

On this basis, the writers have analyzed the results of Smith,⁶ who measured the modulus of copper after a deformation of 1% as a function of time and annealing temperature. The resulting values of β are shown in Fig. 1 for two specimens of different purities. From this one can derive that the defect producing the dislocation pinning has a migration activation energy of 1.0 ev, and it is reasonable to assume that the defects are the deformation-induced vacancies, since the activation energy is the right order of magnitude.

Although this value does not agree exactly with any of the other experimental determinations, it does not seem unreasonable in the light of the recent measurements of Bauerle and Koehler for quenched-in vacancies.⁷ For gold, they find 0.98 ev for the activation energy of formation (U_F) and 0.82 ev for that of migration (U_M) of vacancies. The sum of 1.80 ev agrees rather well with the measured self-diffusion constant of 1.81 ev.⁸ The ratio of the migration to the self-diffusion activation energy is then $U_M/(U_F+U_M)=0.46$. If this ratio is assumed to hold also for copper, which has an activation energy for self-diffusion of 2.05 ev,⁹ one obtains a rather good agreement with the value for the activation energy for the migration of vacancies in copper reported in this note.

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¹ H. B. Huntington, *Phys. Rev.* **91**, 1092 (1953); see also H. B. Huntington and F. Seitz, *Phys. Rev.* **61**, 315 (1942); H. B. Huntington, *Phys. Rev.* **61**, 325 (1942).

² Brinkman, Dixon, and Meehan, *Acta Metallurgica* **2**, 38 (1954). See also F. Seitz and J. S. Koehler, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2; and T. Broom, *Advances in Physics*, edited by W. F. Mott (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 26.

³ Granato, Hikata, and Lücke, *Acta Metallurgica* (to be published).

⁴ A. Granato and K. Lücke, *J. Appl. Phys.* **27**, 583, 789 (1956).

⁵ A. H. Cottrell and B. A. Bilby, *Proc. Phys. Soc. (London)* **A62**, 49 (1949).

⁶ A. D. N. Smith, *Phil. Mag.* **44**, 453 (1953).

⁷ J. E. Bauerle and J. S. Koehler, *Phys. Rev.* **107**, 1493 (1957).

⁸ Makin, Rowe, and LeClaire, *Proc. Phys. Soc. (London)* **B70**, 545 (1957). See also B. Okkerse [*Phys. Rev.* **103**, 1246 (1956)] who finds the smaller value of 1.71 ev.

⁹ Kuper, Letaw, Slifkin, Sonder, and Tomizuka, *Phys. Rev.* **96**, 1224 (1954).

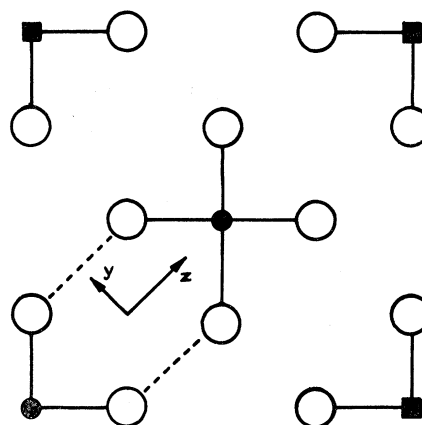


FIG. 1. Section of $(\text{NH}_4)_2\text{Ir}_2\text{PtCl}_6$ in $(1,0,0)$ plane showing nearest-neighbor pair of Ir ions, $\bullet = \text{Ir}$, $\circ = \text{Cl}$, $\blacksquare = \text{Pt}$. The x axis is perpendicular to the plane of the paper. The Ir-Ir separation = 6.9 Å.

$(\text{NH}_4)_2\text{IrCl}_6$. This is a salt with face-centered cubic crystal structure which shows antiferromagnetic behavior in the liquid helium temperature range. The magnetic carriers are the Ir ions, each of which has one unpaired electron with spin $s = \frac{1}{2}$, and it seems likely that the only important exchange interactions are between nearest neighbor Ir ions. The mechanism of the interaction appears to be superexchange¹ via intervening Cl ions; see Fig. 1.

Measurements have been made on a series of crystals of different magnetic dilutions in which diamagnetic platinum ions are substituted for Ir ions. The results are as follows:

(i) In dilute crystals, Ir:Pt \leq 1:100, each Ir is well separated from other Ir's, and there are no exchange interactions. Electron spin resonance measurements show that the unpaired spin is about 70% in 5 d orbits on the central Ir ion, and about 30% in p_π orbits on the surrounding octahedron of six Cl ions belonging to the $(\text{IrCl}_6)^{2-}$ complex.^{2,3}

(ii) In semidilute crystals, Ir:Pt \approx 1:10, there is an appreciable chance of two $(\text{IrCl}_6)^{2-}$ complexes occupying nearest-neighbor positions in the lattice (Fig. 1). There is then overlap of the two unpaired electrons via their transfer onto the intervening Cl ions, and an exchange-coupled pair of Ir ions results. The interaction can be formally written (see, for example, Baker and Bleaney⁴)

$$H_{ex} = J\mathbf{s}' \cdot \mathbf{s}'' + J_x s'_x s''_x + J_y s'_y s''_y + J_z s'_z s''_z \\ = \frac{1}{2}J[S(S+1) - \frac{3}{2}] + \frac{1}{2}[J_x S_x^2 + J_y S_y^2 + J_z S_z^2], \quad (1)$$

where $J_x + J_y + J_z = 0$, $s' = s'' = \frac{1}{2}$ are the spins of the two unpaired electrons, and S is the total spin of the system which can take values 1 or 0. J is the isotropic exchange parameter which splits the levels into a triplet $S=1$ at $J/4$, and a singlet $S=0$ at $-3J/4$ (Fig. 2). J_x , J_y , and J_z are anisotropic exchange parameters which cause a small splitting between the

Superexchange in Ammonium Chloroiridate

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PARAMAGNETIC resonance methods have been used to investigate exchange interactions in

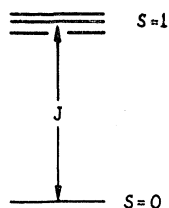


FIG. 2. Energy levels of Ir-Ir pair.

levels of the triplet. Measurement of electron spin resonance transitions between the Zeeman components of the triplet at 20°K and using 1.25 and 3.3 cm wavelengths give $J_x = \mp 0.84$, $J_y = \pm 0.64$, $J_z = \pm 0.20$ cm⁻¹, and also show the axes of the interaction to be as indicated in Fig. 1. Because of the presence of the singlet ($S=0$) the signal intensity R shows a temperature dependence which is approximately of the form (see Bleaney and Bowers⁶)

$$R \propto \frac{1}{T} \left(\frac{1}{3 + e^{J/kT}} \right). \quad (2)$$

Measurements of this intensity down to 2°K yield $J/k = +7.5 \pm 1.0$ °K. The positive sign indicates antiferromagnetic exchange.

(iii) In concentrated crystals each Ir ion has twelve nearest neighbor Ir ions in the face-centered cubic lattice. At high temperatures ($J/kT \ll 1$) the interaction (1) then leads to a Curie-Weiss susceptibility law $\chi = C/(T + \theta)$ with Weiss constant (see, for example, Anderson¹)

$$\theta = zJs(s+1)/3k = 3J/k, \quad (3)$$

where $z=12$ is the nearest-neighbor number. The anisotropic parameters J_x, J_y, J_z do not enter this expression because the g value is isotropic. Measurement of this susceptibility up to 300°K by F. R. McKim shows $\theta = 24$ °K, giving $J/k = 8$ °K, in reasonable agreement with the resonance results on semidilute crystals where the Ir-Ir separation is practically the same. The low-temperature susceptibility measured by A. H. Cooke, F. R. McKim, and W. P. Wolf gives evidence for an antiferromagnetic transition at 2.1°K. The specific heat measured by P. L. Smith and C. A. Bailey shows a λ -type anomaly with its peak at 2.15°K.

The results show that paramagnetic resonance techniques provide a powerful method for investigating exchange interactions in antiferromagnetics if mixed crystals can be grown. A full account of these experiments on ammonium chloroiridate and on some other salts of iridium will be published in due course together with a theoretical analysis of the exchange mechanism which has been made by B. R. Judd. Preliminary measurements of one of these salts, sodium chloroiridate, have been reported previously by Griffiths and Partridge (quoted by Baker and Bleaney⁴).

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¹ P. W. Anderson, Phys. Rev. **79**, 350, 705 (1950).

² K. W. H. Stevens, Proc. Roy. Soc. (London) **A219**, 542 (1953).

³ J. H. E. Griffiths and J. Owen, Proc. Roy. Soc. (London) **A226**, 96 (1954).

⁴ J. M. Baker and B. Bleaney, *Conference on the Physics of Low Temperatures, Paris, 1955* (Centre National de la Recherche Scientifique, and UNESCO, Paris, 1956).

⁶ B. Bleaney and K. D. Bowers, Proc. Roy. Soc. (London) **A214**, 451 (1952).

Excitation of Rotons in Helium II by Cold Neutrons

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THE theories of helium II developed by Landau¹ and Feynman² conclude that in helium below the λ point there exist relatively simple periodic motions of the fluid, *viz.*, phonons and rotons, having mean free paths long compared to their wavelengths. Although in the past few years neutron experiments have clearly demonstrated the existence of phonons in solids, neutron experiments to date³ have failed to demonstrate any marked difference in helium measurements below and above the λ point. Earlier this year Cohen and Feynman⁴ pointed out that on the basis of the Landau-Feynman theories, there should be a measurable difference in the energy distribution of scattered neutrons from He I as compared to He II. This letter is to report the preliminary results of such a measurement.

A neutron beam from the Stockholm Reactor⁵ was filtered through 8 in. of polycrystalline Be, resulting in a neutron spectrum characterized by a sharp rise in intensity at 3.96 Å, and varying approximately as $1/\lambda^6$ for wavelengths greater than 3.96 Å (see Fig. 1). With this incident spectrum, the energy spectrum of the neutrons scattered at 90° from the incoming direction, by a 10-cm sample of liquid helium, at $T=1.2$ and 4.2°K, was measured. The energy was inferred from the measured time-of-flight of the neutrons using the Stockholm slow chopper and associated electronic equipment.⁶

Figure 1 shows the incident spectrum as measured by elastically scattering the incoming beam through 90° by means of a thin vanadium sample. (The scattering amplitude of vanadium is nearly totally incoherent.) The ordinate in the figure represents the counting rate in a 30- μ sec time interval at the time indicated on the abscissa. The sharp rise in intensity at 3.96 Å is the