Penetration depth in proximity-effect superconductors

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We have studied magnetic field penetration into the normal-metal side of proximity-effect sandwiches by measuring the periodic magnetic field dependence of the dc Josephson current in tunnel junctions containing the normal-metal/superconductor bilayers. The systems studied include the bilayers Ag/Pb, Al/Pb, Ag/Sn, and Sn/Pb and the trilayer sandwich Ag/Sn/Pb, in the temperature range down to 0.1 K. We find that the magnetic field is screened out of the normal metal in much the same way as for a type-I superconductor, with a penetration depth that becomes independent of the normal-metal thickness, or the temperature, at sufficiently low temperature. The magnitude of the induced normal-metal penetration depth is a function of the normal-metal parameters and the inverse of the gap in the backing superconductor.

I. INTRODUCTION

Over the past two decades there has been a considerable amount of experimental and theoretical work done on the superconducting proximity effect, the induced superconductivity in a thin normal metal placed in electrical contact with a superconductor.¹⁻⁶ While there are no general microscopic theories of the proximity effect, there are microscopic theories in certain limiting cases,⁷⁻⁹ and the general phenomenological Ginzburg-Landau theory,¹ which has been quite successful in qualitatively describing most situations. The theoretical treatments emphasize that the basic difference between bulk and proximity superconductivity is the inhomogeneous nature of the order parameter in the latter, and produce models of the variation of the order parameter with distance from the normal-metal/superconductor boundary. However, the great majority of proximity-effect experiments to date measure either local properties at the free-normal-metal boundary (tunneling gap,¹⁰ Josephson current amplitude¹¹) or average properties (such as the reduction of T_c and the critical fields¹²).

We have investigated magnetic screening in proximityeffect superconductors in order to compare the behavior of these systems with that of ordinary superconductors and to investigate the spatial dependence which is elusive in other experiments. A rather novel approach using Josephson tunneling allowed a direct measurement of the magnetic penetration depth in the proximity metal.

In previous work on this problem,¹³ we presented data on Ag/Pb proximity sandwiches in the temperature regime above 1 K. We also presented a method for analyzing the results in terms of a self-consistent numerical solution for the magnetic field in the case of a spatially varying pair potential. Our results indicated that there was an "intrinsic" penetration depth for the Ag films 140–160 nm thick (from extrapolation of penetration depth versus temperature data to low temperature) which was independent of the thickness of the Ag films. The previous work raised two questions: (1) was the extrapolation to low temperatures correct (was the model sufficiently accurate?), and (2) what determined the "intrinsic" penetration depth, the Ag itself or both of the metals in the proximity sandwich? Here we attempt to answer these questions.

In Sec. II we describe the experimental techniques as they differ from those of Ref. 13. In Sec. III we present the experimental results on Ag/Pb sandwiches below 1 K. In Sec. IV we present the studies of several other bilayer configurations. In Sec. V we compare our results with other proximity studies and present theories.

II. EXPERIMENTAL TECHNIQUES

The systems studied in this work consisted of Ag/Pb, Al/Pb, Ag/Sn, and Ag/Sn/Pb sandwiches. A number of techniques were employed for these systems and will be described below.

All of the junctions studied in this work used tin as a base electrode. The technique used to create tunneling barriers on many of the samples measured in this work is that of dc glow discharge.¹⁴ To oxidize the tin, we applied about 1000 V dc to an aluminum ring electrode in the evaporation chamber with an oxygen pressure of about 50 mTorr for exposure times on the order of 1 min.

The chief advantage of the glow-discharge technique is that the sample need not be removed from the evaporation system, which speeds up junction fabrication. The main drawback of the technique is that, like thermal oxidation, it gives no feedback while the oxidation progresses.

Perhaps the most difficult aspect of producing the oxide barriers for these junctions is that in order to observe an appreciable Josephson current when the junctions involve very thick ($\sim 1 \ \mu m$) proximity metals, extremely thin oxide barriers are needed. As a result, the oxidation

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must be controlled very precisely, and one needs an extraordinary uniform barrier (devoid of discontinuities or shorts). The glow-discharge technique becomes less effective in this case because of the competitive process in which the ions are energetic enough to "clean off" the oxide as it is formed.

Two solutions to the problem of producing thin, uniform oxide barriers were found, one related to the base electrode and the other using a novel technique for oxidization.

It has been known for some time¹⁵ that favorable structure in thin films is most easily achieved when deposition is performed at an optimum temperature specific to the material being deposited. The optimal temperatures for some common materials include 200 K for Pb, 300 K for Al, and 180 K for Sn. It was found in this work that there was indeed far greater success in producing uniform tin films (and, subsequently, high-quality dielectric barriers) when deposition was done at this reduced temperature.

An improved tunneling barrier was still needed. In collaboration with Wolf, the technique of chemical anodization was applied to this problem.¹⁶ The base electrode is immersed in an ion-rich solution composed of boric acid and ammonium hydroxide to yield a neutral pH solution. The acid provides a source of oxygen that takes part in an electrostatically driven oxidation process much like glow discharge but involving ions of far lower energy. A fixed dc current is applied to the circuit, and the voltage across the two electrodes is continuously monitored. This voltage (V) is related to the constant electric field across the barrier by the relation $V = \mathscr{C}d$; thus the voltage is a direct measure of the oxide thickness (d).

In all cases, fabrication began with vapor deposition of tin in a moderate vacuum $(1 \times 10^{-7} - 5 \times 10^{-6} \text{ Torr})$ onto either whole or cut, standard glass microscope slides. As discussed above, deposition was done with the substrate at a reduced (< 200 K) temperature. The glass substrate was placed in contact with a smooth copper surface maintained at ~80 K by circulating liquid nitrogen. The tin film width was kept on the order of 100 μ m to avoid self-screening effects of the Josephson current.

After the tin deposition, the samples were warmed by flowing room-temperature air through the same copper block. At this point, a thin (70 nm) film of SiO_2 was electron-beam-evaporated in such a way that a pattern of well-defined junction areas was laid out. The film was oxidized, either by glow discharge or by anodization. Finally, the normal-metal/superconductor layers were evaporated. The normal metal (silver, aluminum, or tin) was electron-beam-evaporated at a slow rate (2 nm/sec), and immediately afterwards the backing superconductor (lead or tin) was evaporated from a resistive boat.

In the case of a tin-lead sandwich, care was taken to avoid interdiffusion of the two metals by evaporating the lead at a reduced temperature (again by the use of the cooled cooper block). Such samples were kept cold throughout the steps of removal from the evaporator, mounting to the measurement probe, and installation into the cryostat.

Film thicknesses were monitored and electron-beam-



FIG. 1. Josephson current vs magnetic field in 5000 Å Ag/4000 Å Pb junction.

deposition rates were controlled by the same Sloan 9000 rate controller and thickness monitor. The system was calibrated using an optical interferometer. In all cases, the materials used for the film fabrication were 6-9's grade.

Measurements were performed on an electronic apparatus previously described,¹⁷ which records the maximum Josephsosn current as a function of field. The effective penetration depths were deduced from the Fraunhofer patterns characteristic of the Josephson current-versus-magnetic-field curves by averaging over 10 or more periods. Some samples displayed as many as 50 maxima on each side of zero field. Figure 1 shows data for such a sample. Junctions exhibiting anomalous field dependencies were rejected for his study. High-





quality field-dependence patterns such as seen in Fig. 2 confirm the uniformity of the oxide barrier and the general conformance of the junction to the theoretical interpretation of the periodic pattern.

III. Ag/Pb SANDWICHES

In our previous study of Ag/Pb films above 1 K we found a screening behavior best illustrated by Fig. 5 of Ref. 13. The qualitative description is as follows.

At low thickness, the normal metal is too thin (compared to its characteristic screening length) to screen the applied field, and most of the screening takes place in the lead film which is behind it (λ_{Pb} =40 nm). At large thicknesses the magnetic field penetrates deeply into the normal metal, but is excluded from a region near the interface by screening in the normal metal.

The most interesting aspect was the suggestion of a plateau region in the intermediate-thickness region (200-400 nm). It appears that a penetration depth independent of sample thickness is characteristic in this range.

On the basis of these results alone, it was possible to formulate a theoretical treatment that accounted for these observations.¹³ On the experimental side, however, it still remained to be seen whether the very-low-temperature behavior suggested by these above-1-K results was in fact to be observed; was the characteristic length for each thickness really the same value as the temperature approached zero?

To answer this question about low-temperature screening, we measured junctions in a dilution refrigerator. Junctions were fabricated in the reduced geometry necessary to fit the dilution-refrigerator probe. In other respects, the measurement techniques were identical to the high-temperature experiments.

As Fig. 3 clearly shows, the observed penetration depth for 600-nm, 800-nm, and $1-\mu m$ silver films was essentially identical at the lowest temperatures. The three sets of data all line up at approximately 150 nm as the temperature goes to zero.

The obvious interpretation of this data in conjunction with the high-temperature results is that there is a characteristic penetration depth for proximity superconducting



FIG. 3. Temperature dependence of the magnetic field penetration of Ag/Pb junctions with different Ag thicknesses. Points are data and lines are calculated.

silver of 150 nm. Such a value becomes apparent only at sufficiently low temperature. Furthermore, the thicker the silver film, the lower the temperature must be in order to reach this saturation value. This is evident in our earlier work from the very different temperature dependencies of the various thicknesses measured.

Note that this characteristic length of 150 nm is considerably greater than any of the other penetration depths in the system-40 nm for lead and 55 nm for tin-and is considerably greater than the London-theory value for silver (25 nm). One explanation for this discrepancy is the possibility that the silver film is dirty-that its mean free path is small. To investigate this possibility, additional leads were attached to the silver films in order to make resistivity-ratio measurements from which mean free paths could be estimated by the technique of Toxen, Burns, and Quinn.¹⁸ The results showed mean free paths on the order of 500-800 nm, which certainly do not fall into the category of dirty films. This point will be addressed again in the next section, but it suffices to say here that the inclusion of mean-free-path effects cannot account for the discrepancy between the London value for silver and the observed value.

The fact that strong screening is observed in thick (1 μ m) proximity superconductors raises the question of just how thick the silver films could be and still demonstrate such screening. The theoretical treatment developed in our earlier work demonstrate that for sufficiently low temperatures, *any* thickness of silver should exhibit the same characteristic penetration depth. However, the theory is essentially a zero-field theory. The inclusion of finite fields should certainly have strong effects upon such weakly induced superconductivity; presumably, the magnetic screening itself should become field dependent at even relatively small fields for sufficiently thick films.

A number of attempts were made to observe fielddependent screening in still thicker films. The difficulty is that the induced gap (and the induced pair amplitude at the free surface) in the normal metal becomes smaller and smaller with increased normal-metal thickness. Since the Josephson current is proportional to the pair amplitude at the free surface of the normal metal, it becomes more and more difficult to observe. To compensate for this, lower junction resistances are needed. Unfortunately, even exceedingly low resistances (<0.0005 Ω) did not yield junctions with measurable Josephson currents, for $d > \sim 1$ μ m.

IV. OTHER BILAYERS AND TRILAYERS

The characteristic penetration depth in the Ag/Pb films could come about in three distinct ways: it could be determined by the silver alone, it could be determined by the fact that lead is inducing the superconductivity, or it could be characteristic of the Ag/Pb system. To sort out these three possibilities, several other experiments were performed.

Junctions were prepared using aluminum as the proximity superconductor. The measurements were made above the T_c of the aluminum.

We found virtually no thickness or temperature depen-

dence for the penetration depth observed in the aluminum, but the magnitude of the penetration depth (90 nm) was not the value characteristic of bulk aluminium (50 nm). Figure 4 shows the measured penetration for four thicknesses of aluminum and demonstrates only a slight thickness dependence. We note that the energy gap in the aluminum proximity samples is greater than that for bulk aluminum (0.17 mV); for example, the measured gap in the 800 nm sample was 0.23 mV.

The minimal thickness and temperature dependence is consistent with the usual Ginzburg-Landau treatment of the proximity effect in materials that have a finite T_c . For such materials, the temperature dependencies of the various superconducting properties are determined by $T-T_c$ rather than by the temperature itself. Thus, the aluminum samples at 1.2 K should demonstrate the same behavior as silver films near absolute zero.

The fact that the characteristic penetration depth into proximity aluminum is quite different from that of silver demonstrates that the measurement cannot simply be an artifact of the lead. On the other hand, the lack of agreement with the standard value for aluminum does not rule out the possibility that the value is characteristic only of the aluminum and not the Al/Pb sandwich. The smaller discrepancy in values for this case cannot rule out meanfree-path effects as an explanation.

Another question raised by these aluminum measurements was the effect of a backing superconductor (such as lead) on an intrinsically superconducting proximity metal (such as aluminum) when it is below its T_c . It was experimentally more practical to pursue this issue by investigating tin as the proximity metal rather than aluminum.

Several samples were fabricated such that some of the junctions on a slide where $Sn/SnO_x/Sn$ and some were $Sn/SnO_x/Sn/Pb$. In this way, the effects of the backing superconductor on the tin film could be compared directly with tin films of the same thickness (and presumably all other properties) that were not backed by lead. Since interdiffusion between tin and lead is appreciable at room temperature, steps had to be taken to diminish its effects. Hence, the samples were prepared by cooling the tin film on the substrate holder while evaporating the lead, and keeping the samples at nitrogen temperature throughout the removal from the cryostat.

The result of this experiment was that the presence of the lead had no effect upon the screening at all. The ener-



FIG. 4. Observed penetration vs aluminum thickness for Al/Pb junctions.

gy gap in the tin was enhanced as expected by the proximity effect (0.74 mV rather than 0.6 mV in the 400 nm of tin), but the penetration depth in the tin did not change. Since all the measurements were taken well below the T_c of the tin (3.7 K), no temperature dependence was observed. This is evidence for the claim that the screening is determined only by the proximity metal itself. On the other hand, tin is itself a superconductor and might exhibit different properties for that reason alone. The next step was to substitute for the lead as the backing superconductor.

Junctions were prepared with Ag/Sn layers and magnetic penetration measurements were made. Junctions of 250 and 400 nm of silver backed by tin show practically no temperature dependence and a very large penetration depth (essentially no screening at all). The measured penetration depths were, in fact, the silver thicknesses themselves. However, a thicker (600 nm) film did demonstrate behavior qualitatively similar to the Ag/Pb films. The temperature dependence for this junction was quite similar to the counterpart Ag/Pb junction, but, as Fig. 5 shows, the magnitude of the penetration depths were quite different. In fact, extrapolating the temperaturedependence curves for these samples using the Ag/Pb samples as a guide indicates a characteristic depth of about 350 nm. Therefore, the thinner films behave like Ag/Pb films thinner than 150 nm. The characteristic length is simply greater than the film thickness in this case and much of the screening really occurs in the backing superconductor.

Therefore, the characteristic penetration depth measured is *not* simply a function of the normal metal alone. Substituting tin for lead makes a major difference in the observed screening properties.

The contribution of the superconductor could be a metallurgical one—due to the normal-metal properties of the superconductor—or one directly due to the superconducting properties of the superconductor. To test this, junctions were prepared containing a trilayer normal-metal/superconductor-1/superconductor-2 rather than a simple normal-metal/superconductor sandwich. An Ag/Sn/Pb sandwich was used in which the Sn layer was only 50 nm thick. With such a thin tin layer, the gap in-



FIG. 5. Observed penetration vs temperature for 6000 Å silver backed by tin.



FIG. 6. Observed penetration vs temperature for 6000 Å silver in Ag/Pb (top), Ag/Sn/Pb (middle), and Ag/Sn (bottom) sandwiches.

duced in the tin by the lead is quite large ($\sim 1 \text{ meV}$), so that the silver acts as if it were backed by nearly as strong a superconductor as pure lead. The purpose for this experiment is to use a superconductor to back the silver whose gap is between the gaps of tin and lead. Once again, the low-temperature deposition and mounting technique was used for the tin-lead contact required.

The result was that once more the temperature dependence was qualitatively similar to the previous experiments, but the magnitude of the penetration depth was different (see Fig. 6). This time, the characteristic penetration depth could be extrapolated to approximately 200 nm, a value between the tin and lead values.

The important parameter appears to be the energy gap of the superconductor backing the normal metal. The gap in lead (1.38 meV) is approximately 2.3 times that of tin (0.6 meV), and the penetration depth appears to scale inversely with this ratio. The value measured for the trilayer scales approximately with the induced gap in the Sn/Pb sandwich serving as the backing superconductor.

V. DISCUSSION

In our previous work we presented a method for selfconsistently calculating the magnetic field profile of a normal-metal/superconductor sandwich (or any superconductor with a spatially varying pair amplitude in one dimension). The resultant differential equation for the field with the explicit spatial dependence included is

$$\frac{d^2 H(x)}{dx^2} = \frac{1}{\lambda_0^2} \frac{\cosh[(d_n - x)/\xi_n]}{\cosh^2(d_n/\xi_n)} H(x) + \frac{2}{\xi_n} \tanh\left[\frac{d_n - x}{\xi_n}\right] \frac{dH(x)}{dx}, \quad (1)$$

which is a formidable equation to deal with. The most practical approach to solving this equation proved to be a numerical one. The "cosh" terms represent an approximation for the pair amplitude which gives a local penetration depth of the form

$$\lambda(x) = \lambda_0 \frac{\cosh(d_n/\xi_n)}{\cosh[(d_n-x)/\xi_n]} .$$
⁽²⁾

The numerical solutions require two parameters: the value of λ_0 and the temperature-independent prefactor of the coherence length in the normal metal. The two factors were fitted in the following way.

The data for penetration depth versus silver thickness (Fig. 5 in Ref. 13) is used. Since all the experimental points are at the same temperature, adjusting the two values of λ_0 and the coherence length to best fit the experimental points is a relatively simple procedure. The temperature dependence of the coherence length itself can take two forms: a dirty-limit or a clean-limit expression. Mean-free-path data suggest that neither limit is strictly applicable; the dirty-limit dependence seems to work better.

The dirty-limit expression for the coherence length in proximity silver,

$$\xi(T) = \left(\frac{hv_f l}{6k_B T}\right)^{1/2},\tag{3}$$

yields a value of about 600 nm for films at 1.2 K. Starting from this point, the experimental data is best fitted by a value of about 615 nm, which is within 2% of the expected value.

 λ_0 has already been seen to be on the order of 140–160 nm from the high-temperature data in Ref. 13, and has been shown to be 150 nm by the low-temperature data of Fig. 3.

Therefore, the two parameters needed for the calculation can be deduced in a rather straightforward way from a combination of data and theory. The temperaturedependence data for the Ag/Sn and Ag/Sn/Pb films is also well explained by this numerical treatment. The data are superimposed along with theoretical plots in Figs. 5 and 6. Clearly, the temperature dependence for films of the same silver thickness is scaled only by the λ_0 parameter. The best fits are achieved by the values 338 nm for the Ag/Sn sample and 195 nm for the Ag/Sn/Pb sample. Fitting the temperature-dependence curves allows for a tolerance of about 5% for these values, which is quite accurate enough to characterize how these values depend upon superconducting and normal-metal properties.

The high-temperature end of the Ag/Sn data appears to be flattening out, which corresponds to the penetration depth saturating to the film thickness. Figure 5 shows this data superimposed upon a theoretical plot extended out to 3 K, which clearly demonstrates this phenomenon.

The numerical theory also fits the Al/Pb data at 1.2 K, as Fig. 4 shows. The only thickness dependence apparent at this temperature is at the low-thickness end, which corresponds to the screening saturating in the lead film.

This numerical calculation demonstrates that, unlike the usual tunneling measurements that probe superconducting properties at the free surface, the characteristic penetration depth samples a large region of the proximity metal. We can easily see this by comparing the temperature dependences of two related quantities: the critical Josephson current, which is a measure of the pair amplitude at the tunneling barrier, and the inverse of the characteristic penetration depth, which is determined by the pair amplitude averaged in some sense throughout the material. According to the Werthamer spatial dependence,¹⁹ the latter should not be as sensitive to temperature as the former.

Figure 7 shows these two quantities normalized to their 1-K values and plotted against temperature for a 250-nm Ag/Pb sample. Clearly, the decay of the pair amplitude is more rapid than that of the inverse pentration depth, which is precisely what is expected.

In summary, the numerical calculation of the magnetic field profile in the proximity sandwich yields good agreement with the experimental observations of magnetic penetration in silver. Both the thickness and temperature dependence are accounted for the Ag/Pb, Ag/Sn, and Ag/Sn/Pb systems. Furthermore, this behavior is quite different from that observed previously, although it does not contradict the earlier observations.¹² This is due to the fact that the thickness and temperature range for most of our experiments were outside the range of the Orsay results.

Nevertheless, the most important parameter in the problem—the characteristic penetration depth—is empirically fitted to the data and not predicted by the theory. It is to this point that the remaining discussion will be devoted.

The experimental results of this work give a clear indication of what must be the form of an appropriate theoretical formulation of the problem.

(a) The penetration depth is determined by a combination of normal-metal and superconductor properties. While a given normal-metal and superconductor couple at low temperature exhibit a characteristic penetration depth that is independent of normal-metal-layer thickness and temperature, changing either member of the couple results in a different penetration depth.

(b) The penetration depth is inversely proportional to the energy gap of the superconductor. Table I lists the



FIG. 7. Temperature dependence of the critical Josephson current and the inverse of the observed penetration depth for a 2500-Å silver film backed by lead.

TABLE I. Scaling of effective penetration depth with energygap of the superconductor.

	Ag/Pb	Ag/Sn/Pb	Ag/Sn
λ_n (Å)	1500	1950	3375
$\lambda_n / \lambda_{n Pb}$	1	1.33	2.26
Δ_s (mV)	1.37	1.0	0.6
$\Delta_{\rm Pb}/\Delta_s$	1	1.36	2.28
Ratio of both	1	1.03	1.01

penetration depths, gaps, and ratios of these parameters in Ag/Pb, Ag/Sn/Pb, and Ag/Sn films. The penetration depth scales precisely with the gap, which conclusively demonstrates that the gap comes in inversely without any additional exponent.

(c) According to the results in Sn/Pb films, the enhancement of the energy gap in the proximity metal due to the presence of the stronger superconductor (Pb) does not result in a lower penetration depth. This is consistent with the so-called Gor'kov-Goodman-type saturation²⁰⁻²² in the superconducting properties.

Thus, many of the features of a successful theoretical description of the problem are already present in the high-temperature theory. In particular, the Orsay¹² expression for $\xi(x)$ contains both normal-metal and superconductor properties and scales inversely with the gap in the superconductor. The theory breaks down in the temperature dependence, but probably, as is often the case with Ginzburg-Landau results, yields more information than the assumptions underlying it should allow.

In a proper treatment of the problem one must return to the fundamental expression for the current stated in terms of the Green's function in the normal metal.^{23,24} At low temperatures the coherence length in these materials is much larger than the penetration depth; therefore nonlocal electrodynamics is required for a proper description of the system. Nonlocality always results in weaker screening.

Therefore, at this point, an explicit form for the Green's function at zero temperature should be used, corrections for the anisotropy of the order parameter built into the Green's function, and an additional integration for nonlocal effects performed. Such a calculation is undoubtedly the only way to obtain quantitative agreement with the saturation values of the penetration depth observed in this work. A theoretical calculation of this type is currently underway.²⁵

Quantitatively, there is very little data in other work that can be directly compared to these measurements. All other measurements have yielded either indirect or deduced values for screening lengths. The Orsay measurements¹² cannot be translated into penetration-depth measurements because the film thicknesses were not measured. The only other measurement of the penetration depth in a silver-lead system was from ultrasound measurements.²⁶ The coefficient of the $T^{1/2}$ dependence of λ for their data was 117 nm for films moderately dirtier than those in this research. When mean-free-path effects are included, there is reasonable agreement with the current results.

VI. CONCLUSION

These measurements represent the first direct measurement of the magnetic penetration depth in proximity superconductors. The results clearly demonstrate that the screening is not a phenomenon peculiar to the vicinity of the normal-metal/superconductor interface, but instead approaches the magnetic field response of ordinary type-I superconductors.

The primary difference between proximity and intrinsic superconductors comes from the inhomogeneity of the induced superconductivity. The order parameter is spatially varying, and the superconductivity weakens with distance from the normal-metal/superconductor interface. As the effects of inhomogeneity are accentuated—as by raising the temperature or increasing the normal-metal thickness—the magnetic behavior departs from that of ordinary superconductors. However, as the inhomogeneity is diminished—by lowering the temperature or keeping the film thickness low—the proximity superconductor acts more like an ordinary superconductor.

A numerical calculation for the thickness and temperature dependence of the penetration depth has been performed with good agreement with the experimental results. A complete theoretical calculation for the zerotemperature limit to the screening remains to be done.

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