity of structures similar with those analyzed here is the Rayleigh bi-dimensional theory [1], [2]. Even if the theory doesn't illustrate the frequency dependence of the effective permittivity, its calculated values correspond to the geometrical parameters of the samples and consider samples components nature. We have decided to use this theory for single frequency checking of the simulation results for permittivity. Operating proper modifications in the effective permittivity formula, permittivity resonances can be predicted in this way.

The structure for which the Rayleigh formula is valuable is given in figure 9. The composed material is a regular array of parallel equidistant dielectric cylinders, with radium *a*, distance between basis centres *b* and distance between two successive cylinders walls *d.* Electric permittivity of cylinders material is ε_1 in direct current (DC). Cylinders are placed in a dielectric continuum volume of arbitrary shape and permittivity ε_2 in DC.

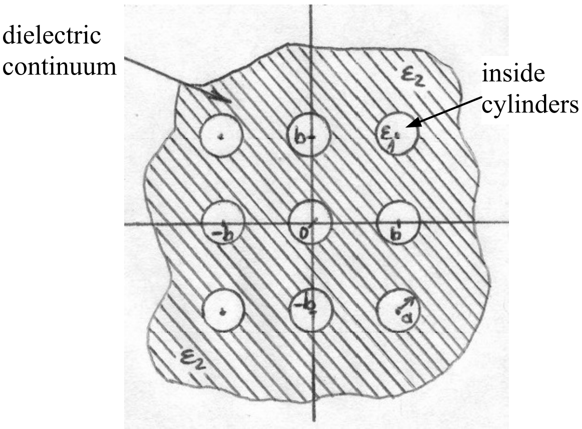

Figure 9. Cross-section of the composed material in the Rayleigh bi-dimensional theory.

The Rayleigh bi-dimensional formula for the relative effective permittivity is:

$$\boldsymbol{\varepsilon}_{r,ef}^{(3)} \square \varepsilon_2 \left[1 - \frac{2\beta_{2,1} w_4}{1 + \beta_{2,1} w_4 - 0.30583 \beta_{2,1}^{2} w_4^{4} - 0.013361 \beta_{2,1}^{2} w_4^{8}} \right] \tag{3}$$

where one denotes:

$$\beta_{2,1} = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + \varepsilon_1} \text{ and } w_4 = \pi \left(\frac{a}{b} \right)^2 . \tag{4}$$

The model can be associated the most fair to the smectic liquid crystal HAB, proper scaled, in the case of the testing field propagating with the $\vec{E}$ vector parallel with long molecules. The difference between Rayleigh model and the HAB smectic consists of not-equidistant distribution of the smectic's long molecules in the same plan.

The nematic liquid crystal structure of HAB is represented not so fair using this model, because the symmetry centres of the long molecules are not-equidistant and no more placed in parallel equidistant planes, as in smectic case.

For the isotropic HAB, the chaotic distribution and orientation of long molecules increases more the imprecision of the result, when this theoretical model is applied. It is important to remark that theoretical argumentation of the model *justifies its using* in all this cases.

Consequently, applying the Rayleigh bi-dimensional theory, the results obtained for the DC values of effective permittivity, in the case of HAB substance are given in table 2 (the long molecules and intermolecular dimensions for each internal order were taken from literature [5]). In the case of the liquid crystals states of HAB, effective permittivity was calculated in both cases: for testing field propagating with the $\vec{E}$ vector parallel, respectively transverse on long molecules. In the case of $\boldsymbol{\varepsilon}_{r,ef}$ transversal we have calculated and introduced in the formula (3) some coefficients imposed by the geometry [7], [9], making the theory still valuable.

Table 2. The DC values of relative effective permittivity for HAB, given by Rayleigh theory.

$\boldsymbol{\varepsilon}_{r,ef}$ for HAB isotropic	14.111
$\boldsymbol{\varepsilon}_{r,ef}$ parallel for HAB smectic	14.149
$\boldsymbol{\varepsilon}_{r,ef}$ transversal for HAB smectic	14.167
$\boldsymbol{\varepsilon}_{r,ef}$ parallel for HAB nematic	14.080
$\boldsymbol{\varepsilon}_{r,ef}$ transversal for HAB nematic	14.135

In microwave range, one proposes the formula (5) for the relative effective permittivity of a substance with a definite internal structure, for well known working conditions. The formula is based on Rayleigh theory and considers the geometrical resonances typical for this frequency range.

$$\boldsymbol{\varepsilon}_{r,ef} \ \square \ \varepsilon_2 \left\{ \sum_i ct_i \frac{M}{\left[\left(f-f_i\right)/f_0\right]\cdot\left(\langle h\rangle/L\right)} + \right.$$
$$+\sum_j ct_j \frac{M\cdot\left(\langle d\rangle/a\right)}{\left(f-f_j\right)/f_0} + \qquad (5)$$
$$\left. +\sum_k ct_k \frac{M}{\left[\left(f-f_k\right)/f_0\right]\cdot\langle\theta_z\rangle/\theta_0} \right\}$$

The following notations are used:

- $\beta_{2,1} = \dfrac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+\varepsilon_1}$ and $\overset{\square}{w}_{4\,av} = \pi\left(\dfrac{a}{\langle b\rangle}\right)^2$ (6)
- $M \overset{(7)}{=} \left[1-\dfrac{2\beta_{2,1}\overset{\square}{w}_{4\,av}}{1+\beta_{2,1}\overset{\square}{w}_{4\,av}-0.30583\beta_{2,1}{}^2\overset{\square}{w}_{4\,av}{}^4-0.013361\beta_{2,1}{}^2\overset{\square}{w}_{4\,av}{}^8}\right]$;
- L is the length of the long molecules;
- $\langle b\rangle$ and $\langle d\rangle$ represents the b, respectively d parameters, averaged over the whole sample volume (for all long molecules in the sample);
- $\langle h\rangle$ is the volume averaged distance between the bottom of a long molecule and the top of the successive one;
- $\langle\theta\rangle$ is the volume averaged angle between long molecular axis and the xOy plane; θ_0 represents this angle in the material polarized at saturation;
- f is the operating frequency;
- f_0 is the fundamental resonance frequency in microwave field (over 0.3 GHz), where geometrical resonance appear;
- f_i, f_j and f_k represents the resonance frequencies of superior order, corresponding to sample geometrical resonances in microwave field, where the effective permittivity presents peaks or valleys;
- ct_i, ct_j and ct_k are semi-empirical determined constants, proportional with every resonance magnitude; these constants establish character of the resonance: peak or valley, and depend on samples constituents nature;

We will have in every sum a number of terms equal with the number of geometrical resonances determined by the corresponding geometrical parameter of the sample, which is volume averaged in sum's terms.

As we have already specified, the formula (5) is valuable in microwave range, where the resonances frequencies are high (of 10^{10} GHz order). This fact ensures us that, in the vicinity of a resonance, only its corresponding term in the sum has to be kept, the other term having to low values. (Only the corresponding $1/\left(f-f_i\right)$ is less then 10^2 times lower than the others.)

We have also to mention that the quantity M includes the physical resonances of the sample (when its denominator vanishes).

Conclusions

A material with properties of liquid crystal was analyzed here (HAB) and its effective permittivity was determined, using a microwave simulation method and with help of the Rayleigh bi-dimensional theory. The considered frequency domain of the testing field was 7.8 - 12.4 GHz.

Material internal order evolves with temperature, being characteristic for an isotropic state (below 74.4 ^{0}C), then, at higher temperatures, for a smectic liquid crystal, then for a nematic liquid crystal and became finally isotropic again. Electrical anisotropy of the material changes also, presenting a frequency evolution for the liquid crystal states and being always negative for these states, fact which indicates an optical negative substance.

Both smectic and nematic states presents effective permittivity decreasing with frequency and have resonances above the considered frequency domain, in the case of testing field propagating with the $\vec{E}$ vector parallel with long molecules. Higher values of the effective permittivity are obtained when the testing field is transverse on long molecules. In this case permittivity resonances are obtained, too: resonant valley for the smectic HAB and resonant peaks for the nematic HAB, characterising the effect of lower, respectively higher material polarisation by the external field.

The first ten resonances of effective permittivity were obtained, for all HAB internal orders. All are geometrical resonances, corresponding to a geometrical parameter of the HAB exposed sample.

A theoretical prediction of effective permittivity resonances imposes, but most of the existing theories in the field give the DC permittivity values and ignore the frequency dependence of this quantity. The Rayleigh bi-dimensional theory, the most proper for the considered structures, was developed here, in order to obtain a complete formula for the effective permittivity. Dependence of material components nature and all geometrical parameters of the structure were taken into account.

Frequency dependence of the effective permittivity was also illustrated. The obtained formula allows also the effective permittivity resonances calculation and is valuable in microwave fields (over 0.3 GHz).

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