given by Eqs. (6):

$$\epsilon_1 = n^2 - k^2, \tag{6a}$$

$$\epsilon_2 = -2nk. \tag{6b}$$

$\mathrm{Im}[1/\epsilon]$ appears to be reaching a maximum just beyond 6.0 ev, corresponding to a small value of ϵ_2 when ϵ_1 is nearly zero. At this energy one should expect a collective or plasma oscillation to occur.[6] Similar maxima should appear for indium antimonide and gallium arsenide at energies just beyond 6.0 ev, $\mathrm{Im}(1/\epsilon)$ for these materials at 6.0 ev being 1.74 and 0.77.

CONCLUSIONS

The results of the investigation reported in this paper can be summarized in the following conclusions:

(a) The optical properties of InAs, InSb, and GaAs can be obtained from measurements of reflectivity and analysis of the data using dispersion relations. This technique can be applied to calculated optical constants where transmission measurements are not possible.

(b) The magnitude and spectral detail of the reflectivity of III–V compounds are strongly dependent on the surface quality, although for a given surface the measured reflectivities are reproducible to within ±0.01.

(c) Calculations of $\mathrm{Im}(1/\epsilon)$ for the three materials studied indicate that there should be a plasma energy at about 7 ev.

(d) Although the optical constants calculated are believed to be nearly correct at energies well below the limit of experimental data, more definitive results require extension of the reflectivity data further into the ultraviolet as well as a more refined extrapolation procedure.

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Sublattice Switching in Antiferromagnets

W. J. O'Sullivan, W. A. Robinson, and W. W. Simmons
Space Technology Laboratories, Inc., El Segundo, California

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An experiment is described which enables an antiferromagnetic sublattice assignment to be monitored in the presence of a small applied magnetic field. Using this technique it becomes possible to observe sublattice switching events, should they occur. It is concluded, despite the negative character of the results, that a particular magnetic sublattice assignment in $CuCl_2 \cdot 2H_2O$ can be considered a stable form at 1.4°K.

In principle, a quantum mechanical resonance effect should exist between a given antiferromagnetic spin arrangement and the configuration which follows upon inverting the magnetization direction of each of its spin sublattices. These two spin arrangements are indistinguishable and, in the case of a system of only two magnetic sublattices, are related by an interchange of sublattices.

Theoretical treatments by Anderson,[1] Kubo,[2] and Van Kranendonk and Van Vleck[3] established the long-term stability against rotation of the magnetization axes in the ground state of three-dimensional antiferromagnets, even in the limit of zero anisotropy energy. The tendency for the magnetization direction to wander is further inhibited by the existence of anisotropic interactions in real systems. The resonance effect between the two indistinguishable spin arrangements is extremely weak, in the ground state occurring only in a perturbation expansion carried to an order N, which is of the order of the number of atoms in the crystal. Although the problem becomes more complicated at higher temperatures,[2] the presence of even a small anisotropy energy tends to make rotations of the magnetization axes through large angles highly improbable. The assignment of definite magnetization directions to particular sublattices in antiferromagnetic materials has been likened[3] to heavy molecular isomers, technically metastable, but considered stable for all practical observation times.

Previous experimentally deduced upper limits placed upon the frequency of sublattice exchange events are too high to be significant. Neutron diffraction studies of antiferromagnetic materials are insensitive to the sublattice switching process if the lifetime of a particular magnetic sublattice arrangement is long compared to the time of interaction between an incident neutron and the spin array. Electron and nuclear magnetic resonance linewidth considerations in antiferro-

[1] P. W. Anderson, Phys. Rev. **86**, 694 (1952).
[2] R. Kubo, Phys. Rev. **87**, 568 (1952).
[3] J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. **30**, 1 (1958).

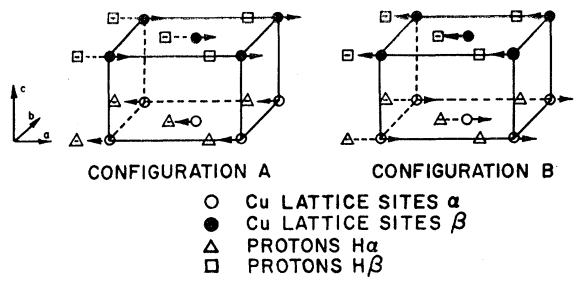

FIG. 1. The two equivalent spin arrangements in $CuCl_2 \cdot 2H_2O$. The proton positions as shown serve illustrative purposes only. They do not correspond to any proton site in an actual $CuCl_2 \cdot 2H_2O$ lattice.

magnets are of little value for gaining insight into the times involved. For example, from the proton nuclear magnetic resonance (NMR) linewidths a lower limit of $\cong 10^{-4}$ sec may be placed upon the lifetime of a magnetic sublattice assignment in antiferromagnetic $CuCl_2 \cdot 2H_2O$.[4]

The purpose of this article is to describe an experiment which monitors a given antiferromagnetic spin arrangement and would respond to a sublattice switching event should one ever occur. Observation times are approximately 10 hours, which, while not of geological magnitude, will be considered long enough.

In Fig. 1 are depicted two equivalent spin arrangements in antiferromagnetic $CuCl_2 \cdot 2H_2O$, the particular material investigated. While the model of the spin arrangement illustrated is that due to Poulis and Hardeman,[5] the canted spin arrangement suggested by Moriya[6] would serve quite as well. The important point is that there is a set of protons (H_α in Fig. 1) bearing the same spatial relationship to the copper sites of sublattice α as the set of protons H_β bears to sublattice β sites.

The sample (a single-crystal $CuCl_2 \cdot 2H_2O$ needle $\cong 0.5$ cm^3) sits within a set of two orthogonal rf coils, each of which forms part of the tank circuit of one of two marginal oscillators operating in the 2–5 Mc/sec range. With this arrangement, both the H_α and H_β protons can be observed simultaneously when an external magnetic field[7] is applied. The sample temperature is held at 1.4°K, and a 230-gauss magnetic field is applied along the crystalline *a* axis. Then the nuclear spin distribution of one set of protons, either H_α or H_β, is continually saturated with one marginal oscillator while the NMR of the second set is observed with the other. This situation, continual saturation of one proton resonance and monitoring of the second, is maintained for the full observation period. If sublattice switching is occurring, there is only one set of conditions we need consider. That is, the time between separate switching transitions is longer than the proton T_1, which is approximately 2 sec under small applied fields at 1.4°K.[8] If the time separating individual transitions is short compared to the T_1 for the protons, it can be shown that both the H_α and H_β NMR would be saturated, should a saturating rf field be applied at either the H_α or H_β resonance frequency. This double saturation effect is not observed. In addition, the conclusions drawn from this experiment remain essentially unaffected whether or not the proton spins are able to follow, adiabatically, the rapidly changing internal field occurring as a consequence of an exchange between the two equivalent configurations *A* and *B*.

If a sublattice exchange occurs, the saturated and unsaturated proton spin systems exchange internal fields. Consequently, the monitored NMR signal should decrease and then build up to its former level in a time characterized by the proton T_1.

In no observation periods were effects detected suggesting a sublattice exchange had occurred. In another experiment we attempted to reduce the anisotropy advantage that the *a* axis holds over the other crystalline axes by applying a field along the *a* axis within 100 gauss of the critical field for flopping of the sublattices to the $\pm b$ direction. In this case also there was no evidence of sublattice switching.

We are led to conclude, then, despite the intrinsically negative character of the experimental results, that a particular magnetic sublattice assignment in $CuCl_2 \cdot 2H_2O$ can be considered a stable form, at least up to 1.4°K.

[4] C. J. Gorter, Revs. Modern Phys. **25**, 332 (1953).
[5] N. J. Poulis and G. E. G. Hardeman, Physica **18**, 201 (1952).
[6] T. Moriya, Phys. Rev. **120**, 91 (1960).
[7] In the presence of an applied magnetic field, the equivalence between the two inverse-related sublattice assignments is lifted by the difference of the electron spin-nuclear spin interactions before and after a sublattice switch, this difference decaying with T_1^{nuclear}. If the nuclear spins cannot follow a switching process, adiabatically, then a transient energy splitting exists, even in the absence of an applied field. In either case, we consider the energy splitting negligible.
[8] N. J. Poulis, G. E. G. Hardeman, W. Van der Lugt, and W. P. A. Hass, Physica **24**, 280 (1958).