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Nonlinear Dependences in Pulse-Pair-Excited Scanning Tunneling Microscopy

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The mechanism underlying an ultrafast decay of a sample parameter appears that in a time-resolved tunnel current signal obtained by pulse-pair-excited scanning tunneling microscopy (PPX-STM) was theoretically and experimentally investigated. It was shown that the nonlinear dependence of the tunnel current on the sample parameter allows us to observe the decay process in a PPX-STM experiment even when the sample parameter itself does not have a nonlinear response to the light intensity. This result promises the applicability of PPX-STM measurement to a wide variety of samples. On the other hand, such a nonlinear dependence makes the analysis of the obtained data very difficult. Since the apparent decay time in the signal can be different from the actual value, the analysis must be carried out with detailed knowledge of dependence of the tunnel current on the sample parameter. [DOI: 10.1143/JJAP.45.1926]

KEYWORDS: scanning tunneling microscopy, ultrafast mesurement, femtosecond pulsed laser

1. Introduction

In order to satisfy the limitless requirement for miniaturizing and speeding up electronic devices, development of femtosecond–angstrom technology has been proposed to be a helpful solution and is gathering attention. The technology combines ultrashort-pulsed lasers and scanning tunneling microscopy (STM) to realize a measurement technique which has both extremely high temporal and spatial resolutions. Among several setups proposed for realizing such measurements,[1–10,12] pulse-pair-excited STM (PPX-STM) has recently made a significant stride by introducing the delay-time-modulation method,[8–10,12] in which the tunnel gap of an STM apparatus is illuminated by a sequence of ultrashort laser pulse pairs and the variation in tunnel current is measured as a function of the delay time between the two pulses in each pulse pair. As a result, picosecond-order ultrafast response in tunnel current was successfully detected for a low-temperature-grown GaN_α-$As_{1-\alpha}$ sample.

As already pointed out,[9,10] however, the analysis of the time-resolved tunnel current signal obtained by PPX-STM is very complex. This is because the obtained time-resolved tunnel current signal does not exactly correspond to the ultrafast variation in tunnel current caused by the laser illumination. Instead, it represents the variation in the temporarily averaged tunnel current as a function of delay time. The dependence of tunnel current on delay time generally involves complex physical processes. The detailed physical processes that occur in a sample must be understood to extract the ultrafast physical property of the sample from the obtained signal. This paper aims to clarify a basic concept for the analysis of time-resolved tunnel current obtained by PPX-STM, where the nonlinear response of the tunnel current to the light illumination in the sample plays a major role. Finally, the experimentally obtained data is interpreted along with the discussion.

2. Pulse-Pair-Excited STM

First, we describe the typical experimental setup for PPX-STM. A sequence of laser pulse pairs, with a pulse width of 10–100 fs, a wavelength of ~800 nm, a repetition rate of ~100 MHz, a delay time between two paired pulses of 0–500 ps and an averaged intensity of 1–100 mW, is targeted onto a spot with a diameter ~50 µm at the tunnel junction in STM. A weak feedback control is applied for the separation between the STM tip apex and the sample surface to maintain the averaged tunnel current constant at a reference value of 0.1–10 nA with a bias voltage of 10–1000 mV. Since the bandwidth of the STM preamplifier is limited, the tunnel current is temporarily averaged so that the possible tunnel current spikes generated by the laser pulse pair cannot be resolved. The averaged tunnel current is, however, still affected by the laser illumination and is measured as a function of the delay time between the two pulses. Since the variation in tunnel current is generally very small, the signal needs to be processed by a lock-in amplifier to reduce the noise level. In the recently proposed shaken-pulse-pair-excited STM (SPPX-STM) method, the lock-in detection is synchronized with the small periodic modulation of the delay time. This new method enables high-sensitivity measurement without introducing the elongation and shrinkage problem of the STM tip due to the heat generated by the exciting laser pulses even under a high-intensity excitation condition.[8] Although some other techniques have also been proposed, the obtained signal in any PPX-STM method is the temporally averaged tunnel currnt that varies as a function of delay time, $\overline{I}_t(t_d)$.

Next, we consider how the ultrafast response of the ultrasmall region of the sample is reflected in the $\overline{I}_t(t_d)$ signal. When a laser pulse impinges the sample surface, the transient tunnel current $I_t(t;t_d)$ is generated in response to it; i.e., the tunnel current increases or decreases due to the sample excitation brought about by the laser pulse and decays in a time scale from femtoseconds to hundreds of picoseconds resulting in an ultrashort tunnel current pulse. The averaged tunnel current is related to the transient tunnel current as

$$\overline{I}_t(t_d) = f_{\text{rep}} \int_0^{T_{\text{rep}}} I_t(t;t_d)\,dt, \tag{1}$$

where the integral is taken over the repetition period of the sequence of laser pulses, $T_{\text{rep}} = 1/f_{\text{rep}}$. As is discussed in detail below, when the averaged tunnel current shows a delay time dependence, the nonlinear response of the transient current $I_t(t;t_d)$ to the laser intensity plays an important role.

Let us assume that the two pulses in a pulse pair have the same intensity, as a simple case. Then, when t_d is sufficiently long such that the two pulses excite the sample independently, there should be two identical current pulses in $I_t(t;t_d)$, each of which corresponds to its respective laser pulse in the pulse pair. In contrast, when t_d is very short, the pulse pair affects the sample as if they are only one pulse with twofold intensity, causing only one current pulse in $I_t(t;t_d)$. Thus, if $I_t(t;t_d)$ has any nonlinearity on the laser pulse intensity, $\overline{I_t}(t_d)$ takes on different values in these two cases. Moreover, when the delay time is increased from zero to a large value, $\overline{I_t}(t_d)$ varies from the former value to the latter value reflecting the time scale of the ultrafast transition of the excited sample state.

3. Nonlinear Dependence of Tunnel Current on Photointensity

What is the nonlinear dependence of transient tunnel current, $I_t(t;t_d)$, on light intensity? Indeed, the relationship between light intensity and tunnel current is generally very complex. Thus, it seems rather improbable that they are linearly related. We classify such a nonlinear dependence into two types: (1) nonlinear dependences of physical parameters of the samples on light intensity, and (2) nonlinear dependences of tunnel current on the physical parameter of the samples. Here, the physical parameter of the sample can be anything that influences the tunnel current, such as photocarrier intensity, plasmon intensity, electron temperature, electric polarization, and lattice vibration amplitude.

The former case, the nonlinear dependence of a sample parameter on light intensity, is easier to understand. For example, the photocarrier generation in a semiconductor sample often shows a saturable nonlinearity with respect to an intense light illumination. This saturable response is caused by the depletion of originally occupied electronic states and by the occupation of originally empty electronic states due to photoexcitation. Previously reported pump-probe reflectivity measurements show that the absorption of the second pulse can be slightly suppressed ($\Delta R/R = 10^{-6}$–10^{-3}) by this effect. In principle, PPX-STM can be sensitive to such a phenomenon. If the absorption of the second pulse is suppressed, the total amount of photocarrier generation due to illumination of two pulses is also suppressed, which will affect the temporally averaged tunnel current. In reality, however, a deviation in tunnel current due to this effect is impossible to detect. Note that when the intensity of the current pulse due to the second pulse deviates by 10^{-6}–10^{-3}, the ratio of the deviation to the total current, including the static tunnel current and the current pulse due to the first laser pulse, is more than one order of magnitude smaller. The inevitable noise level of the tunnel current detection is usually $\sim 10^{-4}$ even with the help of the lock-in detection technique. Consequently, in order to make use of the type-1 nonlinearity, the nonlinearity must be sufficiently large and the change in the physical parameter must affect the tunnel current very efficiently. Multistep excitation of the sample state might be a good candidate that has such properties, where the probability of excitation is largely emphasized when the second pulse arrives within the lifetime of the midstate.

The second type of nonlinearity seems to appear more commonly, which is the focus of this paper. The tunnel current is known to be affected by the physical parameters of the sample discussed above and its dependence generally has strong nonlinearities. For example, an increase in carrier density in a semiconductor sample due to light illumination varies the amount of surface bandbending. This is known as the photovoltage effect, which can be measured by STM at a high spatial resolution.[13–15] While photocarrier generation rate is almost linearly dependent on light intensity, photovoltage is nonlinearly dependent on carrier density. In particular, photovoltage saturates when the original surface bandbending is fully compensated by an intense illumination. Then, the photovoltage modifies the effective bias voltage at the tunnel junction. Since the current-voltage dependence for a semiconductor sample is also strongly nonlinear, tunnel current depends on photovoltage nonlinearly. Consequently, the dependence of tunnel current on light intensity shows a strong nonlinearity on a semiconductor sample.

4. How Nonlinearity Affects PPX-STM Measurement

Let us consider how such a nonlinear dependence type-2 appears in a PPX-STM measurement. We assume that when one laser pulse with unit intensity and a negligible pulse width impinges onto a sample at time zero, some physical parameter of the sample $n(t)$ shows an exponential decay process as

$$n(t) = n_0(t) = \begin{cases} 0 & t < 0 \\ N\exp(-t/\Delta t) & 0 \leq t \end{cases}. \tag{2}$$

By assuming that the dependence of the physical property on light intensity is linear, the decay process caused by a pulse pair with delay time t_d can be expressed as

$$n(t;t_d) = An_0(t) + Bn_0(t - t_d), \tag{3}$$

where the intensity of the first pulse is A and that of the second pulse is B under the condition $t_d > 0$. Additionally, we assume that tunnel current is a function of this sample parameter, i.e.,

$$I_t(t;t_d) = I_t(n(t;t_d)). \tag{4}$$

In reality, $I_t(t;t_d)$ and $n(t)$ should contain a constant that corresponds to their value in the dark condition. Here, we neglect this steady component and concentrate on the photoinduced component for simplicity. With these assumptions, the temporally averaged tunnel current can be obtained by eq. (1).

4.1 *Polynomial dependence*

At first, we examine the cases in which the tunnel current shows polynomial dependences on the sample parameter, i.e., $I_t(n) = n^k$. The condition $k = 1$ corresponds to the linear dependence of tunnel current on the sample parameter and $k > 1$ corresponds to a diverging nonlinear dependence. In the case of $k = 1$, we obtain

$$\overline{I_t}(t_d) = (A + B) f_{\text{rep}} \int n_0(t)\,dt, \tag{5}$$

which suggests that the temporally averaged tunnel current is always constant without any dependence on delay time. In contrast, $k = 2$ leads to

$$\begin{aligned}\overline{I_t}(t_d) &= (A^2 + B^2) f_{\text{rep}} \int n_0(t)^2\,dt \\ &\quad + 2AB f_{\text{rep}} \int n_0(t) n_0(t - t_d)\,dt.\end{aligned} \tag{6}$$

While the first term again does not depend on delay time, the second term does. However, the dependence is always as an even function; i.e., even when the magnitudes of A and B are different, the plot of the time-resolved tunnel current shows perfect symmetry. In the case of $k = 3$, we obtain

$$\begin{aligned}\overline{I_t}(t_d) &= (A^3 + B^3) f_{\text{rep}} \int n_0(t)^3\,dt \\ &\quad + 3AB f_{\text{rep}} \int n_0(t) n_0(t - t_d) \\ &\quad \times \{A n_0(t) + B n_0(t - t_d)\}\,dt,\end{aligned} \tag{7}$$

where the second term depends on delay time. This time, the dependence is not an even function. Thus, when A and B are different, the plot becomes asymmetric. Likewise, the condition $k > 3$ results in an asymmetric function except for some special cases.

Figure 1 shows the calculated curves for $\overline{I_t}(t_d)$ with $k = 2$–5, $A = 1/4$, $B = 1$ and $\Delta t = 1$. Even with the assumption of an exponential decay process for the parameter $n(t)$, the time-resolved tunnel current signal does not decay as an exponential function. Moreover, the apparent decay constant varies with the nonlinear factor k. In general, the larger the k, the smaller the apparent decay constant. As predicted above, different intensities for the two pulses in the pulse pair results in symmetric and asymmetric curves in the cases of $k = 2$ and $k > 2$, respectively. The result for $k = 1$ is not shown because it is a constant function. In the case of $k > 2$, the apparent time constant is smaller when the stronger pulse reaches the sample first. For comparison, the decay process assumed for $n_0(t)$ is also plotted with a broken line on the same time scale with an arbitrary vertical scale. In this example, the signal intensity decreases when k is increased. This is, however, not a generic law. Substituting different values for A and B, the relative intensity changes markedly.

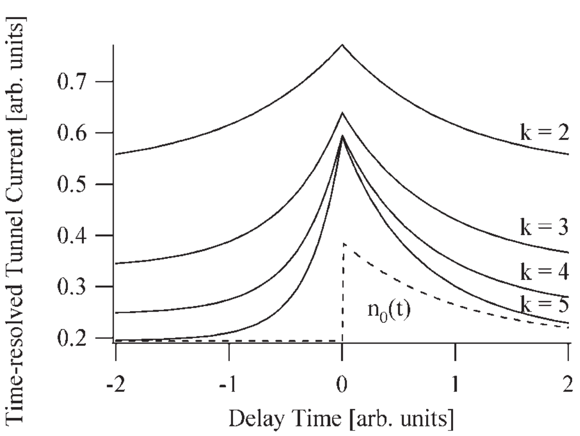

Fig. 1. Time-resolved tunnel current signal expected for polynomial dependences of tunnel current on exponentially decaying sample parameter n: $I_t(t) = n(t)^k$ for $k = 2$–5 (solid lines). The decay process of $n(t)$ is shown with an arbitrary vertical scale as well (broken line).

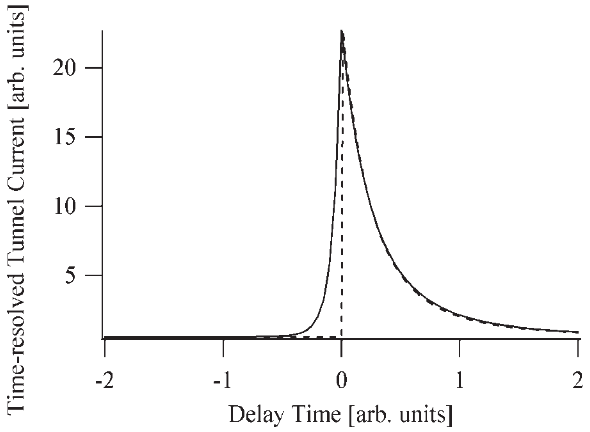

Fig. 2. Time-resolved tunnel current signal expected for exponential dependence of tunnel current on exponentially decaying sample parameter n: $I_t(t) = \exp[n(t)]$ (solid line). The decay process of $\exp[n(t)]$ is shown with an arbitrary vertical scale as well (broken line).

4.2 *Exponential dependence*

What happens when the nonlinearity is even higher than the polynomial dependence? An exponential dependence, $I_t(t) = \exp[n(t)]$, also gives a diverging nonlinearity similar to the polynomial dependences but the nonlinearity becomes much higher when $n \gg 1$. Figure 2 shows the result for $A = 4$, $B = 16$, and $\Delta t = 1$ with a solid line. The broken line corresponds to $I_t(An_0(t))$ with an arbitrary vertical scale. The coincidence of these two curves is not accidental. When the nonlinearity is very high, i.e., $(dI_t/dn)/(I_t/n) \gg 1$ at $n = Bn_0(0)$, the integrated intensity of the function $I_t(Bn_0(t - t_d))$ is dominated by the peak intensity at $t = t_d$ under the condition of $B \gg A$. Introducing another less intense function of $An_0(t)$, the peak intensity of the function $I_t(n)$ becomes $I_t(An_0(t_d) + Bn_0(0))$. When the dependence is exponential, this value is proportional to $I_t(An_0(t_d))$.

As a summary of the diverging nonlinearities, when the nonlinearity is very strong, the time-resolved tunnel current is related to the decay process of the sample parameter in a simple form given by $I_t(An_0(t_d) + Bn_0(0))$. Knowing the function $I_t(n)$, we can directly obtain the decay process of the sample parameter $n(t)$ from the experimental result. On the other hand, if the dependence is second-order polynomial, the result is an autocorrelation function of $n(t)$. In moderate cases, the time-resolved tunnel current signal has a shape between these two cases.

4.3 *Saturable nonlinear dependence*

Finally, we examine the saturable nonlinearities. Figure 3 shows the case of $I_t(t) = n(t)^{1/k}$ with $k = 2$–4, $A = 2$, $B = 8$ and $\Delta t = 1$ (solid lines). In this case, the time-resolved tunnel current has a dip at zero delay time, with apparent decay constant larger than the actual decay constant of the physical parameter (broken line). The plot is asymmetric and, surprisingly, the plot can be convex upward when the strong pulse reaches the sample first. As the intensity ratio between two pulses increases, the plot becomes almost

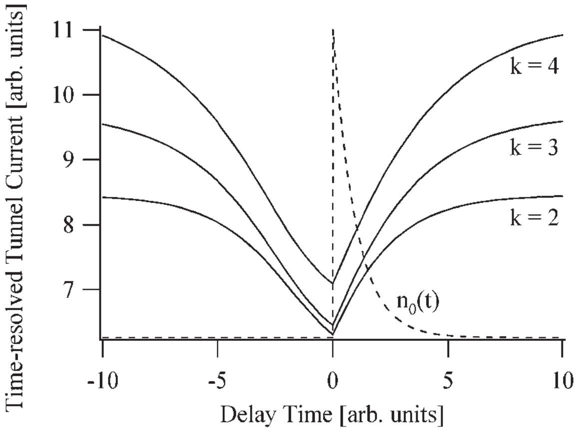

Fig. 3. Time-resolved tunnel current signal expected for saturable dependences of tunnel current on exponentially decaying sample parameter n: $I_t(t) = n(t)^{1/k}$ for $k = 2-4$ (solid lines). The decay process of $n(t)$ is shown with arbitrary vertical scale as well (broken line).

constant in the range of small negative values of t_d. This is because, in the case of a strong saturable nonlinearity, a small increase in n does not contribute to I_t when the absolute value of n is large. Thus, when the intensity of the two pulses are very different, the contribution of the weak pulse becomes negligible if it temporarily overlaps with the stronger pulse. Thus, the constant value of $\overline{I_t}$ for small negative values of t_d is equal to $\overline{I_t(\infty)} - \int_0^\infty I_t(An_0(t))\,dt$ when $A \ll B$. In addition, the plot for positive t_d values approaches $\overline{I_t(\infty)} - \int_{t_d}^\infty I_t(An_0(t))\,dt$. In comparison with a diverging nonlinearity, the saturable nonlinearity correlates the tunnel current *nonlocally* to the decay of the sample parameter. Consequently, the interpretation of the experimental result tends to be more difficult.

5. Experimental Results and Discussion

Here we experimentally examine the light intensity dependence of the time-resolved tunnel current signal obtained by SPPX-STM and interpret them along with the above discussion. The experimental setup was as previously described in detail.[10] The sample was low-temperature-grown $GaN_\alpha As_{1-\alpha}$ ($\alpha = 0.36\%$) and the measurement was performed in air. The reference value of the tunnel current was 0.1 nA with a sample bias voltage of -2.0 V. The sequence of laser pulse pairs had a repetition rate of 80 MHz, an average power of $\sim$10 mW, a center wavelength of 800 nm, and a pulse width of 25 fs. Delay time modulation had an amplitude of 0.7–5 ps at $\sim$20 Hz.

Figure 4 shows the result. The relative intensities of the two pulses were varied from 1 : 1 to 4 : 0.125. In all measurements, the stronger pulse hit the sample later when the delay time was positive. The vertical axis indicates the tunnel current deviation from the steady value. Since SPPX-STM does not provide the absolute value of deviation, the origin of the vertical axis was selected arbitrarily. A positive deviation represents an increase in tunnel current. It can be seen that the all curves show asymmetries for positive and negative delay time regions, even for the intensity ratio of 1 : 1. This was probably due to the difficulty in precise control of light intensity at the tunnel junction. Although the light spots of the two pulses in the pulse pair were carefully

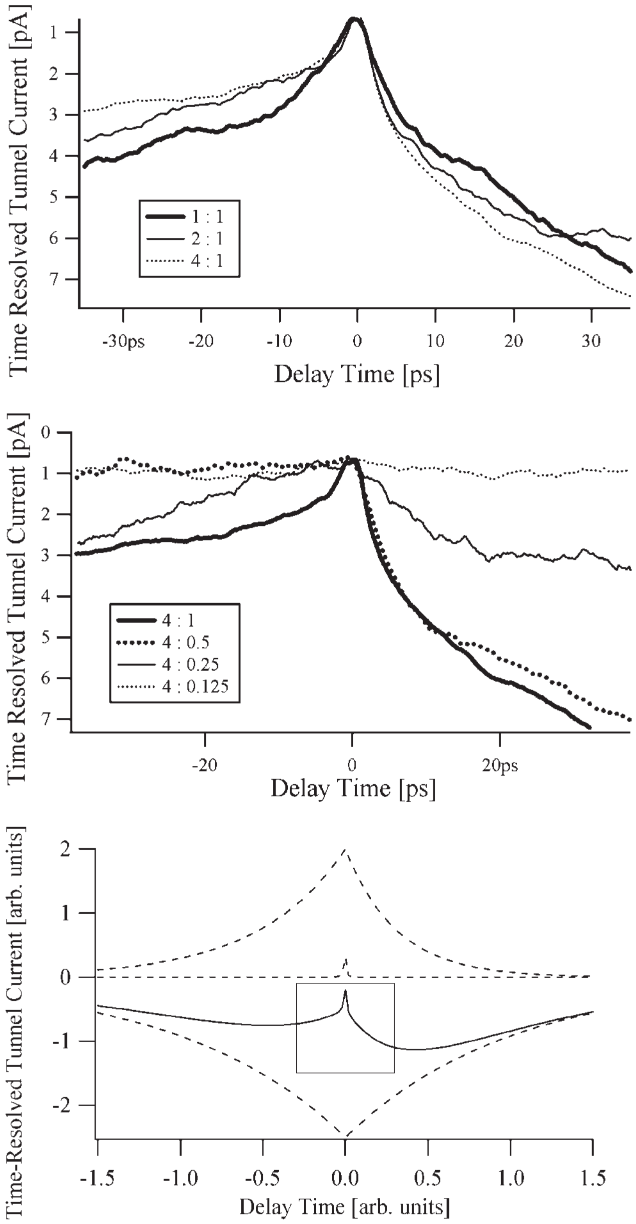

Fig. 4. (a), (b) Experimentally obtained time-resolved tunnel current signals for $GaN_\alpha As_{1-\alpha}$ with varying relative intensities of two pulses from 1 : 1 to 4 : 0.125. (c) Schematics illustrating existence of long-living negative component that explains apparant difference in signal amplitudes on negative and positive sides of delay time.

adjusted at the tunnel junction, the slight difference in the spot diameter or the spot position caused the difference in the effective intensity of the two laser pulses. We also point out that such a precise registration of the laser spots relative to the tunnel junction can temporally fluctuate slightly during the long acquisition time of the time-resolved tunnel current signal. Even so, the plots show a systematic variation against the change in the intensity ratio.[11]

During the attempt to interpret the experimental result in accordance with the above discussion, we noticed the experimental result has several discrepancies from that expected in the above discussion. First, the data for the intensity ratio 1 : 2 was almost perfectly fitted by the combination of a step function and two double exponential functions that decays towards positive and negative infinities of delay time as reported previously.[12] The time constants of the two double exponential functions are almost the same but the amplitudes are different. The amplitude of the step function was equal to this difference such that the two double exponential curves are connected continuously at zero delay time. Although the data for other intensity ratios contained too much noise for curve fitting analysis because of the shorter averaging times, the amplitude instead of the apparent time constant differs between the negative and positive sides as well. Indeed, the curve in one side seems to overlap that of the other side with multiplied by a suitable scaling factor. Secondly, the increase of the intensity of the stronger pulse affected the tunnel current in the opposite direction for the positive and negative regions of delay time in Fig. 4(a), although the numerical models always increased the signal amplitudes with an increase in the intensity of whichever excitation pulse.

Regarding the first issue, the step function in the numerical model was suggested to represent the existence of a decay process longer than the measured delay time range in the previous study.[12] However, such a long-living component should not appear as a step function in a PPX-STM experiment. Since the repetition periodicity of the pulse pair was 12.5 ns, the condition $t_d = -6.25$ ns is physically equivalent to $t_d = +6.25$ ns. Consequently, a time-resolved tunnel current must have the same value at these points. Thus, instead of the step function, the experimental curve might consist of, at least, one long-living component that has a *dip* at zero delay time as illustrated in Fig. 4(c). Within the limited range of the delay time, the signal appears to have different amplitudes in positive and negative sides. Separation of such multiple components with unknown shape is fundamentally very difficult. Future experiments with different wavelengths for excitation might overcome this difficulty by eliminating some of these components. A wider scan of delay time and higher precision for data acquisition must be achived as well. The second issue is due to the different probe-sample separation for PPX-STM measurement with a different excitation intensity. When the excitation intensity is changed, the STM probe thermally elongates or shrinks. Thus, the signal intensity for a different excitation intensity cannot be directly compared. At this moment, no appropriate solution for this problem is known.

6. Conclusions

We examined how an exponentially decaying sample parameter appears in time-resolved tunnel current via a nonlinear dependence between the parameter and tunnel current. It was turned out that PPX-STM has a potential to detect the ultrafast decay processes of any sample parameter that affects the tunnel current, even when the dependence of the parameter on the light intensity is perfectly linear. The light illumination on an STM specimen often suffers from the insufficient light intensity at the tunnel junction to induce the nonlinear response of samples on light intensity. PPX-STM was suggested to be widely applicable even for such cases.

On the other hand, the nonlinear dependence between tunnel current and the sample parameter makes the analysis of the experimental time-resolved tunnel current obtained by PPX-STM difficult, in particular, to extract the ultrafast decay process of the sample. Namely, the exponential decay of a sample parameter does not appear in the time-resolved tunnel current as an exponential function. Furthermore, the apparent time scale of the obtained decaying curve can be different from that of the actual sample parameter. A detailed understanding of the nonlinear dependence is needed for the analysis. In addition, in the experimental results, there can exist multiple components in a time-resolved tunnel current signal. Currently, separating these components to investigate their light intensity dependence remains to be solved in future studies.

In order to apply PPX-STM for a wide variety of specimens other than GaNAs and to obtain more precise experimental results for detailed analysis, experiments with a more intense light source that is capable of varying wavelength and has higher stability for output angle against the fluctuation in the room temperature, a wider scan range of delay time and an ultrahigh vacuum environment are desired. A new experimental system that meets these requirements is under development.

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