Phonon-induced quantum pair correlation in the diffraction of an ultracold atomic beam by a crystalline solid surface

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In this paper, a quantum-field theory is employed to study the diffraction of an ultracold atomic beam by a crystalline solid surface. We show that the crystalline solid surface acts as a nonlinear atomic grating for an ultracold atomic beam with a Bose condensate. The nonlinearity in such a mechanical atomic grating is due to the phonon exchange between the atoms in the ultracold atomic beam via the surface lattice vibration. Such a phonon exchange leads to an attractive interatom interaction in the ultracold atomic beam, which results in the generation of correlated atomic pairs in momentum space. Such correlated atomic pairs have similar properties to the conjugate photon pairs in nonclassical light.

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I. INTRODUCTION

Surface scattering of atomic and molecular beams has been an active topic in surface science over the past decade [1–3]. By detecting the trajectories of the scattered particles, one can study the realistic surface structures and the motion of lattice atoms in the surface. Hence the surface scattering provides a useful tool to "observe" atoms or molecules by themselves. However, the existing experiments and theory on surface scattering of atoms and molecules only concern the thermal beams with a sufficiently low density and a relatively short thermal de Broglie wavelength $\lambda_{dB}=\sqrt{2\pi\hbar^2/mk_BT}$. In this case, the information of some weak surface processes such as the lattice vibrations and superelastic scattering could be missed in the scattering beam since the thermal collisions between the atoms or molecules in the incident beam could be stronger than these surface processes.

Recently, laser cooling and trapping of neutral atoms has made great progress with the recoil limit broken [4–7]. By the current atomic-cooling techniques, a temperature on the order of microkelvin is achievable. In principle, lower temperatures in the nanokelvin and subnanokelvin ranges are also possible. Due to the long thermal de Broglie wavelength, such cold atoms have opened a window to study ultracold atomic collisions [6,8], wavelike behaviors of atoms in atom optics [9], quantum statistics of ultracold atoms in a light wave [10,11], and nonlinear atom optics [12–15]. In this paper, we consider a natural combination between two active topics, *surface scattering and ultracold atoms,* by replacing the thermal atomic beam with an ultracold atomic beam in surface scattering. To include the effects of quantum statistics on the surface scattering of ultracold atoms, a quantum field theory is employed to study the surface scattering processes. For simplicity, we assume that the surface under consideration is very "clean" with a perfectly periodic lattice structure and ignore all the competing processes from surface roughness. In this case, the interaction of the incident ultracold atomic beam with the surface could be understood as two parts: one is the interaction of the atoms in the incident atomic beam with the surface atoms in their thermal equilibrium positions. Under appropriate conditions, that part of the interaction determines the diffraction of the incident beam because of the spatially periodic structure of the surface lattice. The other part of the interaction is due to the thermal vibration of atoms in the surface lattice from their equilibrium positions, which leads to a coupling between atoms in the incident ultracold beam via surface phonon exchange. The main purpose of this paper is to study the collective quantum effect in the surface scattering of the ultracold atomic beam which is caused by the phonon exchange.

The paper is organized as follows. Section II is devoted to the derivations of a formal nonlinear Schrödinger equation for the surface scattering of atoms. Both the atomic beam and the surface lattice vibrations are treated in the quantum-field-theoretic framework. In Sec. III, we discuss the realistic structure of the surface potential. A series of coupled wave equations for the possible diffraction eigenstates are derived by solving the single-atom Schrödinger equation without the phonon-exchange effect. In terms of the single-atom diffraction eigenstates, the quantum pair correlation due to the phonon-exchange nonlinearity in atomic surface scattering is analyzed in Sec. IV. The incident atomic beam is assumed to be composed of a plane-wave Bose condensate with a high atomic density. We show that if there is no phonon nonlinearity or the atomic density of the condensate is so low that the phonon exchange nonlinearity is very weak, the condensate only results in a resonant excitation of diffraction eigenstates. However, when the phonon exchange nonlinearity is strong, further excitation of atoms toward other diffraction eigenstates happens because of

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the nonlinear multiwave mixing. We analyze the formation of correlated atomic pairs in such a multiwave mixing and study its effect on the surface scattering of ultracold atomic beams. The conclusions are included in Sec. V.

II. FORMAL THEORY OF ATOM-SURFACE INTERACTION

In this section, we develop a theoretical formalism for the atom-surface interaction in the framework of quantum-field theory. The motion of the atoms in the surface is mainly the thermal vibrations from the equilibrium positions. Hence the systematic Hamiltonian of an atom in the incident beam interacting with the atoms in the surface lattice can be expressed as

$$H=-\frac{\hbar^2\nabla^2}{2m}+\sum_i\left[\frac{\mathbf{P}_i^2}{2M_i}+\frac{1}{2}f_i\mathbf{u}_i^2\right]+V(\mathbf{r}) +\sum_i\left[\frac{\partial V(\mathbf{r};\mathbf{q}_1,\ldots,\mathbf{q}_i,\ldots)}{\partial\mathbf{q}_i}\right]_0\mathbf{u}_i\ , \tag{1}$$

where $\mathbf{r}$ is the position vector of the atom in the incident beam and $\mathbf{q}_i=\mathbf{q}_i^0+\mathbf{u}_i$ is that of the ith atom in the surface lattice. The ith atom in the surface lattice vibrates thermally in the vicinity of the equilibrium position $\mathbf{q}_i^0$ with the displacement $\mathbf{u}_i$, the momentum $\mathbf{P}_i=M_i(\partial\mathbf{u}_i/\partial t)$, and the modulus f_i. The averaged interaction potential $V(\mathbf{r})=\sum_i V_i(|\mathbf{r}-\mathbf{q}_i^0|)$ is the contribution of all surface atoms at the equilibrium positions. The last term of Eq. (1) describes the correction from the thermal vibrations of surface lattice. Generally, the potential correction term due to the thermal vibrations of surface lattice is small compared to the surface potential $V(\mathbf{r})$ and is treated as a perturbation in this paper. The displacement field and the momentum of the surface atoms in Eq. (1) can be expanded in terms of the surface waves [16–18]

$$\begin{aligned}\mathbf{u}_i&=\sum_{J,\mathbf{W}}\left[\frac{\hbar}{2\rho S v_s\alpha(\mathbf{W})}\right]^{1/2} \times[\beta_J\mathbf{e}_J(\mathbf{W})B_{\mathbf{W}}\exp(i\mathbf{W}^J\cdot\mathbf{q}_i^0)+\text{H.c.}]\ ,\\ \mathbf{P}_i&=\sum_{J,\mathbf{W}}\left[\frac{\hbar\rho v_s}{2S\alpha(\mathbf{W})}\right]^{1/2} \times[\beta_J\mathbf{e}_J(\mathbf{W})B_{\mathbf{W}}\exp(i\mathbf{W}^J\cdot\mathbf{q}_i^0)(-i\mathbf{W}^J)+\text{H.c.}]\ ,\end{aligned} \tag{2}$$

where $\rho=N_0M_i/LS$ is the density of atoms in the crystal, S the surface area, L the thickness of the crystal, v_s the sound velocity of surface waves, $\mathbf{e}_J(\mathbf{W})$ the polarization vector of the surface waves, and $\mathbf{W}^J=(W_x,W_y,iW_z^J)$ the wave vector of the surface waves. The imaginary component of the wave vector $\mathbf{W}^J$ in the direction normal to the crystal surface describes the localization of the surface sound waves. The numerical factor β_J is chosen to satisfy the boundary condition of zero stress at the crystal surface x-y plane and determines an amplitude normalization constant

$$\alpha(\mathbf{W})=\sum_{JJ'}\frac{\beta_J^*\beta_{J'}\mathbf{e}_J(\mathbf{W})^*\cdot\mathbf{e}_{J'}(\mathbf{W})}{W_z^J-W_z^{J'}}\ .$$

In the second quantization description of surface sound waves, $B_{\mathbf{W}}$ and its Hermitian conjugate $B_{\mathbf{W}}^\dagger$ are the annihilation and creation operators of surface phonons. Substituting Eq. (2) into Eq. (1), the second term of Hamiltonian (1) is reduced into

$$H_{\text{ph}}=\sum_{\mathbf{W}}\hbar\omega_{\mathbf{W}}(B_{\mathbf{W}}^\dagger B_{\mathbf{W}}+\tfrac{1}{2}) \tag{3}$$

and the fourth term can be expressed as

$$H_{a\text{-ph}}=\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})B_{\mathbf{W}}+\text{H.c.}\ , \tag{4}$$

where the subscript a-ph denotes the coupling of atom and phonon, $\omega_{\mathbf{W}}=v_s\sqrt{W_x^2+W_y^2}$ denotes the surface phonon frequency, and

$$\eta(\mathbf{r},\mathbf{W})=\sum_i\sum_J\left[\frac{\hbar}{2\rho S v_s\alpha(\mathbf{W})}\right]^{1/2} \times\beta_J\left[\frac{\partial V(\mathbf{r},\ldots,\mathbf{q}_i,\ldots)}{\partial\mathbf{q}_i}\right]_0\cdot\mathbf{e}_J(\mathbf{W}) \times\exp(i\mathbf{W}^J\cdot\mathbf{q}_i^0) \tag{5}$$

is the phonon-induced potential function of the atom in the incident ultracold atomic beam.

In the above discussions, we only consider a single atom in the incident beam interacting with the crystalline solid surface. For an ultracold atomic beam composed of degenerate Bose condensate, the quantum statistics is of crucial importance in the diffraction of ultracold atoms by the surface lattice, which has not yet been studied in the literature of atom-surface scattering. In order to incorporate the quantum statistics into the atom-surface scattering, we adopt the quantum-field-theoretic description of the ultracold atomic ensemble. In terms of the field-theoretic language, the total Hamiltonian of the system composed of the ultracold atoms in the incident beam and the atoms in the crystalline solid surface can be expressed as

$$H_{\text{sys}}=\int d^3r\,\psi^\dagger(\mathbf{r})\left[-\frac{\hbar^2\nabla^2}{2m}+V(\mathbf{r})\right]\psi(\mathbf{r})+H_{\text{ph}} +\int d^3r\,\psi^\dagger(\mathbf{r})\left[\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})B_{\mathbf{W}}+\text{H.c.}\right]\psi(\mathbf{r})\ , \tag{6}$$

where the operator $\psi(\mathbf{r})$ and its Hermitian conjugate $\psi^\dagger(\mathbf{r})$ describe the annihilation and creation of the atoms at the position $\mathbf{r}$. Hamiltonian (6) determines the following Heisenberg equations of motion for the surface phonons and the atoms in the ultracold atomic beam:

$$i\hbar\frac{\partial B_{\mathbf{W}}(t)}{\partial t}=\hbar\omega_{\mathbf{W}}B_{\mathbf{W}}+\int d^3r\,\eta(\mathbf{r},\mathbf{W})^*\psi^\dagger(\mathbf{r},t)\psi(\mathbf{r},t)\ , \tag{7a}$$

$$i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t}=\left[-\frac{\hbar^2\nabla^2}{2m}+V(\mathbf{r})+\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})B_{\mathbf{W}}+\text{H.c.}\right]\psi(\mathbf{r},t)\ . \tag{7b}$$

In Eqs. (7), we see that the surface phonons and the atoms in the ultracold beam are coupled by the potential function $\eta(\mathbf{r},\mathbf{W})$, which is due to the thermal vibrations of the surface atoms. Solving Eq. (7a) and substituting the solution into Eq. (7b), one can eliminate the phonon annihilation and creation operators $B_{\mathbf{W}}$ and $B_{\mathbf{W}}^{\dagger}$ in Eq. (7b). This leads to a nonlinear stochastic Schrödinger equation for the atomic field operator of the incident atomic beam

$$i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t}=\left\{-\frac{\hbar^2\nabla^2}{2m}+V(\mathbf{r})+V_R(\mathbf{r},t)+\left[\int d^3r'\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})\eta(\mathbf{r}',\mathbf{W})^*\frac{1}{i\hbar}\int_0^{\infty}d\tau\, e^{-i\omega_{\mathbf{W}}\tau}\psi^{\dagger}(\mathbf{r}',t-\tau)\psi(\mathbf{r}',t-\tau)+\text{H.c.}\right]\right\}\psi(\mathbf{r},t)\ , \tag{8a}$$

where $V_R(\mathbf{r},t)\equiv\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})B_{\mathbf{W}}(0)e^{-i\omega_{\mathbf{W}}t}+\text{H.c.}$ gives a random potential which is induced by the thermal vibration phonons. The statistical correlation function of the random potential has the form

$$\begin{aligned}\langle V_R(\mathbf{r},t)V_R(\mathbf{r}',t')\rangle&=\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})^*\eta(\mathbf{r}',\mathbf{W})n_{\mathbf{W}}(T_0)e^{i\omega_{\mathbf{W}}(t-t')}\\&\quad+\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})\eta(\mathbf{r}',\mathbf{W})^*[1+n_{\mathbf{W}}(T_0)]e^{-i\omega_{\mathbf{W}}(t-t')}\ ,\end{aligned} \tag{8b}$$

where $n_{\mathbf{W}}(T_0)$ is the number of phonons in the surface wave with wave vector $\mathbf{W}$ and T_0 is the temperature of the crystal surface. The nonlinear term in Eq. (8a) results from the exchange of phonons between the atoms in the ultracold atomic beam, which leads to a direct interaction of atoms in this beam. This case is similar to the photon exchanges between atoms which leads to a ground–excited-state atomic nonlinearity [11–15]. The only difference between phonon exchange and photon exchange is that the phonons can be exchanged directly between ground-state atoms and the photon exchange only happens between ground- and excited-state atoms. In this paper, no electromagnetic interaction is involved and hence no atom in the atomic beam will be excited. As a result, the dipole-dipole interaction due to atomic excitation will not have any effect on the surface scattering processes. In addition, some other many-body effects which possibly affect the scattering processes are the short-range ground–ground-state atomic collisions. However, if we assume that the incident atomic beam is prepared in a macroscopic single quantum state with a Bose condensate, these many-body effects have been incorporated into the formation of such a stable condensate in advance. Hence the dominant atomic nonlinearity in the present problem is the interatom interaction due to phonon exchange. On the other hand, in terms of Eq. (8b), the effects from the random potential $V_R(\mathbf{r},t)$ depend on the temperature of the crystal surface. The random potential causes incoherent scattering of atoms which can destroy the coherence of the atomic beam. To reduce the incoherent scattering, a "cold" surface is required. This can be realized by cooling the crystal in a low-temperature liquid, for example, liquid He. For a cold crystal surface, only few surface phonon modes are excited and a large number of phonons are left in vacuum states. In this case, the random potential $V_R(\mathbf{r},t)$ is negligible compared to the averaged surface potential $V(\mathbf{r})$. Further we assume that the thermal vibration of surface lattice atoms has a large phonon bandwidth $\Delta\omega_{\mathbf{W}}$ so that the energy change of atoms in the atomic beam due to phonon exchange satisfies the relation $E_{\mathbf{k}'}-E_{\mathbf{k}}\ll\hbar\Delta\omega_{\mathbf{W}}$. Then the integral over time τ in Eq. (8a) can be approximated by adiabatically removing the atomic field operators from within the integral. Such an assumption can be valid for the interaction of atoms (or other heavy particles) with wideband phonons. But it is invalid for the interaction of light particles such as electrons with phonons [19]. By these above assumptions, Eq. (8a) can be finally reduced to the form

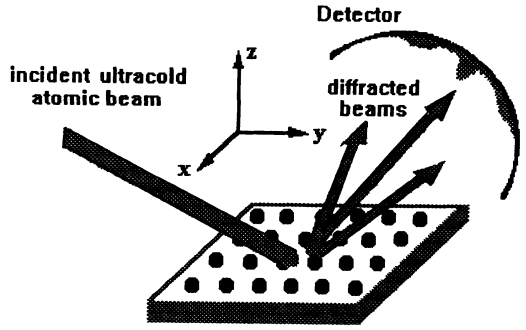

FIG. 1. Schematic diagram for the diffraction of an ultracold atomic beam by a crystalline solid surface atomic grating.

$$i\hbar\frac{\partial\psi}{\partial t}=\left[-\frac{\hbar^2\nabla^2}{2m}+V(\mathbf{r})\right]\psi + \int d^3r' Q(\mathbf{r},\mathbf{r}')\psi^\dagger(\mathbf{r}')\psi(\mathbf{r}')\psi(\mathbf{r}) , \tag{9a}$$

$$Q(\mathbf{r},\mathbf{r}')=-\left\{\sum_{\mathbf{W}}\eta(\mathbf{r},\mathbf{W})\eta(\mathbf{r}',\mathbf{W})^*/\hbar\omega_{\mathbf{W}}+\text{H.c.}\right\} . \tag{9b}$$

The negative sign in expression (9b) indicates that the phonon exchanges induce an attractive force between atoms in the beam. Equations (9) determine the dynamic scattering processes of ultracold atomic beams by a crystalline solid surface. A schematic diagram for such a scattering is shown in Fig. 1.

III. SURFACE POTENTIALS AND DIFFRACTION EIGENSTATES

The basic formalism given in Sec. II is only formal. The exact forms of the surface potential $V(\mathbf{r})$ and the nonlinear correlation coefficient $Q(\mathbf{r},\mathbf{r}')$ depend on the practical structure of the crystal surface, which is chosen as the atomic grating. So far there is no good theory to describe the realistic surface structure and the relevant surface potential. In the literature of surface scattering, the often used models for some solid surfaces can basically be divided into three types. The first type was initiated by Lennard-Jones [20]. This theory is based on a first-order distorted-wave Born approximation. In the theory, the surface potential is presented by a Morse potential. Experiments show that the Lennard-Jones approach is inadequate for a useful description of elastic scattering data. However, because of the simplicity in the theoretic analysis of surface scattering, it is still widely adopted. Another typical model is to simplify the surface potential as a hard wall with a periodic variation [2]. The third type of model is to expand the surface potential in the Fourier transformation in terms of the surface reciprocal lattice vector. In such an expansion, each order Fourier component can be modeled in terms of the realistic experimental data of surface scattering. The model is suitable for arbitrary surface structure, but usually no analytic function is available for the surface potential. In this paper, we employed the general Fourier expansion for the surface potential and the phonon-induced potential in terms of the periodicity of the crystal surface as

$$V(\mathbf{r})=\sum_{\mathbf{G}}V_{\mathbf{G}}(z)\exp(i\mathbf{G}\cdot\boldsymbol{\rho}) , \tag{10a}$$

$$\eta(\mathbf{r},\mathbf{W})=\sum_{\mathbf{G}}\eta_{\mathbf{G}}(z,\mathbf{W})\exp(i\mathbf{G}\cdot\boldsymbol{\rho}) , \tag{10b}$$

where $\boldsymbol{\rho}=(x,y)$ denotes the position vector in the plane parallel to the crystal surface and $\mathbf{G}$ is a reciprocal lattice vector of the surface. The Fourier components $V_{\mathbf{G}}(z)$ and $\eta_{\mathbf{G}}(z,\mathbf{W})$ in expressions (10) depends on the individual interaction of the atom in the atomic beam and the atom in the surface. For a realistic surface, a reasonable choice for the individual interaction is to model it by a van der Waals potential. The scattering experiments on the He-LiF surface interaction and numerical analysis of the scattering data in terms of expansion (10a) show that the Fourier components of the surface potential can be approximated by the analytic functions

$$V_0(z)=d\left[\left(\frac{z_0}{z}\right)^{12}-4\left(\frac{z_0}{z}\right)^{3}\right] ,$$
$$V_{\mathbf{G}}(z)=d_{\mathbf{G}}\left(\frac{z_0}{z}\right)^{14}\quad(\mathbf{G}\neq 0) , \tag{11}$$

where $d=80$ cal/mol, $z_0=2.65$ Å, $d_{1,0}=d_{0,1}=6$ cal/mol, and $d_{1,1}=5$ cal/mol. The higher-order components in $\mathbf{G}$ have very small values and can be ignored. Figure 2 shows the surface potential and its equipotential curves in terms of Eq. (10a) and the parameters given in Eqs. (11). The Fourier components of the phonon-induced potential should have forms similar to Eqs. (11). But so far there has not yet been any analytic functions available to describe them.

In this paper, our purpose is to study the quantum pair correlation induced by the phonon-exchange nonlinearity. At this stage, the exact function forms of these Fourier components of the phonon-induced potential are not important to discuss the quantum correlation in the surface scattering and we can leave them as the adjustable parameters in our discussions. As the beginning of our discussions, we solve the Schrödinger equations without the phonon exchange nonlinearity and find all possible diffraction states determined by the surface potential $V(\mathbf{r})$. If these diffraction states are denoted as $\phi_{\mathbf{K}}$ with eigenvalues $E_{\mathbf{K}}$, we have the following Schrödinger equation:

$$\left[-\frac{\hbar^2\nabla^2}{2m}+V_0(z)+2V_{0,1}(z)[\cos(Gx)+\cos(Gy)] +2V_{1,1}(z)\cos(Gx+Gy)\right]\phi_{\mathbf{K}}=E_{\mathbf{K}}\phi_K , \tag{12}$$

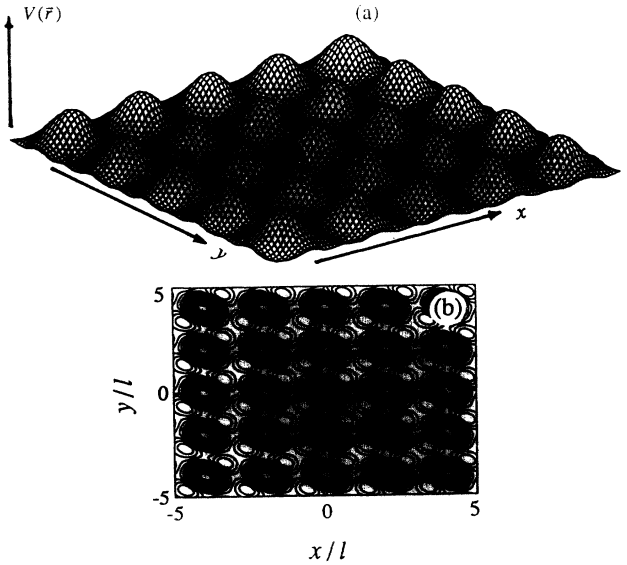

FIG. 2. (a) The surface potential $V(\mathbf{r})$ at $z=2$ Å and (b) the equipotential curves in the x-y plane. The calculation is performed in terms of expressions (10a) and (11).

where we have ignored the higher-order Fourier components of the surface potential and only consider the (0,0), (0,1), and (1,1) components. The reciprocal vector is chosen to have the same project in the x and y direction, i.e., $G_x=G_y=G=2\pi/l$ with l the project of the translation vector in the x and y directions. In terms of the periodicity of the surface potential, the diffraction eigenstates will include the same periodic information as the surface structure. Hence the diffraction states can be expanded into the forms

$$\phi_{\mathbf{K}}(\mathbf{r})=\frac{1}{\sqrt{V}}\exp(i\mathbf{K}\cdot\mathbf{r})u_{\mathbf{K}}(\mathbf{r}) ,$$
$$u_{\mathbf{K}}(\mathbf{r})\equiv\sum_{nm}\varphi^{\mathbf{K}}_{nm}(z)\exp(inGx+imGy) , \tag{13}$$

where the wave vector $\mathbf{K}=(k_x,k_y,k_z)$ is the carrier wave vector of the diffraction states. The physics implicit in Eq. (13) is evident: a plane atomic wave impinging on a crystal surface is modulated by the periodic structure of the surface with the modulation wave function $u_{\mathbf{K}}(\mathbf{r})$. The modulation sideband wave vector is identical to the reciprocal lattice vector of the surface. The case is similar to that of a light wave transmitting a light modulator or reflected by a reflecting grating. Substituting Eq. (13) into (12), the scattering waves in the z direction satisfy the coupled wave equation

$$\left[\frac{d^2}{dz^2}+2ik_z\frac{d}{dz}+\left[k_z'^2-\frac{2mV_0(z)}{\hbar^2}\right]\right]\varphi^{\mathbf{K}}_{nm}(z)$$
$$=\frac{2mV_{0,1}(z)}{\hbar^2}[\varphi^{\mathbf{K}}_{n-1,m}(z)+\varphi^{\mathbf{K}}_{n+1,m}(z)+\varphi^{\mathbf{K}}_{n,m-1}(z)+\varphi^{\mathbf{K}}_{n,m+1}(z)]$$
$$+\frac{2mV_{1,1}(z)}{\hbar^2}[\varphi^{\mathbf{K}}_{n-1,m-1}(z)+\varphi^{\mathbf{K}}_{n+1,m+1}(z)] , \tag{14}$$

where $k_z'^2\equiv(2mE_{\mathbf{K}}/\hbar^2)-(k_x+nG)^2-(k_y+mG)^2-k_z^2$. Equation (14) determines two types of eigenstates: bound states and reflection states. The bound states correspond to the eigenenergy $E_{\mathbf{K}}<0$ and localize at the crystal surface with an imaginary wave vector $i|k_z'|$. The reflection states are the positive eigenenergy states with wave vector k_z' as a real number which connects to the diffraction states of the crystal surface. Here we assume that the incident atomic beam has a kinetic energy E which is large enough to allow the reflection states. In this case, Eq. (14) can be solved by the standard scattering theory with the boundary condition

$$\varphi^{\mathbf{K}}_{nm}(z)\xrightarrow[z\to\infty]{}\exp(-ik_zz)+k_z'^{-1/2}R_{nm}\exp[ik_z'z] . \tag{15}$$

In terms of Eqs. (13)–(15) and (11), numerically one can work out the practical spatial patterns of the diffraction states (13). However, this is not the main task of this paper. Comparing Eq. (13) with the surface potential (10a), one can conclude that the shapes of the diffraction patterns have similar properties to those of the surface potential shown in Fig. 2.

IV. QUANTUM PAIR CORRELATION IN SURFACE SCATTERING OF ULTRACOLD ATOMS

In this section, we consider the exact Schrödinger equation (9) with the phonon-exchange nonlinearity. The Schrödinger equation (9) determines an effective Hamiltonian for the ultracold atomic beam with the form

$$H_{\text{eff}}=\int d^3r\,\psi^\dagger(\mathbf{r})\left[-\frac{\hbar^2\nabla^2}{2m}+V(\mathbf{r})\right]\psi(\mathbf{r})$$
$$+\tfrac{1}{2}\int d^3r\int d^3r'Q(\mathbf{r},\mathbf{r}')\psi^\dagger(\mathbf{r})\psi^\dagger(\mathbf{r}')\psi(\mathbf{r}')\psi(\mathbf{r}) . \tag{16}$$

In terms of the diffraction states (13), we expand the atomic-quantum-field operator in Eq. (16) in the form $\psi(\mathbf{r})=\sum_{\mathbf{K}}a_{\mathbf{K}}\phi_{\mathbf{K}}$. The operator $a_{\mathbf{K}}$ and its Hermitian conjugate $a^\dagger_{\mathbf{K}}$ describe the annihilation and creation of atoms in the diffraction states $\phi_{\mathbf{K}}$. For an incident ultracold atomic beam composed of bosonic atoms, the operators $a_{\mathbf{K}}$ and $a^\dagger_{\mathbf{K}}$ obey the Bose commutation. By employing the operators $a_{\mathbf{K}}$ and $a^\dagger_{\mathbf{K}}$, Hamiltonian (16) can be expressed as

$$H_{\text{eff}}=\sum_{\mathbf{K}}E_{\mathbf{K}}a^\dagger_{\mathbf{K}}a_{\mathbf{K}}$$
$$+\frac{1}{2V}\sum_{\mathbf{K}_1}\sum_{\mathbf{K}_2}\sum_{\mathbf{K}_3}\sum_{\mathbf{K}_4}U(\mathbf{K}_1,\mathbf{K}_2,\mathbf{K}_3,\mathbf{K}_4)$$
$$\times a^\dagger_{\mathbf{K}_1}a^\dagger_{\mathbf{K}_2}a_{\mathbf{K}_3}a_{\mathbf{K}_4} , \tag{17a}$$

where

$$U(\mathbf{K}_1,\mathbf{K}_2,\mathbf{K}_3,\mathbf{K}_4)$$
$$=-\frac{1}{V}\sum_{\mathbf{W}}\frac{1}{\hbar\omega_{\mathbf{W}}}\{\gamma_{\mathbf{W}}(\mathbf{K}_1,\mathbf{K}_4)\gamma_{\mathbf{W}}(\mathbf{K}_3,\mathbf{K}_2)^*+\gamma_{\mathbf{W}}(\mathbf{K}_4,\mathbf{K}_1)^*\gamma_{\mathbf{W}}(\mathbf{K}_2,\mathbf{K}_3)\} ,$$
$$\gamma_{\mathbf{W}}(\mathbf{K}_i,\mathbf{K}_j)=\int d^3r\,\eta(\mathbf{r},\mathbf{W})u_{\mathbf{K}_i}(\mathbf{r})^*u_{\mathbf{K}_j}(\mathbf{r})\times\exp[-i(\mathbf{K}_i-\mathbf{K}_j)\cdot\mathbf{r}] . \tag{17b}$$

The function $\gamma_{\mathbf{W}}(\mathbf{K}_i,\mathbf{K}_j)$ is the general Fourier transformation of the phonon-induced potential. In terms of Eq. (17b), the significant nonlinear terms in Hamiltonian (17a) are approximately determined by the carrier-wave resonance condition $\mathbf{K}_1-\mathbf{K}_4=\mathbf{K}_3-\mathbf{K}_2=\mathbf{K}$ in the diffraction. The other terms off resonance can be neglected since the functions $\gamma_{\mathbf{W}}(\mathbf{K}_1,\mathbf{K}_4)$ and $\gamma_{\mathbf{W}}(\mathbf{K}_3,\mathbf{K}_2)$ do not overlap in the momentum space in this case. Therefore, we further have

$$H_{\text{eff}}=\sum_{\mathbf{K}}E_{\mathbf{K}}a^\dagger_{\mathbf{K}}a_{\mathbf{K}}$$
$$+\frac{1}{2V}\sum_{\mathbf{K}_1}\sum_{\mathbf{K}_2}\sum_{\mathbf{K}}U(\mathbf{K}_1,\mathbf{K}_2,\mathbf{K}_2+\mathbf{K},\mathbf{K}_1-\mathbf{K})$$
$$\times a^\dagger_{\mathbf{K}_1}a^\dagger_{\mathbf{K}_2}a_{\mathbf{K}_2+\mathbf{K}}a_{\mathbf{K}_1-\mathbf{K}} . \tag{18}$$

Evidently, if the diffraction processes are initially excited by an incident single-atom or low-density atomic beam, the nonlinear terms in (18) can be ignored. In this case, for an incident monochromatic beam with the wave vector $\mathbf{K}_0$, the diffraction pattern is determined by $|\phi_{\mathbf{K}_0}(\mathbf{r})|^2$. In this paper, we consider the nonlinear case where the diffraction is excited by an incident ultracold atomic beam. For simplicity, we assume that the ultracold atomic beam is a plane-wave coherent Bose condensate with wave vector $\mathbf{K}_0$. When the coherent Bose condensate impinges on the crystal surface, a diffraction mode $\phi_{\mathbf{K}_0}(\mathbf{r})$ is excited. Hence the initial state of the system described by Hamiltonian (18) has the form

$$|\Phi_0\rangle=|\alpha_{\mathbf{K}_0}\rangle\otimes\prod_{\mathbf{K}\neq\mathbf{K}_0}|0_{\mathbf{K}}\rangle ,$$

where $|\alpha_{\mathbf{K}_0}\rangle$ is the quantum state of the initially excited diffraction mode and for a coherent condensate $a_{\mathbf{K}_0}|\alpha_{\mathbf{K}_0}\rangle=\alpha_{\mathbf{K}_0}|\alpha_{\mathbf{K}_0}\rangle$. The other diffraction modes are initially in vacuum state $\prod_{\mathbf{K}\neq\mathbf{K}_0}|0_{\mathbf{K}}\rangle$ without excitation. The time evolution of this initial quantum state in the diffraction process is given by the Schrödinger equation

$$i\hbar\frac{\partial|\Phi\rangle}{\partial t}=H_{\text{eff}}|\Phi\rangle . \tag{19}$$

Equation (19) can be solved by employing the standard technique developed by Bogoliubov [21]. The basic idea in this treatment is to evaluate the nonlinear interaction term in Hamiltonian (18) by limiting the sum over the momentum space near the condensate wave vector $\mathbf{K}_0$. Such a treatment is valid if there are a large number of atoms coherently condensing in the condensate. By the above assumption, Hamiltonian (18) can be rewritten as

$$\begin{aligned}
&H_{\text{eff}}=H_c+H_s+H_I ,\\
&H_c=(E_{\mathbf{K}_0}+\rho_0U_0)a^{\dagger}_{\mathbf{K}_0}a_{\mathbf{K}_0}\\
&H_s=\sum_{\mathbf{K}>0}(E_{\mathbf{K}_0-\mathbf{K}}+\rho_0U_0)a^{\dagger}_{\mathbf{K}_0-\mathbf{K}}a_{\mathbf{K}_0-\mathbf{K}}\\
&\qquad+\sum_{\mathbf{K}>0}(E_{\mathbf{K}_0+\mathbf{K}}+\rho_0U_0)a^{\dagger}_{\mathbf{K}_0+\mathbf{K}}a_{\mathbf{K}_0+\mathbf{K}} ,\\
&H_I=\frac{U_0}{2V}(\alpha^{*}_{\mathbf{K}_0})^2\sum_{\mathbf{K}>0}a_{\mathbf{K}_0+\mathbf{K}}a_{\mathbf{K}_0-\mathbf{K}}\\
&\qquad+\frac{U_0}{2V}\alpha^2_{\mathbf{K}_0}\sum_{\mathbf{K}>0}a^{\dagger}_{\mathbf{K}_0+\mathbf{K}}a^{\dagger}_{\mathbf{K}_0-\mathbf{K}} ,
\end{aligned} \tag{20}$$

where H_c, H_s, and H_I are, respectively, the Hamiltonians for the condensate, the sideband diffraction modes, and the interaction of the condensate with the sideband diffraction modes. The parameters $U_0\equiv U(\mathbf{K}_0,\mathbf{K}_0,\mathbf{K}_0,\mathbf{K}_0)$ and $\rho_0=N_{\mathbf{K}_0}/V=\alpha^{*}_{\mathbf{K}_0}\alpha_{\mathbf{K}_0}/V$ are the atomic densities in the diffraction mode initially excited by the coherent condensate. Hamiltonians (20) have similar forms to that of optical multiwave mixing or parametric interaction in quantum optics. By analogy to quantum optics, we conclude that Hamiltonians (20) describe the multiwave mixing of atomic waves in the surface scattering. In terms of Eqs. (19) and (20), at time τ, the quantum state for the diffraction modes of the crystal surface has the form in the interaction picture

$$\begin{aligned}
&|\Phi(\tau)\rangle=S|\Phi_0\rangle=|\alpha_{\mathbf{K}_0}\rangle\otimes|\mathbf{O}_s\rangle ,\\
&|\mathbf{O}_s\rangle=S\prod_{\mathbf{K}\neq\mathbf{K}_0}|0_{\mathbf{K}}\rangle ,\\
&S=\prod_{\mathbf{K}>0}\exp(\xi^{*}_{\mathbf{K}}a_{\mathbf{K}_0+\mathbf{K}}a_{\mathbf{K}_0-\mathbf{K}}-\xi_{\mathbf{K}}a^{\dagger}_{\mathbf{K}_0+\mathbf{K}}a^{\dagger}_{\mathbf{K}_0-\mathbf{K}}) ,\\
&\xi_K=\frac{\rho_0|U_0|}{2\Delta E_{\mathbf{K}}}\sin\left[\frac{\Delta E_{\mathbf{K}}}{\hbar}\tau\right]\\
&\qquad\times\exp\left[i\left[2\theta_0+\frac{\Delta E_{\mathbf{K}}}{\hbar}\tau+3\pi/2\right]\right] ,\\
&\Delta E_{\mathbf{K}}=(E_{\mathbf{K}_0+\mathbf{K}}+E_{\mathbf{K}_0-\mathbf{K}})/2-E_{\mathbf{K}_0} ,
\end{aligned} \tag{21}$$

where τ is the interaction time of the condensate with the crystal surface, θ_0 the initial phase of the condensate, and $\Delta E_{\mathbf{K}}$ the averaged energy of the sideband diffraction mode. From (21), we see that the quantum state of the diffraction mode initially excited by the coherent condensate remains unchanged in the time evolution. The result is due to the assumption of a large number of atoms in this mode, which is equivalent to the nondepletion approximation in the multiwave mixing. The vacuum state of the initially unexcited diffraction modes evolves into the well-known multimode squeezed vacuum state [22]. Such an evolution is due to the nonlinear multiwave mixing in the diffraction induced by the phonon exchanges between atoms. This case is similar to the generation of squeezed state of light in the nonlinear optical parametric interaction and four-wave mixing where the nonlinear optical interaction results in the pairing of conjugate photons [23]. In this paper, the bosonic atoms take the role of the photons in the conventional quantum optics and the phonon exchanges produce an attractive interaction which leads to the pairing of correlated atoms. In this sense, the phonon-exchange-induced nonlinearity for bosonic atoms plays a similar role to optical nonlinearity for photons in quantum optics. We can further show the quantum pair correlation by defining the transformations

$$\begin{aligned}
&b_{\mathbf{K}}=S^{\dagger}a_{\mathbf{K}_0+\mathbf{K}}S=\mu_{\mathbf{K}}a_{\mathbf{K}_0+\mathbf{K}}-\nu_{\mathbf{K}}a^{\dagger}_{\mathbf{K}_0-\mathbf{K}} ,\\
&b_{-\mathbf{K}}=S^{\dagger}a_{\mathbf{K}_0-\mathbf{K}}S=\mu_{\mathbf{K}}a_{\mathbf{K}_0-\mathbf{K}}-\nu_{\mathbf{K}}a^{\dagger}_{\mathbf{K}_0+\mathbf{K}} ,\\
&\mu_{\mathbf{K}}=\cosh\left[\frac{\rho_0|U_0|}{2\Delta E_{\mathbf{K}}}\sin\left[\frac{\Delta E_{\mathbf{K}}}{\hbar}\tau\right]\right] ,\\
&\nu_{\mathbf{K}}=\sinh\left[\frac{\rho_0|U_0|}{2\Delta E_{\mathbf{K}}}\sin\left[\frac{\Delta E_{\mathbf{K}}}{\hbar}\tau\right]\right]\\
&\qquad\times\exp\left[i\left[2\theta_0+\frac{\Delta E_{\mathbf{K}}}{\hbar}\tau+3\pi/2\right]\right] .
\end{aligned} \tag{22}$$

Equation (22) is the well-known Bogoliubov transformation which shows that the annihilation (creation) of an

atom in the sideband mode $\mathbf{K}_0+\mathbf{K}$ is always accompanied by the creation (annihilation) of an atom in the conjugate sideband mode $\mathbf{K}_0-\mathbf{K}$. Such an annihilation and creation of atoms in conjugate mode pairs results in the quantum pair correlation in the surface diffraction, which is due to the nonlinear excitation of the atoms in the condensate into the sideband diffraction modes in the atomic momentum space by multiwave mixing induced by phonon exchanges. To study the effect of the quantum pair correlation on the surface scattering, we can compare the diffraction pattern of the atomic beam in the case of a weak phonon-induced nonlinearity and a strong phonon-induced nonlinearity. For a weak nonlinearity, the quantum state of the sideband diffraction modes around the condensate keeps unchanged except a time-dependent phase factor. In this case, we have the averaged density distribution of the atomic beam

$$\rho(\mathbf{r})=\langle\Phi(\tau)|\psi^{\dagger}(\mathbf{r})\psi(\mathbf{r})|\Phi(\tau)\rangle=|\alpha_{\mathbf{K}_0}|^2|\phi_{\mathbf{K}_0}(\mathbf{r})|^2 . \quad (23)$$

For a strong phonon-induced nonlinearity, we have

$$\rho(\mathbf{r})=\langle\Phi(\tau)|\psi^{\dagger}(\mathbf{r})\psi(\mathbf{r})|\Phi(\tau)\rangle$$
$$=|\alpha_{\mathbf{K}_0}|^2|\phi_{\mathbf{K}_0}(\mathbf{r})|^2+\sum_{\mathbf{K}>0}N_{\mathbf{K}}|\phi_{\mathbf{K}_0-\mathbf{K}}(\mathbf{r})|^2$$
$$+\sum_{\mathbf{K}>0}N_{\mathbf{K}}|\phi_{\mathbf{K}_0+\mathbf{K}}(\mathbf{r})|^2 , \quad (24)$$

where the atomic number excited into the sideband diffraction modes from the condensate is given by $N_{\mathbf{K}}\equiv|\nu_{\mathbf{K}}|^2$. In Fig. 3, we show the dependence of the atomic number in the excited sideband diffraction modes on the nonlinearity and the sideband averaged energy $\Delta E_{\mathbf{K}}$. Evidently the stronger the atomic nonlinearity, the larger the excited atomic number in the sideband diffraction modes. The atomic nonlinearity depends on two important factors: the atomic density of the incident condensate ρ_0 and the coefficient U_0, which is determined by the strength of the phonon-induced potential. Hence we conclude that there is an evident difference between the diffraction patterns of the ultacold atomic beam and that of the low-density atomic beam. Such a difference can be employed to detect the characteristics of the surface sound waves, which includes the complete information of the motion of the atoms in the surface lattice. On the other hand, compared to the research on generation of nonclassical light in quantum optics, the results presented in this paper show that the crystalline-surface atomic grating can change the properties of quantum statistics of the incident atomic beam and is useful in generating atomic beams with different quantum states.

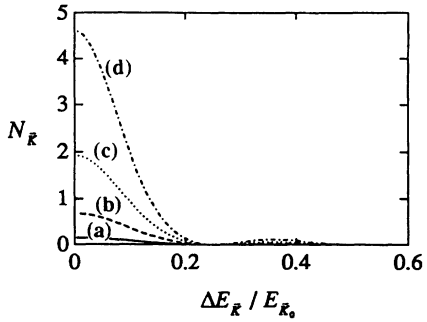

FIG. 3. The dependence of the atomic number $N_{\mathbf{K}}$ in the sideband diffraction modes on the nonlinearity $\rho_0|U_0|$ and averaged sideband energy $\Delta E_{\mathbf{K}}$ for (a) $\rho_0U_0=0.06E_{\mathbf{K}_0}$, (b) $\rho_0U_0=0.12E_{\mathbf{K}_0}$, (c) $\rho_0U_0=0.18E_{\mathbf{K}_0}$, and (d) $\rho_0U_0=0.24E_{\mathbf{K}_0}$. The interaction time is chosen as $\tau=4\pi\hbar/E_{\mathbf{K}_0}$ in all cases.

V. CONCLUSIONS

In this paper, we develop a general quantum-field-theoretic description for the surface scattering of atoms. The many-body quantum collective effect in the surface scattering due to surface phonon exchanges is analyzed. We show that the phonon-exchange-induced atomic nonlinearity can result in the multiwave mixing of diffracted atomic waves by the surface. Such a multiwave mixing for atoms is analogous to the multiwave mixing for photons in nonlinear optics. By the nonlinear multiwave mixing, the atoms in the incident beam can be paired to form correlated atomic pairs in the momentum space. Such correlated atomic pairs have similar behaviors to the correlated photon pairs in squeezed light.

Experimental observation for the atomic quantum pair correlation in the diffraction of ultracold atoms by a crystalline solid surface requires a strict condition. A mechanically clean surface is basically important to avoid the atom sticking on the surface and to achieve a high reflection coefficient. In addition, a low surface temperature should be satisfied to avoid the incoherent scattering due to phonon thermal fluctuations. An ideal surface, which is made available by current low-temperature techniques, is the liquid-He coating surface. On the other hand, an ideal ultracold atomic source composed of Bose condensate is required for such a diffraction experiment. For the current techniques, this is not yet available. However, the results presented in this paper are valuable for pedagogical reasons, as they show another source of atomic nonlinearity besides the photon-induced atomic nonlinearity [11–15]. Specially the quantum-field theory provides a simple way to study surface scattering with thermal lattice vibrations considered. In addition, the general formalism developed here for the surface scattering of ultracold atomic beams also provides possible applications to other topics in studying the many-body collective effect of ultracold atomic ensembles.

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[1] W. A. Steele, Surf. Sci. **38**, 1 (1973).
[2] U. Garibaldi *et al.*, Surf. Sci. **38**, 269 (1973).
[3] R. I. Masel, R. P. Merrill, and W. H. Miller, Surf. Sci. **46**, 681 (1974).
[4] S. Chu, J. E. Bjorkholm, A. Ashkin, and A. Cable, Phys. Rev. Lett. **57**, 314 (1986).
[5] C. Monroe, W. Swann, H. Robinson, and C. Wieman, Phys. Rev. Lett. **65**, 1571 (1990).
[6] P. Lett, P. Jessen, W. Phillips, S. Rolston, C. Westbrook, and P. Gould, Phys. Rev. Lett. **67**, 2139 (1991).
[7] J. M. Doyle, J. C. Sandberg, I. A. Yu, C.L. Cesar, D. Kleppner, and T. J. Greytak, Phys. Rev. Lett. **67**, 603 (1991).
[8] A. Gallagher and D. E. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).
[9] See Appl. Phys. B **54** (5) (1992), special issue on atom optics, edited by J. Mlynek, V. Balykin, and P. Meystre.
[10] Weiping Zhang and D. F. Walls, Quantum Opt. **5**, 9 (1993).
[11] Weiping Zhang, Phys. Lett. A **176**, 225 (1993).
[12] G. Lenz, P. Meystre, and E. M. Wright, Phys. Rev. Lett. **71**, 3271 (1993).
[13] Weiping Zhang, D. F. Walls, and B. C. Sanders, Phys. Rev. Lett. **72**, 60 (1994).
[14] Weiping Zhang and D.F. Walls, Phys. Rev. A **49**, 3799 (1994).
[15] Weiping Zhang and D. F. Walls, in *Quantum Optics VI*, edited by J. D. Harvey and D. F. Walls (Springer, Berlin, in press).
[16] R. G. Steg and P. G. Klemens, Phys. Rev. Lett. **24**, 381 (1970).
[17] A. A. Maradudin and D. L. Mills, Phys. Rev. **173**, 881 (1968).
[18] P. J. King and F. W. Sheard, Proc. R. Soc. London Ser. A **320**, 175 (1970).
[19] H. Haken, *Quantum Field Theory of Solids* (North-Holland, Amsterdam, 1976).
[20] J. E. Lennard-Jones and A. F. Devonshire, Proc. R. Soc. London Ser. A **156**, 6 (1936).
[21] N. N. Bogoliubov, J. Phys. (Moscow) **11**, 23 (1947).
[22] C. M. Caves, Phys. Rev. D **26**, 1817 (1982).
[23] H. P. Yuen, Phys. Rev. A **13**, 2226 (1976); R. E. Slusher *et al.*, Phys. Rev. Lett. **55**, 2409 (1985).

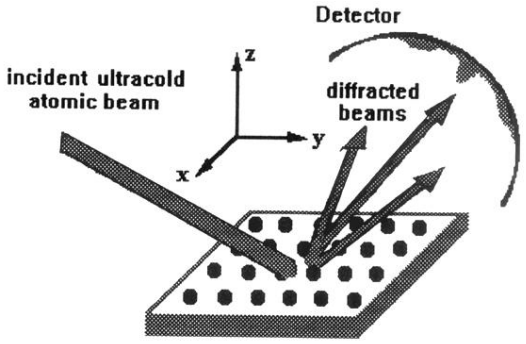

FIG. 1. Schematic diagram for the diffraction of an ultracold atomic beam by a crystalline solid surface atomic grating.

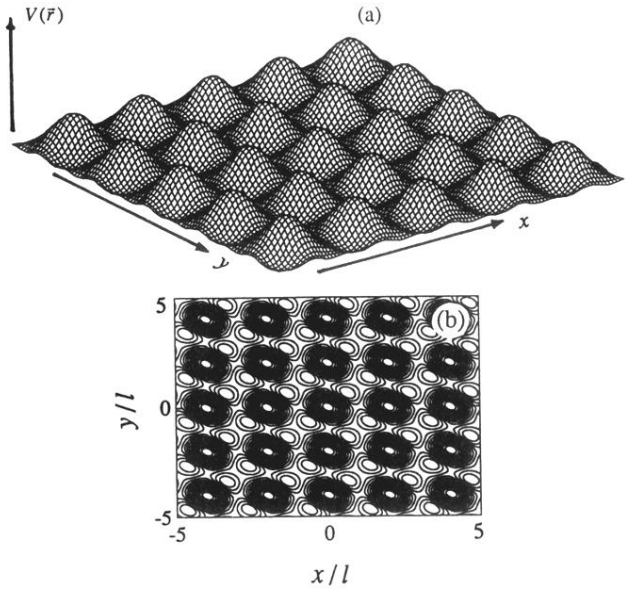

FIG. 2. (a) The surface potential $V(\mathbf{r})$ at $z = 2$ Å and (b) the equipotential curves in the x-y plane. The calculation is performed in terms of expressions (10a) and (11).