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# The role of lock-in phase setting in ac susceptibility measurement

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Method in which lock-in detectors process signals from standard ac susceptometers, particularly in the detection of higher harmonics, is analyzed in details. The exact formulas have been derived and checked experimentally by measurements on soft ferromagnetic sample, using several available lock-in amplifiers. The reasons why the proper phase adjustment has to be implemented in the protocol of higher harmonics measurements have been elaborated. The procedure of the lock-in phase adjustment is described, enabling separation of Fourier or Taylor components of hysteretic ac susceptibility into real and imaginary sectors. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4807752]

## I. THE PROBLEM LESS TACKLED

In understanding the ac magnetic susceptibility measurements there are three most relevant elements:

i. time varying magnetic field, usually pure harmonic $H = H_0\cos(\omega t)$, is generated by some primary coil;
ii. time varying magnetization of the sample, as a response to the latter field; $M = M(H(t))$;
iii. ac voltage induced in the secondary coil(s) proportional to $-dM/dt$. As it is well known and further elaborated later on, induced voltage contains the information on susceptibility of the sample.

In conventional ac susceptometer,[1] which usually consists of a long primary and two spaced, oppositely connected and well balanced secondary coils, induced voltage is of the form

$$V = -\mu_0 \alpha G N_S \frac{dM}{dt}, \tag{1}$$

where $\mu_0$ is the vacuum permeability, $\alpha G$ is the calibration constant typical for a particular device, usually separated in a part dependent on geometry of coils ($\alpha$) and a part dependent on the quantity of sample ($G$), $N_S$ is the number of turns of the secondary coil; altogether, the prefactors can be abbreviated by $C_S$.

In this article we consider a particular situation in which the induced voltage is directly measured by the two-channel phase sensitive detector (PSD or lock-in), such as the digital Ametek/Signal Recovery 7265, Stanford Research 830, or the older analog PAR 5210. These digital lock-ins can measure amplitudes of the nth Fourier component of the signal connected to the input. Analog lock-in can do measurement in one of the available filter-employing modes (BANDPASS, LOPASS) in which the contribution close to or below a chosen frequency can be extracted out or (in NOTCH mode) filtered out of the input signal, or in the FLAT mode in which it bypasses any filtering.

In order to acquire full command over ac susceptibility measurements, as performed by PSD, the role of these three elements needs to be clarified in all necessary details.

The driving field is produced by connecting an ac source to the primary coil. In most cases the lock-in's internal oscillator output voltage is used but any other source can be used as well – in the latter case lock-in reference has to be locked to this external source. The driving field can be generally written as $H = H_0 \cos(\varphi + \varphi_S)$, where $\varphi_S$ is the unknown phase shift referred to the phase to which the lock-in is synchronized and $\varphi = \omega t$. (Hereafter, explicit time dependence is represented by phase dependence. Note that there is always a phase shift between the voltage applied to the primary coil and the current passing through it due to reactance of the primary coil.)

Generally, the magnetization of the sample is a nonlinear function of the field, lagging behind the driving field. Nonlinearity and phase lag of the magnetization can be described formally in different ways; the form appropriate for our discussion is the Fourier expansion:

$$M(\varphi) = M_0 + \sum_{n=1}^{\infty} \{A_n \cos[n(\varphi + \varphi_S)] + B_n \sin[n(\varphi + \varphi_S)]\}. \tag{2}$$

By convention, $A_n$ and $B_n$ can be written as $\chi_n' H_0$ and $\chi_n'' H_0$, respectively, where $\chi_n'$ and $\chi_n''$ are called real and imaginary parts of the nth susceptibility component. ($\chi_n'$ and $\chi_n''$ are components in phase and orthogonal to the driving field, respectively.)

If the magnetization of the sample is given by Eq. (2) then induced voltage, Eq. (1), reads

$$V(\varphi) = C_S \omega H_0 \sum_{n=1}^{\infty} n\{\chi_n' \sin[n(\varphi + \varphi_S)] - \chi_n'' \cos[n(\varphi + \varphi_S)]\}$$
$$= \sum_{n=1}^{\infty} V_n' \sin[n(\varphi + \varphi_S)] + V_n'' \cos[n(\varphi + \varphi_S)]. \tag{3}$$

By connecting this voltage to the two-channel PSD the user expects that voltages, appeared in the channels, give real and imaginary components separated (for any harmonic). But a trivial fact is that separation of input voltage into lock-in channels strongly depends on lock-in phase setting. So, a very important experimental question is the following: if a digital lock-in is set to measure the nth harmonic of induced voltage, Eq. (3), how does one determine the lock-in's referent phase in order to separate real and imaginary components of the susceptibility? And a more important one: does the latter setting remain the same for all Fourier components?

 

These questions are, in fact, related to a specific way in which the lock-in processes the signal. In the so-called phase mixer the input signal is multiplied by the phase shifted nth harmonic of the referent signal and then this product is passed through a LowPass filter. This process (which results with the corresponding voltages obtained in two lock-in channels) can be mathematically described by the relation

$$V_n^{x,y} = C_{\text{int}} \int V_{in}(\varphi) V_{ref}^{x,y}(\varphi) d\varphi.$$

The form of referent voltage is a source of dispute here. If digital lock-in is set to measure harmonic components of the input signal higher than one, it is not clear whether the latter, phase shifted, nth harmonic of the referent signal is of the form

$$V_{ref}^{x}(\varphi) = \sin(n\varphi + \varphi_D) \tag{4a}$$

or (as is expected from simple mathematical Fourier analysis)

$$V_{ref}^{x}(\varphi) = \sin[n(\varphi + \varphi_D)] \tag{4b}$$

(hereby, $V_{ref}^{y}(\varphi) = V_{ref}^{x}(\varphi + \pi/2)$, while $\varphi_D$ represents the lock-in's referent phase setting). In the user-accessible lock-in documentation nothing is mentioned about these alternatives. On the other side, in most articles referring to results on "higher susceptibility components $\chi_n'$, $\chi_n''$," that do pay attention to the problem of proper phase, it is implicitly assumed that the phase setting for the first harmonic remains the same for all higher ones and that this common choice of phase setting will put $\chi_n'$ in the first channel and $\chi_n''$ in the second channel, thus suggesting that the latter form, Eq. (4b), is valid.

However, our experimental insight suggests that this may not be true: We have verified that only a specific Fourier sequence, particularly of the form

$$V(\varphi) \sim \sum_{n=1}^{\infty} \left[ V_n^x \sin(n\varphi + \varphi_D) + V_n^y \cos(n\varphi + \varphi_D) \right],$$

is able to reproduce the full shape of the time dependent multiharmonic signal applied to the lock-in input. $V_n^x$ and $V_n^y$ are signals measured in the first and the second channel, respectively, as obtained by keeping the same lock-in phase setting. This form suggests that there could be mixing of the susceptibility components in measured voltages if the lock-in phase is kept fixed for different harmonics.

Therefore, we decided to test experimentally which one of the options in Eqs. (4) is really applied in the lock-in processing. The calculations were done by assuming that the option, Eq. (4a), for the referent signal is true. To test the alternative option, Eq. (4b), it would be enough to replace $\varphi_D$ with $n\varphi_D$ in final formulas.

So, our starting assumption was that the voltages reported in the two channels of the lock-in, set to measure nth harmonic, are actually

$$V_n^x = C_{\text{int}} \int V_{in}(\varphi) \sin(n\varphi + \varphi_D) d\varphi, \quad \varphi = \omega t, \tag{5a}$$

$$V_n^y = V_n^x \left( \varphi_D + \frac{\pi}{2} \right), \tag{5b}$$

where $C_{\text{int}}$ is the integration constant equal to $1/(\pi\sqrt{2})$ in digital lock-in detection.

## II. EXPERIMENTAL VERIFICATION

### A. Measurement of Fourier components of the susceptibility by digital lock-in

If the induced voltage, Eq. (3), is applied to formulas (5), one obtains

$$V_n^x = C_S \frac{n\omega H_0}{\sqrt{2}} [\chi_n' \cos(n\varphi_S - \varphi_D) + \chi_n'' \sin(n\varphi_S - \varphi_D)]$$
$$= \frac{1}{\sqrt{2}} [V_n' \cos(n\varphi_S - \varphi_D) + V_n'' \sin(n\varphi_S - \varphi_D)], \tag{6a}$$

$$V_n^y = C_S \frac{n\omega H_0}{\sqrt{2}} [\chi_n' \sin(n\varphi_S - \varphi_D) - \chi_n'' \cos(n\varphi_S - \varphi_D)]$$
$$= \frac{1}{\sqrt{2}} \left[ V_n' \sin(n\varphi_S - \varphi_D) - V_n'' \cos(n\varphi_S - \varphi_D) \right]. \tag{6b}$$

These expressions immediately tell that lock-in phase setting, for a decomposition of an arbitrary nth component into real and imaginary part (i.e., making $\sin(n\varphi_S - \varphi_D) = 0$, $\varphi_D = n\varphi_S$), should be n-dependent. At variance, if Eq. (4b) were correct, then it would be enough to replace $\varphi_D$ in Eqs. (6) with $n\varphi_D$; the decomposition would be n-independent now because $\sin n(\varphi_S - \varphi_D) = 0$, $\varphi_D = \varphi_S$.

We propose two ways to test validity of Eqs. (6) or its alternatives. In both cases we need an induced multiharmonic voltage in the pick-up coils. The first way is to measure the voltage in one channel as a function of lock-in phase through entire cycle for a few different source phases and to do that for a few harmonic components. The behavior of those curves for options (4a) or (4b) would be different. The second way is to record induced voltage independently of lock-in measurement and calculate numerically its Fourier components and then to measure the same voltage by lock-in, trying to see whether the same values as from fit can be obtained with lock-in phase setting n-dependent or not.

The sample used was the one from the ferromagnetic amorphous alloy series $Fe_X Ni_{80-X} B_{18} Si_2$ with x = 5. This particular sample has the Curie temperature around 59 K. Below this temperature it is a soft ferromagnet with coercive field of order of a few Oe. Although we could use the own lock-in output, we preferred the use of the Keithley ac current source 6221 and its SINE OUT to feed the primary coil of our ac susceptometer.[2] This current source is able to arbitrarily shift its own TTL signal relative to its own SINE OUT signal. In our measurements the lock-in was synchronized to the latter TTL signal. So, for sample below the Curie temperature, we can change harmonic composition of $V_{in}$ in Eqs. (5) by changing sine out level. By changing the TTL position we can also shift that signal relative to the lock-in phase setting and in that way change the source phase.

The scan of induced voltage was done by high-speed, 12-bit resolution digitizer card NI 6111. The driving frequency was 231 Hz, field amplitude 22 Oe, and sampling frequency 1 MHz. The pattern of voltage as the function of time

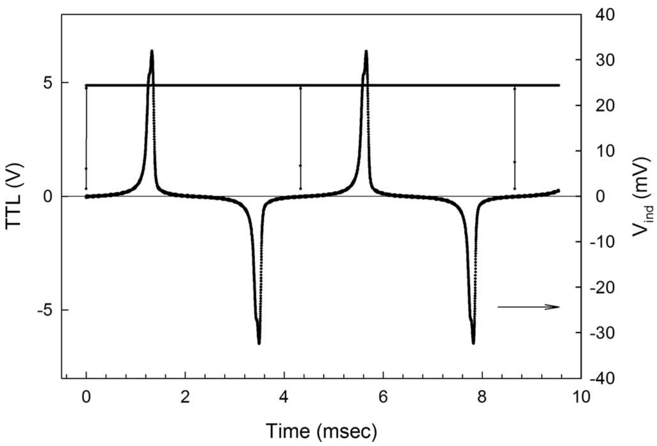


FIG. 1. Voltage induced in secondary coils and corresponding TTL signal as function of time.

on T = 4.2 K, obtained as an average of 100 scans, is given in Fig. 1.

For the first test we measured odd harmonics $V^{y}_{2n+1}$ on our sample as a function of detector phase through entire cycle for a few different source phases (the component of Earth magnetic field in direction of driving field was cancelled and, therefore, even harmonics vanishes, $V^{x,y}_{2n} = 0$). If Eq. (4a) is true, then the zeroes of this voltage should appear, following Eqs. (6) at

$$\varphi_D = (2n+1)\varphi_S - atn\left(\frac{\chi''_{2n+1}}{\chi'_{2n+1}}\right). \qquad (7a)$$

Alternatively, if Eq. (4b) is true then zeroes should appear at

$$\varphi_D = \varphi_S - \frac{1}{2n+1} atn\left(\frac{\chi''_{2n+1}}{\chi'_{2n+1}}\right). \qquad (7b)$$

We see that the behavior of the span between zeroes of different harmonics is decisive. In the first case it will increase exactly proportional to $n$ and in the second case it will decrease with $n$.

In Fig. 2 we show the dependence of the first three odd harmonics on detector phase for four different source phases (in fact, position of TTL signal on Keithley 6221). First position was arbitrary and then increased for 10°, 20°, and 30°. In all three cases the zeroes move in the direction of change of $\varphi_S$ and the span between zeroes of harmonics increases with $(2n+1)\varphi_S$. This finding shows that Eq. (7a) is true, which strongly supports the option in Eq. (4a) and the validity of Eqs. (5).

Now we proceed with detailed examination of Fourier analysis of induced voltage from Fig. 1. First we calculate Fourier components taking the points within TTL markers as the period. Next, we measure the same voltage by lock-in manipulating the phase in a way that the sin terms in Eqs. (6) vanish. We did it in two ways. In both ways we used the fact that phase of real susceptibility is the same as that of the empty coil (real susceptibility just changes the amount of induced voltage, not the phase). First, we set the lock-in phase $\varphi_D$ to zero and set TTL marker such that complete positive voltage of empty secondary coil (in the first harmonic) is positioned in the first channel only (formally $\varphi_S = 0$). That will remove

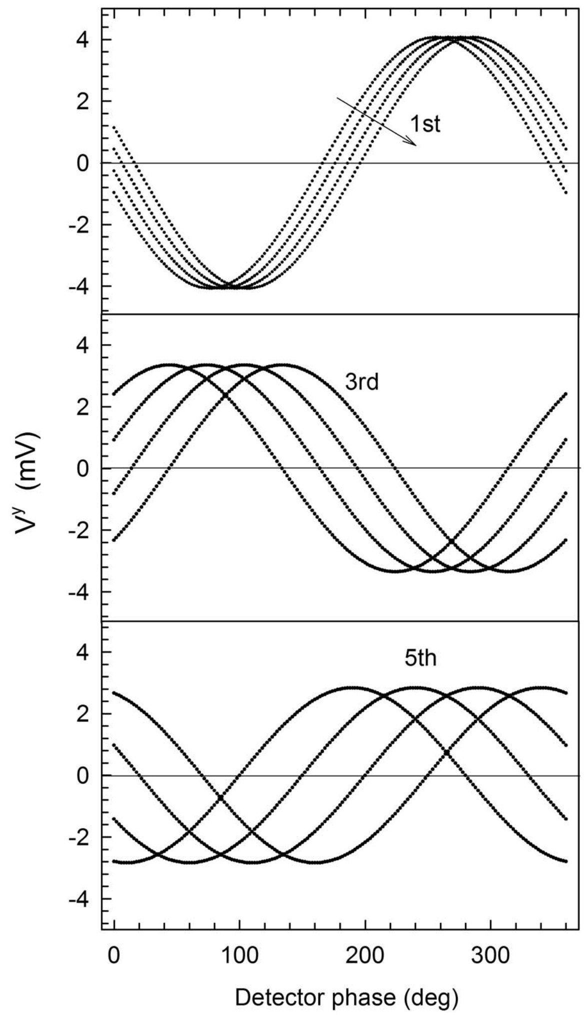


FIG. 2. 1st, 3rd, and 5th even component of induced voltage for arbitrary source phase and phase shifted for 10°, 20°, and 30° as a function of detector phase. Arrow shows shift of source phase, same for all three harmonics.

any phase dependence from Eqs. (6) for any harmonic and the voltages measured in two channels will be proportional to real and imaginary parts of nth susceptibility component. In the second way, lock-in is again set to measure empty secondary coil voltage in the first harmonic and lock-in phase is found such that complete positive voltage is in the first channel only (AUTOPHASE command[3] of the lock-in can be used). That phase is, in fact, equal to the source phase, $\varphi^1_D = \varphi_S$. Now, for nth harmonic measurement it is necessary to set $\varphi_D = n\varphi^1_D$. This will give us the sin terms equal to 0 and the cos terms equal to 1, thus decoupling the components. The voltages measured in these two ways completely coincide for any harmonic. They are

$$V^x_n = C_S \frac{n\omega H_0}{\sqrt{2}} \chi'_n, \qquad (8a)$$

$$V^y_n = -C_S \frac{n\omega H_0}{\sqrt{2}} \chi''_n. \qquad (8b)$$

In Fig. 3 we plot the first 29 odd harmonics measured by lock-in (full symbols) and those calculated from the digitized voltage (open symbols). The latter data were additionally divided

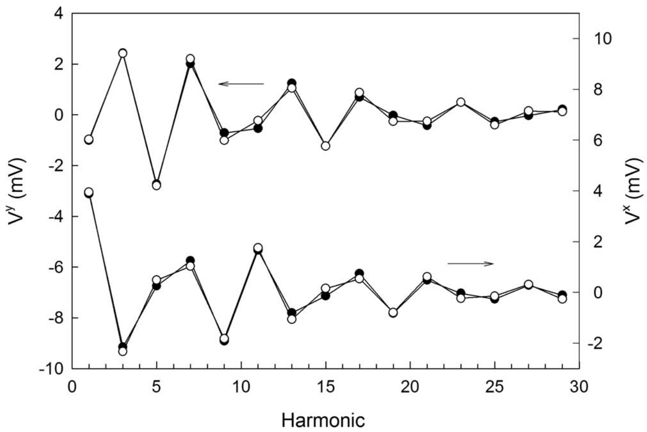


FIG. 3. RMS amplitudes of real (down) and imaginary (up) Fourier components of induced voltage obtained from fit of digitized voltage (full dots) and directly measured on Ametek 7265 lock-in (empty dots).

by the RMS normalization factor. Obviously, the agreement between the two sets of data provides a clear proof that our basic assumption, Eqs. (5), is correct.

We conclude that formulas (6) indeed provide the voltages measured in the two lock-in channels. It is clear that keeping the detector phase fixed would not separate real and imaginary components of all harmonics.

## B. Measurement of Taylor components of the susceptibility by digital lock-in

The above conclusions have important consequence to another representation of hysteresis: the Taylor expansion. This representation is of interest when it is important to extract components of the susceptibility that depend on the powers of the field:

$$M_{\pm} = \pm M_0 + \sum_{k=0}^{\infty} \chi_{2k+1} H^{2k+1} \pm \sum_{k=1}^{\infty} \chi_{2k} H^{2k}. \tag{9}$$

$M_+$ is the magnetization branch for decreasing field and $M_-$ for increasing field. This form follows from the request $M(-H) = -M(H)$ (driving field is again of the form $H = H_0 \cos(\varphi + \varphi_S)$). Voltage measured in the first channel now reads

$$V_n^x = C_S C_{\text{int}} \left[ \int_{\varphi_S}^{\pi-\varphi_S} \left( -\frac{dM_+}{dt} \right) \sin(n\varphi + \varphi_D) d\varphi + \int_{\pi-\varphi_S}^{2\pi-\varphi_S} \left( -\frac{dM_-}{dt} \right) \sin(n\varphi + \varphi_D) d\varphi \right]. \tag{10}$$

The result, after lengthy but otherwise straightforward calculations, is

$$V_{2n+1}^x = C_S C_{\text{int}} \omega \left\{ M_{2n+1} \cos[(2n+1)\varphi_S - \varphi_D] - N_{2n+1} \sin[(2n+1)\varphi_S - \varphi_D] \right\}, \tag{11}$$

$$V_{2n+1}^y = C_S C_{\text{int}} \omega \left\{ N_{2n+1} \cos[(2n+1)\varphi_S - \varphi_D] + M_{2n+1} \sin[(2n+1)\varphi_S - \varphi_D] \right\}, \tag{12}$$

$$M_{2n+1} = \pi \frac{2n+1}{2^{2n}} \sum_{k=0}^{\infty} \frac{1}{2^{2k}} \binom{2n+1+k}{k} \chi_{2n+1+2k} H_0^{2n+1+2k},$$

$$N_{2n+1} = 2 \sum_{k=1}^{\infty} \frac{1}{2^{2(k-1)}} \times \left( \sum_{j=0}^{2k} \frac{k-j}{2n+1+2k-2j} \binom{2k}{j} \right) \chi_{2k} H_0^{2k}.$$

The same procedure with the phase adjustment as described above has to be applied to obtain even and odd terms separated in two channels:

$$V_{2n+1}^x = C_S C_{\text{int}} \omega M_{2n+1} = C_S C_{\text{int}} \frac{2n+1}{2^{2n}} \pi \omega \times \sum_{k=0}^{\infty} \frac{1}{2^{2k}} \binom{2n+1+k}{k} \chi_{2n+1+2k} H_0^{2n+1+2k}, \tag{13}$$

$$V_{2n+1}^y = C_S C_{\text{int}} \omega N_{2n+1} = C_S C_{\text{int}} 2\omega \sum_{k=1}^{\infty} \frac{1}{2^{2(k-1)}} \times \left( \sum_{j=0}^{2k} \frac{k-j}{2n+1+2k-2j} \binom{2k}{j} \right) \chi_{2k} H_0^{2k}. \tag{14}$$

The fact that $V_{2n+1}^x$ comprises components of (2n + 1)th and higher odd orders of the susceptibility enables finding the inverse relation for odd components of the susceptibility. After a few iterations the relation inverse to Eq. (13) is obtained as

$$\chi_{2n+1} = \frac{1}{C_S C_{\text{int}} \pi \omega} \frac{1}{H_0^{2n+1}} \frac{2^{2n}}{2n+1} \times \sum_{j=0}^{\infty} (-1)^j \binom{2n+j}{j} V_{2n+1+2j}^x. \tag{15}$$

This equation shows that, in cases when any nonlinearity in magnetization curve is expected (on phase transitions it is true even for small driving field), it is necessary to measure more harmonics to extract even linear behavior. Up to field and calibration factor, linear and cubic terms should be obtained by measuring a number of Fourier components and composed according to Eq. (15):

$$\chi_1 \sim \left( V_1^x - V_3^x + V_5^x - V_7^x + V_9^x - \cdots \right),$$

$$\chi_3 \sim \frac{4}{3} \left( V_3^x - 3V_5^x + 6V_7^x - 10V_9^x + \cdots \right).$$

## C. Measurement of Taylor components of the susceptibility by analog lock-in

We have also measured the same sample by an analog lock-in (PAR 5210). As this lock-in multiplies input signal by square voltage shifted in phase for $\varphi_D$ it is clear that it, in fact, integrates the derivative of the magnetization and that the result could be numerically related to magnetization. The voltages in the two channels are

$$V_x = C_S C_{\rm int} \int \left(-\frac{dM}{dt}\right) \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{\sin(2n+1)(\varphi+\varphi_D)}{2n+1} d\varphi,$$

$$V_y = V_x \left(\varphi_D + \frac{\pi}{2}\right),$$

and $C_{\rm int}$ for this lock-in is equal to $-1/(4\sqrt{2})$.

The final result is

$$V_x = 4C_S C_{\rm int}\omega \Bigg\{ \sum_{k=0}^{\infty} \chi_{2k+1} H_0^{2k+1} \cos^{2k+1}(\varphi_S - \varphi_D) - \sum_{k=1}^{\infty} \chi_{2k} H_0^{2k} \left[1 - \cos^{2k}(\varphi_S - \varphi_D)\right] \times \frac{4}{\pi} \sum_{k=0}^{\infty} \frac{1}{2k+1} \sin\left[(2k+1)(\varphi_S - \varphi_D)\right] \Bigg\}. \quad (16)$$

The last sum in Eq. (16) represents a square signal which just takes care of the sign of the second factor. Also, due to hysteresis symmetry, $M_+(H_{\max}) = M_-(H_{\max})$, it follows that

$$\sum_{k=1}^{\infty} \chi_{2k} H_0^{2k} = -M_0.$$

It is clear that in Eq. (16) the two sums represent odd and even components of magnetization in the $(\varphi_S - \varphi_D)$ point.

Thus, the analog lock-in PAR 5210, operating in FLAT mode, in fact gives the magnetization at a particular value, dependent on the phase setting. The voltages measured in the two channels can be rewritten in compact form

$$V_x = \frac{1}{\sqrt{2}} C_S \omega M(\varphi_S - \varphi_D), \quad (17a)$$

$$V_y = \frac{1}{\sqrt{2}} C_S \omega M\left(\varphi_S - \varphi_D - \frac{\pi}{2}\right). \quad (17b)$$

Since, within the driving field oscillation, the phase is proportional to the time, the acquisition of either channel voltage, as a function of source, or lock-in phase through a complete cycle, is equivalent to obtaining M(t) or M(−t), respectively. This is shown in Fig. 4. A corresponding field profile is just $H(t) \sim \cos(\varphi_D)$. The only remaining question is a point in which we can uniquely link M(t) and H(t). Again, the phase $\varphi_D^1$, for which the voltage of the empty detection coil is maximal in the first channel, represents the point of maximal field. Consequently, $H = H_0 \cos\left(\varphi_D - \varphi_D^1\right)$ can be taken as a driving field. That means that in $\varphi_D^1$ the sample voltage in the first channel corresponds to the magnetization in maximal field while the voltage in the second channel corresponds to remanent magnetization. If the voltage in Eq. (17a) is displayed against $H_0 \cos\left(\varphi_D - \varphi_D^1\right)$, one obtains a complete hysteresis loop. It is shown in Fig. 5. For comparison, the same hysteresis, as obtained in the other two ways, is also shown. The second representation is composed from the first 50 measured Fourier components:

$$M(\varphi) = \sum_{n=1}^{50} [-V_n^x \cos(n\varphi)/n + V_n^y \sin(n\varphi)/n],$$

and the field profile is just $H(\varphi) = H_0 \cos(\varphi)$. This second representation overlaps the first one. The third hysteresis representation (shown elongated but just for RMS normalization factor) is obtained by numeric integration of the digitized voltage from Fig. 2 and multiplied by $2\pi f$. Here, the field profile is obtained by digitizing the voltage on a standard resistor in primary circuit, which is then scaled to the field amplitude.

It is worth noting that hysteresis, as obtained from Eq. (17a) by changing the detector phase, "goes" in clockwise direction as one scans through corresponding field points. This is a consequence of negative sign of the detector phase. Direction of hysteresis looping can be reversed by sorting the phase in mode opposite to acquisition prior to making the $\cos(\varphi_D)$. If the phase loop is done by changing the source

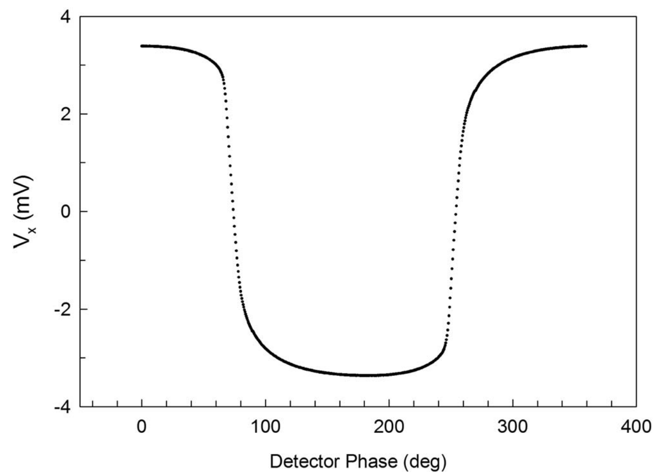


FIG. 4. Voltage measured in first channel of PAR 5210 lock-in (in FLAT mode) as the function of lock-in phase.

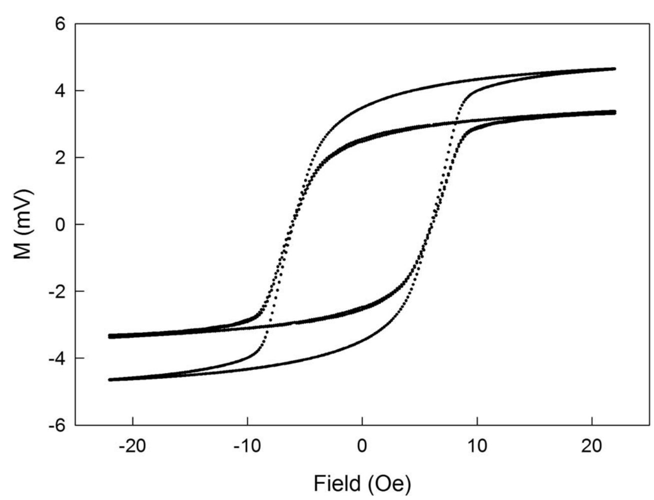


FIG. 5. Hysteresis obtained by: integration of digitized induced voltage (elongated one), constructed from 50 harmonics measured by Ametek 7265, and directly measured by PAR 5210 lock-in. Latter two almost coincide.

phase then hysteresis scans in usual, counterclockwise direction by the same field.

## III. SUMMARY

We proved that the voltages that a digital lock-in (set to measure nth harmonic of the input signal) generates in the two channels are given by

$$V_n^x = \frac{1}{\pi\sqrt{2}}\int V_{in}(\varphi)\sin(n\varphi + \varphi_D)d\varphi,$$

$$V_n^y = V_n^x\left(\varphi_D + \frac{\pi}{2}\right).$$

If $V_{in}$ is a multiharmonic function, then determination of the proper lock-in phase is crucially important. In order to determine Fourier components of input signal, which comes from ac susceptometer, we define the procedure about phase adjustment as follows:

1. If there is an option of changing the phase of input signal, then the best way is to set the lock-in phase $\varphi_D$ to zero and change the source phase such that complete voltage of empty secondary coil (in the first harmonic) is in the first channel only. That will remove any phase dependence from Eqs. (6) and the measured voltages in the two channels are proportional to real and imaginary part of the nth Fourier component of the susceptibility.
2. If there is no possibility to change the phase on the source side (for example, when the lock-in output is used to feed the primary coil), then the proper lock-in phase is found such that complete voltage of the empty secondary coil (in the first harmonic) is positioned in the first channel only. The AUTOPHASE command[3] can be used for this purpose. This lock-in phase is assigned as $\varphi_D^1$. (This actually represents standard procedure for first harmonic ac susceptibility measurement.) Now, for the nth harmonic measurement of the sample voltage it is necessary to set the lock-in phase to $\varphi_D = n\varphi_D^1$. This will again separate real and imaginary part of nth Fourier component of the susceptibility.

In both cases the voltages in the two channels are

$$V_n^x = C_S\frac{n\omega H_0}{\sqrt{2}}\chi_n',$$

$$V_n^y = -C_S\frac{n\omega H_0}{\sqrt{2}}\chi_n''.$$

Proper separation of the Fourier components of induced voltage enables one to find odd Taylor components of sample's susceptibility. This is very important in magnetic phase transitions where magnetization is a nonlinear function of the field even for small fields:

$$\chi_{2n+1} = \frac{1}{C_S}\frac{\sqrt{2}}{\omega}\frac{1}{H_0^{2n+1}}\frac{2^{2n}}{2n+1}\sum_{j=0}^{\infty}(-1)^j\binom{2n+j}{j}V_{2n+1+2j}^x.$$

Validity of all these formulas was confirmed for digital lock-ins Ametek/Signal Recovery models 7225, 7265, and 7280, as well as for Stanford Research Systems 830.

## ACKNOWLEDGMENTS

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[1] M. Nikolo, Am. J. Phys. **63**, 57 (1995).
[2] D. Drobac and Z. Marohnic, Fizika A **8**, 165 (1999).
[3] AUTOPHASE command of the lock-in searches the referent phase of the lock-in in a way that complete positive signal is put in first channel and zero in second channel.