

## Quantum Critical Phenomena in Charged Superconductors

Matthew P. A. Fisher and G. Grinstein

IBM T. J. Watson Research Center, Yorktown Heights, New York 10598

(Received 11 September 1987)

We study the effect of Coulomb interactions on the onset of superconductivity with increasing intergrain coupling in granular systems at  $T=0$ . We show that in three dimensions (3D) the onset transition is described by Euclidean scalar electrodynamics, and so can be either first or second order, depending on parameters. A renormalization-group analysis indicates that in 2D the transition can either be first order, or second order with 3D-XY-like critical behavior. In 1D Coulomb forces are shown to destroy the superconducting state.

PACS numbers: 64.60.Ak, 74.20.-z

In granular or amorphous superconductors, the onset of superconductivity with increasing intergrain coupling can be viewed as a phase transition at zero temperature. This transition has been carefully investigated in recent experiments<sup>1,2</sup> wherein the thickness or normal-state resistance of thin disordered films is systematically varied. One such study<sup>2</sup> found a transition which was continuous, as measured by film resistance, and exhibited apparently universal critical behavior.

A standard model for such systems consists of a regular array of Josephson-coupled grains. The model allows for quantum fluctuations of the phase of the order parameter on each grain.<sup>3</sup> In  $d$  space dimensions this model exhibits a  $T=0$  superconducting transition in the universality class of the  $(d+1)$ -dimensional XY model.<sup>3c</sup> Satisfactory comparison with experiment, however, requires incorporating into the theory several important complications, notably disorder,<sup>4</sup> dissipation,<sup>5</sup> and long-range (Coulomb) interactions.<sup>5b</sup> The purpose of this Letter is to elucidate the important effects of Coulomb forces on the superconducting onset transition at  $T=0$ . We study a model granular superconductor which consists of a fluid of charged bosons, representing the Cooper pairs, hopping on a regular lattice. Our main results follow.

For any dimensionality  $d \leq 3$  the long-range  $1/r$  potential is, to lowest order in the interaction strength, a marginal perturbation about the  $(d+1)$ -dimensional-XY fixed point characterizing the onset transition of the short-range model. In  $d=3$  the  $1/r$  potential is marginally *relevant*; the transition is described by Euclidean scalar electrodynamics, a theory believed<sup>6</sup> to exhibit either first- or second-order transitions, depending on parameters. In  $d=2$  the Coulomb force is marginally *irrelevant*. For some range of parameters the transition is 3D-XY-like, though for others it can be first order. In particular, amorphous (or homogeneous) films should tend to exhibit first-order behavior, whereas a continuous transition is expected in the extreme granular limit. This is consistent with the trend seen experimentally.<sup>1,2</sup> Finally, in  $d=1$  the  $1/r$  potential is marginally *relevant* again, and actually destroys the superconducting state, even at  $T=0$ .

Our model consists of charged bosons hopping on a lattice of grains (with unit lattice spacing) and interacting with the quantized electromagnetic (em) field. The grand canonical partition function (at  $T=0$ ) can be expressed<sup>7</sup> in a path-integral representation as a trace over a complex  $c$ -number Bose field,  $\Phi_i(\tau)$ , and the vector potential,  $\mathbf{A}$ :  $Z = \text{Tr}_{\Phi, \mathbf{A}} \exp(-S)$ , with an action ( $\hbar = c = 1$ )

$$S = \int d\tau \sum_i \Phi_i^* \partial_\tau \Phi_i - \int d\tau H(\Phi, \mathbf{A}) + S_{\text{em}}(\mathbf{A}), \quad (1a)$$

$$H = -t \sum_{\langle ij \rangle} (e^{iA_{ij}} \Phi_i^* \Phi_j + \text{c.c.}) + \sum_{ij} V_{ij} (N_i - \rho_0)(N_j - \rho_0). \quad (1b)$$

Here  $N_i = \Phi_i^* \Phi_i$  is the boson density on the  $i$ th site,  $V_{ij}$  represents the (static) Coulomb interaction,  $e^2/|i-j|$ , with Fourier transform  $V(k) \sim e^2 k^{-(d-1)}$  for  $d > 1$ , and  $\rho_0$  is a neutralizing positive charge background. The fluctuating transverse em field  $\mathbf{A}$  is coupled to the bosons in the usual way,<sup>7b</sup> with  $A_{ij} = e \int_i^j \mathbf{A} \cdot d\mathbf{l}$ . The trace over  $\mathbf{A}$  is taken in the Coulomb gauge,  $\nabla \cdot \mathbf{A} = 0$ . The associated photon action<sup>7b</sup> is then simply

$$S_{\text{em}}(\mathbf{A}) = \frac{1}{2} \int \frac{d^d k d\omega}{(2\pi)^{d+1}} (k^2 + \omega^2) |\mathbf{A}(\mathbf{k}, \omega)|^2. \quad (1c)$$

An interacting Bose fluid in the continuum will in general exhibit only one fluid phase at  $T=0$ —the superfluid.<sup>8</sup> The presence of the lattice (the grains) in (1), however, permits an additional (normal) phase at  $T=0$ , provided  $\rho_0$  is rational (i.e., commensurate with the lattice). In particular, for integer  $\rho_0$ , as will be considered in this Letter, (1) is expected to exhibit a  $T=0$  transition between a (normal) Mott insulating state and a superfluid state as the ratio  $t\rho_0/e^2$  is increased. With purely short-ranged interactions, this transition exhibits

$(d+1)$ -dimensional- $XY$ -like critical behavior.<sup>3c</sup> Thus, for example, the Mott gap  $E_g$  vanishes at the transition with the  $XY$  correlation-length exponent  $\nu$ . This gap controls the activated conductivity [ $\exp(-E_g/k_B T)$ ] in the insulating phase. As argued below, however, the critical properties are more complicated when long-range Coulomb forces are present.

The very existence of the  $T=0$  onset transition *requires* the presence of interactions (i.e.,  $e^2/t\rho_0 \sim 1$ ); thus

the transition is not accessible to perturbation theory in  $e$ . [This can be verified directly from (1).] It is therefore desirable to derive an alternative form for the effective action. The substitution  $\Phi_i = |\Phi_i| \exp(i\phi_i)$  in (1b) makes it apparent that amplitude fluctuations about the most probable value,  $|\Phi|^2 = \rho_0$ , are greatly suppressed by the interaction term. One may therefore expand  $|\Phi|$  about  $\sqrt{\rho_0}$  to quadratic order and integrate out the amplitude fluctuations, leaving an action in terms of the phase degrees of freedom alone, namely,

$$S = \frac{1}{2} \int d\tau \sum_{ij} (V^{-1})_{ij} \dot{\phi}_i \dot{\phi}_j - \int d\tau \sum_{ij} J_{ij} \cos(\phi_i - \phi_j - A_{ij}) + S_{em}, \quad (2)$$

where  $J_{ij} = J \equiv t\rho_0$  for  $i$  and  $j$  nearest neighbors, and  $J_{ij} = 0$  otherwise. Since  $\Phi_i(\tau)$  is periodic in  $\tau$  with period  $\beta \equiv 1/k_B T$ , the phase satisfies the boundary condition  $\phi_i(\beta) = \phi_i(0) + 2\pi m_i$ , for integers  $m_i$ . Note that in general (2) should include the complex term  $i\rho_0 \int \sum_i \dot{\phi}_i$ . For integer  $\rho_0$ , as considered here, however, the periodicity of  $\phi_i$  implies that this term vanishes. Thus, unlike (1), the action (2) is real.

This action can, alternatively, be obtained by our expressing in path-integral form the partition function associated with the Josephson-junction-type Hamiltonian,<sup>3</sup>

$$\hat{H} = \frac{1}{2} \sum_{ij} \hat{n}_i V_{ij} \hat{n}_j - J \sum_{\langle ij \rangle} \cos(\hat{\phi}_i - \hat{\phi}_j - A_{ij}) + \hat{H}_{em}. \quad (3)$$

Here  $[\hat{\phi}_i, \hat{n}_j] = i\delta_{ij}$ , and  $\rho_0$  is an integer; for general  $\rho_0$ ,  $\hat{n}$  should be replaced by  $\hat{n} - \rho_0$  in (3). Because of the boundary condition on the phase, the eigenvalues of the operator  $\hat{n}$  are integers and represent the deviation of the boson density from the mean value  $\rho_0$ .

In the superfluid state (large  $J$ ), it is legitimate to expand the cosine in (2) to quadratic order, producing a Gaussian kernel  $\omega^2 V^{-1}(k) + Jk^2$ , with the plasmon excitation spectrum  $\omega^2 \sim Jk^2 V(k)$ . For short-range interactions,  $V(k=0)$  is finite and  $\omega$  and  $k$  appear symmetrically, which is why the  $T=0$  transition has isotropic  $(d+1)$ -dimensional- $XY$  critical behavior. Coulomb interactions make the kernel anisotropic in space-time, however, producing a potentially different transition.<sup>9</sup> To investigate this possibility, we first recast the action (2) in a form where standard techniques can be applied. Introduction of an auxiliary on-site real field  $A_{0i}(\tau)$  and a complex field  $\psi_i(\tau)$  allows, via the familiar Hubbard-Stratanovich<sup>3c</sup> transformation, the decoupling in (2) of the fields  $\phi_i$  and  $\exp(i\phi_i)$ , respectively, on different sites  $i$ . The partition function is thereby rewritten in the form  $Z = \text{Tr}_{\phi, \mathbf{A}, A_0, \psi} \exp[-(S_1 + S_2 + S_{em})]$ , where

$$S_1 = \frac{1}{2} \int_{k, \omega} [e^{-2V(k)} - 1]^{-1} |A_0|^2 + \frac{1}{2e^2} \int d\tau \sum_i (\dot{\phi}_i - eA_{0,i})^2, \quad (4a)$$

$$S_2 = \frac{1}{2} \int d\tau \sum_{ij} (J^{-1})_{ij} e^{iA_{ij}} \psi_i^*(\tau) \psi_j(\tau) - \sum_i \int d\tau [\psi_i(\tau) e^{-i\phi_i(\tau)} + \text{c.c.}]. \quad (4b)$$

In the following we allow for interactions of arbitrary range, i.e.,  $V(k) \sim e^2/k^\sigma$  for any  $\sigma \geq 0$ , corresponding to a real-space potential varying as  $|i-j|^{-\sigma-d}$ . The  $1/r$  Coulomb potential corresponds to  $\sigma = d-1$ . Treating the last term in (4b) perturbatively, one can now integrate out the  $\{(\phi_i)\}$  to generate a cumulant expansion in the fields,  $\psi_i$ . Keeping only terms up to  $O(\psi^4)$  and replacing the spatial lattice by the usual continuum with a high-momentum cutoff  $\Lambda$  yields the desired field-theoretic representation  $Z = \text{Tr}_{\mathbf{A}, A_0, \psi} \exp(-S)$ , where

$$S = S_0 + \frac{1}{2} \int d^d x d\tau [(\partial_\tau - ieA_0)\psi|^2 + |(\nabla - ie\mathbf{A})\psi|^2 + r|\psi|^2 + u|\psi|^4], \quad (5a)$$

$$S_0 = \frac{1}{2} \int_{k, \omega} k^\sigma |A_0|^2 + S_{em}(\mathbf{A}). \quad (5b)$$

Only the leading small- $k$  limit of the  $|A_0|^2$  term has been kept in (5b). This suffices for the study of the critical properties. We now consider the cases  $d=3, 2$ , and  $1$  in order.

For  $d=3$  the static Coulomb interaction corresponds to  $\sigma=2$ , whereupon model (5) becomes identical to the theory of Euclidean scalar electrodynamics in four (i.e.,  $3+1$ ) dimensions.<sup>6</sup> It is well known<sup>10</sup> that this model describes a 4D superconductor coupled to a fluctuating

transverse em field at *finite temperature*; as such it has been studied via renormalization-group (RG) methods. It has also been extensively considered in the context of lattice-gauge-Higgs theories.<sup>6</sup> The absence of a stable fixed point of the perturbative RG originally suggested the occurrence of a fluctuation-induced first-order phase transition between the normal and superconducting phases. Subsequent Monte Carlo studies show<sup>6</sup> strong

indications that the transition may, depending on parameters, be either first or second order, a tricritical point separating these two regimes. However, the absence of an accessible RG fixed point has so far prevented identification of the universality class of the continuous transition. The possibility of experimental observation of  $T=0$  onset transitions of either first or second order in  $d=3$  is an interesting one nonetheless.

In  $d=2$  (or  $d=1$ ) the very long transverse magnetic penetration depth typical of experimental samples implies that the coupling of the order parameter to the transverse em field manifests itself only unobservably close to the onset transition.<sup>11</sup> The physics is therefore best described by our setting  $\mathbf{A}$  equal to zero in (5), producing a model where the "gauge" field has but one component,  $A_0$ . Hereafter we consider only this simplified limit.

To investigate the resulting critical behavior of model (5) with  $\mathbf{A}=0$ , first note that, for any  $d$  and  $\sigma$ , the model is invariant under the "gauge" transformation

$$\psi = \tilde{\psi} \exp[ie\lambda(\tau)], \quad A_0 = \tilde{A}_0 + \partial_\tau \lambda, \quad (6)$$

for any function  $\lambda(\tau)$ . Now setting the charge  $e$  equal to zero decouples  $A_0$  from  $\psi$ , reducing the  $\psi$  part of  $S$  to the ordinary  $\psi^4$  representation of the  $XY$  model. Thus the model has an isotropic  $(d+1)$ -dimensional- $XY$  fixed point at  $e=0$ . To study the stability of this fixed point for small but nonzero  $e$ , imagine one's performing (still for arbitrary  $d$  and  $\sigma$ ) a  $T=0$  RG transformation<sup>12</sup> by integrating out of the partition function all Fourier components  $\psi(k, \omega)$  and  $A_0(k, \omega)$  with high (spatial) momenta,  $\Lambda/b < k < \Lambda$ , with rescaling factor  $b > 1$ . Rescaling fields, momenta, and frequencies according to  $k = k'b^{-1}$ ,  $\omega = \omega'b^{-z}$ ,  $\psi(k, \omega) = \psi'(k', \omega')b^{\zeta_\psi}$ ,  $A_0 = A'_0 b^{\zeta_A}$ , we choose  $\zeta_\psi$ ,  $\zeta_A$ , and  $z$  to hold the coefficients of  $k^\sigma |A_0|^2$ ,  $(\partial_\tau \psi)^2$ , and  $k^2 |\psi|^2$  fixed at  $\frac{1}{2}$ . The parameter  $z$  allows for the possibility of anisotropic scaling in space-time. It is easy to see that, quite generally, the transformed action  $S'(\psi', A'_0)$  continues to satisfy a gauge invariance of the form (6), but with a renormalized charge  $e' = b^{\zeta_A - d} e$ . Thus the sign of  $\zeta_A - d$  determines the stability of the  $(d+1)$ -dimensional- $XY$  fixed point at  $e=0$ . The isotropy of this fixed point implies  $z=1$ , whereupon, for  $\sigma < 2$ , the invariance of the coefficient of  $k^\sigma |A_0|^2$  immediately gives (since  $k^\sigma$ , being nonanalytic, does not get renormalized)  $2(\zeta_A - d) \equiv 1 + \sigma - d$ . Hence, for all  $\sigma < 2$ , the  $XY$  fixed point is stable for  $\sigma < d-1$  (i.e., for sufficiently short-ranged interactions), and unstable for  $\sigma > d-1$ . The physically interesting Coulomb case,  $\sigma = d-1$ , is precisely marginal for all  $d < 3$ ; a stability determination thus requires more detailed RG calculations. Such calculations are prohibitively difficult directly in  $d=2$  where the  $XY$  fixed point is not perturbatively accessible, but can be performed in an  $\epsilon$  expansion about the upper critical dimension,  $d=3$ , and cautiously extrapolated to  $d=2$ .

When  $d=3-\epsilon$  the marginal Coulomb case has  $\sigma=2-\epsilon$ . More generally, one can write  $\sigma=2-\epsilon_\sigma$ , and perform a double expansion in  $\epsilon$  and  $\epsilon_\sigma$ . Using the above rescalings one readily generates differential recursion relations to lowest nontrivial order:

$$dw/dl = (\epsilon - \epsilon_\sigma)w - w^2/3, \quad (7)$$

$$du/dl = \epsilon u - 10u^2 + \frac{1}{2}wu - w^2/16.$$

Here  $w \equiv e^2$  and  $l \equiv \ln b$ . A schematic flow diagram corresponding to (7) for the Coulomb case  $\epsilon_\sigma = \epsilon$  is shown in Fig. 1. Most of the points in the physically accessible first quadrant flow off to negative values of  $u$ . Since (5) is unstable for  $u < 0$ , this strongly suggests a fluctuation-induced first-order<sup>10</sup> onset transition. There is, however, a wedge of thickness  $\epsilon$  along the  $u$  axis which flows, as shown, into the  $XY$  fixed point at  $w=0, u>0$ . Thus the  $XY$  fixed point is marginally stable. The separatrix between these two regimes flows into a (Gaussian) tricritical point at  $w=u=0$ . Although the domain of attraction of the  $XY$  fixed point is small of  $O(\epsilon)$ , the assumption that this domain grows continuously with  $\epsilon$  implies that, for  $d=2$  ( $\epsilon=1$ ), the  $XY$  fixed point attracts some finite ( $\sim 1$ ) fraction of the first quadrant. Presumably then, the  $d=2$  system can undergo either a 3D- $XY$  superconducting transition (for large enough  $u$ ), or a fluctuation-induced first-order transition (for small  $u$ ). Since  $u$  suppresses amplitude fluctuations of  $\psi$  in (5a), increasing  $u$  corresponds to increased granularity (or inhomogeneity) in the film. Consequently, amorphous (or homogeneous) films should tend to undergo first-order transition, whereas granular films should exhibit continuous  $XY$  transitions. This is consistent with the trend seen experimentally.<sup>1,2</sup> It is of course possible that additional stable fixed points with different critical behavior occur in the region of the

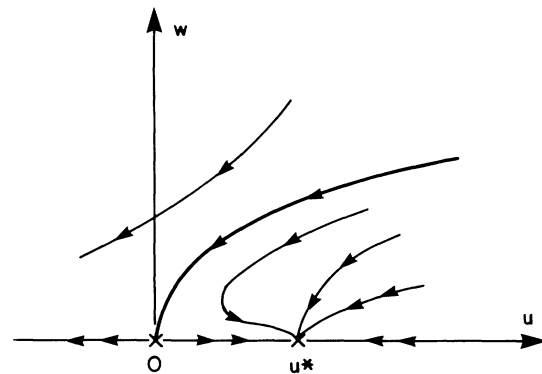


FIG. 1. Flow diagram for  $T=0$  onset transition in  $d=3-\epsilon$  expansion. Separatrix (bold) divides flows to unstable ( $u < 0$ ) region from those attracted to  $(d+1)$ -dimensional- $XY$  fixed point.

figure inaccessible to perturbation theory.<sup>13</sup>

For  $d=1$  (1+1 space-time dimensions), methods developed for treating the two-dimensional  $XY$  model are more useful than representation (5). The idea is to factorize the partition function into a product of a spin wave and vortex contributions in the standard way.<sup>14</sup> [To this end it is convenient to discretize time in the action in (2),

$$S = \frac{1}{2} \int_{k,\omega} [V(k)k^2 + J^{-1}\omega^2] |\theta(k,\omega)|^2 - U \int_{x,\tau} \cos[2\pi\theta(x,\tau)]. \quad (8)$$

Here  $\theta(x,\tau)$  is a scalar field and  $U$  the vortex fugacity.

For short-range interactions,  $V(k=0)$  is finite, and the model reduces to a sine-Gordon theory, with associated Kosterlitz-Thouless transition.<sup>14</sup> For the  $1/r$  interaction,  $V(k)$  is singular ( $\sim \ln k$ ) at small  $k$ . A RG calculation, perturbative in  $U$ , can nevertheless be performed. Integrating out momenta  $\Lambda/b < k < \Lambda$  ( $\Lambda$  is the ultraviolet cutoff), and all frequencies, one obtains the first-order differential recursion relation,  $\partial U/\partial l = 2U$ . Hence, in contrast to the short-range case,  $U$  always increases under the RG flow: The Gaussian fixed line is unstable and the vortices are always unbound. The algebraically ordered superfluid state of the short-range problem has thus been destroyed by the Coulomb interaction. Physically, the Coulomb force suppresses charge (or number) fluctuations at long wavelengths, thereby enhancing quantum fluctuations in the conjugate variable—the order-parameter phase—and destroying the superfluidity. Interactions longer ranged than  $1/r$  also wipe out the ordered state, whereas interactions which fall off faster than  $1/r$  are effectively short ranged, producing the usual Kosterlitz-Thouless transition.

We thank D. S. Fisher, J. Toner, and P. Weichman for helpful conversations. One of us (M.P.A.F.) is grateful to the Aspen Center for Physics where part of this work was completed.

and replace the cosine term by the familiar Villain form.<sup>14]</sup> The vortex contribution describes a neutral two-dimensional plasma with anisotropic interactions between charges in space-time. One then performs a duality transformation<sup>14</sup> on this plasma. In the limit of large vortex core energies the resulting dual model, with continuous time restored, is a generalized sine-Gordon theory<sup>14</sup> with an action

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<sup>4</sup>M. Ma, B. I. Halperin, and P. A. Lee, Phys. Rev. B **34**, 3136 (1986).

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<sup>5b</sup>Matthew P. A. Fisher, Phys. Rev. B **36**, 1917 (1987). Coupled Josephson-junction-type models which ignore dissipative processes (due, say, to the diffusive behavior of unpaired electrons) predict insulator-to-superconductor transitions at  $T=0$  and cannot account for those experiments where a continuous metal-to-superconductor transition is observed, as e.g., in Ref. 2.

<sup>6</sup>See, e.g., J. Jersak, in *Lattice Gauge Theory*, edited by B. Bunk, K. H. Mutter, and K. Schilling (Plenum, New York, 1986), p. 133, and references therein.

<sup>7a</sup>See, e.g., L. S. Schulman, *Techniques and Applications of Path Integration* (Wiley, New York, 1981), Chap. 27.

<sup>7b</sup>C. Itzykson and J. Zuber, *Quantum Field Theory* (McGraw-Hill, New York, 1980), Chap. 8.

<sup>8</sup>For  $T > 0$ , a continuum Bose fluid exhibits a normal (nonsuperfluid) state below a critical density  $\rho_c(T)$ . Since  $\rho_c$  vanishes at  $T \rightarrow 0$ , the associated superfluid-to-normal transition occurs at strictly zero density when  $T=0$ . See, e.g., E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics II* (Pergamon, New York, 1981), Chap. 23; P. B. Weichman, M. Rasolt, M. E. Fisher, and M. J. Stephen, Phys. Rev. B **33**, 4632 (1986); D. D. Vvedensky and R. J. Creswick, Phys. Rev. B **34**, 7760 (1986).

<sup>9</sup>The critical behavior at  $T > 0$  involves only the zero Matsubara frequency, and so is unaffected by the Coulomb interaction. See S.-k. Ma, Phys. Rev. Lett. **29**, 1311 (1972).

<sup>10</sup>B. I. Halperin, T. C. Lubensky, and S.-k. Ma, Phys. Rev. Lett. **32**, 292 (1974). The Coulomb gauge ( $\nabla \cdot \mathbf{A} = 0$ ) makes the coefficients of  $|A_0|^2$  and  $|\mathbf{A}|^2$  in (5b) different, even when  $\sigma=2$ . These coefficients would both be  $k^2 + \omega^2$  in the Lorentz gauge,  $\partial A_0/\partial t + \nabla \cdot \mathbf{A} = 0$ .

<sup>11</sup>M. R. Beasley, J. E. Mooij, and T. P. Orlando, Phys. Rev. Lett. **42**, 1165 (1979).

<sup>12</sup>J. A. Hertz, Phys. Rev. B **14**, 1165 (1976).

<sup>13</sup>See, e.g., C. Dasgupta and B. I. Halperin, Phys. Rev. Lett. **47**, 1556 (1981).

<sup>14</sup>See, e.g., J. V. Jose, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, Phys. Rev. B **16**, 1217 (1978).

<sup>1</sup>See A. F. Hebard and M. A. Paalanen, Phys. Rev. B **30**, 4063 (1984); A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **33**, 3549 (1986); R. C. Dynes, A. E. White, J. M. Graybeal, and J. P. Garno, Phys. Rev. Lett. **57**, 2195 (1986).

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<sup>3a</sup>See E. Simanek, Solid State Commun. **31**, 419 (1979).

<sup>3b</sup>K. B. Efetov, Zh. Eksp. Teor. Fiz. **78**, 2017 (1980) [Sov. Phys. JETP **51**, 1015 (1980)].