Relaxation of conduction-electron magnetization in a metallic plate with different types of surfaces

I. G. Zamaleev and V. N. Lisin

Physicotechnical Institute, Kazan' Division, USSR Academy of Sciences (Submitted 3 August 1980) Zh. Eksp. Teor. Fiz. 80, 1217-1227 (March 1981)

Using as an example a metallic layer of Na with a scatterer sublayer (Ta or Nb) on one of the surfaces, the influence of spin scattering by a part of the sample surface on the relaxation of the magnetization of the conduction electrons is investigated theoretically and experimentally by the conduction-electron parametric-resonance (CEPR) method. Additivity of the contribution of the surface scattering to the total relaxation rate is demonstrated, and an analytic expression is obtained for the surface relaxation time. This expression is valid for arbitrary values of the probability ε of electron spin flip in a single collision with the surface. This time depends on the diffusion coefficient, on the thickness, and on ε , and is inversely proportional to the additional broadening of the CEPR signal. The proportionality coefficient is ≈ 1 . The size and temperature dependences of the CEPR line width are used to determine the diffusion coefficient in the Na volume ($D = 50 \pm 10$ cm² sec⁻¹) and the values of ε ($\varepsilon \ge 8 \times 10^{-2}$ for Na-Ta and $\varepsilon = 8 \times 10^{-3}$ for Na-Nb), with ε independent of temperature. The numerical values of ε are governed by the spin-orbit scattering from the interface and not in the interior of the scatterer. A possible scattering mechanism is proposed.

PACS numbers: 72.10.Fk, 72.15.Qm, 76.30.Pk, 75.70.Dp

1. INTRODUCTION

Because of the high mobility of the conduction electrons in metals, the parameters of the conductionelectron paramagnetic resonance (CEPR) depend on the state of the surface. The CEPR is therefore one of the methods of investigating surface properties. In addition, in the case of strong spin scattering on the surface, an experimental determination of the diffusion coefficient of the conduction electrons in the interior of the metal is possible.¹

The existing theories $^{2-4}$ of the CEPR line shape, which take into account the spin surface scattering of the conduction electrons, were developed for the case when the probability ε of the spin flip in a single collision with the boundary is constant over the entire surface. In the case of strong surface scattering, the magnetization amplitude first decreases and becomes nonresonant on the entire sample boundary. As a result the signal shape is strongly distorted, and the signal amplitude decreases. This makes difficult the observation of the CEPR signal itself, as well as the physical interpretation of the line parameters. For investigations by the CEPR method it is therefore preferable, in our opinion, to cause the conduction-electron spins to be scattered by only part of the sample surface, say from one side of a metallic layer. Then, if the scattering is strong, the CEPR signal is determined by the value of the magnetization of the conduction electron on the nonscattering surface and is neither decreased in amplitude nor distorted in shape. The scattering surface produces an effect because the conduction electrons diffuse only during the time of the spin relaxation, and this leads to broadening of the CEPR line. Conversely, the additional broadening of the CEPR signal can be used to measure directly the time of surface relaxation of the conduction-electron magnetization on the nonscattering surface. The aim of the present paper is a theoretical and experimental study, by the CEPR method, of the surface relaxation in plates with differ-

ent types of surface, using as an example a layer of metallic sodium with a spin-orbit scatterer (Ta, Nb) on one of the boundaries, as well as the determination of the coefficient of diffusion of the conduction electrons in the volume and of the parameters of the interaction of the conduction electrons with a metal-metal interface. The choice of sodium was dictated by the fact that the CERP signal in it has been well investigated; in addition, its Fermi surface is almost spherical and it is of interest to compare the experimental value of the diffusion coefficient of the conduction electrons with that calculated by the free-electron model. Tantalum and niobium were chosen because of their poor solubility in sodium and because of the expected large spin-orbit interaction constant, which is needed for the determination of the conduction-electron diffusion coefficient.

2. THEORY

In the CEPR method one measures directly the derivative, with respect to the constant magnetic field, of the absorbed microwave power averaged over the period of the oscillations. To calculate the latter we must know the amplitude of the electric field on the surface of the sample, determined by simultaneous solution of Maxwell's equations in the case of the normal skin effect and the Bloch equations for the magnetization. According to the experimental conditions, the equations were solved for a metallic layer of thickness d, located in the region $-d/2 \le z \le d/2$, with the alternating microwave field outside the sample and parallel to the surface. The constant magnetic field was chosen, for simplicity, to be perpendicular to the surface, since the experimentally observed CEPR signal was independent of the orientation of the constant magnetic field. To make the result general, the boundary conditions for the microwave field amplitude $\mathbf{H}_{\omega} \exp(-i\omega t) + \mathbf{H}_{-\omega} \exp(i\omega t)$ were chosen in the form

$$\mathbf{H}_{s} = \mathbf{H}_{-s} = b\mathbf{H}_{1}, \quad z = -d/2;$$

$$\mathbf{H}_{s} = \mathbf{H}_{-s} = \mathbf{H}_{1}, \quad z = d/2,$$

where b is a numerical constant; $\omega/2\pi$, $2bH_1$, and $2H_1$ are frequency and the amplitudes of the microwave field on the surfaces z = -d/2 and z = d/2. The spin scattering of the conduction electrons by the surface was taken into account phenomenologically with the aid of the boundary condition for the deviation of the transverse component of the magnetization from the instantaneous local equilibrium value on the surface²⁻⁵:

$$\varepsilon^{\mp} \frac{v_{F}}{2D} (\mathbf{M}_{o} - \chi \mathbf{H}_{o}) = \pm \frac{\partial}{\partial z} (\mathbf{M}_{o} - \chi \mathbf{H}_{o}), \quad \varepsilon^{\mp} \frac{\varepsilon^{\mp}}{1 - \varepsilon^{\mp}};$$

here \mathbf{M}_{ω} and \mathbf{H}_{ω} are the amplitudes of the magnetization and of the microwave field, χ is the statistical susceptibility, ε^{\pm} are the probabilities of spin flip in a single collision of the conduction electron with the surface z = -d/2 and z = d/2, respectively; $v_{\mathbf{F}}$ and D are the Fermi velocity and the diffusion coefficient of the conduction electrons in the volume.

The expression obtained for the derivative of the resonant part of the absorbed microwave power is of the form

$$\partial P/\partial H = \chi H_1^2 \omega SZ, \tag{1a}$$

where S is the area of one boundary of the layer, Z is defined by

$$Z = \operatorname{Re} \frac{\frac{1}{k_{1}} \frac{\partial}{\partial H} \frac{\omega}{D(k_{2}^{*} - k_{1}^{*})}}{\left\{\frac{\partial}{\partial k_{1}} \left[\left(\frac{1+b}{2}\right)^{2} F_{1} + \left(\frac{1-b}{2}\right)^{2} G_{1}\right]\right]}$$
$$+ \frac{2k_{1}}{k_{2}^{2} - k_{1}^{2}} \left[\frac{1+b}{2} (F_{1} - F_{2})A + \frac{1-b}{2} (G_{1} - G_{2})B\right]\right\}.$$
(1b)

The first term is due here to modulation of the depth of the skin layer by the conduction-electron magnetization, and the second is due to the nonlocal coupling of the magnetization with the microwave field, owing to the diffussion of the conduction electrons. The following notation was assumed in (1b)

$$F_{i,2} = k_{i,2} \operatorname{th} \left(\frac{k_{i,3}d}{2} \right), \quad G_{i,2} = k_{i,2} \operatorname{cth} \left(\frac{k_{i,3}d}{2} \right),$$

$$A = \frac{(1+b) \left(F_i^+ G_3^- + F_i^- G_3^+ \right) + (1-b) \left(G_i^+ G_3^- - G_i^- G_3^+ \right)}{2 \left(F_2^+ G_2^- + F_2^- G_3^+ \right)},$$

$$B = \frac{(1+b) \left(F_1^+ F_2^- - F_1^- F_2^+ \right) + (1-b) \left(G_1^- F_2^+ + G_1^+ F_2^- \right)}{2 \left(F_2^- + F_2^- G_3^+ \right)},$$

$$F_{i,2}^* = F_{i,2} + k_*^{\pm}, \quad G_{i,2}^{\pm} = G_{i,2} + k_*^{\pm},$$

$$k_1 = \frac{1-i}{\delta}, \quad k_2 = \frac{\eta + i\xi}{\delta_M}, \quad k_*^{\pm} = \xi^{\pm} \frac{v_F}{2D},$$

$$\eta = \left[\left(1 + x^2 \right)^{v_i} + 1 \right]^{v_i}, \quad \xi = \operatorname{sgn} x \left[\left(1 + x^2 \right)^{v_i} - 1 \right]^{v_i}, \quad x = (\Omega - \omega) T.$$
(2)

Here $\delta = (c^2/2\pi\sigma\omega)^{1/2}$ is the depth of the skin layer; $\delta_{H} = (2DT)^{1/2}$ is the depth of the "magnetic" skin layer; T is the time of the spin relaxation of the conduction electrons in the volume; $\Omega = \gamma H$ is the Zeeman frequency; γ is the absolute value of the gyromagnetic ratio; H is the constant external magnetic field. The expressions for the magnetization amplitudes $M_{\omega}^{-} = M_{\omega}^{x} - iM_{\omega}^{y}$ on the layer boundaries, which are needed for the physical analysis of the surface relaxation, are of the form

$$M_{\omega}^{-} - \chi b H_{1} = \chi H_{1} \frac{i\omega}{D(k_{2}^{2} - k_{1}^{2})} [b - (A - B)], \quad z = -\frac{d}{2},$$

$$M_{\omega}^{-} - \chi H_{1} = \chi H_{1} \frac{i\omega}{D(k_{2}^{2} - k_{1}^{2})} [1 - (A + B)], \quad z = \frac{d}{2}.$$
 (3)

We note that the quantity K_s^- in (2) can be rewritten in

a form similar to k_1 and k_2 :

$$k_s^-=1/\delta_s^-,$$

where the quantity $\delta_s^- = (2DT_s^-)^{1/2}$ will be called the depth of the "surface magnetic" skin layer on the surface z = -d/2, due to the spin scattering of the conduction electron by this surface; δ_s^- is the distance over which an "average" electron diffuses in a time τ_s^- and preserves its spin direction, where

 $\tau_{s}^{-}=2D/v_{F}^{2}(\tilde{\epsilon}^{-})^{2}$

has a simple physical meaning: this is the time during which an electron located at the surface z = -d/2 will collide with it $1/\varepsilon^-$ times and its spin will thus flip. We can introduce analogously the value of the surface magnetic skin layer of the surface z = d/2.

In particular cases when there is no surface relaxation ($\varepsilon^- = \varepsilon^+ = 0$) or with symmetric boundary conditions (b = 1, $\varepsilon^- = \varepsilon^+$), expression (1) for $\partial P/\partial H$ coincides with the results of Ref. 6 and Refs. 2 and 4 respectively, except that in place of the factor ω in (1b) the quantity in the derivative $\partial/\partial H$ (in Refs. 2 and 6) is $\Omega = \gamma H$, and this leads to a noticeable distortion of the form of $\partial P/\partial H$ in the case of broad lines, for example in thin samples with strong surface scattering. This difference is due to the fact that the authors of the cited references failed to take into account the fact that the magnetization of the conduction electrons relaxes not to zero but to a local equilibrium instantaneous value.

For dirty metals $(\delta_{\mu} \ll \delta)$, when the second term in (1b) can be neglected, the observed CEPR signal does not depend on the surface scattering. Thus, the CEPR signal is sensitive to the state of the surface only in pure metals with high mobility of the conduction electrons.

For pure metals

$$\delta_M \gg \delta$$
 (4)

with thickness larger than the depth of the skin layer

$$d \gg \delta$$
, (5)

expression (1) for $\partial P/\partial H$ can be expressed in terms of the amplitude of the magnetization (3) on the sample boundaries:

$$\frac{\partial P}{\partial H} = \operatorname{Re} \frac{\omega S}{ik_1} \frac{\partial}{\partial H} \left\{ \left[M_{\omega}^{-} \left(\frac{d}{2} \right) - \chi H_1 \right] H_1 + \left[M_{\omega}^{-} \left(-\frac{d}{2} \right) - \chi b H_1 \right] b H_1 \right\}.$$
(6)

In the derivation we have used here, in accord with (4) and (5), the inequalities

$$|F_1| = |G_1| = |k_1| \gg |F_2|, |G_2|, |k_2|$$

If the condition of strong surface relaxation is satisfied on the surface z = -d/2 and there is no scattering from the boundary z = d/2, then

$$\min(\delta_M, d) \gg \delta_{\bullet}^{-}; \tag{7}$$

 $\varepsilon^+=0$ when $F_2^-=G_2^-=k_s^-$, and the magnetization can in turn be expressed by

$$M_{\omega}^{-}\left(-\frac{d}{2}\right)-\chi bH_{1}=-\chi bH_{1}\frac{\omega\delta^{2}}{2D}\frac{k_{1}}{k_{s}^{-}},$$
(8a)

$$M_{\omega}^{-}\left(\frac{d}{2}\right) - \chi H_{4} = -\chi H_{4} \frac{\omega \delta^{2}}{2D} \frac{k_{1}}{k_{2}} \operatorname{th}(dk_{2}).$$
(8b)

It is seen from (8) that the amplitude of the magnetization on a strongly scattering surface z = -d/2 is nonresonant, and furthermore, when account is taken of (7), it is smaller in absolute value than the amplitude of the magnetization on the nonscattering surface by a factor δ_s^-/δ_W or δ_s^-/d . Therefore, in accord with (6), the observed CEPR signal is determined by the amplitude of the magnetization of the conduction electrons on the nonscattering surface.

To analyze the observed shape and width of the line, we consider particular cases of (6).

Bulk metal $(d \gg \delta_{\mathbf{k}})$. We replace $\tanh(d\mathbf{k}_2)$ in (8b) by unity. Substituting (8b) in (6), we obtain

$$\frac{\partial P}{\partial H} = \chi H_1^2 \omega^2 \gamma S T^2 \frac{\delta^2}{\delta_M} \frac{(1-r/2) (1+r)^{\gamma_4}}{4}, \qquad (9)$$

where $r = [1 + (\Omega - \omega)^2 T^2]^{1/2}$. This expression for the absorbed microwave power, in contrast to the case of a plate with identical surfaces,⁴ is of the same form as in the absence of surface relaxation, but is smaller by a factor $1 + b^2$.

Intermediate thicknesses $(5 \ll d \ll \delta_{\mu})$. We substitute (8b) in (6) and separate the real part. Expanding the ordinary and hyperbolic sines in the numerator and the cosines in the denominator in the obtained expression, and retaining the first four expansion terms, we have

$$\frac{\partial P}{\partial H} = \chi H_1^2 \omega^2 \gamma S T^{*2} \frac{\delta^2}{d} \frac{1-\alpha^2}{(1+\alpha^2)^2}, \qquad (10)$$

where $\alpha = (\Omega - \omega)T^*$

$$\frac{1}{T^*} = \frac{1}{T_s} + \frac{1}{T} \left[\left(\frac{3}{2} \right)^{\frac{1}{2}} \frac{4}{5} \right] \approx \frac{1}{T_s} + \frac{1}{T},$$
(11)

$$1/T_{\bullet} = 6^{\frac{1}{2}} D/d^2.$$
 (12)

In the derivation of (10) we have neglected the terms of order (d^2/δ_{μ}^2) . Expression (10), with the exception of the factor $(1+b)^2/2$, has exactly the same form as in the case when there is no surface relaxation. However, in place of the usual relaxation time we obtain here the effective time, which is determined by expression (11). It is seen from the latter that strong scattering by one of the surfaces yields for the width of the CEPR line an additive increment (12), proportional to the diffusion coefficient and inversely proportional to the square of the sample thickness.

Thin samples $(d \ll \delta)$. As in this case the microwave field is constant over the sample thickness, we consider the case of symmetrical boundary conditions for the microwave field (b=1). Then $F_{1,2}^- = k_s^-$ and $G_{1,2}^- = k_s^-$. Recognizing that near the peaks of the derivative of the CEPR line we have $|F_2|$, $|G_2| \gg |F_1|$, $|G_1|$, we obtain from (1) and (2)

$$\frac{\partial P}{\partial H} = -2\left(\frac{2}{3}\right)^{\frac{1}{2}} \chi H_1^2 \omega^2 \gamma S T^{*2} d \frac{\alpha}{(1+\alpha^2)^2}.$$
 (13)

It follows therefore that the influence of the strong surface relaxation, just as in the case of intermediate thicknesses, manifests itself only in a line broadening.

We note that in the case of strong surface relaxation, when the total line width is determined by the surface



FIG. 1. Theoretical dependence of the ratio of the line width ΔH at half the maximum of the CEPR line to the relaxation width $(\gamma T^*)^{-1}$ on the value of $d\delta$. The plot was drawn for the following parameter values: b = 1; $\gamma = 1.76 \times 10^7 \ G^{-1} \ sec^{-1}$; $\delta = 1.12 \ \mu$ m; $v_F = 1.07 \ cm/sec$; $(\gamma T)^{-1} = 6.5 \ G$; $D = 50 \ cm^2/sec$; $\epsilon^+ = 0$; δ_s^- is equal to ∞ (curve 1), $9.4 \ \mu$ m (curve 5), $4.7 \ \mu$ m (curve 3), $0.56 \ \mu$ m (curve 4), and $9.4 \times 10^{-5} \ \mu$ m (curve 2).

contribution (12), the intensity of the CEPR signal, as follows from (13), (10), and (9), increases with increasing sample thickness $(\partial P/\partial H \sim d^5)$ for thin samples and $\sim d^3$ for intermediate ones), and in the absence of surface relaxation it assumes a maximum value at $d \sim \delta$ after which it decreases like d^{-1} .

In the case of weak surface relaxation $[\delta_s^- \gg \min(d, \delta_{\underline{u}})]$ the line shape does not change.² The additive contribution to the total line width, due to surface scattering, takes the following form^{2,3,5}:

$$1/T_s = \tilde{\varepsilon}^{-} v_F / 2d. \tag{14}$$

Thus, in the case of strong and weak surface relaxation the line shape is the same as in the absence of surface scattering, and only the additive contributions to the total line width, (12) and (14), are different. One can expect in the intermediate case $[\min(\delta_M, d) \sim \delta_s]$ the line shape to remain unchanged, and the surface contribution to the total line width to be described by the expression interpolated from (12) and (14)



FIG. 2. Theoretical dependence of the asymmetry parameter A/B of the CEPR line on the value of d/δ . The values of the parameters and the symbols are the same as in Fig. 1.



FIG. 3. Theoretical dependence of the asymmetry parameter C/D of the CEPR line on d/δ . The parameter values and the symbols are the same as in Fig. 1; H_{max} is the coordinate of the maximum CEPR peak; $H_0 = \omega/\gamma$ is the resonant value of the magnetic field.

10

$$\frac{1}{T_{\bullet}} = \left[\frac{d^2}{D\sqrt{6}} + \frac{2d}{\overline{\varepsilon}^- v_F}\right]^{-1},$$
(15)

i.e., the total line width $T^{*^{-1}}$ is of the form

$$\frac{1}{T} = \frac{1}{T} + \left[\frac{d^2}{D\sqrt{6}} + \frac{2d}{\varepsilon^{-}v_F}\right]^{-1}.$$
(16)

In fact, numerical calculations of expression (1) for $\partial P/\partial H$ have shown (Figs. 1-4) that the line parameters depnd little on ε^- . The maximum deviation from the parameters at $\varepsilon^- = 0$ is reached at $\delta_s^- \approx \min(d, \delta_M)$ and amounts to $-\delta/\delta_M$ for the case of interest to us, that of thin samples and samples of intermediate thickness. In a bulky sample, the deviation of one of the line asymmetry parameters A/B and of the coefficient of proportionality of the line width ΔH to $(\gamma T^*)^{-1}$ is somewhat larger, this being due to the relative increase of the contribution to the signal from the scattering surface, a contribution that distorts the line shape.

We can thus draw the following conclusions concerning the influence of surface scattering by one of the sides of the sample on the observed CEPR signal in the cases of interest to us, of thin and intermediate-thickness samples:

1) The signal shape is practically the same as in the absence of surface scattering, in contrast to the case of a plate with identical surfaces.

2) The line width $T^{*^{-1}}$ increases on account of the additive contribution from the scattering of the conduction electrons by one of the sample boundaries, $T^{*^{-1}} = T^{-1} + T_s^{-1}$.



FIG. 4. Theoretical dependence of the parameter $g_{\max} = H_{\max}$. $/\Delta H$ on d/δ . The symbols and the parameter values are the same as in Fig. 1.

3) The spin surface relaxation time is well described by the expression

$T_{\bullet}=d^2/D\sqrt{6}+2d/\tilde{\epsilon}-v_F$

and has a simple physical meaning: this is the time during which an "average" electron diffuses from the nonscattering surface to the surface with the scatterer, plus the time during which it undergoes $1/\tilde{\epsilon}^-$ collisions with this surface, i.e., during which its spin relaxes.

3. EXPERIMENTAL PROCEDURE

The layer of metallic sodium with spin scatterers on the surface was produced by sputtering in a vacuum $\approx 1 \times 10^{-6}$ Torr. The sputtering sequence was the following:

1) Five pairs of glass substrates were each coated with a layer of lithium fluoride of thickness $\approx 0.5 \ \mu m$ to obtain a clean surface.

2) On one out of each pair of substrates we deposited a layer spin scatterer, tantalum or niobium, of thickness ≈ 100 Å.

3) After several seconds, we started to sputter all the substrates simultaneously with sodium. After reaching the specified thickness of the sodium layer, one of the substrate pairs was covered by a shutter, but sputtering on the remaining substrates continued. After a second pair of substrates acquired a thicker layer it was covered, and so on. The thicknesses were determined with a quartz resonator by measuring the sputtering time.

4) The films were covered with a thin layer of paraffin.

5) Pure helium gas was admitted into the cover of the setup to a pressure ≈ 1 Torr. The samples were dropped one at a time into thin-wall glass ampoules, which were then sealed-off from the setup.

Thus, in one evacuation act we could obtain up to five samples with identical sodium-scatterer contact, but with different sodium thicknesses. In addition we obtained comparison samples with which we determined the time of the conduction-electron spin relaxation due to the scattering in the interior of themetal.

The CEPR signals were measured with a standard 3-cm microwave spectrometer with high-frequency modulation of the field (100 kHz) in the temperature range 100-300 K. The sample was placed in the antinode of the magnetic field of the resonator in such a way that the microwave field was parallel to the surface. The range of investigated thicknesses of the layers of sodium was $1.7-8.4 \mu m$ for sodium with niobium.

4. RESULTS OF EXPERIMENT

The presence of a scatterer on the sodium surface in the investigated range of thicknesses led to a strong attenuation of the signal on account of the line broadening, so that it was impossible to carry out detailed measurements of the line-shape parameters A/B and C/D, but a qualitative agreement was observed between



FIG. 5. Dependence of the CEPR line whidth on the reciprocal square of the thickness of a sodium film with a Ta (\blacktriangle) or Nb (O) sublayer at room temperature. The lines show the theoretical plots for the values $\varepsilon^{-} = 8 \times 10^{-2}$ (1) and $\varepsilon^{-} = 8 \times 20^{-3}$ (2). The values of the remaining parameters are the same as in Fig. 1.

the experimental and the calculated values. For the same reason, the CEPR signals could be detected only in samples with a sodium thickness exceeding 2δ , when the waveform of the observed signal was almost symmetric, and the most convenient parameter that characterizes the relaxation time was the width ΔH at halfheight of the maximum line peak. The experimental dependences of ΔH on the reciprocal of the square of the thickness of the sodium film, for the Na-Ta and Na-Nb samples, are shown in Fig. 5. For all samples the CEPR line width in sodium without surface scattering, measured on the comparison samples, was $(\gamma T)^{-1}$ $=6.6\pm0.1$ G at room temperature. To explain the observed dependences with the aid of expression (1) one can choose the parameters $D = 50 \text{ cm}^2/\text{sec}$, $\epsilon(\text{Na-Ta})$ $\geq 8 \times 10^{-2}$, ϵ (Na-Nb) = 8 $\times 10^{-3}$. The corresponding calculated dependences are shown in Fig. 5. The same values of the parameters account also for the temperature dependence of the additive line-width increment due to scattering by the surface (Fig. 6). To show that the surface contribution to the line width, when the strong relaxation conditions $(\delta_s^- \ll d)$ are satisfied, is proportional to the conduction-electron mean free path, the plot in Fig. 6 is not against temperature but against the electric conductivity (σ) of bulky sodium (Ref. 7,



FIG. 6. Dependence, on the electric conductivity of sodium, of the additional contribution to the line width $\Delta H_s = \Delta H - \Delta H_V$, due to the spin scattering of the conduction electrons of Na on the boundary with Ta and Nb. Here ΔH_V is the line width in the samples without the scatterer. The thickness of the Na layer is 8.4 μ m (Δ) for the Na-Ta sample to and 7.7 μ m (\bullet) and 11 μ m (\bigcirc) for the Na-Nb samples. The lines show the theoretical dependences for the same thicknesses at $\varepsilon^- = 0.5 \times 10^{-2}$ (Na-Ta) and $\varepsilon^- = 8 \times 10^{-3}$ (Na-Nb); D (300 K) = 50 cm² sec. The values of δ (300 K) and of the remaining parameters are the same as in Fig. 1.

p. 140). These measurements could be made on the samples with the largest sodium thickness 8.4 μ m for the Na-Ta samples and 11.0 μ m and 7.7 μ m for the Na-Nb samples. It was assumed in the numerical calculations that the conduction-electron diffusion coefficient is proportional to the electric conductivity. The line width $(\gamma T)^{-1}$ of pure sodium was determined experimentally from the comparison samples. Typical plots of the line width ΔH against the electric conductivity for sodium 7.8 μ m thick is shown in Fig. 7.

Thus, on the basis of the presented data we can draw the following conclusions:

1. The theoretical calculations given in Sec. 2 describe well the observed experimental relations.

2. The value of the diffusion coefficient of the conduction electrons in sodium at room temperature, with allowance for the measurement errors, is $D = 50 \pm 10$ cm²/sec.

3. The conduction electron spin flip probability in sodium after a single collision is $\epsilon \ge 8 \times 10^{-2}$ for the Na-Ta boundary and $\epsilon \approx 8 \times 10^{-3}$ for the Na-Nb boundary, and does not depend on the temperature.

The experimentally obtained values of ε can be explained by making the following assumptions: 1) the conduction electrons of sodium do not pass in the volume of the scatterer, and spin scattering takes place from the first layer of the Ta or Nb atoms; 2) the effective cross section σ_s for conduction-electron scattering with spin flip, in collisions with Ta and Nb atoms, is equal to its bulk value σ_v . Then $\varepsilon = n\sigma_s \approx n\sigma_v$, where n is the number of scatterer atoms per cm² of the interface. We estimate σ_v in accord with the simple theory of paramagnetic relaxation⁸:

$$2\sigma_{\mathbf{v}}=a^{2}\left(\frac{\Delta_{is}a}{\hbar v_{F}}\right)^{2},$$

where *a* is the lattice constant of sodium, and Δ_{ls} is the atomic spin-orbit splitting of the scatterer. Substituting the value of σ_v in the expression for ε , we obtain

$$2\varepsilon = \theta \left(\frac{\Delta_{isa}}{\hbar v_F}\right)^2; \tag{17}$$

here $\theta = na^2$ is the ratio of the number of scatterer atoms to the number of sodium atoms on the interface. In the estimate we assume $\theta = a^2/a_s^2$, where a_s is the lattice constant of tantalum (niobium). For *d* electrons, Δ_{Is} = 2.5 ξ and, using the spin-orbit interaction constants $\xi(Ta) = 1657 \text{ cm}^{-1}$ and $\xi(Nb) = 475 \text{ cm}^{-1}$ (Ref. 9), we obtain

$$\epsilon$$
 (Na-Ta) =0.93 · 10⁻¹, ϵ (Na-Nb) =7.5 · 10⁻³.

We used in the estimates the following parameter values: $a=4.28\times10^{-8}$ cm, $v_{F}=1.07\times10^{8}$ cm/sec (Na);



FIG. 7. Dependence of the CE PR line width on the electric conductivity of a sodium film without a scatterer, $d = 7.8 \,\mu\text{m}$.

 $a_s = 3.31 \times 10^{-8}$ cm (Ta); $a_s = 3.3 \times 10^{-8}$ cm (Nb).

Thus, the theoretical values of ε agree well with those obtained in experiment. The physical argument in favor of the assumption that the conduction electrons do not enter into the volume of the scatterer is the fact that otherwise ε would depend on the relaxation time in the scatterer volume and would decrease with decreasing temperature. In our case, both the size dependence and the temperature dependence of the total line width are well accounted for by temperature-independent values of ε .

One of the possible mechanisms of the scattering of the sodium conduction electrons by the interface may be resonant scattering by the *d*-orbitals of the first layer of the scatterer which form a band of surface states¹⁰ at the surface of the free scatterer crystal, this band being located near the Fermi level. In the case of the contact of the scatterer layer with sodium, the surface-state band should broaden. The situation recalls resonant scattering by virtual *d* states of the impurity atom. Comparing the known expression for the effective cross section for resonant scattering, with spin flip, by an impurity located in the volume of the sodium, and the value of σ_s calculated from the experimentally obtained value of ε , we can estimate the broadening of the surface-state band: $\Gamma(\text{Na-Nb}) \approx \Gamma(\text{Na-Ta}) = 3 \text{ eV}$. It should be noted that the question of the scattering mechanism calls for further theoretical and experimental research.

- ¹I. G. Zamaleev and E. G. Kharakhash' yan, Pis' ma Zh. Eksp. Teor. Fiz. 27, 677 (1978) [JETP Lett. 27, 641 (1978)].
- ²F. J. Dyson, Phys. Rev. 98, 349 (1955).
- ³M. B Walker, Phys. Rev. B3, 30 (1971).
- ⁴V. V. Ustinov, Fiz. Met. Metalloved. 45, 473 (1978).
- ⁵V. N. Lisin, Zh. Eksp. Teor. Fiz. **72**, 573 (1977) [Sov. Phys. JETP **45**, 300 (1977)].
- ⁶J. H. Pifer and R. Magno, Phys. Rev. **B3**, 661 (1971).
- ⁷Spravochnik po fiziko-tekhnicheskim osnovam kriogeniki (Handbook of Physical and Technical Principles of Cryogenics), M. P. Malkov, ed., Energiya, 1973.
- ⁸D. Pines and F. Slichter, Phys. Rev. 100, 1014 (1955).
- ⁹J. S. Griffith, The Theory of Transition Metal Ions, Cambridge Un. Press, 1961.
- ¹⁰St. G. Louie, Kai-Ming Ho, J. R. Chelikowsky, and M. L. Cohen, Phys. Rev. B15, 5627 (1977).

Translated by J. G. Adashko

Transport of polarized radiation in crystals in the excitonic region of the spectrum. Polariton effects

E. L. Ivchenko, G. E. Pikus, and N. Kh. Yuldashev

A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences (Submitted 3 August 1980) Zh. Eksp. Teor. Fiz. 80, 1228–1245 (March 1981)

A theory is developed for radiation transport in crystals with allowance for polariton effects in elastic scattering of the polaritons by impurity centers. The frequency dependences of the intensity and of the degree of polarization of the resonant secondary emission upon excitation by polarized light are calculated and analyzed. It is shown that the analysis of polariton transport in the absence of spatial dispersion, when the exciton effective mass $M \rightarrow \infty$, reduces to the problem of radiation transport in a turbid medium. At a finite exciton effective mass, account was taken of the additional light waves. In the region below the longitudinal frequency ω_L , the presence of additional waves that attenutate in space at $\omega < \omega_L$ influences only the polariton reflection coefficients. In the frequency region above ω_L an important role is played by scattering processes between two transverse-wave branches and between branches of transverse and longitudinal polaritons.

PACS numbers: 79.60. - i, 71.36. + c, 71.35. + z

§1. INTRODUCTION

In our preceding paper¹ we developed a theory of resonant transport of polarized radiation in crystals, with account taken of the reabsorption and reemission as well as of multiple reflection of light from the surface. The analysis was carried out in the limiting case of weak exciton-photon interaction, when the photons and excitons could be regarded as separate particles. The present article is devoted to a study of transport of resonant radiation in crystals in the case of strong excitonphoton interaction, which leads to a restructuring of the energy spectrum of the photon and excitons, and to formation of collective modes-polaritons (opto-excitons). creased in connection with the observation and investigation of resonant Brillouin scattering of polaritons in III-V semiconductors (GaAs) and II-VI semiconductors (CdS, CdSe).²⁻⁵ In most theoretical work on resonant scattering of polaritons, only single scattering was taken into account.⁶⁻⁸ In the general case, however, before a polariton is emitted into vacuum or recombines nonradiatively, it undergoes multiple scattering. The Brillouin spectrum revealed therefore not only singlephonon but also two-phonon peaks, as well as a broad background corresponding to multiple scattering (the so-called polariton fluorescence).⁴ As shown by experiment,⁹ the distribution function of the polaritons is inhomogeneous in space and has greatly differing values near the boundary and in the interior of the sample.

Interest in the theory of polariton transport has in-