<sup>4</sup>A. J. Epstein, S. Etemad, A. F. Garito, and A. J. Heeger, Phys. Rev. B 5, 952 (1972).

<sup>5</sup>E. Ehrenfreund, E. F. Rybaczewski, A. F. Garito, and A. J. Heeger, Phys. Rev. Lett. **23**, 873 (1972).

<sup>6</sup>E. Ehrenfreund, S. Etemad, L. B. Coleman, E. F. Rybaczewski,

A. F. Garito, and A. J. Heeger, Phys. Rev. Lett. 29, 269 (1972).

<sup>7</sup>A. A. Ovchinnikov, Zh. Eksp. Teor. Fiz. **57**, 2137 (1969) [Sov. Phys.-JETP **30**, 1160 (1970)].

<sup>8</sup>M. Takahasi, Prog. Theor. Phys. **43**, 1619 (1970); Prog. Theor. Phys. **42**, 1098 (1969); Prog. Theor. Phys. **43**, 860 (1970).

- <sup>9</sup>E. F. Rybaczewski, E. Ehrenfreund, A. F. Garito, A. J. Heeger, and P. Pincus, Phys. Rev. B (to be published).
- <sup>10</sup>For example, J. Kimball, Bull. Am. Phys. Soc. 17, 320 (1972).
   <sup>11</sup>A. N. Bloch, R. B. Weisman, and C. M. Varma, Phys. Rev.

Lett. 28, 753 (1972).

<sup>12</sup>I. F. Shchegolev, L. I. Burovov, A. V. Zvarykina, and R. B. Lyubovskii, Zh. Eksp. Teor. Fiz. Pis'ma Red. 8, 353 (1968)

[JETP Lett. 8, 218 (1968)].

- <sup>13</sup>N. F. Mott, Philos. Mag. 19, 835 (1969).
- <sup>14</sup>V. Ambegaokar, B. I. Halperin, and S. J. Langer, Phys. Rev. B 4, 2612 (1971).

<sup>15</sup>E. F. Rybaczewski, E. Ehrenfreund, A. F. Garito, and A. J. Heeger, Bull. Am. Phys. Soc. 17, 332 (1972).

<sup>16</sup>P. S. Acker, R. J. Herder, W. R. Hertler, W. Mahler, L. R. Melby, R. E. Benson, and W. E. Mochel, J. Am. Chem. Soc. **82**, 6408 (1960).

<sup>17</sup>L. R. Melby, R. J. Harder, W. R. Hertler, W. Mahler, R. E. Benson, and W. E. Mochel, J. Am. Chem. Soc. **84**, 3374 (1962).

<sup>18</sup>M. Plischke, Phys. Rev. Lett. 28, 361 (1972).

<sup>19</sup>R. Ramirez, L. M. Falicov, and J. C. Kimball, Phys. Rev. B 2, 3383 (1970).

 $^{20}T.$  M. Rice and W. F. Brinkman, Phys. Rev. B (to be published).

<sup>21</sup>M. C. Gutzwiller, Phys. Rev. 137, A1726 (1965).

<sup>22</sup>D. Kuse and H. R. Zeller, Phys. Rev. Lett. 27, 1060 (1971).

PHYSICAL REVIEW B

## VOLUME 7, NUMBER 5

1 MARCH 1973

# Superconductivity, Ferroelectricity, and the Mott Insulator

Robert A. Bari

Brookhaven National Laboratory, Upton, New York 11973

(Received 11 August 1972)

It is shown that Mott-insulator (or semiconductor) state can become unstable to the formation of Cooper pairs. The transition between the states can be either first or second order. The ferroelectric instability of the Mott insulator is also discussed and a spin-analog model is presented that can account for all three thermodynamic states.

#### I. INTRODUCTION

The strong correlation effects that appear to be present among electrons in partially filled orbitals in certain organic substances [e.g., those containing tetracyanoquinodimethan (TCNQ)] suggest a description in terms of a Hubbard model.<sup>1,2</sup> An advantage of this description is that it incorporates the Mott insulator<sup>3</sup> as a possible ground state for certain organic semiconductors. The simple model does not allow for electron-phonon or electronexciton interactions which may be important in describing the equilibrium and transport properties of a particular material. A novel feature of the organic systems is that thermal promotion of polar states may occur at reasonable temperatures,<sup>2</sup> quite unlike the situation in the transition-metal and rare-earth compounds.

Given that the Mott insulator represents the normal state of the electronic system (i.e., the Coulomb repulsion is much greater than the bandwidth in the half-filled-band Hubbard model), we investigate the instability of this normal state in the presence of interactions extraneous to the Hubbard model. These include indirect interactions via phonons or excitons, as well as electron-electron interactions not included in the Hubbard model. A consideration of these interactions is important in

attaining an understanding of the complex behavior of these substances, as is indicated by experiment. Further, the theoretical investigation represents the first in which the short-ranged Coulomb interaction is treated exactly in a model superconductor. The principal result is that the Mott insulator (or semiconductor) can become unstable to Cooper-pair<sup>4</sup> formation below a certain transition temperature. It is also found that the phase transition can be either first or second order, depending on the parameters. The possibility of a ferroelectric instability has also been considered, and it has been found that, within the molecular-field scheme, the phase boundary between the ferroelectric and superconductor is independent of temperature. It is also seen that the competing interactions that lead to ferroelectricity, or superconductivity in a strongly correlated electron system, can be represented in terms of a spin-analog model.

### **II. INSTABILITY TO THE SUPERCONDUCTOR**

The Hubbard model<sup>5</sup> can be written

$$H_{1} = I_{1} \sum_{i} C_{i}^{\dagger}, C_{i}, C_{i}^{\dagger}, C_{i}, + \sum_{i,j} t_{ij} C_{i\sigma}^{\dagger} C_{j\sigma}, \qquad (1)$$

where  $C_{i\sigma}$  destroys an electron on site  $R_i$  and spin  $\sigma$  and  $I_1$  and  $t_{ij}$  are the intrasite repulsion and hop-

2128

Solid State Commun. 9, 1803 (1971).

ping strength, respectively. The interaction that leads to the Cooper-pair instability is written

$$H_{2} = - (I_{2}/N) \sum_{i,j} C_{i}^{\dagger}, C_{j}^{\dagger}, C_{j}, C_{j}, .$$
 (2)

Here  $I_2 > 0$  and N is the number of Wannier sites. The philosophy of writing the interaction in this form is much like that of the BCS reduced Hamiltonian.<sup>6</sup> Rather than try to give a complete account of the interactions in the electron-phonon (or electron-exciton) system, we consider only that term responsible for the instability in the normal system.  $H_2$  can be derived from the BCS reduced interaction by transforming the latter to the Wannier representation and observing that, in a very narrow band, many k states are within a Debye energy of the single-electron energy excitations. With the familiar assumption that the matrix element of the interaction is momentum independent, one arrives at  $H_2$ . Another interaction, leading to a ferroelectric instability, will be discussed later.

The thermodynamics of  $H_1 + H_2$  can be obtained easily for  $t_{ij} \equiv 0$ . This is partly due to the fact that  $H_2$  is exactly treated in terms of the molecular field.<sup>7</sup> Consequently, in the Wannier-site notation, the effective Hamiltonian (or trial Hamiltonian for the variational principle) is the sum of single-site Hamiltonians. Since  $H_1$  is already of the singlesite form, one straightforwardly treats the strong Coulomb repulsion exactly in this superconductor.

The effective Hamiltonian for the ith site can be written

$$\Im C_{\text{eff}}^{i} = I_1 C_i^{\dagger}, C_i, C_i^{\dagger}, C_i, - I_2 (\alpha C_i^{\dagger}, C_i^{\dagger}, + \alpha^* C_i, C_i, ),$$

where  $\alpha \equiv \langle C_{i}, C_{i}, \rangle$  is evaluated self-consistently by taking the thermodynamic average (in its definition) with the effective Hamiltonian.  $\alpha$  is seen to be the off-diagonal order parameter related to the Cooper-pair instability.

The effective single-site energy levels (including the chemical potential term, since we work in the grand canonical ensemble) are the eigenvalues of the following matrix:

$$\mathcal{H}_{eff}^{i} - \mu(C_{i}^{\dagger}, C_{i}, + C_{i}^{\dagger}, C_{i})$$

$$= \begin{bmatrix} 0 & 0 & 0 & -I_{2}\alpha^{*} \\ 0 & -\mu & 0 & 0 \\ 0 & 0 & -\mu & 0 \\ -I_{2}\alpha & 0 & 0 & I_{1} - 2\mu \end{bmatrix} . \quad (3)$$

Here the matrix elements are taken in the four states of occupancy of the single site. If the state  $|0\rangle$  denotes an unoccupied *i*th site, then these states are  $|0\rangle$ ,  $C_{i}^{\dagger}|0\rangle$ ,  $C_{i}^{\dagger}|0\rangle$ ,  $C_{i}^{\dagger}C_{i}^{\dagger}|0\rangle$ . Other operators can be cast in this representation as well. For example,

The partition function is obtained as the product of the single-site partition functions and the chemical potential is then fixed to give one electron per site, on the average. The partition function is

$$Z = (2e^{\beta\mu} + e^{-\beta\epsilon_+} + e^{-\beta\epsilon_-})^N ,$$

where

$$\epsilon_{\pm} = \frac{1}{2} \{ I_1 - 2\mu \pm [(I_1 - 2\mu)^2 + 4(I_2)^2 |\alpha|^2]^{1/2} \}$$

From the eigenstates of Eq. (3) and the matrix operators of Eq. (4) it is found that  $\langle n_i, \rangle = \langle n_{i,i} \rangle$ =  $\frac{1}{2}$  for  $\mu = \frac{1}{2}I_1$ . The partition function becomes

$$Z = \{ 2 [ e^{\beta I_1/2} + \cosh(\beta I_2 | \alpha |) ] \}^N.$$

For  $I_2 = 0$  this expression reduces to the partition function<sup>8</sup> of the zero-bandwidth Hubbard model.

The thermodynamic average of  $\alpha$  can be found in the same manner and one finds the condition

$$\frac{\Delta}{I_2} = \frac{\sinh(\frac{1}{2}\beta\Delta)}{e^{\beta I_1/2} + \cosh(\frac{1}{2}\beta\Delta)} , \qquad (5)$$

where  $\Delta \equiv 2I_2 | \alpha|$ . An upper bound to the free energy is obtained from the well-known inequality  $F \leq \langle H - \Re_{\text{eff}} \rangle - kT \ln Z$ . In this case  $H = H_1 + H_2$  (with  $t_{ij} = 0$ ) and  $\Re_{\text{eff}} = \sum_i \Re_{\text{eff}}^i$ . Again the thermodynamic average is with respect to the equilibrium ensemble with  $\Re_{\text{eff}}$ . (If we treated  $\alpha$  as a variational parameter in  $\Re_{\text{eff}}$ , then formal variation of  $\alpha$  leads to the condition  $\alpha = \langle C_i, C_i, \rangle$ , i.e., the definition given above.)

Numerical analysis of Eq. (5) yields the solution  $\Delta = 0$  as well as nonzero solutions for certain values of the parameter. The former solution corresponds to the thermodynamic state of the Mott semiconductor and the latter to the superconducting state. We determine the state of the system as that which minimizes the variational bound on the free energy.

It is indeed found that the Mott semiconductor is unstable to Cooper-pair formation. Figure 1 shows the region of the phase diagram in which a nonzero solution of Eq. (4) minimizes the upper bound to the free energy. For large  $I_1$  the system is a Mott semiconductor at all temperatures. For  $I_1/I_2$  $<\frac{1}{2}$ , there is an instability in the Mott semiconductor to the superconductor. At larger values of

2129



FIG. 1. Phase diagram for the superconductor vs Mott semiconductor. To the (left-) right-hand side of the intersection of the dashed line and the phase boundary the transition is (second) first order.

 $I_1/I_2$ , the transition is first order and only becomes second order below a certain value.<sup>9</sup> At  $I_1$ = 0 the transition is second order and the model reduces to the zero-kinetic-energy model of a superconductor studied by Wada and Fukuda.<sup>10</sup>

We do not expect that small but finite bandwidth will qualitatively alter these results, at least for the case  $kT_s > (1/I_1)\sum_j t_{ij}^2$ . Here  $T_s$  is the transition temperature defined on the phase boundary in Fig. 1. A finite-bandwidth generalization of this system can be obtained by modifying the Gorkov Green's-function equations<sup>11</sup> to include the Mott insulator. This is done by replacing the normal free-electron Green's functions that appear in the Gorkov equations by adding a self-energy part to the former such that for  $I_2 = 0$  the solution goes over to that obtained by Hubbard<sup>5</sup> for the Mott insulator. The self-energy part is given by

$$\Sigma(\omega) = \frac{1}{2}I_1\omega/(\omega - \frac{1}{2}I_1)$$

and the normal Green's function undergoes the modification  $G^{-1} + G^{-1} - \Sigma(\omega)$ . The detailed structure of the single-particle spectrum is complicated by the fact that there are now four poles of the Green's function. This is due to the fact that the Hubbard Green's function contains two subbands and the pairing interaction  $I_2$  mixes states of these subbands and gives rise to an additional splitting.

The instability of the Mott insulator can be understood by recognizing that under the condition  $\rho I_2 \gg 1$ , where  $\rho$  is the density of states ~ (bandwidth)<sup>-1</sup>, the superconducting state can exist but that with  $I_1 \sim I_2$ , one has  $\rho I_1 \gg 1$  and the normal state is a Mott insulator. In this connection, we note that Little<sup>12</sup> showed that if a gap exists between occupied and unoccupied states in an ordinary semiconductor, the system can become unstable to superconductivity if the zero-temperature superconductor and semiconductor gaps are comparable. In the Mott insulator the gap is not due to crystal structure but electron correlations. One consequence of this distinction is the first-order transition in the present theory.

# **III. INSTABILITY TO THE FERROELECTRIC**

We now turn to a discussion of the ferroelectric instability of a Mott semiconductor. This theory had been presented earlier by the present author.<sup>13</sup> That the electron-charge ordering of Ref. 13 represents a ferroelectric state can be seen from the facts that alternating sites along a chain have (including the positive ions) opposite charge and that the lattice distortion accompanying the charge alternation yields two alternating nearest-neighbor site spacings. The interaction that gave rise to the ferroelectric instability was an electron-phonon interaction. We do not present the details of that theory and only give the results that are pertinent to this discussion. In Ref. 13 it was shown that the interaction that gives rise to the charge ordering is of the form

$$H_3 = I_3 \sum_{\langle i,j \rangle} (n_{i,+} + n_{i,+}) (n_{j,+} + n_{j,+});$$

here the brackets denote that the summation is restricted to nearest-neighbor electrons.

We have established that  $H_1 + H_2$  leads to Cooperpair formation and that  $H_1 + H_3$  leads to charge ordering (and concomitant ferroelectricity). It is straightforward to show that the total Hamiltonian,  $H = H_1 + H_2 + H_3$ , will exhibit either instability (or no instability) and that the phase boundary between the superconductor and ferroelectric is independent of temperature<sup>14</sup> (i.e., there is no crossover of phases with temperature).

#### IV. SPIN MODEL

Finally, we present a spin analog for H. The spin formalism is quite similar to that of Anderson,<sup>15</sup> except that the pseudospin operators are labeled by site indices instead of wave vectors. Let

$$S_i^z = \frac{1}{2}(n_i + n_i - 1)$$
,  $S_i^+ = C_i^+, C_i^+$ ,  $S_i^- = C_i, C_i$ ,

These pseudospin operators satisfy the usual angular-momentum algebra, but although  $S_i^z$  takes on four values (0, 0,  $\pm \frac{1}{2}$ ), it is easily seen that the spin operators do not simply represent the spin- $\frac{3}{2}$ system. Also note that  $(S_i^*)^2 = (S_i^*)^2 = 0$ , as in the spin- $\frac{1}{2}$  system. In terms of these spin operators the Hamiltonian is written

$$H = 2I_1 \sum_i (S_i^x)^2 - (I_2/N) \sum_{i,j} S_i^* S_j^-$$
$$+ 4I_3 \sum_{\langle i,j \rangle} S_i^x S_j^x + (I_1 + 4I_3q) \sum_i S_i^x + \text{const}$$

where q is the number of nearest neighbors. In the grand canonical ensemble we introduce the additional term  $-2\mu\sum_{i}S_{i}^{z}$ . It is now seen that the system may be represented by a special anisotropic Heisenberg model. The superconductivity term is represented by a planar ferromagnetic interaction and the ferroelectricity term is represented by an antiferromagnetic Ising interaction. These two terms compete to give ordering either in the plane or along the axis. The Mott-insulator term is represented by single-ion quadratic anisotropy, competing against both of the above interactions. The last term is of the form of an external magnetic field in the z direction. The strength of this field is determined by the chemical potential or the number of particles. Thus, for a halffilled band, with  $\langle S_i^z \rangle = 0$  one has  $2\mu = I_1 + 4I_3q$ . The spin representation allows a simple analog for the system of strongly correlated electrons.

## V. DISCUSSION

It has been shown that the half-filled-band Mott insulator can become unstable to a superconducting state. The analysis is based on a Hamiltonian exhibiting strong short-ranged Coulomb repulsion and a pairing interaction between localized states. While the former is familiar from the study of narrow bands, the latter has been obtained rather heuristically from the BCS reduced interaction. Although the range of the interaction in Eq. (2) has been taken to be infinite, the molecular-field treatment of the phase transition would give the same results for a finite-range interaction.

In the small-bandwidth regime, the polarizability of the boson field leads to small polaron effects. Chaikin *et al.*<sup>16</sup> have discussed this for the case of the Hubbard model coupled to excitons. The band is narrowed by a factor  $e^{-S}$ , where S is the ratio of the polaron binding energy to the average free-

<sup>\*</sup>Work performed under the auspices of the U.S. Atomic Energy Commission.

<sup>1</sup>Z. G. Soos and D. J. Klein, J. Chem. Phys. <u>55</u>, 3284 (1971); D. J. Klein and Z. G. Soos, Mol. Phys. 20, 1013 (1971).

- 20, 1013 (1971). <sup>2</sup>A. J. Epstein, S. Etemad, A. F. Garito, and A. J. Heeger, Phys. Rev. B 5, 952 (1972).
- <sup>3</sup>N. F. Mott, Proc. Phys. Soc. (London) <u>A62</u>, 416 (1949).

<sup>4</sup>L. N. Cooper, Phys. Rev. 104, 1189 (1956).

<sup>5</sup>J. Hubbard, Proc. Roy. Soc. (London) <u>A276</u>, 238

exciton excitation energy. Further, the Coulomb repulsion  $I_1$  is reduced by the polaron binding energy  $I_1 \rightarrow I_1 - 2E_B$ . In the present work, we have taken the bandwidth to be zero, which is consistent with reduction of bandwidth due to polarons. In addition, the parameter  $I_1$  can be understood to be renormalized by the polaron binding energy.

It was found that the Mott semiconductor could also have a ferroelectric instability and that the phase boundary between the superconductor and ferroelectric is independent of temperature. Consequently, with decreasing temperature the Mott semiconductor can become unstable to either of the above condensed states. A spin model is used to summarize the interactions that compete to give the three thermodynamic states discussed in this paper. This simple model serves to illustrate that a strongly correlated system of electrons (perhaps the  $\pi$  electrons of partially filled orbitals in certain organic substances<sup>1,2</sup>) that is an intrinsic semiconductor at elevated temperature can show a second- (or first-) order transition to the superconducting state.

Lastly, we mention the work of Bychkov et al.<sup>17</sup> on the possibility of superconductivity in a onedimensional system. These authors study the instability of the normal metal in the presence of a short-range Hubbard-like attractive interaction. They use a weak-coupling approximation to deduce that the metallic state is unstable to the superconductor. They also found a simultaneous Peierls instability with the superconductor. In comparison, we started with an insulating state (not restricted to one dimension) with strong electron correlations present (strong-coupling limit) and found conditions for the insulator to be unstable to superconductivity in the presence of an indirect long-range interaction. The charge-ordered ferroelectric resulted from competing inter- and intraatomic interactions and the superconductor and ferroelectric were found to be mutually exclusive.

#### ACKNOWLEDGMENTS

I am grateful to Dr. M. Blume, Professor L. N. Cooper, and Dr. H. Schuster for reading the manuscript and making helpful comments.

- (1963).
- <sup>6</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. <u>108</u>, 1175 (1957).

<sup>7</sup>N. N. Bogoliubov, Jr., Physica 32, 933 (1966).

<sup>8</sup>T. A. Kaplan and P. N. Argyres, Phys. Rev. B <u>1</u>, 2457 (1970).

 ${}^{9}$ This is representative of tricritical-point behavior; see M. Blume, V. J. Emery, and R. B. Griffiths, Phys. Rev. A <u>4</u>, 1071 (1971).

 $^{10}{\rm Y}.$  Wada and N. Fukuda, Progr. Theoret. Phys. (Kyoto) 22, 775 (1959). A further discussion of this "constant-

kinetic-energy" model is given by D. J. Thouless, The Quantum Mechanics of Many-Body Systems (Academic, New York, 1961), p. 101.

<sup>11</sup>L. P. Gorkov, Zh. Eksp. Teor. Fiz. <u>34</u>, 735 (1958) [Sov. Phys. JETP <u>34</u>, 505 (1958)].

<sup>12</sup>W. A. Little, Phys. Rev. <u>134</u>, A1416 (1964). <sup>13</sup>R. A. Bari, Phys. Rev. B <u>3</u>, 2662 (1971).

<sup>14</sup>A straightforward application of molecular-field theory in the presence of two order parameters gives the free energy (per site)  $f = \Delta^2/4I_2 + \delta^2/4I_{3q} - kT \ln(2\{e^{\beta I_1/2} + \cosh \left[\frac{1}{2}\beta(\Delta^2 + \delta^2)^{1/2}\right]\})$  where  $\delta$  represents the order parameter of the ferroelectric phase. We also note that the

competition between a lattice distortion and superconductivity had been studied in the opposite limit of the Hubbard model  $(I_1=0, t_{ij} \neq 0)$  by D. C. Mattis and W. D. Langer, Phys. Rev. Letters 25, 376 (1970).

<sup>15</sup>P. W. Anderson, Phys. Rev. 112, 1900 (1958).

<sup>16</sup>P. M. Chaikin, A. F. Garito, and A. J. Heeger, Phys. Rev. B 5, 4966 (1972).

<sup>17</sup>Yu. A. Bychkov, L. P. Gorkov, and I. E. Dzyaloshinskii, Zh. Eksp. Teor. Fiz. 50, 738 (1966) [Sov. Phys. JETP 23, 489 (1966)]; I. E. Dzyaloshinskii and A. I. Larkin, Zh. Eksp. Teor. Fiz. 61, 791 (1971) [Sov. Phys. JETP <u>34</u>, 422 (1972)].

PHYSICAL REVIEW B

#### VOLUME 7, NUMBER 5

1 MARCH 1973

# Transferred Magnetically Induced Al<sup>27</sup> Quadrupole Interaction in GdAl,

J. Degani\* and N. Kaplan

The Racah Institute of Physics, The Hebrew University, Jerusalem, Israel (Received 15 August 1972)

A direct observation of a magnetically induced electric field gradient (efg) at a nonmagnetic Al site in ferromagnetic GdAl<sub>2</sub> is described. The efg was determined from accurate measurements of the Al<sup>27</sup> nuclear-spin-echo envelope-modulation frequency, performed on Al in a sites for which the magnetization  $\vec{M}$  and the crystaline efg axis are colinear. Monitored as function of M, it is found that the total efg in *a* sites is given by  $q = q_c - (M/M_{sat})q_m$ , with  $e^2 Q^{27}q_c = 4.19 \pm 0.05$  MHz and  $e^2 Q^{27}q_m = 0.577 \pm 0.05$ MHz. The origin of  $q_m$  is as yet unclear, since simple models leading to induced efg will result in  $M^2$ dependence for the transferred induced efg term, contrary to the linear M dependence presently observed.

In recent years there have been several reports about nuclear-quadrupole-interaction (QI) measurements in which a magnetically induced electric field gradient (efg) was observed at the nuclear site of magnetic ions in solids.<sup>1,2</sup> In these ions the electronic wave functions of the unfilled magnetic shell, and thus the charge distribution around the ionic nucleus, depend on the state of magnetization of the ion, which in turn results in a magnetization-dependent efg. Now, in analogy with the case of magnetic hyperfine interactions, one might also search for a process in which the efg at the nuclear site of *nonmagnetic* ions in a crystal is effected by the state of magnetization of a neighboring magnetic ion. In principle, any crystal in which magnetic-transferred hyperfine interaction has been identified is suitable for such a search provided  $I \ge 1$ , where I is the spin of the nucleus of the nonmagnetic ions. The search to be described was conducted on ferromagnetic GdAl<sub>2</sub>, in which the Gd has the role of the magnetic ions and the sizable M-dependent  $Al^{27}$  QI was detected in the nonmagnetic Al ions. We believe this report provides the first direct experimental evidence for the existence of a transferred magnetically induced efg in solids.<sup>3</sup>

Familiarity with some features of the compound is essential for the understanding of the present

experiment: GdAl<sub>2</sub> is an intermetallic compound of the cubic Laves phases structure<sup>4</sup>; there are 16 Al ions in the cubic cell (Fig. 1), forming four tetrahedra, and the point symmetry of each Al is  $\overline{3}m$ , with the threefold symmetry axes parallel to the principal diagonals of the cubic unit cell; below 176°K, the Gd moments order ferromagnetically



FIG. 1. Unit cell of GdAl<sub>2</sub>. Large circles represent Gd and small shaded circles represent Al ions.