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Optical detection of NMR in high-purity GaAs: Direct study of the relaxation of nuclei close to shallow donors

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Optical detection of NMR has two outstanding features: (i) selective observation of the nuclei in the regions of electronic localization and (ii) extremely high sensitivity. This technique can be applied to semiconductors under circularly-polarized light excitation and involves monitoring the change at resonance of the degree of circular polarization of the luminescence. We have detected optically the resonance of nuclei close to shallow donors in high-purity GaAs at 1.7 K. These nuclei are extremely scarce, of total number of the order of 10^{11}. We have studied their spin-lattice relaxation due to hyperfine contact interaction with the photoelectrons trapped on the donors. We show the very fast modulation of this interaction, of characteristic time of several 10^{-11} s, which is very likely produced by spin exchange between free and trapped electrons. Furthermore, we demonstrate in a very striking way the occurrence of transport of nuclear magnetization by spin diffusion. This is done by switching off the light excitation, since the evolution of the nuclear spin system in the absence of photoelectrons is entirely due to spin diffusion. The analysis of the experimental results yields the value of the diffusion constant, which is comparable with its theoretical estimate.

I. INTRODUCTION

The relaxation of a nuclear-spin system by paramagnetic impurities in solids has been investigated for a long time[1] and has led to the distinguishing of two types of nuclear spins.[2,3] First there are the spins situated close to the relaxing centers, which are directly relaxed by the electrons trapped on these centers, either by dipolar[4] or by hyperfine-contact interaction.[5,6] On the other hand, the relaxation of the bulk nuclei takes place through transport of magnetization by spin diffusion from the former nuclei. The theoretical and experimental investigations of this relaxation have shown that the magnetization of the bulk nuclei increases exponentially as a function of time. An important parameter which describes the system is the diffusion radius, which is the distance from the impurity which sets the limit between the two types of nuclei.

The above-mentioned theory has been extensively verified in a wealth of experimental cases,[4–6] using conventional nuclear-magnetic-resonance (NMR) studies as a function of magnetic field, temperature, and impurity concentration. Since the nuclei close to impurities are in most cases much less numerous than the bulk nuclei, their contribution to the NMR signal is generally negligible, so that the measured relaxation is that of the bulk nuclei. Fundamentally, this has an important drawback. The relaxation of the bulk nuclei involves two distinct processes: existence of fast-relaxing nuclei close to the relaxing centers and transport of nuclear magnetization from these nuclei to the bulk nuclei by spin diffusion. The measured value of the relaxation time of the bulk nuclei depends on these two processes which cannot be studied independently from each other.

A few studies, however, have yielded direct information on the nuclei close to impurities, by using highly doped crystals, in which case these nuclei are more numerous and yield an observable NMR signal. There is first the work of Blumberg,[7] who shows a nonexponential increase of the nuclear magnetization due to polarization of the nuclei close to impurities at the very first stage of the relaxation. In some cases, the resonance signal from these nuclei is directly observed and their relaxation is investigated.[8] On the other hand, the transfer of nuclear magnetization by spin diffusion has been observed directly by several authors. This is done either by using systems where spin diffu-

sion can be compared with atomic diffusion,[9] or by studying the effect of surface relaxing centers on the bulk relaxation as a function of the size of the crystal.[10]

In this work, we present a very unique case where (i) we observe selectively the resonance of only the nuclei situated close to shallow donors in a high-purity crystal, (ii) the magnetic resonance of this extremely small number of nuclei (several 10^{11}) is recorded directly, and (iii) spin diffusion and direct relaxation by electrons trapped on donors can be analyzed separately in a straightforward way. The present study is performed in high-purity gallium arsenide at 1.7 K under light excitation. The three above-mentioned remarkable results are a direct consequence of the fact that the resonance is optically detected from the polarization of the luminescence light. Indeed, the selectivity and extremely high sensitivity, as will be shown below, rely on the fact that the resonance signal depends on the magnitude of the hyperfine-contact interaction between the photoelectrons trapped on donors and the relevant nuclei. The possibility of distinguishing spin diffusion from direct relaxation is determined by the fact that, in the absence of light excitation, no electrons are trapped on the donors. Thus, direct relaxation is completely frozen and the only possible evolution of the nuclear-spin system is due to spin diffusion.

We measure the relaxation speed of the nuclei close to shallow donors and show the transport of nuclear magnetization by spin diffusion. Theoretical analysis of these results is obtained from a numerical resolution of the diffusion equation, and yields the value of the diffusion constant.

In the following section, we recall the principles of our method. The experimental studies of direct relaxation and spin diffusion are presented in Secs. III and IV, respectively. These results are interpreted in Sec. V.

II. PRINCIPLES

Optical detection of NMR in a semiconductor under circularly-polarized light excitation (optical pumping) is based on the measure of the change at resonance of the degree of circular polarization of the luminescence light. The feasibility of this detection relies on three distinct effects:

(i) Absorption of circularly polarized light creates spin-polarized photoelectrons.[11]

(ii) Owing to the hyperfine contact interaction with the photoelectrons, the nuclear spins are dynamically polarized.[12,13] The photoelectrons experience in turn a giant hyperfine field of nuclear origin, as high as several kilogauss in practical cases, which lies along the mean electronic spin.[13,14]

(iii) The change of the direction of this nuclear field in conditions of NMR causes an electronic precession which results in a decrease of the mean value of the electronic spin. This is detected from the corresponding decrease of the degree of circular polarization of the luminescence.[15,16]

The present method has close analogies with electron-nuclear double resonance (ENDOR),[17] in the sense that the electronic-spin system acts as a probe of the nuclear-spin polarization, due to the existence of the hyperfine-contact interaction between the two systems. However, our method has two basic advantages over ENDOR:

(i) Simplicity: There is no need for a double-resonance technique, since the mean electronic spin is conveniently monitored from the measure of the luminescence light polarization. Furthermore, unlike the standard detection of NMR, the measured quantity is not the slight variation of the value of the rotating field B_1, so that there is no need for a sophisticated setup to excite the resonance.

(ii) Extreme sensitivity: This is due to the fact that this method is an optical detection, and is only limited by the intensity of the luminescence light.

On the other hand, the present method has two drawbacks. Firstly, it is limited to cases where optical pumping, that is optical orientation of photoelectrons by circularly polarized light, can be observed. Secondly, the measured quantity is not simply connected with the magnetization of the nuclei which resonate, but with the geometry of the field configuration at resonance.[16] Consequently, the precise measurement of this magnetization is intricate. This is why we do not attempt a precise study of the nuclear relaxation here. We limit ourselves to estimating the characteristic time of the evolution of the resonance signal during the nuclear dynamic polarization. This time, which is obtained within a factor of 2, is of the order of the relevant nuclear relaxation time.

We have applied the present method to the relaxation of arsenic nuclei in high-purity GaAs in the range 10^{14} donors per cubic centimeter, at pumped-helium temperature. We refer to a previous publication for a description of experimental setup and conditions for a complete study of the

resonance line shape.[16] The fundamental feature of this system is that the observed resonance is that of nuclei close to shallow donors. This comes from the fact that the relevant hyperfine interaction takes place between photoelectrons trapped on shallow donors and lattice nuclei situated close to these donors.[14] The nuclear field which is responsible for the observed resonance signal is then the hyperfine field experienced by electrons trapped on donors. This field is of the form

$$B_n = b_n \int p(r) \exp(-2r/a_0^*) r^2 dr \ , \tag{1}$$

where $p(r)$ is the nuclear polarization at distance r from the donor, and obviously depends on the magnetization of the nuclei situated in a sphere centered at the donor, of typical radius of the order of the Bohr radius of the donor, $a_0^* \simeq 100$ Å.

To analyze the time variation of the nuclear field, we recall that under the effect of the hyperfine coupling with the photoelectrons trapped on donors the nuclei are dynamically polarized. The steady-state polarization p_∞ is proportional to the mean electronic spin, which is $\pm\langle S\rangle$ for $\sigma^\mp$ excitation light, and zero for a linearly polarized or unpolarized light. The time evolution of the nuclear polarization is described by the usual diffusion equation[1] which takes into account direct relaxation together with spin diffusion:

$$\frac{dp(r)}{dt} = D\Delta p(r) - \frac{1}{T_1(r)}[p(r) - p_\infty] \ . \tag{2}$$

An approximate value of the diffusion constant D is given by[3]

$$D = \gamma_n \Delta h d^2/30 \ , \tag{3}$$

where γ_n is the nuclear gyromagnetic ratio, d the internuclear spacing, and Δh the width of the standard resonance line of the nuclear spins. Owing to the exponential decrease of the electronic density as a function of distance to the donor, the nuclear relaxation time $T_1(r)$ is of the form

$$T_1(r) = T_1(0) \exp(4r/a_0^*) \ . \tag{4}$$

The relaxation speed at the origin is given by[18]

$$\frac{1}{T_1(0)} = \Gamma_t \Omega^2 \frac{2\tau_c}{1+\omega^2 {\tau_c}^2} \ , \tag{5}$$

where Ω is, in frequency units, the magnitude of the relaxing interaction. The correlation time τ_c is a measure of its fluctuation time. The quantity $\hbar\omega$ is the energy required for a simultaneous reversal of both an electronic and a nuclear spin, and Γ_t is the probability of occupation of the donor.

The time evolution of B_n is completely defined by Eqs. (1) and (2). This evolution is in the general case complex since B_n is the sum of contributions of the nuclei at different distances from the donor, which are not characterized by the same time constant. More precisely, up to a distance from the donor equal to the diffusion radius δ, the evolution of the nuclear magnetization by spin diffusion is negligible so that the increase is due to the sole effect of direct relaxation. On the other hand, beyond δ, the spin diffusion plays a dominant role and the magnetization is homogeneous and increases in a time of the order of the bulk nuclear relaxation time. In this work, we shall consider, for simplicity, that the characteristic time of evolution of B_n, which is measured from the experiment, is the relaxation time of the nuclei which have the largest contribution to the nuclear field. These nuclei are shown to be at a distance from the donor equal to the Bohr radius a_0^*.[19,20] Indeed, closer nuclei are too scarce and only weakly contribute to B_n. Conversely, nuclei situated at $r > a_0^*$, although sufficiently numerous, individually create a vanishingly small nuclear field.

In summary, we have recalled in this section that the magnitude of the optically detected resonance signal is connected to the value of the hyperfine field experienced by electrons trapped on shallow donors, and we have discussed the time evolution of this nuclear field, which is produced by nuclear relaxation. In the following section, we present the determination of the relaxation time of the relevant nuclei, from the measure of the characteristic time of the variation of the resonance signal.

III. EXPERIMENTAL STUDY OF DIRECT RELAXATION

The possibility to study the direct relaxation independently of spin diffusion is based on the assumption that the diffusion radius δ is larger than the Bohr radius a_0^*, which will be verified below. In this framework, the relevant nuclei, at distance $r = a_0^*$ from the donor, are situated inside the diffusion sphere, and their polarization is mainly due to direct relaxation. Thus, the characteristic time of the increase of B_n, after switching on circularly polarized excitation, is of the order of $T_1(a_0^*)$ which is related geometrically to $T_1(0)$ [Eq. (4)], and depends very weakly on the existence of spin diffusion.

Figure 1 shows the experimental measurement of $T_1(a_0^*)$ for arsenic nuclei. One starts with the nu-

clear system at equilibrium under σ^+-polarized-light excitation. At time 0, the excitation light polarization is changed to σ^-. This produces a change of sign of B_n, and one looks for the time t, at which no resonance signal is detected. This time is of the order of $T_1(a_0^*)$. Complete cancellation of this signal is found to be impossible, probably because of inhomogeneities of nuclear relaxation, but a sharp minimum is obtained for $t \simeq 3$ s. Using Eq. (4), we obtain $T_1(0) = 80 \pm 50$ ms.

The results presented here have been obtained for an external magnetic field B equal to 6 kG, but are found to be independent on B up to the maximum available magnetic field, equal to 12 kG. This indicates[18] a very short correlation time τ_c, smaller than 2×10^{-11} s. On the other hand, this correlation time can be estimated from the measured value of $T_1(0)$, if we assume that the rate Γ_t of occupation of the donors is equal to unity, which is valid within less than one order of magnitude. One finds $\tau_c \simeq 2.5 \times 10^{-11}$ s. Thus, the two results above are consistent with each other, and show a very rapid modulation of the hyperfine coupling experienced by the nuclei. This cannot originate from trapping and recombination of electrons, which is at least two orders of magnitude slower.[21] We think that this modulation occurs from spin-exchange processes between free and trapped electrons. The very high efficiency of these mechanisms has been demonstrated in the same sample by a previous study of the optically detected NMR.[16] The estimated spin-exchange

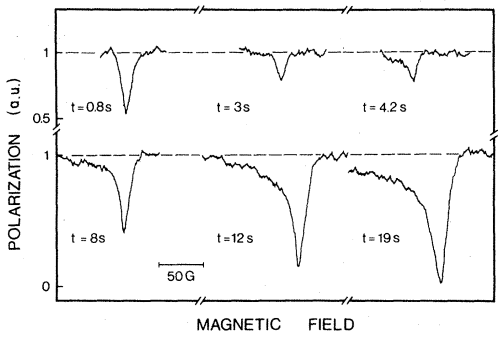

FIG. 1. Measurement of the spin-lattice relaxation time of nuclei close to shallow donors. At time zero, one changes the polarization of the excitation light, which produces a change of sign of the equilibrium nuclear magnetization. The time t, at which no NMR signal is detected, gives an order of the nuclear relaxation time. The resonance is that of ^{75}As nucleus in a magnetic field equal to 6 kG in adiabatic fast-passage conditions.

time is comparable with the above correlation time. This agreement is a further proof of the high efficiency of spin exchange between free electrons and electrons trapped on donors.

IV. EXPERIMENTAL STUDY OF SPIN DIFFUSION

A convenient way to show the effect of spin diffusion is to study the evolution of the nuclear field in absence of light excitation. Indeed, in this case, no photoelectrons are present, so that the above relaxation does not occur. Furthermore, in our pure samples at 1.7 K, the dark spin-lattice relaxation is completely negligible.[22] Consequently, in absence of excitation light, the evolution of the nuclear spin system is completely governed by spin diffusion.[23] Although the total magnetization of the nuclear system remains unchanged, the transfer of magnetization by spin diffusion modifies the nuclear hyperfine field experienced by trapped electrons. This is why this transfer can be demonstrated using our optical technique. We present below two different experimental situations which slow the spin diffusion from the donor, and the spin diffusion to the donor, respectively. All the experimental results have been obtained in the case of arsenic nuclei.

We first start from the case where the nuclei situated inside the diffusion spheres are polarized, and the bulk nuclei are not polarized. This can be accomplished by exciting the crystal during a time comparable with the time, $T_1(a_0^*)$, measured above. We then switch off the excitation light and we measure the characteristic time of the resulting decay of B_n due to spin diffusion. This time is of the form

$$T_{\text{dark}} = \xi \delta^2 / D \ , \tag{6}$$

where ξ is a numerical factor of order unity. The measure of T_{dark} is performed by switching off the excitation light for a time t_{dark} and then by reexciting the crystal with circularly polarized light and by optically detecting the resonance. Figure 2 shows the variation of this signal as a function of t_{dark}. We indeed find a fast decay, and we obtain $T_{\text{dark}} = 10 \pm 5$ s.

The experimental procedure used to evidence spin diffusion *towards* the donor is shown in Fig. 3. This procedure is composed of three different steps:

(i) The nuclei are first polarized by exciting the

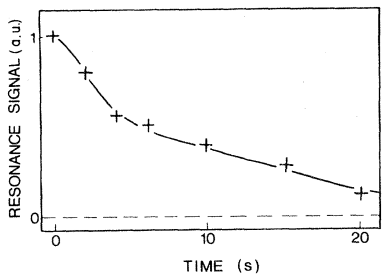

FIG. 2. Effect of spin diffusion from the donors. The nuclei close to the donors are first polarized by light excitation of duration 30 s. The excitation light is then switched off for a time t_{dark}, and the NMR signal is detected. This gives the variation of the observed signal as a function of t_{dark}. Note that, for a polarization time of 20 min, one obtains exactly the same variation, provided one changes the time unit from seconds to minutes.

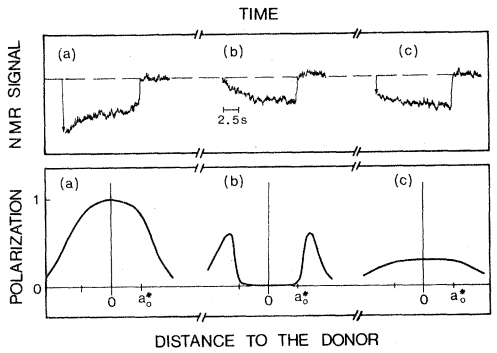

FIG. 3. Spin diffusion to the donor. The upper half represents the resonance signal obtained by sudden application of B_1 (see Ref. 16), and the lower half illustrates the corresponding configuration of the nuclear spin system. Case (a) is the reference signal. Case (b) is obtained after σ/π transient. Case (c) is obtained after σ/π/dark transient. The corresponding times are $t_\sigma = 2$ h, $t_\pi = 1$ min, and $t_{dark} = 20$ s. The resonance signal is zero in case (b) and is indicated by arrows in cases (a) and (c). The fact that step (c), which is obtained from step (b) after switching off the light excitation, shows a nonzero resonance signal, is unambiguous evidence for the existence of spin diffusion.

crystal with circularly polarized light for a time t_σ.

(ii) One illuminates the crystal with linearly polarized light for a time t_π. This creates nonpolarized photoelectrons and depolarizes the nuclear spins, since in this case, $p_\infty = 0$ in Eq. (2). The first depolarized nuclei are those of shorter relaxation time situated close to the donors, so that the effect of linear excitation is to "burn a hole" in the diffusion spheres. As shown in Fig. 3, no signal is detected in this step. However, there exist polarized nuclei, situated too far from the donor to contribute significantly to the nuclear field since their hyperfine coupling with the electron trapped on the donor is negligible.

(iii) Excitation light is then switched off for a time t_{dark}. This results in a transfer of nuclear polarization by spin diffusion towards the donor and allows to reobserve a resonance signal [Fig. 3(c)]. We point out the remarkable feature of the present experiment which shows enhancement of the polarization of the relevant nuclei in absence of photoelectrons, that is, in conditions where this effect is initially unexpected. This can only be due to the transfer of magnetization from other nuclei and is a direct evidence of the existence of spin diffusion independently of all models which describe spin relaxation and resonance line shape.[24]

In summary, we have shown that the evolution of the nuclear-spin system in absence of light excitation is entirely due to spin diffusion, since, in this case, direct relaxation is completely frozen. The observation of a rapid change of the hyperfine nuclear field after switching off the excitation light, is unambiguous evidence for the occurrence of spin diffusion. We point out that the possibility to observe the effect of spin diffusion is a consequence of the fact that the nuclear polarization is not homogeneous in space since the bulk nuclei are not polarized and the only polarized nuclei are those situated close to shallow donors. In the opposite case, if all the nuclei throughout the sample are uniformly polarized, spin diffusion can no longer occur. In this case, the only possible evolution of the nuclear-spin system in the dark is through spin-lattice relaxation which, in absence of photoelectrons, is characterized by times of the order of hours in our high-purity crystals at 1.7 K. Thus, we expect that the polarization of the bulk nuclei should produce a drastic increase of the time T_{dark}.

We have looked for this effect using very-long-lasting light excitations, in order to polarize the bulk nuclei. As expected, we have observed that, after irradiation of the crystal for 20 min, the decay time of the nuclear field in the absence of excitation light is increased by a factor of 60. More precisely, the decay of the resonance signal as a function of the time t_{dark} of absence of excitation light, coincides with that of Fig. 2 if in this figure, one changes the time unit shown in abcissa from seconds to minutes. This drastic increase of the time T_{dark} indicates that the bulk nuclei are polarized and confirms qualitatively the interpretation

of the above effects of spin diffusion. We point out however that, in such a case, free electrons should experience a nonzero nuclear field since the bulk nuclear magnetization is no longer negligible. This last fact can be shown to be inconsistent with the results presented in Fig. 3.[19] This contradiction is not resolved at the present time. The fact that the bulk nuclear magnetization remains very small could be explained if inhomogeneous polarization of the bulk nuclei occurs due to macroscopic fluctuations of nuclear relaxation time. Further experiments are necessary to verify this assumption.

V. INTERPRETATION

We present here the evaluation of the diffusion constant from the results summed up in Figs. 1 and 2. This evaluation is obtained by using the results of a numerical resolution of the diffusion equation [Eq. (2)], which does not have an analytic solution. We recall that previous bulk spin-lattice relaxation measurements were interpreted using simple physical ideas which allow to obtain an approximate value of the diffusion radius, δ.[5,6] This treatment satisfyingly accounts for the bulk nuclear relaxation, but is completely unsufficient for our local study of nuclear relaxation. The results of our numerical calculation are presented in Fig. 4 in the case of a single donor in a sphere of radius R. This figure shows the theoretical variation of the nuclear polarization $p(r)$, for a time ranging between $2.5T_1(0)$ and $2000T_1(0)$. The boundary condition is $\partial p/\partial r \mid_R = 0$. On each curve, a landmark shows the distance $r(t)$ for which the nuclear polarization $p(r)$ has the value $\frac{1}{2}[p(0)+p(R)]$. One sees that for $t < 50T_1(0)$, the bulk nuclei are unpolarized, and $r(t)$ increases. On the other hand, for $t > 50T_1(0)$, the bulk nuclei are polarized and one observes that $r(t)$ remains constant. This verifies the well-known behavior for the nuclear polarization.[7] For the diffusion radius we take δ the constant value of $r(t)$ at long times t. We have repeated this treatment for several values of D and $T_1(0)$ and have found that δ is given in all cases by

$$DT_1(\delta)=\delta^2 , \tag{7}$$

with a maximum error of about 20%.[25] This formula expresses that diffusion is as efficient as direct relaxation at polarizing the nuclei situated immediately beyond δ. We recall that Bagraev

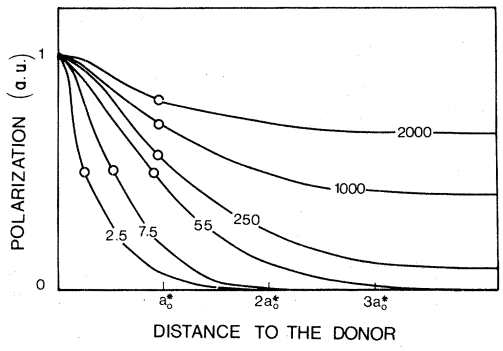

FIG. 4. Numerical resolution of the diffusion equation for various times ranging between $2.5T_1(0)$ and $2000T_1(0)$ in a sphere of radius $R=4a_0^*$. The landmark represents the point of the curve corresponding to $p(r)=\frac{1}{2}[p(0)+p(R)]$. The abscissa of this landmark increases with time as long as the bulk nuclei remain unpolarized. The constant value of this abscissa when the bulk nuclei are polarized gives the value of the diffusion radius.

et al.[6] have used the approximated value $DT_1(\delta)=a_0^{*2}$ and that Lampel[5,20] has obtained, from a first-order resolution of Eq. (2),

$$DT_1(\delta)=a_0^{*2}/16 . \tag{8}$$

However, this last treatment supposed that the width Δr, of the curve $p(r)$, is small as compared with $r(t)$, which is obviously not verified by our numerical calculation. If on the other hand, we assume $\Delta r = r(t)$, this first-order resolution leads to the result of our numerical calculation.

Thus, the two quantities δ and D are related to the measured quantities $T_1(0)$ and T_{dark} by Eqs. (6) and (7), and can be experimentally determined. However, an accurate determination of these parameters requires the knowledge of the quantity ξ, introduced in Eq. (6). This quantity depends on the slope of the function $p(r)$ at the diffusion radius, and on the relative values of both the diffusion radius and Bohr radius. We have performed the determination of D by an iteration procedure supposing first that $\xi=0.5$, and by performing a self-consistent calculation.[19] We find for arsenic nuclei:

$$\begin{aligned} &\delta = 140 \text{ Å} , \\ &D = 10^{-13} \text{ cm}^2/\text{s} . \end{aligned} \tag{9}$$

The measurement of δ is precise to within about 20%. One verifies that, as has been assumed before, the obtained value of δ is larger than the Bohr radius. The precision on the determination of D is essentially determined by the precision on

T_{dark}, which is 50%. This value is a factor of 3 smaller than the estimate obtained from Eq. (3). This agreement is quite satisfactory, since this last estimate only gives an order of magnitude of D.

VI. CONCLUSION

The optical detection of NMR in a semiconductor has allowed direct investigation of spin-lattice relaxation of nuclei close to shallow donors. Although similar investigations have already been achieved using conventional NMR, the present work shows a unique case where this study can be performed in a very straightforward way (in a high-purity crystal), and allows us to study both direct relaxation and spin diffusion separately. We show a very efficient modulation of the hyperfine coupling of these nuclei with electrons, which is due to spin exchange with free electrons. Furthermore, we show the effect of spin diffusion independently of all models describing relaxation and resonance line shape.

The most outstanding aspect of the optical detection of NMR, which has made the present study possible, is that this technique is a local probe of the nuclei situated in the zones of electronic localization. This has been discussed in detail elsewhere,[14] and is based on the fact that the hyperfine-contact interaction of an electron with a given nucleus increases with the localization of this electron. As a consequence, the present method is extremely sensitive. In the present case, the total number of nuclei which produce the reported effects is estimated to be of the order of 10^{11}, which is several orders of magnitude lower than the sensitivity of existing NMR spectrometers. We recall, however, that the applicability of optical detection of NMR is limited to crystals where orientation of photoelectrons by circularly polarized light, together with orientation of the lattice nuclei, is possible. This represents however a wide range of semiconducting crystals, for which optical pumping is observed, and is almost systematically accompanied by orientation of lattice nuclei.[26] Furthermore, the fact that this method is a specific probe of a very small fraction of the total number of nuclei present in the crystal, should have promising applications such as the study of deep centers in semiconductors. In spite of the high localization of these states, the hyperfine nuclear field is in principle expected to be of the same order as in the present case.[14] The optical detection of NMR could then be applied to these systems, which might yield valuable information on nuclear relaxation and electronic wave function.

ACKNOWLEDGMENTS

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[1]N. Bloembergen, Physica 15, 386 (1949).
[2]P. G. DeGennes, J. Phys. Chem. Solids 7, 345 (1958); I. J. Lowe and D. Tse, Phys. Rev. 166, 279 (1968); L. L. Buishvili and D. N. Zubarev, Fiz. Tverd. Tela 7, 722 (1965) [Sov. Phys.—Solid State 7, 580 (1965)].
[3]G. R. Khutsishvili, Usp. Fiz. Nauk 87, 211 (1965) [Sov. Phys.—Usp. 6, 743 (1966)].
[4]S. M. Day, E. Otsuka, and B. Josephson, Phys. Rev. 137, 108 (1965); A. L. Pigg and S. M. Day, Phys. Rev. 11, 3219 (1975); P. Bernier and H. Alloul, J. Phys. F 6, 1193 (1976).
[5]G. Lampel, Thesis, Orsay (1968) (unpublished).
[6]N. T. Bagraev, L. S. Vlasenko, and R. A. Zhitnikov, Zh. Eksp. Teor. Fiz. 76, 552 (1979) [Sov. Phys.—JETP 49, 278 (1979)].
[7]W. E. Blumberg, Phys. Rev. 119, 79 (1960); see also the subsequent work of G. W. Leppelmeier and V. Jeener, Phys. Rev. 175, 498 (1968).
[8]A. R. King, J. P. Wolfe, and R. L. Ballard, Phys. Rev. Lett. 28, 1099 (1972); H. Alloul, *ibid.* 35, 460 (1975).
[9]J. V. Gates and W. H. Potter, Phys. Rev. 15, 4143 (1977).
[10]Y. Roinel and J. M. Winter, J. Phys. (Paris) 31, 351 (1970).
[11]G. Lampel, *Proceedings of the Twelfth International Conference on the Physics of Semiconductors, Stuttgart, 1974,* edited by M. H. Pilkuhn (Teubner, Stuttgart, 1974), p. 743; C. R. Pidgeon, in *Handbook of Semiconductors,* edited by M. Balkanski (North-Holland, Amsterdam, 1980), Vol. 2, p. 223, and references therein.
[12]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, 1961), Chap. 8.
[13]M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. 68, 1514 (1975) [Sov. Phys.—JETP 41, 759 (1976)].
[14]D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, Phys. Rev. B 15, 5780 (1977).
[15]V. L. Berkovits, A. I. Ekimov, and V. I. Safarov, Zh. Eksp. Teor. Fiz. 67, 1912 (1974) [Sov. Phys.—JETP 38, 169 (1974)].
[16]D. Paget, Phys. Rev. B 24, 3776 (1981).
[17]G. Feher, Phys. Rev. 114, 1219 (1959).

[18]A. Abragam, Ref. 12, Chap. 9.

[19]D. Paget, thesis, Orsay 1981 (unpublished).

[20]V. L. Berkovits, C. Hermann, G. Lampel, A. Nakamura, and V. I. Safarov, Phys. Rev. B 18, 1767 (1978).

[21]R. Ulbrich, *Proceedings of the Twelfth International Conference on the Physics of Semiconductors, Stuttgart, 1974,* edited by M. H. Pilkuhn (Teubner, Stuttgart, 1974), p. 376; C. J. Hwang and L. R. Dawson, Solid State Commun. 10, 443 (1972).

[22]J. A. McNeil and W. G. Clark, Phys. Rev. B 13, 4705 (1976).

[23]If the difference of spin energies of two neighboring nuclei is larger than the local field, the spin diffusion can be blocked (Ref. 7). As shown in Ref. 19, this is not the case in high-purity GaAs, at least for the nuclei situated at the Bohr radius, which are responsible for the measured evolution of the nuclear field.

[24]Unlike the result presented in Fig. 2, this interpretation is still valid if there exist traps of unknown origin which are likely to cause irreversible modification of relaxation time after light excitation of the crystal, as reported by T. Kushida and A. H. Silver, Phys. Rev. 137, 1591 (1965).

[25]This holds at least provided $DT_1(0)$ is smaller than a value of the order of $0.04a_0^{*2}$ which is the maximum value of $DT_1(0)$ for which Eq. (7) admits a real solution. For larger values of $DT_1(0)$, which correspond to very efficient spin diffusion, we find as expected that the nuclear polarization is at all times homogeneous in space, so that no diffusion radius can be obtained from the numerical resolution. However, this homogeneous polarization is very unlikely to occur in physical cases, due to the blocking of spin diffusion very close to the donor produced by inhomogeneities of nuclear-spin energy, mentioned in Ref. 23. In this case, one should probably take for δ the value calculated by Blumberg in Ref. 7.

[26]For a review of the effects of nuclear orientation in optical-pumping conditions, in weak external fields, see B. P. Zakharchenya, in *Proceedings of the Fourteenth International Conference on the Physics of Semiconductors, Edinburgh, 1978,* edited by B. L. H. Wilson (Institute of Physics, Bristol, 1978), p. 31. This review shows the effects obtained in Si, GaAs, and $Ga_{1-x}Al_xAs$. Nuclear effects have also been observed in CdTe [A. Nakamura, D. Paget, C. Hermann, C. Weisbuch, G. Lampel, and B. C. Cavenett, Solid State Commun. 30, 411 (1979)]; GaSb (see Ref. 20), $Ga_{1-x}In_xAs$ (V. L. Berkovits, D. Paget, and V. I. Safarov, unpublished); and quaternary alloys (C. Hermann, private communication).