

the tricritical point. In this case, however, it must be assumed that both  $B$  and  $C$  are "accidentally" systematically small compared with the coefficient of  $Q^6$ .

<sup>7</sup>We note that Filev<sup>23</sup> has proposed a method of measuring the exponent  $\gamma$  by an optical procedure, with which it is possible to distinguish between the behavior near the isolated point and the tricritical point.

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## $\mu^+$ -meson spin relaxation in rare earth metals at various temperatures

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We measured the temperature dependences of the rates of relaxation of the  $\mu^+$ -meson spin in Pr, Nd, Sm, Eu, Tb, Dy, Ho, and Er, at  $T = 5-300$  K. We demonstrate the possibility of identifying antiferromagnetic phase transitions and of measuring the Néel temperature  $T_N$  by the  $\mu^+$ -meson method. The value of  $T_N$  of praseodymium measured by the  $\mu^+$  method was found to be 6 K. A method is proposed for measuring the frequency of the oscillations of the electron spin of the atoms of a metal in the paramagnetic state.

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The spin of a  $\mu^+$  meson in a metal relaxes because of the interaction of the magnetic moment of the  $\mu^+$  meson with the magnetic moments of the surrounding electrons and nuclei. A study of these interactions is of interest both for the investigation of the properties of a singly charged impurity particle in a metal and for the investigation of the properties of the metal itself. An example of a study of ferromagnets (iron, nickel, cobalt and gadolinium) with the aid of  $\mu^+$  mesons is Ref. 1.

In this paper we study the relaxation of the  $\mu^+$ -meson spin in rare-earth metals. The metals of this group, depending on the temperature, can be in a paramagnetic,

antiferromagnetic, and ferromagnetic state. We used polycrystalline samples of metals with less than 0.2% impurities. The relaxation rate  $\Lambda$  of the spin of the  $\mu^+$

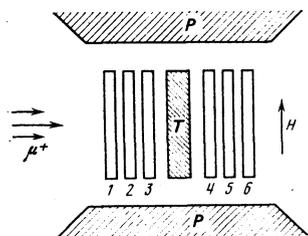


FIG. 1. Experimental setup: T—target, P—poles of electromagnet, 1-6—scintillation counters.

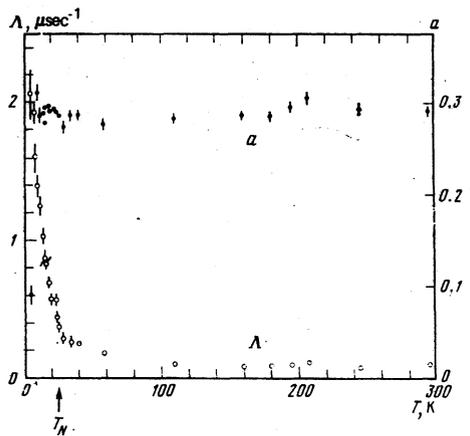


FIG. 2. Temperature dependences of the relaxation rate  $\Lambda$  and of the precession amplitude  $a$  of the  $\mu^+$ -meson spin in praseodymium.  $T_N$ —Néel temperature of the transition into the antiferromagnetic state, measured by the neutron diffraction method.<sup>3</sup>

meson was determined from the damping of the spin-precession amplitude of the  $\mu^+$  meson in a transverse magnetic field  $H = 300$  Oe. Individual results of the described experiments were published before.<sup>2</sup>

The setup for the observation of the  $\mu^+$ -meson precession is shown schematically in Fig. 1. The longitudinally polarized  $\mu^+$  mesons were slowed down and stopped in a target  $T$  of the investigated metal, in the form of a disk of 80 mm diameter and 10 mm thickness. The instant  $t_\mu$  of the stopping of the  $\mu^+$  meson was fixed by a system of scintillation-counter signals 1234 (coincidence of signals 1, 2, and 3 and anticoincidence of signal 4). The instant  $t_e$  of the emission of the positron of the  $\mu^+ \rightarrow e^+$  decay was registered by signals 4563. The parameters characterizing the precession of the  $\mu^+$  meson in the target  $T$  were determined from the experimental dependence of the count  $N_{\text{exp}}(t)$  of the positron telescope 4563 on the time  $t = t_e - t_\mu$ . This dependence was approximated by the expression

$$N(t) = N_0 e^{-t/\tau_0} (1 - a e^{-\Lambda t} \cos \omega t) \quad (1)$$

in the entire investigated interval of temperatures  $T$ . Here  $\tau_0 = 2.2 \times 10^{-6}$  sec is the lifetime of the  $\mu^+$  meson;  $\omega = eH/mc$  is the Larmor precession frequency of the  $\mu^+$

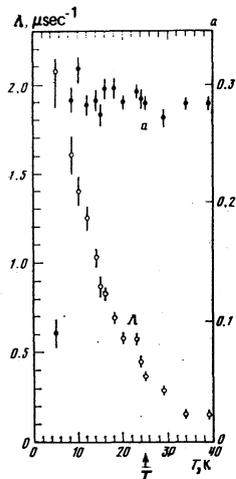


FIG. 3. Temperature dependences of  $\Lambda(T)$  and  $a(T)$  in praseodymium near the Néel temperature.

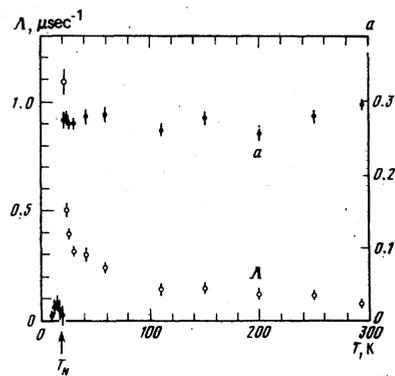


FIG. 4. Temperature dependences of  $\Lambda(T)$  and  $a(T)$  in neodymium.

meson in the external field  $H$ ;  $m$  is the mass of the  $\mu^+$  meson;  $a$  is the experimental asymmetry coefficient of the angular distribution of the positrons of the  $\mu^+ \rightarrow e^+$  decay;  $\Lambda$  is the relaxation rate of the  $\mu^+$ -meson spin in the target material. The values of the parameters  $N_0$ ,  $a$ ,  $\Lambda$ , and  $\omega$  were determined by comparison the  $N(t)$  dependence (1) with the corresponding experimental spectrum  $N_{\text{exp}}(t)$  by the maximum likelihood method.

Figures 2–10 show the temperature dependences of  $a(T)$  and  $\Lambda(T)$ , which characterize the precession of the  $\mu^+$  meson in praseodymium, neodymium, samarium, europium, terbium, dysprosium, holmium, and erbium. The values of  $a$  and  $\Lambda$  shown in these figures were determined by the maximum-likelihood method, as described above. The only exceptions are three values of  $a$  for terbium at  $T = 230$ – $236$  K, which were assumed equal to the mean value  $a = 0.286$  of the parameter  $a$  at  $T > 250$  K. This was done to decrease the number of parameters to be determined in (1) and by the same token to improve the accuracy of the determination of  $\Lambda$  at  $T = 230$ – $236$  K, when the value of  $\Lambda$  in terbium increases sharply. Here and hereafter we indicate only the statistical errors of the parameters  $a$  and  $\Lambda$ . The possible systematic errors  $\delta\Lambda$  due to inhomogeneity of the external magnetic field in the investigated samples does not exceed  $\delta\Lambda \approx 0.03 \mu\text{sec}$ . For each metal, Figs. 2–10 (and also Table II) show the temperatures  $T_N$  of the Néel phase transition from the paramagnetic ( $T > T_N$ ) into the antiferromagnetic ( $T < T_N$ ) state, measured in neutron-diffraction experiments.

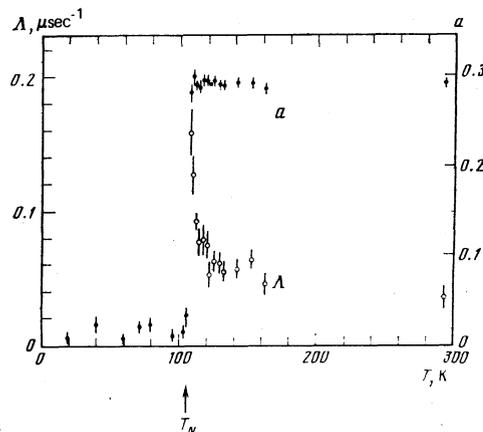


FIG. 5. Plots of  $\Lambda(T)$  and  $a(T)$  in samarium.

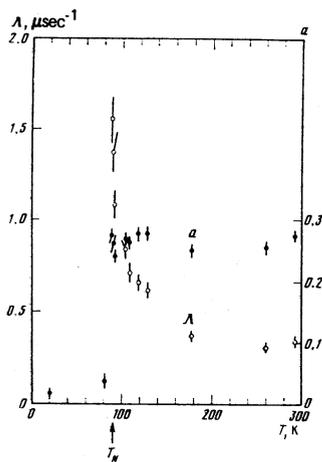


FIG. 6. Plots of  $\Lambda(T)$  and  $a(T)$  in europium.

Figures 7–10 show also the value  $T_C$  of the Curie temperature of the transition of the metal into the ferromagnetic state.

It follows from Figs. 2–10 that the functions  $a(T)$  and  $\Lambda(T)$  are similar for all the rare-earth metals and undergo abrupt changes on going from the paramagnetic into the antiferromagnetic state. In the paramagnetic state is observed a damped precession of the  $\mu^+$ -meson spin, and the damping rate  $\Lambda$  increases with decreasing temperature as  $T \rightarrow T_N$ . When the temperature drops below the Néel point the value of  $\Lambda$  increases so much that observation of the  $\mu^+$ -meson precession becomes impossible, since the amplitude  $a$  of the precession at  $T < T_N$  is practically equal to zero. The experimental values  $a \approx 0.02$  at these temperatures make up the background due to the  $\mu^+$  mesons that are stopped in the cryostat walls. The background values of  $\Lambda$  at  $T < T_N$ , which are of no physical interest, are not shown in Figs. 2–10.

To determine the relaxation rate  $\Lambda$  it is not obligatory that the  $\mu^+$ -meson precession be observed in a transverse magnetic field. It is possible to measure the spectrum (1) of  $N(t)$  in a longitudinal magnetic field  $H_{\parallel}$  (see Table I) or at  $H=0$ . In all these cases the measurement of the large values of  $\Lambda$  typical of the antiferromagnetic state of the rare-earth metals is limited by the dead (unobservable) time  $t_{\text{exp}} \approx 15$  nsec of the electronic apparatus used by us, so that only the lower limit of this quantity can be indicated, namely  $\Lambda(T < T_N) > 10^8 \text{ sec}^{-1}$ . So large a value of  $\Lambda$  means that the relaxation of the  $\mu^+$ -meson spin at  $T < T_N$  is due to interactions with the

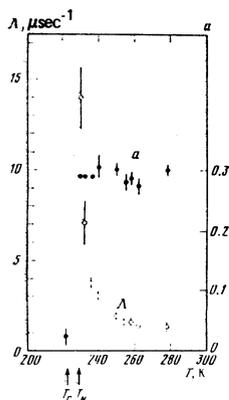


FIG. 7. Plots of  $\Lambda(T)$  and  $a(T)$  in terbium.  $T_C = 223$  K is the Curie temperature of the transition into the ferromagnetic state.

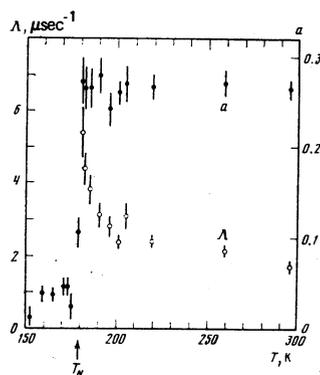


FIG. 8. Plots of  $\Lambda(T)$  and  $a(T)$  in dysprosium,  $T_C = 87$  K.

atomic electrons that are in a magnetically ordered state.

The abrupt changes of the quantities  $\Lambda(T)$  and  $a(T)$  at  $T_N$  allows us therefore to identify quite definitely the antiferromagnetic phase transition by the  $\mu^+$ -meson method. The relaxation of the  $\mu^+$ -meson spin in the paramagnetic state, i.e., at  $T > T_N$ , will be considered below.

Particular notice must be taken of the temperature dependence of the  $\mu^+$ -meson spin relaxation in praseodymium. It is seen from Figs. 2 and 3 that the  $\mu^+$ -meson spin precession in praseodymium is observed down to the temperature  $T \approx 8$  K and only at 5 K does a sharp decrease occur in the experimental precession amplitude and attests to the rapid decrease of the relaxation rate of the  $\mu^+$ -meson spin, i.e., to the transition of the metal into a magnetically ordered state. It follows from Figs. 2 and 3 that the Néel temperature measured by the  $\mu^+$ -meson method is  $T_N = (6 \pm 1)$  K. This result contradicts the previously measured<sup>3</sup>  $T_N = 25$  K of a polycrystalline praseodymium sample. The reason for this discrepancy is not clear and may be due to differences between the investigated praseodymium samples, all the more since this metal is a very "delicate" antiferromagnet and undergoes antiferromagnetic ordering only in polycrystalline form. No transition of single-crystal praseodymium into the ferromagnetic state could be observed down to  $T = 0.4$  K (Ref. 4).

We consider now the relaxation of the  $\mu^+$ -meson spin in rare-earth metals at  $T > T_N$ , i.e., in the paramagnetic state. As seen from Figs. 2–10, the relaxation rate  $\Lambda$ , at sufficiently high temperatures  $T > T_N$ , becomes almost constant and in a number of metals (terbium, dysprosium, holmium, erbium) it amounts to  $\Lambda \geq 10^8 \text{ sec}^{-1}$ .

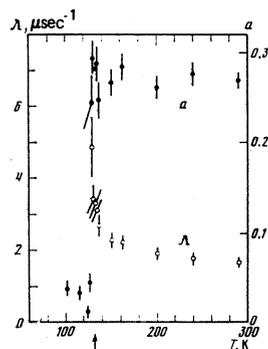


FIG. 9. Plots of  $\Lambda(T)$  and  $a(T)$  in holmium,  $T_C = 20$  K.

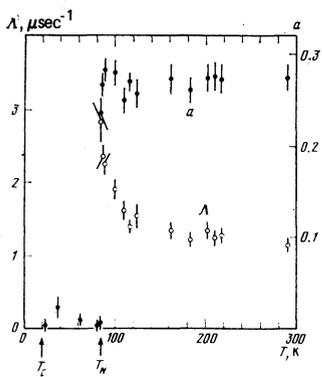


FIG. 10. Plots of  $\Lambda(T)$  and  $\alpha(T)$  in erbium,  $T_C = 19$  K.

For the investigated metals of the cerium subgroup (praseodymium, neodymium, samarium, and europium), the experimental values of  $\Lambda(T > T_N)$  are smaller by approximately a factor of ten. These values of  $\Lambda$  can be attributed to the dipole interactions of the magnetic moments of the  $\mu^+$  meson with the paramagnetic atoms of the metal. The expected relaxation rate  $\Lambda_{\text{calc}}$  at  $T > T_N$  can be estimated at

$$\Lambda_{\text{calc}} = 2\sigma^2/\nu \sim 10^5 \text{ sec}^{-1}. \quad (2)$$

Here  $\sigma = \gamma_\mu H_{\text{el}} \sim 10^9 \text{ sec}^{-1}$  is the rate of relaxation of the  $\mu^+$  meson spin on account of the dipole interactions with the system of non-oscillating spins of the surrounding atoms of the paramagnet;  $\gamma_\mu$  is the gyromagnetic ratio for the  $\mu^+$  meson;  $H_{\text{el}} \approx \mu_{\text{el}}/\gamma^3 \sim 10^5 \text{ Oe}$  is the magnetic field produced at the  $\mu^+$  meson by the electron magnetic moments ( $\mu_{\text{el}}$ ) of the metal atoms;  $r \approx 10^{-8} \text{ cm}$  is the distance from the  $\mu^+$  meson introduced into the crystal cell to the nearest metal atoms. The parameter  $\nu \sim 10^{13} \text{ sec}^{-1}$  is the frequency of the oscillations of the atomic spins at  $T > T_N$  and can be estimated from the relation  $h\nu = kT_N$ , where  $k$  is the Boltzmann constant and  $T_N \approx 100 \text{ K}$  is the magnetic ordering temperature. Formula (2) describes thus the decrease of the rate of dipole relaxation  $\sigma$  of the spin of the  $\mu^+$  meson on account of the oscillations of the spins of the surrounding atoms, and is analogous to the corresponding expression for the decrease of the rate of dipole relaxation on account of diffusion.<sup>5</sup> Formula (2) is valid at  $\nu \gg \sigma$ , which in our case is satisfied with good accuracy.

The described electronic relaxation of the spin of the  $\mu^+$  meson at  $T > T_N$  is confirmed experimentally. Table I lists the values of  $\Lambda$  at  $T > T_N$  for terbium, dysprosium, and holmium as functions of the intensity of the longitudinal magnetic field  $H_{\parallel}$  (along the direction of the  $\mu^+$ -meson spin). It is seen from Table I that longitudinal fields  $H_{\parallel} \lesssim 40\,000 \text{ Oe}$  hardly change the value of  $\Lambda$ . It follows therefore that the local magnetic fields, which lead to the relaxation of the  $\mu^+$ -meson spin in these metals, greatly exceed the value  $H = 4\,000 \text{ Oe}$  and can be

TABLE I. Plot of  $\Lambda(H_{\parallel})$  [ $10^6 \text{ sec}^{-1}$ ] at  $T = 300 \text{ K}$ .

Metal	$H_{\parallel} = 5 \text{ Oe}$	$H_{\parallel} = 4300 \text{ Oe}$
Terbium	$1.2 \pm 0.1$	$1.0 \pm 0.1$
Dysprosium	$2.1 \pm 0.2$	$2.4 \pm 0.2$
Holmium	$1.9 \pm 0.1$	$2.2 \pm 0.2$

TABLE II. Experimental values of  $\Lambda$  at  $T = 300 \text{ K}$ .

Metal	$T_N, \text{K}$	$\mu_{\text{el}}/\mu_B$	$\Lambda(T=300 \text{ K})$ [ $10^6 \text{ sec}^{-1}$ ]	Metal	$T_N, \text{K}$	$\mu_{\text{el}}/\mu_B$	$\Lambda(T=300 \text{ K})$ [ $10^6 \text{ sec}^{-1}$ ]
Praseodymium	27	0.7	$0.09 \pm 0.02$	Terbium	229	9.6	$1.3 \pm 0.1$
Neodymium	19	2.0	$0.10 \pm 0.02$	Dysprosium	179	10.7	$1.9 \pm 0.1$
Samarium	106	0.1	$0.04 \pm 0.02$	Holmium	132	10.9	$1.7 \pm 0.1$
Europium	89	5.9	$0.34 \pm 0.02$	Erbium	84	9.8	$1.2 \pm 0.2$

produced only by electrons. The calculated value  $\Lambda_{\text{calc}} \approx 10^5 \text{ sec}^{-1}$  obtained from formula (2) agrees qualitatively with the experimental values of  $\Lambda$  at  $T > T_N$  (see Figs. 2–10 and Table II); this also confirms the electronic mechanism of the  $\mu^+$ -meson spin relaxation.

Table II gives the values of  $\Lambda(T = 300 \text{ K})$  of the rate of relaxation of the  $\mu^+$ -meson spin at  $T = 300 \text{ K}$ , when the values of  $\Lambda$  of almost all the investigated metals are independent of temperature. The experimental values of  $\Lambda(T = 300 \text{ K})$  listed in Table II are compared with the values of  $\mu_{\text{el}}$  of the electronic magnetic moments of the paramagnetic atoms of the corresponding elements in the metallic state. The correlation between the values of  $\mu_{\text{el}}$  and of  $\Lambda(T = 300 \text{ K})$  which follows from Table II explains qualitatively the observed difference between the relaxation rates of the  $\mu^+$ -meson spins of the investigated paramagnets.

The increase of the experimental values of  $\Lambda$  as  $T \rightarrow T_N$  in the paramagnetic region can be attributed to partial ordering of the spins of the metal atoms, and consequently to the effective decrease of the frequency  $\nu(T)$ :

$$\nu(T) = 2\sigma^2/\Lambda(T). \quad (3)$$

Formula (3) which follows from (2) can be used for an experimental determination of the frequency  $\nu(T)$ . The quantity  $\sigma$  in (3) can be calculated under the assumption that the  $\mu^+$  meson is in one of the interstitial pores of the crystal lattice of the metal. In principle it is also possible to estimate  $\sigma$  experimentally in measurements of the relaxation rate  $\Lambda$  of the  $\mu^+$ -meson spin in the magnetically ordered state at  $T < T_N$ . These measurements, however, require not only a high time resolution but also the ability to measure small time intervals,  $\Delta t \sim 1 \text{ nsec}$ .

We note that in expression (3) for  $\nu(T)$  no account is taken of the diffusion of the  $\mu^+$  meson over the crystal; this diffusion, generally speaking, leads to an additional decrease of the experimentally measured relaxation rate  $\Lambda$ . Neglect of diffusion in this case is justified because of the low frequency of the diffusion jumps of the  $\mu^+$  meson from one unit cell to another, compared with the frequency  $\nu \sim 10^{13} \text{ sec}^{-1}$  of the oscillations of the electron spins of the metal atoms.

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## Dipole-reservoir cooling and dynamic polarization of nuclei in saturation of inhomogeneous EPR line

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The temperature of the electronic dipole-dipole reservoir (DDR) and of the dynamic polarization of nuclei are calculated under conditions of stationary saturation of an inhomogeneous EPR line. The solution is obtained by two methods: using the model of thermal mixing in a rotating coordinate frame, and in terms of spin packets. In the latter case it is shown that allowance for the DDR leaves the spectral-diffusion valid, but only for a definite combination of the Zeeman and dipole-dipole temperatures. Estimates of the maximum attainable DDR cooling coefficients and of the polarization of the lattice nuclei are presented, and it is shown that they do not depend monotonically on the width of the "hole" produced in the EPR line upon saturation.

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### 1. INTRODUCTION AND FORMULATION OF PROBLEM

It is known that not-strictly-resonant saturation of a magnetic resonance line in a solid can lower substantially the temperature  $T_{SS}$  of the spin-spin interaction reservoir (DDR).<sup>1</sup> Upon saturation of the EPR line of a paramagnetic impurity in a magnetically dilute paramagnetic crystal, this cooling can be transferred to the Zeeman subsystem of the nuclear spins of the lattice, thus causing dynamic polarization of the nuclei ("dynamic cooling").<sup>2,3</sup> This phenomenon was reliably established in experiment and plays a substantial role in magnetic resonance and its applications, especially at low temperatures.<sup>4-7</sup>

The theory of effects connected with the cooling of the DDR was initially developed for magnetic-resonance lines homogeneously broadened by dipole-dipole interactions.<sup>1-3</sup> In practice, however, the main contribution to the width of paramagnetic-impurity EPR lines are made as a rule by inhomogeneous mechanisms, and it was therefore necessary to extend the theory to cover this case, too. So far this problem could be solved only for two limiting situations: neglecting the spectral diffusion inside the inhomogeneous line, and for very strong spectral diffusion that covers the entire EPR line.<sup>4,8</sup> At the same time, for the more general case (and perhaps of greatest practical importance) of limited spectral diffusion, corresponding to the appearance of a stationary "burned hole" in the inhomogeneous line, only qualitative estimates were proposed.<sup>8,9</sup> These, as will be shown below, do not agree with the true result even in order of magnitude.

In this paper we solve the problem by two methods. The result is first obtained using a simplified model of thermal mixing in a rotating coordinate frame, after which it is generalized within the framework of the description of an inhomogeneous line in the form of an aggregate of spin packets.

The object that we consider is a solid paramagnet at a temperature  $T_L \equiv \beta_L^{-1}$ , with  $n_I$  nuclear spins  $I$  and  $n_S$  electron spins  $S$  per unit volume ( $n_I/n_S \gg 1$ ), and located in a stationary magnetic field  $H_0 \parallel z$ . We assume that  $S = \frac{1}{2}$ , so that the EPR spectrum consists of a single line, assumed to be inhomogeneously broadened (with width  $2\delta_{inh}$ ), and the distribution of the resonance frequencies of the electron spins does not correlate with their distribution over the crystal volume, so that the electronic DDR is common to all spins  $S$ .<sup>8</sup> Let furthermore the sample be acted upon by a high-frequency magnetic field  $2H_1 \cos \Omega t (H_1 \perp z)$ , that saturates the EPR line with a detuning  $\Delta = \Omega - \omega_0$  relative to its "center of gravity"  $\omega_0$ , determined from the relation

$$\int_{-\infty}^{\infty} \delta G(\delta) d\delta = 0, \quad (1)$$

where  $\delta = \omega - \omega_0$ , and  $G(\delta)$  is the form factor, normalized to unity, of the inhomogeneous line.

It is well known that the action of the saturating field leads under the conditions considered above to the appearance of the so-called "burned line" which is observed when the EPR is recorded with a second (unsaturated) high-frequency field. The width  $2\delta_h$  of this hole depends on the saturation factor  $s(\Omega)$ , on the homogeneous width  $2\delta_s$  of the spin packet, and on the effectiveness