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The γ–α transition in cerium compounds

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Abstract. The γ–α transition and the related volume collapse are explained within a compressible Kondo lattice where the variation of $|J|/D$ with volume is taken into account. We show that, contrary to the promotion model, the Kondo contribution is sufficient to induce a first-order transition at low temperature from a magnetic to a Kondo phase. The α phase is then characterised by an extremely high Kondo temperature.

The calculations are also made at zero and finite temperatures and the existence of a critical point is demonstrated. Finally we propose a generalisation to the effect of a chemical pressure. Applications are made to Ce, $CeAl_2$ and $Ce_{1-c}Th_c$.

1. Introduction

Depending on the pressure, pure cerium exhibits two distinct phases (both FCC): a high-volume local moment γ phase and a low-volume, enhanced Pauli paramagnetic α phase. More generally, in the intermetallic cerium compounds, the effective volume and the magnetic character of cerium can also be characterised as being γ like or α like (see $CeAl_2$—Croft and Jayaraman 1979, $CeIn_3$). Similar results can be obtained using a chemical pressure ($Ce_{1-c}Th_c$—Lawrence *et al* 1975, $Ce_{1-c}Sc_cAl_2$—Croft and Levine 1982) and the inverse transition $\alpha \rightarrow \gamma$ has even been observed when acting with a 'negative' chemical pressure ($Ce_{1-c}La_cPd_3$—Croft and Levine 1982). In each case, the effect of pressure is to induce a transition from a magnetic Kondo type γ phase to a non-magnetic α phase. At low temperature, the transition is of first order with a volume collapse. A critical point may be observed above which the transition becomes second order.

Different models for understanding these two phases and the transition between them have been proposed, based on the following points.

(i) The promotion of part of the 4f electron into the conduction band (the valence change model: Coqblin and Blandin 1968).

(ii) The increase of the 4f band width leading to a Mott transition (Johansson 1974). This idea has also been developed by Glötzel and Podloucky (1980) who interpreted Compton experiments from band calculations on α and γ cerium.

(iii) The increase of the f level d band hybridisation (Hill and Kmetko 1975).

In recent years, the controversy between these theoretical schemes has heightened, especially with photoemission experiments, which have brought new ways of understanding these phenomena. From the photoemission results on $CeAl_2$ (whether or not it is alloyed with Y and Sc—Croft *et al* 1981, CeTh—Parks *et al* 1982) and on pnictides and chalcogenures of cerium (Franciosi *et al* 1981), it seems that the pressure

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does not affect the position of the impurity level but rather acts on the degree of localisation of this 4f state. This goes against the promotion model. Other objections to this model have been made from positron annihilation, Compton and neutron scattering experiments, melting point and cohesive energy arguments (see Shapiro *et al* 1977 and references given by Croft *et al* 1981).

Following these ideas, we propose here a model of a 'compressible' Kondo lattice where the Kondo interaction (i.e. T_K) is enhanced under the effect of the pressure. The phase diagram of the Kondo lattice is given in figure 1 as a function of $|J|/D$ (J being the Kondo exchange interaction and D the half-width of the conduction band). At zero temperature, there is a succession of two phases: the mixed Kondo phase where the magnetic moment is reduced by the Kondo interactions, and the pure Kondo phase where the magnetic moment is zero. A compression of the system leading to an enhancement of $|J|/D$ (see the Slater rules below) can induce a transition from the mixed to the pure Kondo phase. The purpose of this paper is to show that the Kondo contribution, added to the usual elastic energy of the matrix, can lead to a negative curvature (i.e. an instability) in the free energy of the system. The transition is first order and an instability appears in the phase diagram.

In § 2, we give a brief review of the Kondo lattice and in § 3 we examine the effect of the pressure at zero temperature: a short account of this has been published recently (Lavagna *et al* 1982). Applications to Ce and $CeAl_2$ are made and we will discuss the position of the instability region related to the magnetic transition line. The non-magnetic α phase is interpreted as a pure Kondo phase of extremely high T_K : this is the same idea as that suggested some time ago by Schrieffer (1967) for the case of dilute alloys. The study is then extended to finite temperatures in § 4: when the temperature increases, we expect the instability region to become narrower until it vanishes at a critical point. Finally in § 5 this work is generalised to the case of a chemical pressure and applied to compounds such as $Ce_{1-c}Th_c$.

2. Review of the Kondo lattice

The Kondo effect for impurities of dilute rare earths is quite well understood within single impurity models. However, a Kondo effect has also been observed in concentrated alloys or compounds: $CeAl_2$ and TmSe exhibit magnetic ordering at low temperatures with a reduced magnetic moment compared to the free ion; $CeAl_3$ and SmB_6 are non-magnetic down to zero temperature.

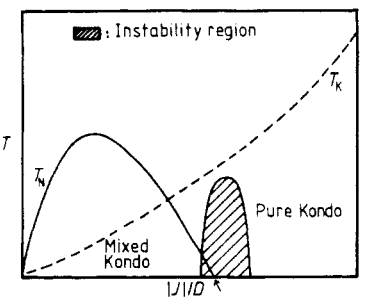

Figure 1. Phase diagram of the Kondo lattice. The arrow indicates $|J|/D_{crit}$.

In order to explain these anomalous behaviours, Doniach (1977) introduced the Kondo lattice model which is a generalisation of the Kondo Hamiltonian to the concentrated case. On each site of the lattice, one rare-earth ion interacts with the conduction electrons. The exchange interaction between the conduction and 4f electrons is responsible for both the Kondo effect and the magnetism through the RKKY interactions. The model is interesting as it gives quite a good description of the competition between the Kondo effect and the magnetism. Two different approaches exist to solve this problem.

(i) Doniach (1977) considered a one-dimensional analogue, the Kondo necklace. In the mean-field approximation, he found that the ground state is antiferromagnetic for low $|J|/D$ and is a non-magnetic Kondo singlet for large $|J|/D$. These results were confirmed by Jullien *et al* (1977a, b) using a renormalisation group treatment.

(ii) Another approach is that of Lacroix and Cyrot (1979) who transformed the initial Kondo interaction into a fictitious d–f hybridisation within a functional integral formalism. The following phase diagram is reported in figure 1. The ground state is magnetic for low $|J|/D$ and pure Kondo above a critical value. An intermediate phase exists called the 'mixed' phase where both effects coexist with a reduced magnetic moment. When the number of conduction electrons per site equals one, the system is insulating.

In this paper, the model of Lacroix and Cyrot is generalised in order to introduce the effect of pressure on the Kondo lattice.

3. Isotherms at zero temperature

The effect of the pressure on the Kondo lattice is studied within the model of Lacroix and Cyrot:

$$H=\sum_{k\sigma}(\varepsilon_k+\tfrac{1}{4}Jm_{-\sigma})c^{\dagger}_{k\sigma}c_{k\sigma}+\sum_{i\sigma}(E_0+\tfrac{1}{4}Jn_{-\sigma})d^{\dagger}_{i\sigma}d_{i\sigma}$$
$$+\tfrac{1}{2}J\sum_i x_i(d^{\dagger}_{i\uparrow}c_{i\uparrow}+c^{\dagger}_{i\downarrow}d_{i\downarrow})+\tfrac{1}{2}J\sum_i y_i(d^{\dagger}_{i\downarrow}c_{i\downarrow}+c^{\dagger}_{i\uparrow}d_{i\uparrow}) \tag{3.1}$$

where $c^{\dagger}_{k\sigma}$, $d^{\dagger}_{i\sigma}$, ε_k and E_0 have their usual meaning; x_i and y_i, determined self-consistently, represent the fictitious f–d hybridisation; $m_\sigma=\langle d^{\dagger}_{\sigma}d_{\sigma}\rangle$ and $n_\sigma=\langle c^{\dagger}_{\sigma}c_{\sigma}\rangle$. The resolution of this Hamiltonian leads to the following d and f densities of states:

$$\rho^{\sigma}_c(\omega)=\frac{1}{2D}$$
$$\rho^{\sigma}_d(\omega)=\frac{J^2x^2}{8D(\omega-E_0-\frac{1}{2}Jn_{-\sigma})^2} \tag{3.2}$$

for $\omega^{\sigma}_1<\omega<\omega^{\sigma}_2$ and $\omega^{\sigma}_3<\omega<\omega^{\sigma}_4$ with

$$\omega^{\sigma}_1=\tfrac{1}{2}[-D+E_0+\tfrac{1}{2}J(n_{-\sigma}+m_{-\sigma})-\{[D+E_0+\tfrac{1}{2}J(n_{-\sigma}-m_{-\sigma})]^2+J^2x^2\}^{1/2}]$$
$$\omega^{\sigma}_2=\tfrac{1}{2}[D+E_0+\tfrac{1}{2}J(n_{-\sigma}+m_{-\sigma})-\{[-D+E_0+\tfrac{1}{2}J(n_{-\sigma}-m_{-\sigma})]^2+J^2x^2\}^{1/2}]$$
$$\omega^{\sigma}_3=\tfrac{1}{2}[-D+E_0+\tfrac{1}{2}J(n_{-\sigma}+m_{-\sigma})+\{[D+E_0+\tfrac{1}{2}J(n_{-\sigma}-m_{-\sigma})]^2+J^2x^2\}^{1/2}] \tag{3.3}$$
$$\omega^{\sigma}_4=\tfrac{1}{2}[D+E_0+\tfrac{1}{2}J(n_{-\sigma}+m_{-\sigma})+\{[-D+E_0+\tfrac{1}{2}J(n_{-\sigma}-m_{-\sigma})]^2+J^2x^2\}^{1/2}].$$

As soon as $x\neq 0$, a gap opens in the density of states which is of the order of T_K at zero temperature. The width of the resonance in the impurity density of states is of the same

order, i.e. T_K. The origin of the gap comes from the hybridisation of the rare-earth level with the conduction band and is directly related to the periodicity of the lattice. The gap may disappear in various situations; for example, if one of the following effects is considered: degeneracy of the f orbitals; intersite interaction between f and d electrons. Nevertheless, the existence of the gap is not fundamental here and the results would not be much different if there was no gap.

The self-consistent conditions on E_0 and E_F, and the resulting expression of the magnetic moments μ_c and μ_d are given in the appendix within the limit $J^2x^2/4n_\sigma m_\sigma D^2 \ll 1$ (which is verified in both Kondo and magnetic phases). In the following the same limit is used in order to simplify the calculations. The Kondo contribution to the free energy has the following form at $T=0$:

$$\Delta F = \sum_\sigma \int_{\omega_1^\sigma}^{\tilde{E}_F} \omega[\rho_c^\sigma(\omega) + \rho_d^\sigma(\omega)]\, d\omega - \tfrac{1}{2}Jx^2 - \tfrac{1}{2}J\sum_\sigma n_\sigma m_{-\sigma}. \tag{3.4}$$

Using the relations (A.3) and (A.4):

$$\Delta F = -\tfrac{1}{2}Jx^2\left[1 - \frac{J}{2D}\left(-1 + \ln\frac{J^2x^2}{16(n_\uparrow n_\downarrow m_\uparrow m_\downarrow)^{1/2}D^2}\right)\right]$$
$$+ \sum_\sigma n_\sigma(n_\sigma - 1)D + \sum_\sigma m_\sigma(E_0 + \tfrac{1}{2}Jn_{-\sigma}). \tag{3.5}$$

For the energy of the mixed and pure Kondo phase we find:

$$\Delta F_{\text{mixed}} = -\frac{Jx^2}{2}\left[1 - \frac{J}{2D}\left(-\frac{1}{2} + \ln\frac{J^2x}{8nD^2}\right)\right] - \frac{J^2}{32D}$$
$$\Delta F_{\text{pure}} = -\frac{Jx^2}{2}\left[1 - \frac{J}{2D}\left(-1 + \ln\frac{J^2x^2}{4nD^2}\right)\right]. \tag{3.6}$$

The zero of the energies is that of the non-magnetic, non-Kondo state. The total number of conduction electrons is n.

At $T=0$, the free energy is reduced to its minimum value which occurs respectively at

$$\frac{J^2x_0}{8nD^2} = \exp(2D/J) \qquad \text{for the mixed Kondo phase}$$
$$\frac{J^2x_0^2}{4nD^2} = \exp(2D/J) \qquad \text{for the pure Kondo phase} \tag{3.7}$$

leading to:

$$\Delta F^0_{\text{mixed}} = D\left[-8\left(\frac{nD}{J}\right)^2 \exp(4D/J) - \frac{J^2}{32D^2}\right]$$
$$\Delta F^0_{\text{pure}} = -nD\exp(2D/J). \tag{3.8}$$

One can easily verify that the two relations (3.7) and (3.8) are effectively continuous at the magnetic transition (for $x=\frac{1}{2}$, cf appendix). For example, the critical value of $|J|/D$ (noted by the arrow on figure 1) is evaluated from equation (3.7) to be 0.46 for $n=1$, and 0.67 for $n=0.5$.

The free enthalpy is obtained by adding the usual elastic energy of the matrix, i.e. of the normal system without Kondo or magnetic effects. Noting B and V_0 the bulk modulus and

the atomic volume of such a system:

$$G = \Delta F^0 + \tfrac{1}{2}BV_0[(V - V_0)/V_0]^2 + PV. \tag{3.9}$$

The model introduced by a 'compressible' Kondo lattice consists of taking into account the volume dependence of $|J|/D$:

$$|J|/D = |J|/D|_0 \exp[-q(V - V_0)/V_0]. \tag{3.10}$$

q is a coefficient which can be approximated from the Slater rules: usually q is taken to be between 6 and 8.

The P–V isotherm is obtained by putting equation (3.10) into $\partial G/\partial V = 0$.

The Kondo effect always leads to a contraction of the system and the reduction of volume at zero pressure agrees with experimental results.

In the same way, the bulk modulus of the system at normal pressure can be deduced from the second derivative of the free energy:

$$B' = -V_0 \frac{\partial^2 F}{\partial V^2} = B - D\frac{q^2}{V_0}\left\{32\left(\frac{D}{J}\right)^2 \exp(4D/J)\left[\left(1+\frac{2D}{J}\right)^2 + \frac{D}{J}\right] + \frac{J^2}{8D^2}\right\}. \tag{3.11}$$

The second term represents the Kondo contribution and is negative: the bulk modulus is always lower than it would be if the system was 'normal'.

Applications are made to the case of cerium and $CeAl_2$. No gap has been observed in these compounds, undoubtedly for the reasons given above but, as explained before, this is not a real difficulty for the validity of the model. The values of the parameters are indicated in table 1.

(i) *$CeAl_2$*. The transition is found to be of first order at a critical pressure of 45 kbar with a volume collapse of 8%. The instability region overlaps the magnetic transition (as in figure 1). The Kondo temperature at ambient pressure is of the order of 1 K, and the one just below the transition is 100 K. At last, the bulk modulus is reduced by the Kondo contribution (which is of the order of −38 kbar). The obtained value of 712 kbar is close to the experimental value of 710 kbar (Penney *et al* 1982).

(ii) *Ce*. The results are similar to the previous case with a critical pressure of −3 kbar and a volume collapse of the order of 15%, in agreement with experimental results.

In summary, the pressure induces a transition from a mixed Kondo phase (γ type) to a pure Kondo phase of extremely high T_K. In both the cases considered here, the Kondo temperature in the α phase, i.e. above the transition, is particularly high, of the order of 1000 K (Ce), and 5000 K ($CeAl_2$). This constitutes our description of the α phase: contrary to the valence change model, the disappearance of magnetism is associated with a very high Kondo temperature.

Table 1. Values of parameters for $CeAl_2$ and Ce ($n = 1$).

	D (eV)	$\lvert J\rvert/D\rvert_0$	q	B (kbar)	$V_0^\dagger$ (Å^3)
$CeAl_2$	2.2	0.26	7	745‡	$\frac{523}{8}$§
Ce	0.4	0.4	7	240	34

† V_0 is the atomic volume at normal pressure of the equivalent 'normal' system without Kondo interaction.

‡ The bulk modulus of 'normal' $CeAl_2$ is extrapolated between the values of $LaAl_2$ and $PrAl_2$ (as for V_0).

§ The value of V_0 at 0 K is extrapolated from that at 300 K with the coefficient of thermal dilatation given by Walker *et al* (1973).

4. Effect of temperature and critical point

When T is different from zero, the problem is a little more complicated and we restrict ourselves to the simple case of a pure Kondo phase. The Kondo contribution to the free energy is:

$$\Delta F = -2kT \int_{\omega_1,\omega_3}^{\omega_2,\omega_4} (\rho_c(\omega) + \rho_d(\omega) \ln\left[\exp\left(\frac{\omega - E_F}{2kT} \right) + \exp\left(-\frac{\omega - E_F}{2kT} \right) \right] d\omega - \frac{Jx^2}{2} - \frac{Jn}{4}. \tag{4.1}$$

This is solved in the particular case $n=1$ where E_0 and E_F are independent of the temperature and equal to 0 and $\frac{1}{4}J$:

$$\Delta F = -\frac{Jx^2}{2}\left[1 - \frac{J}{2D}\left(-1 + \ln \frac{J^2x^2}{4D^2} \right) \right] + 2kT \sum_{p=1}^{\infty} (-1)^p E_2\left(p \frac{J^2x^2}{4DkT} \right) p^{-1}. \tag{4.2}$$

The last temperature-dependent term is a series of exponential second-order integrals. The P–V isotherm can be deduced in the same manner as detailed in § 3, being careful to take the statistical average of ΔF on all the values of x:

$$\Delta F = \frac{\int \Delta F \exp(-\Delta F/kT)\, dx}{\int \exp(-\Delta F/kT)\, dx}. \tag{4.3}$$

Applications are made to $CeAl_2$ and Ce going to the twentieth term in the series of exponential integrals.

(i) *$CeAl_2$*. The values of the parameters chosen are those of table 1 except for V_0 where we have to take into account the thermal dilatation (at 300 K, for example, V_0 is taken equal to 527 Å^3 for eight formulae). At 300 K, the transition is still found to be of first order for a pressure of 65 kbar, in agreement with the experimental results (Croft and Jayaraman 1979). The calculations lead to a critical point at 4000 K and 125 kbar. The critical point occurs well above the fusion temperature of 1800 K. In our model for $CeAl_2$ we expect a first-order transition at all temperatures.

(ii) *Ce*. The choice of the parameters is that given in table 1 except for V_0 (at 300 K for example, the value used is of 35 Å^3). At 300 K, we find a first-order transition at a critical pressure of 11 kbar, close to the experimental value of 8 kbar (Shapiro *et al* 1977). The critical point is found to occur at 700 K and 23 kbar. The resulting isotherms and phase diagram of cerium are reported in figure 2. We should mention that the same type of explanation has been proposed recently by Martin and Allen (1982) for cerium.

5. Introduction of a chemical pressure

The correspondence between chemical and external pressure is drawn following Cyrot (1973).

Using equation (3.10), we have in the elastic limit:

$$\frac{d(|J|/D)}{|J|/D} = \frac{q}{B}\, dP. \tag{5.1}$$

On the other hand, the variation of $|J|/D$ when introducing a concentration c of atoms (Th,

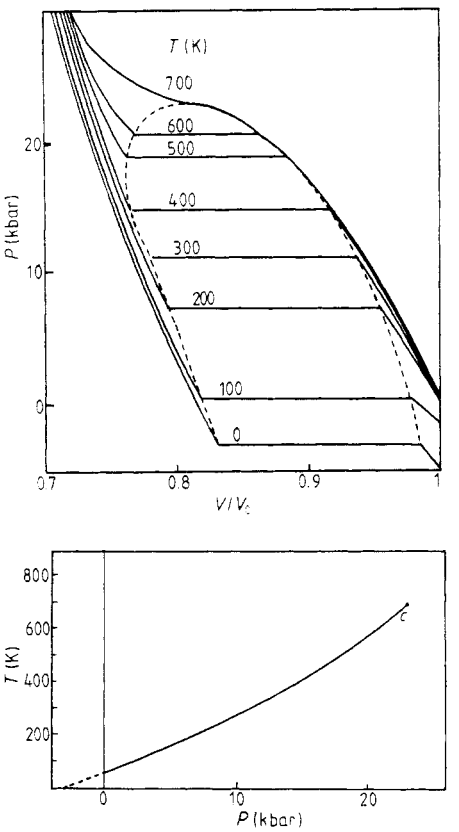

Figure 2. Isotherms and phase diagram of cerium.

Sc . . .) instead of cerium, is assumed to follow the empirical law:

$$\frac{\mathrm{d}(|J|/D)}{|J|/D} = \mathrm{d}c. \tag{5.2}$$

Thus, we have the equivalence between pressure and concentration:

$$c = \int^{\cdot} \frac{q}{B}\,\mathrm{d}P. \tag{5.3}$$

In the case where the bulk modulus in the two extreme limits $c=0$ and $c=1$ is quite different, the problem is solved self-consistently in B. At each value of V, we start with a given B and obtain the corresponding pressure (cf § 3), i.e. concentration (cf equation (5.3)). The related bulk modulus is then deduced using a linear law:

$$B' = (1-c)B_{c=0} + cB_{c=1}. \tag{5.4}$$

The resolution is made in such a way that B' coincides with B.

(i) $Ce_{1-c}Th_c$. The introduction of thorium slightly affects the bandwidth and volume of cerium, but radically changes its bulk modulus: $B_{c=1} = 580$ kbar compared with $B_{c=0} = 240$ kbar. Applications of equations (5.3) and (5.4) and the corresponding self-consistent procedure lead to a critical point at $T = 150$ K and $c = 0.33$. The agreement with the experimental results (Lawrence *et al* 1975: $T = 147$ K and $c = 0.27$) is surprisingly

good, considering that the chemical pressure is treated in a rather crude model. The dramatic change in the critical point when going from pure Ce to CeTh (147 K instead of 700 K) originates from the huge value of B in thorium.

6. Conclusion

In conclusion, the model of a compressible Kondo lattice provides a possible explanation of the anomalous phase diagram of cerium and its compounds. It appears that the Kondo contribution is sufficient to create a thermodynamically unstable region leading to a first-order transition at low temperature. The disappearance of magnetism in the α phase is then explained by an extremely high Kondo temperature. The advantage of the model is that it is consistent with photoemission experiments and leads to reasonable results. However, at the present time, we cannot definitely conclude the nature of the α phase from the experimental results: whether it be a different valence phase or a Kondo phase with high T_K. Further experiments are still needed to provide a better understanding of the already existing results.

Acknowledgments

The authors are grateful to M Croft for useful comments on this work.

Appendix

At $T=0$, the self-consistent conditions on E_0 and E_F are given by:

$$n_\sigma = \frac{1}{2D}(E_F - \omega_1^\sigma)$$
$$m_\sigma = \frac{J^2x^2}{8D}\left(\frac{1}{\omega_1^\sigma - E_0 - \frac{1}{2}Jn_{-\sigma}} - \frac{1}{E_F - E_0 - \frac{1}{2}Jn_{-\sigma}}\right). \tag{A.1}$$

Putting

$$E_F - E_0 - \tfrac{1}{2}Jn_{-\sigma} = t_\sigma$$
$$\omega_1^\sigma - E_0 - \tfrac{1}{2}Jn_{-\sigma} = \tfrac{1}{2}[-z_\sigma - (z_\sigma^2 + J^2x^2)^{1/2}] = \tfrac{1}{2}u_\sigma \tag{A.2}$$

with $z_\sigma = D + E_0 + \frac{1}{2}J(n_{-\sigma} - m_{-\sigma})$, we find

$$E_0 = (-1 + n_\sigma + m_\sigma)D - \frac{1-(n_\sigma/m_\sigma)}{2n_\sigma/m_\sigma}[4n_\sigma^2D^2 + (n_\sigma/m_\sigma)J^2x^2]^{1/2} + \tfrac{1}{2}J(m_{-\sigma} - n_{-\sigma})$$
$$E_F - E_0 = \tfrac{1}{2}Jn_{-\sigma} + n_\sigma D - \tfrac{1}{2}[4n_\sigma^2D^2 + (n_\sigma/m_\sigma)J^2x^2]^{1/2} \tag{A.3}$$

which is a generalisation of the results of Lacroix and Cyrot (1979) to the magnetic case. If we say that equation (A.3) is verified as well by up and down spins, the expressions of the magnetic moments μ_c and μ_d can be deduced in the limit $J^2x^2/4n_\sigma m_\sigma D^2 \ll 1$ (which is in

fact always satisfied):

$$\mu_d = (1-4x^2)^{1/2}$$
$$\mu_c = \frac{|J|}{4D}(1-4x^2)^{1/2}. \tag{A.4}$$

The disappearance of magnetism occurs when $x = \frac{1}{2}$. The correspondence with the critical value of $|J|/D$ is given in § 3.

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