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Secondary resonances in tapping mode atomic force microscopy

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Abstract

This work investigates a dual-frequency excitation scheme for tapping-mode atomic force microscopy (AFM) and demonstrates, through numerical simulations, that it can significantly enhance nanoscale compositional mapping of heterogeneous surfaces compared to conventional single-frequency operation. The AFM cantilever is modeled as an Euler–Bernoulli beam with a nonlinear tip–sample interaction and reduced using a two-mode Galerkin approach. Rather than driving the cantilever solely near its first resonance, we apply two distinct excitations whose difference frequency is tuned close to that resonance. Due to nonlinear intermodulation at the intermittent tip–sample contact, this scheme generates a strong secondary (combination) resonance without directly exciting the cantilever at its natural frequency. The amplitude of this secondary resonance is shown to be highly sensitive to nanoscale variations in both long-range attractive forces (characterized by the Hamaker constant) and short-range repulsive contact forces (described by the effective elastic modulus of the sample). By contrast, the conventional single-frequency tapping response exhibits only weak variations under the same parameter changes. These results indicate that tapping can function as an intrinsic nonlinear amplifier of material contrast, enabling the simultaneous acquisition of high-resolution topography and spatially resolved functional contrast (chemical/mechanical). The proposed method therefore provides a pathway toward quantitative nanoscale property mapping—such as adhesion and stiffness—without requiring continuous high-force contact with the surface.

Keywords: Atomic force microscopy; Nonlinear dynamics; Multi-frequency excitation; Combination resonance

1. Introduction

Atomic Force Microscopy (AFM) is one of the most widely used techniques for nanoscale surface characterization in materials science, thin-film engineering, polymer research, and biological systems because it can provide simultaneous access to surface topography and local physical properties with nanometre-scale spatial resolution. In a typical AFM, a sharp probe mounted on a compliant microcantilever is raster-scanned across the sample while tip–sample interaction forces are continuously monitored. These interactions are then interpreted to recover both geometric information (height, morphology) and functional information (adhesion, viscoelasticity, local stiffness), making AFM essential in contexts where high-resolution, non-destructive probing is required. In particular, tapping-mode (intermittent-contact) AFM has become the standard for imaging delicate or heterogeneous samples because it minimizes lateral drag and wear compared to contact mode, while still maintaining high spatial resolution. During tapping-mode operation, the cantilever is driven near its first resonance and oscillates with controlled amplitude, approaching and withdrawing from the surface in each cycle. The measured oscillation amplitude and phase at this primary resonance are used both for feedback control (to reconstruct surface height) and for generating qualitative contrast related to properties such as adhesion and stiffness [1–4].

Despite its practical success, tapping-mode AFM is governed by a strongly nonlinear and intermittent tip–sample interaction. The probe simultaneously experiences long-range attractive forces, dominated by van der Waals attraction, and short-range repulsive forces associated with contact mechanics once the tip briefly touches or indents the surface. As the cantilever oscillates, it repeatedly transitions between these force regimes within a single vibration cycle. This nonlinear interaction produces higher harmonics, intermodulation products, and even bistable or hysteretic behavior in the cantilever response. From a physical perspective, this nonlinearity is important for two reasons. First, it means the cantilever cannot be treated as a simple, single-degree-of-freedom linear oscillator when high-fidelity interpretation is required. Second, and more importantly, it means that the measured dynamics actually contain rich, property-specific information about local chemistry (surface energy, adhesion, Hamaker constant) and local mechanics (effective elastic modulus, viscoelastic damping). However, in standard single-frequency tapping AFM, only the amplitude and phase of the first flexural resonance are typically monitored and used. This conventional approach is dominated by the linear dynamic response of the first mode and therefore has limited sensitivity to subtle nanoscale variations in adhesion or stiffness. As a result, valuable compositional or nanomechanical contrast may remain weak or even hidden in traditional amplitude-modulation AFM images [3–8].

In recent years, multi-frequency and multi-modal AFM techniques have been developed specifically to address this limitation. The central idea is that nonlinear tip–sample dynamics naturally couple different spectral components and different flexural modes of the cantilever. By deliberately exciting and reading out multiple frequencies or multiple modes, one can extract additional contrast channels that are more directly tied to material properties. For example, bimodal and multifrequency AFM approaches use more than one driven frequency or more than one cantilever mode to separate conservative from dissipative interactions, enhance phase contrast, and improve sensitivity to local stiffness, adhesion, viscoelastic losses, and surface chemistry. These approaches show that, instead of treating the nonlinearities as a nuisance that corrupts the signal, they can be actively exploited as a source of amplified material contrast. The nonlinear response of the cantilever can be engineered into a metrological asset rather than a limitation [6–10, 13, 14].

The present work builds directly on this idea by proposing and analyzing a dual-frequency excitation strategy for tapping-mode AFM that intentionally generates what we refer to as a secondary or combination resonance. Instead of driving the cantilever with a single signal near its first resonance, we apply two distinct drive frequencies, Ω_1 and Ω_2, at higher values such that neither drive is itself resonant. Because the tip–sample interaction is nonlinear, these two drive frequencies

do not remain isolated. They mix through nonlinear intermodulation at the oscillating tip and generate new response components at linear combinations $m\Omega_1 \pm n\Omega_2$. Of particular interest is the difference frequency $\Omega_2 - \Omega_1$. By choosing Ω_1 and Ω_2 so that $\Omega_2 - \Omega_1$ is close to the first natural frequency of the cantilever, we effectively 'synthesize' an internal drive at the fundamental mode without directly exciting that mode. As a result, energy is down-mixed from the two higher-frequency drives into a strong response near the first resonance—the combination resonance—which is inherently nonlinear and does not arise in a purely linear single-mode model [6, 9, 10, 13].

We hypothesize that because this secondary resonance is created by nonlinear intermodulation at the tip–sample contact, it is intrinsically more sensitive to nanoscale variations in both long-range attraction (captured, for example, by the Hamaker constant and thus linked to surface chemistry and adhesion) and short-range repulsive contact mechanics (linked to effective elastic modulus and local stiffness) than the traditional primary resonance channel. To test this hypothesis, we model the AFM cantilever as an Euler–Bernoulli beam, apply a two-mode Galerkin reduction to capture its dominant flexural dynamics, and run numerical simulations of its steady-state response under dual-frequency excitation. We then evaluate how the amplitude and phase of the combination resonance vary when physically meaningful parameters such as the Hamaker constant and the effective elastic modulus are perturbed. The results show that small changes in adhesion or stiffness—which produce only modest shifts in conventional single-frequency tapping—yield strong, clearly distinguishable signatures in the secondary resonance channel. In practical terms, properly tuned dual-frequency tapping can act as an intrinsic nonlinear amplifier of material contrast and can provide, in a single scan, both high-resolution topography and spatially resolved functional maps of adhesion and stiffness at the nanoscale [9, 14, 15, 16].

2. Theoretical Formulation

2.1 Microcantilever Dynamic Model

The classical beam theory based on the Euler–Bernoulli assumptions is used to develop a continuous model for the AFM probe. As shown in Fig. 1, the microcantilever has a rectangular cross-section $b \times h$ and a tip mounted on its free end. The total deflection on $u(x,t)$ is expressed as $u(x,t) = w(x,t) + y(t)$, where w(x,t) is the deflection of the microcantilever relative to a non-inertial reference frame attached to the base. The base excitation $y(t)$ from a dither piezo is assumed to be a dual-frequency harmonic displacement:

$$y(t) = Y_1 \cos(\Omega_1 t) + Y_2 \cos(\Omega_2 t) \tag{1}$$

where Y_1 and Y_2 are the excitation amplitudes and Ω_1 and Ω_2 are the two distinct excitation frequencies. The instantaneous tip/sample separation is $z(t) = Z - u(l, t) = Z - w(l, t) - y(t)$, where Z is the equilibrium separation.

Assuming the surface is flat and the tip is a hemisphere, we model the interaction forces (van der Waals attraction and DMT contact repulsion) as:

$$F_{vdW} = -\frac{HR}{6z^2} \tag{2}$$

$$F_{Contact} = \frac{4}{3} E^* \sqrt{R} (a_0 - z)^{3/2} \tag{3}$$

where E^* is the effective elastic modulus of the tip-sample system, defined by:

$$\frac{1}{E^*} = \frac{1 - v_{tip}^2}{E_{tip}} + \frac{1 - v_{sample}^2}{E_{sample}} \tag{4}$$

The total tip-sample interaction force, $F_{ts}(t)$, is a non-smooth piecewise function dependent on the instantaneous separation *z(t)* relative to the intermolecular distance a_0 (the contact boundary):

$$F_{ts}(t) = \begin{cases} F_{vdW} & z(t) \geq a_0 (Non - Contact) \\ F_{Contact} - \dfrac{HR}{6a_0^2} & z(t) < a_0 (Contact) \end{cases} \tag{5}$$

This non-smooth nature at $z = a_0$ is the primary source of the system's complex nonlinear dynamics.

Using the extended Hamilton's principle, the governing partial differential equation (PDE) for the *relative* motion *w (x, t)* is derived as:

$$EI \frac{\partial^4 w}{\partial x^4} + \rho A \frac{\partial^2 w}{\partial t^2} + c \frac{\partial w}{\partial t} = -\rho A \frac{\partial^2 y}{\partial t^2} - c \frac{\partial y}{\partial t} \tag{6}$$

where ρ is the material density, *A* is the cross-sectional area, *c* is the viscous damping coefficient per unit length, *E* is the Young's modulus, and *I* is the second moment of area of the cross-section. This equation is subject to the following physical boundary conditions:

At the clamped end (*x = 0*):
- Displacement: *w (0, t) = 0*
- Slope: *w' (0, t) = 0*

At the free end (*x = l*):
- Bending Moment: *EIw''(l, t) = 0*
- Shear Force: *EIw'''(l, t) = −F_{ts}(t)*

Here, F_{ts} is the piecewise nonlinear force defined in Eq. (5).

To solve this system, we develop a reduced-order model. First, the Lagrangian (*L = T - V*) and the energy dissipation function (*D*) of the system are formulated. The total kinetic energy *T* is:

$$T = \frac{1}{2} \int_0^l \rho A \left(\dot{w}(x,t) + \dot{y}(t) \right)^2 dx \tag{7}$$

The potential energy *V* includes the elastic strain energy V_e and the tip-sample potential V_{ts}:

$$V = V_e + V_{ts} = \frac{1}{2} \int_0^l EI \left(w''(x,t) \right)^2 dx + \int F_{ts}(z) dz \tag{8}$$

The dissipation function *D* is:

$$D = \frac{1}{2} \int_0^l c \left(\dot{w}(x,t) + \dot{y}(t) \right)^2 dx \tag{9}$$

Next, following the Galerkin procedure, the relative deflection *w (x, t)* is approximated as a finite series expansion using the mode shapes ($\phi_n(x)$) of a standard cantilever beam, truncated to *N* modes:

$$w(x,t) = \sum_{n=1}^{N} \phi_n(x) q_n(t) \tag{10}$$

This assumed solution (Eq. 10) is substituted into the energy functions (Eq. 7-9). Finally, the Lagrange equations are applied for each modal coordinate q_n:

$$\frac{d}{dt}\left(\frac{\partial \mathrm{L}}{\partial \dot{q}_n}\right) - \frac{\partial \mathrm{L}}{\partial q_n} = -\frac{\partial \mathrm{D}}{\partial \dot{q}_n} \quad (for\ n = 1,2,\dots N) \tag{11}$$

where, *L* is the Lagrangian and *D* is the Rayleigh dissipation function. This procedure reduces the governing PDE (Eq. 6) and its complex boundary conditions into a set of *N* nonlinearly coupled ordinary differential equations (ODEs). After normalization, these equations take the form:

$$\ddot{q}_n + 2\zeta_n \omega_n \dot{q}_n + \omega_n^2 q_n + f_{n,nonlinear}(q_1 \dots q_N, y) = f_{n,forcing}(y, \dot{y}, \ddot{y}) \tag{12}$$

where ω_n and ζ_n are the *n*-th natural frequency and modal damping, $f_{n,nonlinear}$ is the generalized nonlinear force from $\mathrm{F_{ts}}$, and $f_{n,forcing}$ is the generalized forcing from the base excitation. In this study, first two modes (*N=2*) are considered for Galerkin discretization, resulting in a system of two coupled ODEs for $q_1(t)$ and $q_2(t)$. It should be noted that we employ two mode shapes for discretization because in the proposed combination resonance, the excitation scheme is such that only first two mode shapes are, effectively, contributes to the cantilever response.

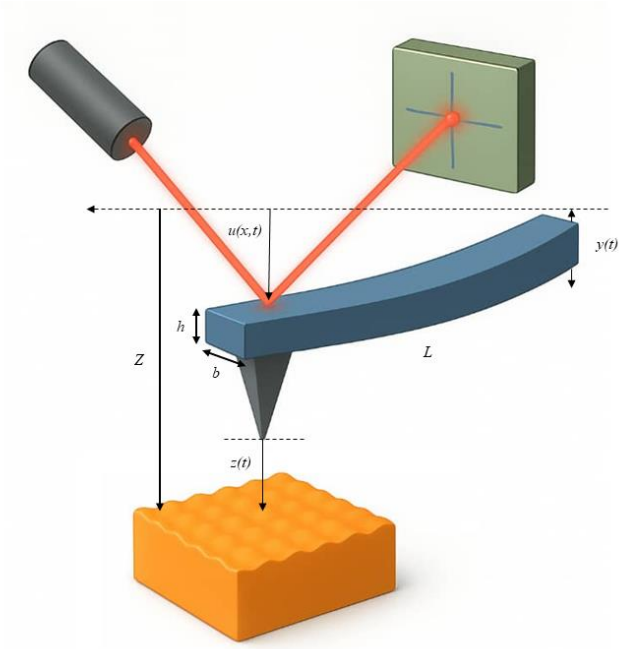

Figure 1. Schematic of atomic force microscope in tapping mode

3. Numerical Results and Discussion

3.1 Natural Frequencies

The set of two nonlinearly coupled ODEs (*N=2*) derived from the Lagrangian procedure was solved numerically to investigate the system's response to the dual-frequency excitation. The physical and geometrical parameters used for the simulation are listed in Tables 1. Based on these values the first three natural frequencies of the microcantilever are obtained as $\omega_{1,2,3}$ = 44.707, 280.228, 786.645 kHz.

Table 1. Characteristics of atomic force microscope microbeam in tapping mode.

Characteristic of microbeam	Size	unit
Microbeam tip radius **(R)**	10×10^{-9}	m
Length **(l)**	250×10^{-6}	m
Cross section **(A)**	8.09×10^{-11}	m^2
The second moment of surface torque **(I)**	3.57×10^{-23}	m^4
Density **(ρ)**	2300	kg/m^3
Elastic modulus **(E)**	130×10^9	Pa
Effective elastic modulus **(E*)**	10.2×10^9	Pa
Air quality factor **(Q)**	33.3	–
Hamaker constant **(H)**	2.96×10^{-19}	J
Intermolecular distance **(a_0)**	3.8×10^{-10}	m

3.2 Combination (Secondary) Resonance

To generate a combination resonance, two excitation frequencies were selected such that their difference closely matched the cantilever's first natural frequency. In our setup, the excitation frequencies were $\Omega_1 = 150$ kHz and $\Omega_2 = 197.707$ kHz, resulting in a difference of $\Omega_2 - \Omega_1 = 47.707$ kHz $\approx \omega_1$. The underlying mechanism is the nonlinear tip–sample interaction, which mixes the two high-frequency excitation components and generates intermodulation products, including a component at the difference frequency $\Omega_2 - \Omega_1$. By tuning this component near the first flexural mode's natural frequency, the cantilever is resonantly driven in its fundamental mode, even though the primary excitation frequencies (Ω_1 and Ω_2) are far from any natural resonance. This energy transfer through nonlinear frequency mixing gives rise to the so-called secondary or combination resonance.

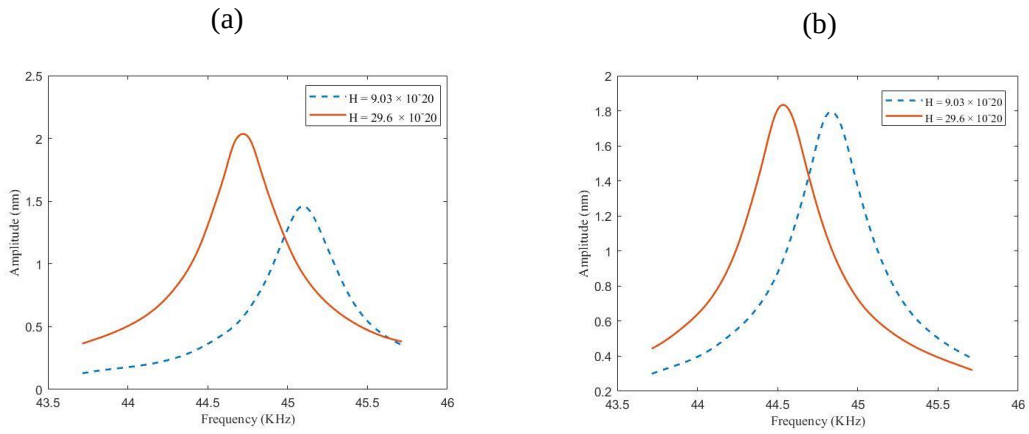

Figure 2. Variations of tip amplitude with excitation frequency for two different Hamaker constants **(a)** for combination resonance (**b)** for single frequency excitation

In the first study (Fig. 2), the Hamaker constant H was varied. The Hamaker constant characterizes the strength of long-range van der Waals attraction between the probe tip and the sample and is therefore linked to local chemical composition and surface energy. Numerical simulations show that even a modest change in H produces a large and clearly distinguishable shift in both the amplitude and the phase of the combination resonance. The same change in H produces only a comparatively mild modification in the traditional single-frequency response. This result implies

that the slope $\partial A/\partial H$ (and similarly $\partial\varphi/\partial H$) is considerably steeper for the secondary resonance channel. In practical terms, this means that the nonlinear, dual-frequency scheme can act as a chemically sensitive contrast mechanism for compositional mapping of heterogeneous surfaces, revealing differences in local adhesion or surface energy that may not be detectable in conventional tapping-mode AFM.

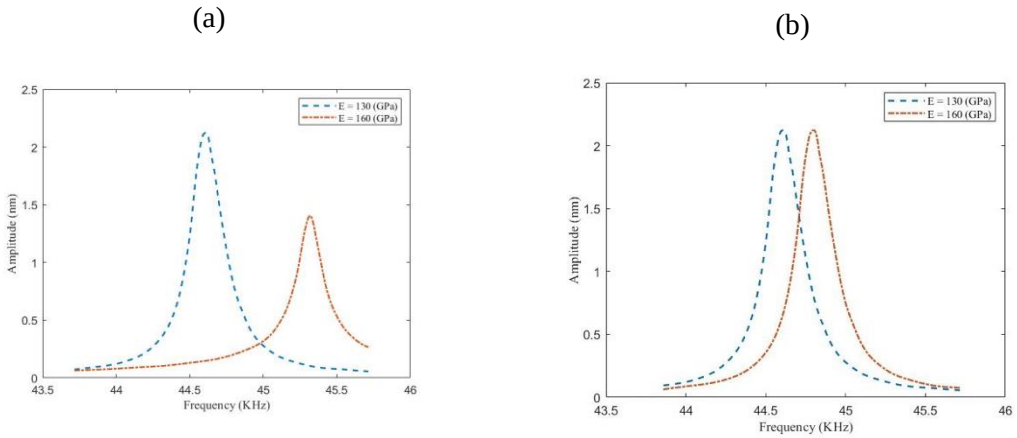

Figure 3. Variations of tip amplitude with excitation frequency for two different elasticity moduli **(a)** for combination resonance (**b)** for single frequency excitation

As shown in figure 3, the effective elastic modulus of the sample, E*, was varied to probe sensitivity to short-range repulsive contact forces. Once again, we observe that changes in E* lead to a strong and easily resolvable variation in the amplitude and phase of the combination resonance, while the conventional single-frequency response near ω_1 exhibits a much weaker dependence on E*. This indicates that the nonlinear intermodulation channel is not only sensitive to long-range attractive forces (through H), but is also highly responsive to local mechanical stiffness encoded in E*. As a result, the same AFM scan can, in principle, provide both high-resolution topography (through the conventional amplitude-control channel) and localized nanomechanical contrast (through the secondary resonance channel) without requiring additional passes or continuous high-force contact with the surface.

4. Conclusion

This study numerically investigates the nonlinear dynamic behavior of an atomic force microscope (AFM) cantilever operating in tapping mode under both the conventional single-frequency excitation and a dual-frequency excitation scheme. By driving the cantilever at two distinct excitation frequencies whose difference frequency is tuned close to the fundamental flexural resonance, the system produces a nonlinear combination (secondary) resonance through intermodulation processes occurring at the tip–sample interaction zone. Unlike the primary resonance, which primarily represents the linear response of the first flexural eigenmode, the secondary resonance arises from the nonlinear and intermittent tip–sample force coupling during each oscillation cycle. Numerical simulations reveal that the amplitude of this secondary resonance is highly sensitive to minute variations in both long-range van der Waals attractions (characterized by the Hamaker constant) and short-range contact mechanics (effective modulus). Small changes in adhesion or local stiffness, which induce only minor variations in the conventional tapping-mode response, generate strong and spatially distinct contrast in the combination-resonance channel. This finding demonstrates that du-

al-frequency (multi-excitation) tapping AFM can function as an intrinsic nonlinear amplifier of material-dependent contrast. From a practical standpoint, a single multi-frequency AFM scan can thus simultaneously provide high-resolution topography and spatially resolved functional contrast (chemical or mechanical), enabling quantitative nanoscale property mapping without continuous high-force contact. In this way, multi-frequency tapping AFM evolves beyond purely morphological imaging into a multimodal nanoscale characterization tool capable of probing both surface structure and material properties

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