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A simple model for the effect of nonuniform mass loading on the response of gravimetric chemical sensors

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ABSTRACT

We reconsider the effects of nonuniform mass loading in gravimetric chemical or biological sensors by applying a mode-matching approach. The approach yields an eigenvalue equation which is solved numerically. The results reveal how an increasing concentration of the mass deposited on top of a microacoustic resonator leads to a frequency shift different from that predicted by the Sauerbrey equation valid for uniform mass loading. Depending on the mode type and the mass distribution, a nonuniform mass loading can lead to a frequency shift higher or lower than that of the uniform loading case. We then derive a point-mass solution which can be considered exact in all practical applications and compare it to available data in the literature. The point-mass solution is used to obtain a closed formula for the frequency shift produced by an arbitrary nonuniform mass loading, and some consequences of this formula are discussed. Altogether, the model presented is quite simple and yet allows one to judge the effects of nonuniform mass loading in gravimetric sensors very efficiently.

1. Introduction

It has been known for decades that the resonance frequency of microacoustic resonators decreases when the resonator adsorbs mass on its surface. The principle has been used in chemical sensors such as quartz crystal microbalances (QCM or QMB), surface acoustic wave (SAW) resonators, and thin-film bulk acoustic wave resonators (FBAR). In these devices, a miniaturized piezoelectric resonator is coated with a thin layer that specifically incorporates the analyte molecules to be detected [1,2].

It was shown by Sauerbrey in 1959 that the change $\triangle f$ of the resonance frequency is directly proportional to the mass increase m in the sensitive coating due to the incorporation of analyte molecules in the layer [3, Eq. (4)]:

$$\frac{\Delta f}{f} = -\frac{m}{M}. \tag{1}$$

Here, M is the resonating mass in the quiescent state (no analyte molecules incorporated in the sensor layer). The quiescent resonance frequency f mainly depends on the materials and acoustic wave types involved and on the transducer geometry.

The validity of the Sauerbrey Eq. (1) hinges on a number of assumptions:

- The equation only describes the effects of mass loading and neglects electrical loading, elastic loading, viscous loading, etc.
- It assumes that the mass distribution after analyte incorporation is uniform.
- It assumes that the acoustic wave attenuation changes only negligibly upon molecule accretion.
- It assumes an acoustically thin layer with $h \ll \lambda$, where λ is the wavelength of the standing acoustic wave [4].
- It is a static model that only describes the steady-state response. No dynamic effects caused by chemical reactions or diffusion are considered [5,6].

In this contribution, we are concerned with the implications of the second assumption or rather its relaxation. The effects of nonuniform mass loading have drawn little attention so far in relation to the vast literature on chemical or biological applications of microacoustic sensors. However, this nonuniformity can be expected to be important in biosensors when larger cells, modeled as point masses, accrete on the sensor surface.

Sauerbrey himself only considered the fundamental mode of AT- or BT-cut quartzes and experimentally investigated the spatial

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dependence of the displacement amplitude by localized deposition of test masses; he did not give a functional expression for the influence of the mass location, though [3].

Van der Steen and coworkers investigated the effect of mass adsorption outside the sensitive electrode area of a QCM, but did not consider a nonuniform (e.g., partial) loading of the electrode area [7].

Dybwad used a simple analogon – two coupled mechanical resonators with lumped masses – to show that the loose adhesion of a point mass lying on a piezoelectric resonator can result in a frequency increase [8]. This coupled-resonator effect can indeed be observed for weekly bonded small particles, but has nothing to do with gravimetric measurement [9].

Later, Vig and Ballato commented with much insight, but only verbally on the effects of nonuniform mass loading and did not give any quantitative details [10].

Today, we are able to investigate the effects of nonunifom mass loading by numerical computations with simultaneous consideration of realistic geometries and anisotropic material parameters. This was shown, for example, by Liu et al. [11,12]. However, such full-fledged solution procedures usually do not further one's basic insight into the existing parameter dependencies. In other words: one needs a simpler model which, nevertheless, covers the dominant phenomena.

Dultsev and Kolosovsky undertook such a model simplification for the case of a point-mass loading of a QCM and concluded that the sensor should respond with a frequency increase [13]. This is surprising as a successive accretion of point masses would then lead to a sequence of positive frequency shifts. It is hardly explainable how the limit of this process, corresponding to an (almost) uniform mass loading, could produce the negative frequency shift required by the Sauerbrey Eq. (1).

The point is not whether microacoustic chemical sensors can respond with a frequency increase – in fact, resonator coupling or elastic loading can lead to such an increase [3,8,9,14] –, but whether they can do so under some exotic form of nonuniform mass loading.

It is the goal of the present contribution to clarify the fundamental effects of the nonuniformity of the mass distribution on the surface of a gravimetric chemical sensor and to reveal the connection between the point-mass loading and the distributed mass loading cases. To this end, we will consider the one-dimensional QCM model presented in Ref. [13]. The simplicity of the model notwithstanding, it will predict the key effects of the nonuniform mass loading and thus enables one to better guide full-fledged simulation efforts based on two- or three-dimensional numerical (e.g., finite-element, or FE) codes. Most numerical codes would fail for very localized (Dirac-like) mass loading anyway and thus cannot be used to investigate this most fundamental of all cases. For instance, Yong et al. [15] used FE calculations to investigate local-mass effects in AT-cut resonators, but did not concentrate the mass to, in linear terms, less than 10% of the resonator diameter.

The 1-d (strip) model of a QCM is shown in Fig. 1. The length L and width w can be chosen such as to make the model predictions match the characteristics of the actual resonator. The length L is very near the electrode diameter as the displacement vibration in the resonator is almost limited to the electrode region (also see Section 2.2 below).

2. Mode matching approach to the problem of nonuniform mass loading

2.1. Basic (uniform) resonator

Assume for the time being that the strip is uniform. We use a Cartesian coordinate system in which the strip length and width are

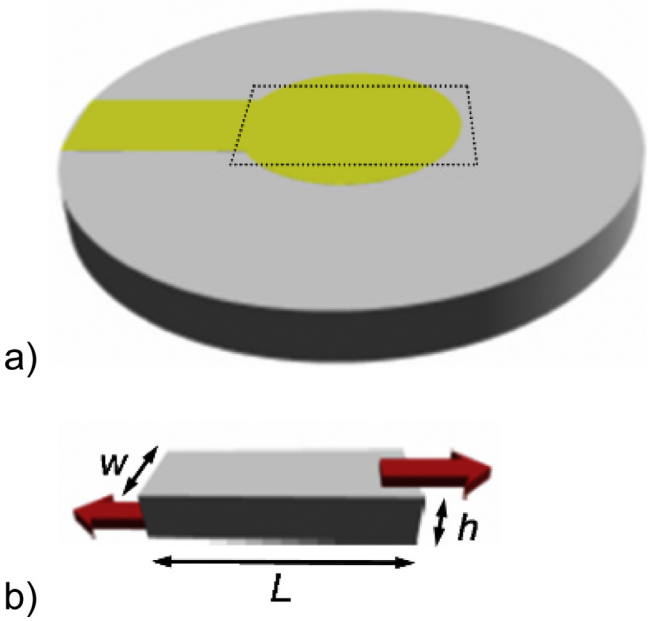

Fig. 1. QCM and equivalent 1-d model. (a) Quartz resonator with typical gold electrode. Another electrode sits on the backside. The electric field between the electrodes leads to mechanical shear vibrations essentially limited to the central area bounded by the dotted lines. (b) One-dimensional strip model equivalent to the vibrating region of (a).

respectively aligned with the x- and y-axis. Consequently, the surface normal points to the z-direction. We are looking for a particle displacement of the form

$$u(x,t) = Re\{\underline{U}(x)\cdot e^{j\omega t}\}. \tag{2}$$

(Note that [13] uses a time-dependence of $e^{-j\omega t}$.) The position-dependent phasor $\underline{U}(x)$ must satisfy the one-dimensional Helmholtz Eq. [(13), Eq. (3); [16] [p. 112], Eqs. (22) and (3)]

$$\frac{d^2\underline{U}}{dx^2} + k^2\underline{U}(x) = 0 \tag{3}$$

with

$$k(\omega) = \frac{\pi}{L}\frac{\omega}{\omega_1} \tag{3a}$$

and

$$\omega_1 = \frac{\pi}{L}\sqrt{\frac{whE}{2\rho_{\ell 0}(1-\sigma^2)}}. \tag{3b}$$

Here, L, w, and h are the strip length, width, and height, respectively. E and σ respectively denote Young's modulus and Poisson's ratio of the strip material, and $\rho_{\ell 0} = M/L$ is the mass per unit length or linear mass density (in x-direction) of the strip (M is the total strip mass). For the uniform resonator considered, ρ_ℓ is a constant independent of x.

The particle displacement associated with the standing wave in a QCM decays fast outside the electrode region [3][3(p. 214), 13, 12]. At the QCM edge and, thus, at the ends of the equivalent strip, it will be zero or very nearly zero, so that Eq. (3) must be solved under the boundary conditions $\underline{U}(0) = \underline{U}(L) = 0\underline{U}(0) = \underline{U}(L) = 0$. There exist infinitely many solutions, the resonance modes. The resonance angular frequency of the n-th mode is determined by $k_nL = n\pi$. This leads to $\omega_n = n\times\omega_1 (n = 1, 2, 3, ...)$. The associated particle displacement is

$$\underline{U}_n(x) = \underline{A}_n \sin(k_n x) = \underline{A}_n \sin(n\frac{\pi x}{L}) \tag{4}$$

with some constant $\underline{A}_n$.