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500 G and rather insensitive to the field gradient and the temperature. At such a field, the corrections to the diffusion terms because of the dipole energy are negligible and also the NMR linewidth term is negligible in comparison to the diffusion terms, since this is what we wanted. Naturally, the above limit for the field is dropping near  $T_c$ , but neglecting the dipole energy (except for the NMR shift) remains a good approximation.

In order to make the above evaluation. I have investigated the NMR linewidth and the NMR shift beyond the hydrodynamic limit used by Leggett.<sup>9</sup> It happens that Leggett's result,  $\omega_R^2 = \omega_L^2$  $+\omega_0^2(T)$ , is still true over the complete range of frequency including the collisionless limit, if terms of order of  $[\omega_0(T)/\omega_L]^4$  are neglected, and that, away from the hydrodynamic region, the NMR linewidth is of order of  $\omega_0^4(T)/\omega_L^3$ . These two facts are correlated by sum rules, as pointed out by Leggett.<sup>12</sup> These results are obtained from Eq. (3) with q = 0. If we neglect higherorder terms in the dipole energy, we can replace  $\overline{\theta}$ , in  $i\Phi^{(0)}\overline{\theta}$ , by its expression as a function of  $\delta \vec{\rho}$  for zero dipole energy, which is given by Eq. (4). Our result is thus independent of any approximation in the collision terms. Note also that  $\omega_0(T)$  is still the longitudinal resonance frequency, since it is always in the hydrodynamic region. Finally, in the collisionless regime, these results can be checked from Maki and Ebisawa's results<sup>13</sup> by making the appropriate approximations.

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- <sup>1</sup>K. Maki and H. Ebisawa, J. Low Temp. Phys. <u>15</u>, 212 (1974).
  - <sup>2</sup>R. Combescot, Phys. Rev. A <u>10</u>, 1700 (1974).
  - <sup>3</sup>R. Combescot, Phys. Rev. Lett. <u>33</u>, 946 (1974).
  - <sup>4</sup>W. F. Brinkman and H. Smith, to be published.
  - <sup>5</sup>K. Maki and T. Tsuneto, to be published.
- <sup>6</sup>A. J. Leggett and M. J. Rice, Phys. Rev. Lett. <u>20</u>, 586 (1968).
- <sup>7</sup>A. J. Leggett, J. Phys. C: Proc. Phys. Soc., London <u>3</u>, 448 (1970). <sup>8</sup>L. R. Corruccini, D. D. Osheroff, D. M. Lee, and

<sup>o</sup>L. R. Corruccini, D. D. Osheroff, D. M. Lee, and R. C. Richardson, Phys. Rev. Lett. <u>27</u>, 650 (1971).

<sup>9</sup>A. J. Leggett, Phys. Rev. Lett. <u>31</u>, 352 (1973). <sup>10</sup>R. Combescot and H. Ebisawa, Phys. Rev. Lett. <u>33</u>, 810 (1974).

- <sup>11</sup>L. R. Corruccini and D. D. Osheroff, to be published.
  <sup>12</sup>A. J. Leggett, Phys. Rev. Lett. <u>29</u>, 1227 (1972).
  <sup>13</sup>K. Maki and H. Ebisawa, Prog. Theor. Phys. <u>50</u>,
- 1452 (1973).

## Propagating Order-Parameter Collective Modes in Superconducting Films\*

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A peak at finite frequency has been observed in measurements of the order-parameter dynamical structure factor of short-mean-free-path superconducting aluminum films. The existence of a propagating, low-frequency order-parameter collective mode can be inferred from these results.

The order-parameter structure factor is the space and time Fourier transform of the orderparameter-order-parameter correlation function. In this Letter we report measurements of the structure factor in superconducting aluminum films which provide the first clear-cut demonstration of the existence of order-parameter collective modes in superconductors. Previously<sup>1,2</sup> we reported measurements of the imaginary part of the pair-field susceptibility of aluminum using a tunneling technique.<sup>3</sup> The structure factor  $S(\omega, q)$  can be related to the imaginary part of the susceptibility,  $\chi''(\omega, q)$ , by the fluctuation-dissipation theorem which in the low-frequency limit  $(\hbar\omega \ll k_B T)$  takes the form  $S(\omega, q) = 2k_B T\chi''(\omega, q)/\omega$ . The advantage of studying  $S(\omega, q)$  rather than  $\chi''(\omega, q)$  is that from the former it is easier both to demonstrate the nature of the collective mode and in principle to ascertain its dispersion relation.

The pair-field susceptibility of aluminum films can be obtained experimentally by using a tunneling junction in which aluminum is one electrode and lead is the other. Near  $T_c$  of aluminum an excess current which is a consequence of pair tunneling is observed in the current-voltage characteristic of the junction. This current is a direct measure of the imaginary part of the pairfield susceptibility,  $\chi''(\omega, q)$ , where  $\omega = 2eV/\hbar$  and  $q = (2e/\hbar c)(\lambda + d/2)H$ . Here V is the dc bias across the junction, and H is a magnetic field applied in the plane of the junction. The penetration depth in the lead film is  $\lambda$  and *d* is the thickness of the aluminum film. In the regime studied, the penetration depth and the coherence length of the aluminum electrode are both greater than its thickness, and the lead electrode is thicker than its penetration depth. Measurements of the excess current carried out below the transition temperature of aluminum require a magnetic field large enough to quench the dc Josephson effect. Experimental procedures and the analysis used to find the excess current have been described elsewhere.<sup>1,2</sup>

Above  $T_c$  order-parameter fluctuations can be described by a diffusive time-dependent Ginzburg-Landau equation.<sup>4</sup> The pair-field susceptibility obtained experimentally is consistent with such a picture of the order-parameter dynamics. Below  $T_c$  a diffusive equation cannot be used to describe the dynamics of the order parameter. Experimentally, three structures are observed in the excess-current-voltage characteristic.<sup>2</sup> There is a low-voltage peak, or sometimes a shoulder, which can be related to longitudinal fluctuations or fluctuations in the amplitude<sup>5,6</sup> of the order parameter. The main peak, which develops continuously from the peak observed above  $T_c$ , can be associated with fluctations in the phase. A third peak occurs at a voltage  $eV = \Delta(T)$ , where  $\Delta(T)$  is the order parameter in the aluminum. This peak, which may be a tunneling anomaly associated with the creation of real pairs by the ac Josephson current,<sup>7</sup> provides a convenient reference to the magnitude of the order parameter at the particular temperature and magnetic field.

The structure factor, which is proportional to the excess current divided by the voltage, is shown in Fig. 1. On the frequency scale used,



FIG. 1. The structure factor in arbitrary units plotted as a function of frequency and temperature. A magnetic field of 125 G is applied parallel to the plane of the junction reducing the transition temperature to 1.7795 K from 1.786 K in zero field. It should be noted that the curves are symmetric about  $\omega = 0$ , although only positive  $\omega$  is shown. The high-frequency peak is out of the range of the graphs.

the peak at the aluminum gap observed in the excess current is not on the curve. Above  $T_c$  the structure factor is a Lorentzian centered at  $\omega$ =0. Below and in the immediate vicinity of  $T_c$ ,  $S(\omega, q)$  has a peak at a nonzero frequency  $\omega_{p}$  in addition to the usual one at the origin. As the temperature is reduced below  $T_c$  this peak broadens and disappears into the tail of the central peak. Usually a peak in the dynamical structure factor at a nonzero frequency implies the existence of a propagating mode. In the present case the mode has been identified with fluctuations in the phase of the order parameter.<sup>5,6</sup> The range of temperature over which the mode is well defined corresponds to that over which the main peak in  $\chi''(\omega, q)$  was found to be distorted from the form exhibited above  $T_c$ .<sup>2</sup>

In Fig. 2 we show the wave-vector dependence of the finite-voltage peak in the structure factor at fixed temperature [Fig. 2(a)] and fixed reduced temperature  $\epsilon(H, T)$  [Fig. 2(b)]. The bending over of the curves in Fig. 2(a) is a consequence of the approach to the superconducting-normal phase boundary with increasing field. Because the applied field shifts  $T_c$  as well as determines the wave vector q, the physically significant curve is believed to be the plot at fixed reduced temperature  $\epsilon(H, T)$  rather than at fixed T. Thus Fig. 2(b) in effect can be used to determine the



FIG. 2. (a) Frequencies in rad/sec of peaks in  $S(\omega,q)$  at several fixed temperatures, plotted as a function of q in units of  $(2e/\hbar c) (d/2 + \lambda)$ .  $\lambda$  is taken to be 390 Å and d=1300 Å. The points A, B, C, and D correspond to temperatures of 1.77104, 1.7674, 1.76379, and 1.75317 K, respectively. (b) Frequencies of peaks in  $S(\omega,q)$  at fixed  $\epsilon(T,H) = [T_c(H) - T]/T_c(H)$ , with  $T_c(0) = 1.786$  K and  $\xi(0) = 5.46 \times 10^{-6}$  cm.  $\epsilon(T,H)$  was calculated from  $\epsilon(T,H) = \epsilon(T, 0) - \frac{1}{3}[e^2d^2\xi^2(0)/\hbar^2c^2]H^2$ .  $\xi(0)$  is the zero-temperature coherence length. The points labeled A and B correspond to  $\epsilon = 6.32 \times 10^{-3}$  and  $1.0 \times 10^{-3}$ , respectively.  $T_c(0)$  and  $\xi(0)$  are quantitatively consistent with the results of extensive measurements above  $T_c$  and with the experimentally determined  $T_c(H)$  curve which is not shown.

dispersion relation for the propagating mode suggested by these results. If the dispersion relation is taken to be linear, propagation velocities can be obtained from the slopes of the curves and they are  $1.74 \times 10^6$  cm/sec for  $\epsilon = 6.32 \times 10^{-3}$  and  $1.35 \times 10^6$  cm/sec for  $\epsilon = 1.0 \times 10^{-3}$ . These velocities are greater than the sound velocity in aluminum but are lower than the Fermi velocity of aluminum and the velocity of Swihart modes in the junction.<sup>8</sup> The propagation velocity appears to be independent of  $\epsilon$  for  $\epsilon > 6 \times 10^{-3}$  to the extent that it is possible to observe a peak in  $S(\omega, q)$ . In Fig. 3 we show the temperature dependence of the peak frequency showing this apparent saturation at low temperatures.

In Figs. 2 and 3 the upper limits on the ranges of q and  $\epsilon$  for which data are plotted are determined by limitations on the quantitative definition of the peak in  $S(\omega, q)$  which is a consequence of the graphical technique used to analyze data. For values of q and  $\epsilon$  as large as 300 Oe and  $6 \times 10^{-2}$ , respectively, an effect is still evident but can be characterized only in a marginal manner. Over the range plotted, the error in the determination of peak frequencies using a graphical technique



FIG. 3. Temperature dependence of the peak of  $S(\omega,q)$  in fields of 150 G (A) and 125 G (B). The solid and dashed lines are calculated from Refs. 5 and 6 for 150 and 125 G, respectively. The depairing parameter was taken to be 0.0250 with use of the theory of Ref. 5.

rather than a fitting scheme is estimated to be at most  $\pm 3 \times 10^9$  rad/sec if all the peaks are Lorentzians or shifted Lorentzians. The errors shown on the graphs are a combination of measurement uncertainties and errors resulting from the analysis. These errors are not large enough to account for the fact that all graphs of  $\omega_{p}$  versus q shown here, and determined for another sample which is not shown, extrapolate to a small negative frequency as q goes to zero. A detailed fitting technique which takes into account all of the peaks in the excess-current-voltage characteristic might result in a significant improvement and is under investigation. A systematic error in the analysis might also be responsible for the negative intercept: A possible contribution from a remnant of the fluctuation-broadened dc Josephson current would make an increasing contribution to the excess current at low voltages as the magnetic field is reduced. This would appear as a contribution to the central peak of  $S(\omega, q)$  and lead to distortion at low frequencies. Measurements of the height of the central peak as a function of temperature suggest that this effect is probably unimportant. The reason for this is that the peak narrows and decreases in height as T is lowered below  $T_c$  when the measurements are carried out in fields in excess of 10 G. The

The lower limits on the ranges of q and Tshown on the graphs are set by the requirement that the total-current-voltage characteristic not contain regions of negative dynamical resistance. The latter appear at low q and T and are accompanied by switching and susceptibility to noise which make reliable measurements impossible. All of the data shown here and in Refs. 1 and 2 were obtained under conditions in which the total dynamical resistance was positive.

At the present time the theoretical situation is unclear. Recently Brieskorn, Dinter, and Schmidt<sup>5</sup> and Maki and Sato.<sup>6</sup> in an effort to explain the results of Ref. 2, calculated  $\chi''$  from the Gor'kov-Eliashberg equations.<sup>9</sup> They obtained a phononlike mode in the order-parameter phase similar to fourth sound in superfluid helium. Fourth sound is a pressure and density wave in which only the superfluid is in motion.<sup>10</sup> The motion of the normal fluid is inhibited by its viscosity, as the geometry of the containing vessel is one in which the liquid fills a porous stationary matrix of solid material. For superconductors in the dirty limit, the motion of the normal electrons is also inhibited as a consequence of their strong scattering by impurities. The solid and dashed curves in Fig. 3 are calculated from the fourthsound velocity  $c^2 = (\Delta^2/\pi) (T_c/T) (8/\hbar) \xi^2(0) \psi'(\rho + \frac{1}{2})$ given in Refs. 5 and 6 with the assumption of a phononlike dispersion relation. The quantities  $\rho$  and  $\Delta$  are the depairing parameter and the order parameter, respectively.  $\psi'(\rho + \frac{1}{2})$  is a trigamma function and  $\xi(0)$  is the zero-temperature coherence length. The theoretical curves are drawn over a range of temperature only 4.5 mK wide which is the upper limit on the possible width of the gapless region, based on a depairing parameter whose upper limit is 0.0250. If the depairing parameter is smaller than 0.0250, the region of validity of the theory is narrower than that plotted and the discrepancy between experiment and theory is less serious. The depairing parameter was determined from a careful comparison of measurements of  $\chi''(\omega, q)$  above  $T_c$ with calculations in Ref. 5 which contain the explicit dependence of  $\chi''(\omega, q)$  on  $\rho$ .

The fact that the experimental frequencies in Fig. 3 are higher than the theoretical ones may be a consequence of the coupling of the collective mode to charge fluctuations which are not con-

tained in the Gor'kov-Eliashberg<sup>9</sup> equations. It should be emphasized that the experiment does not measure the dispersion curve at low q. However, the negative-frequency intercept of the curve suggested by the high-q data is not inconsistent with a model in which a phononlike mode is coupled to a plasmonlike mode.<sup>11</sup>

The details of preparation, characterization, and testing of junctions used in these experiments have been described in detail elsewhere.<sup>1</sup> The results presented here are representative of features observed in the five Pb-Al junctions which have been studied since these effects were discovered. The aluminum films in these junctions were all disordered and exhibited transition temperatures ranging from 1.65 to 1.95 K. However. no sample-to-sample variation in the qualitative features of the effects reported here was observed. Film transition temperatures appear to be in the range usually found for granular films 1200 Å thick and of average grain size of the order of 100 Å.<sup>12</sup> Electron microscope studies and grain counts on the films actually used in the present investigation have not been carried out. However, such granular structure was observed in electron micrographs made of an aluminum film with a transition temperature of 1.84 K which was prepared in the same vacuum system under the same conditions as the films of the present investigation. Granular films with short conduction-electron mean free paths are thought to behave as homogeneous dirty-limit superconductors as long as the grain diameters are smaller than the coherence length  $\xi(T)$ .<sup>13</sup> Such a picture is strongly supported in the present instance by the fact that the same value of  $\xi(0)$  is consistent with the variation of  $T_c$  with magnetic field and the variation of  $\chi''(\omega, q)$  with magnetic field above  $T_c$ .<sup>14</sup>

It should also be noted that in the present studies the lower electrode of the junction was covered with a layer of bismuth oxide so that only a small window in the middle of the crossed-film structure defined the tunneling junction.<sup>1</sup> Thus the measurements never involve tunneling in the vicinity of film edges which may be strained. Such a precaution could not preclude the possibility of strain across the thickness of the film which could be obviated only by using singlecrystal aluminum substrates or by preparing free-standing junctions.

In conclusion we have observed a propagating collective mode which appears in the order-parameter structure factor of a superconductor as

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a peak at a finite frequency. The mode may be identified with propagating phase fluctuations. A complete determination of the dispersion relation has not been made.

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<sup>1</sup>J. T. Anderson, R. V. Carlson, and A. M. Goldman,

J. Low Temp. Phys. 8, 29 (1972).

<sup>2</sup>R. V. Carlson and A. M. Goldman, Phys. Rev. Lett. <u>31</u>, 880 (1973).

<sup>3</sup>D. J. Scalapino, Phys. Rev. Lett. <u>24</u>, 1052 (1970). <sup>4</sup>M. Cyrot, Rep. Progr. Phys. <u>36</u>, 103 (1973).

<sup>5</sup>G. Brieskorn, M. Dinter, and H. Schmidt, Solid State Commun. <u>15</u>, 757 (1974).

<sup>6</sup>K. Maki and H. Sato, J. Low Temp. Phys. <u>16</u>, 557 (1974).

<sup>7</sup>E. Simanek and J. C. Hayward, to be published.

<sup>8</sup>J. C. Swihart, J. Appl. Phys. <u>32</u>, 461 (1961).

 $^9 \mathrm{L}$  . P. Gor'kov and G. M. Eliashberg, J. Low Temp. Phys. <u>2</u>, 161 (1970).

<sup>10</sup>K. A. Shapiro and I. Rudnick, Phys. Rev. <u>137</u>, A1383 (1965).

<sup>11</sup>If modes of frequencies  $\omega_1$  and  $\omega_2$ , where  $\omega_1 \gg \omega_2$ , are coupled together, then the dispersion relation can be of the form  $\omega = \omega_2 - |V|^2 / |\omega_1 - \omega_2|$ , where V is the coupling parameter.

<sup>12</sup>G. Deutscher, H. Fenichel, M. Gershenson, E. Grunbaum, and Z. Ovadiahu, J. Low Temp. Phys. <u>10</u>, 231 (1973).

<sup>13</sup>B. Abeles, R. W. Cohen, and W. R. Stowell, Phys. Rev. Lett. <u>18</u>, 902 (1967).

<sup>14</sup>S. R. Shenoy and P. A. Lee, Phys. Rev. B <u>10</u>, 2744 (1974).

## Substrate Bias Effects on Electron Mobility in Silicon Inversion Layers at Low Temperatures

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The mobility of electrons in silicon inversion layers and conductance activation energy have been measured as a function of substrate bias. The mobility increased and the activation energy decreased as the electrons were forced toward the surface. This seems inconsistent with activated conduction arising from potential fluctuations due to oxide charge.

The conductance of electrons in inversion lavers in metal-insulator-silicon field-effect transistors (MOSFET's) has been the subject of many studies.<sup>1-4</sup> At low temperatures and all but the highest densities, the electrons lie in the lowest quantum state induced by the field perpendicular to the silicon surface. The results below are surprising because they seem in contradiction to other measurements in the temperature and carrier-density range where the conductance is thermally activated.<sup>1,3,4</sup> Several explanations have been offered to explain the decrease of activation energy of conductance with increasing carrier density. All depend on assuming that oxide charges near the interface play a major role. This is true for scattering by potential fluctuations,  ${}^{5_{16}}$  trapping where the screening increases with charge density and lowers the trap energy,  ${}^{7}$ and a percolation model or Mott-Anderson model<sup>8,9</sup> where fluctuations arise from the oxide charges.<sup>10</sup>

If these effects are caused by the fluctuations due to oxide charges, then forcing the electrons closer to the interface should increase the effects. It is well known<sup>11,1</sup> that application of substrate bias,  $V_s$ , between the source contact and the substrate reduces the free-electron charge  $qN_{inv}$  by increasing the depletion charge  $qN_{depl}$ and increases the gate voltage for conductance threshold  $V_t$ . To maintain a given  $N_{inv}$ , the gate voltage must be increased as the substrate is made more negative. Then the free electrons