

Optical orientation of Si^{29} nuclei in n -type silicon and its dependence on the pumping light intensity

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The dynamic polarization of Si^{29} nuclei in phosphorus-doped ($N_d \approx 1.5 \cdot 10^{13} \text{ cm}^{-3}$) n -type silicon induced by optical pumping has been investigated. The dependences of the polarization and spin-lattice relaxation time of the Si^{29} nuclei on the concentration of photoexcited electrons have been determined experimentally. The parts played by various nuclear relaxation mechanisms in light-irradiated silicon are discussed.

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INTRODUCTION

In his work on optical orientation in semiconductors, Lampel^[1] obtained dynamic polarization of Si^{29} nuclei in pure n -type silicon and detected the decrease in the nuclear relaxation time incident to excitation of conduction electrons by light. Moreover, only two papers^[1,2] have been published in which observation of optical orientation of free carriers and nuclear moments by a nuclear magnetic resonance (NMR) method is reported. In other studies^[3-6] optical orientation in semiconductors has been registered via the polarization of the recombination luminescence. However, this method is difficult to employ with certain semiconductors, e.g., with silicon. In addition, the luminescence method does not give enough information on the nuclear relaxation throughout the entire volume of the crystal.

The NMR and magnetic relaxation of Si^{29} nuclei in silicon has been fairly well studied in the absence of irradiation with light.^[7-9] These studies showed that the principal nuclear spin-lattice relaxation mechanisms in n -type silicon are the interactions of Si^{29} nuclei with conduction electrons^[7,8] and with electrons localized at donor centers.^[9] Bloembergen^[10] asserted that the localized electrons are the more effective, and this has been confirmed experimentally.^[11]

However, a silicon specimen in which the donor concentration is low may, under intense illumination, contain considerably more photoexcited conduction electrons than donor centers, and hence more conduction electrons than localized ones. Then it is of interest to know which of these two mechanisms will make the greater contribution to the nuclear relaxation rate under such conditions.

We have used an NMR method to investigate the optical orientation of Si^{29} nuclei in n -type silicon, and have used the results of that study to examine the nuclear relaxation rate as a function of the number of photoexcited electrons. We have also investigated the effect of the light intensity of the limiting degree of dynamic polarization of Si^{29} nuclei in silicon.

EXPERIMENTAL TECHNIQUE

High-resistivity n -type silicon doped with phosphorus ($N_d \approx 1.7 \cdot 10^{13} \text{ cm}^{-3}$) was used in the experiments. The

specimen was a single-crystal silicon plate 14 mm in diameter and 1 mm thick having polished faces.

The specimen was irradiated in a magnetic field of strength $H_0 = 4.9 \text{ G}$ at temperature $T = 77 \text{ K}$ with the unpolarized light from a 1-kW incandescent lamp. For irradiation the specimen was lowered into an extension of a glass Dewar flask containing liquid nitrogen, which was placed in the gap of a permanent magnet. In this case the light beam was perpendicular to the magnetic field H_0 .

The specimen was also irradiated with circularly polarized light. For this an infrared polaroid and a quarter-wave plate for $1\text{-}\mu\text{m}$ waves were placed in the Dewar flask in the path of the light beam before it reached the silicon crystal. In this case the light beam was parallel to a $\sim 5 \text{ G}$ magnetic field produced by Helmholtz coils.

The duration of the irradiation was varied, ranging from 2 to 24 hr. After irradiation, the specimen was warmed to room temperature and placed in the magnet of an NMR radiospectrometer. The magnetization of the Si^{29} nuclei that accumulated during the light irradiation did not change during this operation since the Si^{29} nuclei in our specimen have a long spin-lattice relaxation time at room temperature ($\sim 5 \text{ hr}$) and the specimen was transferred in the earth's magnetic field, which is stronger than the local field in silicon ($H_{\text{loc}} \approx 0.13 \text{ G}$). No more than 3-5 min were required for transferring the specimen and measuring the nuclear magnetization.

The magnetization of the Si^{29} nuclei was measured with a type RYa-2301 broad-line radiospectrometer with crossed coils, using the rapid adiabatic transit method.^[12] The crossed coils are very sensitive and make it possible both to record the Si^{29} NMR signals even with the low abundance of Si^{29} in natural silicon (4.7%) and to determine the sign of the nuclear magnetization.

During illumination of the silicon the magnetization of the Si^{29} nuclei becomes gradually established, following an exponential law with a time constant equal to the spin-lattice relaxation time T_1 of the Si^{29} nuclei. We measured the nuclear magnetization at a fixed light intensity, using exposures of various lengths. By extrapolating the measurements to infinite irradiation

time we could determine the limiting relative nuclear magnetization $\langle I_z \rangle / I_0$ and the relaxation time T_1 of the Si^{29} nuclei.

The accuracy of the measurements and the accuracy achieved in determining $\langle I_z \rangle / I_0$ and T_1 are mainly determined by the signal-to-noise ratio in recording the Si^{29} NMR signals, and this decreases considerably with decreasing light intensity.

In our experiments the light intensity was varied by adjusting the voltage on the lamp. The change in the concentration of photoexcited electrons after changing the light intensity was determined from the change in the resistivity of the specimen on turning on the light; it ranged from $5 \cdot 10^{13}$ to $5 \cdot 10^{14} \text{ cm}^{-3}$.

RESULTS AND DISCUSSION

1. Effect of the intensity of light irradiation on the polarization of Si^{29} nuclei in *n*-type silicon

Figure 1 shows the relative polarization $\langle I_z \rangle / I_0$ of the Si^{29} nuclei as a function of the concentration n_{ph} of photoexcited electrons. The full curve was calculated (see below). The experimental points on Fig. 1 show that irradiating *n*-type silicon containing phosphorus at an atomic concentration of $1.7 \cdot 10^{13} \text{ cm}^{-3}$ in a 4.9-kG magnetic field at 77 K results in dynamic polarization of the Si^{29} nuclei, and that the nuclear magnetization is opposite in direction to the equilibrium Boltzmann magnetization I_0 in the same field at the same temperature. This indicates the presence of the Overhauser effect,^[12] since the magnetic moment of the Si^{29} nucleus is negative. The figure also shows that the polarization of the Si^{29} nuclei in our specimen remains virtually unchanged as the concentration n_{ph} of photoexcited electrons falls from $5 \cdot 10^{14} \text{ cm}^{-3}$ to $2 \cdot 10^{14} \text{ cm}^{-3}$. In this region the nuclear polarization is enhanced as compared with its equilibrium value by the factor -1.86 . With further decrease in the light intensity, and therefore in n_{ph} , the nuclear polarization also decreases.

Let us consider the factors that affect the polarization of the Si^{29} nuclei in silicon under irradiation with light. Irradiating silicon with photons of energy equal to or greater than the width of the forbidden gap ($E_g = 1.19 \text{ eV}$ at 77 K) leads to the appearance of photoexcited electrons in the conduction band. Then the average projection $\langle S_x \rangle$ in the direction of the external magnetic field

H_0 of the spins of the electrons in the conduction band may differ from the equilibrium value S_0 determined by the Boltzmann difference in the populations of the magnetic sublevels with spin projections $+\frac{1}{2}$ and $-\frac{1}{2}$.

As Lampel showed,^[1] the interaction of these electrons with the Si^{29} nuclei leads to dynamic polarization of the latter.¹⁾ In this case the stationary value of the nuclear polarization is determined by the equation^[12]

$$\left(\frac{\langle I_z \rangle}{I_0} \right)_m = 1 - \frac{\gamma_e S_0 - \langle S_x \rangle}{\gamma_n S_0} f, \quad (1)$$

in which γ_e and γ_n are the gyromagnetic ratios of the electron and nucleus, respectively ($\gamma_e/\gamma_n = 3310$), and $f \leq 1$ is the nuclear polarization loss factor, which in our case, as in Lampel's work,^[1] can be taken as unity.

On irradiating the specimen with unpolarized light equal numbers of electrons with spin projections $S_x = +\frac{1}{2}$ and $S_x = -\frac{1}{2}$ are generated in the conduction band. As a result of light irradiation and relaxation, an average value $\langle S_x \rangle_{\text{ph}}$ of the spin projection of the photoexcited electrons is established^[11]:

$$\langle S_x \rangle_{\text{ph}} = S_0 \frac{\tau}{\tau + \tau_s} = \frac{n_{\text{ph}}^+ - n_{\text{ph}}^-}{2n_{\text{ph}}} \frac{\tau}{\tau + \tau_s}, \quad (2)$$

where $S_0 = (n_{\text{ph}}^+ - n_{\text{ph}}^-)/2n_{\text{ph}}$ is the average electron-spin projection at the Boltzmann equilibrium, n_{ph}^+ and n_{ph}^- are the equilibrium concentrations of electrons with spin projections $+\frac{1}{2}$ and $-\frac{1}{2}$, respectively, τ is the lifetime of a photoexcited electron, and τ_s is the spin-lattice relaxation time of the photoexcited electrons.

The conduction band will contain not only photoexcited electrons, but also thermal electrons, whose concentration n_0 at 77 K is roughly equal to the concentration of phosphorus donor atoms. The average spin projection for these electrons is $S_0 = (n_0^+ - n_0^-)/2n_0$.

Let us determine the value of $\langle S_x \rangle$ for all the electrons in the conduction band. Using (2), we have

$$\langle S_x \rangle = \frac{n^+ - n^-}{2(n_0 + n_{\text{ph}})} = \frac{1}{2(n_0 + n_{\text{ph}})} \left[n_0^+ - n_0^- + (n_{\text{ph}}^+ - n_{\text{ph}}^-) \frac{\tau}{\tau + \tau_s} \right] = \frac{S_0}{n_0 + n_{\text{ph}}} \left(n_0 + n_{\text{ph}} \frac{\tau}{\tau + \tau_s} \right). \quad (3)$$

Then substituting this value into (1), we obtain the following expression for the limiting nuclear polarization:

$$\left(\frac{\langle I_z \rangle}{I_0} \right)_m = 1 - \frac{\gamma_e \tau_s}{\gamma_n \tau + \tau_s} \frac{n_{\text{ph}}}{n_0 + n_{\text{ph}}}. \quad (4)$$

It is evident from Eq. (4) that in the absence of light irradiation, when $n_{\text{ph}} = 0$, the nuclear magnetization $\langle I_z \rangle$ will be equal to its equilibrium value I_0 . At high light intensities ($n_{\text{ph}} \gg n_0$) Eq. (4) reduces to $\langle I_z \rangle / I_0 = 1 - (\gamma_e/\gamma_n)\tau_s/(\tau + \tau_s)$, and this expression is the same as that given by Lampel.^[1]

The n_{ph} dependence of $\langle I_z \rangle / I_0$ as calculated from Eq. (4) with $n_0 = 1.7 \cdot 10^{13} \text{ cm}^{-3}$ is shown by the curve in Fig. 1. The value $\tau_s/(\tau + \tau_s) = 0.9 \cdot 10^{-3}$ was determined from the measured value $\langle I_z \rangle / I_0 = -1.86$ for the maximum light intensity ($n_{\text{ph}} = 5 \cdot 10^{14} \text{ cm}^{-3}$) when $n_{\text{ph}}/(n_0 + n_{\text{ph}}) \approx 1$.

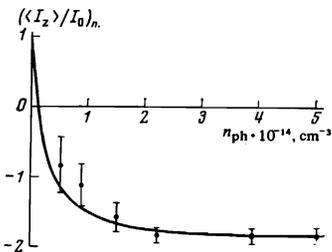


FIG. 1. Limiting dynamic polarization of the Si^{29} nuclei $\langle I_z \rangle / I_0$ vs photoexcited-electron concentration n_{ph} in *n*-type silicon ($N_d \approx 1.7 \cdot 10^{13} \text{ cm}^{-3}$) irradiated with unpolarized light in a 4.9 kG magnetic field at 77 K. The curve was calculated with Eq. (4).

Thus, it follows from the above considerations that the limiting polarization of the Si²⁹ nuclei in *n*-type silicon is determined by the average spin projection of the aggregate of photoexcited and thermal electrons in the conduction band and, in the final analysis, depends on the concentration ratio n_{ph}/n_0 (Eq. (4)).

2. Effect of light intensity on the spin-lattice relaxation time of the Si²⁹ nuclei

As was noted in the Introduction, the principal relaxation mechanisms for Si²⁹ nuclei in silicon are: a) interaction of the Si²⁹ nuclei with conduction electrons, which are uniformly distributed throughout the entire crystal; we denote the relaxation rate due to this process by $1/T_{1e}$; and b) interaction of the Si²⁹ nuclei with electrons localized at donor centers; we denote the relaxation rate due to this mechanism by $1/T_{1d}$. The total spin-lattice relaxation time T_1 for these nuclei is determined by the equation $1/T_1 = 1/T_{1e} + 1/T_{1d}$.

Figure 2a shows the experimental n_{ph} dependence of T_1 obtained on irradiating silicon with unpolarized light in a 4.9 G field H_0 at 77 K. It will be seen that T_1 remains constant as n_{ph} falls from $5 \cdot 10^{14}$ to $2 \cdot 10^{14}$ cm⁻³. As n_{ph} falls further, the relaxation time rises. In the absence of light irradiation ($n_{ph} = 0$) T_1 was greater than 100 hr.

Let us consider the contribution from each of the mechanisms to the spin-lattice relaxation of the Si²⁹ nuclei in more detail.

a) *The conduction-electron contribution.* The interaction of the conduction electrons with the Si²⁹ nuclei is a Fermi-Segre contact interaction between the nuclear spin **I** and the electron spin **S**. The relaxation time of the Si²⁹ nuclei under this interaction was calculated by Bloembergen^[10] and the calculation was improved by Abragam^[12]:

$$\frac{1}{T_{1e}} = \left(\frac{8\pi}{3} \gamma_e \gamma_n \hbar \eta \right)^2 n_e N_c \frac{\hbar}{kT}, \quad (5)$$

where η is the degree of localization of the electron wave function at the Si²⁹ site ($\eta = 186$ ^[7]), n_e is the conduction-electron concentration, N_c is the equivalent density of states in the conduction band ($N_c = 3.6 \cdot 10^{18}$ cm⁻³ at 77 K), k is Boltzmann's constant, and $\hbar = h/2\pi$ is Planck's constant.

Equation (5) gives a good account of the nuclear relaxation rate in silicon for $n_e > 10^{15}$ cm⁻³^[7,8] at $T = 300$ K. The contribution to the relaxation rate from deep paramagnetic impurity centers becomes significant as the conduction-electron concentration falls. However, the contribution from these impurities is negligible at 77 K if the silicon is pure enough.^[11]

As is evident from (5), under the conditions of our experiment the relaxation rate due to the interaction of the Si²⁹ nuclei with the photoexcited conduction electrons should increase with increasing concentration of the latter. However, no such increase in the Si²⁹ relaxation is observed in the region $n_e \approx n_{ph} > 2 \cdot 10^{14}$ cm⁻³ (Fig. 2a).

b) *Relaxation at donor centers.* In accordance with

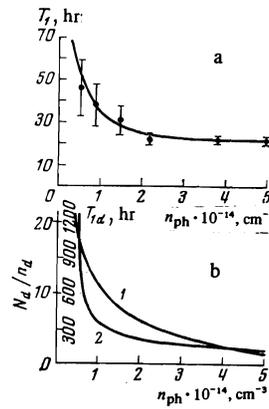


FIG. 2. a) Experimental dependence of the spin-lattice relaxation time T_1 of Si²⁹ nuclei in *n*-type silicon on the concentration n_{ph} of photoexcited electrons; and b) Calculated curves showing the reciprocal degree of fillings of donor centers N_d/n_d (curve 1) and the nuclear relaxation time T_{1d} (curve 2) as functions of the photoexcited electron concentration n_{ph} .

Lampel's results,^[11] let us consider the relaxation of the Si²⁹ nuclei due to interaction with electrons localized at donor centers. The nuclear relaxation in silicon at 77 K is due to contact interaction between electrons captured by phosphorus atoms and the closest Si²⁹ nuclei. Thermal excitation of electrons from donor levels to the conduction band results in time modulation of the contact interaction with a frequency of $\sim 10^{10}$ sec⁻¹. Calculation of the relaxation time for Si²⁹ nuclei lying close to a donor atom gives the following result^[11,12]:

$$\frac{1}{T_{1d}(r_I)} = A \frac{n_d}{N_d} \frac{\tau_c}{1 + \omega_e^2 \tau_c^2} \exp\left(-\frac{4r_I}{b^*}\right), \quad (6)$$

$$A = \frac{1}{2} \left(\frac{8\pi}{3} \gamma_e \gamma_n \hbar \frac{\eta}{b^{*3}} \right)^2,$$

where n_d is the concentration of electrons captured at donor atoms, N_d is the donor-atom concentration, ω_e is the electron Larmor frequency in the magnetic field H_0 , b^* is the Bohr radius of the orbit of an electron in the ground state of the donor atom (for phosphorus in silicon, $b^* = 17 \text{ \AA}$), τ_c is a correlation time that depends on the probability ω_d for thermal excitation of an electron from a donor atom to the conduction band and on the probability ω_{ex} for spin-flip exchange processes ($1/\tau_c = \omega_d + \omega_{ex}$), and r_I is the distance between the nucleus with spin **I** and the electron with spin **S**. In our case, i. e., at relatively low light-irradiation intensities, $\omega_d > \omega_{ex}$ and τ_c is determined mainly by ω_d .

Now let us consider how the relaxation time $T_{1d}(r_I)$ varies with the light intensity. It is easily seen that the quantities appearing in the expression for A are determined by the properties of the medium and are independent of the irradiation. The number of electrons captured at donor centers, on the other hand, does depend on the light intensity.

The concentration of captured electrons is determined by the same Fermi quasilevel as is the number of electrons in the conduction band:

$$n_d = N_d \left[1 + \frac{1}{2} \exp \frac{E_d - E_F}{kT} \right]^{-1}, \quad (7)$$

where $E_d = 10.045$ eV is the energy of the ground state of a phosphorus atom in silicon (energies are reckoned from the bottom of the conduction band). The energy E_F of the Fermi quasilevel can be determined when the conduction-electron concentration $n_{ph} + n_0$ is known:

$$n_{ph} + n_0 = N_c \exp \frac{E_p}{kT}. \quad (8)$$

Eliminating E_p from (7) with the aid of (8), we obtain

$$\frac{n_d}{N_d} = \left[1 + \frac{N_c}{2(n_{ph} + n_0)} \exp \frac{E_d}{kT} \right]^{-1}. \quad (9)$$

The calculated n_{ph} dependence of $(n_d/N_d)^{-1}$ is shown as curve 1 on Fig. 2b.

It can be shown that ω_d , which occurs in (6) (through τ_c), is independent of n_{ph} . Actually, under conditions of dynamic equilibrium, the number of electrons excited from phosphorus atoms per unit time is equal to the number of electrons arriving at those atoms per unit time from the conduction band:

$$n_d \omega_d = (n_{ph} + n_0) \langle v \rangle \sigma_{cap} (N_d - n_d), \quad (10)$$

where σ_{cap} is the cross section for capture of a free electron by an ionized phosphorus atom, $N_d - n_d$ is the concentration of ionized phosphorus atoms, and $\langle v \rangle$ is the thermal velocity of the electrons.

Using Eq. (9) we obtain

$$\omega_d = (n_{ph} + n_0) \langle v \rangle \sigma_{cap} \frac{N_c}{2(n_{ph} + n_0)} \exp \frac{E_d}{kT} = \frac{1}{2} \sigma_{cap} \langle v \rangle \exp \left(\frac{E_d}{kT} \right) N_c.$$

In the first approximation, ω_d can be assumed to be independent of the light intensity. Using data from^[13,14] we take $\tau_c \approx 1/\omega_d \approx 0.2 \cdot 10^{-10}$ sec. Thus, the relaxation rate for Si²⁹ nuclei lying close to donor atoms is mainly determined by the probability n_d/N_d that a donor center is occupied, and this, in turn, depends on the light intensity.

To determine the total relaxation time T_{1d} due to donor centers for all the Si²⁹ nuclei in the silicon crystal we must take account of diffusion of nuclear spin,^[15] as a result of which nuclear polarization propagates from the relaxation centers into all parts of the crystal. The solution of the diffusion equation for the case of free spin diffusion^[15] leads to the following expression for T_{1d} :

$$\frac{1}{T_{1d}} = 4\pi N_d \int_0^\infty \frac{r^2 dr}{T_{1d}(r)}, \quad (11)$$

where $T_{1d}(r)$ is given by Eq. (6) and ρ is the diffusion radius, which can be found from the equation

$$D/a^2 = 1/T_{1d}(\rho), \quad (12)$$

in which D is the diffusion coefficient ($D = 2.4 \cdot 10^{-14}$ cm²/sec for silicon) and a is the distance between Si²⁹ nuclei ($a = 6.5$ Å).

The quantity ρ , defined by Eq. (12), is the distance from a donor center at which the nuclear spin-flip probability due to direct relaxation ($1/T_{1d}(\rho)$) is equal to the nuclear spin-flip probability due to spin-spin interaction between two nearest Si²⁹ nuclei (D/a^2). Using Eqs. (6) and (12), we obtain

$$\rho = \frac{b^*}{4} \ln \frac{B n_d a^2}{N_d D},$$

$$B = A/\omega_d [1 + (\omega_c/\omega_d)^2]. \quad (13)$$

Then, performing the integration in (11), we find the relaxation time T_{1d} for Si²⁹ nuclei:

$$1/T_{1d} = \pi N_d b^* D [1 + (1 + 4\rho/b^*)^2]. \quad (14)$$

The n_{ph} dependence of T_{1d} as calculated from Eq. (14) is shown as curve 2 on Fig. 2b.

On comparing this curve with the experimental n_{ph} dependence of the total spin-lattice relaxation time T_1 of the Si²⁹ nuclei (Fig. 2a) we see that, despite the quantitative discrepancy, which is apparently due to some factors that affect the relaxation rate but were not taken into account (e.g., dipole-dipole interaction between Si²⁹ nuclei and localized electrons, generation of deep paramagnetic centers by light), Eq. (14) gives a good account of the limitation of the relaxation time on increasing the photoexcited electron concentration.

We note that a limitation of the relaxation time of Si²⁹ nuclei with increasing n_{ph} was also observed when pumping with circularly polarized light in a weak magnetic field ($H_0 \sim 5$ G). Here, however, T turned out to be ~ 8 hr at maximum pumping light intensity; this is associated with the dependence of the Si²⁹ relaxation rate on the magnetic field (see Eq. (6), where $\omega_e = \gamma_e H_0$).

Thus, the main results of the present study are as follows:

1. It has been established that electrons captured at donor centers make the main contribution to relaxation of Si²⁹ nuclei in *n*-type silicon even under conditions in which there are many times more conduction electrons than localized ones. This permits the conclusion that the relaxation of the Si²⁹ nuclei on the localized electrons is at least two orders of magnitude more effective than their relaxation on conduction electrons. This conclusion is also confirmed by the fact that no optical polarization of Si²⁹ nuclei in boron-doped *p*-type silicon containing no phosphorus has been detected.

2. It has been found that the decrease of the spin-lattice relaxation time of the Si²⁹ nuclei with increasing concentration of photoexcited electrons is limited, and it has been shown that this limitation is due to filling to saturation of the donor centers by electrons.

3. It has been established that the limiting degree of dynamic polarization of the Si²⁹ nuclei depends on the pumping light intensity. It has been shown both theoretically and experimentally that the limiting degree of nuclear polarization is determined by the spin polarization of both the photoexcited and the thermal electrons in the conduction band.

¹⁾As will be shown below, the dynamic polarization of the Si²⁹ nuclei takes place as a result of their interaction with electrons localized at donor centers. However, the average spin projection for these electrons can be regarded as equal to the average spin projection for free electrons, since at 77 K frequent transitions of electrons back and forth between the donor levels and the conduction band take place and the probability for these transitions ($\sim 10^{10}$ sec⁻¹) is much greater

than the probability for electron spin flip ($1/\tau_s \sim 10^8 \text{ sec}^{-1}$). Moreover, the Zeeman interaction of a localized electron with the external magnetic field $H_0 = 4.9 \text{ kG}$ is stronger than its Zeeman interaction with the nucleus of the phosphorus donor atom,^[9] so we can speak of the spin state of the localized electrons in the external magnetic field.

¹Georges Lampel, Phys. Rev. Lett. 20, 491 (1968).

²B. P. Zakharchenya, M. E. Kompan, and V. G. Fleisher, Pis'ma Zh. Eksp. Teor. Fiz. 19, 734 (1974) [JETP Lett. 19, 378 (1974)].

³R. R. Parsons, Phys. Rev. Lett. 23, 1152 (1969).

⁴B. P. Zakharchenya, V. G. Fleisher, R. I. Dzhoiev, Yu. P. Veshchunov, and I. B. Rusanov, Pis'ma Zh. Eksp. Teor. Fiz. 13, 195 (1971) [JETP Lett. 13, 137 (1971)].

⁵A. I. Ekimov and V. I. Safarov, Pis'ma Zh. Eksp. Teor. Fiz. 12, 293 (1970) [JETP Lett. 12, 198 (1970)].

⁶M. I. D'yakonov, V. I. Perel', V. L. Berkovits, and V. I. Safarov, Zh. Eksp. Teor. Fiz. 67, 1912 (1974) [Sov. Phys. JETP 40, 950 (1974)].

⁷R. G. Shulman and V. J. Wyluda, Phys. Rev. 103, 1127 (1956).

⁸Georges Lampel and Ionel Solomon, C. R. Acad. Sci. (Paris) 258, 2289 (1964).

⁹G. Feher, Phys. Rev. 114, 1219 (1959).

¹⁰N. Bloembergen, Physica 20, 1130 (1954).

¹¹Georges Lampel, Theses, Paris, Orsay, 1968.

¹²A. Abragam, The principles of nuclear magnetism, Oxford, 1961 (Russ. transl. IIL, 1963).

¹³M. Loewenstein and A. Honig, Phys. Rev. 114, 781 (1966).

¹⁴W. D. Davis, Phys. Rev. 114, 1006 (1959).

¹⁵G. R. Khutsishvili, Usp. Fiz. Nauk 87, 211 (1965) [Sov. Phys. Usp. 8, 743 (1966)].

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Some features of x-ray fluorescence in metals near the absorption threshold

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An expression for the x-ray inelastic scattering cross section in metals is derived by taking into account interference of hole states localized at different centers. It is demonstrated that the shape of the spectral band depends on the initial photon frequency ω if the latter is near to the inner level ionization threshold. A shift of the fluorescence threshold is found in alkali metals.

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I. INTRODUCTION

We have previously^[1] considered the dependence of the emission spectrum of a diatomic homonuclear molecule on the frequency ω of the exciting photon. This dependence is due to interference between the x-ray-excited states with holes localized on different atoms. The purpose of the present study was to investigate the influence of this interference on the shape of the x-ray emission band of a metal. The role of the interference effects was qualitatively discussed earlier by Nozieres and Abrahams,^[2] who considered the scattering of a soft x-ray photon in a metal. Nozieres and Abrahams^[2] investigated inelastic scattering of a photon in a metal with allowance for the interaction of the electrons of the valence band with the hole in the internal shell. In view of the complexity of the many particle formulation of the problem, Nozieres and Abrahams actually confined themselves to an investigation of the singularities of the photon scattering cross section in the region

$$\omega - \omega_0 \lesssim \gamma \sim 0.1 \text{ eV} \quad (1)$$

and (or)

$$\omega_0 - \omega' \lesssim \gamma,$$

where the interference effects, in their opinion, are ap-

preciable. Here $\omega_0 = E_F - E_0$, ω, ω' are the frequencies of the initial and final photons; E_F and E_0 are the Fermi energy and the energy of the internal level. γ is the natural width of the x-ray line. In the present paper, confining ourselves to the single-particle approximation, we investigate the cross section for the scattering of an x-ray photon in a metal outside the narrow interval (1). The existing single-particle calculations^[3] of emission bands in a metal agree well with experiment, including the region (1) and the low-energy tails of the emission band. Skinner^[4] attributes these tails to the energy dependence of the lifetime of the single-particle state in the valence bands; for the state at the bottom of the valence band this time is much less than near the Fermi level. Within the framework of the single-particle description, this effect can be easily taken into account by introducing the corresponding damping parameter γ .^[5]

2. K FLUORESCENCE

I. We consider *K* fluorescence of a metal. The cross section for inelastic scattering of a photon by the system is described by the Kramers-Heisenberg formula.^[6] Using the dipole approximation for the atomic matrix element and neglecting the width of the x-ray line, we obtain