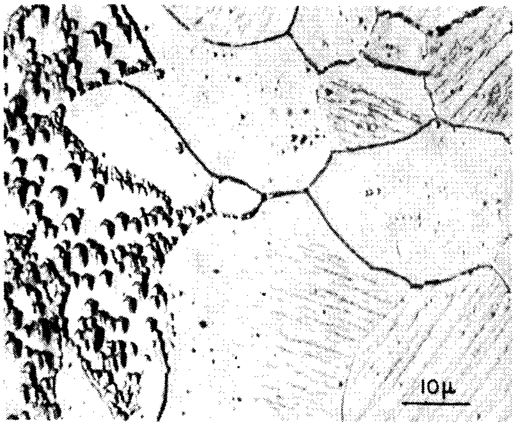

FIG. 12. Etch pits on a partly contaminated surface; etchant A.

area that was partly contaminated with carbon. Note the difference in pits! Carbon from the oil diffusion pump was sufficient to contaminate the sample during the high-temperature annealing.

Carbon analysis made on a few samples that were not deliberately contaminated showed the carbon concentration to range from 10–20 ppm. The concentration of carbon in contaminated samples was, of course, much higher. Microhardness tests showed that some of the heavily striated grains were actually a new phase which was identified as W_2C. It was further observed that the shape and density of etch pits were affected by the presence of carbon even when the concentration was too low for the formation of a second phase.

SUMMARY

Some details of dislocation etch pits in tungsten have been presented. The etchants A and C produce pits defined by crystallographic faces, namely {110} planes. This fact can conveniently be used to determine the crystal orientation. The Millner-Sass etchant does not produce well-defined pits. Etch pits can be produced only on certain crystallographic planes. All three etchants can reveal dislocations, however, there is no guarantee that all pits are formed at dislocation sites. The presence of carbon can markedly affect the size, shape, and density of etch pits. The etchants A and C can produce pyramids on octahedral or nearly octahedral planes. The mechanism for this phenomenon is not understood, and it is not certain that it is related to dislocations.

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On the Problem of Pulsed Oscillations in Ruby Maser*

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It is shown by means of a semiquantitative nonlinear analysis that elementary interaction between an inverted electron spin system and a resonant cavity does not give rise to the pulsed mode of operation of the ruby maser oscillator. It is suggested that the additional nonlinearity necessary for the existence of such a mode resides in the "distant ENDOR," the interaction between the chromium electrons and the Al^{27} nuclei.

THE existence of the pulsed mode of operation of the ruby maser oscillator[1] has been attributed by Statz and DeMars[2] to time-dependent interaction between the inverted population of the electron-spin systems of the paramagnetic substance and the resonant cavity. It has come to this writer's attention that conclusions contained in reference 2 are based solely on analog computer solutions of nonlinear differential equations describing the interaction between the spin system and the cavity. A semiquantitative analysis of these equations shows the computer solutions to be in error, and consequently the conclusions of reference 2 concerning the nature of pulsed oscillations to be incorrect.

The equations in question, as derived by Statz and DeMars, and also independently by this writer[3] are of the form:

$$\begin{aligned} dx/dt &= -c_1xy + c_2(x_0 - x) \\ dy/dt &= c_3xy - c_4y, \end{aligned} \tag{1}$$

where x is the population difference, and y is the magnetic energy in the cavity. The coefficients c_1, c_2, c_3, and c_4, and the constant x_0 are functions of material and circuit parameters, of temperature, and of excitation. Definitions of these quantities may be found in reference 2. For the purposes of the present analysis it is sufficient

* This work was conducted by Project MICHIGAN under a Department of the Army contract administered by the U. S. Army Signal Corps.

[1] C. Kikuchi, J. Lambe, G. Makhov, and R. Terhune, J. Appl. Phys. **30**, 1061 (1959).

[2] H. Statz and G. DeMars, *Quantum Electronics* (Columbia University Press, New York, 1960), p. 530.

[3] G. Makhov, Conference on Electron Tube Research, Mexico City, 1959.

to establish that c_2 is the inverse of spin-lattice relaxation time T_1, and c_4 is inversely proportional to the cavity Q.

In order to account for pulsed oscillations these equations must admit periodic solutions. Furthermore, in order to be in agreement with experimental data, there must occur a transition from the periodic to the aperiodic mode, as the magnitude (but not the sign) of one or more coefficients is changed. It is shown below that neither condition is satisfied by Eqs. (1).

The system of equations (1) is of second order and hence Liapunoff's stability criterion is applicable.[4] There are two singular points: a focus or a node at

$$x_1 = c_4/c_3 \qquad y_1 = (c_3c_2/c_1c_4)(x_0 - x_1) \tag{2}$$

and a saddle point at

$$x_2 = x_0 \qquad y_2 = 0. \tag{3}$$

Only the former singularity is of interest insofar that the latter merely determines the condition of dynamic equilibrium in the absence of signal field. Of course, the condition

$$x_2 > x_1$$

must be satisfied in order for oscillations to occur.

The first-order terms of the Taylor expansion about x_1, y_1 are computed to be

$$a = -c_3c_2/c_4 \qquad b = -c_1c_4/c_3 \qquad c = (c_3^2c_2/c_1c_4)(x_0 - x_1) \qquad d = 0. \tag{4}$$

These quantities are coefficients of the linearized equations of (1).

The characteristic equation is of the form

$$\lambda^2 - \lambda(a+b) + (ad - cb) = 0. \tag{5}$$

Substitution of (4) into (5) yields

$$\lambda^2 + (c_3c_2/c_4)x_0\lambda + c_3c_2(x_0 - x_1) = 0. \tag{6}$$

The coefficient of the linear term in λ is positive for all positive values of c_3, c_2, and c_4. Hence, the singularity is stable. Further examination of Eq. (6) shows that for all practical operating conditions, the singular point is a focus. This implies that the solution tends towards (x_1, y_1) in an oscillatory manner. This behavior corresponds to the *c-w* mode of the oscillator.

It is now inquired whether there exist closed trajectories in the *x-y* plane about (x_1, y_1). It will be recalled that, experimentally, transitions from the *c-w* mode to the pulsed mode are most easily effected by changing the cavity Q. In terms of the coefficients of Eqs. (1) this corresponds to changing c_4. The singularity, however, remains stable for all positive values of c_4; negative values of c_4 would imply negative cavity Q. Hence, the phenomenon of bifurcation, i.e., transformation of a stable focus into an unstable focus surrounded by a stable limit cycle, which might account for the observed behavior of the maser oscillator, is not to be expected.

The above reasoning does not preclude the existence of an arbitrary number of limit cycles about (x_1, y_1). A general proof of nonexistence of such limit cycles is very difficult. Bendixon's theorem fails to yield any useful information in the present case. The following reasoning, however, based on experimental evidence, shows that Eqs. (1) do not have limit cycles in the region of the *x-y* plane important to the operation of the maser oscillator.

From the physical point of view, the only limit cycles that may be reached from the point $(x_1, 0)$ must be situated between this point and (x_1, y_1). However, the existence of such limit cycles will not permit the trajectory to approach the singular point, i.e., aperiodic behavior would be impossible unless the initial conditions were adjusted so as to place the starting point $(x_1, y>0)$ inside the first limit cycle. This is clearly in contradiction with experimental evidence, since transition between the two modes is obtained without a change in initial conditions. Furthermore, the introduction of a microwave bias, or for that matter, of a noise bias corresponding to a nonzero value of the initial condition on y, fails to produce a transition from pulsed to *c-w* mode. It appears, therefore, safe to conclude that Eqs. (1) do not admit periodic solutions which may account for the pulsed mode of operation of the maser oscillator. They do, however, account adequately for the *c-w* mode.

This analysis is readily extended to transients encountered in optical masers.[5] Decaying oscillation pulses observed by Sorokin and Stevenson[6] in the case of uranium doped calcium fluoride bear a striking resemblance to transients of the *c-w* mode of the ruby maser.[7]

It may be stated in support of the above considerations that analog and digital computer solutions of Eqs. (1) carried out at this laboratory failed to reveal periodic solutions demonstrated in reference 2. Furthermore, there exists ample experimental evidence that the pulsed mode of operation of the ruby maser oscillator does not arise from an elementary interaction between the spin system and the cavity as suggested in reference 2. The most pertinent experimental results to this end appear to be the absence of the pulsed mode in the case of maser oscillator using more heavily doped (0.2% Cr) ruby, and the transition between the *c-w* and the pulsed

[4] N. Minorski, *Nonlinear Mechanics* (Edwards Brothers, Inc., Ann Arbor, Michigan, 1947).

[5] H. Statz, C. Luck, C. Shafer, and M. Clifton, Quantum Electronics Conference, Berkeley, California, 1961.

[6] P. D. Sorokin and M. J. Stevenson, Quantum Electronics Conference, Berkeley, California, 1961.

[7] In reference 5 it has been indicated that earlier computer solutions were inaccurate, and that small additional nonlinearities are required to have undamped oscillation pulses.

modes effected by magnetic resonance of Al^{27} nuclei.[8,9] These results provide an indication that the mechanism responsible for the pulsed mode is contained in the paramagnetic materials and bears a connection with ENDOR interactions existing in ruby.[9,10] Accordingly, it is thought that the pulsed mode can be accounted for if the first of the Eqs. (1), the spin system equation, is supplemented by a function of the population difference; the second of Eqs. (1), the cavity equation, remains unchanged. In other words, the modified equations are of the form:

$$\begin{aligned} dx/dt &= -c_1xy+c_2(x_0-x)+c_5f(x) \\ dy/dt &= c_3xy-c_4y. \end{aligned} \tag{7}$$

The singular point of interest is now located at

$$\begin{aligned} x_1' &= c_4/c_3 \\ y_1' &= (c_3/c_1c_4)[c_2(x_0-x_1)+c_5f(x_1)]. \end{aligned} \tag{8}$$

The pertinent characteristic equation, obtained as previously, is

$$\lambda^2+\left\{\frac{c_3}{c_4}[c_2x_0+c_5f(x_1')]-c_5\frac{df(x)}{dx}\bigg|_{x=x_1'}\right\}\lambda +c_3[c_2(x-x_0)+c_5f(x_1')]=0. \tag{9}$$

Here, the coefficient of the linear term in λ may be positive, zero, or negative, depending on the relative magnitudes of the two terms of opposite sign comprising it. Thus, one may expect, respectively, stable, neutrally stable, and unstable behavior. The first corresponds to the *c-w* mode of the oscillator; the second is essentially impossible to obtain in practice; and the third corresponds to the pulsed mode.

Further examination of the coefficient of the linear term in λ provides an indication as to the nature of the function $f(x)$. In order to induce instability, $f(x)$ must have positive slope. Experiment shows that the transition from the *c-w* to the pulsed mode is effected by decreasing the cavity Q, which corresponds to a proportional increase in the coefficient c_4. This suggests that $df(x)/dx$ must increase with x. An elementary example of such a function is the power function $f(x)=x^n$. It is shown below that satisfactory agreement with experiment is obtained if one chooses $n=1+\epsilon$, where ϵ is a small positive number. With the reasonable assumption of $x_0=2x_1$, the ordinate of the singularity is given by

$$y_1'=\frac{c_3}{c_1c_4}(c_2x_1'^{(1+\epsilon)}). \tag{10}$$

[8] G. Makhov, R. Terhune, J. Lambe, and L. Cross, J. Appl. Phys. **31**, 936 (1960).
[9] G. Makhov, Conference on Electron Tube Research, Seattle, Washington, 1960.
[10] J. Lambe, N. Laurance, E. McIrvine, and R. Terhune, Phys. Rev. **142**, 1161 (1961).

It is now recalled that in the case of Eqs. (1), the corresponding quantity y_1 was given by

$$y_1=(c_2c_3/c_1c_4)x_1. \tag{11}$$

This quantity is both measured and computed to be of the order of 10^{-6} ergs. Comparing Eqs. (10) and (11), one can conclude that in order for y_1' to be in agreement with experiment, the second term on the right-hand side of Eq. (10) must be of this order of magnitude or smaller. This implies that $c_2>c_5$, or that the rate of the process responsible for the second term is slower than spin-lattice relaxation. This process is thought to be the interaction between the chromium electrons and the distant aluminum nuclei, or "distant ENDOR." Relaxation time associated with this interaction is of the order of ten seconds.[8,9] Letting $c_5=0.1$ sec^{-1}, $c_2=10$ sec^{-1}, $x_1=10^{18}$, and choosing $\epsilon=1/9$, one has for y_1':

$$y_1'=2\cdot10^{-6}\approx y_1$$

as required. The coefficient of the linear term in λ, given in the present case by

$$c_2-c_5\epsilon x_1'^{\epsilon}$$

is computed to be approximately -90. Thus, the system is unstable, and the oscillator will operate in the pulsed mode. In order to obtain aperiodic operation, cavity Q must be increased; or pumping must be increased; or ϵ must be decreased. The first two conditions are known to be in agreement with experiment; no applicable data is available at the present time which may permit an evaluation of the third condition.

The choice of $f(x)$ made above was, of course, quite arbitrary. It is known, however, that the interaction between the chromium electrons and aluminum nuclei increases rapidly as the polarizations of the two systems become comparable. This suggests that in the case of the maser oscillator this interaction affects the pumping, rather than the signal transition. As emission occurs, the population difference in the latter decreases, and the polarization of the former increases. This leads to a decrease in the intensity of electron nuclear interaction with decrease in signal transition population difference x. This reasoning appears to indicate that the representation of $f(x)$ over a limited range as a monotonically increasing function of x has reasonable validity. In reality, this function is undoubtedly much more complicated. Currently, it is being attempted to determine $f(x)$ experimentally. A detailed study of the dynamic behavior of the ruby maser oscillator will be published in the near future.

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