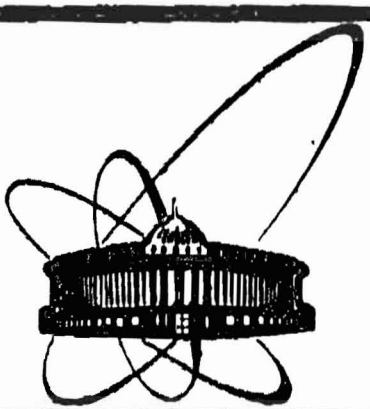

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ON THE EXPLANATION OF ANOMALIES IN INELASTIC NEUTRON SCATTERING BY HIGH-T_c SUPERCONDUCTORS

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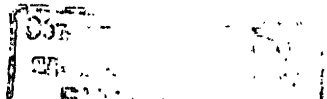
I. INTRODUCTION

Recent inelastic neutron scattering experiments in $La_{2-x}Sr_xCuO_4$ (LSCO) [1] and $YBa_2Cu_3O_{7-\delta}$ (YBCO) [2] have revealed interesting features as anomalous temperature dependence of the differential cross section in the low frequency range, its decreasing with increasing scattering momentum as well as broadening (a double-peak-like structure) of the scattering intensity near 20 meV. A strong dependence of the cross section on the oxygen content has been reported, too (see,e.g.[3]).

Despite some indications of the magnetic nature [1,2,4], there is still no clearness as to the origin of such anomalous behaviour which itself could be important for the understanding of the mechanism of high-T_c superconductivity. On the other hand, the experimental studies of the lattice dynamics provide some evidence for the existence of soft modes in LSCO [5-7] and recently in YBCO [8].In addition, the neutron diffraction data [5] bring about unusually large and highly anisotropic Debye-Waller factors for oxygen ions participating in CuO_6-octahedron rotations in LSCO (the so-called tilting mode) as well as for O(1)-ions vibrating along the x-axis in YBCO [9]. Furthermore, structural studies have shown a tendency of Cu(1)-O(1) chains in YBCO to deviate from linearity with a characteristic transverse displacement length of d~0.1 Å [10] (see Fig.1). These experimental findings suggest a more extensive study of the inelastic neutron scattering (INS) by quasilocal excitations in the frame of the anharmonic model of high-T_c superconductors [11].

In the present paper we concern ourselfes with INS on the 1d



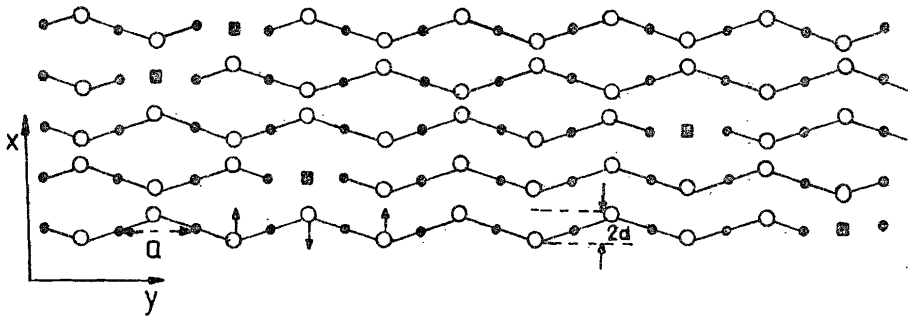


Fig.1.The Cu(1)-O(1) chain ordering in the c=0 plane. Filled circles denote Cu, open circles are O. The squares are oxygen vacancies. a ≈ b ≈ 3.88 Å and d ≈ 0.1 Å.

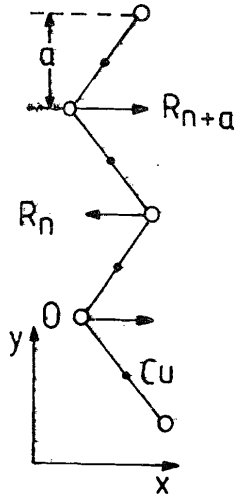


Fig.2.The representation of the Cu-O chain by rigid O-Cu-O molecules.R_n- LNC rotational displacement in the n-th unit cell. The rotational axes go through the Cu ions perpendicularly to the sheet plane.

Cu(1)-O(1) chains in YBCO. The corresponding study of INS on the tilting mode near 6÷8 meV in LSCO will be considered elsewhere.

II.Model Hamiltonian

The essential assumption of our model is that the zig-zag structure of the Cu-O chain in the z=0 plane can be represented by strongly interacting rigid O-Cu-O (OCO) "stick-like" molecules as depicted in Fig.2. Such a zig-zag ordering of the chain seems to be energetically favourable rather than a linear ordering, due to the nearest-neighbour oxygen repulsion, resulting in the unit cell doubling. The latter can be understood as freezing of the soft mode at the BZ boundary $q_0=\pi/a$, a being the lattice constant along the chain (the y-axis).

In order to describe the soft mode involving the rotational vibrations of the OCO molecules, we use the represantation of the local normal coordinate (LNC) as introduced by Pytte and Feder [12]. Then, the oxygen-ion displacements x_n in the n-th unit cell (n=1,2,..N) along the x-axis can be written in the form

$$x_n = \frac{a}{2\sqrt{2ma^2}} (R_n - R_{n+a}), \qquad (1)$$

where R_n is a rotational displacement of the OCO molecule in the n-th unit cell and m is the O ion mass. The Hamiltonian describing the oxygen ion vibrations,

$$H = \sum_n \frac{m}{2} \dot{x}_n^2 + \frac{1}{2} \sum_{n,k} \phi_{nk} x_n x_k + \frac{B}{4} \sum_n x_n^4 , \qquad (2)$$

in the LNC representation reads

$$H = \frac{1}{4} \sum_n \dot{R}_n^2 - \frac{\omega_0^2}{2} \sum_n R_n^2 + \frac{\Gamma}{4} \sum_n R_n^4 + \frac{1}{2} \sum_{n \neq k} U_{nk} R_n R_k \; ; \qquad (3)$$

here

$$\omega_o^2 = \frac{A}{m} = -U_{nn} = \frac{1}{4m}(\phi_{n+a,n} - \phi_{n,n}) > 0 \qquad (4)$$

is the frequency of unstable rotational vibrations in the harmonic approximation and

$$\Gamma = B/4m^2,\ U_{n,k} = (1/8m)[2\phi_{nk} - \phi_{n-a,k} - \phi_{n,k-a}], \qquad (5)$$

are respectively the effective rotational anharmonic and displacement coupling constants. There are two possibilities for the orientation of each OCO molecule above the structural phase transition (T_s). This rotational motion of OCO molecules is the 1d analogue of the CuO_6-tilting in LSCO.

Going over to the pseudo-spin representation (s=1/2)

$$R_n \to 2\xi_{o1} S_n^x , \qquad (6)$$

S_n^x being the Pauli x-matrix and $\xi_{o1} = \langle \psi_o | \xi | \psi_1 \rangle$ - the matrix element between the two lowest eigenstates, the Hamiltonian (3) is cast in the form

$$H = -\Omega \sum_n S_n^x + \sum_{n \neq k} J_{nk} S_n^z S_k^z , \qquad (7)$$

where $\Omega = \varepsilon_1 - \varepsilon_o$ is the excitation energy and $J_{nk} = 4(\xi_{o1})^2 U_{n,k}$ - the effective pseudo-spin interaction. It includes both the intra-chain ($J''_{n,n+a}$) and inter-chain ($J^{\perp}_{n,n+a}$) couplings. The two lowest eigenstates $\psi_{1,o}$ and eigenvalues $\varepsilon_{1,o}$ are determined by the equation

$$[\ \tfrac{1}{2}\hat{R}^2 + U(R)\ ]\ \psi_{1,o} = \varepsilon_{1,o}\psi_{1,o} \qquad (8)$$

for each OCO molecule vibrating in a double-well potential $U(R) = -(\omega_o/2)R^2 + (\Gamma/4)R^4$.

The possibility to describe these vibrations by a two-level system (TLS) (Eq.(7)) has been demonstrated numerically in the case of a realistic R^4-potential [13]; however, in what follows we prefer

simple model estimates. In the nearest-neighbour approximation the Hamiltonian (7) leads to the two-dimensional dispersion law for the anharmonic pseudo-spin mode ω_q

$$\omega_q^2 = \tilde{\Omega}^2 + \Omega\tanh(\Omega/2T)\,(J'' \cos q_{//} b + J^{\perp}\cos q_{\perp} a), \qquad (9)$$

where

$$\tilde{\Omega}^2 = \Omega^2 + (\ 2J\langle S^x\rangle\ )^2;\ J = J'' + J^{\perp}. \qquad (10)$$

For $T > T_s$

$$\langle S_z \rangle = \tfrac{1}{2}\tanh(\Omega/2T) \qquad (11)$$

and $\langle S^x \rangle = 0$, because each site is occupied with equal probability. On the contrary, for $T < T_s$, $\langle S_x \rangle \neq 0$ and $\langle S^z \rangle$ is no longer temperature dependent being equal to $\Omega/2J$. We note that the dispersion along the z-axis has been neglected in Eq.(9) due to the large distance between basal planes bearing Cu-O chains. For the structural phase-transition to occur it is obvious that the condition $\Omega/2J < 1$ has to hold. It should be mentioned that for real anisotropic systems (e.g. YBCO) it is $J'' > J^{\perp}$, whereby, owing to the predominant in-phase ordering of neighbouring chains (see, Fig.1), $J^{\perp} > 0$ also holds. Consequently, the coherence length along the x-axis is shorter than the corresponding one along the chain (the y-axis).

III. Differential cross section

For the sake of simplicity we consider the nuclear neutron scattering on oxygen ions participating in low-frequency rotational vibrations of stick-like OCO molecules, only, i.e., we neglect the explicit effect of other phonons in the same frequency range. As we are to see below, such approach is justified by the fact of possible distinction between peculiar rotational and

phonon degrees of freedom as properly revealed in the neutron scattering intensity of the system. In this context it is believed that the difference of the neutron spectra for $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_6$ can be studied at least qualitatively.

We start from the general expression of the neutron double-differential cross section per unit solid angle O and unit interval of outgoing energy in the very well known form

$$\frac{1}{N}\frac{d^2\sigma}{dOdE} = \frac{p}{p_o} S(\vec{\varkappa},\omega), \qquad (12)$$

where $\hbar\vec{\varkappa} = \vec{p}_o - \vec{p}$ and $\hbar\omega = E_o - E$ are momentum and energy transfers, p_o, $E_o = p_o^2/2m$ and p, $E = p^2/2m$ being neutron initial and final momenta and energies, respectively. The corresponding Van Hove function for neutron scattering by oxygen ion vibrations of rotational type is given by [14]

$$S(\vec{\varkappa},\omega) = \frac{1}{N}\sum_{n,k} a_n a_k e^{i\vec{\varkappa}(\vec{n}-\vec{k})} e^{-2W(\vec{\varkappa})} \cdot \frac{[1+N(\omega)]}{\pi} \cdot Im[\chi_{nk}(\vec{\varkappa},\omega)], \qquad (13)$$

where a_n is the scattering length for the n-th nucleus, $\exp[-2W(\vec{\varkappa})]$ is the anharmonic Debye-Waller factor and $[1+N(\omega)]$- the Bose factor. For the one-quantum scattering process the above introduced displacement correlation function

$$\chi_{n,k}(\vec{\varkappa},\omega) = \langle\langle \vec{\varkappa}\vec{x}_n | \vec{\varkappa}\vec{x}_k \rangle\rangle =$$

$$= \varkappa_x^2 \frac{1}{2m_o} \cdot \frac{1}{4} \langle\langle (\vec{R}_n - \vec{R}_{n+a}) | (\vec{R}_k - \vec{R}_{k+a}) \rangle\rangle_\omega \qquad (14)$$

in the pseudo-spin representation (6) reads

$$\chi_{n,k}(\vec{\varkappa},\omega) = \frac{\varkappa_x^2}{2m}(2\xi_{01})^2 \cdot \frac{1}{N}\sum_q e^{i\vec{q}(\vec{n}-\vec{k})} \sin^2(\frac{\vec{q}\vec{a}}{2}) \chi_q^{xx}(\omega), \qquad (15)$$

where

$$\chi_q^{xx}(\omega) = -\langle\langle S_q^x | S_{-q}^x \rangle\rangle_\omega \qquad (16)$$

is the resonant (xx-component) Green function.

We note that the contribution of the relaxational (zz-component) Green function $\chi_q^{zz}(\omega)$, being of entirely quasi-elastic nature, is omitted in Eq.(15) as it is predominantly responsible for the central peak dynamics (i.e. the zero-frequency range) in addition being weighted by the small pseudo-spin form-factor $4|(\xi_{00})^2 - (\xi_{11})^2| \ll 1$.

For the coherent scattering from Eqs. (13)-(16) one obtains

$$S_{coh}(\vec{\varkappa},\omega) = a_{coh}^2 \Delta(\vec{\varkappa}-\vec{q}) e^{-2W(\varkappa)} \cdot \frac{\varkappa_x^2}{2m}(2\xi_{01})^2 \sin^2(\frac{\vec{\varkappa}\vec{a}}{2}) \cdot$$

$$\cdot \frac{[1+N(\omega)]}{\pi} Im[\chi_q^{xx}(w+i\delta)]. \qquad (17)$$

The corresponding scattering function is given by the expression

$$S_{inc}(\vec{\varkappa},\omega) = \frac{\sigma_{inc}}{4\pi} e^{-2W(\varkappa)} \cdot \frac{\varkappa_x^2}{2m}(2\xi_{01})^2 \cdot \frac{[1+N(\omega)]}{2\omega} G(\omega), \qquad (18)$$

where the weighted spectral density of vibrational states (DOS) $G(\omega)$ is defined as follows:

$$G(\omega) = \frac{1}{N}\sum_{\mathbf{q}} \sin^2(\frac{\vec{q}\vec{a}}{2}) \; 2\omega \; \frac{1}{\pi} Im[\chi_q^{xx}(\omega+i\delta)] =$$

$$= \frac{1}{N}\sum_{\mathbf{q}} \sin^2(\frac{\vec{q}\vec{a}}{2}) \Omega \langle S^z \rangle \delta(\omega-\omega_q). \qquad (19)$$

For polycrystalline samples, the incoherent approximation for the coherent part (17) can be applied. Therefore, the total inelastic scattering function becomes

$$S(\vec{\varkappa},\omega) = (a_{coh}^2 + \frac{\sigma_{inc}}{4\pi}) e^{-2W(\varkappa)} \cdot \frac{\bar{\varkappa}^2}{2m}(\frac{2\xi_{01}}{2\omega})^2 [1+N(\omega)]G(\omega). \qquad (20)$$

In order to compare the obtained results (17)-(20) with experiments [1,2] we also introduce the ordinary DOS

$$Z(\omega)= \frac{1}{N}\sum_{q}\delta(\omega-\omega_q). \qquad (21)$$

After continuumization we arrive at the expression

$$Z(\omega) \approx 2\omega \begin{cases} \dfrac{\coth\frac{\tilde{\Omega}}{2T}}{\tilde{\Omega}\sqrt{J''J^{\perp}}}F(k,\frac{\pi}{2}), & 0<k<1, \\ \dfrac{1}{\sqrt{(\omega^2_{max}-\omega^2)(\omega^2-\omega^2_{min})}}F(\frac{1}{k},\frac{\pi}{2}), & 1<k<\infty, \end{cases} \qquad (22)$$

where $F(k,\frac{\pi}{2})$ is the complete elliptical integral of the first kind

$$F(k,\tfrac{\pi}{2})= \int_0^{\pi/2}\frac{d\varphi}{\sqrt{1-k^2\sin^2\varphi}}, \qquad (23)$$

in which

$$k = \frac{\sqrt{(\omega^2_{max}-\omega^2)(\omega^2-\omega^2_{min})}}{2\tilde{\Omega}\sqrt{J''J^{\perp}}}\coth\frac{\tilde{\Omega}}{2T} \qquad (24)$$

and

$$\omega^2_{\substack{max\\min}} = \tilde{\Omega}^2 \pm (J''+J^{\perp})\tilde{\Omega}\tanh\frac{\tilde{\Omega}}{2T}. \qquad (25)$$

The spectral function $Z(\omega)$ exhibits two strong peaks (logarithmic singularities from the condition $k=1$) at

$$\omega^2_{\pm} = \frac{\omega^2_{max}+\omega^2_{min}}{2} \pm \left[\frac{(\omega^2_{max}-\omega^2_{min})^2}{4} - 4\tilde{\Omega}J''J^{\perp}\tanh^2\frac{\tilde{\Omega}}{2T}\right]^{1/2}. \qquad (26)$$

Thus, the anisotropic dispersion law (9) leads to "splitting"of the DOS (see Fig.3):

$$\omega^2_{+}-\omega^2_{-} = (J''-J^{\perp})\tilde{\Omega}\tanh\frac{\tilde{\Omega}}{2T}. \qquad (27)$$

This splitting is strongly temperature dependent as a mere consequence of the peculiar factor $\tanh(\tilde{\Omega}/2T)$ coming from the pseudo-spin representation. We note that such a strong temperature dependence cannot be obtained within usual harmonic approximation. By assuming $\Omega \approx 20$meV [15], one can estimate that below a temperature near 100K the splitting (27) becomes temperature independent due to saturation of $\tanh(\tilde{\Omega}/2T)$. That may be

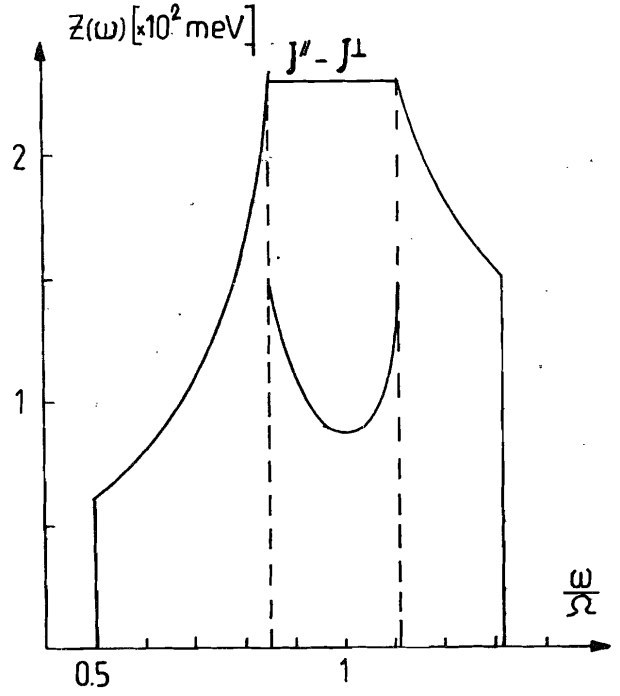


Fig.3. The spectral density of states (DOS) $Z(\omega)$.

interpreted as an order-disorder type instability at T_s~ 100K which might cause favourable conditions for superconducting transition at $T_c < T_s$, e.g. at $T_c \approx$ 90K [16]. In this context it is noteworthy that some experimental findings indicate the existence of an order-disorder type instability near 120K [17] as possibly related to the onset of superconductivity at 90K.

IV. Discussion

In comparison with the ordinary DOS $Z(\omega)$ (21) the weighted DOS $G(\omega)$ (19) comprises the additional factor $\langle S^z \rangle = (1/2)\tanh(\omega/2T)$ which gives rise to the peculiar temperature dependence. Besides, there appears the phase-like factor $\sin^2(\vec{q}\vec{a})$, strengthening the scattering intensity in the vicinity of the wave vector $q \approx q_{BZ} \approx \pi/a$ corresponding to off-phase rotations of neighbouring OCO molecules. The latter can be observed experimentally as anomalous $\varkappa$-dependence behaviour. That can be predominantly revealed in the coherent scattering when $\vec{\varkappa} = 2\pi\vec{\tau} + \vec{q}$ and $\sin^2(\vec{q}\vec{a}/2) \to \langle \sin^2(\vec{\varkappa}\vec{a}/2) \rangle_\theta = (1/2)[1+(\sin\varkappa a/\varkappa a)]$ ($\vec{\tau}$ is the reciprocal lattice vector and the symbol $\langle \ldots \rangle_\theta$ stands for the orientational averaging over $\vec{\varkappa}$, θ being the angle between the chain axis and $\vec{\varkappa}$). Therefore, the pseudo-spin model presented as accounting strongly anharmonic vibrations of oxygen ions in zig-zag chains is able to explain qualitatively the observed neutron scattering anomalies in high-T_c superconductors LSCO and YBCO.

Within this model the double-peak structure as observed in scattering intensity of YBCO can be related to highly anharmonic and anisotropic OCO vibrations of rotational type. So as a result of gradual decreasing of the oxygen content, with O- ions being removed from the chains in YBCO, this vibrational mode vanishes, thus causing drastic changes in the phonon spectrum as has been experimentally observed in neutron scattering intensities [18].

In our view, the main conclusion coming from the results presented is an alternative explanation of some peculiarities in the inelastic neutron scattering in high-T_c superconductors LSCO and YBCO which recently have been proposed to be of magnetic origin [1-3]. A more detailed comparison of our theoretical results with experimental data demands account of real structures of LSCO and YBCO. Namely, in reality a certain ammount of oxygen vacancies always exists. This leads to finite chain-lentghs and off-phase ordering between neighbouring chains. For instance, in $YBa_2Cu_3O_{7-\delta}$ with $\delta \approx 0.1$, the coherent lengths along the x-and y-axes can be estimated to be about ~6Å and ~20Å (see Fig. 1), respectively. Thus, the c=0 plane resembles a glass-like system. Therefore, an additional averaging in Eqs. (17)-(20) over possible chain-configurations should be performed. This, being beyond the scope of this paper, will be the subject of further consideration. This short-range coherent order may effect in such a way that the temperature dependence of the scattering intensity, proportional to $G(\omega)$, possibly weakens, resulting in less sharp peaks than as in Fig.3. Additional smearing out will be caused by the finite resolving ability of the experiment (typically of about 1÷4%).

One of the authors (S.S) would like to thank the Laboratory of Theoretical Physics ,JINR for hospitality.

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Плакида Н.М. и др. E17-88-822

К объяснению аномалии неупругого рассеяния нейтронов на высокотемпературных сверхпроводниках

Рассматривается ядерное неупругое рассеяние нейтронов на сильно ангармонических колебаниях ионов кислорода в высокотемпературных сверхпроводниках. На основе псевдоспиновой модели для описания ротационных колебаний O-Cu-O молекул получены выражения для когерентного и некогерентного сечений рассеяния. Дается объяснение наблюдаемых на эксперименте аномальных температурной и угловой зависимостей интенсивности неупругого рассеяния нейтронов в высокотемпературных сверхпроводниках LSCO и YBCO.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна 1988

Plakida N.M. et al. E17-88-822

On the Explanation of Anomalies in Inelastic Neutron Scattering by High-T_c Superconductors

The nuclear inelastic neutron scattering on highly anharmonic vibrations of oxygen ions in high-T_c superconductors is considered. In the framework of a pseudo-spin model associated with rotational vibrations of O-Cu-O "molecules" both coherent and incoherent scattering cross sections are evaluated. A possible explanation of experimentally observed anomalous temperature and angular dependences of inelastic neutron scattering intensity in high-T_c superconductors LSCO and YBCO is proposed.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Communication of the Joint Institute for Nuclear Research. Dubna 1988