

## The Growth Rate of $^3\text{He}$ Crystals

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*We have measured the relaxation of the shape of  $^3\text{He}$  crystals under the effect of gravity and surface tension, close to the minimum in their melting curve at  $T_{\min} = 0.32$  K. A growth rate is deduced, which is found to be at a maximum at  $T_{\min}$  when the latent heat is zero. We interpret this maximum value ( $k = 0.18 \pm 0.04$  sec/m) as the intrinsic mobility of the liquid-solid interface and compare it with existing theories. We also consider on which side the latent heat is released during growth, and how it may cross the liquid-solid interface.*

We report the first direct measurement of  $k$ , the intrinsic growth rate of  $^3\text{He}$  crystals or intrinsic mobility of the interface between liquid and solid  $^3\text{He}$ . When the temperature is homogeneous in the whole system, and if the surface is rough<sup>1,2</sup>,  $k$  is the ratio of the growth velocity  $v$  to the difference in chemical potential  $\Delta\mu = \mu_L - \mu_S$  between the liquid and the solid ( $\mu$  is taken per unit mass). In ordinary systems, it is generally difficult to have access to this quantity. Indeed, when a pure crystal is grown from its melt, the growth velocity is mainly limited by the slow diffusion of the latent heat in the bulk phases on each side of the moving interface. However, the growth resistance associated with this diffusion may vanish if the heat conductivity is very large and the latent heat very small. This is the case in  $^4\text{He}$  below 1 K, where the growth rate of rough surfaces was shown to be very large by several experiments<sup>4</sup>: the mobility of these  $^4\text{He}$  surfaces is only limited<sup>5</sup> by collisions with thermal excitations (phonons and rotons). In the case of  $^3\text{He}$ , the latent heat vanishes<sup>3</sup> at  $T_{\min} = 0.32$  K; Andreev and Parshin<sup>5</sup> suggested ten years ago that the growth rate  $k$  should be about  $m/p_F$ , the  $^3\text{He}$  mass divided by the Fermi momentum of quasiparticles in the liquid.<sup>6,†</sup> More recently, Puech, Bonfait, and Castaing<sup>7</sup> obtained a lower bound for  $k$  which was much larger than  $m/p_F$ . They explained this

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†Similar problems are encountered for the transmission of spin waves (Heff *et al.*) and for the mobility of the superfluid  $^3\text{He}$  A-B interface (Buchanan *et al.*); see Ref. 6.

discrepancy by supposing that the liquid quasiparticles interact with the crystal lattice more than the interface; their theoretical estimate is

$$k = (3t/4)(m/p_F)(\rho_S\rho_L/(\rho_S - \rho_L)^2) \quad (1)$$

where  $\rho_L$ ,  $\rho_S$  are the respective densities of liquid and solid  $^3\text{He}$  and  $t$  is the sticking probability of quasiparticles colliding with the crystal.

By looking at the shape of a pure single crystal of  $^3\text{He}$ , relaxing in some thirty seconds towards its equilibrium shape, we have measured the effective growth rate in a small temperature domain ( $\pm 0.02$  K) around  $T_{\min} = 0.32$  K. We observed that the growth is a linear function of the applied force. As expected, we found that the growth velocity (or the inverse relaxation time) goes through a maximum at  $T = T_{\min}$  when the latent heat is zero. At this temperature, we interpret the ratio  $v/\Delta\mu = 0.18 \pm 0.04$  sec/m as the intrinsic growth rate  $k$ . We compare it with existing theories and with the experiment by Puech *et al.*<sup>7</sup>. We also present a tentative interpretation of the measured increase in relaxation time on both sides of  $T_{\min}$ , when a nonzero latent heat  $L$  appears.

The experimental cell, where  $^3\text{He}$  crystals are grown, is immersed in a superfluid  $^4\text{He}$  bath which acts as a thermostat (Fig. 1). It is made of two parts. The lower one, where crystals are observed through sapphire windows, is a small cube ( $3.5 \times 3.5 \times 5$  mm<sup>3</sup>) where sintered copper was added for better thermalization. The upper part is a BeCu double membrane, which allows us to change the cell volume by changing the outside  $^4\text{He}$  pressure. This cell was originally built to measure the roughening temperature and surface tension of  $^3\text{He}$  crystals. It is described in more detail in another article.<sup>2</sup> For our present purpose, it is only important to note that we use purified  $^3\text{He}$  which contains only 5 ppm  $^4\text{He}$  and that this residual  $^4\text{He}$  is presumably adsorbed as a small fraction of a monolayer on the sintered copper. Thus, differences in chemical potential due to concentration gradients may be neglected. (In previous experiments, where impure  $^3\text{He}$  with 200 ppm  $^4\text{He}$  was used, our crystals behaved very differently<sup>1</sup>). At the equilibrium, the temperature is homogeneous within less than  $1 \mu\text{K}$  in our cell.<sup>2</sup> Finally, crystal shapes (or values of chemical potentials) depend on surface tension, on the local pressure which itself depends on gravity, and on slight temperature differences which may occur during growth. Gravity  $g$  induces a hydrostatic equilibrium in the liquid; the surface tension  $\gamma$  relates the local difference in pressure  $\Delta P = P_L - P_S$  to the local curvature of the liquid-solid interface through the usual Laplace equation  $\Delta P = \gamma(1/R_1 + 1/R_2)$ . We have assumed<sup>2</sup> that the surface stiffness  $\gamma$  is isotropic, and therefore equal to the surface tension or free energy per unit area. Careful measurements<sup>2</sup> of equilibrium crystal shapes gave us the value  $\gamma = 0.060 \pm 0.011$  erg/cm<sup>2</sup>.

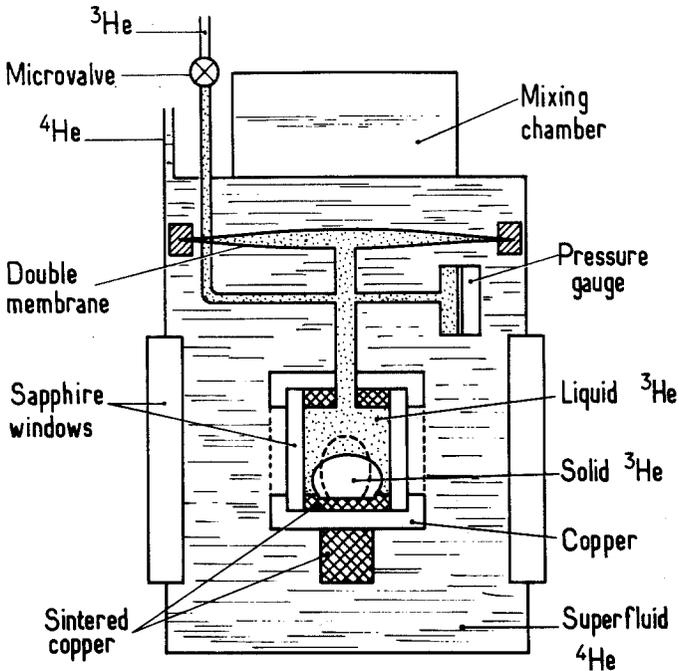


Fig. 1. The experimental cell, in which  $^3\text{He}$  crystals were grown and observed, is immersed in a  $^4\text{He}$  bath which acts as a thermostat. The inner distance between the cell windows is 5 mm; the section of the cell, parallel to them, is a square (3.5 by 3.5 mm). The sintered copper pieces allow a good thermalization, but also adsorb all the residual  $^4\text{He}$  impurities in the cell. Thanks to the Be-Cu double membrane, when the outside  $^4\text{He}$  pressure is changed, the cell volume may vary by about 5%. By measuring the  $^3\text{He}$  pressure, we checked the calibration of our carbon thermometer, which is located in the  $^4\text{He}$  bath. The heat radiation through the windows was minimized by using infrared filters at 4 K. We could force a crystal to grow first in the upper part of the cell and then fall down, through the 1 mm tube, onto the floor in the lower part. The conjunction of gravity and surface tension finally forced a relaxation towards an equilibrium shape. From the study of this phenomenon, we extracted the growth rate of our  $^3\text{He}$  crystals.

The principle of our experiment is to make a slightly deformed crystal and look at its relaxation towards an equilibrium shape. This can be done in the following way. We start with a small crystal at rest in the lower part of the cell at  $0.1 \leq T \leq 0.2$  K. We then fix the temperature regulation at the desired temperature, close to 0.32 K. Since the upper part is thinner, its typical warming time is smaller (a few seconds typically) than that of the lower part (about one minute). As a first result, the crystal melts in the lower part and a new crystal grows in the upper one (remember that, below  $T_{\min}$ ,

the latent heat is negative and  $^3\text{He}$  crystallizes on hot spots). However, one or two minutes later, the temperature equalizes in the two parts and gravity again becomes relevant: the crystal tends to fall down through the tube ( $\varnothing = 1$  mm). It was surprising to us that this actually occurs, provided  $T$  is not more than about 30 mK away from  $T_{\min}$ . Since the crystal "flow" results from local melting and recrystallization, a liquid channel must remain open in the tube for the necessary mass transfer. Whatever exactly happens during this stage, a nice symmetrical  $^3\text{He}$  crystal drips from the tube orifice, 5 to 10 minutes after having fixed the temperature to its new regulation point, and gently lands on the bottom of the lower part of the cell. It is usually elongated in the vertical direction and relaxes to a flatter shape under the effect of gravity and surface tension. This is the typical event which we have analyzed. However, let us make a few further remarks.

When  $T$  is really close to  $T_{\min}$ , the crystal drips as one single drop. When  $T$  is 10 to 20 mK away from  $T_{\min}$ , the crystal drips in 4 or 5 steps; each new drop perfectly wets the previous fallen one, since it generally has the same crystalline orientation. Each landing of a new drop is analyzed with a video camera, recorder, and image analyzer. Note here that the thermal conductivity in the solid<sup>7</sup> ( $K_S \approx 20$  W/Km) is much larger than in the liquid<sup>11</sup> ( $K_L \approx 4.5 \times 10^{-3}$  W/Km). This simple fact makes different crystals evolve independently when the latent heat  $L$  is not strictly zero. Indeed, any mass transfer from, say, a crystal already fallen to another one still in the upper part would need a slow heat transfer. In contrast, during the shape relaxation of a given crystal, mass is easily transferred from the melting top to the growing sides since the corresponding latent heat is easily conducted through the solid. The fascinating result is that, during most of these relaxations, solid  $^3\text{He}$  appeared to be a dripping viscous liquid rather than a high quality, single crystalline solid. In a few cases, however, the last little drop fell down with a crystal orientation different from the previous crystal drops. Instead of wetting and relaxing, it then rolled before shrinking. We did not analyze the latter events, but used all the others. We saw no systematic difference between the mobility found for the first drop and those found for the following ones.

Let us now come to the quantitative measurements. From usual thermodynamics,  $d\mu = dP/\rho - SdT$ . A simple way to analyze the shape relaxation would be to suppose that the liquid pressure  $P_L$  is constant in time and that the curvature and temperature effects are small. The height  $z$  of the crystal top would then simply obey the equation

$$v = dz/dt = -k_{\text{eff}}(\rho_S - \rho_L)g(z - z_{\infty})/\rho_S \quad (2)$$

where  $z_{\infty}$  is the final height at the end of the relaxation;  $k_{\text{eff}}$  is an effective mobility. We first plotted  $\log(z - z_{\infty})$  versus time, obtained rather well

defined straight lines and relaxation times, and got rough estimates of  $k_{\text{eff}}$  which appeared to be about twice as large as those we obtained from the following, more rigorous, analysis. We redefine the effective growth rate by

$$1/k_{\text{eff}} = -[(\rho_S - \rho_L)g(z - z_0) + \gamma C]/\rho_S v \quad (3)$$

where  $C$  is the interface curvature ( $C = 1/R_1 + 1/R_2$ ) and  $z_0$  is the equilibrium height of a flat interface. This inverse effective growth rate is the total growth resistance, which is the sum of the intrinsic growth resistance  $1/k$  and a thermal resistance that is mainly associated with the evacuation of the latent heat. It may also be seen as the ratio of the applied departure from equilibrium, or mechanical part of the difference in chemical potential  $\Delta\mu_m$ , to the growth velocity. Since  $z_0$  may be time dependent, because  $P_L$  may vary during the relaxation, we proceeded by taking the difference between two different points  $z_1$  and  $z_2$  on the crystal. In practice, we often took  $z_1$  as the top and  $z_2$  as an intermediate height with negligible velocity. We thus used the relation

$$v_1 - v_2 = -k_{\text{eff}}[(\rho_S - \rho_L)g(z_1 - z_2) + \gamma(C_1 - C_2)]/\rho_S \quad (4)$$

The velocities  $v_1$  and  $v_2$  were obtained from finite differences. Our main difficulty was that only one of the two curvature radii could be seen on the video recordings. We assumed a revolution symmetry for small crystals which did not touch the side walls (the first drops). For bigger crystals, we kept the revolution symmetry only for the top ( $R_1 = R_2$ ) and assumed that the curvature in the plane perpendicular to the visible profile, near the line of contact with the copper walls, was nearly constant during the relaxation. Since the cell depth is 5 mm, which is large compared to the capillary length  $(\gamma/g(\rho_S - \rho_L))^{1/2} \approx 1$  mm, this particular curvature has no drastic effect on our estimate of  $k_{\text{eff}}$ . It is simply evaluated from the equilibrium condition at the end of the relaxation. The uncertainty in our measurement mainly arises from this unknown curvature and the uncertainty in  $\gamma$  itself.

Figure 2 shows a typical graph obtained at  $T = 0.33$  K. The relation between  $v$  and the departure from equilibrium  $\Delta\mu_m$  is linear, and the slope gives us  $k_{\text{eff}}$ . Note that  $10^{-3}$  J/kg =  $10$  cm<sup>2</sup> sec<sup>2</sup> corresponds to a height difference of about 2 mm, which is a very small departure from the equilibrium pressure ( $\delta P/P \approx 10^{-6}$ ). Figure 3 shows the variation of  $1/k_{\text{eff}}$ , the effective growth resistance, as a function of  $T - T_{\text{min}}$ . The drawn parabola is

$$1/k_{\text{eff}} = 5.5 + 3.9 \times 10^4 (T - T_{\text{min}})^2 \quad (5)$$

in MKS units. As expected, the growth rate goes through a maximum value at  $T = 0.32$  K.

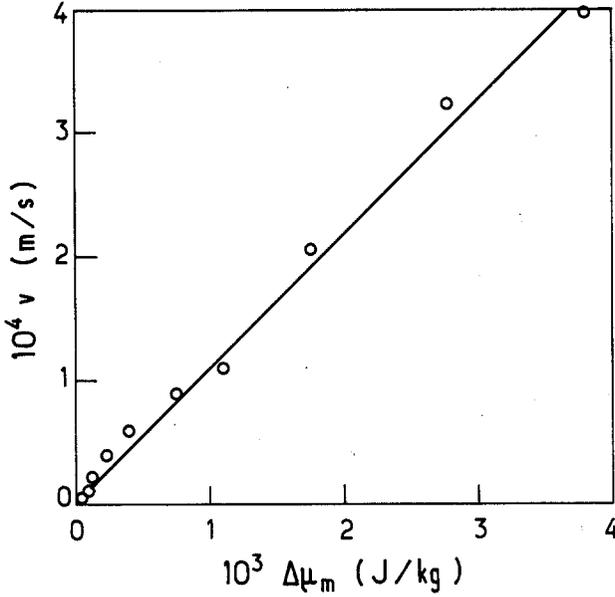


Fig. 2. The growth velocity is proportional to the applied departure from equilibrium ( $\Delta\mu_m$  is the mechanical part of the difference in chemical potential  $\Delta\mu = \mu_L - \mu_S$  between the liquid and the solid). The ratio  $k_{\text{eff}} = v/\Delta\mu_m$  is the effective growth rate of the crystal, which is equal to the intrinsic mobility of the liquid-solid interface when the latent heat is zero, but also accounts for the thermal resistance which controls the transport of this latent heat when it is nonzero.

Finally, let us try to interpret our experimental results. The mass current  $J = \rho_S v$  and total heat current  $J_E = J_Q + TSJ$  through the interface are related<sup>9</sup> to the differences  $\Delta T$  and  $\Delta\mu$  by

$$\Delta T = R(J_E - \lambda J) \quad (6)$$

$$J = k \left( \Delta\mu + \lambda \frac{\Delta T}{T} \right) \quad (7)$$

Here,  $R$  is the thermal impedance, or Kapitza resistance, of the interface;  $k$  is the isothermal growth rate or intrinsic mobility, and  $\lambda$  is the coefficient which determines how much heat is liberated or absorbed on each side of the interface. The effective growth resistance in our experiment is given by

$$\frac{1}{k_{\text{eff}}} = \frac{1}{k} + \rho_S \left[ \frac{RZ_L(TS_L - \lambda)^2 + RZ_S(TS_S - \lambda)^2 + Z_L Z_S L^2}{T(R + Z_L + Z_S)} \right] \quad (8)$$

where  $Z_L$  and  $Z_S$  are the thermal impedances of the liquid and solid phases, respectively, and  $L$  is the latent heat.

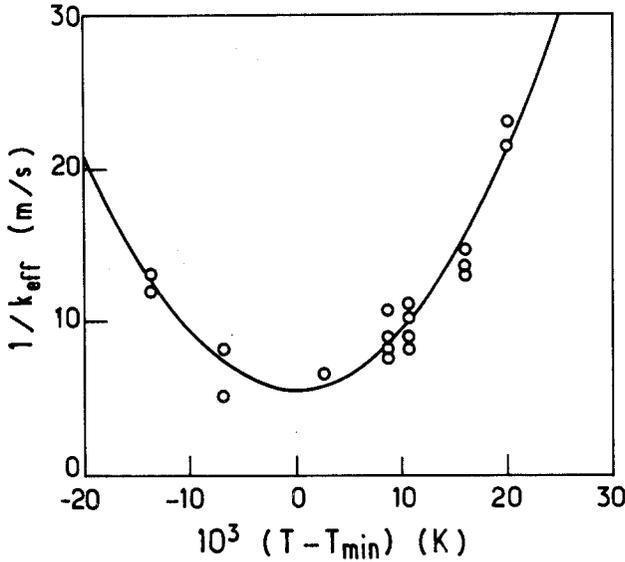


Fig. 3. The effective growth resistance  $1/k_{\text{eff}}$  as a function of temperature ( $T_{\text{min}} = 0.32$  K). This resistance is minimum when the latent heat is zero. The parabola drawn through the points is  $1/k_{\text{eff}} = 5.5 + 3.9 \times 10^{-4} (T - T_{\text{min}})^2$  in mks units.

There are various possible interpretations for our results. The latent heat may be liberated in the liquid ( $\lambda = TS_S$ ) or in the solid ( $\lambda = TS_L$ ) or shared between both sides (the example of  $^4\text{He}$  at zero temperature corresponds to  $R = \infty$  and  $\lambda = 0$ , which means that the interface is impermeable to heat). According to Bowley, Edwards, and Nozières,<sup>10</sup> all the latent heat is liberated in the liquid. It then has to go through the interface and be conducted through the solid. Indeed, values of  $Z_S$  and  $Z_L$  can be found using  $Z = d/K$  with  $d$  being a typical length scale and  $K$  the thermal conductivity. Using values of  $K_S$  from Ref. 7 and  $K_L$  from Ref. 11, one finds  $Z_S = 0.5 \text{ cm}^2 \text{ K/W}$  and  $Z_L = 2200 \text{ cm}^2 \text{ K/W}$ . Therefore  $Z_S \ll Z_L$ . With this scenario, we obtain

$$1/k_{\text{eff}} = 1/k + (R + Z_S)L^2/T \quad (9)$$

Since  $L = 900(T - T_{\text{min}})\text{J/kg}$ , our results are well interpreted by this formula if

$$R = 1 \text{ cm}^2 \text{ K/W}$$

The minimum shown on Fig. 3 gives the value of the intrinsic growth rate

$$k = 0.18 \pm 0.04 \text{ sec/m}$$

which is compatible with Puech's formula<sup>7</sup> if the sticking probability  $t$  is about 0.1 (we have used<sup>8</sup>  $p_F = 9.25 \times 10^{-25}$  kgm/sec). Our value for the Kapitza resistance is close to the result of a calculation from acoustic mismatch theory.<sup>12</sup> Now, it is also much smaller than the value  $R = 24.4 \text{ cm}^2 \text{ K/W}$  which can be extrapolated from Puech's data. However, their experimental results seem questionable: the growth rate is not large enough to significantly change the prediction from the acoustic mismatch theory (contrary to the <sup>4</sup>He case). Let us also note that the hypothesis  $\lambda = TS_L$  would lead to a temperature variation of  $1/k_{\text{eff}}$  about three times smaller than what we observed. In any case, at 0.32 K where  $S_L = S_S = S$ , we have  $\rho_S R (TS - \lambda)^2 / T \leq 1/k_{\text{eff}}$ , so that  $\lambda$  cannot be very different from  $TS$

$$|1 - \lambda / TS| \leq 2 \times 10^{-2}$$

Finally, Puech *et al.*<sup>7</sup> also gave the experimental lower bound  $k \geq 0.7 \text{ sec/m}$ ; however, without seeing, they could not be sure that their solid was one single crystal with a single liquid-solid interface of known area (their "level" measurement was only an average density measurement); we thus consider their lower bound only as an order of magnitude.

In summary, we have measured the growth rate of <sup>3</sup>He crystals close to  $T = 0.32 \text{ K}$ . Our results are well interpreted by supposing that, during growth or melting, the latent heat is released on the liquid side of the moving interface. We obtain an isothermal growth rate  $k = 0.18 \pm 0.04 \text{ sec m}$  for helium 3 crystals, in good agreement with a recent theory by Puech *et al.*,<sup>7</sup> who suppose that the mobility of the <sup>3</sup>He liquid-solid interface is controlled by the collisions of quasiparticles with the solid. Within this theory, our result indicates that the sticking probability of the quasiparticles to the solid is about 0.1. We also obtain a value  $R = 1 \text{ cm}^2 \text{ K/W}$  for the Kapitza resistance of the liquid-solid interface, which seems close to a prediction from the acoustic mismatch theory.<sup>12</sup>

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### REFERENCES

1. E. Rolley, S. Balibar, and F. Gallet, *Europhys. Lett.* **2**, 247 (1986).
2. E. Rolley, S. Balibar, F. Gallet, F. Graner, and C. Guthmann, *Europhys. Lett.* (to appear).
3. E. R. Grilly, *J. Low Temp. Phys.* **4**, 615 (1971).
4. K. O. Keshishev, A. Ya. Parshin, and A. B. Babkin, *Zh. Eksp. Teor. Fiz.* **80**, 716 (1981). [*Soviet Phys.-JETP* **53**, 362 (1981)]; B. Castaing, *J. Phys. Lett.* **45**, L233 (1984); J.

- Bodensohn, P. Leiderer, and D. Savignac, in *Proceedings of 4th International Conference Phonon on Scattering of Condensed Matter* (Springer, New York, 1984), p. 266; P. Leider, *Physica* **126B**, C, 92 (1984).
5. A. F. Andreev and A. Ya. Parshin, *Zh. Eksp. Teor. Fiz.* **75**, 1511 (1978) [*Soviet Phys.-JETP* **48**, 763 (1978)]; R. M. Bowley and D. O. Edwards, *J. Phys. (Paris)* **44**, 723 (1983); A. F. Andreev and V. G. Knizhnik, *Zh. Eksp. Teor. Fiz.* **83**, 46 (1982) [*Soviet Phys.-JETP* **56**, 226 (1982)].
  6. A. Heff, D. Candela, D. O. Edwards, and S. Kumar, *Europhys. Lett.* **4**, 1043 (1987); D. S. Buchanan, G. W. Swift, and J. C. Wheatley, *Phys. Rev. Lett.* **57**, 341 (1986); S. Yip and A. J. Leggett, *Phys. Rev. Lett.* **57**, 345 (1986).
  7. L. Puech, G. Bonfait, and B. Castaing, *J. Low Temp. Phys.* **62**, 315 (1986).
  8. J. Wilks, *The Properties of Liquid and Solid Helium* (Clarendon, Oxford, 1967), p. 677.
  9. B. Castaing and P. Nozières, *J. Phys. (Paris)* **41**, 701 (1980); D. O. Edwards, S. Balibar, and P. E. Wolf, in *75th Jubilee Conference on  $^4\text{He}$* , J. G. M. Armitage, ed. (World Scientific, Singapore, 1983), p. 70; P. Nozières, unpublished lectures at the College de France.
  10. R. M. Bowley, D. O. Edwards, and P. Nozières, private communications.
  11. A. C. Anderson, J. I. Connolly, O. E. Vilches, and J. C. Wheatley, *Phys. Rev.* **147**, 90 (1966).
  12. R. M. Bowley, F. Graner, and P. Nozières, to be published.