Cryogenics and Nuclear Physics

Solid-state and nuclear physics advance together through research at ultralow temperatures.

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One of the most powerful stimuli to progress in scientific research is the bringing together of two ostensibly widely differing fields of endeavor. Frequently this involves the adaptation of a specialized experimental technique to an area quite different from that in which it had been previously applied. Well-known examples are the use of microwaves in solid-state spectroscopy (with an eventual repayment of the debt by way of the maser); of nuclear magnetic resonance in structural chemistry; of ultrasonic absorption to study electron-lattice interaction in metals; of cryogenic liquids for high-energy particle detection in the bubble chamber; and of radioactive materials as "tracers" in many fields of study.

The marriage of nuclear physics and cryogenics has been a particularly fruitful one. In addition to the development of the bubble chamber already cited, notable advances in the field of radioactivity have been made possible through nuclear orientation at low temperatures, while oriented radioactive nuclei may be applied, in turn, to investigating a variety of solid-state problems. Since the first successful experiments in 1951, low-temperature nuclear orientation studies have yielded much useful information on nuclear spins, moments, and decay modes of gamma emitters. When, in late 1956, the technique was applied (under the stimulus of a bold theoretical speculation) to the study of beta decay, the consequent demonstration of the nonconservation of parity resulted in a phenomenal burst of activity in research on the weak interactions generally.

At present, in many laboratories throughout the world, exploratory work is in progress to produce a useful degree of nuclear polarization by combining microwave and radiofrequency engineering with cryogenics and solid-state physics. When this method ("dynamic polarization") is perfected, both low-energy and high-energy nuclear physicists will seize it with enthusiasm, while along the way the atomic physicist is discovering much of interest concerning interactions and relaxation processes.

Orientation of Nuclei at Low Temperatures

Static methods. According to the third law of thermodynamics, systems in internal equilibrium approach a state of perfect order (zero entropy) as the temperature is lowered toward absolute zero. There are certain classes of inorganic salts which are almost ideal paramagnetics—that is, their susceptibility χ follows the Curie law

$$\chi = kT^{-1}$$

even at temperatures as low as 1°K. Thus, in zero external magnetic field their electronic magnetic moments remain completely disordered, and the associated entropy will not fall toward zero (as required by the third law) until the weak internal constraints (interactions) become comparable with the thermal energy. This will be achieved between 0.001° and 0.1°K, depending upon the specific properties of the individual substances. The possibility of forcing the electronic magnetic moments into a state of order at 1°K by means of an externally applied magnetic field, however, leads to the technique of cooling by isentropic demagnetization (removal of the field from a thermally isolated, initially magnetized specimen) into the region of very low temperatures indicated above.

Nuclear magnetic moments are in general about 1000 times smaller than their electronic counterparts. Hence, nuclear polarization demands a corresponding increase in the value of H/T, and one would need to apply, for example, 10,000 gauss at 0.001°K to produce a high degree of nuclear polarization. The technical difficulties besetting this direct or "brute force" method are evidently very great, and only small polarizations have been achieved in this way to date. Fortunately, here nature is for once helpful to the experimenter, providing a coupling between nuclear and electronic moments which may be made use of in specific instances to automatically align the nuclei if the electron spins are first aligned. The latter can be accomplished by producing a very low temperature by adiabatic demagnetization, and then utilizing either a relatively small external magnetic field (the Gorter-Rose method) or anisotropy, when it exists, in the crystalline electric field (the Bleaney method). A fourth method, developed by Pound, is to make use of the gradient of crystal field acting upon the nuclear electric quadrupole moment. The Bleaney and Pound methods give rise to *alignment*

$$\langle I_z \rangle_{av} = 0, \ \langle I_z^2 \rangle_{av} \neq 0$$

while the "brute force" and Gorter-Rose methods yield *polarization*

$$\langle I_z \rangle_{av} \neq 0$$

Here I_z is the component of the nuclear spin along the orientation axis.

Subsequently yet another technique was found—the production of nuclear polarization in *ferromagnetic* materials. Very recently, Russian scientists have shown that even nuclei of diamagnetic atoms are subjected to comparably intense polarizing forces (effective fields of the order of 10^6 gauss) when such atoms are alloyed in small concentrations in ferromagnetic host substances. It is thought that here one is encountering a polarization of the inner-core electrons by the "conduction electrons." The *s*-electrons need be only slightly

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affected to yield a powerful resultant force on the nucleus, and in fact, experience to date also suggests that it is this core polarization effect which gives the dominant contribution to H_{eff} at the nuclei of ferromagnetic substances.

Dynamic methods. The energy levels of a paramagnetic ion showing hyperfine structure are characterized (to take the simplest example) by the values of the electronic and nuclear angular momentum projection quantum numbers M and m ($=J_z$ and I_z). In a magnetic field of the order of 10^4 gauss, states of differing M are split apart, with separations in the microwave region of energy of approximately 1 cm^{-1}. Under conditions of thermal equilibrium, a Boltzmann distribution of ions over the various energy levels exists, maintained by "relaxation processes" which "flip" spins and exchange energy with the crystal lattice. These processes may be transitions involving a change (*a*) in M alone, (*b*) in m alone, or (*c*) in both M and m simultaneously.

When microwave energy is fed into this system, transitions are induced and the population distribution is disturbed. It is possible to equalize the populations of two separated levels, and the transition is then said to be "saturated." By bringing about an unequal distribution over states of positive and negative m, we automatically achieve nuclear polarization. There are two different ways of proceeding (we particularize to the case of $J = \frac{1}{2} = I$ for simplicity).

1) Select the microwave frequency to excite a transition of type *a*, reversing M only. Then if relaxation mode *c* is the dominant one, the partition over the $m = +\frac{1}{2}$ and $m = -\frac{1}{2}$ states is progressively driven away from the thermal equilibrium condition of equality, and nuclear polarization results. This is a method originally conceived by Overhauser for metals.

2) Excite a type of transition, reversing both spins and thus establishing a nuclear polarization. The latter is doubled when relaxation takes place by way of type *a* processes, where only the electron spin reverses. This method was developed by Jeffries.

Cases 1 and 2 are illustrated in Figs. 1 and 2, respectively, wherein $\Delta = g\beta H$, the electronic Zeeman energy, and δ is the hyperfine splitting.

Another case for the dynamic polarization of nuclei (proposed by Abragam) is that of an electron and a nucleus under weak dipolar coupling. The nuclear splitting is then just $g_n\beta_n H$. As in the above cases, the polarization achieved may be positive or negative, depending on which transition is selected for saturation. The enhancement factor is $\Delta/\delta = g\beta/g_n\beta_n$. This situation is found in paramagnetic crystals where the magnetic ions are surrounded by the protons of water of crystallization, and in various materials containing paramagnetic impurities. The latter are perhaps of most interest to the nuclear physicist, for by irradiating simple hydrocarbon materials to produce *F*-centers it is possible to have an assembly of electron spins and protons that are adulterated only by carbon and are entirely free of heavy elements.

Finally, one may mention a transient method developed by Feher. In this, a system such as was discussed at the beginning of this section is subjected to two successive rapid changes of the direct-current magnetic field in a process known as "adiabatic fast passage." The first change inverts the populations of a pair of electronic levels; the second has a similar effect on the populations of a pair of nuclear levels. Nuclear polarization results, and the system relaxes back to the equilibrium state.

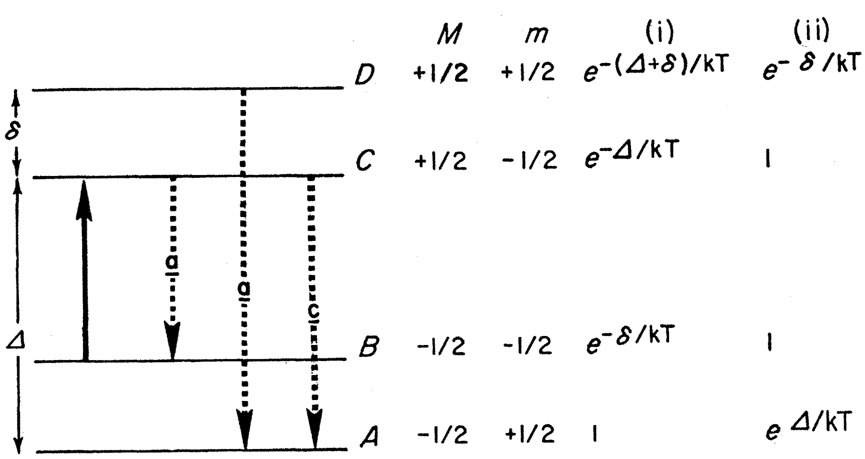

Fig. 1. (Solid arrow) "Allowed" transition selectively excited to saturation; (broken arrows) relaxation processes comprising (*a*) normal process, flipping only the electron spin, and (*c*) transition which flips electronic and nuclear spins in opposite directions ($C \rightarrow A$). (i) Thermal equilibrium populations (unnormalized); (ii) populations upon application of microwave field. The requirements are that n_C is now equal to n_B and that ratios n_D/n_A and n_C/n_A remain unaltered from the original state. Nuclear polarization, $\Sigma n_m m/I\Sigma n_m, \approx +\Delta/4kT$, for δ, $\Delta << kT$. Since the polarization for the thermal equilibrium state (i) $\approx \delta\Delta/4k^2T^2$, an enhancement by a factor kT/δ is achieved which is of the order of 100 at liquid helium temperatures. (If relaxation process $D \rightarrow B$ should have a probability equal to that of $C \rightarrow A$, the method fails.)

Low Temperature Nuclear Orientation Experiments

General. A nucleus of a radioactive element resembles a radio antenna in that the probability of emission has a characteristic directional dependence. The analogy becomes more satisfactory if one considers an *assembly* of such nuclei 100 percent polarized, for then the characteristic anisotropic pattern is evidenced by the observed intensity, as in the radio propagation case. The pattern is rotationally symmetric around the polarization axis. For randomly oriented nuclei, as at room temperature, the emission is averaged out to spherical symmetry and an intermediate situation obtains for partially polarized nuclear ensembles. Alpha-ray and gamma-ray anisotropy may be observed with aligned or polarized nuclei; beta-ray asymmetry is produced only by polarized systems (*1*).

The precise details of the angular variation with respect to the system axis depend upon the spins of the initial, intermediate, and final states of the nucleus under study; upon the angular momentum carried away in the transition; and upon the properties of the crystal of which the parent atoms are constituents. For the method to be feasible it must be possible, of course, to satisfy the solid-state requirements, which are in general quite restrictive. Experiments to date have thus dealt chiefly with nuclei of elements in the iron group and in the rare-earth and transuranic series. It is probable that developments in the ferromagnetic alloying method and in the dynamic

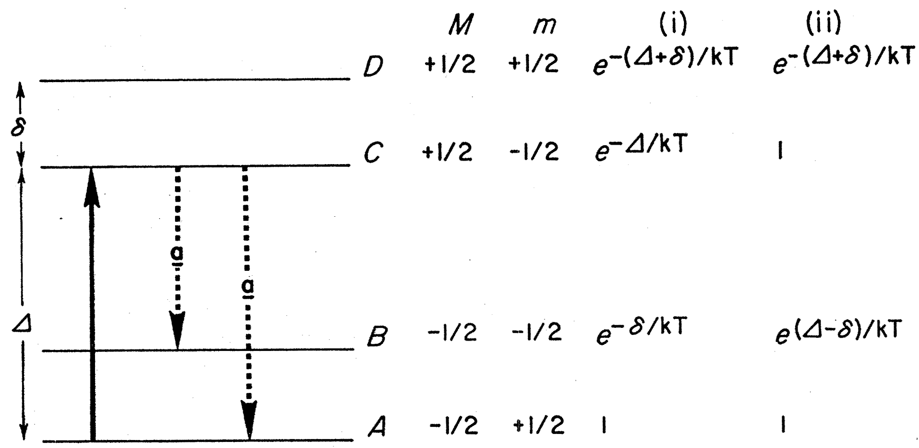

Fig. 2. (Solid arrow) "Forbidden" transition selectively excited to saturation; (broken arrows) normal relaxation process, flipping only the electron spin. (i) Thermal equilibrium populations (unnormalized); (ii) populations upon application of microwave field. The requirements are that n_o is now equal to n_A and that ratios n_C/n_B and n_D/n_A remain unaltered from the original state. Nuclear polarization $\approx -\Delta/2kT$ for $\delta, \Delta << kT$.

polarization field will substantially increase the number of nuclear candidates for study at low temperatures.

The orientation of a nucleus may change upon its emitting a beta particle prior to gamma decay. Thus, the "orientation parameters" determining the gamma emission will differ from those calculated for the parent nuclei under the given experimental conditions, but this can usually be taken into account quite rigorously. If, however, the lifetime of the intermediate state is long as compared with the precession period of the daughter nucleus in the polarizing field ($\tau > 10^{-9}$ sec), this is no longer true, and indeed the emission anisotropy may be considerably attenuated.

If a gamma-ray transition is "mixed" (for example, if it contains both magnetic dipole and electric quadrupole modes), the emission pattern will be affected, though the anisotropy may be either attenuated or enhanced by interference. By studying the plane polarization of the gamma radiation, too, one may obtain an independent estimate of the mixing ratio. This approach is especially useful when the mixing is such as to severely attenuate the anisotropy, an effect which might otherwise be dismissed as merely evidence that the nuclei were not being oriented.

Diminished orientation can arise from solid-state causes such as imperfect crystals, or (more fundamentally) dipolar interactions among the paramagnetic ions. When radioactive specimens are prepared by neutron bombardment of inactive crystals, it is possible to observe an increase in the radiation anisotropy with successive experiments. This is due to healing from the "damage" of recoil displacement of the target ions, and hence it is possible to draw conclusions concerning the precise nature of such damage, activation energies, and so on.

Alpha-particle emission has been studied to check the theory of the effect of nuclear shape on the probability of emission and to check ideas concerning the bonding in heavy element compounds. Measurements of the spin-dependence of the slow-neutron-capture cross section tell one the spin of the compound nucleus.

The emission anisotropy may be used as a thermometric parameter, once the details have been well established in calibration experiments. The activity must always be kept low to avoid a significant heating of the specimen, hence the thermometer is a vanishingly small appendage to the system and is in intimate thermal contact, while the indication is as remote as could be desired—circumstances which approach ideality from the point of view of practical thermometry.

In sum, it is evident that not only may the numerous aspects of radioactive decay be studied by nuclear orientation methods but, conversely, the nuclear properties may be made use of in such diverse studies as crystal structure; electron-nucleus, ion-ion, and ion-lattice interactions; radiation damage; thermal equilibrium and thermometry.

Beta decay and the nonconservation of parity. The case of beta asymmetry is worthy of separate discussion in that it involves theoretical considerations of most fundamental importance, while its observation demands very special experimental conditions. The theory of beta decay was developed by Fermi and others in the 1930's and remained unaltered basically for some twenty years. Within the framework of this theory the interaction describing the decay of a neutron into a proton plus a beta particle and a neutrino can be formulated in five distinct ways, according to the Lorentz tranformation properties. There is a second set of five formulations for which the Hamiltonian function changes sign upon inversion of the coordinate system, and which were therefore rejected because of the violation of the principle of parity conservation (or space inversion invariance).

When, in 1956, Lee and Yang were facing a baffling problem in the decay of K mesons, they were tempted to question the heretofore sacrosanct principle, at least in the "weak interactions," which include K-meson and beta decays. Upon investigation, they discovered that the point had never actually been put to experimental test. Let us describe the energy of the system by the *general* Hamiltonian function, which contains both parity-conserving and parity-violating terms. We represent these by C and C', scalar and pseudoscalar, respectively. The general expression for an observable as a function of the system's parameters (for example, spin, momentum, energy, and so on) will contain terms in C^2, $(C')^2$, and CC'. Only the last-mentioned will change sign under space-inversion (being pseudoscalar while *both* the others are scalar) and give rise to an asymmetry which may then be detected. Beta-decay experiments carried out prior to the Lee-Yang investigation, not involving pseudoscalar combinations of parameters, could not demonstrate the nonconservation of parity even if it existed. Lee and Yang proposed several experiments which could provide the necessary test: the distribution function for beta particles (of momentum $\mathbf{p}$) from polarized nuclei (of spin $\mathbf{I}$), for example, contains a term in $(\mathbf{I} \cdot \mathbf{p})$ which is pseudoscalar. In view of the fact that the interference term being sought might well be a small one, it was this experiment which offered the best hope of success as far as the size of an observable effect was concerned.

An important consideration here is frequently lost sight of in retrospect.

While it is true that experimentalists are not invariably deterred from "speculative" investigations, even in the face of theoretical deterrents, they are not inclined to undertake such an experiment if it is exceptionally difficult *and* flies in the face of a most hallowed tenet of theoretical physics, until there is at least some reason to question the latter. Thus it remained for Lee and Yang to raise the necessary doubt, and, in the event, the experiment proved to be somewhat less formidable than prior consideration suggested.

It proved possible to adapt low-temperature nuclear orientation techniques to the polarization of nuclei in thin surface layers of material, to maintain the polarized state for a usefully long period, and to peform the counting of the emitted beta particles within the inner chamber of the cryostat. The intensity of emission was found to be asymmetric with respect to the nuclear polarization vector, and the nonconservation of parity was established as a fact in nuclear beta decay. The effect was large, indeed a maximum ($C' = C$), and this circumstance made feasible a host of other experiments, in which the achievable asymmetry is much smaller than in the primary experiment. In addition to the flood of investigations into the weak interactions (β, π, and μ decays; properties of the neutrino) that followed, considerable activity was engendered in related studies (invariance principles in nuclear physics; elementary particles, and so on), which is still continuing.

Nuclear Cryogenics

Just as very low temperatures may be produced by the isentropic demagnetization of electronic paramagnetics, so, in principle, may ultralow temperatures be achieved through "nuclear cooling." Referring back to the discussion of static methods of orienting nuclei at low temperatures, however, one sees that the initial entropy reduction by isothermal magnetization can only be obtained with available magnetic-field intensities at starting temperatures of the order of 0.001°K. Thus, one is led to a two-stage process in which an electronic stage produces the starting temperature for the second, or nuclear, stage. Because the magnetizing field for the latter must not impinge upon the first stage and so reheat it, this field cannot be made very large, except with extremely careful magnet design, and the two stages must be separated by a thermal link. Ideally, this link should have the properties of a switch, providing excellent thermal contact while the nuclear stage is cooling down to its starting temperature and complete isolation thereafter.

The many severe technical problems were finally surmounted some 5 years ago (*2*) with the first successful experiments in Oxford, when a nuclear-spin temperature of about 10^{-5} °K was produced. More recently the range has been extended down to 10^{-6} °K, with starting conditions of 30 kilogauss at 0.01 °K. The nuclear-spin system was provided by a copper specimen which formed its own (temperature-dependent) thermal link to a compress of chromic potassium alum, the first stage. Such experiments yield information on the interaction between nuclear and electronic spins, and between the nuclear spins themselves, at extremely low temperatures. When still lower temperatures are reached, one should be able to observe the nuclear paramagnetism give way to ferromagnetic or antiferromagnetic ordering. One intriguing aspect of this type of investigation is the fantastic degree of thermal insulation demanded. For example, if the ordered state should only be manifested below, say, 10^{-7} °K, then it could be destroyed by a heat influx as small as 10 ergs per mole. But heat leaks occur by way of the crystal lattice or the electron assembly, or both, and the experiments show that the nuclear spins are very well isolated from these other systems at the very lowest temperatures.

Future Prospects

For the immediate future one may expect a continuation of conventional (as they are already regarded) experiments in which nuclear and other data are derived from alpha, beta, and gamma decays. The newer techniques for polarizing nuclei at low temperatures—the dynamic method and the incorporation of trace elements into ferromagnetics—will undoubtedly both broaden this field and open up new avenues of interest.

In the case of ferromagnetics, experiments are currently being conducted to find whether the method has general applicability to elements throughout the periodic table; whether the effective field is positive or negative in each specific case; and what the determining factors are. Since solubility of the subject element in iron (or such other ferromagnetic materials as may prove to be useful) is an essential condition, such experiments will have implications of general interest in metal physics. Development of the dynamic polarization method will bring with it much useful information in the realm of solid-state physics.

The development of the helium-3 cryostat has made available isothermal baths at temperatures down to 0.3 °K. Certain ions having very large nuclear and electronic magnetic moments can thus be used with readily available magnetic fields to yield useful degrees of nuclear polarization, which may be maintained as long as there is liquid helium in the cryostat. This will give a notable improvement in counting statistics for radioactive-decay experiments and in studies of the resonance absorption of neutrons and gamma rays.

It will be possible to study the scattering of polarized protons from polarized-proton targets when the dynamic method, applied to electron centers in hydrogen-rich materials, is perfected. The apparatus will probably incorporate helium-3 refrigeration. There are obvious problems in connection with getting the protons onto the target, and with minimizing the effect of stray field from the polarizing magnet upon the proton machine. So often in recent years, however, has the impossibility of today become the commonplace of tomorrow that such points are small cause for pessimism.

Notes

1. The terms anisotropy and asymmetry refer, respectively, to situations in which the emission is, or is not, symmetric with respect to the median plane (perpendicular to the *z*-axis). A beta-ray *anisotropy* is possible in the case of certain "forbidden" transitions.
2. This article was prepared in November 1960.

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