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of the arbitrary extension of $S_{ce}(k)$ to high k and uncertainties in the exact slope of S(k) at high k. Extreme variations of that slope due to possible systematic experimental uncertainties were imposed on the S(k) data. While changes in the exact size, $\pm 25\%$, and position, $\pm 0.3rk_{\rm F}$, of g(r)were observed; all had peaks with $r < \overline{r}$. The results, therefore, do strongly indicate the phenomena associated with the anomalous plasmon dispersion and the peaking of $S_{ce}(k)$ at $2k_{\rm F}$ correspond to features of the electron-electron interaction at distances shorter than \overline{r} . It is possible that interaction with the localized, bound core electrons is responsible for the short-range correlation. Arguing against this interpretation is the observations in $S_{e}(k,\omega)$ studies⁵ and in this work that the results look very similar when scaled to electron-gas parameters. However, a possible interpretation is that in real solids, as opposed to the idealized electron gas with its jellium background, the effect of electron correlation is not to condense to a Wigner lattice at r $=\overline{r}$, but rather to make a Mott-type transition to an atomiclike state. However, on the basis of these experiments, we can only make a general observation that the characteristic distance associated with the anomalies is shorter than \overline{r} .

It is hoped that this measurement of $S_{ce}(k)$ will stimulate further theoretical interest in the nature of interactions in the electron gas and that the observation in this work that short-range interactions are important will be useful. In a broader sense it is hoped that the general technique described here will be extended to study

other Fermi liquids as well as other electron systems. In principle $S_e(k)$ studies can provide a testing ground for our microscopic understanding of electronic interactions.

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¹D. Pines, Elementary Excitations in Solids (Benjamin, New York, 1964), p. 131.

²R. Weiss, X-ray Determinations of Momentum Distribution (Wiley, New York, 1966), p. 79.

³P. Eisenberger, P. M. Platzman, and K. C. Pandey, Phys. Rev. Lett. 31, 311 (1973).

⁴P. Eisenberger, P. M. Platzman, and P. Schmidt, Phys. Rev. Lett. 34, 18 (1975).

⁵P. M. Platzman and P. Eisenberger, Phys. Rev. Lett. 33, 152 (1974).

⁶P. Eisenberger and P. M. Platzman, Phys. Rev. B 13, 934 (1976). ⁷P. Eisenberger, unpublished results.

⁸P. M. Platzman and P. Eisenberger, Solid State Commun. 14, 1 (1974).

⁹P. Eisenberger and P. M. Platzman, Phys. Rev. B 2, 415 (1970).

¹⁰G. E. Ice, M. H. Chen, and B. Crasemann, Phys. Rev. A 17, 3325 (1977).

¹¹D. M. North, J. E. Enderby, and P. A. Egelstaff, J. Phys. C 1, 784 (1968).

¹²W. C. Marra, P. Eisenberger, and R. J. Birgeneau, Phys. Rev. B 15, 3325 (1977).

Effect of Charging Energy on Kosterlitz-Thouless Transition Temperature in Granular Films

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A relationship between the Kosterlitz-Thouless transition temperature $T_{\rm KT}$ and the sheet resistance R_{\Box}^{N} of a granular superconducting film is derived taking into account the electrostatic charging energy. The zero-point phase fluctuations produce a drop of the areal superfluid density at a critical value of the sheet resistance in agreement with the recent observation on granular lead films.

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Recently Beasley, Mooij, and Orlando¹ and Halperin and Nelson² considered the possibility of the vortex unbinding transition in superconducting films. A simple relation has been established between the Kosterlitz-Thouless transition temperature T_{KT} and the sheet resistance R^N of the film.¹ On comparing this relation with the broadening of the resistive transition, taken from a wide varie-

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ty of observations, Beasley, Mooij, and Orlando¹ were only able to obtain a qualitative agreement. More recently Hebard and Vandenberg^{3, 4} studied the resistive transitions in granular lead films. By changing the degree of oxidation they measured $T_{\rm KT}$ vs R^N for a sequence of resistive transitions on the same 250-Å-thick film. The results, partly reproduced in Fig. 1, show a distinct drop near $R^N \simeq 13 \ {\rm k}\Omega$, which is in strong disagreement with the smooth dependence, predicted by the relation of Ref. 1.

In this Letter, I derive a new relationship between $T_{\rm KT}$ and R^N which is in agreement with the experimental results of Refs. 3 and 4. The sudden drop of T_{KT} is attributed to the zero-point fluctuations of the phase of the order parameter which occurs as a result of the electrostatic charging energy.⁵ Hebard and Vandenberg^{3,4} presented a conclusive evidence for the formation of clusters, whose average size l_c decreases as the sheet resistance increases. By assuming a regular array of clusters separated by a 10-Å layer of oxide, they estimate the value of l_c below which the charging energy disrupts the long-range superconducting order. For $R^N \simeq 13 \text{ k}\Omega$, their results^{3,4} imply $l_c \lesssim 250$ Å, a value too small for the expected cluster size, which seems to suggest that charging energy is not involved in the sudden drop of T_{KT} . I believe, however, that although the above model of the tightly coupled clusters finds it quantitative justification^{3,4} for the low-resistance $(R_{\Box}^{\ N} \simeq 3.1 \text{ k}\Omega)$ films it underestimates



FIG. 1. The temperature ratio $T_{\rm KT}/T_{c0}$ vs R_{\Box}^{N} for resistive transitions in lead film. Curve *A*, the SCHA result given by Eq. (13). Curve *B*, the MFA result of Eq. (20). Curve *C*, the experimental points of Ref. 3. Curve *D*, previous theory (Refs. 1, 2, and 3) with no account of the charging energy.

the charging energy in the region of $R_{\Box}^{\ N} \gtrsim 10 \text{ k}\Omega$. As $R_{\Box}^{\ N}$ increases, the disorder tends to disrupt more superconducting links until a percolation threshold is approached, in which case the picture of a homogeneous network of nodes connected by narrow channels (links) seems more appropriate.⁵ The latter model presumably allows for larger charging effects taking place especially along the narrow channels. Correspondingly, I adopt a model of a regular array of clusters of size l_c the capacitance of which is effectively reduced compared to that of Refs. 3 and 4. The Hamiltonian of this array is⁶

$$\mathcal{BC} = \frac{1}{2} U \sum_{i} n_{i}^{2} + \sum_{ij} E_{ij} [1 - \cos(\varphi_{i} - \varphi_{j})], \qquad (1)$$

where n_i is the electron-number operator and φ_i is the phase for the ith cluster. The Josephson coupling strength between nearest-neighbor clusters is denoted by $E_{ij} = E_1 \delta_{i,j \pm 1}$. I note that Hamiltonian (1) includes only the diagonal or Hubbardlike part of the charging energy, the Coulomb interaction between different grains being neglected. In previous works⁶ the arrays described by Hamiltonian (1) were shown to exhibit at T=0, a mean-field phase transition characterized by the vanishing of the average order parameter $\langle \cos \varphi \rangle$ when the cluster charging energy U exceeds the value $U_c = zE_1$ where z is the number of nearest neighbors in the array. That this also implies an instability of phase locking as R_{\Box}^{N} increases towards certain critical value can be seen as follows. First of all the dependence of E_1 on R_{\square}^{N} is given by the relation³

$$E_1 = \left[\pi R_0 \Delta(0) / 8 R_{\Box}^{N} \right] f(T), \tag{2}$$

where $R_0 = \hbar/e^2$ and f(T) is defined as¹

$$f(T) = \left[\Delta(T)/\Delta(0)\right] \tanh[\Delta(T)/2k_{\rm B}T].$$
 (3)

Moreover, the electrostatic charging energy U is expected to increase with $R_{\Box}^{\ \ N}$, since it is proportional to $d_{\rm ox}/l_c$, where $d_{\rm ox}$ is the thickness of the oxide layer separating the clusters.^{3,4} This implies, in conjunction with Eq. (2), that the parameter

$$\alpha(0) = \frac{zE_1(T=0)}{U} = \frac{\pi R_0 \Delta(0)}{8R_{\Box}^N U(R_{\Box}^N)}$$
(4)

decreases with the sheet resistance R_{\Box}^{N} . For R_{\Box}^{N} larger than a critical value $(R_{\Box}^{N})_{crit}$ [for which Eq. (4) yields $\alpha(0) \simeq 1$] Ref. 6 implies a complete destruction of the long-range phase coherence of the sample.

To calculate the modification of the superfluid

sheet density n_s^{2D} , by the effects of the charging energy, I start from the expression²

$$\vec{j}_{s} = (\hbar e/2m)n_{s}^{2D}[\nabla \varphi - (2e/\hbar c)\vec{A}].$$
(5)

Below I derive this relation from Hamiltonian (1), by employing the self-consistent harmonic approximation⁶ (SCHA) which replaces the Josephsoncoupling term of (1) by an effective Hamiltonian

$$\mathscr{K}_{\sigma} = \frac{1}{2} \sum_{i,j} K_{ij} (\varphi_i - \varphi_j)^2, \tag{6}$$

where $K_{ij} = K_1 \delta_{i,j+1}$ is the Josephson-coupling constant renormalized by the zero-point phase fluctuations. K_1 is calculated self-consistently from the pair of equations⁶

$$K_1 = E_1 e^{-D/2} \tag{7}$$

and

$$D = 2N^{-1}\sum_{i} (1 - \cos \vec{k} \cdot \vec{l}_{c}) \langle q_{k} q_{-k} \rangle, \qquad (8)$$

where $\langle q_k q_{-k} \rangle = (1/k l_c)(U/K_1)^{1/2}$. Performing the two-dimensional k sum in Eq. (9) with the use of the Debye approximation, I find $D \simeq 0.75(U/E_1)^{1/2}$. With introduction of a dimensionless parameter $\gamma = 4/D^2$, Eqs. (7) and (8) yield

$$0.55\gamma = \alpha \exp(-\gamma^{1/2}).$$
 (9)

A numerical solution of Eq. (9) shows that the ratio $k_1 = K_1/E_1 = \exp(-\gamma^{1/2})$ decreases as α approaches the critical value $\alpha_{\rm crit} \simeq 1$ from above and drops discontinuously to zero at $\alpha_{\rm crit}$. This discontinuity is a sign of a spurious first-order transition,⁶ a well-known artifact of the SCHA. However, for values of α well above 1 the SCHA yields a fairly accurate description of this "phase-locking" transition.

With the interaction of the supercurrent with the vector potential \vec{A} taken into account, the effective coupling of Eq. (6) gives rise to the following Ginzburg-Landau free-energy term:

$$F_c = \frac{1}{2} K_1 \int d^2 \boldsymbol{r} (\nabla \varphi - 2e\vec{\mathbf{A}}/\hbar c)^2.$$
 (10)

With use of this expression the supercurrent in the array is calculated to be

$$\vec{j}_{s} = -c\delta F_{c}/\delta\vec{A} = (2eK_{1}/\hbar)(\nabla\varphi - 2e\vec{A}/\hbar c).$$
(11)

Comparison of Eqs. (5) and (11) yields

$$n_s^{2D} = (4m/\hbar^2)K_1 = (4mE_1/\hbar^2)k_1.$$
 (12)

Equations (2) and (12) and the relation between $T_{\rm KT}$ and n_s^{2D} , derived in Ref. 2, produce the fol-

lowing relation:

$$(T_{\rm KT}/T_{c0})_{\rm SCHA} = (\pi\hbar^2/8m)n_s^{2D}$$

= $2.18 \frac{R_0}{R_{\rm C}} f(T_{\rm KT})k_1(R_{\rm C}^{N}),$ (13)

where $T_{c0} = \Delta(0)/1.76k_{\rm B}$. Equation (13) agrees with Eq. (8) of Ref. 1 except for the factor k_1 which represents the modification due to charging effects. To plot k_1 as a function of $R_{\Box}{}^N$ I use the relation (4). Near the transition, $\alpha(0) \approx 1$ and $(R_{\Box}{}^N)_{\rm crit} \approx 13 \text{ k}\Omega$ as indicated by the experiment.³ This implies for the charging parameter at the transition $U(R_{\Box}{}^N = 13 \text{ k}\Omega) \approx \Delta(0)$. Neglecting the slow variation of $d_{\rm ox}$ with $R_{\Box}{}^N$, I expect $U(R_{\Box}{}^N)$ $\sim l_c^{-1}$. The variation of l_c with $R_{\Box}{}^N$ can be deduced from Fig. 3 of Ref. 3 to be $l_c^{-1} \sim R_{\Box}{}^N$. Inserting the resulting dependence $U(R_{\Box}{}^N) \sim R_{\Box}{}^N$ into Eq. (4) yields

$$\alpha(0) = 13\pi R_{\rm o} / (R_{\rm o}^{N})^2.$$
(14)

With use of this result to translate the $\alpha(0)$ dependence of k_1 [obtained from Eq. (9)] into a $R_{\Box}^{\ N}$ dependence, Eq. (13) yields curve A of Fig. 1. The latter is in qualitative agreement with the experimental points. The experimental tail occurring for $R_{\Box}^{\ N} > 13 \text{ k}\Omega$ is, however, not reproduced by the present theory.⁷

Because the SCHA is expected to fail in the vicinity of $(R_{\Box}^{\ N})_{crit}$ it is useful to use in this region a mean-field approximation (MFA) to calculate the dependence of $T_{\rm KT}$ on $R_{\Box}^{\ N}$. As argued in Reg. 6, the MFA is justified for a three-dimensional array since the quantum mechanical nature of the order parameter effectively adds an extra dimension to the problem.⁸ I assume that MFA gives a fairly good picture of the T=0 transition for the two-dimensional film as well, since the zero-point fluctuations $\langle q_k q_k \rangle \sim k^{-1}$ do not produce an infrared divergence of the quantity D in Eq. (8).

The derivation of the dependence of T_{KT} vs R_{\Box}^{N} in the MFA involves steps similar to those of Ref. 3.

First the critical current density is calculated in the MFA from the Josephson relation⁹

$$J_0 \sin \varphi = \frac{1}{z l_c d_F} \frac{2c}{\hbar} \frac{\delta \mathcal{H}_c^{MFA}}{\delta \varphi}, \qquad (15)$$

where d_F is the film thickness and \mathcal{H}_c^{MFA} is the MFA for the coupling energy of Hamiltonian (1) (see Ref. 6)

$$\mathfrak{H}_{c}^{\mathrm{MFA}} = -2zE_{1}(\cos\varphi)\sum_{i}\cos\varphi_{i}.$$
 (16)

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Equations (15) and (16) yield

$$J_0 = (J_0)_{\rm H} \langle \cos\varphi\rangle, \tag{17}$$

where $(J_0)_{\rm H}$ coincides with Eq. (7) of Hebard's work.³ Using this result in the relation¹⁰

$$\lambda_{\rm eff}^2 = \hbar^2 / 8\pi J_0 l_c \tag{18}$$

and inserting the latter result into the relation ^ between the bulk superfluid density and $\lambda_{eff}{}^2$ I obtain

$$\left(\frac{T_{\mathrm{KT}}}{T_{c0}}\right)_{\mathrm{MFA}} = \frac{2.18R_0}{R_{\Box}^{N}} f(T_{\mathrm{KT}}) \langle \cos\varphi \rangle_{T=0}.$$
 (19)

The MFA for $\langle \cos \varphi \rangle_{T=0}$ obtained in Ref. 6 reads¹¹

$$\langle \cos\varphi \rangle_{T=0} \simeq 1.07 [(\alpha - 1)/\alpha^3]^{1/2}. \tag{20}$$

Relating the parameter α to R_{\Box}^{N} with the use of expression (14), Eqs. (19) and (20) yield the curve B of Fig. 1. In contrast to the SCHA result the MFA shows a continuous transition at $(R_{\Box}^{N})_{crit}$. For values of $R_{\Box}^{N} < 10 \text{ k}\Omega$ the MFA values for T_{KT} are, however, less reliable than the SCHA due to the fact that Eq. (20) is valid only for α very close to 1. Since the experimental values of $T_{\rm KT}$ for $R_{\Box}^{N} < 13 \text{ k}\Omega$ are obtained with a 1% of R_{\Box}^{N} criterion they are an overestimate of the actual $T_{\rm KT}$ which may account, at least partly, for the lack of complete quantitative agreement with the present calculation. The other possible reasons for this discrepancy may be the use of a simple linear dependence of U upon R_{\Box}^{N} leading to Eq. (14). A faster dependence would bring curves A and Bcloser to the experimental data.

Finally one should mention the possibility of the interplay between the Coulomb correlation, considered here, and the electron localization by disorder.¹² Since the electron level splitting caused by quantization in the large clusters is negligible compared to the charging energy, it is the latter which plays a dominant role in the electron localization of granular films.¹³

In conclusion, the present work shows that the zero-point phase fluctuations, due to the granular charging energy, can explain the observed rapid decrease of the Kosterlitz-Thouless transition temperature with the sheet resistance of the granular lead films.

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¹M. R. Beasley, J. E. Mooij, and T. P. Orlando, Phys. Rev. Lett. 42, 1165 (1979).

²B. I. Halperin and D. R. Nelson, J. Low Temp. Phys. <u>36</u>, 599 (1979).

³A. F. Hebard, in *Inhomogeneous Superconductors* —1979, edited by T. L. Francavilla, D. V. Gubser, J. R. Leibowitz, and S. A. Wolf, AIP Conference Proceedings No. 58 (American Institute of Physics, New York, 1979), p. 129.

 ${}^{4}A.$ F. Hebard and J. M. Vandenberg, Phys. Rev. Lett. <u>44</u>, 50 (1980).

⁵A. Skal and B. I. Shklovskii, Fiz. Tekh. Poluprovodn. <u>8</u>, 1586 (1974) [Sov. Phys. Semicond. <u>8</u>, 1029 (1975)]; see also P. G. de Gennes, J. Phys. (Paris), Lett. <u>37</u>, L1 (1976).

⁶E. Šimánek, Phys. Rev. B <u>22</u>, 459 (1979). See also P. G. de Gennes, private communication cited by T. Worthington, P. Lindenfeld, and G. Deutscher, Phys. Rev. Lett. 41, 316 (1978).

⁷The finite values of $T_{\rm KT}$ for $R^{\rm N} > 13 \ {\rm k}\Omega$ may be an experimental artifact. This conjecture is supported by preliminary measurements done in Ref. 4 on a sample with $R^{\rm N} > 12 \ {\rm k}\Omega$ showing a resistance which flattens out in the millikelvin range in spite of the finite value of $T_{\rm 2D}$ predicted on the basis of the higher-temperature data. I am indebted to A. F. Hebard for this remark.

⁸Specifically, the T = 0 properties of the model Hamiltonian (1) in *d* dimensions are identical to the finite temperature properties of the *XY* model in *d*+1 dimensions [see, e.g., A. Luther and D. J. Scalapino, Phys. Rev. B <u>16</u>, 1153 (1977)].

⁹B. D. Josephson, Adv. Phys. <u>14</u>, 419 (1965).

¹⁰B. Giovannini and L. Weiss, Solid State Commun. <u>27</u>, 1005 (1978).

¹¹The prefactor 1.07 is a correction of the value 1.2 used in Eq. (2.7) of Ref. 6.

¹²D. C. Liciardello and D. J. Thouless, Phys. Rev. Lett. <u>35</u>, 1475 (1975). Localization by disorder is mentioned in Ref. 3 as a possible source of the sudden drop of $T_{\rm KT}$ at $(R_{\Box}^{\ N})_{\rm crit}$.

¹³B. Abeles and Ping Sheng, in *Electrical Transport* and Optical Properties of Inhomogeneous Media, edited by J. C. Garland and D. B. Tanner, AIP Conference Proceedings No. 40 (American Institute of Physics, New York, 1978), p. 360.