THE OVERHAUSER EFFECT AND THE ELECTRON PARAMAGNETIC RESONANCE "SECONDARY SIGNAL"

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Submitted to JETP editor July 8, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 40, 32-33 (January, 1961)

The relative increase in the polarization of the protons in a free radical as the electron paramagnetic resonance is saturated has been measured by observing the increase in the nuclear magnetic resonance signal. It is shown that at large saturating power an electron paramagnetic resonance "secondary signal" is observed simultaneously with the nuclear magnetic resonance signal, corresponding to the emission or absorption of an additional quantum by the electron spin system.

WE have observed dynamic nuclear polarization (d.n.p.) in solid diphenylpicrylhydrazyl at room temperature in fields of approximately 3300 oe. The apparatus described in reference 1 has enabled us to measure the d.n.p. of the protons by means of the relative amplitude of the nuclear magnetic resonance (n.m.r.) signal on an oscillograph screen.

The coil with the sample containing 30 mm^3 of diphenylpicrylhydrazyl placed into a teflon container was situated within a rectangular three centimeter resonant cavity excited in the H_{102} mode. The sample was cooled by a stream of air. The amplitude of the high frequency magnetic field H_1 was determined from the power dissipated in the resonator. The position of the coil in the resonant cavity and its dimensions are such that there should be no significant variations in the magnitude of the high frequency magnetic field over the volume of the sample where H_1 is perpendicular to the axis of the coil, and this is confirmed by the observation of the electron paramagnetic resonance (e.p.r.) signal with the coil present in the cavity.

The dependence on H_1 of the increase in the n.m.r. signal is shown in Fig. 1. The accuracy of determining H_1 and the relative increase in the signal amounts to approximately 10%. As an example, we show in Fig. 2 an oscillogram of the n.m.r. signal in the absence of the high frequency field. In order to obtain reliable results at higher values of the high frequency power and field in the resonator we shall need more intensive cooling, since starting with $H_1 > 2$ oe appreciable heating begins to be observed, while for $H_1 > 3$ oe partial melting of the sample is observed, as can be inferred from the n.m.r. line.

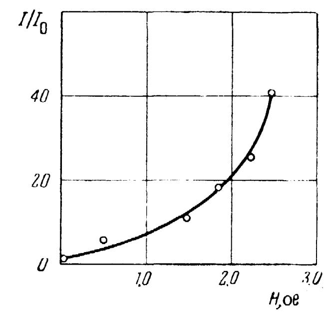

FIG. 1. Dependence of the relative increase in the nuclear magnetic resonance signal due to protons in diphenylpicrylhydrazyl on the amplitude of the high frequency magnetic field saturating the electron paramagnetic resonance.

We have also observed at $H_1 > 1.5$ oe the "double quantum emission" effect (9300 and 14 Mc/sec) in e.p.r. described by Winter.[2] This effect occurs when the electron spin system, situated in a constant magnetic field H_0, is simultaneously acted upon by electromagnetic radiation of frequency ω_1 and another radiation of frequency ω_2. The condition for the appearance of a "secondary signal" at the frequency ω_2 is $\omega_1 \pm \omega_2 = \gamma_e H_0$. Fig. 3 shows an oscillogram of the signal which

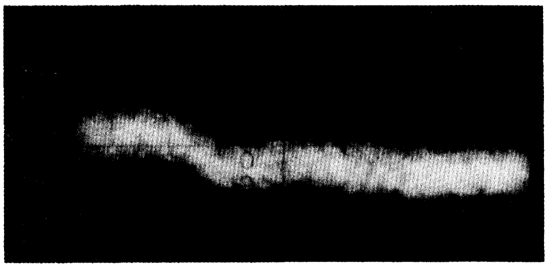

FIG. 2. Oscillogram of the nuclear magnetic resonance signal due to protons in solid diphenylpicrylhydrazyl. Amplification K = 30.

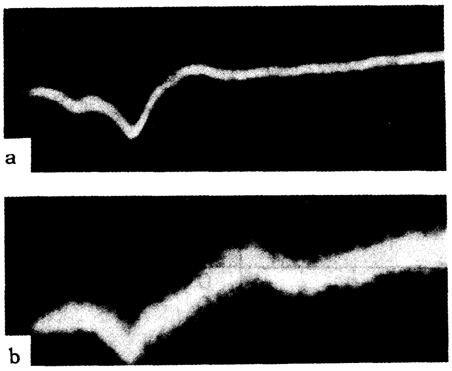

FIG. 3. a – Signal representing the superposition of the nuclear magnetic resonance signal (middle peak) and of the electron paramagnetic resonance "secondary signal" (outer peaks). Amplification K = 1 (the signal is applied to the oscillograph plates), $H_1 = 2.5$ oe; b – Electron paramagnetic resonance "secondary signal," $H_1 = 2.2$ oe, amplification K = 3.

represents the superposition of an n.m.r. signal with a maximum in the field H_0 and of an e.p.r. "secondary signal" at the same frequency ν_n which consists of the absorption and emission signals displaced respectively by $\pm \omega_n/\gamma_e = 5$ oe from the field H_0. The detuning of the autodyne n.m.r. detector by several tens of kilocycles does not change the shape and the position of the e.p.r. "secondary signal", but sharply affects the amplitude (and, of course, the position) of the n.m.r. signal.

In our opinion, the signals described in reference 3 represent a superposition of an n.m.r. signal and an e.p.r. "secondary signal", with the n.m.r. signal displaced from the maximum value corresponding to the Overhauser effect.

I wish to express my gratitude to Professor E. I. Kondorskiĭ and E. S. Goryunov for active participation in this work.

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[2] I. M. Winter, J. phys. radium **19**, 843 (1958).

[3] You Hing-Tchao and J. Herve, Compt. rend. **250**, 700 (1960).

Translated by G. Volkoff
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