Electronwave Amplitude Oscillation during Tunneling due to Interference

H.Nejo*

National Research Institute for Metals, 1-2 Sengen, Tsukuba, 305 Japan
H.Hori
Faculty of Engineering, Yamanashi University, 4-3-11 Takeda, Kohu 400 Japan
D.Baksheyev
Novosibirsk State University, P.O.Box 761, Novosibirsk 630090 Russia

Abstract

By using the electronwave interference in a tunnel barrier, we have measured the conductance oscillation. As a scanning tunneling microscope (STM) tip gives coherent beam and acts as the point source of a spherical wave, we can cause interference using a liquid crystal molecule inside the potential barrier. The adjacent potentials of this molecule act the same as slits in a conventional interference experiment. This result also shows the existence of the electron inside the potential barrier, since we understand that the result reflects of the electron being not observed during tunneling
*Corresponding author:nejoh@nrim.go.jp

1. Introduction

It is impossible to probe an electron inside the potential barrier, since the tunneling is quantum phenomenon and once it is observed it is not the tunneling electron anymore[1] Then the only possible way to detect the electron inside the potential barrier is measuring the interference of the electron wave inside the potential barrier.

For observing the interference it is required that a double-slit is placed in the electron wave [2]. For observing interfrence in the potential barrier, we have prepared the double-slit inside the potential barrier by putting a molecule where the electron wave passes through coherently. Then the observed conductance oscillates as a function of the bias voltage applied between an electron source and a detector.

This result is understood that this oscillation is caused by the phase difference between two paths with slight different potentials during the clock working outside the potential barrier by applying the Aharanov-Bohm effect (scalar potential case) [3,4,5].

The concept of the traversal time is a big theme in quantum mechanics and a lot of theoretical works has been done [6]. Although the definitive description has not been given, BÅEtiker gives Larmor-clock idea and it has been shown experimentally using neutron tunneling through a magnetic film [7].

Our phase shift is caused by the potential difference between two paths so that this difference is equivalent to the time difference which is measured by the clock outside the potential barrier compared to the effect caused by vector potential. This may suggest that the traversal time can be measured by this method. It is not clear if there is a kind of similarity with the Larmor-clock method but they are relative methods.

Although we have reported previously the oscillation of the conductance in a double potential barrier, it does not show any suppression of the conductance around the zero bias voltage [8] Although we attributed this oscillation to the incremental changing of a molecule, we can give a better explanation for the result together with some other experimental results.

2. Experiments and Results

Here we have used as a slit the liquid crystal (LC) molecule 4'-n-heptyl-4-cyanobiphenyl (7CB, where 7 is the number of carbons in the alkyl group), 2CB and 12CB. These compounds show a nematic LC phase near room temperature, and previous STM studies have shown that the molecules form ordered layers on graphite. The platinum (111) surface was polished to a roughness of $1\,\mu$ m, chemically etched in a solution of HNO_3, and HCl (ratio 1:3)[9], rinsed in ultra-pure water and dried in air. A droplet of LC molecules was placed on the platinum surface immediately after drying. A commercial STM (Nanoscope II, Digital instruments) was used, with a tip of etched platinum-iridium wire. The I-V curves were taken at ambient pressure, and the temperature could be controlled within a certain range. The curves were obtained as the bias voltage was scanned

while the tip height was held constant [10]. An STM tip was positioned above an LC molecule on the platinum substrate. This system of STM tip and molecule behaves like an electron source and slits, since the highest Occupied Molecular Orbital (HOMO) and the Lowest Unoccupied Molecular Orbital (LUMO) locate at the end of the molecule and those are favorable for electron tunneling.

Figure 1 shows an STM image of 7CB molecule on graphite. Due to the nature of 7CB molecule, the molecule shows zigzag arrangement by facing cyano-group each other. Since the graphite surface is atomically flat, this set up does not give oscillation of conductance. This is equivalent to that a detector is large enough so that the interference is obscured in conventional double-slit experiment.

Figure 2 shows the differential conductance as a function of the bias voltage. The oscillation of the conductance with periodicity of about 0.2 V is clearly seen. Since the Pt surface is not atomically flat, it is difficult to get molecular resolution image. The attempt was made so as the conductance oscillation is gotten whereas the tip position differs from time to time.

For observing interference by two wavefunctions, it is necessary that two wave-packet should arrive at the detector within the coherence time. The coherence time t is defined as the monochromaticity of the wave packet
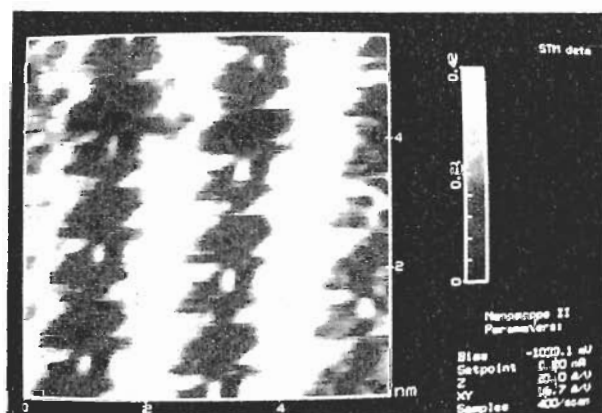

Fig.2 The di/dv-V curve of 2CB, 7CB and 12CB on Pt(111). The periodic peaks separation corresponds to the 2π phase. The sample temperature is 21 ℃. The bias voltage is -1V and the set point current is 0.2 nA. The bias modulation is 50 mV and the sample period is 250 micro seconds. All the parameters for getting the di/dv curves in this paper are the same. (Inset (a)) The molecular surface potential image. The nitrogen atom and the center of the biphenyl group have negative potential, hydrogen atoms and carbon atoms of the biphenyl group have positive potential. A numerical potential value (EP) versus color is green, $EP \leq -0.65$(eV); blue,-$0.65 < E \leq -0.39$; light blue, $-0.39 < EP \leq -0.13$; yellow, $-0.13 < EP \leq 0.13$; violet, $0.13 < EP \leq 0.39$; red, $0.39 \leq EP$. (Inset (b)) Molecular orbital density $|\phi|^2 = 8.1$ (bohr radius) of the one energy level above LUMO. (Inset (c)) That of LUMO, $|\phi|^2 = 8.1$ (bohr radius).

Fig.1 STM image of 7CB molecule on a graphite surface. 6nmx6nm. Bias voltage=-1V (sample negative). Setpoint current=0.2nA. (Due to the nature of 7CB molecule, the molecule shows zigzag arrangement by facing cyano-group each other. Note that the conductance oscillation is not gotten by this set up, instead it is gotten on Pt surface.
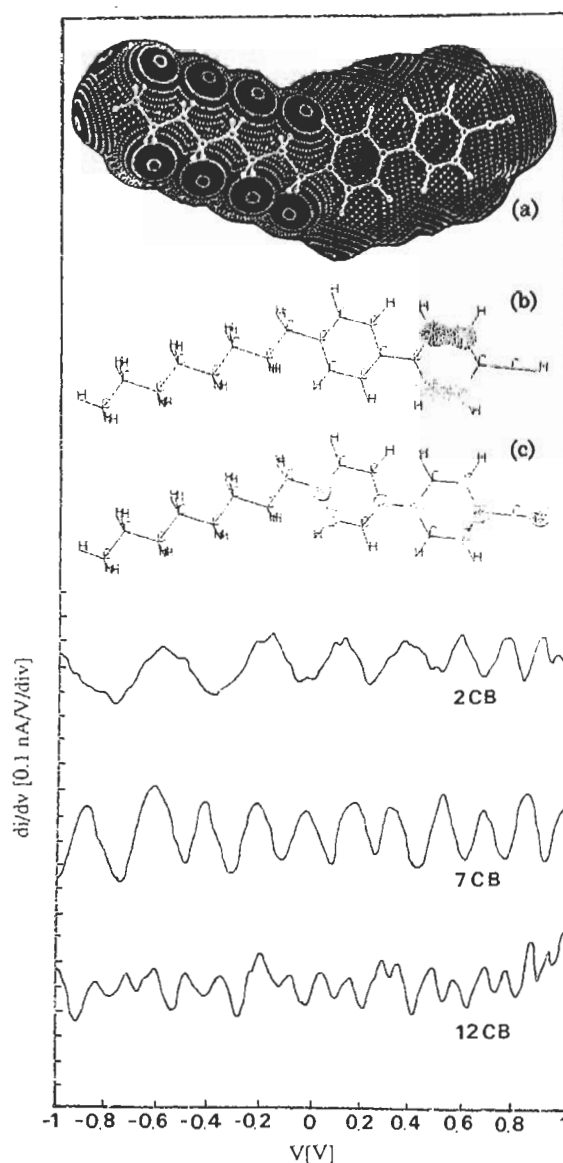
like t=h/Δ E, where Δ E is the energy dispersion determined by the Fermi distribution function [2]. The t is calculated to 2.5 x 10^{-14} s at the room temperature. The arrival time difference between two-wave packets is determined experimentally to around 1.7x10^{-14}s so that the two wave packets arrive within the coherence time.

Fig. 2 shows the bias voltage (potential energy) dependence on the phase difference; the larger the bias voltage, the larger the phase difference. This shows that the phase difference is caused by the effect of the potential surfaces $V_{c,d}$ as below; the potential surface (inset(a) of Fig. 2) reflect the total electron density as described in the next section. But as shown, the electron density becomes more nonuniform at one energy level above the LUMO (Inset (b) in Fig.2) than at the LUMO (Inset(c)). This leads the more potential surface difference at higher energy. Then when the bias voltage rises, the electronwave feels the more different potentials $V_{c,d}$. Accordingly, the phase difference becomes smaller at larger bias voltage.

As shown in Fig.2, the shorter the molecular length (2CB<7CB<12CB), the larger the period of oscillation. Namely, it takes more voltage change to return to the same phase. This is interpreted using the potential idea. As shown in the inset(a) of Fig.2, the cyano-group has more negative potential than the biphenyl-group and alkyl-group. The shorter the alkyl-chain, the less the potential difference between cyano-group and others. For the electronwave which reaches the cyano-group and others, the path depends on the molecular length. The phase difference Φ_c-Φ_d where c corresponds to the cyano-group and d corresponds to others, becomes smaller when the molecular length becomes shorter. When potential difference is smaller, the bias voltage should be larger in order to return to the same phase.

For confirming the effect of the potential surface on the phase, we have varied the sample temperature. When the temperature of the sample decreases, the period of the oscillation increases (Fig.3)[11].

Since the 7CB molecule shows a nematic nature, the molecule orders more at lower temperatures by a zigzag of facing cyano-groups. At the nematic phase, although the molecular axes point almost in the same direction, they deviates from molecule to
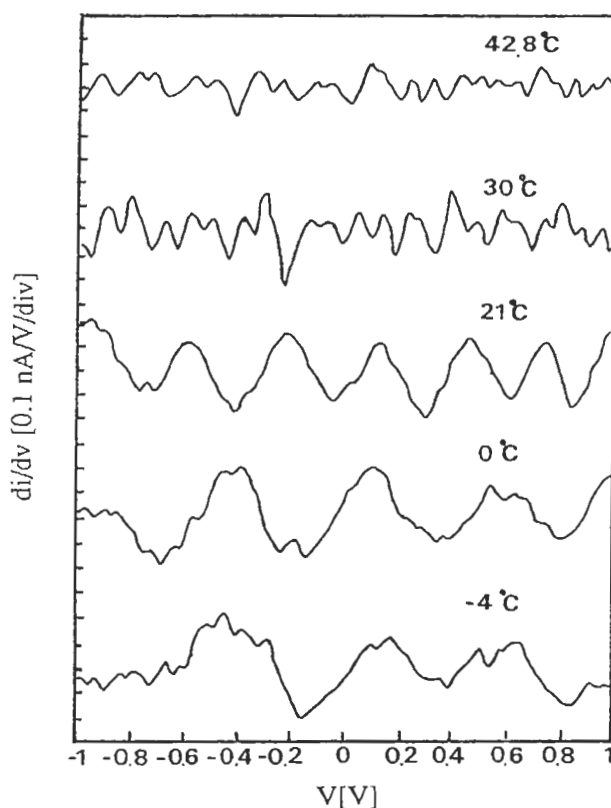

Fig.3 The temperature dependence of the di/dv curve of 7CB on a Pt(111) substrate. di/dv curves of 7CB on Pt were taken at -4, 0, 21, 30 and 42.8℃. They show that the lower the temperature, the wider the period of oscillation. The bias voltage is -1V and set point current 0.2 nA.

molecule, and also this phase does not show a layered structure so that the cyano-groups cannot face each other. High-resolution STM images of the molecule show that there is an orbital hybridization between the N atom of the cyanine-group and the C atom in the biphenyl-group of the neighboring molecule [12].As the surface potential image shows (inset(a) in Fig.2), the potential of the N atom is most negative and that of the C atom of the biphenyl-group is most positive. By ordering, and the consequent orbital hybridization, these potential differences become averaged. These averaged potentials make the phase difference smaller. Then, to return to the same phase, the bias voltage should change more and this is well seen at the lower temperature.

Next, the sample was irradiated by monochromated IR light and the suppression of the conductance oscillation is observed until the bias voltage reaches to the light energies (Fig.4).
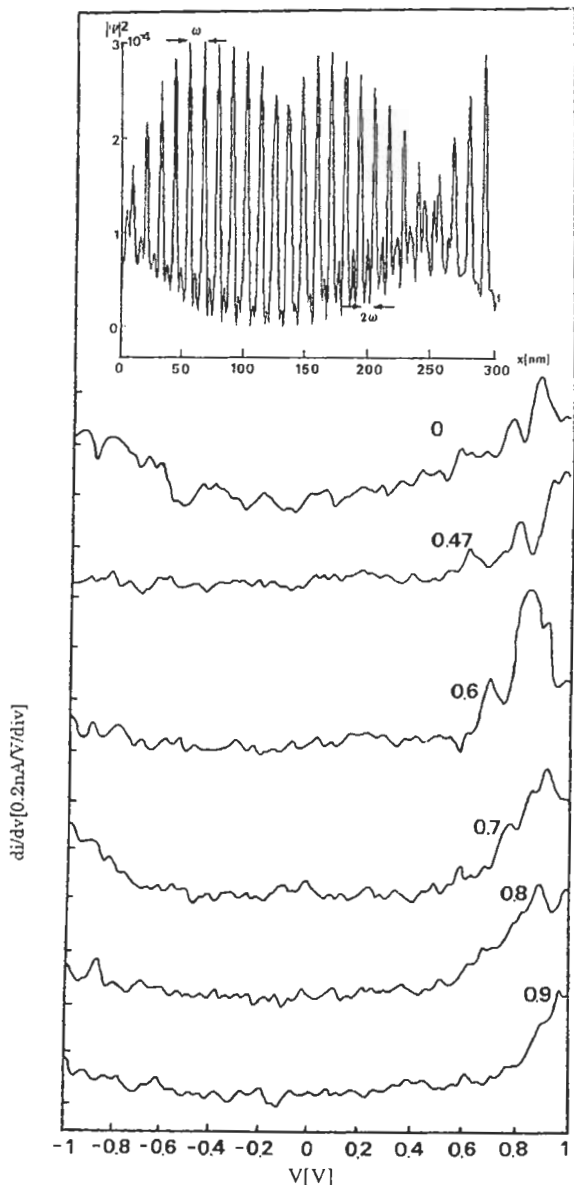
Fig.4 The di/dv curve with and without irradiation by monochromated light with various photon energies from 0.47 to 0.9 eV. It is observed that conductance oscillation effects are suppressed when the magnitude of the applied bias is less than the photon energy. The photon energies for each curve are indicated. (Inset) $|\Psi|^2$ oscillations to the right of the structure. This is calculated by summing the amplitude via the $\hbar\omega$ virtual state and that via the $2\hbar\omega$ virtual state, and squaring.

In Fig.4, the first peak appears at 0.6, 0.69, 0.77, 0.88, and 0.97 eV which corresponds to irradiation of 0.47, 0.6, 0.7, 0.8, and 0.9 eV, respectively. Also, the second peak is observed following the first peak. The intervals between the first and second peaks are 0.19, 0.15, 0.14, and 0.12 eV for $\hbar\omega$ irradiation of 0.47, 0.6,

0.7, and 0.8 eV, respectively. The explanation of disappearance of the oscillation for this set up is given in the next section.

3. Discussions
III-1 Phase differences in the potential barrier

The path integral or amplitude K(a,b) is written as

$$K(a,b) = \exp(-iS[a,b]/\hbar)$$

,where the action S is written as

$$S[a,b] = \int_a^b dt L \qquad (1)$$

where L is the Lagrangian L=T-V, T is the kinetic energy and V is the potential energy[13].

The inset(a) of Fig.2 shows the surface potential image of a liquid crystal molecule based on ab initio calculation[14]. When an electron is emitted from an STM tip, the electronwave can be treated as a spherical wave in the tunnel barrier of the 1nm wide and 0.2nm-distance of different potentials[15]. Then the same wave-plane reaches to the different potential surface of the molecule. For the electronwave, the different potential $\phi(\mathbf{r})$ works differently for shifting the phase:

$$e\phi(\mathbf{r}) = \int \frac{eq(\mathbf{r'})d\mathbf{v'}}{|\mathbf{r}-\mathbf{r'}|} \qquad (2)$$

where e is the elemental change, and q(**r'**) is the effective charge determined by the Mulliken population analysis at **r'**. These different potentials act the same as the slits same in a conventional interference experiments. Namely, when the electronwave reaches the different potential surfaces, it becomes the second source for the new electronwave. Since the potential $V=e\phi$ varies, the action varies and hence these actions bring phase differences when the wave reaches the substrate.

Next, the action is the function of the potential energy and this causes the energy dependence of the action. When an electron is emitted from the point a and reaches the different positions c and d on the same molecule, the action difference is

$$S[a,c]-S[a,d] = \int_a^c dt(T-V_c) - \int_a^d dt(T-V_d) \qquad (3)$$

Since the action difference depends on the potential V_c, V_d, this action difference brings the oscillation of the conductance as a function

of the bias voltage. The potential V_c, V_d, change as the function of the bias voltage is determined by solving the Poisson equation at this boundary condition; $\nabla^2(\phi_c - \phi_d) = -\rho/\varepsilon = -CV/S\varepsilon$ (4) where $e\phi_{c,d} = V_{c,d}$, ζ is the charge density at the tip and the substrate, ε is the dielectric constant, C is the capacitance between the tip and the substrate, V is the bias voltage by assuming the tunnel junction by a capacitance and S is the area of the interest.

The path integral K(a,b) is defined as eq.(1) and when an electron with energy T tunnels through the double-potential barrier from a to b, the traversal time t is associated with it. Here the traversal time is the time which is measured *by the observer outside the potential barrier.*

III-2 Light irradiation effect

The interference under irradiation is different from that caused by passing the different potential surfaces; in the former case, the action $\int dt L$ differences occur due to that the wavefunction passes different virtual states, namely the ground state E and the excited state $E+\hbar\omega$.

The reason for the suppression of the oscillation until the bias voltage reaches the energy of the photon is considered below; the action for the interaction between the electron and the electromagnetic field is given as

$$S_2 = \int dt \int d^3\mathbf{R}[\ \rho\,(\mathbf{R},t)\ \phi\,(\mathbf{R},t)\text{-}\mathbf{j}(\mathbf{R},t)\cdot\mathbf{A}(\mathbf{R},t)]\ (5)$$

where ρ is the charge density, ϕ is the scalar potential, $\mathbf{j}$ is the current density and $\mathbf{A}$ is the vector potential. When the transition element of this action is considered, the transition element of $\mathbf{j}$ and $\mathbf{A}$ is of the order of the small time interval and $\mathbf{j}$ and $\mathbf{A}$ fluctuate on the path all the way[15] so that the two electron waves loose their interference nature. This is why we cannot see conductance oscillation under IR irradiation.

If there are path integrals which pass via virtual states, their contribution as the second order perturbation makes interference. The calculation shows that there are two virtual states which corresponds to $\hbar\omega$ and $2\hbar\omega$ emission at positive polarity but only $\hbar\omega$ absorption at negative polarity [16].

The inset of Fig.4 shows the calculated electronwave amplitude oscillation as a function of distance from the collector when the incident electron energy matches $\hbar\omega$. It

shows the interference between path integrals via the virtual states $\hbar\omega$ and $2\hbar\omega$. This interference can occur as follows; the path integrals passes virtually via the $\hbar\omega$ virtual state and the $2\hbar\omega$ virtual state. Since we *do not make observation* during the path, the actual transition does not occur. Then the time which elapses for the transition is not required. If it is required, it changes the action from transitions to the various virtual states, and the action differences destroy the interference. Then the interference due to two virtual states at positive polarity makes interference possible but not at negative polarity. This is why the peak, which corresponds to interference via the virtual state, appears only at positive polarity [17].

In summary, these results show the existence of the electron inside the potential barrier, since we understand that the result reflects of the electron being not observed during tunneling

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9. The Pt surface is not atomically flat. This gives the situation that the each protrusion acts as the detector small enough to detect the merged electron wave at each position. When the molecule is supported on an atomically flat substrate (HOPG), the conductance oscillation is not observed. This is thought to be that the detector is large enough to detect electron wave as a whole.

10. Since the tip height is kept constant, the oscillation is not caused by the layer thickness but it is caused the effect of path length change as described below.

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