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The NMR detection characteristics of weakly driven marginal and Robinson oscillators

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Abstract A theoretical analysis of weakly driven marginal and Robinson oscillators is made using the two-sideband approximation. Expressions are derived for the frequency pulling and for the amplitudes of the sidebands and the free oscillation signal. The effect of NMR occurring at the free oscillation frequency is investigated theoretically, and it is shown that the NMR signal obtained by linear envelope detection of the output of an ideal Robinson oscillator should be independent of the size of the driving signal. For the marginal oscillator, an NMR enhancement proportional to the square of the amplitude of the driving signal is predicted. A spectral analysis of a driven Robinson oscillator showed only fair agreement between experiment and theory. When the driving frequency was 50 kHz below that of the free oscillation, our Robinson oscillator showed a large increase in NMR sensitivity which was proportional to the square of the amplitude of the driving signal. Little variation in NMR sensitivity was observed when the driving frequency was 50 kHz above that of the free oscillation. The disagreement between experiment and theory is attributed to imperfections in the oscillator which were neglected in the theoretical treatment.

1 Introduction

Following the early work of Pound (1950), several nuclear magnetic double resonance (NMDR) studies (e.g. Sarles and Cotts 1958, Itoh and Kusaka 1959, Holcolm *et al.* 1961, Hughes and Reed 1971) have been performed using steady-state self-oscillating NMR detectors. In such work it has been customary to reduce the coupling between the two RF circuits by means of a crossed-coil arrangement. Even so, the residual coupling has usually been sufficient to cause frequency pulling and in some cases phase-locking, phenomena which are characteristic of driven or forced oscillators. An associated phenomenon is the dependence of the size of the NMR signal on the magnitude of the driving signal. There appears to be a general tendency for the NMR signal intensity to increase with increasing driving signal, and in our laboratory we have observed NMR enhancements of over 50% (Reed 1970, Smith and Hughes 1972 unpublished) with both a Pound–Knight–Watkins type of marginal oscillator† (Pound and Knight 1950, Watkins and Pound 1951, Watkins 1952) and a limited or Robinson oscillator (Robinson 1959, 1965, Faulkner and Holman 1967). This kind of NMR enhancement is independent of the nuclear spin system being investigated. Rather, it is a purely electronic effect occurring within the oscillator, and the present work was undertaken to explain this surprising phenomenon. Further motivation was provided by our desire to develop, for NMDR work, a spectrometer whose sensitivity is relatively unaffected by a small driving signal.

The driven or forced oscillator problem has received little attention until recently, and much of §2 is devoted to a simple theoretical treatment of weakly driven oscillators. We then calculate the effect of NMR on such oscillators, before considering the signal obtained by envelope detection of the oscillator output. Some experimental results obtained with a Robinson spectrometer are presented and compared with theory in §3.

2 Theory

The early theory of Van der Pol (1927, 1934) successfully accounted for the suppression of the oscillation, and subsequent phase-locking of a marginal oscillator by a driving signal. However, to explain the phenomenon of frequency pulling (Adler 1946, Stover 1966) and to account for the form of the spectrum of the 'unlocked' driven oscillator (Stover 1966, Armand 1969, Biswas 1970), it is necessary to modify the Van der Pol treatment.

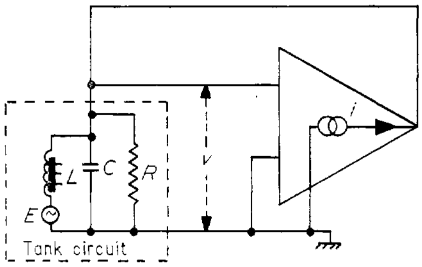

Figure 1 Schematic representation of a driven oscillator

Consider the circuit shown in figure 1. The oscillator consists of a resonant RLC circuit fed by an infinite impedance current generator, while the driving signal is represented by the voltage source

$$E = E_0 \sin \omega_2 t \tag{1}$$

in series with the inductance L. Since we are primarily interested in weakly driven oscillators, we assume that the steady-state signal v across the tank circuit consists of a carrier at frequency ω_1, plus two symmetrically disposed sidebands, one of which is at the driving frequency ω_2. That is, we assume that

$$v = A_1 \cos \omega_1 t + A_L \cos\{(\omega_1 + \Omega)t + \delta_L\} + A_S \cos\{(\omega_1 - \Omega)t + \delta_S\} \tag{2}$$

where

$$\Omega = \omega_2 - \omega_1. \tag{3}$$

(The subscripts L and S refer to the relative size of the two sidebands, the sideband at the driving frequency always

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† In this paper we use the term marginal oscillator to describe a steady-state free-running oscillator whose oscillation amplitude is governed by the nonlinearity of a smooth characteristic.

having the larger amplitude.) Equation (2) can be conveniently written in the form

$$v=(A_1+A_2\cos\Omega t+A_3\sin\Omega t)\cos\omega_1 t + (B_2\cos\Omega t+B_3\sin\Omega t)\sin\omega_1 t \quad (4)$$

in which case

$$A_L=\{(A_2-B_3)^2+(A_3+B_2)^2\}^{1/2}/2 \quad (5)$$

and

$$A_S=\{(A_2+B_3)^2+(A_3-B_2)^2\}^{1/2}/2. \quad (6)$$

Justification for the two-sideband approximation was obtained in our laboratory by a spectral analysis of an unlocked, driven Robinson oscillator (see also Stover 1966). The sideband amplitudes A_L and A_S were found to be of the same order of magnitude (provided $\Omega\ll\omega_1$), and were much larger than those of the higher order sidebands at $\omega_1\pm2\Omega$, $\omega_1\pm3\Omega$, etc., for small values of E_0. Near the locking region, the amplitudes of the higher order sidebands are comparable with A_L and A_S, and equations (2) and (4) are then obviously inappropriate.

2.1 *Application of the two-sideband approximation to weakly driven oscillators*

The behaviour of the tank voltage v is governed by the equation

$$\frac{d^2v}{dt^2}+(RC)^{-1}\frac{dv}{dt}+\omega_0^2 v=\omega_0^2E_0\sin\omega_2 t+C^{-1}\frac{di}{dt} \quad (7)$$

where i is the feedback current and $\omega_0=(LC)^{-1/2}$, the natural frequency of the tank circuit. In the narrow-band approximation ($\Omega\ll\omega_1$), equation (4) can be expressed as

$$v=A(t)\cos\{\omega_1 t-\zeta(t)\} \quad (8)$$

where $A(t)$ is a slowly varying amplitude and

$$\tan\zeta(t)=\frac{B_2\cos\Omega t+B_3\sin\Omega t}{A_1+A_2\cos\Omega t+A_3\sin\Omega t}. \quad (9)$$

For an ideal Robinson oscillator with no phase shift (see for example Hughes and Smith 1971), the corresponding feedback current is given in the harmonic approximation by

$$i=\kappa\cos\{\omega_1 t-\zeta(t)\} \quad (10)$$

where κ is a constant which depends upon the limiter output. By expanding equation (10), it is found that the feedback current has components at frequencies ω_1, $\omega_1\pm\Omega$, $\omega_1\pm2\Omega$, etc. In the two-sideband approximation we retain only those at ω_1 and $\omega_1\pm\Omega$, and, by substituting in equation (7) and separately equating to zero the coefficients of $\cos\omega_1 t$, $\cos\omega_1 t\cos\Omega t$, $\cos\omega_1 t\sin\Omega t$, $\sin\omega_1 t$, $\sin\omega_1 t\cos\Omega t$ and $\sin\omega_1 t\sin\Omega t$, we find

$$A_1=A_0\{1-(B_2^2+B_3^2)/4A_1^2\} \quad (11)$$

$$\omega_1-\omega_0=\omega_0(A_2B_2+A_3B_3)/4QA_1^2 \quad (12)$$

$$A_2=-QE_0/(1+4Q^2\xi^2) \quad (13)$$

$$A_3=-2Q^2\xi E_0/(1+4Q^2\xi^2) \quad (14)$$

$$B_2=-E_0/2\xi \quad (15)$$

$$B_3=Q\xi E_0/2(1+4Q^2\xi^2) \quad (16)$$

provided

$$\frac{Q^2(E_0/A_0)^2}{(1+4Q^2\xi^2)^2},\quad \frac{(E_0/A_0)^2}{16\xi^2}\ll1. \quad (17)$$

Here, $A_0=\kappa R$, the oscillation amplitude in the absence of a driving signal, $Q=\omega_0RC$, the Q factor of the tank circuit, and $\xi=\Omega/\omega_0$.

For a marginal oscillator with a characteristic of the form

$$i=\alpha v+\beta v^2+\gamma v^3, \quad (18)$$

a similar calculation shows that

$$A_1=A_0\{1-(3A_2^2+3A_3^2+B_2^2+B_3^2)/4A_1^2\} \quad (19)$$

with A_0 given by

$$A_0^2=4(\alpha-R^{-1})/(-3\gamma). \quad (20)$$

However, $\omega_1-\omega_0$, A_2, A_3, B_2 and B_3 for the marginal oscillator are still given by equations (12)–(16) respectively, provided Q is replaced by $Q'=Q/2(\alpha R-1)$, the so-called effective Q factor of the oscillator (Watkins 1952). It can be seen that the frequency pulling and the suppression of A_1 are of second order in E_0/A_0. Also, the frequency pulling is the same for both types of oscillator provided $4Q'^2\xi^2\ll1$. If this condition is not satisfied, the marginal oscillator is less susceptible to frequency pulling on account of its narrower bandwidth. On the other hand, the suppression of the oscillation is greater for the marginal oscillator than for the Robinson oscillator, as can be seen from equations (11) and (19). This is due to the action of the limiter in the Robinson oscillator which removes from the feedback current the amplitude modulation represented by the A_2 and A_3 terms in equation (4). The frequency modulation represented by the B_2 and B_3 terms is of course unaffected by the limiter.

2.2 *The effect of* NMR *on weakly driven oscillators*

The resonant absorption of energy associated with χ'', the imaginary part of the nuclear spin susceptibility, can be regarded in the usual way as a small change $-4\pi\eta\chi''(\omega)R^2/\omega L$ in the shunt resistance of the tank circuit, η being the filling factor of the coil. In practice, $\chi''(\omega)$ is nonzero over only a small frequency range and, to correspond to the usual experimental situation, we assume that NMR occurs at the carrier frequency but not at the sideband frequencies. The resistance R therefore changes by

$$dR=-4\pi\eta\chi''(\omega_1)R^2/\omega_1L \quad (21)$$

at ω_1 but is unchanged at $\omega_1\pm\Omega$. Using equation (7), one finds for the Robinson oscillator that the change in A_1 caused by NMR is given, to second order in E_0/A_0, by

$$\frac{dA_1}{A_0}=\{1+(B_2^2+B_3^2)/4A_0^2\}\frac{dR}{R} = -4\pi\eta Q\chi''(\omega_1)(1+E_0^2/16A_0^2\xi^2). \quad (22)$$

A similar calculation for the marginal oscillator shows that

$$\frac{dA_1}{A_0}=-4\pi\eta Q'\chi''(\omega_1)\left\{1+\frac{E_0^2(1+92Q'^2\xi^2+64Q'^4\xi^4)}{16A_0^2\xi^2(1+4Q'^2\xi^2)^2}\right\}. \quad (23)$$

We see that there is in both cases an enhancement of the NMR signal *at the frequency* ω_1, which is proportional to E_0^2.

Associated with NMR absorption is a change $4\pi\eta\chi'(\omega)L$ in the inductance of the coil, where χ' is the real part of the nuclear spin susceptibility. This leads to a change in ω_1, and also a change in A_1 via the ξ dependence of B_2. For example, the change in A_1 for the Robinson oscillator is given by

$$\frac{dA_1}{A_0}=4\pi\eta\chi'(\omega_1)\frac{E_0^2}{16A_0^2\xi^3}. \quad (24)$$

This χ' contribution will not in general be negligible, since it is comparable to the enhancement in the χ'' response (see equation (22)), provided $\xi\leqslant Q^{-1}$.

Because of the coupling between the carrier and sidebands within the oscillators, NMR at the carrier frequency will produce simultaneous changes in the sidebands, and it can be shown that the response to χ'' at the sideband frequencies is given by

$$\frac{dA_S}{A_S} = -(1+16Q'^2\xi^2)\frac{dA_L}{3A_L} = -\{4\pi\eta Q'\chi''(\omega_1)\}\frac{12\xi^2Q'^2}{1+4Q'^2\xi^2} \quad (25)$$

for the marginal oscillator. A similar though smaller effect occurs for the Robinson oscillator. We see from equations (22) and (25) that NMR absorption should cause a decrease in A_1 and A_S and an increase in A_L.

2.3 *Observation of* NMR *after linear detection*

Conventional NMR spectrometers are equipped with quasi-linear RF detectors and therefore do not respond to A_1 alone. A detector fed by a narrow-band signal of the form $A(t)\cos\{\omega t - \zeta(t)\}$ responds only to the slow variations in $A(t)$. Thus, for an input voltage of the form given by equation (4), the detector output D of a linear detector is, in second order, proportional to

$$A_1\{1+(B_2{}^2+B_3{}^2)/4A_1{}^2\}. \quad (26)$$

It follows from equations (22), (23) and (26) that the change in the detector output produced by χ'' is

$$dD \propto -4\pi\eta Q\chi''(\omega_1)\, A_0 + \text{terms of order } E_0{}^4/A_0{}^4 \quad (27)$$

for the Robinson spectrometer and

$$dD \propto -4\pi\eta Q'\chi''(\omega_1)\, A_0\left\{1+\frac{Q'^2E_0{}^2(43+24Q'^2\xi^2)}{8A_0{}^2(1+4Q'^2\xi^2)^2}\right\} \quad (28)$$

for the marginal oscillator spectrometer. We see that the NMR signal of a marginal oscillator spectrometer is enhanced whereas that of the Robinson spectrometer is not. This is because a perfect limiter mixes the carrier and sidebands in such a way as to compensate for the mixing produced by a linear detector. Indeed, a perfect limiter and a linear detector are in this sense complementary devices.

The χ' contribution considered in §2.2 is likewise removed by the cancellation occurring within a Robinson spectrometer. In the marginal oscillator spectrometer, on the other hand, there is only partial cancellation, and a second order enhancement of χ'' is expected, in addition to a χ' contribution.

3 Experimental

A quantitative verification of the theory was attempted with the tube-type Robinson spectrometer used for previous NMDR studies (Hughes and Reed 1971). The circuit was similar to that described by Howling (1966), apart from the limiter stage whose load consisted of a pair of crossed Schottky diodes. To correspond with the assumption made in the theory, the phase shift ϕ around the feedback loop was always made zero by suitable adjustment of the anode loads in the oscillator section. The response of the spectrometer to a calibrator signal (Watkins 1952) at various oscillation levels showed that the limiter was working satisfactorily, and the unit appeared to be operating in the Robinson mode. For the measurements to be described, values of 21 mV and 7·6 MHz were selected for the free oscillation amplitude and frequency; the effective Q factor of the tank circuit was 48.

According to equations (12)–(16), the frequency pulling $\delta = \omega_1 - \omega_0$ should be given by

$$(\delta\Omega)^{-1} = 8A_0{}^2(1+4Q^2\xi^2)/E_0{}^2\omega_0{}^2. \quad (29)$$

Preliminary studies indicated that the magnitude of the frequency pulling was by no means symmetric with respect to a change in the sign of Ω. It can be shown theoretically that a nonzero phase shift in the feedback loop would cause such asymmetric behaviour. However, since the oscillator was always accurately 'phased-in' ($|\phi| < 3°$), this cannot be the correct explanation. After several unsuccessful trials, approximately symmetric behaviour was obtained when the anode loads of the first and second stages of the oscillator were made highly inductive and highly capacitative, respectively. This arrangement, which was used for all the measurements to be described in this paper, gives minimum phase distortion $\partial\phi/\partial\omega$ (at $\phi = 0$) and this seems to be a necessary condition for symmetric frequency pulling. As can be seen in figure 2, $(\delta\Omega)^{-1}$ varies linearly with Ω^2 in agreement with equation (29), except near $\Omega = 0$ where the condition $(E_0/A_0)^2 \ll 16\xi^2$ is no longer satisfied. The value of the intercept $8{\cdot}3\times10^{-3}$ kHz^{-2} is in satisfactory agreement with the theoretical value of $7{\cdot}9\times10^{-3}$ kHz^{-2}. On the other hand, the experimental value of the slope $1{\cdot}9\times10^{-6}$ kHz^{-4} is in poor agreement with the theoretical value of $1{\cdot}3\times10^{-6}$ kHz^{-4}. Indeed, the experimental value of the slope points to a Q value of 59 rather than the value 48 ± 1 obtained by direct observation of the response curve of the tank circuit. We attribute the discrepancy to imperfections in the oscillator which have been ignored in the theory. This is also indicated by the fact that the frequency pulling could be significantly changed by minor modifications to the oscillator circuit (in which $|\phi|$ was kept near zero).

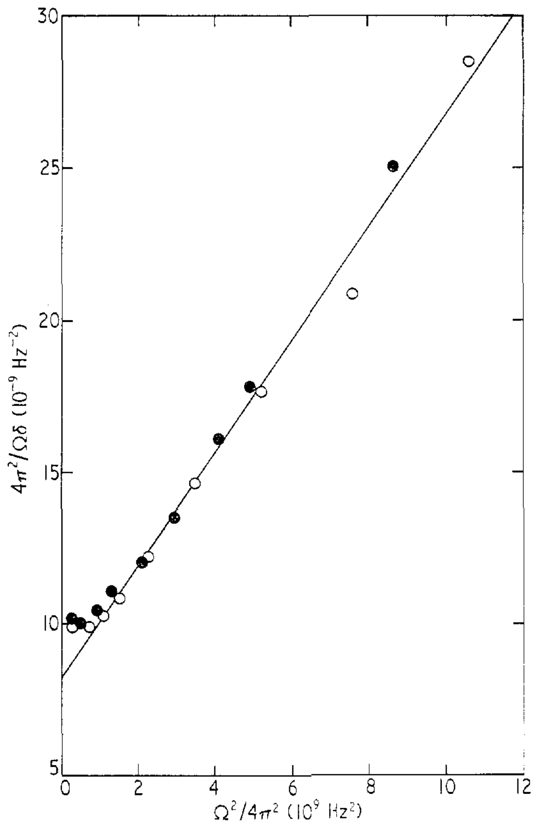

Figure 2 $4\pi^2/\Omega\delta$ shown as a function of $\Omega^2/4\pi^2$ for the Robinson oscillator described in the text. Closed and open data points refer to positive and negative values of Ω respectively

The NMR sensitivity of our Robinson oscillator was measured as a function of E_0 using the ^{23}Na resonance from a doped aqueous solution of $NaNO_3$. (The NMR detection characteristics cannot be studied using a calibrator circuit since NMR is frequency-selective whereas the calibrator signal is not.) Resonances were recorded as the first derivative of the AM signal using conventional phase-sensitive detection in conjunction with sinusoidal field modulation and a linear frequency sweep. (A correction was applied to take account

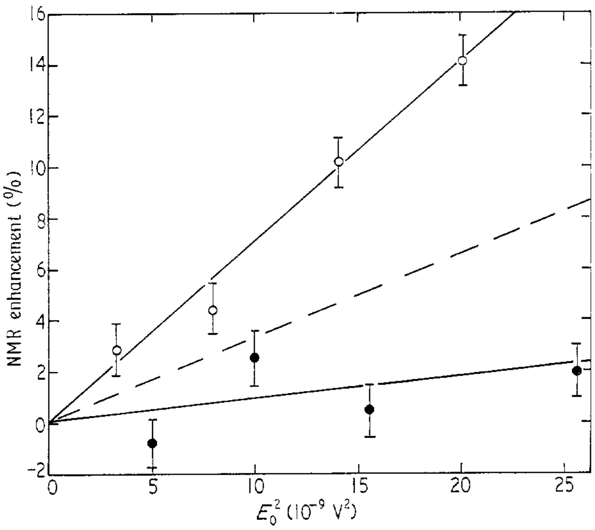

Figure 3 Enhancement of the peak-to-peak NMR signal shown as a function of $E_0{}^2$ for $|\Omega|/2\pi = 50$ kHz. Closed and open data points refer to positive and negative values of Ω respectively; the error bars show estimated errors. The broken line represents the theoretical behaviour (equation (22)) expected if the AM detector responds only to the free oscillation signal. (No enhancement is expected for an ideal Robinson spectrometer)

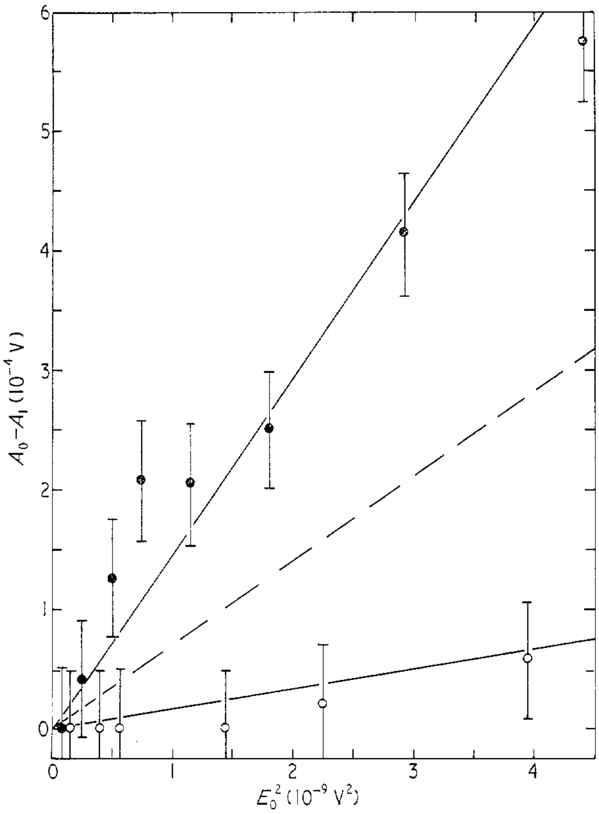

Figure 4 The reduction in the amplitude of the free oscillation signal shown as a function of $E_0{}^2$ for $|\Omega|/2\pi = 50$ kHz and $A_0 = 21$ mV. Closed and open data points refer to positive and negative values of Ω respectively; the error bars show estimated errors. The broken line represents the theoretical behaviour given by equations (11), (15) and (16)

of small saturation effects.) It is evident from figure 3 that the peak-to-peak NMR signal intensity is by no means independent of E_0, as theory would predict, neither is it symmetric with respect to a change of sign of Ω. However, the enhancement does seem to be proportional to $E_0{}^2$, as might be expected if the cancellation referred to in §2·3 is incomplete. The maximum value of E_0 in these measurements is about 0·15 mV (roughly half the value required to phase-lock the oscillator), and measurements of the frequency pulling and the sideband amplitudes, reported elsewhere (Hughes *et al.* 1973) indicate that the two-sideband approximation remains valid at such large values of E_0. (Earlier measurements (Reed 1970, Smith and Hughes 1972 unpublished) showed an enhancement which was proportional to $E_0{}^2$ and whose magnitude was comparable to that shown in figure 3 for $\Omega/2\pi = -50$ kHz. However, no attention was paid to the oscillator phasing in this early work.) The broken line in figure 3 shows the enhancement (due to χ'') expected if the contribution of the sidebands to NMR is completely neglected (see equation (22)). In cases where a large enhancement was obtained, the resonances were visibly asymmetric due to the expected admixture of χ'. (Such an admixture does not in first order affect the peak-to-peak intensity of the resonance.)

In order to check that the detector was responding properly to the sidebands, we measured the variation of the AM detector output with E_0. According to equations (11) and (26), the DC output of the detector should be independent of E_0 for a Robinson oscillator, at least in second order. Although a small reduction in the detector output was observed, it was less than 20% of that expected if the detector was responding to A_1 alone. This suggests that the anomalous behaviour is not caused by limitations of the detector or limiter.

The behaviour of the oscillator was therefore checked by measuring A_L, A_S and A_1 as a function of E_0 using a Hewlett-Packard spectrum analyser model number 8554L/8552A. While A_L and A_S were found to be proportional to E_0 in agreement with theory, there were unexplained differences of up to 25% between the experimental and theoretical values of the constants of proportionality. The reduction in A_1 is shown as a function of $E_0{}^2$ in figure 4. Again there is a large and unexplained discrepancy between the data for $\Omega/2\pi = \pm 50$ kHz, though both sets do lie on either side of the theoretical prediction represented by the broken line. Indeed, the similarity between figures 3 and 4 may not be completely coincidental.

By feeding the oscillator output into a communications receiver we were able to detect signals at the sideband frequencies $\omega_1 \pm \Omega$, $\omega_1 \pm 2\Omega$ and $\omega_1 \pm 3\Omega$ when NMR was occurring at ω_1. As expected, the signals became progressively weaker with increasing order, and consisted in each case of a mixture of χ' and χ''. Since considerable mixing was also introduced by 'slope detection' of the FM signal within the communications receiver, no analysis of the data was made.

The observations made with our Pound–Knight–Watkins spectrometer are entirely qualitative and are in agreement with theory. In the first place, a large NMR enhancement was obtained with small values of E_0. Secondly, and this is in complete contrast to the Robinson spectrometer, the DC output of the detector fell by about 15% and then increased sharply to above the original value as E_0 was progressively increased. The original decrease is presumably associated with the A_2 and A_3 terms in equation (19), the subsequent increase being due to higher order terms ignored in the theory.

Finally we consider the question whether the enhancement obtained with a driven oscillator results in an improved NMR signal-to-noise ratio. Experimentally, there was no evidence of this with either the Robinson or the Pound–Knight–Watkins spectrometer. Indeed, since the NMR signal is caused by an EMF induced in the coil by the precessing nuclei, and since the noise can also be regarded as an EMF in series with the coil, one would not expect an improved signal-to-noise ratio.

4 Conclusion

The theory developed in §2 fails to account satisfactorily for the behaviour of our Robinson spectrometer. Whilst the discrepancy may in part be due to limitations in the detector and limiter, we believe that it is largely due to other effects not considered in the theory. One possibility is a frequency-dependent gain within the oscillator amplifier; if a narrow-band AM signal of the form

$$A' \cos \omega t + A'' \cos \omega t \cos \Omega t \tag{30}$$

is applied to an amplifier with gain $G(\omega)$, the output is

$$G(A' \cos \omega t + A'' \cos \omega t \cos \Omega t) - (\partial G/\partial \omega)\, \Omega A' \sin \omega t \sin \Omega t \tag{31}$$

and an FM signal proportional to $\partial G/\partial \omega$ is generated. (Phase distortion (a nonzero $\partial \phi/\partial \omega$) produces a similar conversion of AM to FM and vice versa.) Also, the tank circuit parameters, R and L say, are in practice frequency dependent; no account of this was taken in our theory. Further experimental and theoretical work is obviously required before the behaviour of real driven oscillators is properly understood. On the basis of our experience, we offer the following remarks. Firstly, it seems advisable to concentrate attention on driven oscillators which are 'phased-in', since a phase shift in the feedback loop introduces several complications. Secondly, measurements at a lower frequency, perhaps near 1 MHz, are desirable since greater control of circuit parameters would then be possible.

In the context of NMR, we have been unable to achieve the zero enhancement expected for an ideal Robinson spectrometer, although we have approached it in our measurements at $\Omega/2\pi = +50$ kHz.

Acknowledgments

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