2900-104-R

Memorandum of Project MICHIGAN

OPTICAL EFFECTS ON F-CENTER SPIN RESONANCE AT LOW TEMPERATURES

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November 1959

SOLID-STATE PHYSICS LABORATORY

Willow Run Laboratories

THE UNIVERSITY OF MICHIGAN

Ann Arbor, Michigan

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PREFACE

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Robert L. Hess
Technical Director
Project MICHIGAN

ABSTRACT

Because of possible maser applications, the interaction of light with spin systems has been of considerable interest. In this connection, experiments have been carried out to study the effect of light on the spin resonance of F-centers. These experiments were done at liquid-helium temperatures because, at this temperature, the F-center has a long spin-lattice relaxation time. It was found that the spin population could be significantly affected by optical illumination. It is possible that such an effect could be useful in pumping a three-level maser system from an optically "heated" two-level spin system.

1
INTRODUCTION[1]

The subject of the interaction of light with spin systems has been of considerable interest because of possible maser applications. Effects which come under the general heading of "optical pumping" may permit the use of light sources as the pump in maser systems. Although such effects have been extensively studied in gases, the work in solids is much less extensive.

This memorandum contains a discussion of measurements made on a system consisting of F-centers in KCl. Such centers have well established spin-resonance and optical properties. Of special interest is that the F-center has very strong optical absorption and a long spin-lattice relaxation time at 4°K. A value of 15 sec has been reported (Ref. 1).

Basically, the objective is to alter the spin temperature of the F-centers by irradiation with F-band light. This is a crude but simple way to see whether optical pumping will occur. It should be noted that the optical excited states are very broad in energy, thereby making the more elegant type of experiments, such as are carried out in sodium vapor, unfeasible. Thus it is doubtful that one could optically invert the spin population in the F-center case.

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[1]A speech presenting the material of this memorandum was presented at the International Conference on Quantum Mechanics, High View, New York, 14-16 September 1959.

The question of the nature of the excited electronic states of the F-center is of fundamental importance in determining whether spin-flips can occur in the optical excitation process. This is dependent on the mixing of spin states in the excited state. The usual procedure has been to consider the first excited state as a p state, and on this basis calculations of the energy of this state have been made. No calculations appear to have been made on the spin-orbit coupling in the excited state. For these purposes a situation analogous to an alkali-metal atom with broadened energy levels is assumed. On this basis, significant spin-optical interaction would exist.

In addition to the occurrence of spin-flipping during optical transitions, there is the problem of what happens to the spin of the F-center due to the local "heating" of the F-center; that is, the F-center absorbs 2.5 ev of energy and emits about 1 ev as light (Ref. 2). The rest of the energy is coupled to the lattice surrounding the F-center. The surrounding atoms vibrate quite strongly, and the F-center finally cools off. This process may increase the temperature of the spin system. Since little is known about such a process, the present type of measurement may aid in understanding such effects.

2
EXPERIMENTAL METHOD

The apparatus used consisted of a magnetic resonance spectrometer operating at about 9400 mc and an arc light source with appropriate filters to select a band of radiation in the F-center wavelength region.

The microwave cavity had the dimensions of standard X-band waveguide and was operated in the TE_{012} mode. The cavity was made of a ceramic type of material which was silver coated. A window was cut in the broad face of the cavity to permit illumination of the sample (Fig. 1).

It was necessary to use superheterodyne detection, since small microwave power was used to minimize saturation problems. The power level used was between 0.05 and 0.005 μw. Although this was still somewhat excessive, reasonable data could be obtained under these conditions.

Crystals used were of commercially available KCl, which were cleaved to about 3-mm thickness and 1 cm on a side. These crystals were x-rayed for about an hour to produce a peak F-band optical density of about 1.2 at room temperature. The crystals were kept in the dark or in red light until immersion in liquid helium. This was necessary in order to avoid growth of M- and R-bands which are known to effect the F-center luminescence.

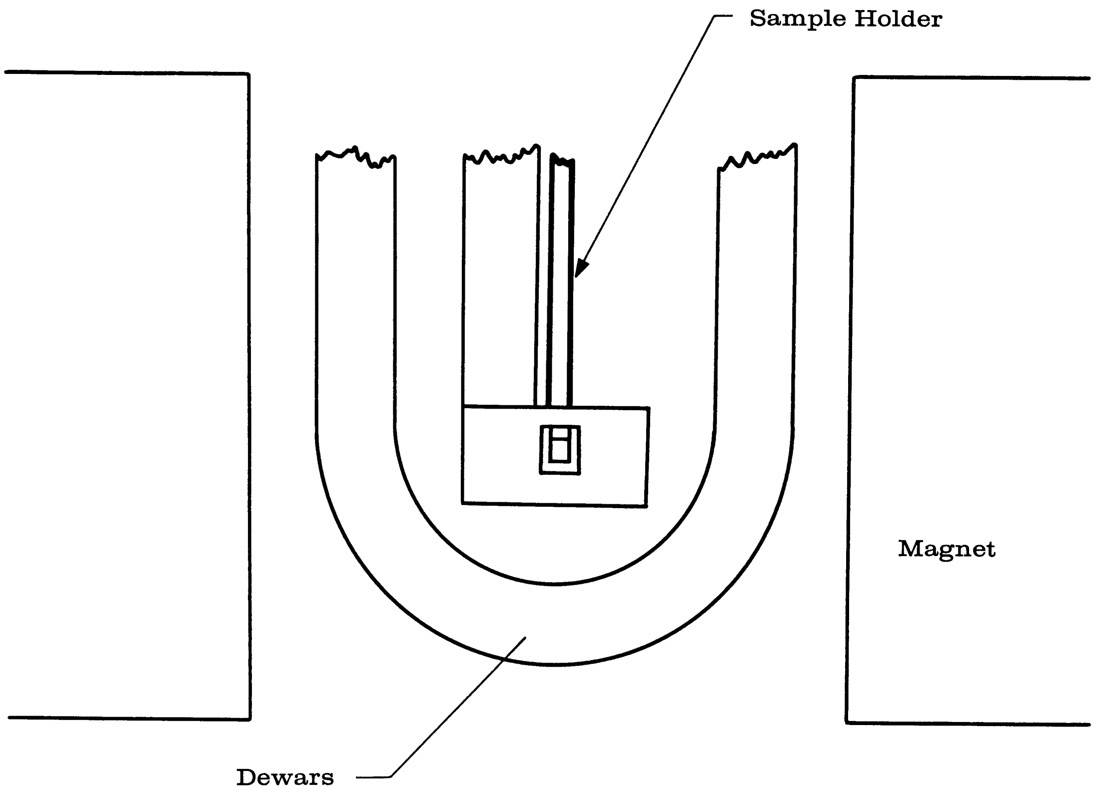

FIG. 1. APPARATUS USED FOR OPTICAL SPIN RESONANCE EXPERIMENT. Cavity and sample are immersed in liquid helium. Dewar system is slit silvered to permit sample illumination. The cavity is shown with the connecting waveguide.

The crystals were then put into the cavity, which was in the liquid-helium dewar, so that the crystals were completely immersed in liquid helium. The spin resonance signal was then observed with light on and off the crystal.

A Zr arc used together with the optical system gave an illumination intensity of about 1 mw/cm^2 at the crystal. The light had to pass through the dewar system (the dewars were slit silvered) and onto the crystal.

3
EXPERIMENTAL RESULTS

The first runs were made simply to observe the spin resonance of the F-center. At this point it was noted that numerous other spin-resonance signals were present from the sample. A typical spectrum is shown in Fig. 2. The F-center resonance is distorted due to saturation effects. The other lines are present in the KCl even before x-raying. These signals were found in commercially available KCl from 3 different sources. The origin of this spectra remains unknown, but these lines serve the useful purpose of a lattice thermometer for the experiment. The unknown center has a spin-lattice relaxation time which is much shorter than the F-center so it can function as a thermometer for the lattice.

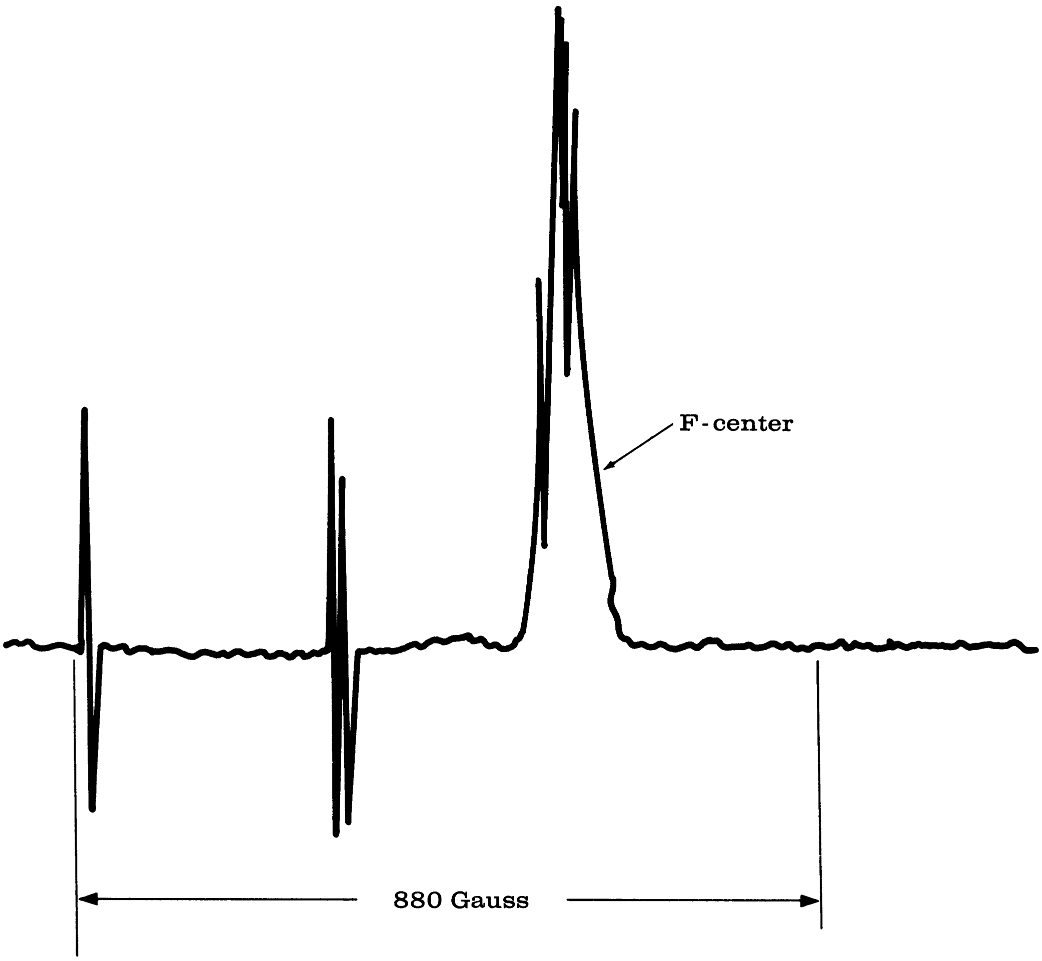

FIG. 2. TYPICAL KCl SPECTRA AT 4.2°K. The F-center absorption is distorted. The other narrow line spectra is angular dependent.

The effect of light on the crystal is shown in Fig. 3. Here the microwave bridge was adjusted to observe dispersion signals. The F-center signal was distorted, but was still useful as a population measurement. It was observed that the F-center resonance signal decreased by about 50%. The "thermometer signal" remained essentially unaffected. From this it was concluded that the spin population of the F-center system had been altered. The effect was equivalent to a 4°K increase in temperature.

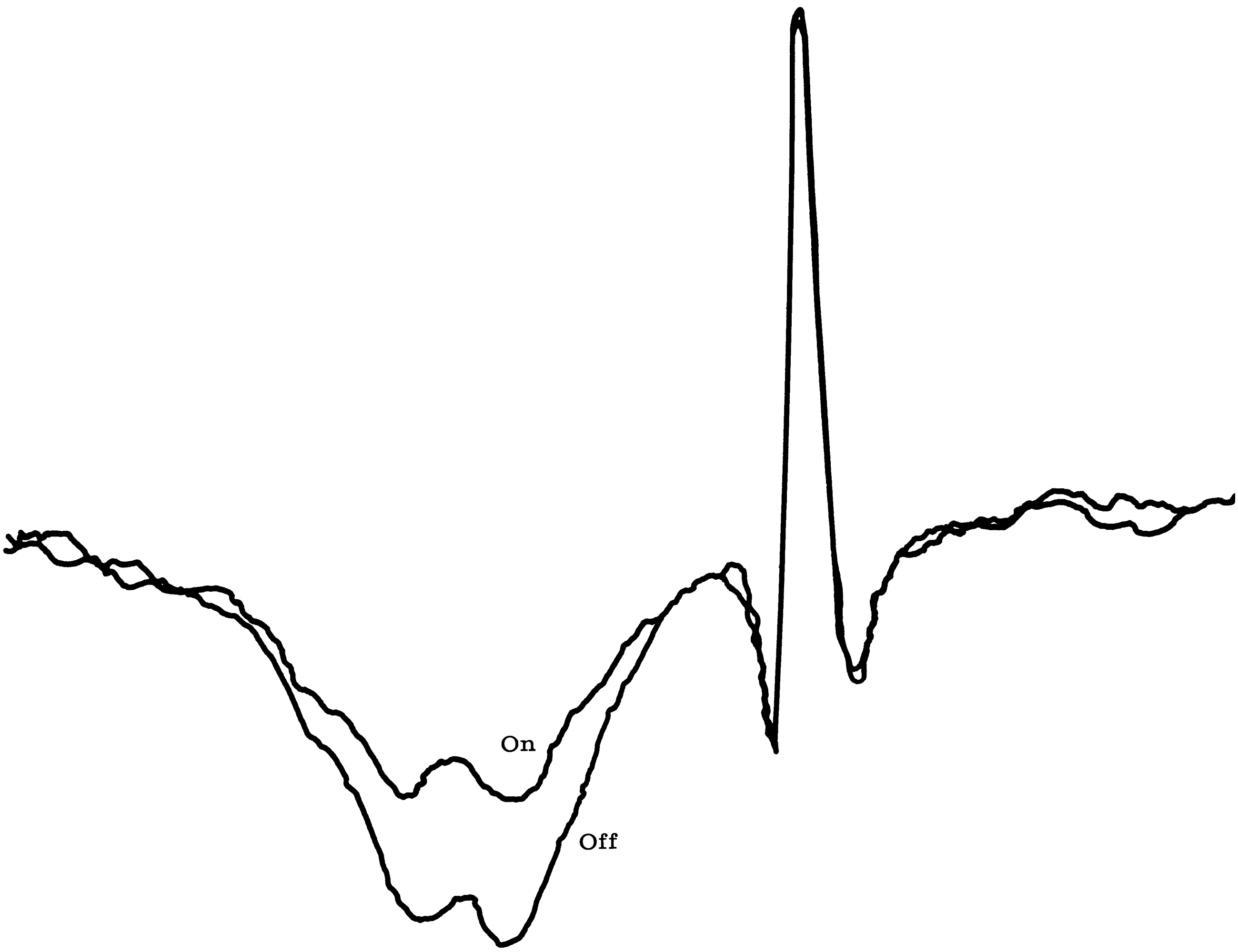

FIG. 3. DISPERSION SIGNAL OBTAINED WITH LIGHT ON AND LIGHT OFF. The signal to the high-field side of the F-center is used as a thermometer.

After the light was turned off, the F-center resonance returned to its original form. About 1 min was required to remove all trace of the light effect. On the basis of the long spin-lattice relaxation time of the F-center, this was to be expected.

4
DISCUSSION

As reported in the foregoing sections, an effect on the F-center spin-resonance signal was observed. It is of interest to discover what a crude calculation would predict as to the expected magnitude of the effect.

The system under discussion contains about 10^{16} F-centers, and was absorbing 1 mw of green light. This means about 2×10^{15} quanta per second are absorbed. The situation can be analyzed as follows.

Let n_1 = number of spins in lower state

n_2 = number of spins in upper state

F = rate of spin flipping by light per center.

Then
$$\frac{dn_1}{dt} = -Fn_1 + Fn_2 - w_{12}n_1 + w_{21}n_2,$$

and
$$\frac{dn_2}{dt} = -Fn_2 + Fn_1 + w_{12}n_1 - w_{21}n_2.$$

Let $\Delta n = n_1 - n_2$

τ = spin-lattice relaxation time (15 sec).

This yields
$$\frac{d\Delta n}{dt} = -2F\Delta n - \frac{1}{\tau}(\Delta n - \Delta n_0).$$

An estimate of F is given by
$$F = \frac{2 \times 10^{15} \text{ photons/second}}{10^{16} \text{ centers}} \times \frac{1}{2}.$$

The factor of 1/2 is a statement of the assumption that the optically excited center has equal probability of going back to either spin state. Then
$$\frac{d\Delta n}{dt} = -\frac{1}{5}\Delta n - \frac{1}{15}(\Delta n - \Delta n_0);$$

at equilibrium $d\Delta n/dt = 0$, so that
$$\Delta n = \frac{1}{4}\Delta n_0.$$

Thus the observed effect is of the proper order of magnitude. However, the data is not good enough to provide a severe test of the assumed model.

The local heating effect may also be contributing to the observed spin temperature effect. An interesting experiment would be to see if the nuclear spin temperature is affected by the fact that nuclei are set in rapid motion in the neighborhood of the paramagnetic center. In this way more information might be obtained on local heating around the F-center.

It should be pointed out that there is little hope of optically inverting the F-center spin system. Yet the fact that a spin system can be heated optically has an important implication for optical pumping in solids. A very interesting system could be achieved by placing such a heated spin system in contact with a three-level maser system. By matching levels 1 and 3 to the hot two-level system, cross relaxation effects could tend to saturate between levels 1 and 3. Maser action could then be achieved between levels 2 and 3. This would be of special interest in cases where the separation between 1 and 3 is very large, since in this case, optical "spin heating" becomes more feasible. Thus optical "spin heating" may be used where the usual optical pumping inversion techniques are not applicable.

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(Memorandum no. 2900-104-R)
(Contract DA-36-039 SC-78801) Unclassified memorandum

Because of possible maser applications, the interaction of light with spin systems has been of considerable interest. In this connection, experiments have been carried out to study the effect of light on the spin resonance of F-centers. These experiments were done at liquid-helium temperatures because, at this temperature, the F-center has a long spin-lattice relaxation time. It was found that the spin population could be significantly affected by optical illumination. It is possible that such an effect could be useful in pumping a three-level maser system from an optically "heated" two-level spin system.

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