## Discussion 49

P. V. MASON, *California Institute of Technology:* Am I right in believing in all cases you assume specular reflection?

TOXEN: You are right. In all of the calculations I have assumed specular reflection. However, I would like to point out that the results with diffuse reflection are quite similar and one really wouldn't expect to find any striking differences.

MASON: In some work I have done, I have found a rather considerable difference. Although I admit that the difference here was that the film had a field applied on only one side. But in this case there was a rather sizeable qualitative difference between diffuse and specular reflection.

TOXEN: Well, if you go back and look in Schrieffer's paper on the calculation of the susceptibility in the superconducting state, and if you look at his curves, you'll find that those for the specular and diffuse reflection are nearly the same. However, here we are not considering the one-sided case, but rather the case in which the field is the same on either side of the film. I have also carried out some calculations for the diffuse scattering case and verify Schrieffer's results.

LYNTON: With regard to the thickness dependence of the critical field in the thin field limit, I believe that you use the customary procedure of saying that in the limit of a very short mean free path the coherence length is essentially equal to the mean free path. There is reason to believe as a number of people have pointed out (de Gennes, Goodman, and others) that the coherence length should in fact be taken as the square root of the bulk coherence length times the mean free path. Would that affect your results in any sense?

TOXEN: Actually I didn't do that. Following the spirit of Schrieffer's calculations the coherence length is that of the bulk material and the coherence length which I used was not taken to depend on thickness. Actually I took $\xi_0$ to be the same for all of the films. The effective coherence distance $\xi$ was then calculated by determining the intrinsic mean free path. That is, I measured the film resistivity in the normal state, measured the film thickness, calculated the intrinsic mean free path, assuming boundary scattering, and then calculated the effective coherence distances in the straightforward manner.

LYNTON: In what straightforward manner?

TOXEN: Well the reciprocal of the effective coherence distance was the sum of the reciprocals of $\xi_0$ and the mean free path.

LYNTON: I would like to hear Dr. de Gennes on whether this indeed is the correct way of calculating $\xi$.

DE GENNES: For phenomena involving the thickness of a transition layer of a superconductor of the first kind, the correct coherence length is $(\xi_0 l)^{\frac{1}{2}}$, while if we compute the current at a point in terms of the vector potential at other points the formula which we must use is $1/\xi = 1/\xi_0 + 1/l$. The reason is as follows: (1) For the transition layer problem, we compute the order parameter $\Delta(\mathbf{r})$ at point $\mathbf{r}$ in terms of $\Delta(\mathbf{r}')$ at surrounding points. The corresponding kernel involves essentially the correlation function:

$$\langle \delta[\mathbf{r} - \mathbf{r}(0)]\delta(\mathbf{r}' - \mathbf{r}(t)) \rangle$$

where $\mathbf{r}(0)$ and $\mathbf{r}(t)$ are successive *positions* of one electron, in the normal state, and the average is taken on electrons at the Fermi energy. The relevant time $t$ is of order $\hbar/k_B T_c$. In a "dirty" alloy the correlation function is ruled by a diffusion process, and the range of the kernel is $\sim(Dt)^{\frac{1}{2}} \sim (\xi_0 l)^{\frac{1}{2}}$ (where $D = \frac{1}{3}v_F l$ is the diffusion coefficient). (2) For the calculation of the current $\mathbf{j}(\mathbf{r})$ in terms of the potential $\mathbf{A}(\mathbf{r}')$, what comes in the kernel is a correlation function between *velocities:* this correlation is essentially destroyed by one collision, and the range of the kernel in the "dirty" limit is the transport mean free path $l$.

# Microwave Nonlinearities in Thin Superconducting Films

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Nethercot[1] has shown that superconducting tin films thicker than 2000 Å can produce second harmonics of 10 kMc/sec microwaves when they are placed in a cavity and properly biased with a magnetic field.

In the present experiment a much thinner tin film is placed directly across an $X$-band waveguide so that the waveguide opening is completely covered. The film is not biased. In a certain power range the film transmission is found to be highly nonlinear and third harmonics of the fundamental frequency are generated. No detectable second harmonic is generated.

[1] A. H. Nethercot, Jr., Phys. Rev. Letters **7**, 226 (1961).

If the film's behavior is to be substantially different in the superconducting and normal states the normal resistance per square should not be too small compared with the guide impedance, which is about 500 Ω. To control the agglomeration of the tin and thereby obtain films with resistances per square of 10 to 110 Ω at helium temperature, the substrates were prepared for the tin evaporation by first evaporating upon them a very thin nonconducting gold layer. The substrates were of fused or crystal quartz and had four gold patches evaporated on them to provide dc connections to the film. The substrate was at room temperature during the tin evaporation. After the tin evaporation the films were removed from the vacuum system, mounted in the waveguide, and inserted into the cryostat. These films are reasonably rugged and survive exposure to air, lengthy storage in low vacuum, and repeated cyclings between room temperature and helium temperatures.

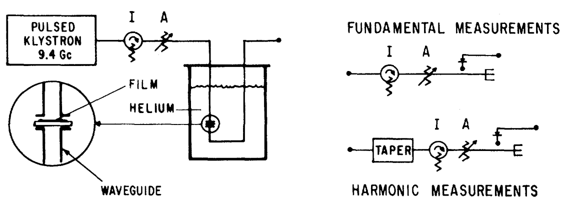


FIG. 1. Schematic diagram of the experimental apparatus. I, isolator; A, precision attenuator.

A schematic drawing of the apparatus is shown in Fig. 1. Usually transmission measurements were made since changes in the film conductivity produce large changes in the transmitted power but only small changes in the reflected power. The frequency was 9.4 kMc/sec. To avoid heating effects, microwave pulses 1 to 10$\mu$ sec long repeated 100 or 1000 times a second were used. The incident power level was varied by a precision attenuator up to the maximum klystron peak power of about 12 W. The transmitted power was measured by varying the output attenuator so as to keep the signal at the crystal detector constant.

Experimental measurements of fundamental transmission and harmonic output for a particular film are shown in Fig. 2. Above $T_c$, the transmission is independent of power. Below $T_c$, the transmission is constant at low powers while the film is in the superconducting state. As the incident power is increased, the transmission increases, approaching the value characteristic of the normal state. This increase in transmission indicates that the film has made a transition from the superconducting state to the normal state. If the transitions are being made during a cycle of the incident field, harmonics of the fundamental frequency should appear in the transmitted field. In the power range where the transmission is increasing, third harmonics are, in fact, observed.

The harmonics were measured in the same way as the fundamental except that the wave guide was tapered. No second harmonics were detected. The results of the third harmonic measurements are also shown in Fig. 2. At about the input power level at which the nonlinearity in the fundamental transmission began, harmonics appeared in the transmitted wave. The absolute harmonic power increased to a maximum and then decreased as the input power level was raised further. Typically, as the temperature was lowered, the maximum harmonic power increased and tended to saturate for further decreases in the temperature. The harmonic power reached a maximum at higher input powers for lower temperatures even when the maximum harmonic power was limited. In general, the harmonic output was measured at successively lower temperatures until the klystron power was insufficient or the lowest attainable temperature was reached (1.75°K).

The temperature dependence of the maximum harmonic power is shown in Fig. 3. Data for three films are shown. The major solid curve is the temperature variation of the square of the critical current density as calculated from the temperature variation of the critical field. This calculation gives an expression for the square of the critical current density of the form

$$J_c^2 = J_c^2(0)(1 - t^2)^3(1 + t^2) ,$$

where $t = T/T_c$ and $J_c(0)$ is the value of the critical current density at $T = 0$°K. The experimental points were fitted by choosing a value for $J_c(0)$. The transition temperatures were determined from a dc measurement.

The qualitative effects reported here may be understood by using a plane-wave treatment and any

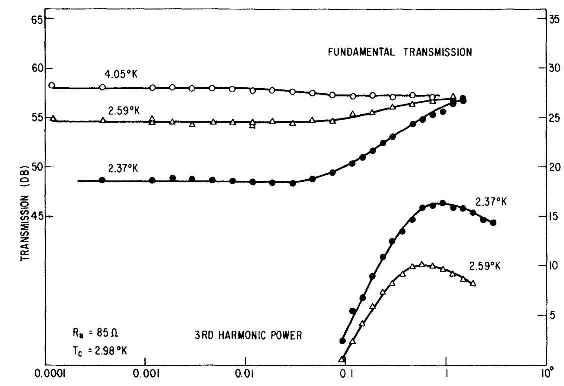


FIG. 2. Transmission and harmonic measurements for a particular film.

of several simple models of the film behavior. The boundary conditions on $E$ and $H$ at the film require that the transmitted electric field be

$$E_t = E_0 \sin wt - (\tfrac{1}{2} Z)J , \tag{1}$$

where $E_0$ sin wt is the incident electric field, $Z$ is the ratio $E/H$ for a plane wave or the impedance of free space, and $J$ is the surface current density. If we have a model for the behavior of $J$ we obtain $E_t$ directly from this equation. One such model is shown in Fig. 4 where the behavior of the film is shown as a function of time.

At low temperatures and low field strengths, the imaginary part of the film conductance associated with the supercurrent is so large compared with $1/Z$ that the film may be regarded as a perfect conductor. In this approximation the supercurrent will be in phase with the incident field and the wave will be perfectly reflected because the incident field is so small that the supercurrent density never reaches $J_c$, and the film is always in the superconducting state [Fig. 4(a)]. If the film were normal the current would be smaller and a part of the incident wave would be transmitted. For larger incident fields the required supercurrent may exceed $J_c$ during part of the cycle. If this occurs it is assumed that the current limits at $J_c$, provided $J_c$ is larger than the current density which would obtain if the film were in the normal state [Fig. 4(b)]. For still higher fields the film may

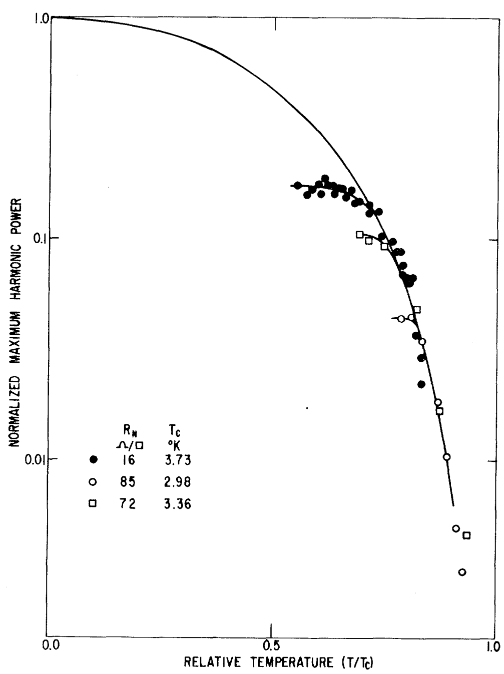


FIG. 3. Temperature dependence of the peak harmonic power.

become completely normal during some parts of the cycle since the normal current density will in this case be greater than $J_c$ [Fig. 4(c)].

Using Eq. (1) and this model we can now calculate $E_t$, Fourier analyze the result, and obtain the harmonic coefficients. This simple model predicts the qualitative features of the experimental results, notably a rise in transmission to the normal value, the generation of odd harmonics, a peak in the third harmonic power, and a maximum third harmonic power proportional to $J_c^2$. The maximum in the third

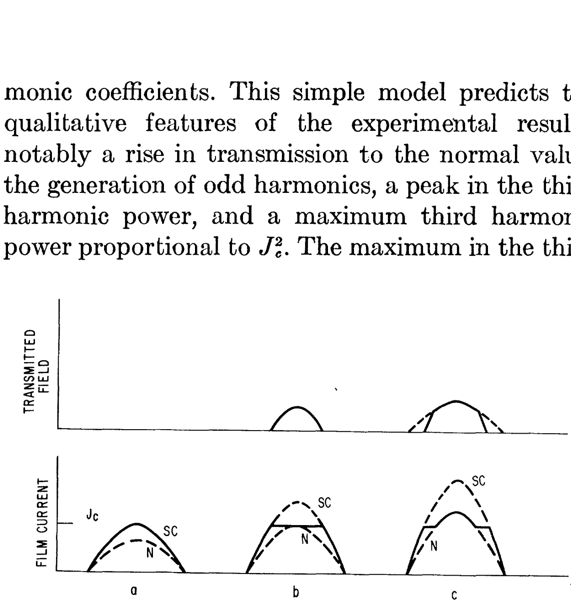


FIG. 4. Simple model of film behavior as a function of time. N, normal film; SC, perfectly conducting film.

harmonic power is associated with the appearance of the normal state since as the film spends more time in the normal state it is more nearly linear in behavior. The only film parameters required by this model are $J_c$ and the resistance per square in the normal state. All of these qualitative predictions are fairly general and do not depend on the details of the model used to describe the transitions. For this particular model, the predicted harmonic power and fundamental transmission rise about an order of magnitude more sharply than they do experimentally. Further, if the critical current density is chosen so that the predicted harmonic maximum occurs at the same incident power level as the observed maxima then the predicted harmonic power is about an order of magnitude larger than the observed value.

When the Ginzburg–Landau theory is applied to the problem of critical currents[2] the density of the superfluid component in the two-fluid model is found to be a function of the common velocity of the ground state $v_s$. The density of the superfluid component decreases in proportion to the square of $v_s$ so that the supercurrent has a maximum as a function of $v_s$. This maximum supercurrent is the critical current. This theoretical treatment assumes that $dv_s/dt$ is sufficiently small so that the equilibrium quasiparticle distribution obtains and the electric fields which are present are very small. In the present experiment, therefore, one may be justified in applying this treatment when the transmitted field is small.

[2] J. Bardeen, Rev. Mod. Phys. 34, 667 (1962). This review gives a clear presentation of the theory and many additional references.

This results in a differential equation for $v_s$ which contains a term in $v_s^3$. This equation can be solved exactly in the absence of the incident field and approximately when there is an incident field, but the transmitted field is small. The third harmonic power appears initially proportional to the cube of the transmitted fundamental power. The fundamental transmission departs gently from the linear region with its slope increasing with incident power very much as the experimental curve does. It is mathematically more difficult and physically unreasonable to apply this approach in the region of the harmonic peak where the transmitted field is large.

In the absence of any detailed model of the experiment which is applicable over the entire power range it is difficult to decide whether the experiment is being unduly influenced by film structure. Calculations have been made using the simple model but taking into account the field distribution in the waveguide and, although the maximum harmonic power is reduced and the effect broadened, the changes are not substantial.

The prediction that the maximum harmonic power is proportional to the square of the critical current should be quite general and not dependent on the details of the model. The fact that the maximum harmonic power does not behave this way at low temperatures suggests that there are some limitations on the film's ability to return to the superconducting equilibrium state which were not present at higher temperatures. As the temperature is reduced the equilibrium number of quasi-particle excitations above the ground state is reduced. Since the probability of quasi-particle recombination should be proportional to the number of quasi-particle excitations,[3] the excitation recombination time is increased. At still lower temperatures one can imagine that this increased recombination time will be long enough to keep the number of excitations from decaying to its equilibrium value in the time available ($\sim 3 \times 10^{-11}$ sec). Since the final number of excitations will be greater than the equilibrium number of excitations at that temperature, an energy gap and consequently a critical current will be reached which depend on the time available for excitation decay rather than directly on the temperature.

[3] D. M. Ginsberg, Phys. Rev. Letters **8**, 204 (1962).

## Discussion 50

LITTLE: How many watts do you get into the third harmonic?

M. D. SHERRILL, *General Electric Research Laboratory:* A few tenths of a milliwatt at best.

BARDEEN: What's the order of the electric field strength at the surface of your film?

SHERRILL: At 1 W transmitted power I believe it's about 10 V per cm. The fields are never larger than this in the experiment.

MAXWELL: Have you made any estimates of the thermal effects when the film goes momentarily normal? Joule heating develops in the film and this has to be dissipated before the next cycle. The point is you have to relax that heat every cycle, otherwise you might expect perhaps some degradation of the harmonic generation.

SHERRILL: Well, let me say that we have observed on occasion pulse shapes on the output harmonic which don't look like the input fundamental power. Sometimes they decay and sometimes we get a spike on the trailing end indicating something even more peculiar is occuring. This is the reason in fact that we went to crystal quartz substrates. We find that we can under the proper circumstances get the same effects there. Although there's certainly the possibility that this is due to heating, there are other effects which we can't rule out. What we've tried to do is to discuss the experiment when these pecularities were absent or minimal.