Chapter 2
Methods of Read-Out

Abstract Resonance frequency and resonance bandwidth can be interrogated in three different ways, which are based on oscillator circuits, impedance analysis, and ring down. The techniques are described and compared.
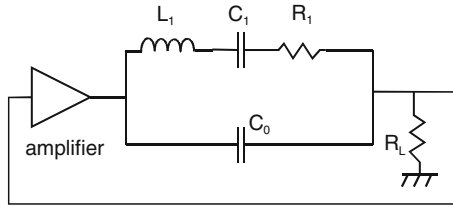
2.1 Oscillator Circuits

QCRs in time and frequency control are always driven by oscillator circuits. An oscillator consists of amplifier and a resonator, where the latter is part of the feed-back loop (Fig. 2.1). A self-sustained oscillation occurs if the so-called Barkhausen condition is fulfilled. The gain of the amplifier must compensate the losses in the feed-back circuit and the phase shift accumulated in the feed-back circuit must be a multiple of 2π (The Barkhausen condition is a necessary, but not a sufficient condition for stable oscillation). An overview of different types of oscillators is given in Chaps. 1 and 5 of Ref. [1]. Oscillator circuits in a wider sense are covered in Ref. [2].

In order to be useful for sensing, oscillator circuits must be combined with frequency counters. Rather than counting the MHz frequency directly, one sometimes down-mixes the signal from the oscillator with a second, stable reference and counts the difference, where the latter typically is in the kHz range.

For sensing in liquids, oscillators have some disadvantages:

- Usually, the circuit is optimized for one particular overtone. Switching between overtones requires additional elements.
- Employing simple oscillator circuits one only obtains the resonance frequency, not the bandwidth. The bandwidth can be determined from the gain of the amplifier needed to drive the resonance because the amplifier's gain compensates the system's losses. Such devices have been built. Other instruments measure the motional resistance, R_1. However, the circuits providing this

Fig. 2.1 In an oscillator circuit, the resonator and an amplifier together form an oscillator. Often the crystal is part of the feed-back circuit. The figure is much simplified. For a more detailed account of oscillator circuits see Ref. [1]. Adapted from Ref. [5]

information are more expensive. Also, the conversion from amplifier gain or motional resistance to resonance bandwidth is not trivial.

- A somewhat subtle problem follows from the fact that the oscillation frequency, f_{osc}, is not necessarily the same as the acoustic resonance frequency, f_r. In this book, the resonance frequency, f_r, is synonymous to the series resonance frequency, f_s (Note: This terminology deviates from other sources. In electronics, f_r often is what is named $f_{\varphi=0}$ in Fig. 2.2. For characteristic frequencies and their definitions see Fig. 9 in Ref. [3]). At the series resonance frequency both the amplitude of oscillation and the resonator's electrical conductance attain its maximum (Fig. 2.2, Sect. 4.5.3). Again, f_s is not strictly equal to the oscillation frequency. The Barkhausen condition is different from the electrical conductance being at its maximum. A difference between f_s and f_{osc} is not a problem as long as it remains constant, regardless of how the properties of the sample change. In sensing, one is only interested in frequency *shifts*. Unfortunately, $f_{osc} - f_s$ *does* depend on both the damping of the resonance and on the parallel capacitance (Sect. 4.5.4). These two parameters usually respond to the presence of a sample. For the parallel capacitance, there are compensation schemes [4], but the issue still requires attention.

These difficulties acknowledged, oscillator circuits may be not only cheaper than the passive schemes like impedance analysis and ring-down, but also more stable [5]. Of course this requires that the damping and the parallel capacitance are under control. Among the reasons is, that impedance analysis and ring-down turn the resonance on and off periodically, thereby introducing variable sources of heat and stress. This does not happen with oscillators. Also, the high-quality oscillators mostly employ the SC-cut, not the AT-cut. The SC-cut is not only temperature-compensated, but also insensitive to certain components of stress. However, the SC-cut is not of the thickness-shear type and can therefore not be used in liquids. More generally, the stability of oscillators has been optimized over many years [6]. Passive interrogation schemes cannot beat the frequency stability achieved in this long-lasting effort. Stability in this context can mean a number of different things, but the oscillator circuits out-perform the passive instruments, regardless of what definition is used.
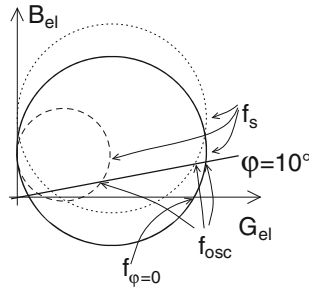
Fig. 2.2 Admittance diagram (see also Sect. 4.5.4). The frequency of oscillation of an oscillator, f_{osc}, is not usually the same as the series resonance frequency, f_s. It was drawn here as one of the two frequencies, where the phase of $\tilde{Y}_{el}$ is $\varphi = 10°$. The details depend on the resonator circuit. f_{osc} often is larger than f_s. The difference between the two frequencies *changes*, when the parallel capacitance increases (*dotted*), and also, when the motional resistance increases (*dashed*). These shifts usually amount to artifacts. The user is mostly interested in f_s

With regard to stability and precision, there is a fundamental difference between the study of complex samples with an advanced QCM, on the one hand, and gravimetric sensing on one single overtone, on the other. When studying complex samples, one fits a model with certain parameters to the shifts of frequency and bandwidth. Most of the time, the systematic errors are larger than the statistical errors and the accuracy of the results therefore does not improve much when the scatter on the individual frequency readings improves. Gravimetric sensing on one overtone is different. Here, the repeatability of this one frequency sets the limit of detection. If the damping and the parallel capacitance are under control, a good oscillator circuit then is the method of choice.

2.2 Impedance Analysis

Mapping out the electrical admittance of the resonator as a function of frequency is the most direct and transparent way of interrogation [8, 9]. An impedance analyzer sweeps the frequency across the resonance and measures the resonator's electrical admittance, $\tilde{Y}_{el}(\omega)$. There are different ways to connect the crystal to the analyzer. Reflectometry as shown in Fig. 2.3a is particularly easy. Reflectometry is efficient if the impedance of the device under test is in the range of 50 Ω. The resonator's impedance is calculated from the reflectivity and the impedance of the cable (often 50 Ω), making use of Eq. 4.2.11. In liquids, the impedance of resonator usually is much larger than 50 Ω. The series-through configuration (Fig. 2.3b) then yields more precise values of the device's impedance than reflectometry. Note: When wiring the resonator in the series-through mode, one needs to worry about grounding of the front electrode (see also Fig. 14.2b).

(a)   reflection
bridge

network
analyzer

(b)
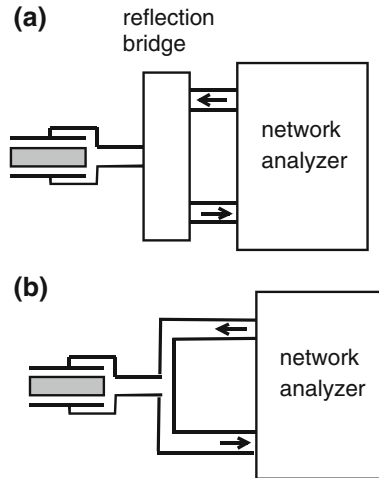
network
analyzer

Fig. 2.3 In impedance analysis, a network analyzer (also: "impedance analyzer") determines the complex electrical admittance of the crystal. **a** The crystal is wired such that the measurement occurs in reflection. In that case, the resonator's electrical impedance is calculated from the reflected electrical amplitude by an equation analogous to Eq. 4.2.11. **b** A resonator wired in the series-through mode. Reference [7] discusses the comparative advantages of the different electrical configurations

At the resonance frequency, the real part of the electrical admittance displays a maximum. The corresponding frequency is the series resonance frequency, f_s. On resonance, the electrodes draw a large current in order to compensate the large piezoelectrically-induced polarization. There is a phase shift between current and voltage. The admittance, $\tilde{Y}_{el}(\omega) = G_{el}(\omega) + iB_{el}(\omega)$ with G_{el} the "conductance" and B_{el} the "susceptance" therefore is complex (Fig. 2.4). We come back to the exact dependence of G_{el} and B_{el} on frequency in the context of the 4-element circuit (Sect. 4.5.4).

Impedance analysis is particularly advantageous if the experimental configuration is complicated in one way or another. For instance, if there are long cables or if anharmonic sidebands interfere with the measurement, the user can recognize and diagnose potential problems based on the admittance curve. For an example see Fig. 7.3b. Oscillator circuits under such conditions often just stop to oscillate (or, worse, drift in frequency, although they should not) and the user cannot easily understand why this happens.

There are various ways to improve on impedance analysis, for instance by adding elements compensating for the parallel capacitance [10], by smart schemes of interrogation [11], and by detection schemes which are intermediate between oscillators and impedance analysis. There is also considerable activity in making impedance analyzers cheaper, possibly allowing for multichannel devices [12–14]. For a recent review see Ref. [15].
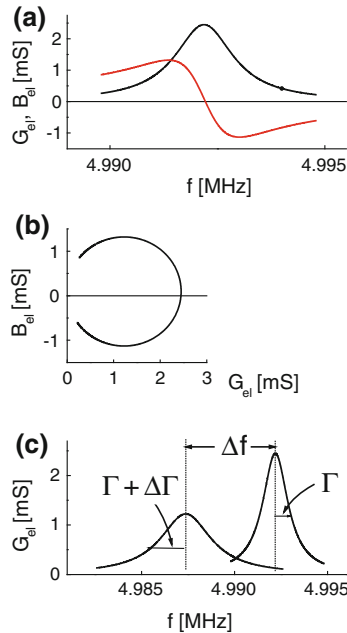
Fig. 2.4 Impedance analysis is based on the admittance curve as shown in panel (**a**). The electrical admittance, $\bar{Y}_{el} = G_{el} + iB_{el}$, is the inverse of the electrical impedance, $\bar{Z}_{el}$, where both the impedance and the admittance are complex (Sect. 4.5.4). One typically analyzes the admittance curve because its real part (the conductance, $G_{el}(\omega)$) forms the well-known, symmetric resonance curve. $G_{el}(\omega)$ is at its maximum at the series resonance frequency. The polar diagram of the admittance curve shows a circle (**b**). Of primary interest in sensing are the shifts of resonance frequency Δf and half-bandwidth, $\Delta\Gamma$ (**c**). Panel **c** only shows the conductance curves for clarity. The imaginary part of the admittance curve (the susceptance, $B_{el}(\omega)$) displays the same shift and the same broadening

2.3   Ring-Down

Ring-down experiments are carried out in many physics undergraduate laboratories. The central piece of equipment is a pendulum, which the students set in motion by hand. They count the number of oscillation cycles per minute and measure the time which it takes, until the amplitude has decayed to half its original value. Sometimes, they also check whether the oscillation period depends on the excursion angle. It does (slightly) because the pendulum is a slightly nonlinear device. Nonlinear behavior is ignored in the following. Similar setups exist, where the conventional pendulum is replaced by spring-driven pendulum (often a torsion pendulum). From the resonance frequency and the known mass (or the known moment of inertia) the students infer the elastic stiffness of the spring. From the decay time they infer the loss modulus (Sect. 3.7).
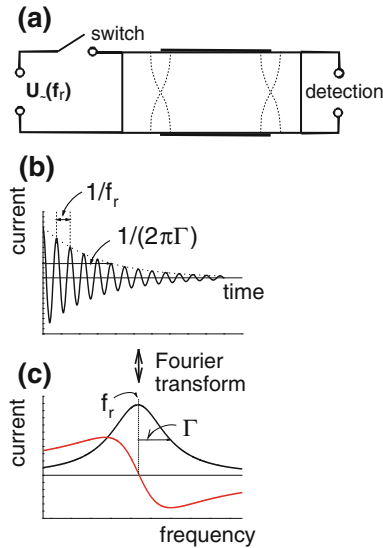
Fig. 2.5 In ring-down, the resonator is excited by an RF-pulse, the frequency of which is close to the expected resonance frequency [18]. In a second step, the excitation is shut off and the oscillation is allowed to decay (**a**). The current into electrodes is recorded. The decay time (the time when the envelope has decayed by a factor of e) is equal to $1/(2\pi\Gamma)$ (**b**). The decaying cosine observed in ring-down is the Fourier transform of the resonance curve as observed with impedance analysis (**c**)

Ring-down is conceptually simple and was employed in acoustic sensing as early as 1954 (Ref. [16], see also Ref. [17]). Q-Sense has commercialized an advanced QCM based on ring-down [18]. It is the QCM-D, where "-D" stands for "with Dissipation Monitoring". The term QCM-D is a trademark owned by Q-Sense. Excitation occurs with a radio-frequency-pulse (RF pulse) having a frequency matching the expected resonance frequency (Fig. 2.5a). Ring-down is called "impulse excitation" in Refs. [19] and [20]. Q-Sense sometimes calls the process "pinging". Once the excitation is turned off, the resonance decays freely. The current into the electrodes is recorded digitally (Fig. 2.5b). (The process of interrogation actually is slightly more complicated, but the details are unessential). The resonance frequency and the decay time follow from fitting the current-versus-time trace with a decaying cosine. Importantly, the information obtained from ring-down is equivalent to the information derived from impedance analysis. The dissipation factor, D, is equal to $2\Gamma/f_r$.

Ring-down can be faster than impedance analysis. The acquisition time per data point can be as low as a few times the inverse half-bandwidth, Γ^{-1}, while it takes about one second to sweep the frequency across the resonance in impedance analysis. Some of this speed is lost in an averaging process, but the user can trade precision against data acquisition rate. This trade-off is more difficult in impedance analysis.

Ring-down deserves a side remark because it is also widely practiced in nuclear magnetic resonance (NMR), where it is called "free induction decay". There is a difference though. In NMR, many resonances (many spins in different environments, having different Larmor frequencies) are excited at the same time by application of an RF-pulse, which is sufficiently broad in frequency to cover all resonances of interest. The data trace (the free induction decay) is a super-position of all decays originating from the different spins. The spectrum is retrieved from the decay by Fourier transformation. The Fourier transform of the decay contains many different lines. "Induction" here is magnetic induction, sometimes also called magnetic polarization. It is the equivalent of the current into the electrodes of a piezoelectric resonator. Ring-down as implemented in the QCM-D is slightly different from free-induction decay. In the QCM-D, excitation occurs with a narrow RF-pulse, selectively exciting the one resonance of interest. Of course an approximate a priori knowledge of the resonance frequency is needed to do this, but this is not a problem in practice because the resonance frequency changes slowly. The reason to not excite all resonances at the same time is that out of the very many resonances of the crystal only a few are of interest. These are the 01-modes (Sect. 7.2). It is more efficient to only excite the resonance of interest and to repeat this process for the different overtones, than it is to excite all resonances at the same time and to disentangle them by Fourier transformation. (Broadband excitation of all resonance has been recently reported by Resa et al. [21]; see the reference for details.)

Ring-down and impedance analysis are equivalent, in principle, but of course there are differences. Frequency-domain data (as acquired in impedance analysis) and time-domain data (as acquired in ring-down) are strictly equivalent to each other as long as the system obeys linear response. However, the QCM is a slightly nonlinear device and the consequences can be observed in both impedance analysis and ring-down if one looks closely enough. The consequences are not strictly the same in both types of interrogation. A second set of differences is linked to noise sources, errors, and error propagation. When stating that ring-down and impedance analysis were equivalent, we simplified the matter a little bit.

Glossary

Variable	Definition (Comments)
B_{el}	Electrical susceptance
C_0	Parallel capacitance (see Sect. 4.5.3)
C_1	Motional capacitance (see Sect. 4.5.3)
el	As a subscript: electrical
f	Frequency

f_{osc}	Oscillation frequency
f_r	Resonance frequency
f_s	Series resonance frequency (same as resonance frequency in this book)
G_{el}	Electrical conductance
L_1	Motional inductance (see Sect. 4.5.3)
R_1	Motional resistance (see Sect. 4.5.3)
R_L	Load resistance
t	Time
$U_\sim$	AC-voltage
$\tilde{Y}_{el}$	Electrical admittance ($\tilde{Y}_{el} = G_{el} + iB_{el}$)
$\tilde{Z}_{el}$	Electrical impedance ($\tilde{Z}_{el} = 1/\tilde{Y}_{el}$)
Δ	As a prefix: a shift induced by the presence of a sample
Δf	Shift of resonance frequency (might have been called Δf_r; the index r was dropped for brevity)
$\Delta\Gamma$	Shift of the half-bandwidth (might have been called $\Delta\Gamma_r$; the index r was dropped for brevity)
Γ	Half-bandwidth ($2\pi\Gamma$: decay rate in a ring-down experiment)
ω	Angular frequency

References

1. Arnau, A.: Piezoelectric Transducers and Applications. Springer, Heidelberg (2004)
2. Gottlieb, I.M.: Practical Oscillator Handbook. Newnes, Boston (1997)
3. http://txccrystal.com/term.html. Accessed 10 Aug 2013
4. http://www.thinksrs.com/downloads/PDFs/Manuals/QCM200m.pdf. Accessed 14 Feb 2013
5. Montagut, Y.J., Garcia, J.V., Jimenez, Y., March, C., Montoya, A., Arnau, A.: Frequency-shift versus phase-shift characterization of in-liquid quartz crystal microbalance applications. Rev. Sci. Instrum. **82**(6), 064702 (2011)
6. Rubiola, E., Giordano, V.: On the 1/f frequency noise in ultra-stable quartz oscillators. IEEE Trans. Ultrason. Ferroelectr. Freq. Control **54**(1), 15–22 (2007)
7. http://cp.literature.agilent.com/litweb/pdf/5988-0728EN.pdf. Accessed 14 Feb 2013
8. Beck, R., Pittermann, U., Weil, K.G.: Impedance Analysis of Quartz Oscillators, Contacted on One Side with a Liquid. Ber. Bunsen Phys. Chem. Chem. Phys. **92**(11), 1363–1368 (1988)
9. Zimmermann, B., Lucklum, R., Hauptmann, P., Rabe, J., Buttgenbach, S.: Electrical characterisation of high-frequency thickness-shear-mode resonators by impedance analysis. Sens. Actuators B Chem. **76**(1–3), 47–57 (2001)

10. Calvo, E.J., Etchenique, R., Bartlett, P.N., Singhal, K., Santamaria, C.: Quartz crystal impedance studies at 10 MHz of viscoelastic liquids and films. Faraday Discuss. **107**, 141–157 (1997)
11. Kankare, J., Loilkas, K., Salomaki, M.: Method for measuring the losses and loading of a quartz crystal microbalance. Anal. Chem. **78**(6), 1875–1882 (2006)
12. Auge, J., Dierks, K., Eichelbaum, F., Hauptmann, P.: High-speed multi-parameter data acquisition and web-based remote access to resonant sensors and sensor arrays. Sens. Actuators B Chem. **95**(1–3), 32–38 (2003)
13. http://n2pk.com/. Accessed 29 Nov 2013, http://www.makarov.ca/vna.htm. Accessed 29 Nov 2013
14. http://sdr-kits.net/VNWA3_Description.html. Accessed 29 Nov 2013
15. Arnau, A.: A review of interface electronic systems for AT-cut quartz crystal microbalance applications in liquids. Sensors **8**(1), 370–411 (2008)
16. Sittel, K., Rouse, P.E., Bailey, E.D.: Method for determining the viscoelastic properties of dilute polymer solutions at audio-frequencies. J. Appl. Phys. **25**(10), 1312–1320 (1954)
17. Hirao, M., Ogi, H., Fukuoka, H.: Resonance emat system for acoustoelastic stress measurement in sheet metals. Rev. Sci. Instrum. **64**(11), 3198–3205 (1993)
18. Rodahl, M., Hook, F., Krozer, A., Brzezinski, P., Kasemo, B.: Quartz-crystal microbalance setup for frequency and q-factor measurements in gaseous and liquid environments. Rev. Sci. Instrum. **66**(7), 3924–3930 (1995)
19. http://en.wikipedia.org/wiki/Impulse_excitation_technique. Accessed 14 Apr 2013
20. Lucklum, R., Eichelbaum, F.: Interface circuits for QCM sensors. In: Steinem, C., Janshoff, A. (eds.) Piezoelectric Sensors, vol. 5, pp. 3–47. Springer, New York (2007)
21. Resa, P., Castro, P., Rodriguez-Lopez, J., Elvira, L.: Broadband spike excitation method for in-liquid QCM sensors. Sens. Actuators B Chem. **166**, 275–280 (2012)

The Quartz Crystal Microbalance in Soft Matter Research
Fundamentals and Modeling
Johannsmann, D.
2015, XX, 387 p. 123 illus., 9 illus. in color., Hardcover
ISBN: 978-3-319-07835-9