Tricritical Points in Compressible Magnetic Systems

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The compressible Ising-like model of Larkin and Pikin is equivalent to a constrained Ising model, as was recently shown by Sak. I introduce finite pressure effects into this system and find that the Ising transition is first order at low pressures and second order "renormalized" at high pressures. The tricritical pressure is equal to the rigidity modulus for the model, but will be lower for more realistic systems. The spherical tricritical point and Gaussian tetracritical point are also observable in principle as the pressure is varied.

The modifications of a regular second-order magnetic phase transition due to the coupling of the magnetic system to the elastic degrees of freedom are not very well understood at the present time. Many conflicting results exist regarding this question. The first discussions of these effects^{1•3} predicted that close enough to the transition temperature T_c , the transition will become first order if the specific heat of the incompressible system diverges at T_c (as is the case for the Ising model). Later,^{4,5} arguments have been put forward against this first-order transition on the grounds that, for example, the internal fluctuations should prevent the instability that is connected with the first-order transition. In an exactly solvable compressible Ising model, due to Baker and Essam,⁶ and in a related class of constrained systems,⁵ one has the exact results^{7,8} that the transition is first order at negative pressures (see, however, Ref. 8, and Bergman, Imry, and Gunther⁹) and second order for positive pressures. These models are unphysical because they have a vanishing rigidity. On the other hand, for models that are infinitely rigid, "magnetothermomechanics"³ or the Domb-Rice ideas hold leading to a first-order transition for all pressures. It was stated in Ref. 7 that a simple continuity argument would suggest that a first-order transition at low pressures becoming second order at a tricritical pressure P_t would be consistent with all these results, provided that $P_t = 0$ for zero rigidity and $P_t \rightarrow \infty$ when the rigidity modulus μ becomes large. This conjecture was consistent with approximate results of Baker and Essam,¹⁰ but it appears to be at odds with the results of Larkin and Pikin¹¹ who gave an essentially exact treatment of a more realistic model with a finite arbitrary value of μ and found that the transition was first order for any value of P. More recently, Wegner¹² and Hörner¹³ have confirmed the Larkin-Pikin result, giving a special emphasis to the surface effects. Sak,¹⁴ using the fact that the Larkin-Pikin model is equivalent to a constrained Ising model (this model was treated by Rudnick, Bergman, and Imry¹⁵), also obtains the result that for finite rigidity the transition is always first order.

It is the main purpose of this Letter to attempt to settle this theoretical question by showing that taking into account finite deformation effects in the Larkin-Pikin model leads to the existence of a finite P_t equal to the rigidity modulus μ . The order of the transition depends on $P - \mu$ via the pairing term¹⁵ in the Hamiltonian [see Eqs. (9)and (10)]. This exact result is in line with the conjecture of Ref. 7 and thus agrees with both the Baker-Essam model $(\mu \rightarrow 0)$ and the Domb-Rice result (valid for $\mu \rightarrow \infty$). The tricritical point at P_t is one which occurs in a constrained system. The general theory^{5,8,9} of the critical and tricritical phenomena in such systems is thus applicable here. Our treatment is also relevant to the question of the dependence of the results on the boundary conditions at the lattice surfaces. In agreement with general statistical mechanical arguments,¹⁶ no such dependence appears to exist. The dependence is only on the macroscopic constraint, P, but not on the surface effects.

The value of P_t obtained is quite high, on the order of 10^5 bar for typical materials. Thus, in order to make contact with existing experimental results,¹⁷ we show that the value of P_t is sensitive to higher-order terms, not included in the model, and to lattice anisotropy. Anharmonic effects may also be important.

We assume the Hamiltonian treated by Larkin and Pikin¹¹ and Sak¹⁴:

$$H = H_m + H_{e1} + H_{int} , (1)$$

where H_m is a usual Ising-like field-theoretic model in a *d*-dimensional space,

$$H_{m} = \int_{\mathbf{V}_{o}} d^{3}x \left[\frac{1}{2} r_{0} \psi^{2} \left(\vec{\mathbf{x}} \right) + \frac{1}{2} (\nabla \psi)^{2} + u_{0} \psi^{4} \right].$$
⁽²⁾

 H_{el} is the elastic part, with $\tilde{u}(\bar{x})$ the deformation vector; for P = 0, H_{el} favors a volume V_0 for the crystal:

$$H_{\rm el} = \int_{\boldsymbol{v}_0} d^3 x \left[\left(\frac{K}{2} - \frac{\mu}{d} \right) (\nabla \cdot \vec{\mathbf{u}})^2 + \mu \sum_{\alpha,\beta} \left(\frac{\partial u_{\alpha}}{\partial x_{\beta}} \right)^2 \right], \tag{3}$$

where K is the bulk modulus and μ the rigidity modulus. H_{int} is the magnetoelastic coupling (assumed to depend only on the longitudinal strains) characterized by a coupling constant g:

$$H_{\text{int}} = g \int_{V_0} d^3 x \, \psi^2(x) \nabla \cdot \vec{u} \,. \tag{4}$$

It is convenient to separate, for a given crystal volume V, the elastic deformation into two parts: a uniform, possibly large, dilation (from the volume V_0) characterized by constant values $u_{\alpha\alpha}$ of $\partial u_{\alpha}/\partial x_{\alpha}$ (uniform shear deformations are easily integrated upon and lead to no relevant effects), and elastic vibrations $\delta \tilde{u}(\bar{x})$ within the volume V. The latter can be expanded in normal modes:

$$5\vec{\mathbf{u}}(\vec{\mathbf{x}}) = \sum_{k \neq 0} \vec{\mathbf{u}}_k e^{i\vec{\mathbf{k}}\cdot\vec{\mathbf{x}}} , \qquad (5)$$

where the allowed values of \vec{k} are determined by the boundary conditions. It is convenient to adopt periodic boundary conditions, but it is *essential*, when V is varied, either to change the boundary conditions accordingly or to keep defining $\delta \vec{u}(\vec{x})$ for x in the volume V_0 , with the same boundary conditions. We shall use the latter method, with periodic boundary conditions in the volume V_0 . As usual in solid state physics, bulk properties will not depend significantly on the details of the boundary conditions. The elimination of the elastic degrees of freedom is now done in two steps.

(a) Integration on \bar{u}_k : The Gaussian integrals on the transverse components are irrelevant for the critical behavior. When integrating over the longitudinal components one obtains, after completing the square, a ψ^4 -like term, which can be written as

$$-g^{2}\left[4\left(\frac{K}{2}+\frac{d-1}{d}\mu\right)\right]^{-1}\left[\int_{V_{0}}d^{3}x\,\psi(x)^{4}-\frac{1}{V_{0}}\sum_{\vec{p},\vec{q}}\psi_{\vec{p}}\psi_{-\vec{p}}\psi_{\vec{q}}\psi_{-\vec{q}}\right],\tag{6}$$

where the second term had been added and subtracted. The first term is a modification to u,

$$u_{\rm eff} = u_0 - g^2 \left[4 \left(\frac{K}{2} + \frac{d-1}{d} \mu \right) \right]^{-1} , \qquad (7)$$

which, as noted by Aharony,¹⁸ will lead to a firstorder transition, and possibly to a tricritical point, if g is large enough to make $u_{eff} \le 0$. Let us assume that g is small enough such that $u_{eff} > 0$. Were an experiment done at a constant volume, $u_{\alpha\alpha}$ would be constants and the effective Hamiltonian would be (1) + (6) [with u_{eff} from (7)]. This is the same as the constrained Ising model^{14,15} and will lead, because the coefficient of the quartic term u_{eff} is positive, to a "renormalized Ising" critical behavior.⁵ The same kind of behavior was also obtained for the Baker-Essam model at constant volume.^{6,7} (b) If one is interested in a constant pressure, the components $u_{\alpha\alpha}$ have also to be integrated upon, in the following way: A term +PV is added to the Hamiltonian; we express V in terms of the $u_{\alpha\alpha}$,

$$V = V_0 \left[1 + \sum_{\alpha} u_{\alpha\alpha} + \sum_{\alpha \neq \beta} u_{\alpha\alpha} u_{\beta\beta} + O(u^3) \right], \qquad (8)$$

and integrate over $u_{\alpha\alpha}$. The $O(u^2)$ terms are important for finite deformations that may be due to high pressures or strong magnetoelastic coupling. They were neglected by Larkin and Pikin,¹¹ Wegner,¹² and Sak,¹⁴ whose treatments thus apply only for low pressures. The $u_{\alpha\alpha}$ integrals lead to a new pairing term in the effective magnetic Hamiltonian [it is important to include the term $\frac{1}{2}K(\sum_{\alpha\alpha})^2$ in the energy of the uniformly strained crystal] which, when added to the one in (6),

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gives

$$H_{p} = \frac{\theta}{V_{0}} \sum_{\vec{p}, \vec{q}} \psi_{\vec{p}} \psi_{-\vec{p}} \psi_{\vec{q}} \psi_{-\vec{q}} , \qquad (9)$$

with

$$\theta = \frac{g^2}{4} \left[\left(\frac{K}{2} + \frac{d-1}{d} \mu \right)^{-1} - \left(\frac{K}{2} + \frac{d-1}{d} p \right)^{-1} \right]$$

The total effective magnetic Hamiltonian is

$$H_{m, eff} = \int_{V_0} d^3x \left[r \psi^2(x) + \frac{1}{2} (\nabla \psi)^2 + u \psi^4 \right] + H_{\flat} , \quad (10)$$

where $u = u_{eff} [Eq. (7)]$ and $r = r_0 - gP/2 \{\frac{1}{2}K + [(d-1)/d]p\}$.

 $H_{m, eff}$ leads to a first- or second-order transition according to whether $\theta < 0$ or $\theta > 0$, respectively; thus the tricritical pressure is $P_t = \mu$, which is an exact result for this model. This tricritical point exists for any value of g. The critical and tricritical behaviors around this tricritical point are those characteristic of constrained systems.⁸ In the renormalization-group language, the tricritical point corresponds to the Ising fixed point.¹⁵ The tricritical behavior is Isinglike and the critical behavior is "renormalized-Ising-like" (as for the case of constant volume discussed above). In addition, for $P > P_t$, if g changes as a function of P or other thermodynamic variables such as to drive u_{eff} negative [Eq. (7)], another tricritical point is possible. This corresponds to the "spherical" fixed point.¹⁵ It is very interesting that the observation of "spherical" critical behavior is thus possible in principle. Finally, the Gaussian fixed point, at which both u_{eff} and θ vanish, is a fourth-order critical point where two tricritical lines ($\theta = 0, u_{eff} > 0$ and $u_{eff} = 0, \theta > 0$) meet.¹⁹ This is the point found by Aharony.¹⁸ We note that the change in u_{eff} [Eq. (7)] does not appear in the Baker-Essam case, which is not a field-theory model.

It is important to emphasize that the rather high value of $P_t = \mu$ is an exact result only for the model characterized by Eqs. (1)-(4). If higherorder terms are included, new terms will appear in the effective Hamiltonian. An important one is, e.g., the sixth-order pairing term

$$\zeta \sum_{\vec{\mathfrak{q}}_1 \vec{\mathfrak{q}}_2 \vec{\mathfrak{q}}_3} \psi_{\vec{\mathfrak{q}}_1} \psi_{-\vec{\mathfrak{q}}_1} \psi_{\vec{\mathfrak{q}}_2} \psi_{-\vec{\mathfrak{q}}_2} \psi_{\vec{\mathfrak{q}}_3} \psi_{-\vec{\mathfrak{q}}_3};$$

although "irrelevant" (in the renormalization group sense), it will change the value of the tricritical θ , and will make it negative. This is because this term will feed positive contributions into the renormalization group recursion relations for θ . Also, higher-order magnetoelastic terms and lattice anisotropy have been shown^{8,9} to shift significantly the tricritical pressure. Anharmonic terms may also have an important effect.

Thus, unfortunately, we are not able to compute P_t and compare with experiment. But we know that P_t is finite and that the order of the transition depends upon the pressure. The tricritical point in NH₄Cl,¹⁷ with $P_t \cong 1.5$ kbar, may well be of this kind, although other interpretations are possible.^{19, 20} Another important guestion is what is the "size" of the first-order transition. For the Baker-Essam and related models it is given⁷ by $A^{1/\alpha}$ where α is the specific-heat index of the Ising model and A a parameter proportional to g^2 and to $|P - P_t|^{21}$ Because of the probable large values of $1/\alpha$ (the best current estimate is $1/\alpha \cong 8$), $A^{1/\alpha}$ is a sensitive function of A, and the first-order transition may be impossible to observe for $A \sim 0.1$. However, once $A \sim 1$ (as may well be the case for NH_4Cl), thr firstorder transition and the region where renormalization is important for the second-order transition increase sharply with $|P - P_t|$ and become easily observable. We have not discussed here the case in which $n \ge 2$, where the situation is different¹⁴ because probably $\alpha < 0$. The relevance of our results there is that the value of θ can be manipulated by varying the pressure, which may enable one to get, e.g., a renormalized Heisenberg critical behavior for an appropriate value of P.

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¹O. K. Rice, J. Chem. Phys. <u>22</u>, 1535 (1954). ²C. Domb, J. Chem. Phys. <u>24</u>, 783 (1956).

³D. C. Mattis and T. D. Schultz, Phys. Rev. <u>129</u>, 175

(1963).

⁴H. Wagner and J. Swift, Z. Phys. <u>239</u>, 182 (1970).

⁵M. E. Fisher, Phys. Rev. <u>176</u>, 257 (1968).

⁶G. A. Baker, Jr., and J. W. Essam, Phys. Rev. Lett. <u>24</u>, 447 (1970).

 $\overline{}^{T_a}$ L. Gunther, D. J. Bergman, and Y. Imry, Phys. Rev. Lett. <u>27</u>, 558 (1971),

^{7b}D. J. Bergman, Y. Imry, and L. Gunther, J. Stat. Phys. <u>7</u> 337 (1973).

⁸Y. Imry, O. Entin-Wohlman, and D. J. Bergman, J. Phys. C Proc. Phys. Soc., London 6, 2846 (1973).

- ⁹D. J. Bergman, O. Entin-Wohlman, and Y. Imry, J. Phys. C: Proc. Phys. Soc., London 7, 1621 (1973). ¹⁰G. A. Baker, Jr., and J. W. Essam, J. Chem. Phys.
- 55, 861 (1971). ¹¹A. I. Larkin and S. A. Pikin, Zh. Eksp. Teor. Fiz.

56, 1664 (1969) [Sov. Phys. JETP 29, 891 (1969)]. ¹²F.J. Wegner, J. Phys. C: Proc. Phys. Soc., London 7, 2109 (1974).

¹³H. Hörner, private communication.

¹⁴J. Sak, to be published.

¹⁵J. Rudnick, D. J. Bergman, and Y. Imry, Phys. Lett. 46A, 449 (1974).

¹⁶D. Shalitin and Y. Imry, Phys. Rev. B (to be pub-

lished); M. Luban and H. Novogrodsky, Phys. Rev. B 6, 1130 (1972).

¹⁷See. C. W. Garland and R. J. Pollina, J. Chem. Phys. 58, 5002 (1973), for previous references on NH_4Cl .

- ¹⁸A. Aharony, Phys. Rev. B 8, 4314 (1973).
- $^{19}\mathrm{T.}$ S. Chang and H. E. Stanley, to be published.
- ²⁰A. Huller, Z. Phys. <u>254</u>, 456 (1972).

 21 From Eq. (80) of Ref. 7b one finds, for the model of Ref. 7a, that up to a numerical factor of order unity, $A = \gamma_m^2 (T_c \theta_L / \theta_D^2) P / K$, where γ_m is the magnetic Grüneisen constant $\gamma_m = \partial \ln T_c / \partial \ln V$, θ_D is of the order of the Debye temperature, and θ_L is a characteristic lattice temperature, $k_{\rm B}\theta_L = \hbar^2/Ma^2$, where M is the mass of the atom and a is the lattice constant. For a model where $P_t > 0$, P should be replaced by $|P - P_t|$.

Test of Parity Conservation in p-p Scattering*

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We report a result of $(1 \pm 4) \times 10^{-7}$ for the parity-nonconserving component in the *p*-*p* nuclear cross section at 15 MeV. Our experiment uses rapid spin reversal of a longitudinally polarized proton beam and an unpolarized H₂ target. Sources of systematic error are discussed and found to be $<10^{-7}$.

The presence of weak interactions between hadrons, which is implied by the current-current form of weak interaction theory,¹ has been established by observations of parity mixing in many nuclei.² However, no quantitative agreement exists between theory and experiment, either for heavy nuclei, where nuclear-structure effects complicate calculations, or for the lightest system with a reported effect, $np \rightarrow d\gamma$.^{3,4} This lack of agreement emphasizes the necessity for studying the nucleon-nucleon system through p - p scattering is estimated to be a few parts in 10^7 , the actual effect, which is sensitive to the details of the interaction including the possible presence of neutral currents, could be considerably different. The smallest previous experimental upper limit on the magnitude of the parity admixture in p-pscattering is 5×10⁻³ at 210 MeV.⁷ Order-ofmagnitude improvements are needed to test the various models of the weak interaction.

Our experiment is sensitive to the pseudoscalar term $\hat{\sigma} \cdot \hat{\rho}$ in the total nuclear cross section arising from the interference between the parityconserving and parity-nonconserving parts of the scattering amplitude. ($\hat{\sigma}$ is the spin and \hat{p} the momentum of the incident proton.) This interference is observed by scattering a longitudinally polarized proton beam on an unpolarized hydrogen target and detecting the change in the total nuclear cross section when the polarization is reversed.

A 200-nA beam of longitudinally polarized protons from a Lamb-shift ion source⁸ is accelerated to 15 MeV at the Los Alamos tandem Van de Graaff accelerator and strikes an H₂ gas target (Fig. 1). A two-stage fast-steering system with feedback stabilizes the beam position and angle at our detector, reducing movement of the beam by a factor of ~ 50 .

A measure of the total cross section is obtained by detecting the scattered beam over a solid angle of nearly 4π . To realize adequate statistical accuracy in a reasonable time, the scattered beam current (~ 3×10^9 protons/sec) is measured