Theory of Nonlinear Effects in Crystals in the Second-Quantization Representation

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A method for an exact theoretical treatment of nonlinear (anharmonic) effects in crystals in the second quantization representation is presented. The method represents essentially a generalization of Bogolyubov's method of approximate second quantization. In addition, the Fermi representation of elementary excitation creation and annihilation operators is given.

INTRODUCTION

THE basic idea in the application of the second-quantization method in theoretical solid-state physics is to reduce the description of phenomena in a system consisting of many interacting particles to the description of phenomena in a gas of quasiparticles. For this method to be applicable to the calculation of the relevant physical characteristics of the system, it is necessary that the quasiparticles obey Bose or Fermi statistics. In the rigorous formulation of the second-quantization method,[1,2] the creation and annihilation operators of excitations in molecules (these should not be confused with the creation and annihilation operators of molecules in given states) satisfy neither Bose nor Fermi commutation relations. These operators and the corresponding commutation relations will be called quasi-Pauli operators and quasi-Pauli commutation relations, respectively. The appearance of quasi-Pauli operators leads to serious difficulties in the application of the second-quantization method to the theoretical description of phenomena in solids. These difficulties are of two different kinds. First, the quasi-Pauli commutation relations are not invariant with respect to the standard transformation from the lattice space into the reciprocal lattice space, and this, as is well known, is the only transformation which enables one to use, in the simplest and the most efficient way, the translation symmetry of the crystal for finding collective modes of the crystal. Secondly, even if we could find another transformation to collective coordinates, which is canonical for these operators, we would still be faced with the difficulty that statistics have not been developed for this type of quasiparticle, and hence, we would not be able to use the standard statistical formulas for the calculation of the relevant physical characteristics of the system. The only way out of such a situation then is to express somehow the quasi-Pauli operators through Bose or Fermi operators. In this way, one substitutes a system of quasi-Pauli particles by an equivalent system of bosons or fermions. This equivalence should hold at least with respect to the integral physical characteristics of the corresponding systems. Such integral physical characteristics are, for example, internal energy, magnetization, dielectric constant, etc. For low excited states, i.e., for low concentrations of quasi-Paulions, the equivalent system is a system of noninteracting bosons. Formally, this means that in this case, the deviation of quasi-Pauli commutation relations from Bose commutation relations can be neglected. (see Ref. 1, p. 203.) This approximation is called the method of approximate second quantization.[1,3] In fact, one may say that until now the method of second quantization has been successfully used mainly in this approximation. There exist, however, physical situations in which the interaction of elementary excitations (nonlinear or anharmonic effects) cannot be neglected, because of their relatively high concentration. Such a situation exists, for instance, in the vicinity of the transition temperature in magnetic and ferroelectric materials and in molecular crystals when they are illuminated with laser beams, etc. In these cases the method of approximate second quantization, obviously, is not applicable. Because of the development of experimental techniques, which has been very intensive in recent years, just those physical situations, in which the concentration of quasiparticles is relatively high, are becoming more and more the subject of interest in both experimental and theoretical solid-state physics. It is, therefore, necessary to find an adequate method for the theoretical treatment of nonlinear effects within the frame of second quantization. In this work we suggest such a method.

In Sec. 1, Bogolyubov's method of second quantization in theoretical solid-state physics is presented. In Sec. 2 the exact Bose and Fermi representations of quasi-Pauli operators are given and Hamiltonians of dynamic, dynamico-kinematic, and kinematic interaction between bosons are formulated. In the Conclusion, we summarize possibilities of application of the method developed in Sec. 2.

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1. BOGOLYUBOV'S METHOD OF SECOND QUANTIZATION

In this section we shall present essential parts of the second-quantization method in solid-state physics, as developed by Bogolyubov[1,2]

Let us consider a crystal composed of N identical molecules.[4] The positions of crystal molecules shall be denoted by g. In the general case $g\equiv(\mathbf{n},\alpha)$, where $\mathbf{n}$ is the position vector of an elementary cell of the crystal, and α denotes the position of a molecule within the elementary cell. Throughout the paper we shall suppose that molecules do not oscillate around their equilibrium positions, i.e., phonon effects will be neglected.

For the basic system of functions we shall take the wave functions of isolated molecules, i.e., the solutions of the eigenproblem

$$\mathcal{H}_g\varphi_{\nu g}(x_g)=E_{\nu g}\varphi_{\nu g}(x_g)\,, \tag{1.1}$$

where $\mathcal{H}_g$ is the Hamiltonian of the gth isolated molecule, x_g is the set of internal coordinates of the gth molecules, and ν is a set of quantum numbers which characterizes the state of the molecule. The ground state of the molecule will be symbolically denoted by $\nu=0$.

In further treatment, we shall limit ourselves only to states of isolated molecules which are described by antisymmetric wave functions in the coordinates x_g. In most cases, though not necessarily, these are electronic states of the molecule, i.e., x_g are the coordinates of electrons in the molecule.

Let us designate by $\mathfrak{N}_{\nu g}$ the number of molecules in position g in the state ν and assume that ν takes a finite and ordered set of values $\nu=0,\,1,\,2,\,\cdots W$.[5] Let us also form the antisymmetrized and orthonormalized products

$$\Psi_{\dots\mathfrak{N}_{\nu g}\dots}(x_1\cdots x_g\cdots x_N)=[(NA)!]^{-1/2}\sum_P(-1)^P P\prod_{\nu g}\varphi_{\nu g}(x_g), \tag{1.2}$$

where A is the number of considered objects (electrons) inside the molecule, which are described by antisymmetric functions, and P is the permutation operator for a pair of objects (electrons).

In order to exclude from consideration the cases when two molecules in different states are in the same position g, we impose the following conditions on numbers $\mathfrak{N}_{\nu g}$ which take values 0 or 1

$$\sum_\nu\mathfrak{N}_{\nu g}=1;\quad\sum_{\nu g}\mathfrak{N}_{\nu g}=N. \tag{1.3}$$

Any state function of the considered system can be expanded in terms of the functions (1.2), i.e.,

$$\Psi(x_1\cdots x_g\cdots x_N)=\sum_{\cdots\mathfrak{N}_{\nu g}\cdots}C(\cdots\mathfrak{N}_{\nu g}\cdots)\Psi_{\dots\mathfrak{N}_{\nu g}\dots}\times(x_1\cdots x_g\cdots x_N), \tag{1.4}$$

where $C(\cdots\mathfrak{N}_{\nu g}\cdots)$ are the wave functions in the second-quantization representation.

Taken with an accuracy to two-particle interactions, the Hamiltonian of the considered system can be written in the form

$$\mathcal{H}=\sum_{g_1}\mathcal{H}_{g_1}+\tfrac{1}{2}\sum_{g_1g_2}{}'V_{g_1g_2}, \tag{1.5}$$

where $V_{g_1g_2}$ characterize the interactions of molecules in the positions g_1 and g_2. In the second-quantization representation the Hamiltonian (1.5) has the form (see Ref. 1, p. 191, and also Ref. 3, p. 48)

$$\mathcal{H}=\sum_{g_1\nu_1\nu_2}A_{g_1}(\nu_1,\nu_2)a_{\nu_1g_1}^\dagger a_{\nu_2g_1}+\tfrac{1}{2}\sum_{g_1g_2\nu_1\nu_2\nu_3\nu_4}B_{g_1g_2}(\nu_1\nu_2;\,\nu_3\nu_4)a_{\nu_1g_1}^\dagger a_{\nu_2g_2}^\dagger\times a_{\nu_4g_2}a_{\nu_3g_1}. \tag{1.6}$$

The operators $a_{\nu g}$ are defined in the following way:

$$\mathfrak{a}_{\nu g}C(\cdots\mathfrak{N}_{\nu g}\cdots)=(-1)^{\sum_{(\nu'g')<(\nu g)}\mathfrak{N}_{\nu'g'}}\times C(\cdots 1-\mathfrak{N}_{\nu g}\cdots)$$

$$\mathfrak{a}_{\nu g}^\dagger C(\cdots\mathfrak{N}_{\nu g}\cdots)=(-1)^{\sum_{(\nu'g')<(\nu g)}\mathfrak{N}_{\nu'g'}}(1-\mathfrak{N}_{\nu g})\times C(\cdots 1-\mathfrak{N}_{\nu g}\cdots). \tag{1.7}$$

They satisfy Fermi commutation relations in both indices,[6] and the matrix elements A_{g_1} and $B_{g_1g_2}$ $(g_1\neq g_2)$ are given by

$$A_{g_1}(\nu_1,\nu_2)=\int d\tau_{g_1}\varphi_{\nu_1g_1}^*\mathcal{H}_{g_1}\varphi_{\nu_2g_1}, \tag{1.8}$$

$$B_{g_1g_1}(\nu_1\nu_2;\nu_3\nu_4)=\int d\tau_{g_1}d\tau_{g_2}\varphi_{\nu_1g_1}^*\varphi_{\nu_2g_2}^*V_{g_1g_2}\varphi_{\nu_3g_1}\varphi_{\nu_4g_2}-\text{exchange terms}. \tag{1.9}$$

The operators $\mathfrak{a}_{\nu g}^\dagger$ generate Hilbert space $\mathfrak{R}$ of fermino states which is wider than that in which the conditions (1.3) are satisfied.[7] However, we shall consider only the part $\mathcal{F}$ of the space $\mathfrak{R}$ in which the conditions (Eq. 1.3) are satisfied. This subspace shall be

[4] $N=M\mathcal{S}$, where M is the number of elementary cells in crystal, and $\mathcal{S}$ is the number of molecules in an elementary cell.

[5] The case when ν takes an infinite number of different values can, in principle, be treated in the same way, but in practical calculations this leads to complications.

[6] In Ref. 2, the Hamiltonian (1.6) is expressed in terms of Pauli operators $b_{\nu g}$, which for the same g and ν obey Fermi commutation relations, while for different g and/or ν they commute. The connection between operators $b_{\nu g}$ and $\mathfrak{a}_{\nu g}$ is:

$$b_{\nu g}=(-1)^{\sum_{(\nu',g')<(\nu,g)}\mathfrak{N}_{\nu'g'}}\mathfrak{a}_{\nu g}.$$

The notation $(\nu'g')<(\nu g)$ (ν and g are ordered) means: if $g'<g$ then $(\nu',g')<(\nu,g)$; if $g'=g$ then $(\nu',g')<(\nu,g)$ if $\nu'<\nu$ (see Ref. 1, p. 166).

[7] The numbers $\mathfrak{N}_{\nu g}$, which appear in (Eq. 1.3), are the eigenvalues of the operator $\hat{\mathfrak{N}}_{\nu g}=\mathfrak{a}_{\nu g}^\dagger\mathfrak{a}_{\nu g}$.

called the space of physical states. The complementary subspace $\mathcal{N}$ of the space $\mathcal{R}$ will be called the space of nonphysical states. From the structure of the Hamiltonian (1.6) it is not difficult to see that it is closed inside $\mathcal{F}$.

If we introduce new Fermi operators $\mathcal{A}_{\mu g}$ by means of the unitary transformation

$$\mathcal{A}_{\nu g}=\sum_{\mu=0}^{W}\theta_g(\mu,\nu)\mathcal{A}_{\mu g}\,, \tag{1.10}$$

where the functions $\theta_g(\mu,\nu)$, apart from the unitarity condition, also satisfy the following conditions:

$$\sum_{\nu_3}A_{g_1}(\nu_1,\nu_3)\theta_{g_1}(\mu,\nu_3)+\sum_{g_2\nu_1\nu_2\nu_3(g_1\neq g_2)}B_{g_1g_2}(\nu_1\nu_2;\nu_3\nu_4)\times\theta_{g_2}^*(0\nu_2)\theta_{g_2}(0\nu_4)\theta_{g_1}(\mu\nu_3)=\lambda_{g_1}(\mu)\theta_{g_1}(\mu,\nu_1); \tag{1.11}$$

$\mu=0,\,1,\,2\cdots W$ (see Ref. 1, pp. 194, 196, and 197) and take into account the conditions [Eq. (1.3)] written in $\mathcal{R}$ by means of new Fermi operators $\mathcal{A}_{\mu g}$:

$$\sum_{\mu=0}^{W}\mathcal{N}_{\mu g}=1;\quad\sum_{\mu g}\mathcal{N}_{\mu g}=N\,, \tag{1.12}$$

then the Hamiltonian (1.6) can be written in the form:

$$\mathcal{H}_I=\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3\,, \tag{1.13}$$

where

$$\mathcal{H}_I=\mathcal{H}-E_0\,, \tag{1.13a}$$

$$\begin{aligned}\mathcal{H}_1=\sum_{g_1\mu_1}\mathcal{S}_{g_1}(\mu_1,0)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_1g_1}+\tfrac{1}{2}\sum_{g_1g_2\mu_1\mu_2}{}'\,&[T_{g_1g_2}(\mu_1\mu_2;00)\\ \times\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{0g_1}\mathcal{A}_{\mu_2g_2}^\dagger\mathcal{A}_{0g_2}&+T_{g_1g_2}(00;\mu_1\mu_2)\mathcal{A}_{0g_1}^\dagger\mathcal{A}_{\mu_1g_1}\\ \times\mathcal{A}_{0g_2}^\dagger\mathcal{A}_{\mu_2g_2}&+2T_{g_1g_2}(\mu_10;0\mu_2)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{0g_1}\\ &\times\mathcal{A}_{0g_2}^\dagger\mathcal{A}_{\mu_2g_2}]\,,\end{aligned} \tag{1.13b}$$

$$\begin{aligned}\mathcal{H}_2=\tfrac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3}{}'\,&[T_{g_1g_2}(\mu_1\mu_2;\mu_30)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_2g_2}^\dagger\mathcal{A}_{0g_2}\mathcal{A}_{\mu_3g_1}\\ &+T_{g_1g_2}(\mu_1\mu_2;0\mu_3)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_2g_2}^\dagger\mathcal{A}_{\mu_3g_2}\mathcal{A}_{0g_1}\\ &+T_{g_1g_2}(\mu_10;\mu_2\mu_3)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{0g_2}^\dagger\mathcal{A}_{\mu_3g_2}\mathcal{A}_{\mu_2g_1}\\ &+T_{g_1g_2}(0\mu_1;\mu_2\mu_3)\mathcal{A}_{0g_1}^\dagger\mathcal{A}_{\mu_1g_2}^\dagger\mathcal{A}_{\mu_3g_2}\mathcal{A}_{\mu_2g_1}]\,,\end{aligned} \tag{1.13c}$$

$$\mathcal{H}_3=\tfrac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3\mu_4}{}'\,T_{g_1g_2}(\mu_1\mu_2;\mu_3\mu_4)\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_2g_2}^\dagger\times\mathcal{A}_{\mu_4g_2}\mathcal{A}_{\mu_3g_1}\,, \tag{1.13d}$$

$$\mathcal{S}_{g_1}(\mu,0)=\lambda_{g_1}(\mu)-\lambda_{g_1}(0)\,, \tag{1.13e}$$

$$E_0=\lambda_{g_1}(0)\,, \tag{1.13f}$$

$$T_{g_1g_2}(\mu_1\mu_2;\mu_3\mu_4)=\sum_{\nu_1\nu_2\nu_3\nu_4}B_{g_1g_2}(\nu_1\nu_2;\nu_3\nu_4)\theta_{g_1}^*(\mu_1,\nu_1)\theta_{g_2}^*(\mu_2,\nu_2)\times\theta_{g_1}(\mu_3,\nu_3)\theta_{g_2}(\mu_4,\nu_4)\,. \tag{1.13g}$$ [8]

[8] Let us remark that in all formulas (1.13b–1.13g), $\mu_i\neq0$.

Let us now introduce the operators

$$\mathcal{P}_{\mu g}=\mathcal{A}_{0g}^\dagger\mathcal{A}_{\mu g}\quad\text{and}\quad\mathcal{P}_{\mu g}^\dagger=\mathcal{A}_{\mu g}^\dagger\mathcal{A}_{0g}\,. \tag{1.14}$$

It is evident that the operators $\mathcal{P}_{\mu g}^\dagger$ and $\mathcal{P}_{\mu g}$ are the creation and annihilation operators of the excitations of type μ on the molecule g and, as we said in the Introduction, we shall call them quasi-Pauli operators.

Since $\mathcal{A}_{\mu g}^\dagger$ and $\mathcal{A}_{\mu g}$ satisfy Fermi commutation relations, it can easily be seen that in the space $\mathcal{R}$ the operators $\mathcal{P}_{\mu g}^\dagger$ and $\mathcal{P}_{\mu g}$ satisfy the following commutation relations:

$$[\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_2}^\dagger]=\delta_{g_1g_2}(\hat{\mathcal{N}}_{0g_1}\delta_{\mu_1\mu_2}-\mathcal{A}_{\mu_2g_1}^\dagger\mathcal{A}_{\mu_1g_1})\,, \tag{1.15a}$$

$$[\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_2}]=[\mathcal{P}_{\mu_1g_1}^\dagger,\mathcal{P}_{\mu_2g_2}^\dagger]=0\,, \tag{1.15b}$$

$$\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_1}=\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_2g_1}^\dagger=0\,, \tag{1.15c}$$

$$\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_1}^\dagger=\hat{\mathcal{N}}_{0g_1}\mathcal{A}_{\mu_1g_1}\mathcal{A}_{\mu_2g_1}^\dagger\,,\quad\mu_1\neq\mu_2\,, \tag{1.15d}$$

$$\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_1g_1}=\hat{\mathcal{N}}_{\mu_1g_1}(1-\hat{\mathcal{N}}_{0g_1})\,. \tag{1.15e}$$

In the subspace $\mathcal{F}$ the relation (1.15e) becomes

$$\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_1g_1}=l_{\mu_1g_1}=\hat{\mathcal{N}}_{\mu_1g_1}=\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_1g_1} \tag{1.16e}$$

so that in this subspace the conditions (1.12) can be written as

$$l_{\mu g}=0\text{ or }1;\quad\sum_{\mu=1}^{W}l_{\mu g}=0\text{ or }1;\quad\sum_{\mu g}l_{\mu g}\leq N. \tag{1.12a}$$
$$\mu\neq0$$

Taking this into account, we see that operators $\mathcal{P}_{\mu g}$ satisfy in $\mathcal{F}$ the following commutation relations:

$$[\mathcal{P}_{\mu_1g_1},\mathcal{P}_{\mu_2g_2}^\dagger]=\delta_{g_1g_2}[\delta_{\mu_1\mu_2}(1-\sum_{\mu=1}^{W}l_{\mu g_1})-\mathcal{P}_{\mu_2g_1}^\dagger\mathcal{P}_{\mu_1g_1}]\,, \tag{1.16a}$$

$$[\mathcal{P}_{\mu_1g_1},\mathcal{P}_{\mu_2g_2}]=[\mathcal{P}_{\mu_1g_1}^\dagger,\mathcal{P}_{\mu_2g_2}^\dagger]=0\,, \tag{1.16b}$$

$$\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_1}=\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_2g_1}^\dagger=0\,, \tag{1.16c}$$

$$\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_1}^\dagger=0,\ (\mu_1\neq\mu_2)\,. \tag{1.16d}$$

In addition, the following is true in $\mathcal{F}$:

$$\mathcal{A}_{\mu_1g_1}^\dagger\mathcal{A}_{\mu_2g_1}=\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_2g_1}\,,\quad\mu_1\neq\mu_2\,. \tag{1.17}$$

Written in terms of the operators $\mathcal{P}_{\mu g}$, Hamiltonian (1.13) has the following form in $\mathcal{F}$:

$$\mathcal{H}=\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3\,, \tag{1.18}$$

where

$$\begin{aligned}\mathcal{H}_1=\sum_{g_1\mu_1}\mathcal{S}_{g_1}(\mu_10)\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_1g_1}+\sum_{g_1g_2\mu_1\mu_2}{}'\,&[T_{g_1g_2}(\mu_10;0\mu_2)\\ \times\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_2g_2}&+\tfrac{1}{2}T_{g_1g_2}(00;\mu_1\mu_2)\mathcal{P}_{\mu_1g_1}\mathcal{P}_{\mu_2g_2}\\ &+\tfrac{1}{2}T_{g_1g_2}(\mu_1\mu_2;00)\mathcal{P}_{\mu_1g_1}^\dagger\mathcal{P}_{\mu_2g_2}^\dagger]\,,\end{aligned} \tag{1.18a}$$

$$\mathcal{H}_2=\frac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3}{}' \big[T_{g_1g_2}(\mu_1\mu_2;\mu_30)\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_3g_1}\mathcal{P}_{\mu_2g_2}{}^{\dagger} + T_{g_1g_2}(\mu_1\mu_2;0\mu_3)\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\mathcal{P}_{\mu_3g_2} + T_{g_1g_2}(\mu_10;\mu_2\mu_3)\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_2g_1}\mathcal{P}_{\mu_3g_2} + T_{g_1g_2}(0\mu_1;\mu_2\mu_3)\mathcal{P}_{\mu_2g_1}\mathcal{P}_{\mu_1g_2}{}^{\dagger}\mathcal{P}_{\mu_3g_2}\big], \quad (1.18b)$$

$$\mathcal{H}_3=\frac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3\mu_4}{}' T_{g_1g_2}(\mu_1\mu_2;\mu_3\mu_4) \times\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_3g_1}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\mathcal{P}_{\mu_4g_2}. \quad (1.18c)$$

It is obvious from the procedure presented above that the operators $\mathcal{P}_{\mu g}$ are introduced in order to include as many terms of the fourth order (in operators $\mathcal{A}_{\mu g}$) as possible into the quadratic (in operators $\mathcal{P}_{\mu g}$) part of the Hamiltonian. Since the quadratic part of the Hamiltonian describes essentially the system of noninteracting quasiparticles, the transition to the operators $\mathcal{P}_{\mu g}$ makes it possible to include most of particle interactions into the energy of free quasiparticles. On the other hand, the transition to the operators $\mathcal{P}_{\mu g}$ leads, as we have already stated, to difficulties in connection with the commutation relations for these operators, which are of neither Bose nor Fermi type. The first difficulty arises when one attempts to diagonalize the quadratic form (1.18a). In order to accomplish this diagonalization, the quadratic form has to be expressed either through Bose or Fermi operators. This can be achieved, of course, simply by substituting Bose operators $\mathcal{B}_{\mu g}$ for quasi-Pauli operators $\mathcal{P}_{\mu g}$. Such an approach is called the approximate second quantization method. Obviously, the substitution $\mathcal{P}_{\mu g}=\mathcal{B}_{\mu g}$ introduces a certain error, because the commutation relations for $\mathcal{B}_{\mu g}$ and $\mathcal{P}_{\mu g}$ are not identical. This error, however, is small insofar as we consider only states for which $\langle \hat{l}_{\mu g}\rangle \approx 0$ ($\mu \neq 0$). It can be easily seen from the relation (1.16a) that in that case the substitution $\mathcal{P}_{\mu g}=\mathcal{B}_{\mu g}$ is justifiable. The same condition $\langle \hat{l}_{\mu g}\rangle \approx 0$, $\mu \neq 0$ which gives us right to make the substitution $\mathcal{P}_{\mu g}=\mathcal{B}_{\mu g}$ requires, on the other hand, the rejection of terms $\mathcal{H}_2$ and $\mathcal{H}_3$ in the Hamiltonian (1.18) so that the approximate second quantization method can be consistently applied only for the description of the system of noninteracting quasiparticles. For the description of the quasiparticle interaction effects (nonlinear or anharmonic effects), it is necessary to find an exact transition from the operators $\mathcal{P}_{\mu g}$ to Bose or Fermi operators, i.e., if we express $\mathcal{P}_{\mu g}$ as functions of Bose or Fermi operators, then these functions must satisfy all the commutation relations (1.16a–1.16e). The next section is devoted to the solution of this problem.

We shall end this section with the explanation of the origin of the term "quasi-Pauli operators," used here. Assuming that, besides the ground state, only one excited state (say μ_0) of the crystal molecules is effective (two-level scheme), one gets for the corresponding operators $\mathcal{P}_{\mu_0 g}$ the following commutation relations:

$$[\mathcal{P}_{\mu_0g_1},\mathcal{P}_{\mu_0g_2}{}^{\dagger}]=(1-2\mathcal{P}_{\mu_0g_1}{}^{\dagger}\mathcal{P}_{\mu_0g_1})\delta_{g_1g_2}, \quad (1.20a)$$

$$[\mathcal{P}_{\mu_0g_1},\mathcal{P}_{\mu_0g_2}]=[\mathcal{P}_{\mu_0g_1}{}^{\dagger},\mathcal{P}_{\mu_0g_2}{}^{\dagger}]=0, \quad (1.20b)$$

$$\mathcal{P}_{\mu_0g_1}{}^{2}=\mathcal{P}_{\mu_0g_1}{}^{2}=0. \quad (1.20c)$$

The operators which satisfy these commutation relations are called Pauli operators. In view of the fact that the commutation relations (1.20a–1.20c) are a special case of more general commutation relations (1.16a–1.16e), we have found it convenient to use term "quasi-Pauli operators" for the operators satisfying (1.20a–1.20c). Let us note that for the Pauli operators there exist the exact Bose[9] and Fermi representations.[10]

2. BOSE AND FERMI REPRESENTATIONS OF QUASI-PAULI OPERATORS

The states of the physical system with the Hamiltonian (1.18) are described by vectors from $\mathcal{F}$. For practical reasons, however, it is more convenient to work with the whole space $\mathcal{R}=\mathcal{F}\oplus\mathcal{N}$, generated by the operators $\mathcal{A}_{\mu g}{}^{\dagger}$. In that case it is necessary to exclude the contribution of states from $\mathcal{N}$, which are considered as nonphysical. In order to accomplish this, instead of working with the Hamiltonian $\mathcal{H}_I$ in the space $\mathcal{F}$, we shall work with the Hamiltonian

$$\tilde{\mathcal{H}}_I=\hat{G}\mathcal{H}_I\hat{G} \quad (2.1)$$

in the whole space $\mathcal{R}=\mathcal{F}\oplus\mathcal{N}$, where $\hat{G}$ is the projector on the $\mathcal{F}$, i.e., $\hat{G}=1$ in $\mathcal{F}$ and $\hat{G}=0$ in $\mathcal{N}$. The Hamiltonian $\tilde{\mathcal{H}}_I$ has the form

$$\tilde{\mathcal{H}}_I=\tilde{\mathcal{H}}_1+\tilde{\mathcal{H}}_2+\tilde{\mathcal{H}}_3, \quad (2.2)$$

where:

$$\tilde{\mathcal{H}}_1=\sum_{g_1\mu_1} \mathcal{S}_{g_1}(\mu_10)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_1g_1}\hat{\gamma}_{g_1}+\sum_{g_1g_2\mu_1\mu_2}{}' \big[T_{g_1g_2}(\mu_10;0\mu_2)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}\hat{\gamma}_{g_2}+\tfrac{1}{2}T_{g_1g_2}(\mu_1\mu_2;00) \times\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\hat{\gamma}_{g_2}+\tfrac{1}{2}T_{g_1g_2}(00;\mu_1\mu_2)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}\hat{\gamma}_{g_2}\big], \quad (2.2a)$$

$$\tilde{\mathcal{H}}_2=\frac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3}{}' \big[T_{g_1g_2}(\mu_1\mu_2;\mu_30)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_3g_1}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\hat{\gamma}_{g_2}+T_{g_1g_2}(\mu_1\mu_2;0\mu_3)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\mathcal{P}_{\mu_3g_2}\hat{\gamma}_{g_2} +T_{g_1g_2}(\mu_10;\mu_2\mu_3)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_2g_1}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_3g_2}\hat{\gamma}_{g_2}+T_{g_1g_2}(0\mu_1,\mu_2\mu_3)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_2g_1}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_1g_2}{}^{\dagger}\mathcal{P}_{\mu_3g_2}\hat{\gamma}_{g_2}\big], \quad (2.2b)$$

$$\tilde{\mathcal{H}}_3=\frac{1}{2}\sum_{g_1g_2\mu_1\mu_2\mu_3\mu_4}{}' T_{g_1g_2}(\mu_1\mu_2;\mu_1\mu_4)\hat{\gamma}_{g_1}\mathcal{P}_{\mu_1g_1}{}^{\dagger}\mathcal{P}_{\mu_3g_1}\hat{\gamma}_{g_1}\hat{\gamma}_{g_2}\mathcal{P}_{\mu_2g_2}{}^{\dagger}\mathcal{P}_{\mu_4g_2}\hat{\gamma}_{g_2}, \quad (2.2c)$$

[9] V. M. Agranović and B. S. Tošić, Zh. Eksperim. i Teor. Fiz. **53**, 149 (1967) [English transl.: Soviet Phys.—JETP **26**, 104 (1968)].
[10] B. S. Tošić and R. B. Žakula, Phys. Status Solidi **27**, 623 (1968).

and[11]

$$\hat{\gamma}_g=\prod_{\mu=1}^{W-1}\prod_{\mu'=\mu+1}^{W}(1-\hat{l}_{\mu g}\hat{l}_{\mu' g}). \tag{2.2d}$$

All the physical results which one can obtain working with the Hamiltonian (2.2a–2.2c) in the space $\mathfrak{R}$ are identical to the results which gives the Hamiltonian (1.18) restricted to the space $\mathfrak{F}$.

A. Bose Representation of Quasi-Pauli Operators

Let us introduce Bose operators $B_{\mu g}^\dagger$ and $B_{\mu g}$ which satisfy the well-known Bose commutation relations:

$$[B_{\mu_1 g_1},B_{\mu_2 g_2}^\dagger]=\delta_{g_1g_2}\delta_{\mu_1\mu_2};$$
$$[B_{\mu_1 g_1},B_{\mu_2 g_2}]=[B_{\mu_1 g_1}^\dagger,B_{\mu_2 g_2}^\dagger]=0, \tag{2.3}$$

where the eigenvalues $N_{\mu g}$ of the operator $\hat{N}_{\mu g}=B_{\mu g}^\dagger B_{\mu g}$ take values 0, 1, $2\cdots$, etc. In the Hilbert space $\mathfrak{B}$, which is spanned by eigenstates of the operators $\hat{N}_{\mu g}$, we define the operator function $\hat{Z}_{\mu g}$ in the following way:

$$\hat{Z}_{\mu g}=\sum_{\rho=0}^{\infty}\frac{(-2)^\rho}{(1+\rho)!}B_{\mu g}^{\rho+1\dagger}B_{\mu g}^{\rho+1}. \tag{2.4}$$

The operator $\hat{Z}_{\mu g}$ is diagonal in the representation of the occupation number $N_{\mu g}$ and has eigenvalues 0 for all states with even number of bosons and eigenvalues 1 for all states with odd number of bosons.[9]

From the Hilbert space $\mathfrak{B}$ we shall single out a subspace $\mathfrak{B}_1$ in which the following condition is satisfied:

$$\sum_{\mu=1}^{W}Z_{\mu g}=0 \text{ or } 1. \tag{2.5}$$

The complementary part of the space $\mathfrak{B}$ we shall denote with the symbol $\mathfrak{B}_2$ ($\mathfrak{B}=\mathfrak{B}_1\oplus\mathfrak{B}_2$).

Let us now introduce the projector on the space $\mathfrak{B}_1$:

$$\hat{\Pi}_g=\prod_{\mu=1}^{W-1}\prod_{\mu'=\mu+1}^{W}(1-\hat{Z}_{\mu g}\hat{Z}_{\mu' g}). \tag{2.6}$$

It can easily be seen from the properties of the operator $\hat{Z}_{\mu g}$ that the condition $\hat{\Pi}_g^2=\hat{\Pi}_g$ is satisfied and that $\hat{\Pi}_g$ has eigenvalues 1 in $\mathfrak{B}_1$ and 0 in $\mathfrak{B}_2$.

Let us further introduce the operator functions $\hat{P}_{\mu g}$, $\hat{P}_{\mu g}^\dagger$, and $\hat{L}_{\mu g}$:

$$\hat{P}_{\mu g}=(1-\sum_{\mu'\neq 0,\mu}^{W}\hat{Z}_{\mu' g})\hat{Y}_{\mu g}^{1/2}B_{\mu g}, \tag{2.7}$$

$$\hat{P}_{\mu g}^\dagger=(1-\sum_{\mu'\neq 0,\mu}^{W}\hat{Z}_{\mu' g})B_{\mu g}^\dagger\hat{Y}_{\mu g}^{1/2}, \tag{2.8}$$

$$\hat{L}_{\mu g}=(1-\sum_{\mu'\neq 0,\mu}^{W}\hat{Z}_{\mu' g})\hat{Z}_{\mu g}, \tag{2.9}$$

where

$$\hat{Y}_{\mu g}=\sum_{\rho=0}^{\infty}\frac{(-2)^\rho}{(1+\rho)!}B_{\mu g}^{\rho\dagger}B_{\mu g}^{\rho}. \tag{2.10}$$

We shall first show that in $\mathfrak{B}_1$ the following is true:

$$\hat{L}_{\mu g}=\hat{P}_{\mu g}^\dagger\hat{P}_{\mu g}=(1-\sum_{\mu'\neq 0,\mu}^{W}\hat{Z}_{\mu' g})\hat{Z}_{\mu g}=\hat{Z}_{\mu g}. \tag{2.11}$$

Indeed, for all states in $\mathfrak{B}_1$, for which $Z_{\mu g}=0$ the operator $\hat{L}_{\mu g}$ has also eigenvalues $L_{\mu g}=0$, while for all states in $\mathfrak{B}_1$ for which $Z_{\mu g}=1$, the operator

$$(1-\sum_{\mu'\neq 0,\mu}^{W}\hat{Z}_{\mu' g})$$

also has eigenvalues 1, and thus, for these states $L_{\mu g}=1$. Hence the operators $\hat{L}_{\mu g}$ and $\hat{Z}_{\mu g}$ are equal in $\mathfrak{B}_1$, since they have the same eigenstates and the same eigenvalues.

We shall now establish the commutation relations for the operators $\hat{P}_{\mu g}$ in the space $\mathfrak{B}_1$. Let us first show that the following is valid:

$$\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_2 g_1}=\hat{P}_{\mu_1 g_1}^\dagger\hat{P}_{\mu_2 g_1}^\dagger=0. \tag{2.12}$$

Since

$$\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_2 g_1}=(1-\sum_{\mu'\neq 0,\mu_1}^{W}\hat{Z}_{\mu' g_1})\hat{Y}_{\mu_1 g_1}^{1/2}B_{\mu_1 g_1} \times(1-\sum_{\mu'\neq 0,\mu_2}^{W}\hat{Z}_{\mu' g_1})\hat{Y}_{\mu_2 g_1}^{1/2}B_{\mu_2 g_1} \tag{2.12a}$$

it can easily be seen that the operator (2.12a) applied on the states for which $N_{\mu_2 g_1}$ is even (i.e., for which $Z_{\mu_2 g_1}=0$) is equal to zero, because after the action of the operator $B_{\mu_2 g_1}$ the operator $\hat{Y}_{\mu_2 g_1}^{1/2}$ acts on the state with an odd number of bosons of the type $\mu_2 g$ and thus gives zero.[12] If $N_{\mu_2 g_1}$ is odd (i.e., $Z_{\mu_2 g_1}=1$) then $N_{\mu_1 g_1}$ must be even (in $\mathfrak{B}_1$) and the action of the operator $\hat{Y}_{\mu_1 g_1}^{1/2}$ on such states gives zero because the operator $\hat{Y}_{\mu_1 g_1}^{1/2}$ is zero on the states with an odd number of bosons. With the same kind of reasoning one can easily show that the operators (2.12a) give zero for the states for which $N_{\mu_1 g_1}$ and $N_{\mu_2 g_1}$ are even. With this the proof that $P_{\mu_1 g_1}P_{\mu_2 g_1}=0$ is completed for all the states from $\mathfrak{B}_1$.

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[11] Strictly speaking, in Eq. (2.2a–2.2c) one should put the projectors $\gamma_g'=\prod_{\mu=0}^{W-1}\prod_{\mu'=\mu+1}(1-\hat{\mathfrak{N}}_{\mu g}\hat{\mathfrak{N}}_{\mu' g})$. However, it can easily be seen that the following is true in $\mathfrak{R}$:

$$\hat{\gamma}_g'\mathcal{P}_{\mu g}\hat{\gamma}_g'=\hat{\gamma}_g\mathcal{P}_{\mu g}\hat{\gamma}_g;\quad \hat{\gamma}_g'\mathcal{P}_{\mu g}^\dagger\hat{\gamma}_g'=\hat{\gamma}_g\mathcal{P}_{\mu g}^\dagger\hat{\gamma}_g$$

and

$$\hat{\gamma}_g\mathcal{P}_{\mu g}\hat{\gamma}_g\hat{\gamma}_g\mathcal{P}_{\mu' g}\hat{\gamma}_g=\hat{\gamma}_g\mathcal{P}_{\mu g}^\dagger\mathcal{P}_{\mu' g}\hat{\gamma}_g.$$

[12] The operators $\hat{Y}_{\mu\nu}^{1/2}$ have eigenvalues 0 for the states with an odd number of bosons and eigenvalues $(1+N_{\mu g})^{-1/2}$ for the states with an even number of bosons (for details see Ref. 9).

Let us now prove the second part of the assertion (2.12), i.e.,

$$\hat{P}_{\mu_1 g_1}{}^\dagger \hat{P}_{\mu_2 g_1}{}^\dagger = (1-\sum_{\mu'\neq 0,\mu_1}^{W} \hat{Z}_{\mu' g_1}) B_{\mu_1 g_1}{}^\dagger \hat{Y}_{\mu_1 g_1}{}^{1/2} \times (1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}) B_{\mu_2 g_1}{}^\dagger \hat{Y}_{\mu_2 g_1}{}^{1/2} = 0. \quad (2.12b)$$

Let us first consider the states for which all $N_{\mu g_1}$ are even. After the action of all operators standing in Eq. (2.12b) on the right-hand side of the operator

$$1-\sum_{\mu'\neq\mu_1,0}^{W} \hat{Z}_{\mu' g_1}$$

a state is obtained for which all $Z_{\mu g_1}$ are equal to zero except $Z_{\mu_1 g_1}$ and $Z_{\mu_2 g_1}$, which are equal to unity (i.e., $N_{\mu_1 g_1}$ and $N_{\mu_2 g_1}$ both are odd). For such a state the operator

$$1-\sum_{\mu'\neq\mu_1,0}^{W} \hat{Z}_{\mu' g_1}$$

gives zero. If we now consider a state for which $N_{\mu g_1}$ $(\mu\neq\mu_1,\mu_2)$ is odd [all other $N_{\mu' g_1}$ $(\mu'\neq\mu)$ must be even], then the operator

$$1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}$$

gives zero for such a state, because the application of the operator $B_{\mu_2 g_1}{}^\dagger \hat{Y}_{\mu_2 g_1}{}^{1/2}$ does not change the value of the number $N_{\mu g_1}$. If $N_{\mu_2 g}$ is odd (and all others are even) then (2.12b) is satisfied as a result of the action of the operator $\hat{Y}_{\mu_2 g_1}{}^{1/2}$. Finally, if $N_{\mu_1 g_1}$ is odd (and all the others are even), we conclude that the operator $B_{\mu_2 g_1}\hat{Y}_{\mu_2 g_1}{}^{1/2}$ does not change the number $N_{\mu_1 g_1}$ and therefore

$$1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}$$

gives zero for such states. With this we have completed the proof of the relation (2.12b).

On the basis of the above proof and the fact that the Bose operators when related to different g commute, it follows trivially that

$$[\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_2 g_2}] = [\hat{P}_{\mu_1 g_1}{}^\dagger, \hat{P}_{\mu_2 g_2}{}^\dagger] = 0. \quad (2.13)$$

Let us now prove that in $\mathfrak{B}_1$

$$\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_2 g_1}{}^\dagger = 0, \quad \mu_1\neq\mu_2. \quad (2.14)$$

From Eqs. (2.7) and (2.8) follows:

$$\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_2 g_1}{}^\dagger = (1-\sum_{\mu'\neq 0,\mu_1}^{W} \hat{Z}_{\mu' g_1}) \hat{Y}_{\mu_1 g_1}{}^{1/2} B_{\mu_1 g_1} \times (1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}) B_{\mu_2 g_1}{}^\dagger \hat{Y}_{\mu_2 g_1}{}^{1/2}. \quad (2.14a)$$

For the states for which $N_{\mu_2 g_1}$ is odd the relation (2.14) is fulfilled due to presence of the operator $\hat{Y}_{\mu_2 g_1}{}^{1/2}$. For the states for which $N_{\mu_1 g_1}$ is odd, the relation (2.14) is fulfilled because $B_{\mu_2 g_1}{}^\dagger \hat{Y}_{\mu_2 g_1}{}^{1/2}$ does not change the value of the number $N_{\mu_1 g_1}$ and the operator

$$1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}$$

gives zero when applied on such states. For the states for which one of the numbers $N_{\mu' g_1}$ $(\mu'\neq\mu_1\mu_2)$ is odd we obtain zero because of the action of the operator

$$1-\sum_{\mu'\neq 0,\mu_2}^{W} \hat{Z}_{\mu' g_1}$$

and the fact that the operator $B_{\mu_2 g_1}{}^\dagger \hat{Y}_{\mu_2 g_1}{}^{1/2}$ does not change the value of the number $N_{\mu' g_1}$ $(\mu'\neq\mu_1\mu_2)$ and finally, if all $N_{\mu g_1}$ are even, then after the application of all the operators which in Eq. (2.14a) are standing on right-hand side of $\hat{Y}_{\mu_1 g_1}{}^{1/2}$, this latter acts on the state with odd $N_{\mu_1 g_1}$ and thus gives zero.

Let us now consider the commutator:

$$\hat{K} = [\hat{P}_{\mu_1 g_1}, \hat{P}_{\mu_2 g_2}{}^\dagger]. \quad (2.15)$$

If $g_1\neq g_2$, then $\hat{K}=0$ because Bose operators with different indices commute. If $g_1=g_2$, and $\mu_1\neq\mu_2$, then from Eq. (2.14) follows:

$$\hat{K} = -\hat{P}_{\mu_2 g_1}{}^\dagger \hat{P}_{\mu_1 g_1}. \quad (2.15a)$$

Hence, it remains to consider the case when $g_1=g_2$ and $\mu_1=\mu_2$. For this case we shall show that

$$[\hat{P}_{\mu_1 g_1}, \hat{P}_{\mu_1 g_1}{}^\dagger] = 1-\sum_{\mu'=1}^{W} \hat{P}_{\mu' g_1}{}^\dagger \hat{P}_{\mu' g_1} - \hat{P}_{\mu_1 g_1}{}^\dagger \hat{P}_{\mu_1 g_1} \quad (2.15b)$$

which reduces to

$$\hat{P}_{\mu_1 g_1}\hat{P}_{\mu_1 g_1}{}^\dagger = 1-\sum_{\mu'=1}^{W} \hat{P}_{\mu' g_1}{}^\dagger \hat{P}_{\mu' g_1}. \quad (2.15c)$$

Taking into accounts Eqs. (2.7), (2.8), and (2.11), the relation (2.15c) can be written in the form

$$(1-\sum_{\mu'\neq 0,\mu_1}^{W} \hat{Z}_{\mu' g_1})(1+\hat{N}_{\mu_1 g_1})\hat{Y}_{\mu_1 g_1} = 1-\sum_{\mu'=1}^{W} \hat{Z}_{\mu' g_1}. \quad (2.15d)$$

From Eqs. (2.4) and (2.10) it can easily be seen that

$$B_{\mu_1 g_1}{}^\dagger \hat{Y}_{\mu_1 g_1} B_{\mu_1 g_1} = \hat{Z}_{\mu_1 g_1} \quad (2.16)$$

from which it follows that

$$B_{\mu_1 g_1} B_{\mu_1 g_1}{}^\dagger \hat{Y}_{\mu_1 g_1} B_{\mu_1 g_1} B_{\mu_1 g_1}{}^\dagger = B_{\mu_1 g_1} \hat{Z}_{\mu_1 g_1} B_{\mu_1 g_1}{}^\dagger. \quad (2.17)$$

Taking into account the spectrum of the operator $\hat{Z}_{\mu_1 g_1}$ one can conclude that in $\mathfrak{B}_1$

$$\hat{Z}_{\mu_1 g_1} B_{\mu_1 g_1}{}^\dagger = B_{\mu_1 g_1}{}^\dagger (1-\hat{Z}_{\mu_1 g_1}). \quad (2.18)$$

By combining Eqs. (2.17) and (2.18), we obtain

$$(1+\hat{N}_{\mu_1 g_1})\hat{Y}_{\mu_1 g_1}=1-\hat{Z}_{\mu_1 g_1}. \qquad (2.19)$$

From Eq. (2.19) and the fact that in $\mathfrak{B}_1$

$$\hat{Z}_{\mu_1 g_1}\hat{Z}_{\mu_2 g_1}=\hat{Z}_{\mu_1 g_1}\delta_{\mu_1\mu_2} \qquad (2.20)$$

we finally conclude that the left-hand side of the expression (2.15d) is identically equal to its right-hand side. All the derived properties of the commutator $\hat{K}$ can be written concisely in the following form:

$$\hat{K}=[\hat{P}_{\mu_1 g_1},\hat{P}_{\mu_2 g_2}{}^\dagger]$$
$$=\delta_{g_1 g_2}[\delta_{\mu_1\mu_2}(1-\sum_{\mu=1}^{W}\hat{Z}_{\mu g_1})-\hat{P}_{\mu_2 g_1}{}^\dagger\hat{P}_{\mu_1 g_1}]. \qquad (2.21)$$

On the basis of Eqs. (1.16a–1.16e), (2.21), (2.13), (2.12), (2.14), and (2.11) we can establish the following correspondence between the operators $\mathcal{P}_{\mu g}$ from $\mathfrak{F}_1$ and the operators $P_{\mu g}$ from $\mathfrak{B}_1$:

$$\mathcal{P}_{\mu g}\to\hat{P};\quad \mathcal{P}_{\mu g}{}^\dagger\to\hat{P}_{\mu g}{}^\dagger;\quad l_{\mu g}\to\hat{L}_{\mu g}=\hat{Z}_{\mu g}. \qquad (2.22)$$

This correspondence shall be called the Bose representation of quasi-Pauli operators.

Let us now define the operator:

$$H_I=H_1+H_2+H_3, \qquad (2.23)$$

where

$$H_1=\mathcal{H}_1(\mathcal{P}\to\hat{P}), \qquad (2.23a)$$

$$H_2=\mathcal{H}_2(\mathcal{P}\to\hat{P}), \qquad (2.23b)$$

$$H_3=\mathcal{H}_3(\mathcal{P}\to\hat{P}). \qquad (2.23c)$$

The notation $\mathcal{H}(\mathcal{P}\to\hat{P})$ means that we get the operators H when we substitute for the operators $\mathcal{P}_{\mu g}$ the operators $\hat{P}_{\mu g}$ in the corresponding $\mathcal{H}$ from Eqs. (1.18a–1.18c).

Since the operators $\hat{P}_{\mu g}$ in $\mathfrak{B}_1$ have the same commutation relations as the operators $\mathcal{P}_{\mu g}$ in $\mathfrak{F}_1$, the Hamiltonian H_I in $\mathfrak{B}_1$ is the Bose equivalent of the Hamiltonian $\mathcal{H}_I$ in $\mathfrak{F}$. For practical reasons, however, it is more convenient to work with the whole space $\mathfrak{B}=\mathfrak{B}_1\oplus\mathfrak{B}_2$. In order to exclude in that case the contribution of states from $\mathfrak{B}_2$, which are considered to be nonphysical, and which correspond to nonphysical states from $\mathfrak{R}$, we shall, instead of working with H_I in $\mathfrak{B}_1$, work in the whole space $\mathfrak{B}$ with the Hamiltonian:

$$\tilde{H}_I=\tilde{H}_1+\tilde{H}_2+\tilde{H}_3, \qquad (2.24)$$

where

$$\tilde{H}_1=\tilde{\mathcal{H}}_1(\mathcal{P}\to\hat{P},\mathcal{Y}\to\hat{\Pi}), \qquad (2.24a)$$

$$\tilde{H}_2=\tilde{\mathcal{H}}_2(\mathcal{P}\to\hat{P},\mathcal{Y}\to\hat{\Pi}), \qquad (2.24b)$$

$$\tilde{H}_3=\tilde{\mathcal{H}}_3(\mathcal{P}\to\hat{P},\mathcal{Y}\to\hat{\Pi}). \qquad (2.24c)$$

The notation (2.24a–2.24c) used here has the same meaning as in formulas (2.23a–2.23c).

On the basis of the results obtained, one can say that $B_{\mu g}$, $B_{\mu g}{}^\dagger$, and $\tilde{H}_I$ are the Bose equivalent of the system of quasi-Paulions with the Hamiltonian (1.18a–1.18c). It is obvious that the correspondence between spaces $\mathfrak{F}_1$ and $\mathfrak{B}_1$ is not isomorphic. In view of this, the mentioned equivalencey has a limited meaning, i.e., it can be said with certainty that it is valid only with respect to the statistical mean values of the physical characteristics of the system. As for other characteristics of the system (the dispersion law, for example), the question remains open whether the real excitations in the crystal are bosons or quasi-Paulions. The dilemma exists, of course, only in the case of high excited states of the system, because for low excited states (small concentrations of elementary excitations) the difference between quasi-Paulions and bosons is negligibly small (see the Introduction and Sec. 1).

If the Hamiltonian (2.24) is expanded into a power series in terms of Bose operators (the details of the expansion of the function $\hat{Y}_{\mu g}{}^{1/2}$ into the power series are given in Ref. 9) we obtain:

$$\tilde{H}_1=h_1{}^{(2)}+h_1{}^{(4)}+\cdots+h_1{}^{(2n)}+, \qquad (2.25)$$

$$\tilde{H}_2=h_2{}^{(3)}+h_2{}^{(5)}+\cdots+h_2{}^{(3+2n)}+, \qquad (2.26)$$

$$\tilde{H}_3=h_3{}^{(4)}+h_3{}^{(6)}+\cdots+h_3{}^{(4+2n)}+, \qquad (2.27)$$

where the index m denotes the order, in Bose operators, of a given term. For example, $h_1{}^{(4)}$ contains only the products of four Bose operators.

The Hamiltonian $h_1{}^{(2)}$ is identical with the Hamiltonian of the approximate second quantization method of Bogolyubov *et al.* (see Refs. 1, 2, and 3). The remaining terms in Eq. (2.25) are called the kinematic interaction because their appearance is the exclusive consequence of the specific commutation relations for the quasi-Pauli operators. The Hamiltonians $h_2{}^{(3)}$ and $h_3{}^{(4)}$ represent the dynamic interaction of elementary excitations and the remaining terms in Eqs. (2.26) and (2.27) shall be called the Hamiltonian of dynamico-kinematic interaction. The appearance of the Hamiltonians of the kinematic and the dynamico-kinematic interactions is characteristic for the Bose representation of quasi-Pauli operators. It is important to note that the effect of projector is felt only in the terms of the sixth and higher order in Bose operators. Hence, if we limit ourselves to work with an accuracy up to the terms of fourth order we then can leave out the projector in Eq. (2.24).[13]

B. Fermi Representation of Quasi-Pauli Operators

Let us introduce pairs of Fermi operators $F_{\mu g}$ and $f_{\mu g}$ which commute with each other and let us form out

[13] B. S. Tošić, Fiz. Tverd. Tela **9**, 1713 (1967) [English transl.: Soviet Phys.—Solid State **9**, 1346 (1967)].

of them the following operators:

$$Q_{\mu g}=(f_{\mu g}+f_{\mu g}^\dagger)(1-\sum_{\mu'\neq\mu}^{W} F_{\mu' g}^\dagger F_{\mu' g})F_{\mu g}, \qquad (2.28)$$

$$Q_{\mu g}^\dagger=(f_{\mu g}+f_{\mu g}^\dagger)F_{\mu g}^\dagger(1-\sum_{\mu'\neq\mu}^{W} F_{\mu' g}^\dagger F_{\mu' g}), \qquad (2.29)$$

$$\hat{D}_{\mu g}=Q_{\mu g}^\dagger Q_{\mu g}=(1-\sum_{\mu'\neq\mu}^{W} F_{\mu' g}^\dagger F_{\mu' g})F_{\mu g}^\dagger F_{\mu g}. \qquad (2.30)$$

These operators are defined in the Hilbert space ϕ which is the direct product of the Hilbert spaces generated by the operators $F_{\mu g}^\dagger$ and $f_{\mu g}^\dagger$. From the space ϕ we shall single out a subspace ϕ_1 by the condition

$$\sum_{\mu} F_{\mu g}^\dagger F_{\mu g}=0 \text{ or } 1. \qquad (2.31)$$

By the similar reasoning, as in the case of Bose representation, it can be shown that in ϕ_1 operators $Q_{\mu g}$ and $Q_{\mu g}^\dagger$ satisfy the following commutation relations:

$$[Q_{\mu_1 g_1}, Q_{\mu_2 g_2}^\dagger]=\delta_{g_1 g_2}[\delta_{\mu_1\mu_2}(1-\sum_{\mu=1}^{W}\hat{D}_{\mu g_1}) - Q_{\mu_2 g_2}^\dagger Q_{\mu_1 g_1}], \qquad (2.32)$$

$$[Q_{\mu_1 g_1}, Q_{\mu_2 g_2}]=[Q_{\mu_1 g_1}^\dagger, Q_{\mu_2 g_2}^\dagger]=0, \qquad (2.33)$$

$$Q_{\mu_1 g_1}Q_{\mu_2 g_2}=Q_{\mu_1 g_1}^\dagger Q_{\mu_2 g_2}^\dagger=0, \qquad (2.34)$$

$$Q_{\mu_1 g_1}Q_{\mu_2 g_2}^\dagger=0. \qquad (2.35)$$

In addition, the following is valid in ϕ_1:

$$\hat{D}_{\mu g}=Q_{\mu g}^\dagger Q_{\mu g}=F_{\mu g}^\dagger F_{\mu g}. \qquad (2.36)$$

Let us also introduce the projector on the subspace ϕ_1

$$\hat{L}_g=\prod_{\mu=1}^{W-1}\prod_{\mu'=\mu+1}^{W}(1-\hat{D}_{\mu g}\hat{D}_{\mu' g}) \qquad (2.37)$$

and the operator $\bar{H}$ which is obtained from Eq. (2.2) by the substitution:

$$\mathcal{P}_{\mu g}\rightarrow Q_{\mu g}, \quad \mathcal{P}_{\mu g}^\dagger\rightarrow Q_{\mu g}^\dagger \quad \text{and} \quad \hat{\gamma}_g\rightarrow \hat{\Gamma}_g.$$

The operator $\bar{H}$ in the space ϕ is the Fermi equivalent of the Hamiltonian (2.2) in the space $\mathcal{R}$. As in the case of the Bose representation, one can say that the operators $F_{\mu g}$, $F_{\mu g}^\dagger$, $f_{\mu g}$, $f_{\mu g}^\dagger$, and $\bar{H}$ are the Fermi equivalent of the system of quasi-Paulions with the Hamiltonian (1.18).

CONCLUSION

The Bose and Fermi representations of quasi-Pauli operators which are developed in Sec. 2 make it possible to study the nonlinear effect with the desired accuracy. In this way, the basic difficulties preventing further progress in application of the second quantization method in studies of solid state phenomena are solved. These difficulties, as we mentioned in the Introduction, are, first, noninvariance of the quasi-Pauli commutation relations with respect to the transformation which performs the transition from the lattice space to the reciprocal lattice space, necessary for the diagonalization of the quadratic part of the Hamiltonian, and, secondly, nonexistence of statistical formulas for quasi-Paulions.

The Bose representation of quasi-Pauli operators represents, in fact, a generalization of Bogolyubov's method of approximate second quantization [1], [3]. This generalization enables us to study the effects of interactions of elementary excitations in crystals. At the present time these effects are becoming more and more one of the central subjects of investigation in solid state physics. The Bose representation of quasi-Pauli operators is especially suitable for the description of the nonlinear effects in those cases in which the concentration of elementary excitations is sufficiently high (of the order of 10^{-6}–10^{-4}) that the nonlinear effects cannot be neglected but is still sufficiently low that the physical situation can be adequately described by using first few terms of the Hamiltonian (2.24), expanded into the power series in terms of Bose operators. We have such a situation, e.g., in nonlinear optics where the laser beams can produce concentrations of excitons of the order of 10^{-4}. In nonlinear optics the Bose representation of quasi-Pauli operators is also convenient for another reason. The proper description of nonlinear optical effects requires the examination of the whole system consisting of the crystal, the field of transverse photons, and their interaction. Since photons are bosons, this interaction can be most conveniently taken into account if exciton operators are expressed in terms of Bose operators (for details see Ref. 2).

The method developed here can obviously be applied, in a straightforward manner, in the quantum theory of magnetism and also in studies of ferroelectric phenomena in crystals. What concerns the first application, the method presented here represents a generalization of the method developed in Ref. 14. This will be discussed in more detail in forthcoming papers.

The Fermi representation of quasi-Pauli operators is suitable for the description of critical phenomena in magnetic and ferroelectric materials, because the concentration of elementary excitations in the vicinity of the transition temperature is close to unity, and in the Fermi representation the highest-order terms of the Hamiltonian of the system (taken with the accuracy to two-particle interactions) are of the fourth order in Fermi operators.

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