Spectral distribution of polarization of nuclei in impurity absorption of light in silicon

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Dynamic polarization of Si²⁹ nuclei in silicon following photoionization of impurity centers was observed for the first time ever. The magnetic field was seen to influence the dependence of the degree of optical polarization of the nuclei on the wavelength of the pump light.

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We describe here results of experiments on optical polarization of the nuclear moment in silicon following polarization of the impurity centers by light with quantum energy lower than the width of the forbidden band.

The experiments were performed with silicon containing $\sim 10^{14}$ cm⁻³ phosphorus atoms and compensated with samarium having approximately the same concentration. The sample was illuminated with unpolarized monochromatic light at T=77 K in various magnetic fields H_0 . The light source was a 50 W incandescent lamp. To obtain monochromatic light, whose wavelength was varied in the experiment between 0.5 and 2 μ m, a prism monochromator was used. The sample was illuminated with light of fixed wavelength λ for 30 minutes and then heated to room temperature and transferred to an NMR microwave spectrometer that measured the nuclear magnetization. During the transport and measurement time, which amounted to ~ 2 min, the nuclear magnetization could not decrease noticeably, since the spin-lattice relaxation time of the Si²⁹ nuclei was ~ 1 hour.

The magnitude of the Si²⁹ NMR signals observed by the fast adiabatic passage method,¹ and consequently also the degree of polarization P_n of the nuclei, depended on the wavelength λ of the pump light. Plots of P_n against λ , obtained at two values of the magnetic field, $H_0 = 1500$ Oe and $H_0 = 20$ Oe are shown in Figs. 1(a) and 1(b). We note that the largest NMR signals of the Si²⁹ nuclei observed in these experiments were 10-15 times larger than the equilibrium NMR signals obtained in the same sample in a magnetic field of 5 kOe at 300 K.

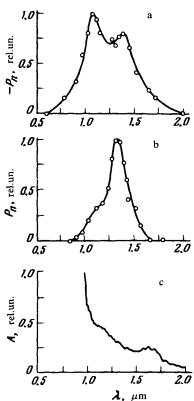


FIG. 1 Dependence of the degree of optical polarization P_n of Si²⁹ nuclei on the wavelength λ of the pump light in a magnetic field 1500 Oe (a) and 20 Oe (b); c—optical absorption in silicon compensated with samarium.

The experimental plots shown in Figs. 1(a) and (b) demonstrate that dynamic polarization of the Si²⁹ nuclei occurs when the crystal is illuminated with light having a quantum energy lower than the width of the forbidden band of silicon ($E_g \approx 1.19$ eV, $\lambda_g \approx 1.04 \,\mu\text{m}$). Thus, in a magnetic field $H_0 = 1500$ Oe [see Fig. 1(a)] optical polarization of the Si²⁹ nuclei is observed when the sample is exposed to light with $\lambda \approx 1.7 \,\mu\text{m}$, corresponding to a transition of the electrons from the impurity level of samarium ($E_v + 0.45$ eV, Ref. 2), to the conduction band. The presence of such phototransitions is evidenced by the spectrum of the optical absorption in the investigated silicon sample, obtained at T = 77 K with a two-beam spectrophotometer, and shown in Fig. 1(c). This spectrum reveals an increase of the light absorption with wavelengths less than 1.75 μ m.

With decreasing pump wavelength, the degree of optical polarization P_n of the nuclei increases and reaches a maximum at $\lambda \approx 1.4~\mu m$ [Fig. 1(a)], after which it decreases somewhat. The decrease of P_n when λ is varied from 1.4 to 1.25 μm is due to the fact that the electrons, which are not at equilibrium with respect to spin and are excited from the samarium level into states higher than the bottom of the conduction band, assume an equilibrium state when they are thermalized at the bottom of the conduction band. It is known^{1.3} that their nuclei are dynamically polarized when they interact with electron spins that are not at equilibrium. Consequently, when the wavelength of the pump light decreases the degree of optical polarization of the nuclei decreases.

With further decrease of λ , electron transitions from the valence band to the conduction band begin to contribute to the optical polarization of the Si²⁹ nuclei. This corresponds to the second maximum of P_n at $\lambda \approx 1 \,\mu\text{m}$ in Fig. 1(a). The decrease of P_n at $\lambda < 1 \,\mu\text{m}$ is also connected with the relaxation of the spins of the photoexcited "hot" electrons when the latter become thermalized.

The direction of the nuclear magnetization which is produced by optical pumping with unpolarized light of varying wavelength in a magnetic field 1500 Oe is opposite to the direction of the equilibrium magnetization of the Si²⁹ nuclei. This corresponds to a contact interaction of the Si²⁹ nuclei with electrons captured from the conduction band by the shallow phosphorus donor levels.³

If the optical pumping is produced in a magnetic field $H_0 = 20$ Oe, then the spectral dependence of the degree of optical polarization of the Si²⁹ nuclei [Fig. 1(b)] changes significantly. The direction of the nuclear magnetization produced in this case is the opposite of the direction of magnetization of the Si²⁹ nuclei optically pumped in a magnetic field 1500 Oe, and agrees with the direction of the equilibrium nuclear magnetization; this is evidence of the dipole-dipole character of the hyperfine interaction of the Si²⁹ nuclei with the electron spins.¹ Dipole-dipole interaction predominates for the electrons captured by deep impurity levels and leads to polarization of the Si²⁹ in weak magnetic fields.³ Thus, the optical polarization of the Si²⁹ nuclei optically pumped in a magnetic field 20 Oe is due to their interaction with electrons captured from the conduction band on the samarium level $(E_n + 0.45 \text{ eV})$. As seen from Fig. 1(b), dynamic polarization of the Si²⁹ nuclei is predominantly the result of impurity absorption of light, and irradiation of a crystal in a 20-Oe magnetic field by light of wavelength $\sim 1 \,\mu\text{m}$, corresponding to the intrinsic absorption, hardly polarizes the Si²⁹ nuclei. The apparent reason lies in the different photoexcited-electron recombination processes which occur when they are excited from the impurity level of samarium and when they are excited from the valence band into the conduction band.

Thus, our investigations have shown that the dynamic polarization of the silicon lattice nuclei can arise not only in interband absorption of light, but also as a result of impurity absorption of light with quantum energy lower than the width of the forbidden band. We note also that in view of the small absorption coefficient of the "impurity" light the nuclei can be optically polarized in a large volume of the crystal.

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