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A linear spin-wave theory for $LiNiPO_4$ utilizing a nonextensive distribution

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This work is concerned about the lithium-nickel phosphate, $LiNiPO_4$, a very important magnetic-electric material due to its promising applications to tailor the next-generation lithium-nickel batteries. In the Hamiltonian operator, it is considered the interactions of Ni^{2+}, the unique magnetic ion in the phosphate. Specifically, it is taken into account the Heisenberg interactions along with the single-ion anisotropy term. Furthermore, the utilization of the Holstein-Primakoff formalism leads to spin-wave scattering. To investigate the spin-wave intensities, we utilize the nonextensive probability distribution what originates some bulges in the curves of the intensities.
Keywords: Spin waves, neutron diffraction, magnetic materials, quantum statistical mechanics.

Una teoría lineal de ondas de espín para el $LiNiPO_4$ utilizando una distribución no extensiva

Este trabajo se basa en el estudio del fosfato de Li-Ni, $LiNiPO_4$, un material magnético-eléctrico promisorio en la próxima generación de baterias de litio-niquel. El operador hamiltoniano considera las interacciones del único ión magnético, Ni^+, en el fosfato. Específicamente se toman en consideración las interacciones de Heisenberg con los términos de anisotropía del ión. Asimismo, la utilización del formalismo de Holstein-Primakoff conduce al scattering de las ondas de espín. Para investigar las intensidades de ondas de espín, utilizamos la distribución de probabilidad no extensiva que origina unos bultos en las curvas de las intensidades.
Palabras claves: Ondas de espín, difracción de neutrones, materiales magnéticos, mecánica estadística cuántica.

Nowadays, the studies to obtain low-cost and non-contaminant rechargeable batteries is very intense, for these investigations have become vital to prevent the pollutant materials which destroy our environment. Particularly, two phosphate compounds, very important when tailoring the next-generation batteries, have focussed the researchers' interest: $FeNiPO_4$ and $LiNiPO_4$. Herein, we will only concern with the Li-Ni phosphate, and we carry out computer simulations for this compound. The references [1, 2] furnish us a current review respecting that material as well as information about its synthesis and the experimental techniques utilized to investigate it. Concretely, to study the $LiNiPO_4$ we will utilize a procedure based on spin waves what was developed by T. B. S. Jensen [3] as well as the nonextensive statistical mechanics invented by C. Tsallis [4].

The nonextensive statistics is also known as Tsallis statistics. It is named after its inventor, a Brazilian researcher of Greek origin. It is by far the most studied generalized statistics. There are four versions about this alternative to the Boltzmann-Gibbs-Shannon statistics; basically, they differ in the way of defining the thermal mean value [5]. The Tsallis statistics has support as theoretical as experimental in areas like Econophysics, Cosmology, Quantum Field theories, etc. Specially, we cite [6] as the reference containing applications of Tsallis statistics on magnetic materials and spin waves in the field of Condensed Matter Physics.

Apart from this introductory section, this article is organized into the following way: in the section we provide the theoretical frame; the subsection contains the background on both the composite $LiNiPO_4$ and the spin waves; the focusing on the Tsallis statistics is done in subsection ; in subsection we display the essential formulas for obtaining the spin-wave intensities originated in the Li-Ni phosphate. The section exhibits the computer simulations of spin waves utilizing the inno-

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vative statistic. Displaying the conclusions about this paper, we have the section . And, finally, in the section we acknowledge the persons that contributed to enhance the present work.

Theory

The phosphate composite $LiNiPO_4$

This crystal presents the magnetoelectricity, a phenomenon that is the interplay between the electric polarization and the magnetization. Its crystal structure is orthorhombic with lattice parameters a=10.032 Å, b=5.854 Å and c=4.677 Å; its crystal symmetry is described by the space group *Pnma* [7]. In the compound $LiNiPO_4$ the following ions aren't magnetic: Li^{1+}, P^{5+} and O^{2-}. But, the ions Ni^{2+} are the magnetic ions which possess a total spin S=1 [8]. So, each of these ions has a magnetic dipolar moment what interacts with its surroundings, and even more they can interplay with neutrons, for these elemental particles present a magnetic dipolar moment at that. Therefore, it is possible to take into account the neutron diffraction when this phosphate is targeted by a neutron beam from a source. For instance, T. B. S. Jensen *et al.* carried out experimental measurements in the Paul Scherrer Institute, Switzerland [9]. They utilized the triple-axis spectrometer RITA (**R**e-**I**nvented **T**riple **A**xis) – a device built at Risø National Laboratory DTU (Dutch acronym translated as Technical University of Denmark) – coupled to the SINQ (German acronym translated as Swiss Spallation Neutron Source). The flux of the SINQ is approximately 1014 n/(cm^2-s) [10].

The full Hamiltonian operator we consider for the compound $LiNiPO_4$ is the one proposed in [3, 9]:

$$\hat{H}_{\text{total}} = \hat{H}_{yz} + \hat{H}_y + \hat{H}_z + \hat{H}_{xz} + \hat{H}_{xy} + \hat{A}_x + \hat{A}_y + \hat{A}_z, \quad (1)$$

where the circumflex symbol indicates a quantum operator; above $\hat{H}_{xy}$ stands for the Heisenberg Hamiltonian of interactions between two ions Ni, which are next-nearest neighbors along the axes x and y (indeed it is along axes a and b); $\hat{H}_{xy}$, $\hat{H}_{yz}$, $\hat{H}_z$ and $\hat{H}_y$ are defined in an analogous manner. $\hat{A}_x$, $\hat{A}_y$ and $\hat{A}_z$ indicate single-ion anisotropies. So on, we specify the mentioned terms like

$$\hat{H}_{yz} = J_{yz} \sum_{i,j} \vec{S}_i . \vec{S}_j, \quad (2)$$

$$\hat{H}_{xy} = J_{xy} \sum_{i,j} \vec{S}_i . \vec{S}_j, \quad (3)$$

$$\hat{H}_{xz} = J_{xz} \left(\sum_{i,i'} \vec{S}_i . \vec{S}_{i'} + \sum_{j,j'} \vec{S}_j . \vec{S}_{j'} \right), \quad (4)$$

$$\hat{H}_y = J_y \left(\sum_{i,i'} \vec{S}_i . \vec{S}_{i'} + \sum_{j,j'} \vec{S}_j . \vec{S}_{j'} \right), \quad \text{and} \quad (5)$$

$$\hat{H}_z = J_z \left(\sum_{i,i'} \vec{S}_i . \vec{S}_{i'} + \sum_{j,j'} \vec{S}_j . \vec{S}_{j'} \right), \quad (6)$$

where it must be implied $\vec{S}_i$ is an operator standing for a spin in the site i; analogously for $\vec{S}_j$. Also, both i and i' represent sites of spin up; both j and j' means sites with spin down; J_{yz}, J_{xy}, J_{xz}, J_y and J_z are the usual exchange constants.

Furthermore, the three terms of single-ion anisotropy are given by

$$\hat{A}_x = D_x \left(\sum_i (S_i^x)^2 + \sum_j (S_j^x)^2 \right), \quad (7)$$

$$\hat{A}_y = D_y \left(\sum_i (S_i^y)^2 + \sum_j (S_j^y)^2 \right) \quad \text{and} \quad (8)$$

$$\hat{A}_z = D_z \left(\sum_i (S_i^z)^2 + \sum_j (S_j^z)^2 \right) = 0, \quad (9)$$

where D_x, D_y and D_z are the single-ion anisotropy constants in the directions of the crystal axes a, b and c, respectively. Finally, it was taken into account A_z=0 since it was assumed D_z=0.

On the other side, it is possible deploy the Holstein-Primakoff transformations to pass from a spin problem toward a boson problem [11], because the commutation rules for bosons are simpler than commutation rules for spins. We must also consider Ni^{2+} originates two magnetic types of ions: the ones with spin up and the ones with spin down. Thereupon, we can expand the spin operators in terms of two class of boson operator:

$$S_i^+ = \sqrt{2S} \sqrt{1 - \frac{a_i^\dagger a_i}{2S}} a_i, \quad (10)$$

$$S_i^- = \sqrt{2S} a_i^\dagger \sqrt{1 - \frac{a_i^\dagger a_i}{2S}} \quad (11)$$

$$S_i^z = S - a_i^\dagger a_i; \quad (12)$$

$$S_j^+ = \sqrt{2S} b_j \sqrt{1 - \frac{b_j^\dagger b_j}{2S}}, \quad (13)$$

$$S_j^- = \sqrt{2S} \sqrt{1 - \frac{b_j^\dagger b_j}{2S}} b_j^\dagger \quad (14)$$

$$S_j^z = -S + b_j^\dagger b_j \ . \quad (15)$$

It is absolutely essential four operators because two operators – both creation and destruction – are utilized for each spin. Namely, for the spin up, the operator $a_i^\dagger \equiv a_{\vec{r}_i}^\dagger$ denotes the creation of a boson in the position $\vec{r}_i$, and the operator $a_i \equiv a_{\vec{r}_i}$ symbolizes the annihilation of a boson in the same position; analogously for the ions with spin down. In Eqs. (10) we have evidenced it

is possible expand it in a power series and, taking into account only linear terms, we can get a linear spin-wave theory:

$$S_i^+ = \sqrt{2S}a_i, \tag{16}$$

$$S_i^- = \sqrt{2S}a_i^\dagger \tag{17}$$

$$S_i^z = S - a_i^\dagger a_i \tag{18}$$

$$S_j^+ = \sqrt{2S}b_j^\dagger, \tag{19}$$

$$S_j^- = \sqrt{2S}b_j \quad \text{and} \tag{20}$$

$$S_i^z = -S + b_i^\dagger b_i \ . \tag{21}$$

Next, we can express each of the respective ladder operators as a Fourier transform:

$$a_i^\dagger = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{-i\vec{k}.\vec{r}_i}a_{\vec{k}}^\dagger \tag{22}$$

$$a_i = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{i\vec{k}.\vec{r}}a_{\vec{k}}, \tag{23}$$

$$b_j^\dagger = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{-i\vec{k}.\vec{r}_j}b_{\vec{k}}^\dagger \tag{24}$$

$$b_j = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{i\vec{k}.\vec{r}_j}b_{\vec{k}}, \tag{25}$$

where $a_{\vec{k}}^\dagger$ and $a_{\vec{k}}$ represent the respective creation and annihilation of a magnon with wave vector $\vec{k}$; likewise for $b_{\vec{k}}^\dagger$ and $b_{\vec{k}}$. The magnon is a collective excitation and to evincing it we can explicitly set the inverse of Eqs.(22):

$$a_{\vec{k}}^\dagger = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{i\vec{k}.\vec{r}_i}a_i^\dagger \ , \tag{26}$$

$$a_{\vec{k}} = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{-i\vec{k}.\vec{r}}a_i \ , \tag{27}$$

$$b_{\vec{k}}^\dagger = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{i\vec{k}.\vec{r}_j}b_j^\dagger \ , \tag{28}$$

$$b_{\vec{k}} = \sqrt{\frac{2}{N}}\sum_{\vec{k}} e^{-i\vec{k}.\vec{r}_j}b_j \ . \tag{29}$$

There upon, replacing the equation sets (16) and (22) into Eq. (1), we obtain the total Hamiltonian operator for $LiNiPO_4$ in the frame of a linear spin-wave theory. That Hamiltonian expression can be compacted in the following matrix way:

$$\hat{H}_{\text{total}} = \frac{1}{2}\sum_{\vec{k}} \mathbf{a}^\dagger \mathcal{H}\mathbf{a} \tag{30}$$

with

$$\mathbf{a}^\dagger = \begin{pmatrix} a_{\vec{k}}^\dagger & a_{-\vec{k}} & b_{\vec{k}}^\dagger & b_{-\vec{k}} \end{pmatrix} \quad \mathbf{a} = \begin{pmatrix} a_{\vec{k}} \\ a_{-\vec{k}}^\dagger \\ b_{\vec{k}} \\ b_{-\vec{k}}^\dagger \end{pmatrix}, \tag{31}$$

where $\mathcal{H}$ indicates a matrix constituted by no operators; it has the following expression:

$$\mathcal{H} = \begin{pmatrix} A & B & 0 & D \\ B & A & D & 0 \\ 0 & D & A & B \\ D & 0 & B & A \end{pmatrix}, \tag{32}$$

the matrix entries will be specified subsequently. We want to stress just now is $\mathcal{H}$ doesn't contain the eigenvalues of $LiNiPO_4$ because this case is very much different to the elemental expression for a given arbitrary Hamiltonian operator:

$$\hat{\mathbb{H}} = \sum_m \mathbf{P}_m^\dagger \mathbb{H}_{mm}\mathbf{P}_m \tag{33}$$

where $\mathbb{H}$ has to be a matrix represented in the eigenvector basis of $\hat{\mathbb{H}}$ with elements $\mathbb{H}_{mm} = \langle E_m|\hat{\mathbb{H}}|E_m\rangle$, and

$$\mathbf{P}_m^\dagger = \begin{pmatrix} |E_1\rangle\langle E_1| & |E_2\rangle\langle E_2| & \ldots & |E_m\rangle\langle E_m| \end{pmatrix} \tag{34}$$

$$\mathbf{P}_m = \begin{pmatrix} |E_1\rangle\langle E_1| \\ |E_2\rangle\langle E_2| \\ \ldots \\ |E_m\rangle\langle E_m| \end{pmatrix}; \tag{35}$$

certainly, the matrix $\mathbb{H}$ contains the arbitrary eigenvalues. Therefore, to find out the eigenvalues of $LiNiPO_4$ it will be necessary to effectuate a transformation that guarantees the boson commutation rules. Before delineating that procedure, we must specify the matrix components of $\mathcal{H}$:

$$A = 4S(J_{yz} + J_{xy}) - 2S\Big\{J_y\Big[1 - \cos(\vec{k}.\vec{r_5})\Big] + J_z\Big[1 - \cos(\vec{k}.\vec{r}_6)\Big] + \ J_{xz}\Big[2 - \cos(\vec{k}.\vec{r}_7) - \cos(\vec{k}.\vec{r}_8)\Big]\Big\} + 2D_x\Big(S - \frac{1}{2}\Big) + 2D_y\Big(S - \frac{1}{2}\Big) \ , \tag{36}$$

$$B = 2D_x\Big(S - \frac{1}{2}\Big) - 2D_y\Big(S - \frac{1}{2}\Big) \ , \tag{37}$$

$$D = 2J_{yz}S\Big[\cos(\vec{k}.\vec{r}_1) + \cos(\vec{k}.\vec{r}_2)\Big] + 2J_{xy}S\Big[\cos(\vec{k}.\vec{r}_3) + \cos(\vec{k}.\vec{r}_4)\Big] \ ; \tag{38}$$

with the vectors $\vec{r}_i$ meaning

$$\vec{r}_1 = (0, \frac{b}{2}, \frac{c}{2}), \quad \vec{r}_2 = (0, \frac{b}{2}, \frac{-c}{2}),$$
$$\vec{r}_3 = (\frac{a}{2}, \frac{b}{2}, 0), \quad \vec{r}_4 = (\frac{a}{2}, \frac{-b}{2}, 0),$$
$$\vec{r}_5 = (0, b, 0), \quad \vec{r}_6 = (0, 0, c),$$
$$\vec{r}_7 = (\frac{a}{2}, 0, \frac{c}{2}) \quad \text{and} \quad \vec{r}_8 = (\frac{a}{2}, 0, \frac{-c}{2}) \, . \quad (39)$$

Immediately, we are going to apply the transformation proposed in [3]: $\mathbf{a} = \mathbf{T}\boldsymbol{\alpha}$. As a result of it the full Hamiltonian, Eq. (30), become

$$\hat{H}_{\text{total}} = \frac{1}{2} \sum_{\vec{k}} \boldsymbol{\alpha}^\dagger \mathbf{T}^\dagger \mathcal{H} \mathbf{T} \boldsymbol{\alpha} \equiv \frac{1}{2} \sum_{\vec{k}} \boldsymbol{\alpha}^\dagger \mathbf{H} \boldsymbol{\alpha} \quad (40)$$

with $\mathbf{H} = \mathbf{T}^\dagger \mathcal{H} \mathbf{T}$ standing for the Hamiltonian matrix in the eigenvector basis; $\mathbf{T}$ is a real transformation matrix, and hence $\mathbf{T}^\dagger = \mathbf{T}^t$; $\boldsymbol{\alpha}^\dagger$ and $\boldsymbol{\alpha}$ are a set of Bosons operators, as well. In detail, these last mathematical entities are denoted as

$$\mathbf{T} = \begin{pmatrix} T_{11} & T_{12} & T_{13} & T_{14} \\ T_{21} & T_{22} & T_{23} & T_{24} \\ T_{31} & T_{32} & T_{33} & T_{34} \\ T_{41} & T_{42} & T_{43} & T_{44} \end{pmatrix}; \quad (41)$$

$$\boldsymbol{\alpha}^\dagger = \begin{pmatrix} \alpha^\dagger_{\vec{k}} & \alpha_{-\vec{k}} & \beta^\dagger_{\vec{k}} & \beta_{-\vec{k}} \end{pmatrix}, \quad \boldsymbol{\alpha} = \begin{pmatrix} \alpha_{\vec{k}} \\ \alpha^\dagger_{-\vec{k}} \\ \beta_{\vec{k}} \\ \beta^\dagger_{-\vec{k}} \end{pmatrix}. \quad (42)$$

Now, there are 16 boson commutator relations originated from commutations between elements of $\mathbf{a}$ and $\mathbf{a}^\dagger$ in the Eq. (31). All of them can be compacted as follows:

$$\begin{pmatrix} T_{11} & -T_{12} & T_{13} & -T_{14} \\ -T_{21} & T_{22} & -T_{23} & T_{24} \\ T_{31} & -T_{32} & T_{33} & -T_{34} \\ -T_{41} & T_{42} & -T_{43} & T_{44} \end{pmatrix} \times \begin{pmatrix} T_{11} & T_{21} & T_{31} & T_{41} \\ T_{12} & T_{22} & T_{32} & T_{42} \\ T_{13} & T_{23} & T_{33} & T_{43} \\ T_{14} & T_{24} & T_{34} & T_{44} \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \quad (43)$$

from this expression, we perceive two facts: the first one is there appears the transpose of $\mathbf{T}$:

$$\mathbf{T}^t = \begin{pmatrix} T_{11} & T_{21} & T_{31} & T_{41} \\ T_{12} & T_{22} & T_{32} & T_{42} \\ T_{13} & T_{23} & T_{33} & T_{43} \\ T_{14} & T_{24} & T_{34} & T_{44} \end{pmatrix}; \quad (44)$$

the second one is the inverse matrix of $\mathbf{T}^t$ also is defined from Eq. (43), for $(\mathbf{T}^t)^{-1}\mathbf{T}^t = \mathbf{I}_{4x4}$; explicitly we have

$$(\mathbf{T}^t)^{-1} = \begin{pmatrix} T_{11} & -T_{12} & T_{13} & -T_{14} \\ -T_{21} & T_{22} & -T_{23} & T_{24} \\ T_{31} & -T_{32} & T_{33} & -T_{34} \\ -T_{41} & T_{42} & -T_{43} & T_{44} \end{pmatrix} . \quad (45)$$

Next, we want to point out an important conclusion from Eqs.(44) and (45): $\mathbf{T}$ isn't a unitary transformation since $\mathbf{T}^t \neq (\mathbf{T}^t)^{-1}$. This fact will have repercussion on the Hamiltonian matrix of $LiNiPO_4$ as we will display later.

Now from the diagonal matrix, with lambda eigenvalues, $\mathbf{H} = \mathbf{T^t}\mathcal{H}\mathbf{T}$ it can be inferred that

$$\mathcal{H}\mathbf{T} = (\mathbf{T}^t)^{-1}\mathbf{H} = \begin{pmatrix} T_{11} & -T_{12} & T_{13} & -T_{14} \\ -T_{21} & T_{22} & -T_{23} & T_{24} \\ T_{31} & -T_{32} & T_{33} & -T_{34} \\ -T_{41} & T_{42} & -T_{43} & T_{44} \end{pmatrix} \times \begin{pmatrix} \lambda_1 & 0 & 0 & 0 \\ 0 & \lambda_2 & 0 & 0 \\ 0 & 0 & \lambda_3 & 0 \\ 0 & 0 & 0 & \lambda_4 \end{pmatrix}; \quad (46)$$

after we have

$$\mathcal{H}\mathbf{T} = \left(\lambda_1 \begin{pmatrix} T_{11} \\ -T_{21} \\ T_{31} \\ -T_{41} \end{pmatrix} \lambda_2 \begin{pmatrix} -T_{12} \\ T_{22} \\ -T_{32} \\ T_{42} \end{pmatrix} \lambda_3 \begin{pmatrix} T_{13} \\ -T_{23} \\ T_{33} \\ -T_{43} \end{pmatrix} \times \lambda_4 \begin{pmatrix} -T_{14} \\ T_{24} \\ -T_{34} \\ T_{44} \end{pmatrix} \right) \quad (47)$$

but from matrix properties we have also

$$\mathcal{H}\mathbf{T} = \begin{pmatrix} \mathcal{H}\mathbf{T}_1 & \mathcal{H}\mathbf{T}_2 & \mathcal{H}\mathbf{T}_3 & \mathcal{H}\mathbf{T}_4 \end{pmatrix} = \left(\mathcal{H} \begin{pmatrix} T_{11} \\ T_{21} \\ T_{31} \\ T_{41} \end{pmatrix} \quad \mathcal{H} \begin{pmatrix} T_{12} \\ T_{22} \\ T_{32} \\ T_{42} \end{pmatrix} \quad \mathcal{H} \begin{pmatrix} T_{13} \\ T_{23} \\ T_{33} \\ T_{43} \end{pmatrix} \quad \mathcal{H} \begin{pmatrix} T_{14} \\ T_{24} \\ T_{34} \\ T_{44} \end{pmatrix} \right), \quad (48)$$

where it is clear the meaning of $\mathbf{T}_1$, $\mathbf{T}_2$, $\mathbf{T}_3$ and $\mathbf{T}_4$. By contrasting the Eqs. (47) and (48) we obtained four matrix equations:

$$\mathcal{H}\mathbf{T}_1 = \lambda_1 \begin{pmatrix} T_{11} \\ -T_{21} \\ T_{31} \\ -T_{41} \end{pmatrix}, \mathcal{H}\mathbf{T}_2 = \lambda_2 \begin{pmatrix} -T_{12} \\ T_{22} \\ -T_{32} \\ T_{42} \end{pmatrix}$$
$$\mathcal{H}\mathbf{T}_3 = \lambda_3 \begin{pmatrix} T_{13} \\ -T_{23} \\ T_{33} \\ -T_{43} \end{pmatrix} \text{ and } \mathcal{H}\mathbf{T}_4 = \lambda_4 \begin{pmatrix} -T_{14} \\ T_{24} \\ -T_{34} \\ T_{44} \end{pmatrix}; \quad (49)$$

from this last four expressions, we found, respectively, the column vectors $\mathbf{T}_1$, $\mathbf{T}_2$, $\mathbf{T}_3$ and $\mathbf{T}_4$ on the right side of the respective matrix identity:

$$\mathcal{H}\mathbf{T}_1 = \lambda_1\mathbf{I}_1\mathbf{T}_1, \quad \mathcal{H}\mathbf{T}_2 = \lambda_2\mathbf{I}_2\mathbf{T}_2, \\ \mathcal{H}\mathbf{T}_3 = \lambda_3\mathbf{I}_1\mathbf{T}_3, \quad \text{and} \quad \mathcal{H}\mathbf{T}_4 = \lambda_4\mathbf{I}_2\mathbf{T}_4\ , \quad (50)$$

with

$$\mathbf{I}_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \quad \text{and} \quad (51)$$

$$\mathbf{I}_2 = \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (52)$$

We notice Eq. (50) may be converted in the following one:

$$\mathbf{I}_1\mathcal{H}\mathbf{T}_1 = \lambda_1\mathbf{T}_1, \quad \mathbf{I}_2\mathcal{H}\mathbf{T}_2 = \lambda_2\mathbf{T}_2, \\ \mathbf{I}_1\mathcal{H}\mathbf{T}_3 = \lambda_3\mathbf{T}_3, \quad \text{and} \quad \mathbf{I}_2\mathcal{H}\mathbf{T}_4 = \lambda_4\mathbf{T}_4; \quad (53)$$

and, by taking into account that $\mathbf{I}_1 = -\mathbf{I}_2$,

$$\mathbf{I}_1\mathcal{H}\mathbf{T}_1 = \lambda_1\mathbf{T}_1, \quad \mathbf{I}_1\mathcal{H}\mathbf{T}_2 = -\lambda_2\mathbf{T}_2, \\ \mathbf{I}_1\mathcal{H}\mathbf{T}_3 = \lambda_3\mathbf{T}_3, \quad \text{and} \quad \mathbf{I}_1\mathcal{H}\mathbf{T}_4 = -\lambda_4\mathbf{T}_4; \quad (54)$$

renaming $\mathfrak{H} = \mathbf{I}_1\mathcal{H}$, we can rewrite the earlier equation as follows:

$$\mathfrak{H}\mathbf{T}_1 = \lambda_1\mathbf{T}_1, \quad \mathfrak{H}\mathbf{T}_2 = -\lambda_2\mathbf{T}_2, \\ \mathfrak{H}\mathbf{T}_3 = \lambda_3\mathbf{T}_3, \quad \text{and} \quad \mathfrak{H}\mathbf{T}_4 = -\lambda_4\mathbf{T}_4\ . \quad (55)$$

Therefore, it has been formed four eigenvalue equations with their respective eigenvalues λ_1, $-\lambda_2$, λ_3 and $-\lambda_4$. Furthermore, it has been obtained the *true* hamiltonian matrix $\mathfrak{H}$ that represents the full Hamiltonian of the Eq. (1). But it isn't represented in the eigenvector basis, for it is non-diagonal; that is, we have to diagonalize else

$$\mathfrak{H} = \mathbf{I}_1\mathcal{H} = \begin{pmatrix} A & B & 0 & D \\ -B & -A & -D & 0 \\ 0 & D & A & B \\ -D & 0 & -B & -A \end{pmatrix}\ . \quad (56)$$

We want to emphasize the matrix $\mathfrak{H}$ – got by T. S. Jensen [3] – is non-Hermitian; it is a consequence of the fact the transformation matrix, Eq. (41), isn't unitary, but its usage have an experimental support [3]. Besides the non-hermitian quantum mechanics is an emerging field of research currently [12]. Now, mathematically, we have four real eigenvalues for Eq. (56):

$$\epsilon_1 = \lambda_1 = \sqrt{A^2 - (B+D)^2}, \quad (57)$$

$$\epsilon_2 = -\lambda_2 = -\sqrt{A^2 - (B+D)^2}, \quad (58)$$

$$\epsilon_3 = \lambda_3 = \sqrt{A^2 - (B-D)^2} \quad \text{and} \quad (59)$$

$$\epsilon_4 = -\lambda_4 = -\sqrt{A^2 - (B-D)^2}. \quad (60)$$

However, we will only consider the positive values expressing the dispersion of the spin waves; we will denote them as

$$\epsilon_{\alpha,\beta} = \sqrt{A^2 - (B \pm D)^2}\ . \quad (61)$$

Next, following the developed work by T. S. Jensen [3], it can be formed, with the associated eigenvectors of $\mathfrak{H}$, a provisional transformation matrix $\mathbf{P}$. Logically, if it is to be the true matrix, it will be renamed $\mathbf{T}$. Then, let P_1, P_2, P_3 and P_4 denoting the associated eigenvectors of $\mathfrak{H}$ so that $\mathbf{P}$ is defined as

$$\mathbf{P} = \begin{pmatrix} P_1 & P_2 & P_3 & P_4 \end{pmatrix} = \\ \begin{pmatrix} -\dfrac{\epsilon_\alpha + A}{B+D} & -\dfrac{-\epsilon_\alpha + A}{B+D} & \dfrac{\epsilon_\beta + A}{B-D} & \dfrac{-\epsilon_\beta + A}{B-D} \\ 1 & 1 & -1 & -1 \\ -\dfrac{\epsilon_\alpha + A}{B+D} & -\dfrac{-\epsilon_\alpha + A}{B+D} & -\dfrac{\epsilon_\beta + A}{B-D} & -\dfrac{-\epsilon_\beta + A}{B-D} \\ 1 & 1 & 1 & 1 \end{pmatrix}. \quad (62)$$

Now, the true transformation matrix must confirm the relation $\mathbf{I}_1 = \mathbf{T}\mathbf{I}_1\mathbf{T}^t$. So, we proceed to verify if $\mathbf{P}$ satisfies it:

$$\mathbf{P}\mathbf{I}_1\mathbf{P}^t = \begin{pmatrix} -x_1 & -x_2 & x_3 & x_4 \\ 1 & 1 & -1 & -1 \\ -x_1 & -x_2 & -x_3 & -x_4 \\ 1 & 1 & 1 & 1 \end{pmatrix} \times \\ \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \begin{pmatrix} -x_1 & 1 & -x_1 & 1 \\ -x_2 & 1 & -x_2 & 1 \\ x_3 & -1 & -x_3 & 1 \\ x_4 & -1 & -x_4 & 1 \end{pmatrix} \neq \mathbf{I}_1, \quad (63)$$

where we have set the following parameters:

$$x_1 = \frac{\epsilon_\alpha + A}{B+D}, \quad x_2 = \frac{-\epsilon_\alpha + A}{B+D} \quad (64)$$

$$x_3 = \frac{\epsilon_\beta + A}{B-D} \quad \text{and} \quad x_4 = \frac{-\epsilon_\beta + A}{B-D}\ . \quad (65)$$

Certainly, the provisional matrix $\mathbf{P}$ doesn't satisfy the mentioned relation for $\mathbf{I}_1$ and, therefore, it doesn't

contain the sought eigenvectors. In [3] is shown that the true transformation matrix is

$$\mathbf{T} = [\mathbf{T_1}\ \ \mathbf{T_2}\ \ \mathbf{T_3}\ \ \mathbf{T_4}] = \mathbf{PM} \tag{66}$$

where $\mathbf{M} = \sqrt{\mathbf{I_1 P^{-1} I_1 (P^t)^{-1}}}$.

Thus to find the true eigenvectors contained in $\mathbf{T}$, it has to be utilized a computational approach because the inverse matrices appearing in the last formula for $\mathbf{M}$.

The Nonextensive Statistical Mechanics

As indicated in the section 1, this statistical proposal has four versions currently. However, it is the third one what has been more studied; we utilized it in this article. Then, the theoretical construction starts by postulating the Tsallis entropy [4, 5]:

$$S_q = k_B \frac{1 - \sum\limits_i (p_i^q)}{q-1}, \tag{67}$$

where p_i^q is the probability distribution to find the system in i-th state, p_i, powered to the entropic index q; k_B is the Boltzmann constant; $\sum\limits_i (p_i^q)$ symbolizes the quantum operation of trace over the matrix p_i^q. As we apply the limit q tending to 1 for Eq.(67), we recover the well known Boltzmann-Gibbs-Shannon entropy,

$$S = -k_B \sum_i (p_i \mathrm{Ln} p_i). \tag{68}$$

The nonextensive probability distribution p_i is obtained via maximum entropy method, which was invented by the American Edward T. Jaynes [15]. In that procedure, we consider the following constraints,

$$\sum_i p_i = 1 \qquad \text{and} \qquad E_q = \frac{\sum\limits_i p_i^q \epsilon_i}{\sum\limits_i p_i^q}, \tag{69}$$

where ϵ_i are the energy eigenvalues and E_q is the internal energy. By the means of the aforementioned method, we obtain the probability distribution given by

$$p_i = \frac{\left[1 - (1-q)\beta' \epsilon_i\right]^{\frac{1}{1-q}}}{Z_q}, \tag{70}$$

where Z_q stands for the partition function

$$Z_q = \sum_i \left[1 - (1-q)\beta' \epsilon_i\right]^{\frac{1}{1-q}}; \tag{71}$$

where β' is a parameter defined as the inverse temperature

$$\beta' = \frac{1}{k_B T}\ . \tag{72}$$

However, in the Tsallis statistics it exists other definitions for temperature [16].

On the other side, we want to stress in the Eqs. (70) and (71) it is necessary to guarantee the positivity of probabilities. It is get via the Tsallis cut-off:

$$\left[1 - (1-q)\frac{1}{k_B T}\epsilon_i\right] > 0\ . \tag{73}$$

Therefore, the distribution of probability can be expressed as

$$p_i = \begin{cases} \frac{[1-(1-q)\frac{1}{k_B T}\epsilon_i]^{\frac{1}{1-q}}}{Z_q} & , \quad \text{if } [1-(1-q)\frac{1}{k_B T}\epsilon_i] > 0 \\ 0 & , \quad \text{otherwise} \end{cases} \tag{74}$$

In addition, we can rewrite the relationships for S_q and p_i through the functions known as q-exponential and q-logarithm, respectively:

$$\exp_q(x) = [1 + (1-q)x]^{\frac{1}{1-q}} \quad \text{and} \tag{75}$$

$$\mathrm{Ln}_q(x) = \frac{x^{1-q} - 1}{1-q}; \tag{76}$$

thereupon, S_q and p_i, Eq. (67) and Eq. (70), verify

$$S_q = -k_B \sum_i \left[p_i \mathrm{Ln}_q(p_i)\right] \quad \text{and} \tag{77}$$

$$p_i = \frac{\exp_q(-\frac{1}{k_B T}\epsilon_i)}{\sum\limits_i \exp_q(-\frac{1}{k_B T}\epsilon_i)} \tag{78}$$

which, clearly, remember both the entropy and probability distribution for the Boltzmann-Gibbs-Shannon statistics.

Quantum mean values

In the third version of the nonextensive statistical mechanics, the thermal mean values of an observable O, represented by the operator $\hat{O}$, in the Hilbert space are calculated by means of this expression:

$$O_q = \langle \hat{O} \rangle_q = \frac{\sum\limits_i p_i{}^q O_i}{\sum\limits_i p_i^q}, \tag{79}$$

where O_i betoken the eigenvalues of the observable $\hat{O}$. The limit $q \to 1$ of the formula above becomes the standard expression:

$$O = \langle \hat{O} \rangle = \sum_i p_i O_i. \tag{80}$$

An example of thermal mean value is the internal energy what is defined by

$$E_q = \frac{\sum_i \left[1-(1-q)\frac{1}{k_BT}\epsilon_i\right]^{\frac{q}{1-q}}\epsilon_i}{\sum_i \left[1-(1-q)\frac{1}{k_BT}\epsilon_i\right]^{\frac{q}{1-q}}} \quad ; \qquad (81)$$

clearly this is an iterative formula, so the Newton-Raphson method can be utilized to find E_q. But, herein, we are more concerned with the energy eigenvalues than the internal energy.

Spin-wave intensities

In this subsection it will be utilized some expressions from the two early subsections. To quantify the spin-wave intensities it must be done a beam of neutrons comes into contact with the $LiNiPO_4$; see e.g. [9]. That stream of particles is scattered inelastically by the material and its energy loss excites a magnon in the phosphate. The spin-wave intensity is expressed by the differential scattering cross-section that is defined as follows

$$\frac{d^2\sigma_m}{d\Omega dE'}(\vec{K},\omega) = \frac{E_f}{E_i}(\gamma r_0)^2\left|\frac{g}{2}F_{(\vec{K})}\right|^2 \frac{e^{-2W_{(\vec{K})}}}{\hbar}\times \sum_{\mu\nu}\left(\delta_{\mu\nu}-\hat{\mathbf{k}}_\mu\hat{\mathbf{k}}_\nu\right)S^{\mu\nu}_{(\vec{K},\omega)}, \qquad (82)$$

where $\vec{K}$ is the momentum transfer, ω is the dispersion frequency, Ω is a solid angle, E_f and E_i are the final and incoming neutron energies, respectively; $F_{(\vec{K})}$ is the dynamical structure factor; $\hat{\mathbf{k}}_\mu$ and $\hat{\mathbf{k}}_\nu$ are unit vectors in the axes μ and ν, respectively; being $\mu,\nu \equiv x,y,z$. γ is the neutron magnetic dipolar moment; r_0 is the classical electron radius; g is the Landé g-factor; $W_{(\vec{K})}$ is the Debye-Waller factor represented in the reciprocal space. In addition, $S^{\mu\nu}_{(\vec{K},\omega)}$ is the scattering matrix (also known as S-matrix); it can be too expressed as $S^{\mu\nu}_{(\vec{K},t)}$, that is,

$$S^{\mu\nu}_{(\vec{Q},w)} = \int dt e^{iwt} S^{\mu\nu}_{(\vec{Q},t)} \quad \text{and} \qquad (83)$$

$$S^{\mu\nu}_{(\vec{Q},t)} = \sum_{\vec{r}\vec{r}'} e^{-\vec{Q}.(\vec{r}-\vec{r}')}\langle S^\mu \vec{r} S^\nu_{\vec{r}}(t)\rangle. \qquad (84)$$

with $\vec{Q} = (Q_x, Q_y)$ representing a reciprocal lattice vector and t meaning a time variable. The cross section indicates the probability that a magnetic interaction happens. This interaction can give origin whether the creation or annihilation of a magnon. For instance, we have the creation of a magnon α is revealed by the following expression

$$\frac{d^2\sigma^{\alpha+}}{d\Omega dE'}(\vec{K},\omega) = \Gamma_{(\vec{K},\omega)}\left[n_{(\omega^\alpha_{\vec{K}})}+1\right]\delta_{(\omega-\omega^\alpha_{\vec{K}})}\times \sum_{\vec{\tau},\vec{K}}\Big[(1-Q_x^2)|M+N|^2+(1-Q_y^2)|M-N|^2 - 2Q_xQ_y\mathrm{Im}(MN^*)\Big] \qquad (85)$$

where $n_{(\omega^\alpha_{\vec{K}})}$ is the Bose-Einstein population of magnons in accordance to Tsallis statistical distribution; it indicates the average number of magnons in the quantum state with an energy $\hbar\omega^\alpha_{\vec{K}}$. Both M and N are auxiliary parameters. All of these last parameters are defined as follows

$$n_{(\omega^\alpha_{\vec{K}})} = \frac{\sum_{n=0}^{\infty} n[1-(1-q)\frac{n}{k_BT}(\hbar\omega^\alpha_{\vec{K}})]^{\frac{q}{1-q}}}{\sum_{n=0}^{\infty}[1-(1-q)\frac{n}{k_BT}(\hbar\omega^\alpha_{\vec{K}})]^{\frac{q}{1-q}}} \qquad (86)$$

and

$$M = T_{11}F_\uparrow(\vec{\tau}) + T_{41}F_\downarrow(\vec{\tau}) \quad \text{and} \qquad (87)$$

$$N = T_{21}F_\uparrow(\vec{\tau}) + T_{31}F_\downarrow(\vec{\tau}) \qquad (88)$$

where $\vec{\tau}$ also is a reciprocal lattice vector defined like $\vec{\tau} = \vec{K}-\vec{k}$; $F_\sigma(\vec{\tau})$ is the spin-dependent structure factor. Likewise, for the destruction of a magnon α we have the following formula

$$\frac{d^2\sigma^{\alpha-}}{d\Omega dE'}(\vec{K},\omega) = \Gamma_{(\vec{K},\omega)}n_{(\omega^\alpha_{\vec{K}})}\delta_{(\omega+\omega^\alpha_{\vec{K}})}\times \sum_{\tau,\vec{K}}\Big[(1-Q_x^2)|M+N|^2+(1-Q_y^2)|M-N|^2 - 2Q_xQ_y\mathrm{Im}(MN^*)\Big] \qquad (89)$$

this time M, N and $\vec{\tau}$ are redefined in the following way

$$M = T_{11}F^*_\uparrow(\vec{\tau}) + T^*_{41}F_\downarrow(\vec{\tau}) \ , \qquad (90)$$

$$N = T_{21}F^*_\uparrow(\vec{\tau}) + T_{31}F^*_\downarrow(\vec{\tau}) \quad \text{and} \qquad (91)$$

$$\vec{\tau} = \vec{K}+\vec{k} \qquad (92)$$

The expressions for the creation and the annihilation of a magnon β are similar.

Numerical results

In this section we introduce some numerical calculations framed in the linear spin-wave theory for the compound $LiNiPO_4$. We apply the nonextensive statistical mechanics in the simulations. So we calculate the energy eigenvalues as well as the intensities for scattering vectors. These late parameter are analyzed for several values of the entropic index q.

Figure	$\vec{k}$	T	J_{yz}	J_y	J_z	J_{xz}	J_{xy}	D_x	D_y	D_z
1a)	(0 1 k_z)	14	1.036	0.6701	-0.0469	-0.1121	0.2977	0.1696	0.9097	0
1b)	(k_x 1 0)	6	1.036	0.6701	-0.0469	-0.1121	0.2977	0.1696	0.9097	0
1c)	(0 k_y 0)	6	1.040	0.6700	-0.0500	-0.1100	0.3000	0.1700	0.9100	0

Table 1: Data for figure 1.

$\vec{k}$	T	J_{yz}	J_y	J_z	J_{xz}	J_{xy}	D_x	D_y	D_z
(0 k_y 0)	6	1.040	0.6700	-0.0500	-0.1100	0.3000	0.1700	0.9100	0

Table 2: Data for figure 2.

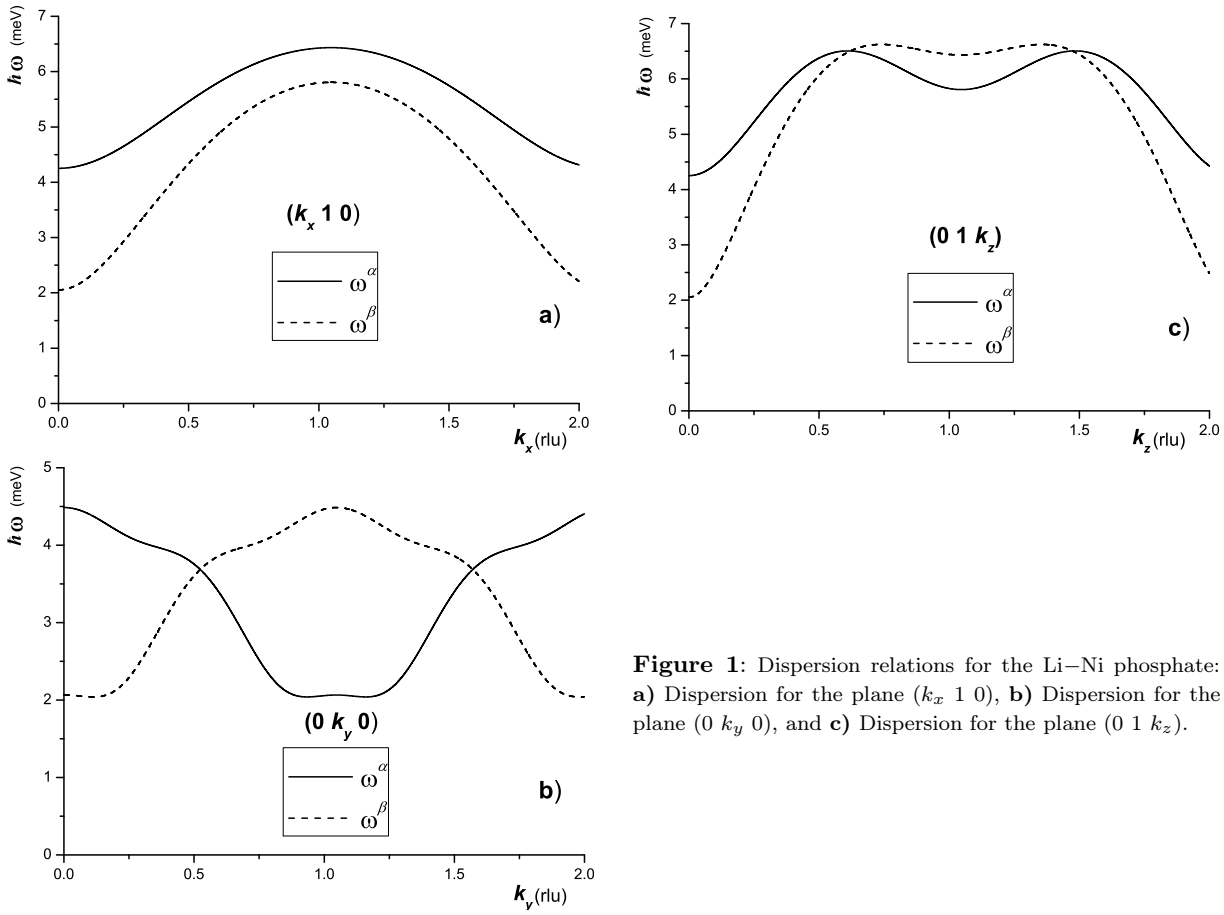

Figure 1: Dispersion relations for the Li−Ni phosphate: **a)** Dispersion for the plane (k_x 1 0), **b)** Dispersion for the plane (0 k_y 0), and **c)** Dispersion for the plane (0 1 k_z).

In the Fig. 1, we have the spin-wave dispersion relations, i.e., the eigenenergies that are excited when the neutrons interact with the Li-Ni phosphate. The solid lines indicate the magnon α and the dashed lines, the magnon β. Concretely, in the Figs. 1a), 1b) and 1c) we have the dispersion relations on the reciprocal planes (0 k_y 0), (k_x 1 0) and (0 1 k_z), respectively. Inside the Table 1 we have all the relevant parameters for the Fig. 1. Respecting to the used units in the table, the temperature T is in kelvins, the wave vector is expressed in reciprocal lattice units, rlu; the exchange constants as well as the single-ion anisotropy constants are in meV. It is obvious the spin-wave dispersion relation doesn't depend on the entropic index q, so this last parameter isn't in the table 1.

The Table 2 show the parameters used in the Fig. 2a) which displays the intensity of the magnons α created during the interaction between the neutrons and the

magnetic structure. This spin-wave intensity has been calculated numerically in the plane (0 k_y 0); we have deployed the following values of the entropic index q: 0.7, 1.3, 1.7 and 2.0. We notice the more q increases, the more the hight peak increases. At $q = 1$ it is clear the intensity of the magnons α is a smooth curve without changes very abrupt. However, when we take into account values of $q > 1$ the landscape is different: it appears some lateral bulges in the graphics. At q=1.3 and 1.7 the bulges are slight but at q=2.0 they are pretty much notorious. For the Fig. 2b) we also utilize the same data from Table 2. It represents the intensity of the magnon β created during the interaction between the plane (0 k_y 0) and the incident neutrons. It is apparent no magnons β were created. This fact is evidenced by the bold lines having all of them a value zero.

These computer simulations can be contrasted with experimental measurements of Li-Ni phosphate done by T. S. Jensen [3]. So, the nonextensive distribution makes a good fitting for experimental data.

Conclusions

In this paper we have applied the nonextensive statistical mechanics for investigating the compound $LiNiPO_4$ by means of a linear spin-wave theory. By considering two sublattices for the unique magnetic ion Ni^{2+}, we have studied the modifications appearing in the computer simulations of that phosphate. For q=1 the results are similar to those obtained early by other authors. However, simulations with $q \neq 1$ turned out to give rise to some lateral bulges in the graphics of spin-wave intensities.

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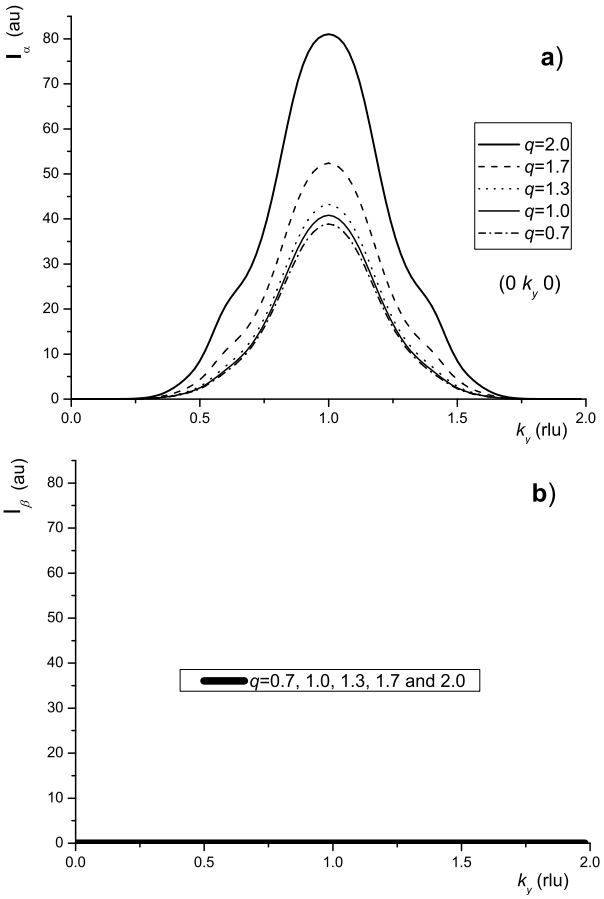

Figure 2: **a)** Intensities, calculated numerically, for the magnons α created through the interaction of the reciprocal plane (0 k_y 0) and the neutrons falling upon it. **b)** Intensities for the magnons β. No magnons β are observed.

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