## AMPLITUDE -DEPENDENT ULTRASONIC ATTENUATION IN SUPERCONDUCTING LEAD\*

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During a study of the ultrasonic attenuation in several superconductors, an unexpected amplitude dependence of the attenuation was observed in lead. Love and Shaw, independently and earlier, noted this phenomenon while studying the superconducting energy gap in lead with ultrasonic waves.<sup>1</sup> The interpretation of our original measurements made necessary an extensive study of the amplitude dependence, and an explanation of the effect is proposed here.

A large ultrasonic attenuation due to electronphonon interactions is well known to occur in pure metals when the temperature is lowered into the liquid-helium range.<sup>2</sup> In superconductors, this mechanism is suddenly quenched at the transition temperature; and according to the Bardeen-Cooper-Schrieffer theory, this drop in attenuation reflects the temperature dependence of the ratio of "normal" to "super conducting" electrons. With the application of the critical magnetic field, the normal conducting state is restored, and the ultrasonic attenuation continues to rise with decrease in temperature until it becomes constant near 1'K. Most superconductors of sufficient purity follow, in general, this same pattern of behavior, differing characteristically only in the value of the superconducting transition temperature. In contrast, the ultrasonic attenuation in lead shows a significant amplitude-dependent deviation in the superconducting state, but remains true to the general behavior in the normal conducting state, as is illustrated in Fig. 1. The normal-state attenuation and the dashed curve for the superconducting state represent the typical behavior of most superconductors. The experimental curve in the superconducting state shows the attenuation at an intermediate amplitude of the ultrasonic wave. If the power in the ultrasonic wave is varied, a corresponding amplitude dependence in the attenuation results. This amplitude dependence is hardly noticeable above the superconducting transition temperature, but increases suddenly with the onset of superconductivity and becomes very strong near  $1^\circ K$ , as is seen in Fig. 2.



FIG. 1. Temperature dependence of ultrasonic attenuation at medium amplitude in 99.999% pure singlecrystal lead.

This anomalous behavior, in particular the strong influence of temperature on the amplitude dependence of the ultrasonic attenuation, is not explicable in terms of a simple modification of the Bardeen-Cooper-Schrieffer theory. One mechanism which is known to give strong amplitude-dependent effects at high temperatures —dislocation damping —seems to be ruled out at first glance because of its absence in the normal conducting state. A consideration of the influence of the conduction electrons on dislocations, however, suggests a plausible model for the absence of dislocation damping in the normal state and its presence in the superconducting state. According to this model, the conduction electrons behave as a viscous gas, and the dislocation lines as vibrating strings,



FIG. 2. Temperature dependence of attenuation in 99.999% pure single-crystal lead.

whose motions are damped by the gas. With the onset of superconductivity and as the temperature is lowered further, the number of "normal" electrons decreases in favor of the number of Cooper pairs, which are assumed not to interact with the lattice. Consequently,

the dislocations become increasingly free to move and to engage in the mechanism leading to amplitude dependence of the ultrasonic attenuation. Such a mechanism has been proposed by Granato and Lücke<sup>3,4</sup> and is well supported experimentally at higher temperatures and lower frequencies.

According to their model, the dislocations are pinned down by impurity atoms and by the nodes of the dislocation network; in most cases, impurities will pin the dislocations at more frequent intervals than the network nodes. An incident ultrasonic wave causes a bowing out of the dislocation loops of average length  $L_c$ . However, if the applied stress is increased sufficiently, the dislocations are tom away from the impurity atoms. This unpinning process is assumed to be also assisted by the thermal phonons. The dislocations are now pinned only at the nodes of the dislocation network and vibrate with length  $L_n$ . The unpinning process absorbs ultrasonic energy which is estimated at 0.1 eV per impurity atom. The higher the sound-wave amplitudes, the more unpinning occurs and the higher is the damping. Granato and Lücke make two simplifying assumptions<br>-the distance  $L_n$  between network nodes is constant for a given sample specimen, and the distribution of the impurities along the dislocations follows an exponential law. They obtain for the ultrasonic attenuation

$$
\Delta_H = \frac{\Omega \Lambda L^3 n}{\pi^3 L_c} \left( \frac{Kna}{L_c \epsilon_0} \right) \exp \left( - \frac{Kna}{L_c \epsilon_0} \right)
$$

where  $\Omega$ , K, n, and  $\alpha$  are crystal structure factors,  $\Lambda$  is the dislocation density, and  $\epsilon_0$ is the maximum value of the oscillating strain. Thus, the amplitude dependence of the dislocation damping is strong, indeed, and depends characteristically on the exertion of sufficient stress on the dislocation to allow unpinning processes. One would expect a large damping of the dislocation motion in the normal state when the rexlaxation time for electron-dislocation interaction is comparable to the vibrational period of a dislocation loop at resonance.

An estimate of this relaxation time was made under two main assumptions'. The electrons interact with the stress field of a dislocation and not with its core directly; and the mean free path of the electrons is very long so that they may be described by Bloch-type wave functions. A formula for the resonant frequency of a dislocation loop has been derived by Mason.<sup>6</sup> The so-calculated vibrational period is of the order of magnitude of our estimated relaxation time.

A variety of experiments were conducted to test the identification of the observed amplitude dependence of the ultrasonic attenuation with the Granato-Lücke mechanism. A Granato-Lücke plot was constructed which examines the qualitative agreement of the observed amplitude dependence with their expression for the ultrasonic attenuation. Figure 3 shows the expression again and the data points. The Granato-Lücke theory would predict that the amplitude dependence should diminish with an increase in the impurity concentration. Measurements on 99.9% pure lead crystals doped with tin as impurity show a strong reduction in amplitude dependence. The theory also predicts that the effect should be frequency independent. Within the accuracy of our measurements, the amplitude dependence remains, indeed, approximately constant with frequency. The ranges studied were 30 Mc/sec to 1.0 Gc/sec for the impure lead and 30 to 150 Mc/sec for the pure



FIG. 3. Granato-Lucke plot of the ultrasonic attenuation of 99.999% pure single crystal at 4.2'K.

lead. Preliminary experiments of deforming the crystal, i.e., increasing the dislocatio density, show an enhancement of the amplitude dependence for moderate deformations and a reduction for large deformations. In the latter case, it is assumed that the dislocation density  $\Lambda$  becomes so great that for many dislocations the node distance  $L_n$  is smaller than  $L_c$ , the impurity pinning distance. As a result, the number of unpinning processes is drastically reduced and the amplitude dependence becomes very weak.

So far no investigation has been made about the nature of pinning-point imperfections. The pinning force has to be weak in order to explain the breakaway at such low temperatures where the assistance by thermal phonons is almost negligible.

A detailed experimental and theoretical account of this effect will be published soon.

The authors wish to express their gratitude to Dr. Paul Shlichta for orienting several of the samples. The authors also wish to thank

Mr. Floyd Lacy for his help, especially in the sample preparation. One of us (B.R.T.) would like to thank Professor W. F. Libby for support out of National Aeronautics and Space Administration Grant No. NASA NSG-237 during part of this work.

\*This research is supported in part by Grant No. NSF GP 2391 from the National Science Foundation.

 ${}^{1}R$ . E. Love and R. W. Shaw, Rev. Mod. Phys. 136, 260 (1964).

<sup>2</sup>R. W. Morse, Progress in Cryogenics, edited by K. Mendelssohn (Academic Press, Inc., New York, 1959), Vol. I, pp. 221-259, for example.

 $3A$ . Granato and K. Lucke, J. Appl. Phys. 27, 583 (1956).

 $4A.$  Granato and K. Lucke, J. Appl. Phys. 27, 789 (1956}.

 $5$ The authors are indebted to Professor T. Holstein for the calculation of the electron-dislocation relaxation time.

 $^{6}$ W. P. Mason, Bell System Tech. J. 34, 903 (1955).

## POSSIBILITY OF LONG-RANGE SPIN POLARIZATION IN A DEGENERATE ELECTRON GAS\*

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In a recent issue of this journal, Dreyfus, Maynard, and Quattropani' have arrived at the conclusion that under certain circumstances it is possible for an induced long-range spindensity distribution to exist in a nonferromagnetic metal. This novel result is surprising when regarded from the standpoint of the theory of Ruderman and Kittel<sup>2</sup> and Yosida.<sup>3</sup> In this theory the spin density generated at large distances from a localized spin perturbation is of an oscillatory nature. Dreyfus, Maynard, and Quattropani suggest that the difference between their result and that of Ruderman and Kittel results from the nonperturbative nature of the problem which they treated. But we find this explanation not completely convincing, as the behavior of the free-electron gas in the asymptotic region at considerable distance from the disturbance would nevertheless be expected to be characteristic of an only slightly perturbed degenerate gas, regardless of the strength of the distant perturbation.

For this reason we have re-examined the calculation of Dreyfus, Maynard, and Quattro $pani<sup>1</sup>$  with the goal of reconciling it with the Ruderman-Kittel theory. In particular, we have evaluated an additional contribution to the total spin density which, although discussed by Dreyfus, Maynard, and Quattropani, was evidently not computed by them. This is the spin density contributed by the "nonevanescing waves." The motivation of our calculation was to explore the possibility that the nonevanescing waves might contribute a long-range polarization which just cancels that coming from the "evanescing waves" —thereby leaving only