$$\operatorname{Re}\sigma(\omega)=\frac{\pi}{\omega}|\langle j\rangle|^2\exp\left\{-\left(\frac{\mu}{E_0}\right)^{1/2}L(\mu)\right\}\cdot$$

$$\times\exp\left\{-\frac{2}{3}\left(40\frac{{}^3/_4\mu-\hbar\omega}{\mu}\right)^2L^2(\mu)\right\}. \quad (45)$$

At $\Delta E_1>(\overline{\Delta E_2^2})^{1/2}$ and at frequencies lower than ω_{01} the line shape is also described by (45). For frequencies higher than ω_{01} the drastically varying quantity is the last exponential factor in (44), which takes into account the broadening due to the deviation of the shape of the fluctuation from spherical. The absorption line shape duplicates in this case the variation of the state density

$$\operatorname{Re}\sigma(\omega)=\frac{\pi}{\omega}|\langle j\rangle|^2\exp\left\{-\left(\frac{4\hbar\omega}{3E_0}\right)^{1/2}L\left(\frac{4}{3}\hbar\omega\right)\right\}. \quad (46)$$

In a region far from the center of the absorption line, at $E_2>1/4E_1(\omega<\omega_{01})$ we have

$$\operatorname{Re}\sigma(\omega)=\frac{\pi}{\omega}|\langle j\rangle|^2\exp\left\{-\left(\frac{\mu}{E_0}\right)^{1/2}L(\mu)\right\}$$

$$\times\exp\left\{-40\frac{[{}^3/_4\mu-\hbar\omega]}{\mu}L(\mu)\right\}, \quad (47)$$

and at $\omega>\omega_{01}$

$$\operatorname{Re}\sigma(\omega)=\frac{\pi}{\omega}|\langle j\rangle|^2\exp\left\{-\left(\frac{\mu}{E_0}\right)^{1/2}L(\mu)\right\}\exp\left\{-80\frac{[\hbar\omega-{}^3/_4\mu]}{\mu}L(\mu)\right\}. \quad (48)$$

It follows from these equations that the optical-absorption line shape should be asymmetrical on the wings and have a slower decrease on the long-wave side. The line width is small compared with the level spacing in accord with the parameter $(E_1/E_2)^{1/4}L(E_1)\gg 1$. For this reason we can assume that higher excited states will also have a small width and will appear in the absorption spectrum in the form of a hydrogenlike series.

For experimental observation of the absorption spectra connected with transitions inside the small-scale fluctuations it is necessary to satisfy a large number of rather stringent conditions. The formulas obtained are valid when the depth of the ground-state level is much less than the width of the forbidden band E_g. On the other hand, the small-scale fluctuations of the multiply charged atom type determine the state density far enough from the edge of the forbidden band. It follows from (3) that at $Na^3\approx 0.1$ and $T_0/E_0\approx 10$ the Fermi level passes through an energy interval in which the state density is controlled by the small-scale fluctuations at a degree of compensation $1-k<10^{-3}$. The concentration of the small-scale fluctuations, which determines the absorption coefficient, is then of the order of $(1-k)N\approx 10^{-3}N$. Since the coefficient of light absorption by the fluctuation wells is close to the absorption coefficient of shallow impurities having the same concentration, observation of this spectrum calls for measurement apparatus of high sensitivity. In semiconductors whose donor activation energy lies in the submillimeter band, such measurements are feasible by using submillimeter technology.[4]

The authors than A. L. Éfros for helpful discussions.

[1]B. I. Shklovskiĭ and A. L. Éfros, Élektronnye svoĭstva legirovannykh poluprovodnikov (Electronic Properties of Doped Semiconductors), Nauka, 1979.
[2]I. M. Lifshitz, Zh. Eksp. Teor. Fiz. **53**, 743 (1967) [Sov. Phys. JETP **26**, 462 (1968)].
[3]L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics), Part 1, Nauka, 1976 [Pergamon].
[4]E. M. Gershenzon, G. N. Gol'tsman, and N. G. Ptitsina, Zh. Eksp. Teor. Fiz. **64**, 587 (1973) [Sov. Phys. JETP **37**, 299 (1973)].

Translated by J. G. Adashko

Dynamic characteristics of EPR signals of saturating systems

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(Submitted 12 September 1979)
Zh. Eksp. Teor. Fiz. **78**, 1237–1239 (March 1980)

An EPR investigation of the dynamics of the saturation of spin systems has revealed that, in contrast to the Torrey model, the frequency of the oscillations of the absorption signal does not depend on the detuning from resonance, and that there are no oscillations of the dispersion signals. The results are explained by taking into account the nonequilibrium acoustic waves excited in the sample by the radio-frequency field.

PACS numbers: 76.30. – v, 72.50. + b, 43.35.Qv

Direct investigations of the dynamics of saturation of spin systems are carried out by observing, in time scale, the establishment of their quasistationary state under the influence of radio-frequency radiation after a jumplike establishment of resonance conditions. A transition process, frequently called Torrey oscillations or transient nutations, can then be observed.[1-4] This effect is connected with the nutational motion of the magnetic-moment vector, which manifests itself in oscillations of the dispersion (χ') and absorption (χ'')

 0038-5646/80/030623-03$02.40

signals with a frequency

$$\Omega=[(\gamma H_1)^2+(\omega_0-\omega)^2]^{1/2}, \quad (1)$$

where γ is the gyromagnetic ration, H_1 is the intensity of the magnetic component of the radio-frequency field of frequency ω, $\delta\omega = \omega_0 - \omega$ is the detuning from resonance. In this case the time dependence of χ' and χ'', with allowance for the relaxation processes, can be determined analytically by solving the Bloch equations.[1]

The conclusions of this model were partially confirmed by Torrey, who was the first to observe oscillations in NMR.[1] When this effect was observed in EPR, the experimental results differed from those expected, and the deviations were attributed either to inaccuracy in the solution of the Bloch equation, or to instrumental effects.[2] In the present paper we report observation of fundamental inconsistencies in the existing model when it comes to saturation of spin systems.

The investigations were performed with experimental installations in which the EPR were registered in a time scale. In contrast to Refs. 1–4, the jumplike establishment of the resonant conditions was effected by varying the polarizing magnetic fields, with the microwave power constant. The preliminary results were obtained with a setup using a homodyne microwave channel. Their fundamental difference from the published results, and also the difficulty of monitoring the separation of the absorption and dispersion signals, made it necessary to repeat these experiments with an improved setup using a superheterodyne microwave circuit. This setup made possible exact separation of the signals and monitoring of the separation with the aid of a ruby crystal placed together with the investigated sample and remaining practically unsaturated under the experimental conditions. This ensured simultaneous registration of the absorption and dispersion signals. Microwave power up to 40 mW at a frequency 10 GHz was fed to the measuring resonator. The H_1 component of the microwave field in the sample was determined from the values of the power fed to the resonator and from the Q of the loaded resonator. The measurements were made at a fixed value of the dc component of the polarized magnetic field, which was modulated by rectangular pulses with approximate amplitude 2 Oe, duration 5 μsec, and rise and fall times 0.2 μsec.

We investiaged E_1' centers produced in crystalline quartz by neutron bombardment and serving as a suitable object for the production of the oscillations.[5,6] The concentration of these centers, determined by comparison with a calibrated DPPH sample, was approximately 2×10^{17} spins/cm^3. The sample was so oriented that its spectrum consisted of three lines of approximate width 0.3 Oe, spaced 1 Oe apart. The temporal oscillograms of the dispersion signals (a) and of the absorption signals (c), obtained for these centers at room temperature, are shown in the figure. The signals were obtained for one and the same point of the line at $\delta\omega$ corresponding to the maximum dispersion signal. The oscillogram b was registered in the dispersion-signal channel when the spin system was off resonance. The drift of the null line of oscillograms

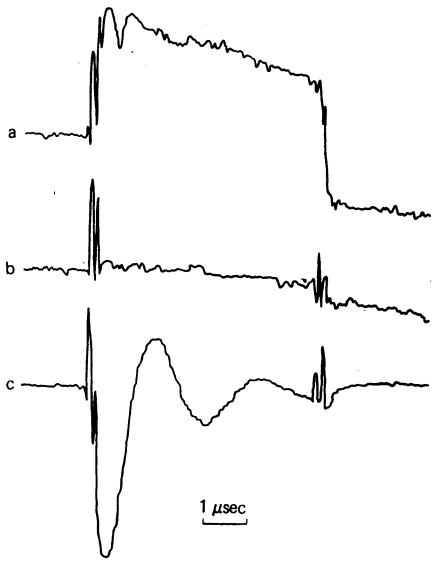

FIG. 1.

a and b is due to the change in the frequency of the working resonator on account of the synchronous shift of the modulation loops inside the resonator when the current pulses were passed. The inflection on the initial section of the dispersion-signal pulse is a consequence of the non-uniformity of the top of the modulating pulse. It is seen that the dispersion signal does not oscillate in the section with the existing model.

An investigation of the absorption signals shows that the frequency of the oscillations is equal to γH_1 and, in contrast to Eq. (1), does not depend on $\delta\omega$. Thus, in our case, at exact resonance, it is equal to 0.45 MHz and should amount to 0.62 MHz at a detuning 0.15 Oe from resonance. At the same time, no change in this frequency, with accuracy not worse than 3%, was observed on any line of the investigated spectrum.

We note that when the microwave power is changed the amplitude of the nonequilibrium signals on the stationary sections coincides with the curves of the translational saturation, obtained by using video registration without modulation, and agrees with the experimental data of Portis.[7] The difference between the presented dispersion signal and that given in Ref. 2 is attributed by us to the use, in the latter, of sinusoidal modulation of the magnetic field, which led to a mixing of the dispersion and absorption signals. Similar results were obtained for the paramagnetic centers of heat-treated 5-phenolresorcin-formaldehyde polymer and for $P3$ centers in silicon (T= 77°K).

The obtained data can be interpreted in the following manner. Since the absorption is proportional to the population difference of the spin levels, it follows, as shown by us earlier,[6] that the oscillations constitute an inversion of a two-level system (a two-level maser). This process is the result of alternating accumulation of the absorbed electromagnetic energy in the form of coherent acoustic waves (absorption), and the reaction

of the latter on the spin system with conversion of their energy again into electromagnetic energy (amplification). It is possible only when the microwave power absorbed by the spin system does not manage to become irreversibly dissipated by relaxation mechanisms, and is preserved in the sample in the form of resonant phonons during a time longer than $1/\gamma H_1$. The period of the oscillations is therefore independent of the detuning but is determined only by the pumping rate. At the same time, the dispersion, being a phase characteristic of the resonant system (by virtue of the conservation, at each instant of time, of the number of spin transitions produced by the electromagnetic and nonequilibrium acoustic fields), should be constant both in the absorption regime and in the regime of amplification of the exciting radiation. The result is a linear dependence of the dispersion signal amplitude on the radiofrequency field.

[1]H. C. Torrey, Phys. Rev. **76**, 1059 (1949).
[2]P. W. Atkins, A. J. Dobbs, and K. A. McLachlan, Chem. Phys. Lett. **25**, 105 (1974).
[3]P. W. Percival and J. S. Hyde, Rev. Sci. Instrum. **46**, 1522 (1975).
[4]G. Berthel, N. P. Blanc, J. Gallice, H. Robert, C. Thibaud, J. M. Fabre, and L. Giral, Sol. St. Commu. **22**, 251 (1977).
[5]D. A. Bozanic, D. Mrgerian, and R. W. Minarik, Phys. Rev. Lett. **21**, 541 (1968).
[6]I. Z. Rutkovskii and G. G. Fedoruk, Dokl. Akad. Nauk Belorus. **22**, 127 (1978).
[7]A. M. Portis, Phys. Rev. **91**, 1071 (1953).

Translated by J. G. Adashko