responding to vortex spacings of 6.8 to 3.4  $\mu$ m) is consistent with the above peak structure.

Thus on the present sample both the microscopic measurements with the atomic beam and the macroscopic measurements with the voltage probes indicate that the expression  $\rho_f = \rho_n (B/H_{cs})$ is not valid. However, the more fundamental relation  $\vec{E} = \vec{v} \times \vec{B}/c$  which is not measured by the macroscopic measurements is consistent with the microscopic data.

Future measurements in which both the voltage drop across the sample and the internal magnetic field are measured in situ should allow us to investigate further some of the questions which the present experiments raise but do not answer. These include the washing out of the peaks in the relative transition probability when a current is flowing, the independence of the resistivity to magnetic field, the lack of signals at higher fields, and the occasional nonconstant velocity shift when a current is flowing. This latter should be particularly interesting since the probable explanation is a large Hall voltage which causes the lattice to flow at an angle to the atomic beam. In addition, we plan to observe samples for which the expression  $\rho_f = \rho_n (B/H_{cz})$  is valid. We hope these experiments will allow a microscopic check of the present theories of flux flow.

In conclusion, we feel that the first application of this method demonstrates some of its inherent possibilities. For example, merely by changing our beam from potassium to cesium we should be able to observe periodicities down to 100 Å. Further applications might include the study of how vortices are created and destroyed at the edges of a superconductor, possible collective motions of the vortex lattice, fluctuations near the Curie point in a ferromagnet, domain boundary motion in ferromagnets, etc. A further possibility would be to observe electric fields in much the same manner using a beam of molecules which are sensitive to oscillating electric fields.

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## Critical Concentration Versus Interaction Range for Random Systems

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We consider the problem of interacting particles occupying random sites on either a plane square or a simple cubic lattice. The critical concentration  $p_c$  is defined as that concentration below which the cooperative transition which normally occurs in the system can no longer take place. In this paper we obtain an expression for  $p_c$  as a function of the interaction range r, exact when  $r^d \gg 1$  (d=dimensionality, r measured in units of lattice constant).

The study of the statistical behavior of interacting particles which occupy randomly a fraction p of the sites of an otherwise empty lattice is of interest for two reasons. First, the properties of such systems may shed some light on the nature of cooperative phenomena in fully occupied lattices.<sup>1</sup> Secondly, these random systems are related to problems involving substitutional defects in regular crystal lattices. For example, OH<sup>-</sup> ions may replace halide ions in alka-

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<sup>&</sup>lt;sup>1</sup>For a general reference on atomic beam techniques see N. F. Ramsey, *Molecular Beams* (Oxford U. Press, Oxford, England, 1956).

<sup>&</sup>lt;sup>3</sup>For a general review of research in this area see Y. B. Kim and M. J. Stephen, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 19. See also H. Meissner, J. Low Temp. Phys. <u>2</u>, 267 (1970).

<sup>&</sup>lt;sup>4</sup>Y. B. Kim, C. F. Hemstead, and A. R. Strnad, Rev. Mod. Phys. <u>36</u>, 43 (1964), and Phys. Rev. Lett. <u>12</u>, 145 (1964), and Phys. Rev. <u>139</u>, A1139 (1965), and Phys. Rev. Lett. <u>13</u>, 794 (1964).

li halide crystals,<sup>2</sup> creating random dipolar impurities. Transition-ion impurities in noble metals comprise a similar magnetic problem.<sup>3</sup>

Because of the increased mathematical difficulties involved in the treatment of random systems, results in this area have been limited. Rigorous theorems concerning the properties of the various thermodynamic functions for random systems have been proved,<sup>4</sup> and very dilute systems have been studied by diagrammatic expansions.<sup>5</sup> Also, Griffiths<sup>6</sup> has shown that in a random Ising system, the magnetization ceases to be analytic at a temperature greater than the associated critical temperature.

Another aspect of the problem involves the determination of the critical concentration  $p_c$ , the concentration below which the cooperative transition which normally occurs in the system cannot take place. Numerical estimates of  $p_c$  have been obtained for Ising models on several types of lattices,<sup>7</sup> and certain inequalities for  $p_c$  have been derived.<sup>8</sup> Exact values of  $p_c$  have been obtained for two-dimensional triangular and honeycomb lattices,<sup>9</sup> and for several pseudolattices.<sup>10</sup>

In this note, we wish to report a rigorous evaluation of  $p_c$  as a function of the interaction range r valid for  $r^d \gg 1$  (d = dimensionality, r measured in units of lattice constant). The approach we utilize involves an evaluation of the radius of convergence of the zero-temperature susceptibility series in the concentration p, derived for Ising and Heisenberg ferromagnets by Rushbrooke and Morgan.<sup>11</sup> This radius of convergence is just  $p_c$ since at  $p = p_c$  the phase transition, with associated divergent susceptibility, occurs at T = 0.

For simplicity, we consider a system of Ising spins interacting via the ferromagnetic Hamiltonian $^{12}$ 

$$H = -\sum_{i < j} V_{ij}(r) \mu_i \mu_j + H_0 \sum_i \mu_i, \qquad (1)$$

where, as usual,  $\mu_i = \pm 1$  and r is the range of the interaction. That is, the interaction between two spins is identically zero if they are more than r lattice constants apart. Very close to T=0, the magnetic susceptibility in zero field may be written as a sum over clusters<sup>13</sup> of spins in the form<sup>11</sup>

$$\chi = \beta \sum_{n} n^2 \sum_{t_n} N_{n,t_n}, \qquad (2)$$

where the sum includes all possible distinct<sup>14</sup> n-particle clusters which can be found on the lattice,  $N_{n,t_n}$  is the number of clusters of type  $t_n$ , and  $\beta$  is the reciprocal of Boltzmann's constant times the temperature. The  $n^2$  term is the susceptibility of an *n*-particle cluster at T = 0, with the magnetic moments implicitly included. If the sites are randomly occupied, then there is a probability p that any given site contains a spin, and a probability 1-p that a site is vacant. Therefore,  $N_{n,t_n}$  is given by the expression

$$N_{n,t_n} = N p^n (1-p)^{\nu_n}, \tag{3}$$

where  $\nu_n$  is the perimeter of the cluster, i.e., the number of vacant sites within a distance r of at least one of the occupied sites. It follows that two clusters are distinct if and only if their perimeters are different, and the sum over  $t_n$  in Eq. (2) can be written as a sum over  $\nu_n$  in the form

$$N^{-1}\chi(0) = \beta \sum_{n} n^{2} p^{n} \sum_{\nu_{n}} (1-p)^{\nu_{n}}.$$
 (4)

We shall determine the radius of convergence of Eq. (4) by finding radii of convergence  $R_c^{\ u}$  and  $R_c^{\ l}$  for power series in p which bound Eq. (4) below and above, respectively. Then it will be seen that, when  $r^d \gg 1$ ,  $R_c^{\ l} = R_c^{\ u}$ . The calculations will be performed for simple cubic (SC) and plane square (PS) lattices only, but the same methods may be applied to more general lattices. To this end we note that for sufficiently small p, Eq. (4) is bounded by

$$\sum_{n} n^{2} p^{n} (1-p)^{\nu} {n^{(\max)}}$$

$$\leq \sum_{n} n^{2} p^{n} \sum_{\nu_{n}} (1-p)^{\nu} {n} \leq \sum_{n} n^{2} p^{n} M(n, r), \qquad (5)$$

where M(n, r) is the number of distinct *n*-particle clusters, and  $\nu_n(\max)$  is the maximum perimeter size. As *p* increases, as long as all the series converge, the inequalities remain valid. Thus the first series to diverge must be the series on the right, and its radius of convergence is a lower bound for  $p_c$ . Similarly, the radius of convergence of the series on the left is an upper bound for  $p_c$ .

We now examine the series in Eq. (5) to find these bounds. In order to obtain  $\nu_n(\max)$  and M(n, r), we first determine, for given interaction range r, the maximum number of sites which are available to an *n*-particle cluster,  $\eta(r)$ . By this quantity we mean the number of sites, occupied and unoccupied, in the cluster in which each particle pair is the maximum distance r apart. If ris measured in units of the lattice constant, then  $\eta(r)$  for the PS and SC lattices is given by ntimes the volume of a d-dimensional hypersphere minus the overlap volume. This is illustrated



FIG. 1. The maximum number of sites for a cluster for d=2. Occupied sites are shown by the large dots. Lined areas are the overlapping parts, which are subtracted from  $n\pi r^2$  to give  $\alpha_2(r)$ .

for d = 2 in Fig. 1. From elementary calculus we have then

$$\eta(r) = n\alpha_d(r),\tag{6}$$

where<sup>15</sup>

$$\begin{aligned} \alpha_2(r) &= r^2 \left(\frac{2}{3}\pi + \frac{1}{2}\sqrt{3}\right), \\ \alpha_3(r) &= r^3 \left(\frac{41}{24}\pi - \frac{3}{2}\sqrt{3}\right). \end{aligned} \tag{7}$$

From our earlier definition of perimeter,  $\nu_n(\max)$  is just the perimeter of this maximum cluster, namely

$$\nu_n(\max) = n\alpha_d(r) - n. \tag{8}$$

Furthermore, an upper bound for M(n, r) is given by the number of ways one can place n objects on the  $\eta(r)$  available sites,

$$M(n, r) \leq \binom{n\alpha_d(r)}{n}, \tag{9}$$

where

$$\binom{j}{k} \equiv \frac{j!}{k!(j-k)!}.$$

Substituting these results into Eq. (5) we obtain

$$\sum_{n} n^{2} p^{n} (1-p)^{n \left[\alpha_{d}(r)-1\right]}$$

$$\leq \sum_{n} n^{2} p^{n} \sum_{\nu_{n}} (1-p)^{\nu_{n}} \leq \sum_{n} n^{2} p^{n} \binom{n \alpha_{d}(r)}{n}. \quad (10)$$

To evaluate the radii of convergence of the series above we use the Cauchy test<sup>16</sup> for convergence of a power series,

$$R_c = \limsup_{n \to \infty} |a_n|^{-1/n}, \tag{11}$$

where  $a_n$  is the coefficient of  $x^n$  and  $R_c$  is the radius of convergence. Applying this test to the series on the right in Eq. (10), we obtain

$$R_c^{\ l} = \lim_{n \to \infty} \left[ n^2 \binom{n \alpha_d(r)}{n} \right]^{-1/n} = \frac{1}{\alpha_d(r)}, \quad (12)$$

where Stirling's formula has been used to evaluate the combinatorial term. We now consider the series on the left in Eq. (10), which we rewrite by expanding the  $(1-p)^{\nu_n (\max)}$  term as

$$\sum_{m} a_{m} p^{m}$$

$$= \sum_{m} p^{m} \left[ \sum_{k=0}^{m} (-1)^{k} (m-k)^{2} \sum_{j=0}^{k} \binom{\nu_{j}(\max)}{k} \right]. \quad (13)$$

The radius of convergence of this series is found by factoring the maximum combinatorial term out of  $a_m$ . The remaining terms may increase at most like some power of m. But since

$$\lim_{m\to\infty}(m^k)^{-1/m}=1,$$

the limit in Eq. (13) will be determined by the limit of the maximum combinatorial term as  $m \rightarrow \infty$ . From Eq. (13) this term is

$$\binom{\nu_m(\max)}{m}$$
,

and therefore the radius of convergence is given by

$$R_c^{\ u} = \frac{1}{\alpha_d(r) - 1} \approx \frac{1}{\alpha_d(r)} \quad (r^d \gg 1). \tag{14}$$

Equations (12) and (14) allow us to conclude that for  $r^d \gg 1$ ,

$$p_c = 1/\alpha_d(r). \tag{15}$$

We point out here that the above result is purely a property of the underlying lattice structure, and in particular does not depend on the exact nature of the cluster susceptibility functions, so long as these quantities do not increase as  $n^n$ . Thus Eq. (15) should apply to paraelectric defects in alkali-halide crystals and other such systems. In these materials the pure dipole-dipole interaction is shielded by correlations between defect and host. Our results, coupled with experimental determination of  $p_c$ , would yield an estimate of this shielding.

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<sup>&</sup>lt;sup>1</sup>For a review, see M. E. Fisher, Rep. Progr. Phys. <u>30</u>, 615 (1967).

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<sup>6</sup>R. B. Griffiths, Phys. Rev. Lett. 23, 17 (1969).

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<sup>10</sup>M. E. Fisher and J. W. Essam, J. Math. Phys. 2,

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<sup>11</sup>G. S. Rushbrooke and D. J. Morgan, Mol. Phys. <u>4</u>, 1 (1961).

<sup>12</sup>It will become apparent later that the results obtained are valid for any interacting system which can undergo a phase transition to an ordered phase.

<sup>13</sup>The term "cluster" denotes a set of particles on the lattice which can be linked together by bonds of length  $\leq r$ .

 $^{14}$ A definition of distinct clusters is given later in the paper.

<sup>15</sup>The integral associated with  $\alpha_3(r)$  is evaluated only approximately.

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## Field-Dependent Central-Cell Corrections in GaAs by Laser Spectroscopy\*

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We report the first definite observations of the magnetic field dependence of centralcell corrections in shallow donors. Exploiting the extreme resolution inherent in magnetospectroscopy employing infrared gas laser sources, we have observed previously unresolved transitions arising from four distinct "hydrogenic" donor species occurring in high-purity epitaxial GaAs. We show that the species-dependent differences in transition energies are directly proportional to the probability of finding an electron at the donor site to which it is loosely bound. This probability increases with increasing magnetic field because of magnetic squeezing of the wave function of the bound electron.

Recent magneto-optical studies<sup>1,2</sup> of the 1s-2pdonor<sup>3</sup> transitions in epitaxial n-GaAs demonstrate that the donor states involved are hydrogenic to within experimental accuracy. We have re-examined some of these transitions both in photoconductivity and transmission in experiments of much higher resolution and with very precise magnetic-field measuring capability in order to check critically the limits of validity of the effective-mass-hydrogenic-atom model for GaAs donors. We find that under high resolution the absorption and photoconductivty previously ascribed to a single donor is actually made up of four, closely spaced, previously unresolved transitions, each arising from a different donor species. The existence of these hyperfine separations (hfs) (of the order of  $0.5 \text{ cm}^{-1}$ ) in the 1s-2p transition cannot be understood from the hydrogenic isolated-donor model even when corrections for band nonparabolicity are introduced.

Thus splittings offer a unique opportunity for investigating perturbations on the simple hydrogenic-donor impurity model.

The most direct evidence for our claim that the hfs arises from small variations in the 1s-2ptransition energies for different donor species<sup>4</sup> present in our samples comes from the observation that the hyperfine peaks change in relative intensity but not in energy from sample to sample. Study of the hyperfine separations in different optical transitions shows that these spacings are attributable to differences among donor ground-state energies. Analysis of the field dependence of the hyperfine separations indicates that the perturbations giving rise to the observed relative shifts in the ground-state energy are (1) of short range compared to the hydrogenic Bohr radius  $a_0$  and are (2) centered either at or very near to the donor site. The observed behavior with magnetic field of the hyperfine sepa-