Quantum Fluid Dynamics Approach to Time-Dependent electronic State Problems in Surface Systems

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Quantum Fluid Dynamics Approach to Time-Dependent electronic State Problems in Surface Systems

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Dynamical properties associated with the electron motion at sufaces are discussed by solving the time-dependent Schrödinger equation based on the quantum fluid dynamics(QFD) scheme. The classical description for the quantum system provides us clear physical picture such as the eletron tunneling, migration, effects of external field. Two dimensional model studies of the electron current are presented at W(001) surface.

KEYWORDS: QFD, DFT, surfaces, time-dependent electronic structures, electronic state excitation

1. Introduction

The recent development in nanotechnology at surfaces and clusters requires us theoretical analyses of dynamical properties in electronic states associated with the electron transport problems, effects of the external fields, tunneling microscopies, and excited states problems. It is of great importance to develop calculation methods to solve the time-dependent Schrödinger equation in the atomic scale. Numerical treatment is the most effective in this case. It is well known[1)-3)] that the single particle Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t) = [-\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r},t)]\psi(\mathbf{r},t) \quad (1)$$

is transformed into a pair of fluid dynamical equations, the quantum fluid dynamics(QFD), by substitution of the polar form of the wave function, $\psi = \sqrt{\rho}\, e^{iS/\hbar}$, as

$$\frac{\partial\rho}{\partial t} + \nabla\cdot\mathbf{j} = 0 \quad (2)$$

$$m(\frac{\partial\mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v}) = -\nabla(V + V_q) \quad (3)$$

$$\text{where} \quad \mathbf{v} \equiv \nabla S/m \quad (4)$$

$$\mathbf{j} \equiv \rho\mathbf{v} \quad (5)$$

The quantum potential V_q is a functional of the electron density $\rho(\mathbf{r},t)$.

$$V_q(\mathbf{r},t) = \frac{\hbar^2}{4m}\frac{\nabla^2\sqrt{\rho}}{\sqrt{\rho}} \quad (6)$$

Without V_q, a pair of equations (2) and (3) just correspond to the solution of the Hamiltom-Jacobi type classical particle motion. The role of the quantum potential V_q is the main subject in this scheme. Unlike the classical fluid, ρ dependence of the pressure term is complicated enough to make the wave packet tunnel through the potential barrier.

In order to study the role of V_q, one dimensional model calculation is illustrated in Figs.1 to 4. The electron tunneling through the potential barrier

$$V(\mathbf{r}) = \frac{u_0}{(\cosh ax)^2} \quad (7)$$

is calculated using equations (2) and (3).

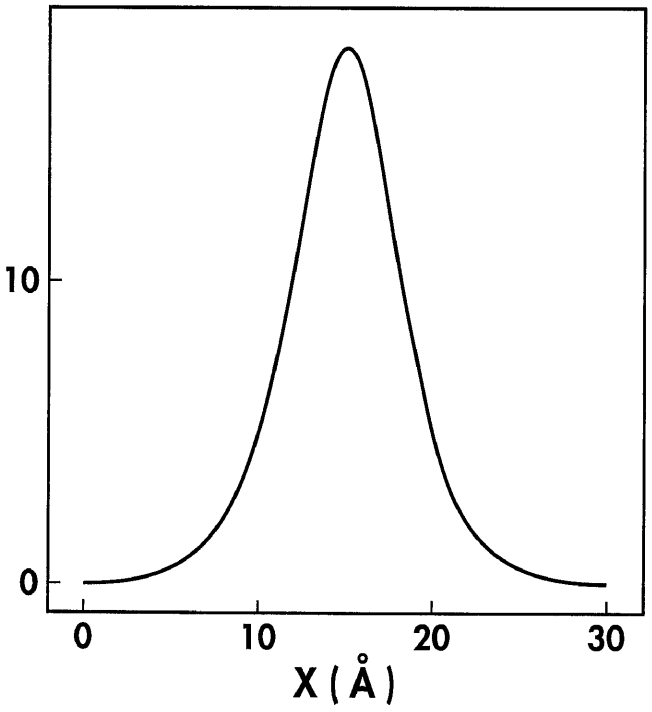

Fig.1 External potential V in Eq.(1). Maximum of the peak is 1 eV.

The initial velocity condition of the wave packet corresponds to 0.68 eV which is lower than the potential barrier, u_0 =1 eV. Each number on curves represents the time step in the unit of $\Delta\ t = 0.5fs$. The time evolution of ρ and velocity v in Figs.2 and 3 provide us information how electron tunnels through the potential

barrier.

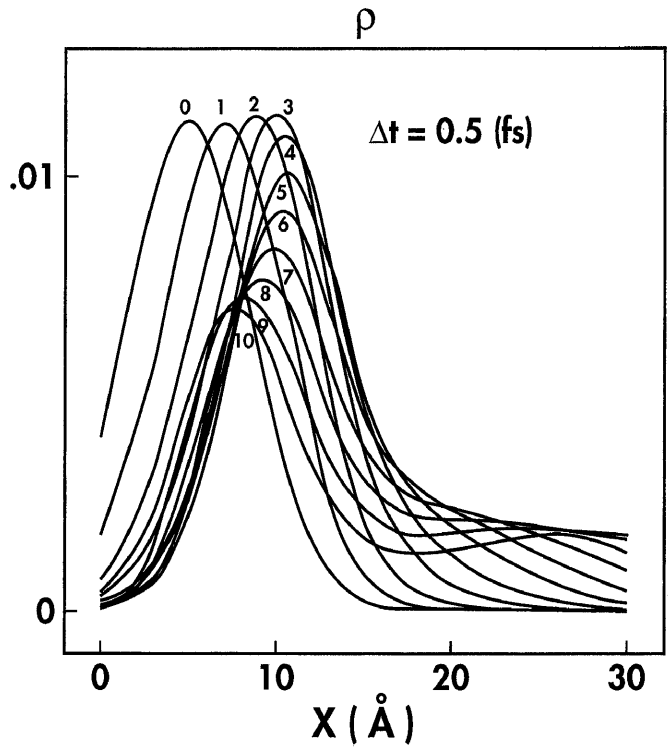

Fig.2 Charge density ρ in Eq.(2). Maximum of the peak is 1 eV.

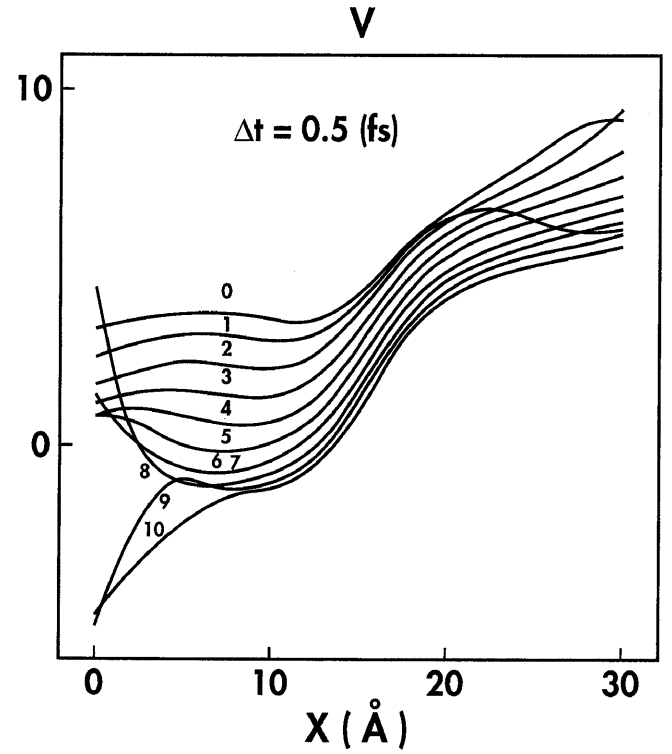

Fig.3 Wave packet velocity v in Eq.(3). v corresponds to 0.67 eV at t=0.

The most striking feature is the change of V_q. It draws the tail of charge density towards the barrier side strongly in the initial stage, which is considered to be a main mechanism of the electron tunneling.

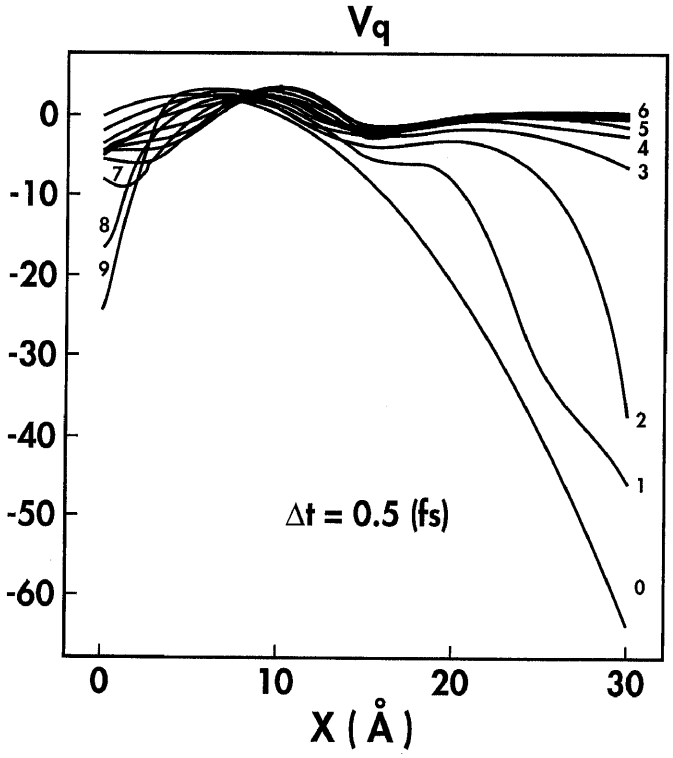

Fig.4 V_q in Eq.(5).

2. Many electron system

2.1. Theoretical model

In many electron case, the basic principle in the density functional theory(DFT) for the time-dependent system was explicitly proved in the form as

$$\frac{\partial \mathbf{j}(\mathbf{r},t)}{\partial t} = \mathcal{P}[\rho(\mathbf{r},t)] \tag{8}$$

where $\mathcal{P}[\rho(\mathbf{r},t)]$ is a unique functional of $\rho(\mathbf{r},t)$[4]. There is no explicit functional form being feasible for the computation of realistic sysytems. The explicit functional form of $\mathcal{P}$, however, is represented in terms of the single particle density matrix $P_1(\mathbf{r}_1,\mathbf{r}_2,t)$, which allows us to deal with the system as a natural extension of the DFT[5]. The time-dependent Schrödinger equation for the many electron case is transformed into

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}P_1(\mathbf{r}_1,\mathbf{r}_2,t) &= -\frac{\hbar^2}{2m}(\nabla_1^2-\nabla_2^2)P_1(\mathbf{r}_1,\mathbf{r}_2,t) \\ &+ W(\mathbf{r}_1,\mathbf{r}_2,t) \end{aligned} \tag{9}$$

where $W(\mathbf{r}_1,\mathbf{r}_2,t)$ is the sum of Coulomb, exchange-correlation, and external potential interactions given by the two particle density matrix $P_2(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_1,\mathbf{r}_2,t)$. Transforming $P_1(\mathbf{r}_1,\mathbf{r}_2,t)$ to the polar form similar to the single electron case

$$P_1(\mathbf{r}_1,\mathbf{r}_2,t) = \rho(\mathbf{r}_1,\mathbf{r}_2,t)exp[iS(\mathbf{r}_1,\mathbf{r}_2,t)/\hbar] \tag{10}$$

and changing coordinate to $\mathbf{r} \equiv (\mathbf{r}_1+\mathbf{r}_2)/2$, $\mathbf{s} \equiv (\mathbf{r}_2-\mathbf{r}_1)$, we can obtain QFD type equations as the first order approximation of small s limit.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{j} = 0 \tag{11}$$

$$\mathbf{j} \equiv \rho(\mathbf{r})\mathbf{v} \tag{12}$$

$$\rho(\mathbf{r}) = \lim_{\mathbf{s}\to 0} \rho(\mathbf{r}+\mathbf{s}/2, \mathbf{r}-bfs/2, t) \tag{13}$$

$$\mathbf{v}(\mathbf{r}) = \lim_{\mathbf{s}\to 0} \nabla_s S(\mathbf{r}+\mathbf{s}/2, \mathbf{r}-\mathbf{s}/2, t) \tag{14}$$

$$\rho \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v} = -\rho\nabla V(\mathbf{r},t) - \mathbf{F}_q(\mathbf{r},t) \tag{15}$$

$$\mathbf{F}_q(\mathbf{r},t) = \nabla_r \left[\int \frac{P_2(\mathbf{r},\mathbf{r}';\mathbf{r},\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}' + 2\mathbf{T} \right] \tag{16}$$

$\mathbf{T}$ is the kinetic energy tensor.

$$T_{ij} \equiv -\frac{\hbar^2}{2m} \lim_{\mathbf{s}\to 0} \frac{\partial^2 \rho(\mathbf{r}+\mathbf{s}/2, \mathbf{r}-\mathbf{s}/2, t)}{\partial s_i \partial s_j} \tag{17}$$

2.2. Electric current at surface

The approximation for T_{ij} is given by the rotationally symmetrical property as

$$T_{ij}(\mathbf{r},t) = \delta_{ij}\frac{1}{3}T^0(\mathbf{r},t) \tag{18}$$

We can apply Thomas-Fermi-Weizsacker type functional form for T^0 and Hatree-Fock type one for P_2.

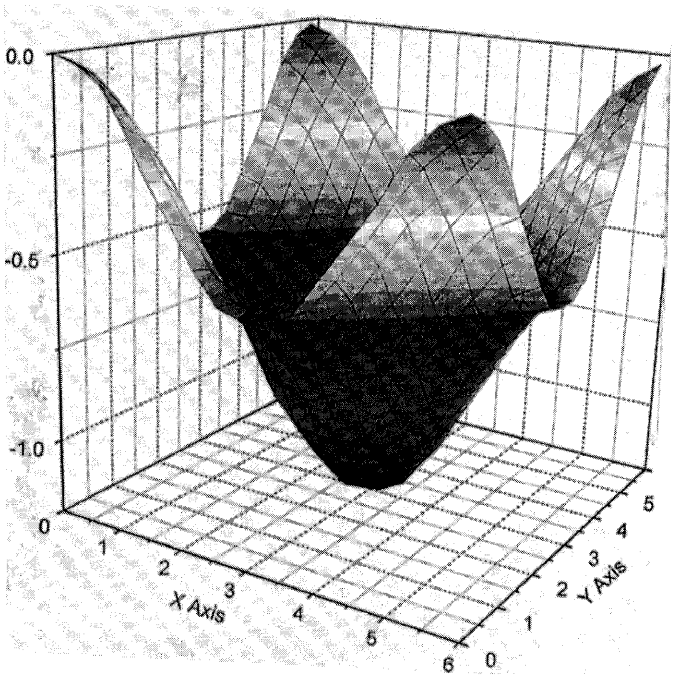

Fig.5 Two dimensional model potential energy given by self-consistently determined W(001) surface at the hight z=5(a.u.)from the surface.

In order to study the time-dependenct electric current of the actual system, two dimensional model calculation has been performed using self-consistently determined W(001) surface potential energy as shown in Fig.5, which is evaluated at z=5 (a.u.) plane above the surface.

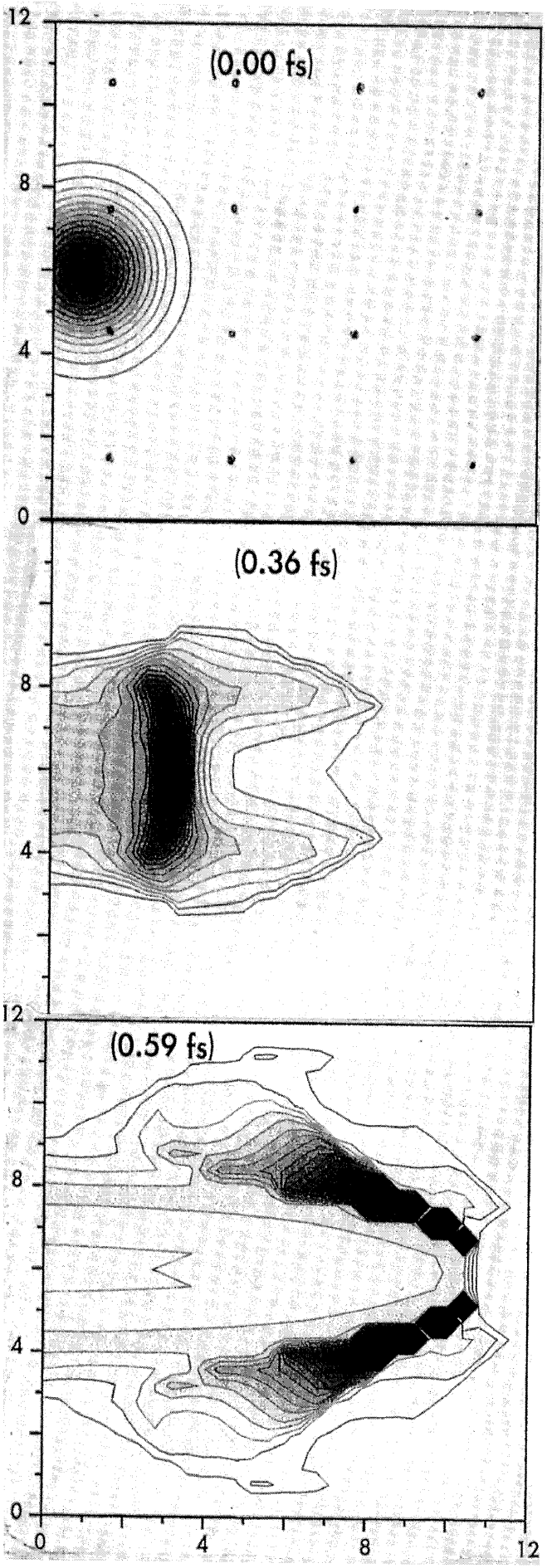

Fig.6 $\rho(\mathbf{r},t)$. Dots are W atoms. Unit length is A.

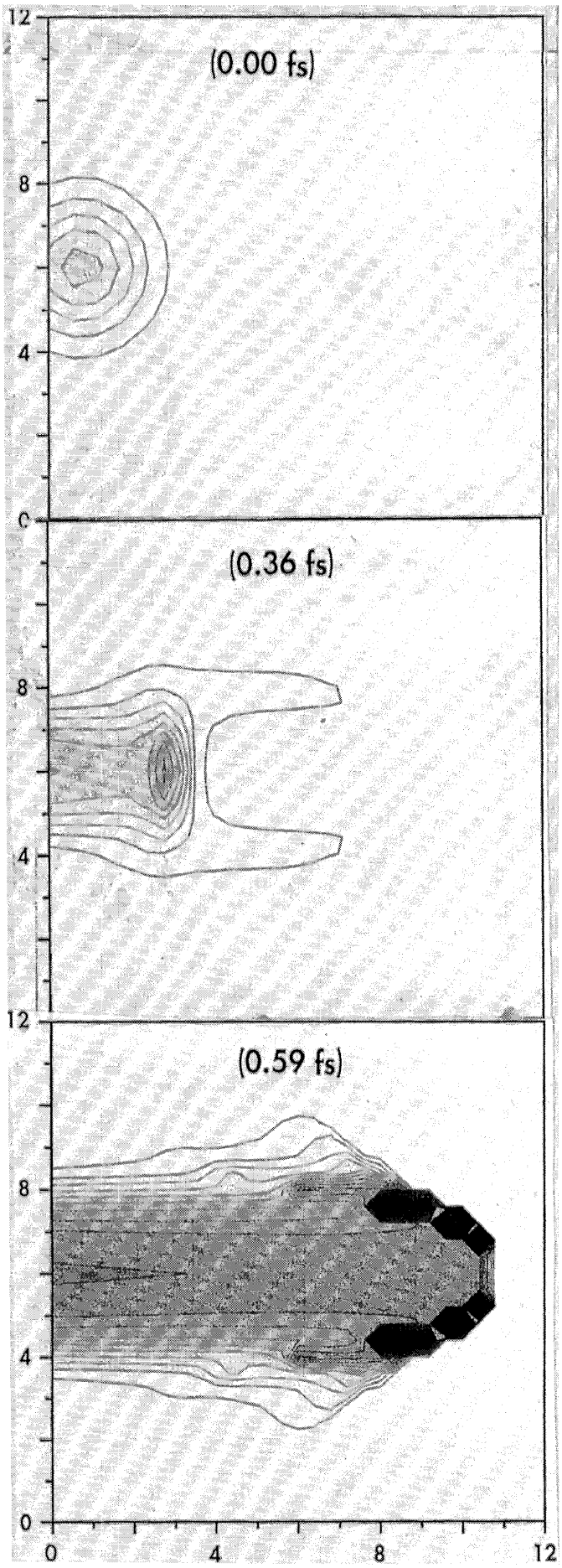

Fig.7 $j_x(\mathbf{r},t)$.

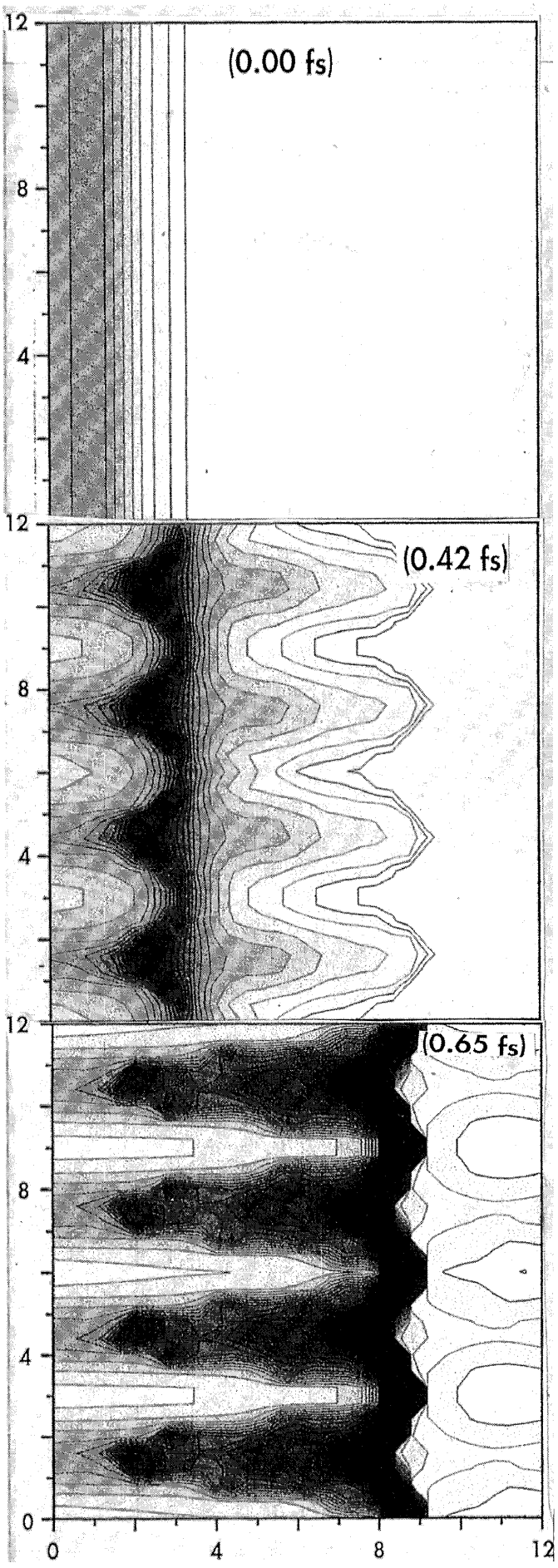

Fig.8 $\rho(\mathbf{r},t)$ for one dimensional wave packet at the initial state.

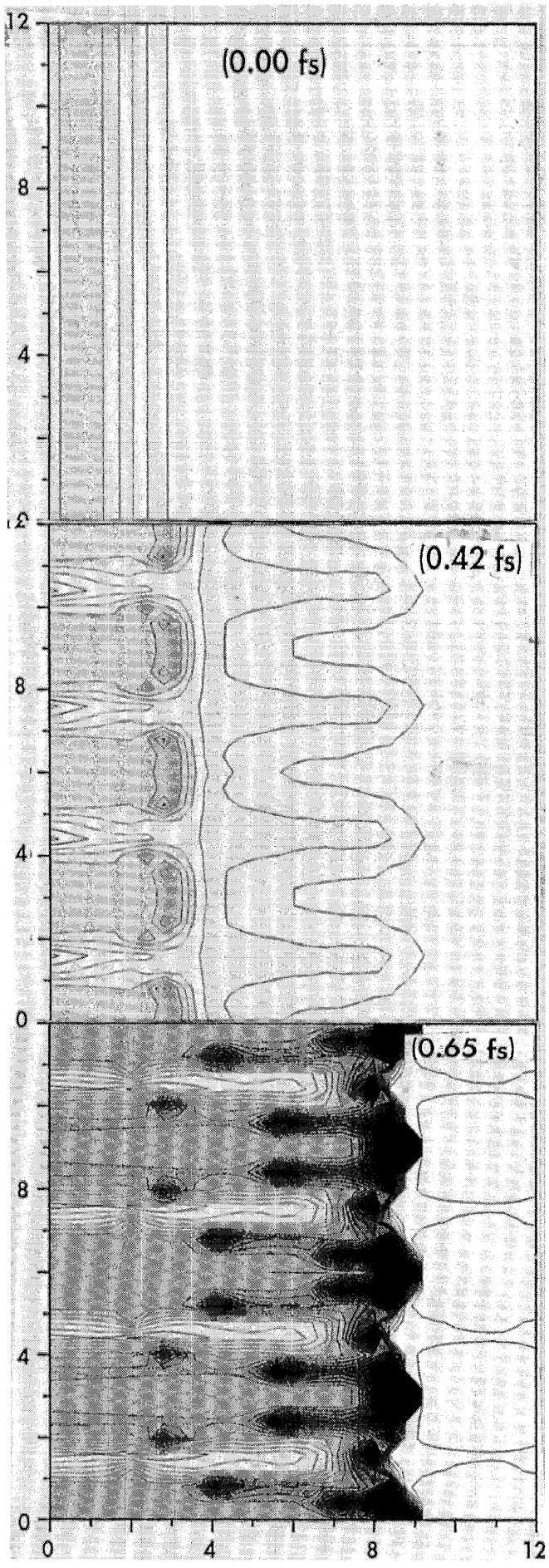

Fig.9 $j_x(\mathbf{r},t)$.

Figs.6 and 7 show the time dependent ρ and J_x, respectively, with the initial velocity corresponding to 4 eV for the wave packet. Periodic dots represent W positions. The change of the one dimensional wave packet at the initial stage case is shown in Figs.8 and 9. It is interesting that the periodicity of the potential field induces several different patterns in J_x.

3. Discussion

Since many electron current calculation is restricted to two dimensional model under the given potential without the additional Coulomb interaction between electrons at surface region, further studies will be necessary by considering three dimensional effects with Coulomb interactions. Experimental studies on electron excitation will help to examine other effective models for T^0. It is very interesting to investigate effects of magnetic field and the geometrical topology for the electron current path within this scheme bacause of inducing the vorticity into the system.

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