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# ON THE LIBRON SPECTRUM IN SOLID ORTHOHYDROGEN AND PARADEUTERIUM

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# ON THE LIBRON SPECTRUM IN SOLID ORTHOHYDROGEN AND PARADEUTERIUM

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К теории либронного спектра в ортоводороде и парадейтерии

Вычисляется либронный спектр в твердом ортоводороде и парадейтерии при $\vec{k}=0$ и $T=0$ с использованием диаграммного метода для спиновых и псевдоспиновых решеточных систем, развитого одним из авторов (К.Валясек).

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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On the Libron Spectrum in Solid Orthohydrogen and Paradeuterium

The $\vec{k}=0$ libron spectrum at $T=0$ in solid orthohydrogen and paradeuterium is calculated by using the diagrammatic method for spin and pseudospin lattice systems developed by one of the authors (K.Walasek).

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

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The low lying orientational excitations in the ordered phase of solid orthohydrogen and paradeuterium, the so-called librons, have been object of a number of studies, both experimental [1-3] and theoretical [1-8]. At the present stage it is well known that the libron-libron interactions considerably affect the single libron spectrum and calculations based on simple spin-wave theories [4-6] fail to explain experimental data of a Raman scattering. Because of the complexity of the electrostatic quadrupole-quadrupole (EQQ) Hamiltonian for systems under consideration the problem of calculation of the renormalized libron spectrum is rather difficult. The first attempt to take into account the libron-libron interactions has been made in ref. [7], where the authors transform the Hamiltonian to the Bose representation and apply the diagrammatic perturbation theory. In their method the spurious kinematic terms appear which slightly decrease the value of the average libron gap.

Quite recently the single libron spectrum at $\vec{k}=0$ has been calculated by using RPA for the retarded Green functions after a suitable unitary transformation of the EQQ Hamiltonian [8]. However, this approach does not predict the existence of the two-libron excitations. Nevertheless, the results of these works are generally in good agreement with the experimental data [1-3]. The purpose of this letter is to present the method and results of our calculations of the zero-temperature libron spectrum applying the diagrammatic technique developed for an arbitrary spin and pseudospin lattice system by one of us [9]. This method was already successfully applied to the calculation of the free energy and investi-

gation of the order-disorder phase transition in solid $o\text{-}H_2$ and $p\text{-}D_2$ [10]. The small parameter of our theory is $1/z$, where $z = 12$ is the nearest-neighbour number for an f.c.c. lattice. The zeroth-order (in $1/z$) libron spectrum corresponds to that obtained from R.P.A. To calculate the Green function, we used the Wick-like reduction theorem for the generalized Pauli operators which allows us to avoid the spurious kinematic terms. The Green function matrix $\hat{K}(\vec{k},\omega_\nu)$ can be written as follows

$$\hat{K}(\vec{k},\omega_\nu) = [\hat{M}^{-1}(\vec{k},\omega_\nu) + 2\hat{\gamma}(\vec{k})]^{-1} \qquad (1)$$

where $\hat{K}(\vec{k},\omega_\nu)$, $\hat{M}(\vec{k},\omega_\nu)$ and $\hat{\gamma}(\vec{k})$ are 20x20 matrices with elements $K^{m,-n}_{\alpha\alpha'}(\vec{k},\omega_\nu)$, $M^{m,-n}_{\alpha\alpha'}(\vec{k},\omega_\nu)$ and $\gamma^{m,-n}_{\alpha\alpha'}(\vec{k})$, respectively, which are defined as

$$K^{m,-n}_{\alpha\alpha'}(\vec{k},\omega_\nu) = \sum_{\vec{R}_\alpha - \vec{R}'_{\alpha'}} \int_0^\beta d\tau\, e^{i\vec{k}(\vec{R}_\alpha - \vec{R}'_{\alpha'}) + i\omega_\nu\tau} \times$$

$$\times \langle \delta\tilde{O}^m(\vec{R}_\alpha,\tau)\,\delta O^{-n}(\vec{R}_{\alpha'})\rangle \qquad (2)$$

$$\delta\tilde{O}^m(\vec{R}_\alpha,\tau) = e^{\tau H}\delta O^m(\vec{R}_\alpha)e^{-\tau H} \qquad (3)$$

$$\delta O^m(\vec{R}_\alpha) = O^m(\vec{R}_\alpha) + 2\delta_{m,0} \qquad (4)$$

$$\langle \ldots \rangle = \frac{\mathrm{Tr}\, e^{-\beta H}}{\mathrm{Tr}\, e^{-\beta H}}, \qquad (5)$$

$H$ is the EQQ Hamiltonian, $O^m(\vec{R}_\alpha)$ $(m = 0, \pm 1, \pm 2)$ is the molecular quadrupole momentum operator (in $J = 1$ manifold of states) [5] related to the site of the crystal lattice denoted by the vector $\vec{R}_\alpha$, $\alpha = 1,2,3,4$ numbers



of the orientational sublattices, $M^{m,n}_{\alpha\alpha'}(\vec{k},\omega_\nu)$ is the Fourier-transform of the two-point irreducible (with respect to the interaction lines) part [9] and $\gamma^{m,n}_{\alpha\alpha'}(\vec{k})$ is the Fourier-transformed EQQ coupling constant $\gamma^{m,n}_{\alpha\alpha'}(\vec{R}'_{\alpha'} - \vec{R}_\alpha)$ [5,6]. Following Raich and Etters [5] we introduce the quasi-Pauli operators $a^+_i, b^+_i$ which create on the $i$-th molecule the $J = 1$ rotational state with $J^z_i = \pm 1$, respectively. We calculate $M^{m,n}_{\alpha,\alpha'}(\vec{k},\omega_\nu)$ to the first order in $1/z$ neglecting the wave structure of the librons. Before calculating $M^{m,n}_{\alpha\alpha'}(\vec{k},\omega_\nu)$ we obtain the renormalized propagator $\tilde{\mathcal{G}}_0(\omega_\nu)$ for the localized libron by summing some infinite class of the diagrams. The result is

$$\tilde{\mathcal{G}}_0(\omega_\nu) = \frac{-a_1}{i\omega_\nu - \tilde{\epsilon}_0} - \frac{a_2}{i\omega_\nu - \tilde{\epsilon}_2}, \qquad (6)$$

where

$$\tilde{\epsilon}_0 = 0.728\,\epsilon_0 = 16.91\,\Gamma \qquad (7)$$

is the renormalized energy of the localized libron,

$$\epsilon_0 = -12\sum_j \gamma^{oo}_{ij} = 21.2\,\Gamma \qquad [11] \qquad (8)$$

$$\tilde{\epsilon}_2 = 38.83\,\Gamma \qquad (9)$$

$\Gamma$ is the EQQ coupling parameter,

$$a_1 = 0.7713 \quad \text{and} \quad a_2 = 0.2287.$$

The presence in $\tilde{\mathcal{G}}_0(\omega_\nu)$ of the pole $\tilde{\epsilon}_2 = 2\tilde{\epsilon}_0$ points to the existence, in the system of two-libron excitations [7,12]. Of course, if $i\omega_\nu$ lies in the single libron band, the second part of $\tilde{\mathcal{G}}_0(\omega_\nu)$ can be neglected. To the first order in $1/z$ the matrix $\hat{M}(\vec{k},\omega_\nu)$ can be written as follows

$$\hat{M}(\omega_\nu, \vec{k}) = \| \delta_{\alpha\alpha'} M_{m,-n}(\omega_\nu) \| \qquad (10)$$

with non-zero functions

$$M_{m,-m}(\omega_\nu) \quad \text{and} \quad M_{1,2}(\omega_\nu) = M_{2,1}(\omega_\nu) = M^*_{-1,-2}(\omega_\nu) =$$

$$= M^*_{-2,-1}(\omega_\nu).$$

More detailed considerations show that for the frequency $i\omega_\nu$ of the libron band nearly total contribution to $\hat{M}$ is due to $M_{1,-1}(\omega_\nu) = M_{-1,1}(\omega_\nu) = M_1(\omega_\nu)$. The other functions $M_{mn}(\omega_\nu)$ contribute to higher orders in $1/z$.

Thus, we have

$$\hat{M}(\omega_\nu, \vec{k}) = M_1(\omega_\nu) \| \delta_{\alpha\alpha'} (\delta_{m,1} + \delta_{m,-1}) \|. \qquad (11)$$

Hence it is easy to see that in $1/z$ approximation the libron frequencies will be expressed by $\gamma^{m,n}_{\alpha\alpha'}(\vec{k})$ with $n,m = \pm 1$. The $M_1(\omega_\nu)$ to the first order in $1/z$ has the form (we use the renormalized propagator $\tilde{\mathcal{G}}_0(\omega_\nu)$ (6)).

$$M_1(\omega_\nu) = \frac{-4\tilde{\epsilon}_0 c}{(i\omega_\nu)^2 - \tilde{\epsilon}_0^2}, \qquad (12)$$

where

$$c = a_1 \left(1 - \frac{2B}{\tilde{\epsilon}_0^2} a_1^3\right) = 0.7694 \qquad (13)$$

with

$$B = \sum_j \left( |\gamma^{11}_{ij}|^2 + |\gamma^{1,-1}_{ij}|^2 \right). \qquad (14)$$

The $\vec{k}=0$ libron frequencies for $T_g^{(2)}$, $T_g^{(1)}$ and $E_g$ modes are $22.55\Gamma$, $13.38\Gamma$ and $10.60\Gamma$, respectively. The libron frequencies with $J=1 \to J=3$ corrections (see a note added in proof of ref. [3]) are calculated to be $23,12\Gamma$, $14,16\Gamma$ and $11,56\Gamma$ for $o-H_2$ and $12,87\Gamma$, $15,23\Gamma$ and $23,89\Gamma$ for $p-D_2$. By fitting to the Raman experiment we obtain the average (nonweighted) quadrupolar coupling parameter $\Gamma$ as 0.56 $cm^{-1}$ for $H_2$ and 0.70 $cm^{-1}$ for $D_2$ which are in good agreement with other experimental values. The average libron gap $\epsilon_L$ is calculated to be:

$$\epsilon_L = \tilde{\epsilon}_0 \left[1 + \frac{\epsilon_0^2}{\tilde{\epsilon}_0^2} (I_{+1} - 1)\right] = 16.29\Gamma, \qquad (15)$$

where $I_{+1}$ is the first moment of the density function for the harmonic libron spectrum [6]. This value is very close to that obtained in ref. [7].

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