

## Letters to the Editor

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### Surface Paramagnetism of Germanium Films

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**I**N connection with a fundamental investigation of the physical and chemical properties of clean germanium surfaces, we have studied the *ortho-para* conversion of hydrogen when adsorbed on sputtered germanium films. The films were sputtered from germanium electrodes in an atmosphere of helium onto the walls of a glass container which was kept in a liquid nitrogen bath. We found a conversion at low temperatures which has all the characteristics of a magnetic spin inversion.<sup>1</sup> We attribute the effect to the presence of unpaired electrons in the atomically clean parts of the film surface. A rough estimate shows that there may be up to one unpaired electron per germanium surface atom.

With a thin film of germanium (about 100 Å thick), in a reaction vessel of about 100 cc volume, we found a conversion half-life of 20 hours at 77°K. When an H<sub>2</sub>+D<sub>2</sub> mixture was exposed to the same surface, no measurable amount of HD was found (<0.2%) after a contact time of 16 hours, indicating that no chemical mechanism was involved. This finding alone would not completely exclude the possibility that a chemical mechanism may be involved in the *para*-hydrogen conversion; the desorption of HD might be very slow compared with the H<sub>2</sub> desorption. This latter possibility is excluded by the observation that the *ortho*-deuterium conversion was fast too (about one-half as fast as the *para*-hydrogen conversion). Evaluation<sup>2</sup> of the temperature dependence and reaction order found leads to a reasonable value for the heat of molecular adsorption of hydrogen of 1.4 kcal/mole.

Cleaner and more active films were obtained by sputtering germanium onto a germanium coating. (The coating was produced by a very strong gas discharge which at the same time further cleaned the germanium electrodes.) The conversion kinetics in this case is not as simple as for thin films, but the fast conversion found at 77°K (half-life 13 minutes) is also magnetic in

origin. Addition of small amounts of oxygen reduced the reaction rate at any temperature; this shows that the activity of the films is not due to adsorbed oxygen. Owing to the complex structure of the films the quantitative determination of the magnitude of the surface paramagnetism is subject to uncertainties. From a comparison with previous results<sup>3</sup> we come to the rather striking conclusion that the conversion on our films is almost as fast as the conversion on intrinsically paramagnetic surfaces. This means that there would be roughly one unpaired electron per germanium surface atom.

This result perhaps does not necessarily mean that the surface is truly paramagnetic. The spins in a (111) surface, for example, may be so far apart from each other that even in case of an antiparallel orientation of two neighbors (in an antiferromagnetic array) the inhomogeneous field between the spins might cause a conversion of the observed magnitude.

<sup>1</sup> See A. Farkas, *Orthohydrogen, Parahydrogen, and Heavy Hydrogen* (Cambridge University Press, New York, 1935), pp. 79-96.

<sup>2</sup> Y. L. Sandler, *J. Chem. Phys.* **21**, 2243 (1953).

<sup>3</sup> Y. L. Sandler, *J. Phys. Chem.* **58**, 54 (1954); *Can. J. Chem.* **32**, 257 (1954).

### Noise Temperature Measurement on a Solid State Maser\*

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**N**OISE measurements have been made on a three-level,<sup>1</sup> 2800-Mc/sec solid state maser with sufficient accuracy to establish that its noise temperature does not exceed 20°K. The maser was operated as a reflection cavity amplifier at 1.25°K, with 9000-Mc/sec saturating power, and used K<sub>3</sub>Co<sub>0.995</sub>Cr<sub>0.005</sub>(CN)<sub>6</sub> as the mixed paramagnetic salt. The amplifier and oscillator characteristics are the subject of a forthcoming article.<sup>2</sup>

The maser noise temperature  $T_M$  was determined by measuring the noise outputs of the system,  $N_1$  and  $N_2$ , corresponding to two noise inputs with temperatures  $T_1$  and  $T_2$ , respectively. The output noise  $N_1$  is proportional to  $(T_1 + T_M)G_M + T_R$ , and  $N_2$  is proportional to  $(T_2 + T_M)G_M + T_R$ , where  $G_M$  is the gain of the maser and  $T_R$  is the noise temperature of the receiver following the maser.  $T_M$  is then given by

$$T_M = (T_2 - T_1)/(N_2/N_1 - 1) - T_1 - T_R/G_M. \quad (1)$$

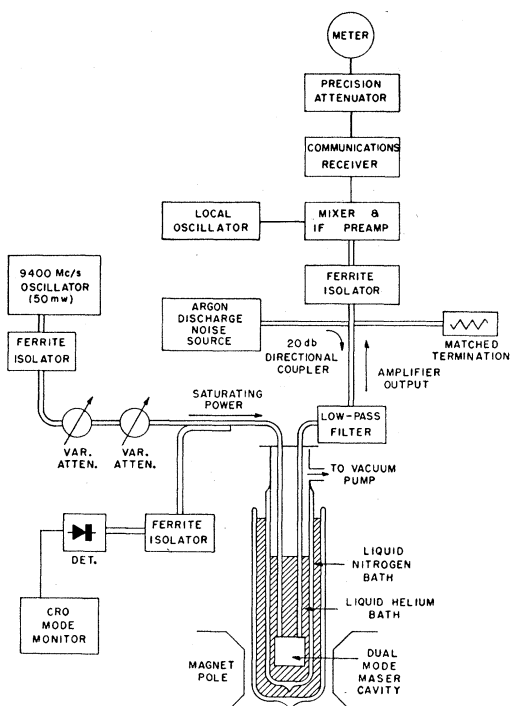


FIG. 1. Block diagram of microwave instrumentation.

Figure 1 shows the experimental arrangement which was used. Temperature  $T_1$  corresponded to the noise input with the argon discharge tube off, so that it was primarily the noise generated by the room temperature ( $30^\circ\text{C}$ ) ferrite isolator, while  $T_2$  was the total noise input with the argon tube energized. With 15 milliwatts of 9000-Mc/sec pumping power, enough to insure full saturation but not enough to cause heating of the salt, the external coupling at the amplifying frequency was adjusted to give a maser gain of 28 db with about 60-kc/sec band width. A communications receiver was used to provide a receiver band width small in comparison with that of the maser. The entire receiver had a noise temperature of about  $1800^\circ\text{K}$  (8.5 db noise figure). As a result of many measurements made with the precision attenuator by two different operators, the ratio  $N_2/N_1$  was determined to be  $1.125 \pm 0.025$  db.

The output of the argon discharge noise source through the directional coupler was calibrated by using a matched load, thermostated in an oil bath to  $\pm 0.1^\circ\text{C}$ ,

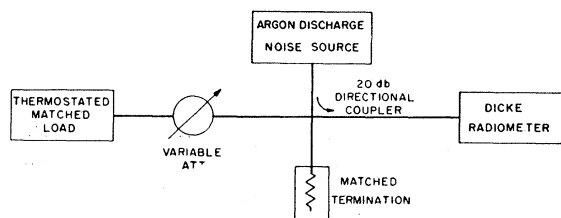


FIG. 2. Block diagram of calibration measurement.

and a Dicke radiometer in the arrangement shown in Fig. 2. The temperature of the thermostated load was adjusted until the radiometer output obtained with the variable attenuator set at zero and the argon tube off, equalled the output obtained with the attenuator fully in and the argon tube on. Knowing the temperature of the thermostated load and of the variable attenuator, one could then determine the noise output of the discharge tube by this essentially null type of measurement. At the frequency of the maser, the noise from the discharge tube through the coupler was  $99.5 \pm 1.5^\circ\text{K}$ .

In order to establish the noise temperature of the maser itself,  $T_1$  and  $T_2$  must be corrected for the attenuation in the input line and the thermal noise generated in this line. The input circuit after the directional coupler consisted of a wave-guide-to-coax coupling, a low-pass filter, and a section of coaxial cable, having a combined attenuation of 0.15 db at room temperature, followed by a 27-inch section of silver-plated stainless steel coaxial line leading down through the helium Dewar to the maser cavity. This line had an attenuation of 0.7 db at room temperature and 0.4 db under operating conditions. Since it was not known what temperature distribution corresponded to the 0.4-db attenuation, two extreme cases were calculated for the noise generated by the line. The most unfavorable case, from the standpoint of a low calculated value for  $T_M$ , is to assume that the entire attenuation takes place at  $0^\circ\text{K}$ . The most favorable case, consistent with the physical situation, was assumed to be a uniform temperature gradient over the entire length of the coaxial line, with the attenuation varying as the square root of the temperature. Upon using  $N_2/N_1 = 1.125$  db, and  $99.5^\circ\text{K}$  for the noise from the argon tube, Eq. (1) gives for the first case  $T_M = 8^\circ\text{K}$ , while for the second,  $T_M = -8^\circ\text{K}$ . When the uncertainties in the other quantities are taken into consideration, the calculated value of  $T_M$  is  $0 \pm 19^\circ\text{K}$ . Hence an upper limit of about  $20^\circ\text{K}$  can be established for the maser noise temperature.

If one neglects the small thermal radiation from the cavity walls, spontaneous emission at the amplifying frequency should be the only source of noise in the maser.<sup>3</sup> Using Bloembergen's notation,<sup>1</sup> the theoretical noise temperature under fully saturated conditions would then be

$$T_M = T_B \frac{\nu_{32}(w_{21} + w_{32})}{w_{21}\nu_{21} - w_{32}\nu_{32}},$$

where  $T_B$  is the helium bath temperature. Assuming the  $w$ 's are equal, we have  $T_M = 2^\circ\text{K}$ .

The  $20^\circ\text{K}$  uncertainty is a result of three main contributions, all approximately equal: the measurement of  $(N_2/N_1 - 1)$ , the calibration of the argon discharge noise source, and the uncertainty in the noise generated by the input line. To obtain the best accuracy in the measurement of  $T_M$ , one should use noise inputs

comparable with the maser noise, but the present necessity of using a room-temperature isolator, which gives a minimum noise input of about 300°K, prevented this. Work is now in progress to develop a good room-temperature circulator or a low-temperature isolator. Such circuit elements, and the subsequent use of low-temperature noise sources, would also permit the generation of noise temperatures  $T_1$  and  $T_2$  by thermostated matched loads at accurately known temperatures, thus eliminating the calibration of a secondary standard. In the masers now being designed, the replacement of the coaxial input line with a wave guide will eliminate most of the input attenuation and hence the uncertainty arising from noise generation in the input circuit. These improvements should reduce the uncertainty in the measurement of  $T_M$  to about 5°K.

We wish to thank Professor L. D. Smullin for the use of his Dicke radiometer in the calibration of the argon tube.

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<sup>1</sup> N. Bloembergen, Phys. Rev. **104**, 324 (1956).

<sup>2</sup> A. L. McWhorter and J. W. Meyer, Phys. Rev. (to be published).

<sup>3</sup> R. V. Pound, Ann. Phys. **1**, 24 (1957); M. W. Muller, Phys. Rev. **106**, 8 (1957); M. W. P. Strandberg, Phys. Rev. **106**, 617 (1957); Shimoda, Takahasi, and Townes, J. Phys. Soc. Japan **12**, 686 (1957).

## Resonant Absorption of Gamma Rays\*

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IN recent years several ingenious methods<sup>1-5</sup> have been used to observe the resonant absorption or fluorescence when nuclei are irradiated by gamma rays. In this investigation we have used in essence a source of gamma rays with a discrete but variable energy to observe the resonant absorption by a direct measurement of the transmission through an absorbing sample.

In the radiative capture of particles of fixed energy and direction of motion, the gamma rays emitted at a given angle are monoenergetic. Moreover, the energy of the radiation varies smoothly with the angle of emission. The energy of the incident particles is fixed either by using a resonant capture process, or, if the capture is nonresonant, by employing a monoenergetic beam of particles and a thin target. It is not at all necessary<sup>4</sup> to use a reaction which is the inverse of the subsequent resonant absorption. It is usually convenient to do so, however, since the available variation in energy is limited but, in the case of reciprocal reactions, sufficient to cover the increase in energy needed to compensate for the energies lost to the recoiling nuclei in emission and in absorption.

The source of radiation was the reaction  $C^{13}(p,\gamma)N^{14}$  at the resonant energy  $E_p=1.76$  Mev corresponding to the level in  $N^{14}$  at  $E_{ex}=9.18$  Mev. The 9.18-Mev radiation was detected at an angle  $\theta$  through a slit of angular width  $\Delta\theta$ , with a NaI counter and a 10-channel analyzer. The inverse absorption  $N^{14}+\gamma\rightarrow N^{14*}$  was obtained by interposing between the source and the slit a quantity of liquid nitrogen, 27.0 cm thick, contained in two Dewar flasks. The energy of the emitted radiation was then varied by changing the angle of observation with the results given in Fig. 1. Data are presented for three different widths of the slit. The measurements at the top were obtained with water instead of liquid nitrogen in the Dewar flasks.

The number of gamma rays emitted at the angle  $\theta$  with energy in the interval between  $E$  and  $E+dE$  will be represented by the single-level formula

$$N(E,\theta) = \frac{N_R}{[2(E-E_R)/\Gamma]^2 + 1},$$

where  $\Gamma$  is the intrinsic width of the level and the resonant energy  $E_R = E_0 - (E_0^2/2Mc^2) + (E_0v/c)\cos\theta$ , to a good approximation. The energy  $E_0$  is the unmodified energy of the transition, and  $v$  is the speed and  $M$  the mass of the radiating compound nucleus. The second term in the expression represents the energy of

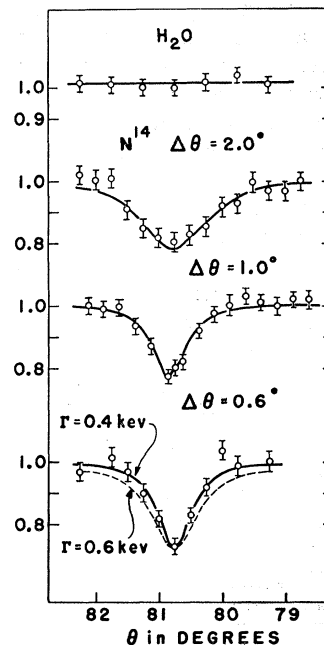


FIG. 1. Resonant absorption in the process  $N^{14}+\gamma\rightarrow N^{14*}$  ( $E_{ex}=9.18$  Mev), using gamma rays from the inverse reaction  $C^{13}(p,\gamma)N^{14}$ . The abscissa represents the angle of emission of the radiation. One degree corresponds to a change of 0.7 keV in the energy of the radiation.

recoil in the emission, and the last term gives the shift in energy due to the motion imparted to the compound nucleus in the capture of the incident particle.

We may neglect the purely electronic absorption, which, although large, is nevertheless essentially con-