Abnormal temperature dependence of elastic properties of cuprates superconductors

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In cuprates superconductors (as polycrystal and single crystal samples) unusual hysteretic temperature behavior of elastic properties is frequently observed. In this work from the uniform point of view detailed analysis this abnormal temperature behavior of elastic properties is given. It is shown that the hysteretic temperature dependencies of elastic properties of YBa\(_2\)Cu\(_3\)O\(_{7−δ}\) crystal are strongly anisotropic and are connected to hysteretic behavior of the C\(_{3333}\) modulus while the C\(_{1111}\) and C\(_{2222}\) moduli have no such dependence. The analysis of the elastic constant tensor on the basis of the microscopic model has allowed to draw a conclusion, that the hysteretic behavior of the C\(_{3333}\) modulus of YBa\(_2\)Cu\(_3\)O\(_{7−δ}\) crystal is caused by temperature dependent renormalization of constants of interaction of apex oxygen atoms with copper atoms and are connected apparently to formation of charge ordering.

I. Introduction

Already the first investigations of high temperature superconductors\[1\][2][3][4] have revealed a variety of essential features in the temperature dependencies of velocity (\(v(T)\)) and attenuation (\(α(T)\)) of ultrasound. So for the most well investigated isostructural series of HTSC compounds (RE)Ba\(_2\)Cu\(_3\)O\(_{7−δ}\) (RE - rare earth atom) in the temperatures region 190 - 230 K step changes of \(v(T)\) have been observed\[5][6], indicating on lattice instabilities and/or phase transitions at temperatures previous to the superconducting transition. Surprising there was that at heating and cooling of a sample the temperature dependencies of velocity (attenuation) did not coincide. A distinct thermal hysteresis was observed between 60 and 230 K\[7][8][9]. Further investigations have shown that the hysteretic behavior of \(v(T)\) is observed for all isostructural series (RE)Ba\(_2\)Cu\(_3\)O\(_{7−δ}\)\[10][11], and also for the Bi\[12][13][14][15], Pb\[16], Tl\[17] HTSC compounds. Also appeared that for polycrystal samples the value of the hysteresis depends on structure of polycrystal samples. So hysteretic behavior of \(v(T)\) was observed in textured polycrystal samples\[18] and polycrystal samples with relatively large crystal grains (~50 μm)\[19]. In fine grinding, dense polycrystal samples\[20] and also polycrystal samples obtained by MPMG (melt-powder-melt-growth) method\[21] such features were not observed. The results marked above specified a possible connection of anomalous temperature dependencies \(v(T)\) with physical processes proceeding on crystal grain boundaries and, hence, for single crystals hysteretic behavior should not be observed. However, further investigations have revealed the hysteretic behavior of \(v(T)\) in (RE)Ba\(_2\)Cu\(_3\)O\(_{7−δ}\) single crystals\[22][23]. So in analyses for ultrasound with frequency 10 MHz the hysteretic behavior of the C\(_{3333}\) and C\(_{1212}\) elastic constants was observed, while hysteretic behavior of C\(_{1111}\), C\(_{2222}\), C\(_{1313}\) and C\(_{1212}\) was not observed. When studying of elastic properties of single crystals YBa\(_2\)Cu\(_3\)O\(_{7−δ}\) at 100 Hz hysteretic behavior of both elastic and torsion modulus were observed\[23]. This fact disagrees with results of\[22\], where the hysteretic behavior of the C\(_{1313}\) and C\(_{1212}\) modules was not observed (module of torsion is determined by these elastic constants). Observation in single crystals of hysteretic behavior of \(v(T)\) puts under doubt the models offered before for an explanation of these dependencies (see, for example,\[19\]). Really, it is generally believed that in cuprate superconductors anomalous temperature behavior of ultrasonic wave velocity is result from redistribution of oxygen atoms in the Cu-O chains or/and phase transitions in twinning area of crystals\[24][25]. In this connection it is necessary to note, that the hysteretic behavior of velocity of ultrasound also was observed in (RE)Ba\(_2\)Cu\(_4\)O\(_8\) polycrystals\[26][27]. This material is stable down to 850 °C and crystals are untwined. Besides while investigating of a linear expansion coefficient of YBa\(_2\)Cu\(_3\)O\(_{7−δ}\) single crystals in\[28\] was shown that in chains disordering processes of oxygen atoms proceed at temperatures above 300 °C. Let’s pay also attention that the hysteretic behavior of \(v(T)\) recently was observed by us and in Ba\(_x\)K\(_{1−x}\)BiO\(_3\) compound\[29\]. For Ba\(_x\)K\(_{1−x}\)BiO\(_3\) and (RE)Ba\(_2\)Cu\(_4\)O\(_8\) compounds the hysteretic behavior of ultrasound can not be closely related to redistribution of oxygen atoms or twinning. Earlier we offered a phenomenological model describing hysteretic temperature behavior of elastic constants of HTSC single crystals\[30\]. However the microscopic mechanism resulting in so anomalous temperature behavior of velocity of ultrasound and essential distinction in elastic properties of polycrystal and single crystal
samples of HTSC compounds remains obscure till now. The solution of this task is important for the following reasons. Recently for an explanation of the phase diagram of cuprate superconductors (remaining mysterious after 13 years of research [21]) the local micro-strain of Cu-O bonds and charge ordering (formation of striped phases) [31] were used. The micro-strain of Cu-O bonds should result in appreciable changes of elastic properties of crystal. Besides, hysteretic behavior of wave velocity of ultrasound was discovered recently in MgB$_2$ compound [32]. The huge temperature hysteresis of velocity of ultrasound was discovered recently in MgB$_2$ compound [32]. The huge temperature hysteresis of velocity of ultrasound was discovered recently in MgB$_2$ compound [32].

In the given work is shown that the hysteretic behavior of ultrasonic velocity and to analyze temperature dependencies of ultrasonic wave velocity in polycrystal crystals but as I have marked in introduction, from [22] and [23] directly it is impossible to make an unambiguous inferences about the temperature dependencies of elastic modules. First of all I show that the experimental results of [22] are consistent with results [24] quantitatively in the assumption, that anomalous temperature dependence the elastic modulus C$_{3333}$ has only.

Let’s calculate resonant frequencies of a sample at deformation of a torsion and compression in view of inertia of cross movement of elements of a sample. We will consider the case when the laboratory system of coordinates is oriented arbitrarily in reference to the crystallographic system of coordinates and the calculations are feasible by a variation method.

A. Orientations $\vec{u} \perp \vec{e} \perp \vec{k}$, $\vec{e} \perp \vec{c}$, $\vec{u} \parallel \vec{c}$

For the given experimentally investigated in [23] orientations of laboratory and crystallographic systems of coordinates $\{\vec{u}\}$ is the displacement vector of an ultrasonic wave; $\vec{c}$ is the unit vector of the crystallographic direction [001]; $\vec{k}$ is the wave vector) in a sample the deformation of a torsion and a bend is created. For evaluation of natural frequencies of oscillations of a sample as generalized coordinates it is convenient to pick the tensor $\Omega_{ij} = \frac{\partial \varphi_i}{\partial x_j}$ (see, for example, [35]), which is closely related to the deformation tensor $(u_{ij}) \Omega_{ij} = \delta_{ikl} \frac{\partial u_{ij}}{\partial x_k}$. If the tensor $\Omega_{ij}$ is known, that the deformation may be restored to within moving a sample as the whole [37]. When measuring natural frequencies of a sample value $\Omega_{ij}$ is usually small and constant. For small values of $\Omega_{ij}$ we can expand a vector and tensor of deformation into a Taylor series of $\Omega_{ij}$ and take into account linear terms only

$$u_i = B_{ijk}(x_1, x_2, x_3)\Omega_{jk};$$
$$u_{ij} = A_{ijkl}(x_1, x_2, x_3)\Omega_{kl},$$  \hspace{1cm} (1)

Then the free energy of the deformed sample may be written as follows

$$F = \frac{1}{2} \int_V C_{ijkl}u_{ij}u_{kl}dV =$$
$$= \frac{1}{2} \left( \int_V C_{ijkl}A_{ijmn}A_{klqr}dV \right) \Omega_{mn}\Omega_{rq},$$  \hspace{1cm} (2)

where $C_{ijkl}$ is a tensor of elastic modules. When writing down kinetic energy as

$$T = \frac{1}{2} \int_V \rho \dot{u}_i^2 dV = \frac{1}{2} \int_V \rho B_{ijk}\Omega_{jk}B_{iml}\Omega_{ml}dV$$  \hspace{1cm} (3)

we obtain the following expression for a Lagrangian

$$L = T - F = \frac{1}{2} \rho \dot{B}_{jklm}\Omega_{jk}\Omega_{ml} - \frac{1}{2} \tilde{C}_{sqrp}\Omega_{sq}\Omega_{rp},$$  \hspace{1cm} (4)

where

$$\dot{B}_{jklm} \equiv \int_V B_{ijk}B_{iml}dV;$$
$$\tilde{C}_{mnrn} \equiv \int_V C_{ijkl}A_{ijmn}A_{klrn}dV.$$  \hspace{1cm} (5)

The Lagrangian (4) leads to the equation of motion for $\Omega_{ij}$

$$\rho \ddot{B}_{ijml}\Omega_{ml} + \tilde{C}_{ijrp}\Omega_{rp} = 0.$$  \hspace{1cm} (6)

Now using the equations of motion (5) we may obtain the characteristic combined equations for definition of natural frequencies of oscillations of a sample

$$\left( -\omega^2 \rho \ddot{B}_{ijml} + \tilde{C}_{ijml} \right) \Omega_{ml}^0 = 0.$$  \hspace{1cm} (7)

For the analysis of experimental results [24] we shall choose a sample in the shape of the elliptic cylinder. It will allow to take into account all characteristic dimensions of experimentally studied samples [24] and to derive an expression for $\Omega_{ij}$ in the closed form, that essentially simplifies evaluation of resonant frequencies. Generally
the stress tensor is determined by the following equations and boundary conditions [36]:

\[
\begin{cases}
\frac{\partial \sigma_{ij}}{\partial x_k} = 0, \\
\delta_{kn} \delta_{lm} S_{mnpq} \frac{\partial^2 \sigma_{pq}}{\partial x_k \partial x_l} = 0, \\
\sigma_{ij} u_j = p_i,
\end{cases}
\]

(a) (b) (c)

where \(u_j\) is the normal unit vector to a surface of a sample, \(p_j\) is the force acting on unit area of a sample. For the case of an isotropic elliptic cylinder the \(\sigma_{ij}\) stress tensor is determined only by the equations (7a), which, generally speaking, have the same form as for isotropic, and an anisotropic material. Hence, the tensor of strains and for the anisotropic elliptic cylinder will be a linear function of coordinates and may be found on the basis of the analysis of boundary conditions and has the following form

\[
u_{ij} = \frac{2|\mathbf{M}|}{\pi a^2 b^2} \left[ \begin{array}{c}
\frac{b}{a} x_1 S_{ij23} - \frac{a}{b} x_2 S_{ij13}
\end{array} \right].
\]

(9)

where \(\mathbf{M}\) is the torsional moment; \(S_{ijkl}\) is the tensor of elastic compliances in laboratory system of coordinates and is related to the tensor in crystallographic system through the transformation matrix as \(S_{ij'}k'k'' = A_{i}^j A_{j'}^k A_{k'}^l A_{l}^i S_{ijkl}\) By using (1) the tensor \(\Omega_{ij}\) may be written as

\[
\Omega_{ij} = \frac{2|\mathbf{M}|}{\pi a^2 b^2} \times \begin{vmatrix}
- \frac{b}{a} S_{1313} & - \frac{a}{b} S_{2313} & - \frac{a}{b} S_{3313} \\
- \frac{a}{b} S_{1323} & - \frac{b}{a} S_{2323} & - \frac{b}{a} S_{3323} \\
\frac{a}{b} S_{1223} & \frac{b}{a} S_{2223} & \frac{b}{a} S_{3223} \\
+ \frac{b}{a} S_{1113} & + \frac{a}{b} S_{2113} & + \frac{b}{a} S_{3113}
\end{vmatrix}
\]

(10)

In the case of \(\mathbf{u} \perp \mathbf{e}, \mathbf{k} \parallel \mathbf{e}\) the equation of motion for \(\Omega_{ij}\) (6) leads to the following expression for natural frequencies of sample oscillations

\[
\omega^2 = \frac{1}{\rho L^2} \frac{C_{1313} C_{2323}}{\left(\frac{a}{b} S_{1313} + \frac{b}{a} S_{2323}\right)} \frac{12 a b}{a^2 + b^2},
\]

(11)

where \(\rho\) is the density of a sample, \(L\) is the length of a sample along a torsion axis; \(C_{ijkl}\) is the tensor of elastic modules expressed in laboratory system of coordinates. In the case \(\mathbf{k} \perp \mathbf{e}, \mathbf{u} \parallel \mathbf{e}\) the characteristic equation for resonant frequencies of a sample have been obtained in a similar way and because of complexity has been solved by numerical methods.

B. Orientations \(\mathbf{u} \parallel \mathbf{e}, \mathbf{k} \parallel \mathbf{e}\)

For the given orientation of laboratory system of coordinates longitudinal vibrations are raised in a sample.

Calculation of natural frequencies of longitudinal vibrations of a sample was performed on the basis of the equation of motion for a vector of strains:

\[
- \frac{\partial^2 u_3}{\partial t^2} + \frac{C_{ijkl} \Pi_{ij} \Pi_{kl}}{\rho (1 + \Pi_{13}^2 + \Pi_{23}^2)} \left(\frac{\partial^2 u_3}{\partial x_3^2}\right) + \frac{S^2_{ij}}{12} \frac{\partial^2 + \partial^2}{\partial x^2} \frac{\partial^2 + \partial^2}{\partial y^2} \frac{\partial^2 + \partial^2}{\partial z^2} = 0;
\]

(12)

where the \(\Pi_{ij}\) tensor is determined on the basis of the condition \(u_{ij} = -\Pi_{ij} u_{33} ; \vartheta_{11} = \Pi_{13}^2 - \Pi_{11}^2 + \vartheta_{12} = \vartheta_{21} = \Pi_{13} \Pi_{23} - \Pi_{12} ; \vartheta_{22} = \Pi_{23}^2 - \Pi_{22}^2 ; S_{ij} \) is the cross-sectional area of a sample. The equation (11) is obtained by a variation method and for isotropic materials coincides with the Relay equation [38]. The harmonic solution of the equation (11) can be deduced by the method of partitioning variables and results in the following expression for natural frequencies of oscillations of a sample

\[
\omega_n = \frac{2 \pi n}{L} \sqrt{\frac{C_{ijkl} \Pi_{ij} \Pi_{kl}}{\rho} \times \frac{\sqrt{1 + \Pi_{13}^2 + \Pi_{23}^2 + (\frac{2 \pi n}{L})^2 \frac{S^2_{ij}}{12}} \frac{\partial^2 + \partial^2}{\partial x^2} \frac{\partial^2 + \partial^2}{\partial y^2} \frac{\partial^2 + \partial^2}{\partial z^2}}}{\frac{\partial^2 + \partial^2}{\partial x^2} \frac{\partial^2 + \partial^2}{\partial y^2} \frac{\partial^2 + \partial^2}{\partial z^2}}}
\]

(13)

where \(n\) is an integer.

Let’s analyze temperature dependencies of the natural frequencies \(\omega(T)\), calculated on the basis of the expressions obtained above . First of all, one additional remark need to be made. Recently we have shown [30], that the \(C_{3333}(T)\) temperature dependencies observed in [22] may be qualitatively described within the framework of model of a strongly correlated bistable sublattice and, hence, simulation of natural frequencies \(\omega(T)\) of a sample also may be performed within the framework of the given model. However here I shall act as follows. At calculation of temperature dependencies of natural frequencies it is convenient to choose as a modelling temperature dependence of \(C_{3333}(T)\) the \(C_{3333}(T)\) dependence measured in [22], that enable not only to simulate temperature dependencies but in details allows to compare results of works [22] and [23]. The calculated temperature dependencies \(\omega(T)\) are depicted in figure 1. For all investigated orientations calculated and experimental dependencies \(\omega(T)\) are in excellent agreement. I pay attention, that for the orientation \(\mathbf{u} \parallel \mathbf{e}, \mathbf{k} \perp \mathbf{e}\) a strong temperature dependence of \(\omega(T)\) is caused by excitation of bending oscillations of a sample which natural frequency is determined by the \(C_{3333}(T)\) module .

III. Analysis of abnormal temperature dependencies of ultrasonic velocity in polycrystal samples

For calculation of effective values of a tensor of elastic
modules of polycrystal materials, it is necessary to know topological type of the heterogeneous medium. From topological point of view all heterogeneous mediums may be subdivided on matrix and statistical mediums. In matrix mediums the main phase envelops all other phases and forms connected space at any volume concentration of the main phase. In statistical medium all phases are distributed by a casual fashion not forming regular structures. For definition of topological type of the polycrystal medium we shall break volume of a sample on elementary cubic regions (domains) with the characteristic size $L_0$, equal to the average size of a crystal grain. Let the crystal grain has orthorhombic symmetry. Each separate crystal grain is oriented arbitrarily relative to the chosen axis $x_3$ determining propagation of an ultrasonic wave. All set of domains of dissection breaks up to three subsets in which domains with orientation of crystallo-

graphic axes of a crystal grain $(\vec{a}, \vec{b}, \vec{c})$ belonging to the intervals:

\[
\text{Intr}(\vec{a}) \in \begin{cases} 
0 \leq \angle(\vec{a} \vec{x}_3) \leq \frac{\pi}{4}, \frac{7\pi}{4} \leq \angle(\vec{a} \vec{x}_3) \leq 2\pi 
\end{cases};
\]

\[
\text{Intr}(\vec{c}) \in \begin{cases} 
0 \leq \angle(\vec{c} \vec{x}_3) \leq \frac{\pi}{4}, \frac{7\pi}{4} \leq \angle(\vec{c} \vec{x}_3) \leq 2\pi 
\end{cases};
\]

\[
\text{Intr}(\vec{b}) \in \begin{cases} 
0 \leq \angle(\vec{b} \vec{x}_3) \leq \frac{\pi}{4}, \frac{7\pi}{4} \leq \angle(\vec{b} \vec{x}_3) \leq 2\pi 
\end{cases}. \quad (14)
\]

The given splitting can be performed always as it is possible to show that for Euler angles the given relation is always fulfilled:

\[
\left( \frac{\pi}{4} \leq \theta \leq 0 \right) \Rightarrow \left( \varphi \geq \frac{\pi}{4}, \psi \geq \frac{\pi}{4} \right). \quad (15)
\]

It is obvious, that for the chosen subset of domains the distribution function of orientations of crystal grains

\[
\Delta \omega / \omega, \% \quad \vec{k} \parallel \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{k} \perp \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{u} \parallel \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{u} \perp \vec{c}
\]

Figure 1. Temperature dependencies of the relative resonant frequency $(\omega(T) - \omega(T_0))/\omega(T_0)$ of oscillations of a single crystal sample for various orientations of the wave vector $\vec{k}$ and the vector of displacement $\vec{u}$ of a ultrasonic wave. Lines represent simulations; points are experimental results [23]; the direction of change of temperature is shown by arrows.

\[
\Delta \omega / \omega, \% \quad \vec{k} \parallel \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{k} \perp \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{u} \parallel \vec{c}
\]

\[
\Delta \omega / \omega, \% \quad \vec{u} \perp \vec{c}
\]
$f(\theta)$ has a maximum in the intervals (13). And, texture of a sample is determined by a relation of numbers of filling of the subsets defined above. Further, for the given subset of domains we replace the tensor of elastic modules of each domain with average value of the tensor of elastic modules $< C_{ijkl} >_\eta (\eta = Intr(\vec{a}), Intr(\vec{b}), Intr(\vec{c})).$

\[ \langle C_{ijkl} \rangle = \frac{1}{8 \pi^2} \times \]
\[ \times \int_0^{2\pi} \int_0^\pi \int_0^\pi C_{ijkl} (\theta, \varphi, \psi) f(\theta, \varphi, \psi) \sin(\theta) d\varphi d\psi d\theta. \] (16)

When performing concrete calculations the distribution function $f(\theta, \psi, \varphi)$ was chosen in the form $f(\theta, \psi, \varphi) = [\cos(\theta)]^{2n} [39]$, $n$ - parameter (at $n \to 0$ or $n \to \infty$ we have random or textured distribution of orientations of crystal grains, respectively). As a result, polycrystal material can be considered as the three-phase heterogeneous medium. Topological type of medium is determined by a relation of the relative volumes $V_\vec{a}, V_\vec{b}, V_\vec{c}$ and the percolation threshold $X_c (X_c = 0.307 \pm 0.325$ for the site problem on a cubic lattice [10]). So the conditions for matrix and statistical mediums have the form $V_\vec{a} < X_c$ (matrix medium concerning phases $V_\vec{b}, V_\vec{c}$) and $V_\vec{a}, V_\vec{b}, V_\vec{c} > X_c$, respectively.

C. Finely grained dense polycrystal ceramics

For YBa$_2$Cu$_3$O$_{7-\delta}$ compound at $\delta \approx 0$ crystal grains has orthorhombic symmetry. Orientation of crystal grains is arbitrary for a nontextured sample and for relative volumes of separate phases we have the following condition $V_\vec{a} \approx V_\vec{b} \approx V_\vec{c} \approx \frac{1}{3}$. The calculation of average values of a tensor of elastic modules has shown, that for compound YBa$_2$Cu$_3$O$_{7-\delta}$ we have the following relation $< C_{1111} > \approx < C_{2222} >$. Hence, in this case polycrystal material is matrix medium and the matrix is formed by crystal grains with relative volume $V_\vec{a} + V_\vec{b} > X_c$. For the matrix material the effective elastic modulus may be calculated using the virial approach based on decomposition of an effective tensor of elastic modules in a series on relative volume of a phase $V_0 < X_c$ and in linear approach expression for the effective module looks like [39]

\[ C^* = \langle C_{1111,2222} \rangle \left[ 1 + \frac{3(\langle C_{3333} \rangle - \langle C_{1111,2222} \rangle)}{2 \langle C_{3333} \rangle + \langle C_{1111,2222} \rangle} V_c \right]. \] (17)

Using expression (14) and average values of a tensor of elastic modules (15) we have calculated temperature dependence of ultrasonic velocity for dense finely grained polycrystal materials. At calculation was considered, that the temperature dependencies of elastic modules of separate crystal grains and single crystals are identical. Results of calculation are depicted in figure 2. The main feature of temperature dependencies in this case is absence of abnormal behavior of temperature dependencies of ultrasonic velocity though the elastic modules $C_{3333}$ of separate crystal grains had pronounced hysteretic temperature dependence. Let’s pay attention, that the calculated temperature dependencies are in a good agreements with experimental ones [20].

Thus, for not textured finely grained polycrystal materials the temperature dependencies of velocity of ultrasound are determined by average values of the $< C_{1111} >$, $< C_{2222} >$ crystalline elastic modules.

D. Textured polycrystal materials

In the case of textured polycrystal material for the relative volume occupied by the crystal grains oriented, for example, in $\vec{c}$ direction the following condition is fulfilled $V_\vec{c} > V_\vec{a}, V_\vec{b}$ and the relative volumes $V_\vec{a}, V_\vec{b}$ are much less than the percolation threshold $X_c$. In this case we have the matrix topology of polycrystal medium, but the matrix is formed by crystal grains with average value of the elastic modulus $C_{3333}$. For calculation of temperature dependencies expression (14) can be used in which the following replacement $< C_{3333} > \leftrightarrow < C_{1111}, C_{2222} >$ have been performed. Calculated temperature dependencies of ultrasonic velocity for textured polycrystal samples with relative volume $V_c \approx 0.9$ are depicted in figure 2b. In this case hysteretic behavior of $\upsilon(T)$ is observed. Similar temperature dependencies were observed experimentally for textured samples in [13].

E. Materials with topology of a statistical mixture

Let’s consider polycrystal materials, when for relative volumes of separate sets of crystal grains the following condition is satisfied $V_\vec{c} \approx 0.5 > X_c; V_\vec{a} \cup V_\vec{b} \approx 0.5 > X_c$. In general case for calculation of an effective tensor of elastic moduli detailed distribution of inhomogeneities in a sample must be known [35]. However temperature dependence of ultrasonic wave velocity can be analyzed on the basis of the percolation theory. Really, from the percolation theory follows immediately [10] that the whole volume of a sample of a statistical mixture can be divided into two connected clusters with different orientation of crystal grains $Intr(\vec{c})$ and $Intr(\vec{a}), Intr(\vec{b})$, respectively. And, apparently, that properties of a statistical mixture should be symmetrical concerning replacement $< C_{3333} > \leftrightarrow < C_{1111}, C_{2222} >$. For a wave length of ultrasound the following relation $\lambda_{sound} < L_{cluster} \approx L_{sample}$ is fulfilled and elastic waves propagate in both clusters. When an elastic plane wave propagates in nonuniform material wave front will be deformed and the spatial modulation of amplitude will appear. It will take place even in a case when end faces of a sample are absolutely parallel and flat. The value of
a signal received by a detector sensitive to amplitude of strains will be proportional to a wave amplitude averaged over the detector surface

$$u_0 = k_0 \langle A \rangle_S ; \langle A \rangle_S = \frac{1}{S} \sum_i A_i(x,y) dxdy,$$

where summation is carried out over all connected clusters. Elastic waves propagating in connected clusters are detected as a single wave with effective half-width and effective standing of a maximum <t>. The average value <t> is determined by a relationship of amplitudes $$A_i$$ and times $$t_i$$ of elastic waves propagating in each cluster and in a linear approach may be represented as the ensemble average

$$\langle t \rangle = \sum_i \left( \frac{A_i}{\langle A_i \rangle_S} \right)^\gamma t_i,$$

where $$\gamma$$ is the parameter of the given detector system. Expressing the amplitude of a passed wave as $$A_i = A_0 \exp(-\alpha_i L)$$ the following expression for average velocity in a case of a statistical mixture can be obtained

$$\langle v(T) \rangle = \frac{L}{\langle t \rangle} = \left( \frac{S_0}{S} \right)^\gamma \left[ 1 + \exp(\alpha_1 L_1 - \alpha_2 L_2) \right]^{\gamma} \left[ 1 + \left( \frac{v_1(T)}{v_2(T)} \right) \exp[\gamma(\alpha_1 L_1 - \alpha_2 L_2)] \right] v_1(T),$$

where $$v_1, \alpha_1$$ are the velocity and the decay factor of an ultrasound wave, $$L_i$$ is the average size of a cluster; $$S_0$$ is the average sectional area of a cluster; $$S$$ is the effective area of a detector. As follows from equation (19), <\langle v(T) \rangle > temperature dependence is being determined by $$v_1(T)$$ or $$v_2(T)$$ temperature dependence depending on a relation of $$\alpha_i(T)$$ decay factors. I have simulated <\langle v(T) \rangle > temperature dependencies of ultrasonic wave velocity taking into account experimentally measured temperature dependencies of a decay factor [19, 22] (see Figure 2). Temperature dependence at high temperatures is determined by temperature dependence of ultrasonic wave velocity in the cluster with orientation of crystal grains $$\text{Intr}(\vec{c})$$. At decreasing of temperature the decay factor of ultrasound in the cluster with orientation of crystal grains $$\text{Intr}(\vec{c})$$ is sharply increasing while in the

![Figure 2. Temperature dependencies of the relative velocity of ultrasound \( (v(T) - v(T_0))/v(T_0) \) for a,b and \( V(T)/v(T_0) \) for c) in \( YBa_2Cu_3O_7-\delta \) polycrystal samples. Lines represent simulations; points are experimental results: a - for dense, small crystal grains sample [20]; b - textured sample [18]; c - sample with relatively large crystal grains (∼50µm) [19]; the direction of change of temperature is shown by arrows.](image)
cluster with orientation $\text{Intr}(\vec{a})$, $\text{Intr}(\vec{b})$ is decreasing [19, 22] and as a result velocity of ultrasound will be determined by the cluster of the type $\text{Intr}(\vec{a})$, $\text{Intr}(\vec{b})$. The given temperature dependence of $v(T)$ represents in some sense a combination of the temperature dependences considered above for textured and finely grained materials. It is interesting to note, that a similar sort of temperature dependencies were observed experimentally in samples with large crystal grains (50 micron) [19] (see fig. 2). Thus, quantitative analysis of experimentally observed hysteretic temperature dependencies of ultrasonic wave velocity in YBa$_2$Cu$_3$O$_{7-\delta}$ polycrystal materials (finely grained, textured, macrocrystalline) at frequencies from 100 kHz up to 20 MHz has shown, that as well as in case of single crystal samples, the hysteretic behavior is determined by hysteretic temperature behavior of the modulus $C_{3333}$ while the other elements of a tensor of elastic moduli have no abnormal temperature dependence.

F. On microscopic mechanism of hysteretic behavior

The fact of existence of strong anisotropy of hysteretic behavior of elastic modulus is important from the point of view of clearing up of the microscopic mechanism of so unusual behavior. Really, the essential contribution to anisotropy of a tensor of elastic modulus is given by interactions of atoms of the nearest coordination spheres of crystal. Having expressed a tensor of elastic moduli as a function of constants of pair interactions of atoms in YBa$_2$Cu$_3$O$_{7-\delta}$ crystal it can easily be shown that the abnormal temperature behavior of the elastic modulus $C_{3333}$ is caused by change with temperature of force constants of the apical oxygen atoms. In used approach the modulus $C_{3333}$ is determined by the interaction of apex oxygen atoms with copper and barium atoms but as abnormal temperature behavior of the elastic moduli $C_{1111}$, $C_{2222}$ is not observed, a sole opportunity is temperature renormalization of the O(4) oxygen - copper force constants. The given conclusion is consistent with experimental results [41] (also references in it) [42, 43, 44] in which features in dynamics of O(4), O(1) oxygen atoms in YBa$_2$Cu$_3$O$_{7-\delta}$ were observed also. From the point of view of study of dynamics of atoms in YBa$_2$Cu$_3$O$_{7-\delta}$ the recently published article [45] is of interest. In [45] it is shown that in the temperature region 100 - 200 K incoherent lattice fluctuations essentially surpass thermal ones. Growth of incoherent lattice fluctuations $u(T)$ were observed at temperature $T_1^* \approx 200K$ and the critical temperature $T_1^*$ is attributed in [45] to the formation of charge ordering. It is very interesting that the temperature interval in which incoherent lattice fluctuations exceeds thermal coincides with the interval in which hys-

Figure 3. Temperature dependence of incoherent lattice fluctuation in YBa$_2$Cu$_3$O$_{7-\delta}$ crystals. Points are experimental results [45]. Lines are calculated temperature dependencies: 1 - Debye model; 2,3 - model of strongly correlated sublattice without and with taking into account disordering of oxygen atoms in Cu(1)-O(1) chains. On the inset - expansivity versus temperature of untwined YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals for $\delta \sim 6.95$ (curve 1) and $\delta \sim 7$ (curve 2) [28].
teristic behavior of speed of ultrasound is observed. And the temperature of formation of charge ordering $T_\text{I}$ corresponds to the temperature of opening of a hysteresis loop. In this connection I shall pay attention to the experimental result [40] where hysteretic behavior of ultrasonic wave velocity in CuO crystals was observed. And recently in this compound charge-ordered domains were revealed [47]. Charge-ordered domains with necessity results in occurrence of correlated states of lattice degrees of freedom, that may be the reason of formation in a crystal of strongly correlated groups of atoms. Recently, we offered the model of strongly correlated sublattice for an explanation of hysteretic behavior of ultrasonic wave velocity and thermal conductivity in YBa$_2$Cu$_3$O$_7$-$\delta$ crystals [40]. Within the framework of the given model I have calculated temperature dependence $u(T) = \sqrt{\sigma(T)}$ (for more details see [40]). The calculated curve $u(T)$ describes experimental results for temperatures $T < T_\text{I}$. However at higher temperatures on the experimental dependence $u(T)$ essential growth of incoherent lattice fluctuations is observed. The given growth of fluctuations may be caused by a disordering of oxygen atoms in the Cu(1)-O(1) chains. Really, a quiet large anomaly of a linear expansion coefficient was observed in nearly optimally doped ($\delta$=0.68) of YBa$_2$Cu$_3$O$_{7-\delta}$ untwinned single crystals in the same temperature region [28]. But in fully oxygenated ($\delta$=7.0) crystals such anomaly were absent (see inset in fig. 3). I have revaluated the $u(T)$ temperature dependence with taking into account a disordering of vacancies of atoms of oxygen (Fig. 3, curve 3). One can see that the calculated temperature dependence well enough describes experimental results.

Thus, the analysis performed shows that observed experimentally incoherent lattice fluctuations in YBa$_2$Cu$_3$O$_{7-\delta}$ crystals may be caused by particular dynamics of oxygen atoms and I can speculate that the hysteretic behavior of the $C_{3333}$ elastic modulus may be attributed to the formation of charge ordering at temperatures $T < T_\text{I}$.

IV. Conclusion

The joint analysis of temperature dependencies of sample resonant frequencies and ultrasonic wave velocity as in YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal samples with various twinning structures and in polycrystal samples measured over a broad range of frequencies (100 kHz $\div$ 20 MHz), shows strong anisotropy of hysteretic behavior of elastic moduli. Namely, the $C_{3333}$ and in much smaller degree $C_{2323}$ moduli have hysteretic temperature dependence. While the moduli $C_{1111}, C_{2222}, C_{1212}$ and $C_{1313}$ do not show abnormal temperature dependence. The analysis of a tensor of elastic moduli of YBa$_2$Cu$_3$O$_{7-\delta}$ crystal on the basis of the microscopic model which is taking into account interaction of atoms of the nearest coordination spheres has shown that the anomalies in behavior of an elastic modulus are caused by a temperature-dependent renormalization of force constants of apex oxygen atoms with Cu(1), Cu(2) copper atoms and apparently are attributed to the formation of charge ordering in YBa$_2$Cu$_3$O$_{7-\delta}$ crystals.

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