Non-resonant frequency components observed in a dynamic Atomic Force Microscope

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Abstract: The oscillating behavior of a micro-cantilever probe plays a central role in the atomic force microscope for studying a nanoscale sample. The oscillatory phenomena in the microscope are numerically investigated by exciting the probe with a single frequency. We observe the non-resonant frequency components, which correspond to a frequency of transient beats superimposed on the stable solution, around the natural frequency of the probe, when the probe is close to the sample. The difference between the non-resonant frequency and the natural frequency changes when the tip-sample distance decreases. Furthermore, we investigate the originating point of the non-resonant frequency components as a function of the tip-sample distance. In addition, we perform an actual experiment for observing the frequency components near the resonant frequency.

Key Words: Dynamic atomic force microscope, cantilever probe, Lennard-Jones potential, side-band frequency, bistability, lightly damped nonlinear oscillator

1. Introduction
An atomic force microscope (AFM) is a powerful tool for examining the structure of a nanoscale object [1, 2]. There are several possible methods for using an AFM depending on the specific purpose. The dynamic AFM mode is a distinctive mode in which a micro-cantilever probe is excited. The subsequent oscillatory phenomena of the probe play a central role in this measurement method. An essential feature of the dynamic AFM mode is that information from the interaction force between the probe tip and the sample is obtained and is recorded. When the probe is excited by the driving force with a large-amplitude oscillation, various nonlinear phenomena can emerge because the interaction force is extremely nonlinear [3–6].

The amplitude of the driven probe decreases monotonically depending on the tip-sample distance [7]. Furthermore, when the distance decreases, two distinctive periodic oscillations appear [7–11]. One
of the periodic solutions is the monotonically decaying oscillation, and the other corresponds to a tapping mode in which the probe intermittently collides with the sample. The later alternative is undesirable when the sample is composed of a soft material because the probe tip is required to move without the collision. Therefore, it is important to determine the tip-sample distance from the decaying oscillation mode (non-contacting oscillation mode). In the dynamic AFM mode, the probe tip is typically excited with a single frequency approximately equal to the natural frequency of the cantilever probe. The response of the probe tip has high-order frequency components due to the nonlinear interaction [12, 13]. The purpose of this study is to investigate the spectral characteristic of the decaying oscillation.

In this study, we investigate the oscillatory behaviors of the probe tip with an approximated dynamical model, which is given by an ordinary differential equation with one degree of freedom [7, 8, 11, 13, 14]. We assume the magnitude of the interaction force between the probe tip and the sample surface from a Lennard-Jones potential. When we set the driving frequency to the natural frequency of the probe, two periodic solutions appear depending on the tip-sample distance. In particular, we calculate the frequency components of one of the solutions. It is verified that two side-bands appear around the fundamental resonant point, which correspond to a frequency of transient beats superimposed on the stable solution. These side-bands are obtained from the time series, excluding 10 times larger transient time than the time constant with no interaction force, though the side-bands of the solution can not be observed after an extremely larger transient time. Such unusual side-bands are attractive because they contain information about the nonlinear interaction. Although the side-bands near the fundamental resonant point were reported in the presence of a water layer [15, 16], the unusual components observed in this study originated from the natural interaction force. The difference between the side-band and the natural frequency increases when the probe tip gradually approaches to the sample surface. Moreover, the originating point of the side-band is investigated in terms of the tip-sample distance, and is determined to originate at the value close to the undesirable solution that intermittently collides with the sample. In addition, we perform the associated experiments to investigate the oscillating probe tip in the atmosphere in the dynamic AFM mode. The obtained result near the sample surface contains the frequency components of the side-bands.

2. Dynamical model of oscillating probe tip influenced by sample

The equation describing the mechanical behavior of the micro-cantilever can be approximated by a mass-spring model with one degree of freedom [7, 8, 11, 13, 14]. Figure 1 shows the mass-spring model, where a sample is put on the $x$-$y$ plane. The probe tip is displaced vertically along the $z'$-axis. The effective mass and linear spring constant are represented by $m$ [kg] and $k$[kg s$^{-2}$], respectively. $z_0$ [m]

![Fig. 1. Mass-spring model as an equivalent model of oscillating behaviors of a cantilever probe in the dynamic AFM mode.](image-url)
corresponds to the equilibrium position of the probe tip with no interaction force. In the dynamic AFM mode, a periodic external force $l \cos \omega t$ [m] is applied to the probe tip. We assume that the frictional force is proportional to the derivative of the position of the probe tip, namely $-C \dot{z}'/d\tau$. The constant $C$ [kg s$^{-1}$] corresponds to the coefficient of viscosity.

Generally, the probe tip is influenced by the sample surface. In this study, we assume that the interaction force originates from the Lennard-Jones potential ($= U_0(r)$ [J]), which is the potential between a pair of neutral atoms [17]:

$$U_0(r) = 4\varepsilon_{LJ} \left\{ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right\},$$

where $r$ [m] is the distance between two neutral atoms. $\varepsilon_{LJ}$ [J] and $\sigma$ [m] correspond to the cohesion energy of a neutral atom and the distance between atoms in equilibrium, respectively. In this study, the sample surface and the probe tip are approximated by an infinite circle plane on the $x$-$y$ axis and a circular cone, respectively, because the sample surface is much larger than the probe tip. The following potential ($= U(z')$) is derived under this assumption:

$$U(z') = \frac{2}{3} R_0 \pi^2 \varepsilon_{LJ} n_0^2 \sigma^5 \left\{ \frac{1}{210} \left( \frac{\sigma}{z'} \right)^7 - \left( \frac{\sigma}{z'} \right)^{2} \right\},$$

where $R_0$ [m] and $n_0$ [m$^{-3}$] are the radius of the curvature of the tip and the number density of atoms, respectively. Then, the interaction force between the probe tip and the sample surface as a function of $z'$ ($= F(z')$ [N]) can be given by the following:

$$F(z') = -\frac{dU(z')}{dz'} = \frac{2}{3} R_0 \pi^2 \varepsilon_{LJ} n_0^2 \sigma^4 \left\{ \frac{1}{30} \left( \frac{\sigma}{z'} \right)^8 - \left( \frac{\sigma}{z'} \right)^{2} \right\}. \quad (1)$$

In this study, we set $\sigma = 0.25 \times 10^{-9}$, $n_0 = 5.00 \times 10^{28}$, and $\varepsilon_{LJ} = 1.602 \times 10^{-21}$ by assuming that both the probe tip and the observed sample are composed of silicon. In addition, we also set $m = 5.17 \times 10^{-12}$ and $R_0 = 15 \times 10^{-9}$ because of specifications of the actual micro-cantilever probe OMCL-AC240TM [18]. Figure 2 shows the characteristic curve of $F(z')$ as a function of $z'$ with these parameters.

From the Newtonian law, the motion equation of the forced probe tip can be given by

$$m \frac{d^2 z'}{dt^2} = -\frac{m \omega_0}{Q} \frac{dz'}{dt} - m \omega_0^2 (z'-z_0) + m \omega_0^2 l \cos \omega t + F(z'), \quad (2)$$

where $\omega_0 (= \sqrt{k/m}$) [rad/s] corresponds to the natural angular frequency with no interaction force. The quality factor of this model is represented by $Q (= m \omega_0/C)$.

Substituting

$$t = \frac{\tau}{\omega_0}, \quad z' = \sigma z, \quad \varepsilon = \frac{z_0}{\sigma}, \quad \mu = \frac{2}{3m \omega_0^2} R_0 \pi^2 \varepsilon_{LJ} n_0^2 \sigma^3, \quad a_e = \frac{l}{\sigma}, \quad \omega_1 = \frac{\omega}{\omega_0}, \quad (3)$$

into Eq. (2) yields the following normalized equation:

$$\ddot{z} + \frac{1}{Q} \dot{z} + (z - \varepsilon) = a_e \cos(\omega_1 \tau) + \mu \left\{ \frac{1}{30} z^{\sigma} - z^{-\sigma} \right\}, \quad \left( \cdot = \frac{d^2}{dt^2}, \quad \cdot = \frac{d}{d\tau} \right). \quad (4)$$

![Fig. 2. Interaction force between the probe tip and sample surface.](image-url)
Fig. 3. Free oscillation of the probe tip ($\mu = 6.1756, Q = 200, a_e = 0.2, \omega_1 = 1$, and $\varepsilon = 80$), where the initial conditions are $z(0) = 79.69$ and $\dot{z}(0) = 39.99$.

Fig. 4. Two coexisting periodic solutions around the saddle-node bifurcation point ($\mu = 6.1756, Q = 200, a_e = 0.2$, and $\omega_1 = 1$).

Fig. 5. Coexisting solutions for $\varepsilon = 10$ and $a_e = 0.20$. The initial conditions of (a) and (b) are $z(0) = 18.68$, $\dot{z}(0) = 1.977$, and $z(0) = 3.486$, $\dot{z}(0) = 1.016$, respectively.

3. Numerical results

In this section, we numerically investigate the behaviors of the driven probe tip in Eq. (4) by using the fourth-order Runge-Kutta method with a step size of $2\pi/(8192\omega_1)$. Assuming the same conditions as those in Fig. 2, we set the parameter $\mu = 6.1756$ in the following results. We also set the parameters $Q = 200$ and $\omega_1 = 1$ by referring to the operating condition of an AFM in the dynamic mode.

First, we use the parameter $\varepsilon$, which corresponds to $z_0$ (the distance between the probe tip and the sample surface), as a control parameter, for $a_e = 0.2$. When $\varepsilon$ is large, the amplitude of $z$ is almost equal to $a_eQ = 40$, because the influence of the interaction force between the probe tip and the sample surface is negligible as shown in Fig. 3. For smaller $\varepsilon$, the influence of the interaction force becomes stronger. To investigate this influence in further detail, we calculate the Poincaré mapped
point at a constant time interval of $2n\pi/\omega_1$, where $n$ is a natural number. Then, we check the stabilities of these solutions using the procedure [19]. Figure 4 shows the existing periodic solutions. The green curve corresponds to a stable periodic solution, which originates from the free oscillation as shown in Fig. 3. The blue and yellow curves are the periodic solution and the corresponding saddle solution, respectively. These solutions disappear via a saddle-node bifurcation at the point $\varepsilon_c = 40.65$ for $a_e = 0.2$. Therefore, the distinctive periodic solutions coexist for $\varepsilon < \varepsilon_c$. Figures 5(a) and (b) show the coexisting solutions with the same value of $\varepsilon = 10$. Comparing both the time series, it is observed that the amplitude of $z$ in Fig. 5(b) is smaller than that in Fig. 5(a). To distinguish between the coexisting solutions, we name the solutions in Figs. 5(a) and (b) as large-amplitude and small-amplitude solutions, respectively. Figure 6 shows the corresponding trajectories on the phase plane of the solutions in Fig. 5. In the figure, the large-amplitude solution collides with the sample surface ($z = 0$).

Figure 7 shows the values of peak to peak $z = z_{pp}$ of the small-amplitude solution as a function of $\varepsilon$ for five different values of $a_e$. For sufficiently large $\varepsilon$, which means that the probe tip is far from the sample surface, $z_{pp}$ is almost equal to the value of $a_eQ$, namely the free oscillation. In contrast, when $\varepsilon$ decreases, $z_{pp}$ decreases almost linearly with any value of $a_e$. Figures 3 and 8 show the time series of the small-amplitude solutions, when we set $a_e = 0.2$, for $\varepsilon = 80$ and $\varepsilon = 20$, respectively. It is seen that the values of $z_{pp}$ for $\varepsilon = 80$ and 20 are considerably different from each other. Therefore, the amplitude decay implies the distance between the probe tip and the sample surface. This phenomenon is important to measure the uneven sample surface in the dynamic amplitude modulation AFM mode [7, 8].

Figure 9 shows both values of $z_{pp}$ for the small- and large-amplitude solutions as a function of $\varepsilon$ for $a_e = 0.2$. The coexistence of the two stable solutions are presented for $\varepsilon < \varepsilon_c$ in Fig. 4. In Ref. [8], it is verified that the two stable branches appear when the probe tip approaches the sample surface.

![Fig. 6. Trajectories of Figs. 5(a) and (b) on the phase plane.](image1)

![Fig. 7. Variations in the amplitude of the small-amplitude solution as a function of $\varepsilon$.](image2)
In addition, the fracture of the small-amplitude solution is shown to be observed under a large free oscillation amplitude. As the obtained result in Fig. 9 is consistent with the result in Fig. 3(a) in [8], the all characteristic curves in Fig. 7 are regarded as a case of relatively small free oscillation amplitude in literature because the all curves in Fig. 7 are for $a_c \leq 0.2$. For larger $a_c$, the fracture of the small-amplitude oscillation is observed in Eq. (4).

Next, we investigate the spectral characteristic of the solutions as a function of $\varepsilon$ by setting $a_c = 0.2$. With no interaction force, Eq. (4) corresponds to a linear differential equation. The transient term of this equation can be derived as $Ae^{-\tau/2Q} \sin(\beta \tau + \phi)$, where $\beta$ is obtained as $\sqrt{1 - 1/(4Q^2)}$. Amplitude $A$ and initial phase $\phi$ are determined through the initial condition. The value $2Q$ corresponds to a time constant because the amplitude of the transient term decreases by $e^{-1}$ when $\tau$ arrives at $2Q$. We use the fast Fourier transform (FFT) with a frequency resolution of $6.519 \times 10^{-7}$ and time interval $\Delta \tau = 1.534 \times 10^6$, excluding the transient time ($= \tau_{\text{tran}}$) = 4000 to obtain the results. Therefore, $\tau_{\text{tran}}$ is 10 times larger than the time constant when $Q = 200$. Figures 10(a), (b), (c), and (d) show the power spectra ($= P [\text{dB}]$) around the fundamental resonant point, namely $\omega_1 = 1$ for $\varepsilon = 10, 20, 35$, and 60. Note that there are small both side-bands near the resonance at $\omega_1 = 1$ for $\varepsilon = 10, 20, 35$, and 60. Moreover, the positions of the side-bands in terms of $\omega_1$ differ slightly in the results. In contrast, no side-band appears in the FFT result for $\varepsilon = 60$ in Fig. 10(d). Furthermore, Fig. 11 shows the power spectrum of the large-amplitude solution for $\varepsilon = 10$ around the resonant point. The shape of the power spectrum is observed to be distinct from the results of the small-amplitude solutions because there are the several peaks seemingly originating from the repulsive force between the probe tip and the sample surface.

We calculate the power spectra of the small-amplitude solutions for $10 \leq \varepsilon \leq 60$ around the fundamental resonant point as shown in Fig. 12 in order to obtain more details. The brighter color in this figure indicates the larger frequency component. It should be noted that the difference between the resonant point and the side-band gradually increases when $\varepsilon$ decreases from $\varepsilon = 60$. Moreover,
to investigate the originating point of the side-bands, we plot the variation of frequency deviation of the upper side-band from $\omega_1 = 1$ ($= \Delta \omega$) in terms of $\epsilon$, as shown in Fig. 13. $\Delta \omega$ corresponds to the frequency of transient beats superimposed on the stable small-amplitude solution. Therefore, for an extremely large value of $\tau_{\text{tran}}$, approximately 20 thousands times larger than the transient time, $\Delta \omega$ cannot be obtained for any value of $\epsilon$. Nevertheless, for a smaller $\tau_{\text{tran}} (\leq 2Q \times 10^4)$, the value of $\Delta \omega$ is changed as a function of $\epsilon$, as shown in Fig. 13. From this figure, when $\epsilon$ increases from 10, $\Delta \omega$ gradually decreases. However, we are unable to calculate the value of $\Delta \omega$ at $\epsilon \approx 37.0$ because both side-bands are merged with the resonant peak around $\omega_1 = 1$. Figure 14 shows the variation of the fourth power of $\Delta \omega$ as a function of $\epsilon$, where the value of $\Delta \omega^4$ exponentially decays to the originating point. Moreover, the eigenvalues of the stable large-amplitude solutions in the complex plane move around the unit circle, as shown in Fig. 15. This phenomenon is qualitatively consistent with the jumping behavior of lightly damped oscillators [20]. As the point where the side-band disappears is close to $\epsilon^c$, the originating point of the side-bands implies that two distinctive stable solutions coexist.
4. Experimental results

In this section, we investigate the spectral characteristics of the oscillating probe tip in the non-contact AFM mode by performing experiments using the environment control unit AFM5300E [21]. Figure 16(a) shows the power spectrum of the oscillating probe tip when the distance between the probe tip and the sample surface is sufficiently large. The horizontal axis is normalized by the natural frequency, which is approximately equal to $76,958 \times 2\pi$ rad/s in this figure. Therefore, this axis can be regarded as $\omega_1$ in the numerical results. Then, we move the position of the probe tip toward the sample surface. Figure 16(b) shows the power spectrum near the sample surface. It is seen that both side-bands appear around $\omega_1 = 1$. Further detailed investigation of the side-bands in the experiment will be performed as our future work.
Fig. 14. Characteristic curve of $\Delta \omega^4$ obtained from the upper side-band of the small-amplitude solution as a function of $\varepsilon$ ($a_e = 0.2$).

Fig. 15. Complex conjugate eigenvalues of the stable large-amplitude solution for $a_e = 0.2$ and the unit circle (dashed circle) in the complex plane for $10.0 \leq \varepsilon < \varepsilon^c$.

Fig. 16. Experimentally obtained results in the dynamic non-contact AFM mode.

(a) Far from the sample surface. (b) Near the sample surface.

5. Conclusions
We investigated the oscillating behaviors of the micro-cantilever probe in the dynamic AFM mode. The existing periodic solutions were investigated by using the nonlinear mass-spring model with one degree of freedom. We focused on one of the coexisting periodic solutions. Then, we investigated the amplitude as a function of $\varepsilon$ (the equilibrium position of the probe tip), and the corresponding spectral characteristics. In particular, we observed the side-bands near the fundamental resonant point. It was confirmed that the difference between the non-resonant frequency and the natural frequency increased for smaller values of $\varepsilon$. Moreover, we discussed the originating point of the side-bands in terms of $\varepsilon$, and we confirmed that the originating point was close to the bifurcation point of the undesirable solution (the large-amplitude solution). In addition, we performed the experimental measurements and observed the side-bands when the probe tip was close to the sample surface. From a practical
point of view, the large-amplitude solution is undesirable for scanning a soft material. Therefore, the originating point of the side-bands is used as a prediction point of the coexistence of the undesirable solution in the dynamic non-contact AFM mode.

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