Ultrasonic attenuation and the resistive transition in a superconducting granular lead film

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We present a model for the temperature dependence of the resistance and ultrasonic attenuation of a superconducting granular lead film. Treating the film as a network of random resistors and using percolation methods, we postulate that while the dc resistance is the resistance of an infinite network, the surface acoustic wave measures the average resistance of finite "subnetworks," the sizes of which are on the order of the acoustic wavelength. The resistance of the infinite network will vanish when more than some critical fraction of the resistors in the network becomes equal to zero. However, when the resistance of the infinite network vanishes, some of the subnetworks will still have nonzero resistance. Consequently, the ultrasonic attenuation should be nonzero even when the dc resistance vanishes. This is in agreement with our experimental data.

I. INTRODUCTION

There has been a considerable amount of interest in systems of low dimensionality, and particularly in granular films[1] as a realization of these systems. This interest motivated surface-acoustic-wave (SAW) experiments on a superconducting granular lead film.[2]

The data on the resistance and ultrasonic attenuation of this film, as functions of temperature, are shown in Figs. 1 and 2. The film was approximately 500 Å thick and had a normal-state sheet resistance of 1000 Ω. The ultrasonic attenuation in the normal state was approximately 4.4 dB/cm, as measured with the SAW at a frequency of 700 MHz. The attenuation of the SAW comes from the coupling of the piezoelectric field in the substrate to the film.[3] This produces Joule losses proportional to the sheet resistance. The attenuation should therefore also be proportional to the sheet resistance. However, comparing the data of Figs. 1 and 2, the observed attenuation in the superconducting state is not proportional to the sheet resistance and, in fact, remains finite below the temperature at which the sheet resistance goes to zero. It has been shown[2] that the excess attenuation in the superconducting state cannot be accounted for by the presence of thermally excited vortex-antivortex pairs. In this paper we present a percolation-theoretic model which accounts for this excess attenuation and also describes the resistive transition in the film.

Our assumption is that long-range superconducting order sets in via classical percolation in a network of Josephson junctions.[4,5] Quantum phase fluctuations, and thus all complications related to phase locking[6] (Kosterlitz-Thouless transition), are neglected. Under this assumption, the model essentially reduces to a classical random resistor network. Above the transition temperature of the grains T_g, all resistors (i.e., all Josephson junctions) are in their normal state and their values are distributed according to some function $W^n(r)$. When the temperature is lowered below T_g, a fraction of the junction resistors go superconducting and the macroscopic sheet resistance R decreases. At some temperature T_c, the fraction of superconducting junctions reaches the percolation threshold p_c and an infinite superconducting cluster appears in the network. The resistance then drops to zero and remains zero for $T < T_c$.

However, the SAW attenuation remains finite at $T = T_c$ and then slowly decreases to zero. We explain this

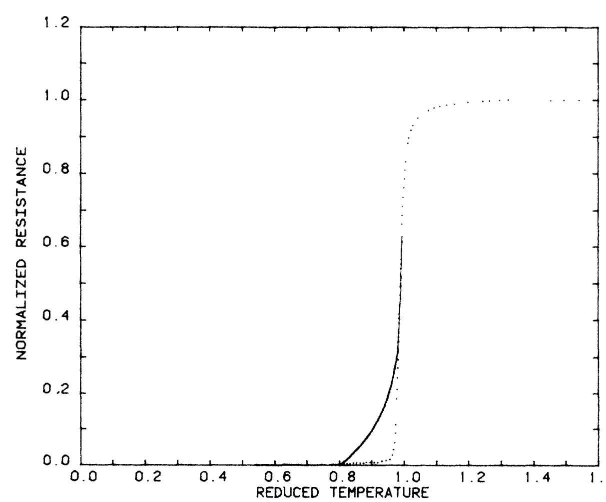

FIG. 1. Normalized resistance (dots) and theory (solid curve) as a function of reduced temperature.

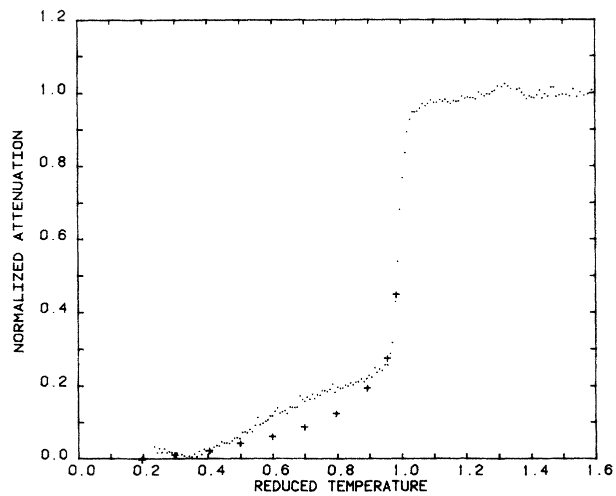

FIG. 2. Normalized attenuation (dots) and theory (crosses) as a function of reduced temperature.

behavior with the following model. The acoustoelectric fields produced by the SAW propagating on the piezoelectric substrate have wavelengths equal to that of the SAW. These fields sample the resistance of small sections of the film, the dimensions of these sections being comparable to the SAW wavelength. The resistance of some of these small sections remains finite, even when the macroscopic sheet resistance of the film vanishes. If, for example, we assume that the network resistors form a square lattice, then 50% of the resistors would still be in the normal state at T_c. The SAW is sensitive to the resistance of these small sections since the SAW is not confined to move along the path of least resistance (the infinite superconducting cluster). Therefore the attenuation should be proportional to the arithmetic average of the resistance of these small sections and will be nonzero at T_c.

In Sec. II of this paper we analyze the resistance of a granular film in the normal and superconducting states. In Sec. III we analyze the ultrasonic attenuation of the film in the normal and superconducting states. Our results are summarized in Sec. IV. Throughout this paper, individual junction resistances will be denoted by r, and the sheet resistance of the network (either finite or infinite) will be denoted R.

II. RESISTANCE IN GRANULAR FILMS

A. Normal-state resistance (AHL model)

In the normal state, the sheet resistance R^n of a random resistance network can be described by the theory of Ambegaokar, Halperin and Langer (AHL).[7] This is a percolation argument for handling a very broad distribution of resistors whose values may range over many orders of magnitude. The AHL theory proceeds as follows. First, let us consider an infinite random resistor network and disconnect all the resistors. We then reconnect them one by one, starting from the smallest ones. At some stage in this process, adding one more resistor will form an infinite cluster connecting the ends of the network. The value of this crucial resistor will be denoted by r_c. The claim is that for a two-dimensional network, R^n is roughly given by r_c. To see this, note that resistances $r \gg r_c$ do not matter much since they are shunted by the smaller resistances of order r_c. On the other hand, resistances $r < r_c$ by themselves cannot provide transport to macroscopic distances. Thus, R^n is determined from the requirement that resistances $r \le r_c$ would constitute a fraction p_c of all resistances, that is

$$\int_{r_1}^{r_c} W^n(r)dr = p_c \,, \tag{1}$$

where $r_c \simeq R^n$. For simplicity, we shall not distinguish between r_c and R^n hereafter.

Let us assume some specific form for the distribution function $W^n(r)$. Following AHL, we take the junction resistance as

$$r = r_1 e^{\xi} \,, \tag{2}$$

where ξ is a random variable related to the distance between neighboring metallic grains. Equation (2) is reasonable since we view the barriers between grains as tunnel junctions. We will assume that ξ is uniformly distributed from zero to some maximum value $\ln(r_2/r_1)$:

$$W(\xi) = \begin{cases} 1/\ln(r_2/r_1), & \text{for } 0 < \xi < \ln(r_2/r_1) \\ 0, & \text{otherwise .} \end{cases} \tag{3}$$

From Eqs. (2) and (3), the distribution function for the junction resistors is

$$W^n(r) = \begin{cases} \dfrac{1}{r}\ln(r_2/r_1), & \text{for } r_1 < r_2 \\ 0, & \text{otherwise .} \end{cases} \tag{4}$$

In Eqs. (3) and (4), r_1 and r_2 are, respectively, the smallest and largest resistances of the junctions. Substituting this distribution into (1) we get

$$p_c = \ln(R^n/r_1)/\ln(r_2/r_1) \,. \tag{5}$$

For our film, we have estimated[8] that $r_1 \simeq 0.5\ \Omega$. Given this value for r_1 and $R^n \simeq 1\ \text{k}\Omega$, and assuming that $p_c = 0.5$, Eq. (5) yields $r_2/r_1 = 4 \times 10^6$.

B. Resistance in the superconducting state

Our picture of the superconducting film below the transition temperature of the grains T_g is one in which the phases of the superconducting energy gap fluctuate from one grain to another. As the temperature falls, successive pairs of grains become phase locked and their junction resistance vanishes. The temperature $T_J(r)$ at which a Josephson junction becomes superconducting depends upon the junction resistance r in the normal state and is determined by the condition[5]

$$E(r, T_J) = \gamma k_B T_J \,. \tag{6}$$

Here, γ is a constant of order unity and E is the Josephson coupling energy given by[9]

$$E(r,T)=\frac{\pi\hbar\Delta(T)}{4e^2r}\tanh\left[\frac{\Delta(T)}{2k_BT}\right], \quad (7)$$

where $\Delta(T)$ is the energy gap of the bulk superconductor.[10] In Eq. (6) we have neglected the charging energy of the grains. It follows from (6) and (7) that at a temperature $T<T_g$, all junctions whose normal-state resistance is smaller than

$$r_J(T)=\frac{\pi\hbar\Delta(T)}{4e^2\gamma k_BT}\tanh\left[\frac{\Delta(T)}{2k_BT}\right] \quad (8)$$

will be superconducting. The fraction $p(T)$ of superconducting junctions is then given by

$$p(T)=\int_{r_1}^{r_J(T)}W^n(r)dr=\frac{\ln[r_J(T)/r_1]}{\ln(r_2/r_1)} \quad (9)$$

and the critical temperature T_c for the onset of macroscopic superconductivity is given by setting $p(T_c)=p_c$ in (9). It follows from (5) that

$$r_J(T_c)=R^n\ . \quad (10)$$

The AHL model would imply a temperature-independent sheet resistance $R(T)=R^n$ for all $T>T_c$. As soon as T falls below T_c, $R(T)$ would be zero since we should now have an infinite cluster of short-circuited junctions. Clearly, as seen in Fig. 1, this is not what happens in our system. $R(T)$ decreases continuously from its normal-state value R^n for $T\geq T_g$ to zero at $T=T_c$.[11] Thus, to reproduce this continuous drop of $R(T)$, an approach different from AHL is needed. The approach that we use will be based on the effective-medium approximation.[12] We treat the problem in two stages. First, we replace the actual distribution $W^n(r)$ by a binary distribution

$$W^s(r)=p(T)\delta(r)+[1-p(T)]\delta(r-R^n)\ . \quad (11)$$

That is, the distribution of normal resistances is represented by a single, typical resistance. This typical resistance is chosen to be R^n to ensure, at least, the correct value for the normal state resistance (when $p=0$). The binary distribution (11) can be easily handled by the effective-medium approximation, the result being

$$R(T)=\left[1-\frac{p(T)}{p_c}\right]R^n\ . \quad (12)$$

This holds, of course, only for $p<p_c$, while for $p>p_c$, $R(T)$ is equal to zero.

From Eqs. (5), (9), and (12) it follows that

$$\frac{R(T)}{R^n}=\ln[R^n/r_J(T)]/\ln(R^n/r_1)\ . \quad (13)$$

Equation (13) gives the normalized sheet resistance of the film in the region $T<T_g$, $r_J(T)$ being given by (8). In Fig. 1 we plot the normalized resistance $R(T)/R^n$ as a function of reduced temperature T/T_g. T_g was taken to be that value which gave the best agreement between theory and experiment. In our case $T_g=6.72$ K. The solid curve in Fig. 1 gives the normalized resistance as predicted by (13). In evaluating (13) we must know the value of γ. The value of γ was fixed by requiring that $R(T_c)/R^n=0$ at a reduced temperature of 0.8 as seen from the experimental data in Fig. 1. Since at $T=T_c$ we have $r_J(T_c)=R^n$, (8) tells us that $\gamma=3.3$. It appears that for our particular choice of the distribution of ξ, and therefore $W^n(r)$, the theory predicts a more gradual decrease in resistance than is seen experimentally.[13] The result (13) for the normalized resistance in the range $T_c<T<T_g$ may be sensitive to the distribution[14] of ξ and to the effective-medium approximation. Near p_c, the resistance behaves as $R\sim(p_c-p)^s$ with $s\simeq1.3$, rather than $s=1$ as predicted by the effective-medium approximation.[15] Choosing s greater than one somewhat improves the agreement with experiment.

III. ULTRASONIC ATTENUATION IN GRANULAR FILMS

A. Normal state

Adler[3] has shown that the SAW attenuation in a homogeneous film is proportional to its sheet resistance. But for granular films, the predicted proportionality between the SAW attenuation and the sheet resistance does not hold quantitatively in the normal state. In our case, the normal-state attenuation is 4.4 dB/cm, whereas Adler would predict 2.3 dB/cm.[2] Furthermore, as seen in Figs. 1 and 2, the ultrasonic attenuation is still rather large even when the macroscopic sheet resistance of the film is equal to zero.

We propose a reinterpretation of Adler's result which will bring it into accord with the experimental data. We replace the macroscopic sheet resistance, which is the sheet resistance of an infinite network, by the average resistance of a square of side L cut out of the film. The reasoning behind this is as follows. The macroscopic resistance measures the response of the film to a dc potential, i.e., an ac potential of infinite wavelength. On the other hand, when a surface wave propagates, it produces an alternating current of finite wavelength, namely the acoustic wavelength. We may regard this alternating current as an ensemble of direct currents, each of which selects a section of the film and samples its local sheet resistance. The size of each "sample" should be $L\simeq\lambda/2\pi$, where λ is the wavelength of the SAW.[16] Furthermore, since the film is not homogeneous, the resistance $R(L)$ of each "sample" will change from location to location. Hence, it is the average sheet resistance $\overline{R}(L)$ of small sections of the film (each of which is a finite resistor network) and not the macroscopic sheet resistance of the infinite resistor network that governs the attenuation of the surface wave.

The problem at hand, therefore, is to try to determine $R(L)$ for $T>T_g$. More precisely, the problem is stated as follows. Given the distribution $W(r)$ for the individual resistors of the network, determine the distribution $W(L,R)$ of the resistances of squares of size L. There are two extreme cases.

(i) $L=L_0$ (a microscopic distance comparable to the grain size). In this limit the distribution is that for the individual resistors $W^n(r)$, i.e., Eq. (4).

(ii) $L \to \infty$. In this limit, $W(L,R)$ reduces to a δ function, $\delta(R - R^n)$, since in the thermodynamic limit, the normal-state sheet resistance is a well-defined quantity with no fluctuations.

In order to estimate $R(L)$ for intermediate L, we will use some of the ideas of Shklovskii and Efros.[17] These ideas are essentially an extension of the AHL argument to a finite network of size L. For convenience, we shall measure L in terms of the grain size so that a square of size L contains L^2 junction resistances. In the present experiments $\lambda \simeq 5$ μm which, with an average grain size of 500 Å, implies that $L \simeq 16$. An important difference—as compared to the case of the infinite network—is that now the percolation threshold $p_L = p_c(L)$ is not well defined but rather fluctuates from one finite resistor network to another. Indeed, it is only in the thermodynamic limit that percolation always starts strictly at $p = p_c$, i.e., when precisely a fraction p_c of the bonds become randomly "occupied." For a finite system, the percolation threshold p_L is a statistical variable distributed according to some distribution ϕ_L. This distribution has been studied numerically by Reynolds *et al.*[18] Even for L as small as 10, the distribution is close to a Gaussian[17]

$$\phi_L = \frac{1}{\sqrt{2\pi}\sigma_L} \exp\left[-\frac{(p_L - \bar{p}_L)^2}{2\sigma_L^2} \right] . \tag{14}$$

The width of the distribution is

$$\sigma_L = BL^{-1/\nu} , \tag{15}$$

where B is a constant of order unity and $\nu = \frac{4}{3}$ is the exponent for the correlation length in the two-dimensional percolation problem. When L increases, σ_L approaches zero while $\bar{p}_L$ approaches p_c as

$$\bar{p}_L = p_c + AL^{-1/\nu} , \tag{16}$$

where A is another constant of order unity. The constants A and B are not expected to be universal, i.e., they should depend upon the type of lattice as well as the details of the averaging procedure. We assume that the width of the distribution plays a more important role than the shift of $\bar{p}_L$ from p_c (i.e., we set $\bar{p}_L = p_c$) so that we only have one undetermined constant B. We fix B from the experimental $\bar{R}^n(L)$ (the average sheet resistance in the normal state) and use this value of B to determine $\bar{R}(L)$ (the average sheet resistance in the superconducting state).

Analogous to the infinite network, we define $r_c(L)$ to be the value of the crucial resistor for which percolation occurs in the finite network. As for the case of the normal-state resistance, we shall not distinguish between $r_c(L)$ and the normal-state sheet resistance of the finite network $R^n(L)$. According to Shklovskii and Efros, $R^n(L)$ is then a statistical variable given by the upper limit of integration in (1) when p_c is replaced by p_L on the right-hand side, p_L being distributed according to (14). Taking $W^n(r)$ from (4) and substituting into (1), we obtain

$$\ln[R^n(L)/r_1]/\ln(r_2/r_1) = p_L , \tag{17}$$

which, then solved for $R^n(L)$, yields

$$R^n(L) = r_1 \exp[p_L \ln(r_2/r_1)] . \tag{18}$$

The average sheet resistance in the normal state is then

$$\bar{R}^n(L) = \int dp_L R^n(L) \phi_L , \tag{19}$$

which, by (14) and (18), yields

$$\bar{R}^n(L) = R^n \exp\{ \tfrac{1}{2} [\sigma_L \ln(r_2/r_1)]^2 \} . \tag{20}$$

We see that when $L \to \infty$, $\sigma_L \to 0$ and $\bar{R}^n(L) \to R^n$ as it should. Furthermore, since the attenuation α is proportional to the sheet resistance, we have the following relation:[19]

$$\frac{\alpha(\text{expt})}{\alpha(\text{Adler})} = 1.9 = \frac{\bar{R}^n(L)}{R^n} . \tag{21}$$

Using this relation in (20), we find that $\sigma_L = 0.075$.

B. Superconducting state

Let us now estimate the average sheet resistance $\bar{R}(L,T)$ below T_g, i.e., when some fraction of the junctions are superconducting. To do this, we take (12) but replace p_c by p_L and R^n by $R^n(L)$. We then have

$$R(L,T) = \begin{cases} \left[1 - \dfrac{p(T)}{p_L} \right] R^n(L), & \text{for } p(T) < p_L \\ 0, & \text{otherwise} . \end{cases} \tag{22}$$

Its average, $\bar{R}(L,T)$, is then

$$\bar{R}(L,T) = \int_{p(T)}^{\infty} dp_L R(L,T) \phi_L ,$$

which by (14) and (22) becomes

$$\bar{R}(L,T) = \frac{r_1}{\sqrt{2\pi}\sigma_L} \int_{p(T)}^{\infty} [1 - p(T)/p_L] \exp\left[-\frac{(p_L - p_c)^2}{2\sigma_L^2} + p_L \ln(r_2/r_1) \right] dp_L . \tag{23}$$

Making the change in variable $x = p_L - p$ and completing the square in the argument of the exponential, we obtain

$$\bar{R}(L,T)/\bar{R}^n(L) = \frac{1}{\sqrt{2\pi}\sigma_L} \int_0^{\infty} \left[\frac{x}{x+p} \right] \exp\left[-\frac{(x + p - \tilde{p}_c)^2}{2\sigma_L^2} \right] dx , \tag{24}$$

where $\tilde{p}_c = p_c + \sigma_L^2 \ln(r_2/r_1) = 0.585$ and $p = p(T)$. Since the attenuation should be proportional to the average sheet resistance, (24) gives the normalized attenuation for $T < T_g$. We can get an analytic expression for (24) by considering the limits of p. For $p \geq \tilde{p}_c$, (24) can be written (see the Appendix) as

$$\bar{R}(L,T)/\bar{R}^{n}(L) \simeq \frac{\sigma_L}{\sqrt{2\pi} p} \exp\left[-\frac{(p-\tilde{p}_c)^2}{2\sigma_L^2}\right] - \left[\frac{p-\tilde{p}_c}{2p}\right]\left[1-\operatorname{erf}\left[\frac{p-\tilde{p}_c}{2\sigma_L}\right]\right], \quad (25)$$

and when p is somewhat smaller than $\tilde{p}_c$, (24) can be written as

$$\bar{R}(L,T)/\bar{R}^{n}(L) \simeq 1 - p/\tilde{p}_c \ . \quad (26)$$

Our theoretical results for the normalized attenuation are plotted in Fig. 2. We see that there is qualitative agreement between theory and experiment. The theory predicts that the attenuation should smoothly decrease to zero but will still be nonzero around T_c.

IV. SUMMARY

In summary, we have presented a percolation-theoretic model for the temperature dependence of the dc resistance and ultrasonic attenuation of a superconducting granular lead film. The film is treated as a random resistor network; the resistors of the network represent the insulating barriers between neighboring grains. In the normal state we assume that the values of these resistors are given by $r_1 e^{\xi}$, where ξ is a random variable taken to be constant over an interval from 0 to $\ln(r_2/r_1)$. This yields a distribution function $W^n(r)$ for the resistors to the network. As the temperature is lowered below the transition temperature of the grains T_g, neighboring grains whose Josephson coupling energy is greater than the thermal energy will become coupled. In the resistor network picture, this corresponds to short-circuiting those resistors which represent the junctions between the coupled grains. When the fraction of shorted junction resistors reaches the percolation threshold (at some temperature $T_c < T_g$) an infinite superconducting cluster appears in the network and the dc resistance drops to zero and remains zero for $T < T_c$. Furthermore, while the dc resistance measurements sample the resistance of the entire (infinite) network, the SAW is effectively measuring the average resistance of finite "subnetworks" whose dimensions are of the order of the acoustic wavelength. When macroscopic superconductivity occurs (as determined by the dc resistance measurement), there are still regions of the film which contain junction resistors that have not been shorted and these regions will produce an attenuation. Thus, one of the predictions of our model is that the ultrasonic attenuation should be nonzero even when the dc resistance is equal to zero.

Returning to Fig. 1, it appears that our model predicts a more gradual decrease in normalized resistance than is seen experimentally. Since the result (13) for the normalized resistance in the region $T < T_g$ is sensitive (to some extent) to the choice of the functional dependence of the random variable ξ, a more "realistic" choice for this function may give a sharper decrease in the normalized resistance. Also, as discussed immediately after Eq. (13), the critical exponent s for percolation is rather greater than unity and this will tend to reduce the resistance below that predicted by the effective-medium approximation. We are currently investigating the effect that different choices for the functional dependence of ξ would have on the resistance curves. We are also looking at the region near p_c in more detail.

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APPENDIX

Here we discuss the evaluation of the integral in (24). The key to the analytic treatment of the integral lies in the smallness of σ_L. Consider first the case when p is large, such that $p > \tilde{p}_c$. Then the argument of the exponential increases as we increase x from zero to infinity. The main contribution to the integral comes from the region of small x up to a few σ_L (for larger x, the integrand is already exponentially small). Thus, in this case, $x/(x+p)$ can be replaced by x/p since[20] $\sigma_L \ll p$, and (24) reduces to

$$\bar{R}(L,T)/\bar{R}^{n}(L) \simeq \frac{1}{\sqrt{2\pi}\sigma_L p} \int_0^\infty x \exp\left[-\frac{(x+p-\tilde{p}_c)^2}{2\sigma_L^2}\right] dx \ . \quad \text{(A1)}$$

Performing the change of variables $y = (x+p-\tilde{p}_c)/\sqrt{2}\sigma_L$, (A1) becomes

$$\bar{R}(L,T)/\bar{R}^{n}(L) \simeq \frac{\sigma_L}{\sqrt{2\pi} p} \exp\left[-\frac{(p-\tilde{p}_c)^2}{2\sigma_L^2}\right] - \left[\frac{p-\tilde{p}_c}{\sqrt{\pi} p}\right] \int_{y_1}^\infty e^{-y^2} dy \ , \quad \text{(A2)}$$

where $y_1 = (p-\tilde{p}_c)/\sqrt{2}\sigma_L$. The second term on the right-hand side of (A2) is related to the error function. Thus, we write (A2) as

$$\bar{R}(L,T)/\bar{R}^{\,n}(L) \simeq \left[\frac{\sigma_L}{\sqrt{2\pi}p}\right]\exp\left[-\frac{(p-\tilde{p}_c)^2}{2\sigma_L^2}\right]-\left[\frac{p-\tilde{p}_c}{2p}\right][1-\text{erf}(y_1)] \ . \tag{A3}$$

Now we consider the case when p is very small (at least several σ_L smaller than $\tilde{p}_c$). For small p the integrand of (24) has a sharp maximum (at $x=\tilde{p}_c-p$) within the integration region. In this case, (24) can be rewritten as

$$\bar{R}(L,T)/\bar{R}^{\,n}(L) \simeq \left[\frac{1}{\sqrt{2\pi}\sigma_L}\right]\left[\frac{\tilde{p}_c-p}{\tilde{p}_c}\right]\int_0^\infty \exp\left[\frac{(x+p-\tilde{p}_c)^2}{2\sigma_L^2}\right]dx \simeq 1-p/\tilde{p}_c \ . \tag{A4}$$

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[19]In Eq. (21), α(Adler) has been calculated assuming an "effective" dielectric constant $\epsilon \simeq 36$. $LiNbO_3$ is very anisotropic. The appropriate "effective" dielectric constant is $\epsilon=(\epsilon_{\parallel}\epsilon_{\perp})^{1/2}$ [see, e.g., S. Kino and T. M. Reeder, IEEE Trans. Electron Devices, ED-**18**, 909 (1971)]. Under stress-free conditions $\epsilon \simeq 36$, but under strain-free conditions the value is $\simeq 50$ [see, e.g., V. M. Ristic, *Principles of Acoustic Devices* (Wiley, Toronto, 1983), Chap. 6]. It is not obvious to us which of these extremes is closer to a realization of the experimental conditions. However, some Monte Carlo simulation experiments currently in progress give tentative support to our fit of the SAW data based on the lower value.

[20]Actually, such replacement would still be valid for somewhat smaller p such that $(p-\tilde{p}_c)<0$ but $p-\tilde{p}_c \ll p$. In this case the argument of the exponential first decreases to zero (at $x=\tilde{p}_c-p$) and then increases. The main contribution to the integral is still coming from the region of small x.