Investigation of the resonance properties of electrons localized above liquid ³He and ⁴He

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The cyclotron resonance of electrons localized above ³He and ⁴He is investigated in the temperature range 0.36–0.6 K at frequencies of 18.5 and 37.75 GHz. In weak clamping electric fields E_1 the cyclotronresonance line width, ΔH , is ~8 Oe for ⁴He at T = 0.4 K and ~20 Oe for ³He at T = 0.36 K. In strong clamping fields ΔH increases by tens of times, and is $\propto kTE_1^2/H\sigma$ (σ is the surface-tension coefficient). It is found that, for $E_1 \gtrsim 20$ V/cm, $\Delta H(E_1)$ decreases by a factor of one and a half to two as E_1 increases. The resonance magnetic field, H, that coincides with the free-electron cyclotron-resonance field, H_e , for $E_1 \rightarrow 0$ decreases with increasing E_1 according to the law $H-H_e$ [Oe] = $8.4-7.8 \times 10^{-5}$ E_1^2 [V²/cm²] for electrons above ⁴He and $H-H_e$ [Oe] = $-2.04 \times 10^{-4}E_1^2$ [V²/cm²] for electrons above ³He. Cyclotron absorption is found to increase when the electrons are irradiated at a frequency ~125-170 GHz, which corresponds to the transition energy between hydrogen-like energy levels. The photoresonance-line widths at $T \approx 0.36-0.4$ K are ~0.45 and ~1.6 GHz respectively for electrons above ⁴He and ³He, which exceed the theoretical predictions by a factor of 30-50. The electron-relaxation (in terms of energy) time is determined from the results of an investigation of the heating of the electrons by the measuring signal and an investigation of the transient period of the signal at the time of photoresonance, and is found to be $\tau_E \approx 10 \ \mu$ sec.

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1. INTRODUCTION

Electrons located above the surface of liquid helium are attracted to the liquid under the action of the electrostatic-image forces. The corresponding potential energy at distance large compared to the interatomic distances has the form^{1,2}

$$U(z) = -e^{2}(\varepsilon - 1)/4(\varepsilon + 1)z = -\Lambda_{o}/z, \qquad (1)$$

where Z is the coordinate along the normal, N, to the liquid surface and ε is the permittivity, which is equal to 1.0572 for ⁴He and 1.043 for ³He. Since the penetration of an electron into the liquid is hindered by a barrier of height 1 eV, a low-energy electron finds itself in a one-dimensional energy well with hydrogen-like energy levels, given to within 5% by the expression

$$\hbar\omega_{\perp l} = -m\Lambda_0^2/2\hbar^2 l^2. \tag{2}$$

Spatially, the electrons are localized at a distance determined by the parameter $1/\gamma = \hbar^2/m\Lambda_0$, and equal to 7.6×10^{-7} cm for ⁴He and 1×10^{-6} cm for ³He. Numerically, the average values of the z coordinate for l=1, 2, 3 are 114, 456, 1026 Å and 150, 600, and 1350 Å respectively for ⁴He and ³He.

The motion parallel to the surface, i.e., in the (x, y) plane remains free. A magnetic field H||N does not change the motion along the z axis, but it leads to the quantization of the transverse motion, with Landau levels:

$$\frac{e\hbar H}{mc}\left(n+\frac{1}{2}\right) = \hbar\Omega\left(n+\frac{1}{2}\right)$$
(3)

and with a characteristic localization dimension in the ground state (n=0) given by

$$L_{H}^{2} = 2\hbar c/eH.$$
(4)

Correspondingly, two types of resonance effects can be observed: cyclotron resonance (CR) at the frequency $f=\Omega/2\pi$

(5)

and photoresonance upon the coincidence of the electromagnetic-field frequency with a transition frequency in the hydrogenic spectrum:

 $F_{ii} = (\omega_{\perp i} - \omega_{\perp i})/2\pi. \qquad (6)$

In experiments, the fulfillment of the condition (5) is usually achieved by varying the magnetic field, while the fulfillment of the condition (6) is achieved by varying the strength of a supplementary clamping field $E_{\perp}||N$, which gives rise to the linear Stark effect and a shift in the eigenfrequencies.³

Both of these resonance effects have been studied only for electrons localized above ⁴He at temperatures $T \ge 1.2 \text{ K.}^{3,4}$ Under these conditions the dominant role in the relaxation processes determining the resonanceline width is played by the collisions with the atoms of the vapor, which has quite a high density: ~10¹⁹ cm⁻³.

In Refs. 5 and 6 the present author describes the first results of experiments on the investigation of the cyclotron resonance of electrons above ³He and ⁴He at $T \approx 0.4$ K. Under these conditions the dominant role is played by the collisions with the thermal oscillations of the liquid surface—the ripplons with the spectrum

$$\omega_q^2 = \sigma q^3 / \rho, \tag{7}$$

where σ is the surface-tension coefficient, ρ is the liquid density, and q is the wave vector of the ripplon with frequency ω_q . It is also established in Ref. 5 that the clamping field causes the effective mass to decrease. In the present paper we describe the results of a similar investigation of the cyclotron resonance of electrons above ³He and ⁴He at $T \approx 0.36 - 0.7$ K and frequencies of 18.5 and 37.7 GHz in fields $E_1 \leq E_{\rm cr}$, where $E_{\rm cr}$ is the field value corresponding to the loss of stability of the charged liquid surface.^{7,8}

As established in Ref. 9, electrons localized above helium are easily superheated. In the process, owing to the fact that the electrons move farther away from the surface as they are heated, their interaction with the surface weakens, and this leads to the increase of the cyclotron-resonance amplitude. This circumstance was used in the present investigation to observe the photoresonance through the change occurring in the cyclotron absorption during the irradiation of the electrons at the frequency F_{11} . Photoresonance was observed in this way at $T \approx 0.4$ K for electrons localized above ³He and ⁴He.

2. EXPERIMENTAL SETUP

The setup of the experiment is illustrated by Fig. 1. A given quantity of helium is condensed into a resonant cavity, so that its level is located at a height h above the bottom, which is insulated from the housing. Between the bottom and the housing is applied a potential difference U_1 , which produces a field E_1 . When a voltage potential ~0.5 - 1 kV is fed to the discharge gap, a cold discharge is ignited from which electrons are drawn by the constant electric field to charge the helium surface. The charging proceeds until the electron density attains a limiting value, equal on the cavity axis to

$$\eta_{\rm max} = U_{\perp} / 4\pi he, \tag{8}$$

which corresponds to complete screening of the static field above the helium surface by the electron layer. (Here we neglect the small deviation of ε from unity). After this the discharger is switched off. The electron concentration, given by the formula (8), was preserved for an indefinite length of time, until a decrease in U_1 occurred, which was accompanied by a proportional decrease in n_{max} .

The cyclotron resonance manifested itself in the resonance absorption of a SHF signal that passed through the cavity as the magnetic field, produced by a superconducting solenoid, was varied (Fig. 2). The direction of the field H was set parallel to N to within 10 -20', as determined from the anisotropy in the effective mass.

The source of the microwave signal was a three-centimeter-band generator based on a traveling-wave tube, and stabilized by a super-conducting resonant cavity.¹⁰ The output signal was modulated at a frequency of 10



FIG. 1. Schematic drawing of experimental setup. Two values of λ were used: 1.5 cm and 8 mm.



FIG. 2. Typical recordings showing the dependence of a signal that has passed through the cavity on the magnetic field during the observation of the cyclotron resonance. The measurement conditions are indicated beside the curves.

kHz, and its frequency was doubled in a crystal diode to which a signal of frequency 100-200 MHz was simultaneously fed, so that one of the components of the output signal had a frequency equal to a natural frequency of the cavity. After passing through the cavity, the signal was picked up by a P5-14A superheterodyne receiver with a sensitivity of $\sim 10^{-14}$ W, separated from the cavity by an auxiliary ferrite rectifier, and operating in the continuous regime. This ensured the reduction of the level of that signal of the receiver's beat-frequency oscillator which passed through the cavity to $\sim 10^{-15}$ W, which had to be done in order to avoid possible parasitic effects. The output signal of the P5-14A was amplified by a narrow-band amplifier (at a frequency of 10 kHz), synchronously detected and registered on the v coordinate of a two-coordinate recorder. along whose x coordinate was registered a signal proportional to the current of the solenoid, or the current passing through an auxiliary scanning coil in the presence of a frozen-in field. The receiving system was calibrated with the aid of a precision attenuator inserted between the microwave-signal source and the cavity. The nonlinearity of the receiver's characteristics, which was ~10%, was taken into account during the processing of the measurement results.

In the experiments at the frequency of 37.7 GHz, we used a klystron oscillator that could frequency sweep through the resonance curve of the cavity and a direct-coupling amplification receiver capable of a narrow-band signal discrimination at the modulation frequency. The sensitivity of the receiver in this case was estimated to be $10^{-8} - 10^{-9}$ W.

Although the Q of the cavity was, as a result of the presence of the insulation between the bottom and the

walls, relatively low, and was $Q_0 \approx 2 \times 10^3$, the additional losses due to the resonance absorption at $f = \Omega/2\pi$ were so high that the power of the transmitted signal could, depending on the helium level, change by a factor running into tens. This led to a considerable distortion of the shape of the cyclotron-resonance line. The contribution of the electrons to the resonance parameters can be decreased by decreasing the height h. If, when doing this, $h \ll d/2$ (d is the cavity height), then the microwave field acting on the electrons decreases by a factor of 2h/d, while the losses decrease by a factor of $(2h/d)^2$. However, when $h \le 1$, the square, E_{cr}^2 , of the critical field corresponding to the stability threshold for the charged surface decreases in porportion to h (Refs. 7 and 8). Therefore, a considerable decrease in h is undesirable, since it leads to a limitation on the range of admissible E_1 fields in which the measurements are possible.

For $h \leq 1.5$ mm, the change induced in the natural frequency of the cavity by a change in the reactive component of the conductivity of the system of surface electrons leads, according to the measurements, to ~10% changes in the transmitted signal, and these distortions can be neglected. Taking this circumstance into account, we can easily relate the magnetic-field dependence of the height of a signal that has passed through the cavity with the cyclotron-resonance line shape, which we assume to be Lorentzian, i.e., to be described by the formula

$$P(\mu) \propto 1/(1+\mu^2),$$
 (9)

where P is the power absorbed by the electrons, μ is the reduced detuning:

$$\mu = e\tau (H - H_0) / mc = (H - H_0) / \Delta H,$$

 τ is the relaxation time, and ΔH is the halfwidth of the CR line with respect to the magnetic field. Substituting (9) into the well-known expression for the transmission coefficient expressed in terms of the power of the microwave wave propagating through the cavity (in the case of weak coupling),

$$T(\mu) \propto Q^2(\mu), \tag{10}$$

and allowing for the fact that $Q(\mu) = Q_0 / [1 + kP(\mu)]$ (k is a coefficient that is of no interest to us now), we obtain a formula relating the CR line shape with the measurable relative changes in the transmission coefficient:¹⁾

$$\frac{1}{1+\mu^2} = \frac{[T_0/T(\mu)]^{\nu_0}-1}{[T_0/T(0)]^{\nu_0}-1},$$
(11)

where T_0 is the transmission coefficient under conditions far from resonance conditions (i.e., for $|\mu| \gg 1$), or in the absence of electrons.

The object of the investigation was to ascertain the dependence of ΔH and the location of the resonance on E_{\perp} . In order for the experimentally obtainable quantities to be referrable to a definite value of E_{\perp} , the field should be constant over as large a section of the surface occupied by electrons as possible. Since the configuration of the constant field in the volume of the cavity (of diameter 20 mm and height 9 mm) is quite far

from that of a field in a plane-parallel capacitor, the field E_1 can be made uniform as far as possible by decreasing the height, h, of the helium layer. If, when doing this, the charge completely screens off the field above the helium, then at the center of the cavity

$$E_{\perp} = U_{\perp}/2h. \tag{12}$$

The fringe effects can have a noticeable influence on the results of the experiment. Thus, for h=1.4 mm, according to model measurements in an electrolytic bath, the E_1 distribution along the radius r is described by the expression (R is the cavity radius):

$$E_{\perp} = E_{\perp}(0) \left[1 - e^{-i\theta(1 - r/R)} \right].$$
(13)

Although even at a distance of $\sim 2 \text{ mm}$ from the edge E. differs from the field at the center by only ~13% and the electron density decreases in proportion to E_1 as r decreases, because of the rapid narrowing of the cyclotron-resonance line as E_1 decreases and the resulting growth of the contribution to the resonance amplitude, the fringe effects turn out to be quite significant. Thus, resonance-line shape calculations carried out on a computer with allowance for the results obtained in the course of the investigation and the formula (13) showed that, for $E_1 = 1000 \text{ V/cm}$ and the H_{111} cavity oscillation mode used in the experiments, the measurable width turns out to be 8%, and the line shift 20%, less than the true value. The influence of the fringe effects turned out to be less than the measurements errors when a cavity with a fin (Fig. 1) bounding the region with the charge was used.

Another cause of the E_1 nonuniformity was the deviation from the horizontal plane of the surface of the lower electrode. Thus, a deviation of 1° corresponds to a variation in the thickness of the helium layer equal to 0.35 mm, which is more than 20% of the total thickness when h=1.5 mm. However, by varying the tilt of the whole instrument, and measuring the values of the critical field in which the charged surface loses its stability, and discharges,⁸ we could adjust the position of the cavity to within 1-2', which corresponds to variations in h of $\Delta h \leq 0.01$ mm, which are comparable to the nonplanarity of the bottom itself.

The variation of h in the course of the experiment can also occur as a result of the vibration of the helium surface. The vibration amplitude a can be estimated in a CR measurement in a magnetic field inclined at an angle ϑ to the normal to the helium surface. For a cavity radius R=1 cm, the characteristic angles by which the liquid surface deviates from the horizontal are $\sim a/R$. Because of the dependence of the effective mass on the angle ϑ according to a law of the form m $\propto 1/\cos\vartheta$ (Ref. 4), there appears upon the inclination of the magnetic field a mass spread, $\pm a \vartheta/R$, that leads to a corresponding broadening of the cyclotron-resonance line. The vibration-induced broadening was indeed observed in our first experiments, but after suspending the cryostat on weak springs (the natural vibration frequencies of the whole apparatus was ~1 Hz), it completely vanished. Taking into account the fact that a

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~5% change in ΔH could have been observed in the experiments, for $\vartheta = 3^{\circ}$ and $\Delta H/H \approx 10^{-3}$ we obtain the estimate $a \leq (1-2) \times 10^{-3}$ cm. Thus, the *h* variations due to the mechanical vibrations did not exceed ~1%, and could not give rise to an additional measurement error.

Because of the thermomechanical effect, the liquid helium could rise in the admission tube to the warmer part of the cryostat, which led to the variation of the level during changes in the temperature, and, for a time, to an uncertainty in h. In order to reduce this effect to a minimum, the helium was condensed through a capillary with an internal diameter of 0.25 mm and with an inner core of diameter 0.22 mm inserted in it. The change in h even when the capillary was completely filled with liquid helium was $\leq 3 \times 10^{-3}$ cm. In order to avoid getting the coaxial lines filled, owing to the flow of a superfluid film, and the accumulation of macroscopic quantities of helium above the foamed polystyrene separators, which were necessary for the centering of the inner conductor, the lines terminated in the cavity in vacuum-tight platinum-glass seals.

The electrostatic forces acting on the electrons should lead to the deformation of the liquid surface and to a change in $h(E_1)$. Using the formula (13), which describes the distribution of E_1 in a real situation, we can easily calculate, neglecting the surface-tension forces, that, for $E_1 \approx 1 \text{ kV/cm}$ and a level located below the cavity fin, the drop in the liquid level at the center of the cavity is 3×10^{-3} cm. But if in the absence of a clamping field the liquid level coincides with the fin (as shown in Fig. 1), or lies above it, then the sag of the surface under the influence of E_1 will be significantly less.

Thus, the measures that were taken ensured the constancy of the depth, h, of the helium layer during the experiment to within 1 - 2%. In accordance with the formula (12), this ensured the determination of the relative magnitude of E_1 in the same experiment with the same accuracy.

To determine the absolute magnitude of E_{\perp} , we measured the U_{\perp} dependence of the transition frequency F_{12} in the hydrogenlike spectrum (see Sec. 4). Comparing the dependence obtained by us with the results given in Ref. 3, we could determine the relation between U_{\perp} and E_{\perp} to within 1 - 2%. We learned in the process that the procedure used by us earlier in Refs. 5 and 6 to compute h from the geometric dimensions of the cavity and the quantity of condensed gas led to errors of up to 0.1 - 0.2 mm. This was due, apparently, to the appearance of gaps between the parts of the cavity during the cooling.

Since it was of interest to elucidate the effect of the electron density, n, on the width and location of the CR line, we performed experiments with a fixed total charge over the helium surface. For this purpose, the charge was produced at some voltage potential U_{10} , then the helium was warmed up to $T > T_{\lambda}$, which led to the disappearance of the superfluid helium film on the cavity walls and the departure of the electrons localized above it.¹¹ After this the cavity was cooled again, and the CR was studied at $U_1 \ge U_{10}$. Because of the nonuni-

formity of the static field, the electrons collected, as U_{\perp} was increased, at the center of the cavity, but their concentration turned out to be lower than the concentration given by the formula (8). For the determination of n and E_{\perp} in this case a model experiment was performed in an electrolytic bath. It was found that, for $h \approx 1-1.5$ mm,

$$n = \frac{U_{\perp 0}}{4\pi h e} \left(\frac{U_{\perp}}{U_{\perp 0}}\right)^{\nu_{h}}, \quad E_{\perp} = \frac{U_{\perp}}{d} \left[1.5 + \left(\frac{4.9}{h} - 1.4\right) \left(\frac{U_{\perp 0}}{U_{\perp}}\right)^{0.45}\right]$$
(14)

to within ~5-10%. Such a dependence for E_1 was obtained when the photoresonance frequency $F_{12}(U_1)$ was measured under conditions of total-charge conservation.

In the measurements with ³He the depth h was assumed to be equal to the depth of the layer in the experiments with ⁴He, if the computed volumes of the liquid ³He and ⁴He used were equal to each other.

3. DEPENDENCE OF THE WIDTH AND LOCATION OF THE CR LINE ON THE CLAMPING FIELD

In Figs. 3 and 4 we show the results of the measurement of the CR line width ΔH and the shift, δH , of the line from the H_{e} -field value corresponding to the free-electron resonance, i.e., for $E_{1} \rightarrow 0$. The results presented were obtained in the processing of experimental recordings of the type shown in Fig. 2.

The CR for electrons above 4 He was observed by us in the temperature range 1.3-0.4 K. The value of the line



FIG. 3. Dependence of the halfwidth of the cyclotron-resonance line on E_{\perp} for electrons above ⁴He for f = 18.5 GHz: **①**) T = 0.72 K, **④**) T = 0.4 K; the remaining points were obtained in various experiments at T = 0.42-0.43 K. The depth, h (in mm), of the helium layer was equal to: **①**, Δ , **▲**) 1.15 ± 0.05 , **○**) 1.25 ± 0.05 , **④**) 1.35 ± 0.02 , **□**, **■**) 1.59 ± 0.02 ; **□**) ΔH for electrons in the l = 1 ground state, **■**) ΔH for electrons in the l = 1 ground state, **■**) ΔH for electrons in the l = 1 aresonance transitions. The dashed curves indicate the doubled ΔH values computed from the formula (19) from Ref. 16. The dot-dash curve represents the values computed from the formula (14) from Ref. 16 with $p^2 = \sigma H/c$.



FIG. 4. The E_1 dependence of the line halfwidth ΔH (top) and the resonance shift δH (bottom). $\nabla, \Delta, +, \nabla, \Theta, \times, \Delta, \Phi$), for electrons above ⁴He and frequency 18.5 GHz; \Box), \blacksquare) for same electrons and frequency 37.7 GHz; O) for electrons above ³He, frequency 18.5 GHz, and T = 0.36 K; ∇) T = 0.74 K, **A**) T = 0.62 K, **O**) T = 1.2 K, and the remaining points are for T = 0.4 - 0.43 K. The depth, h (in mm), of the liquid layer was: ∇, ▲, +) 1.22 ± 0.03; ∇, □, ●) 1.14 ± 0.02; ●) 1.34 ± 0.02; △) 0.96 ± 0.02 ; x) 1.67 ± 0.03 ; A) 1.32 ± 0.02 ; O) 1.40 ± 0.04 . The charge density was: Δ) $\approx (E_{\perp}/90)^{1/2} \times 10^8 \text{ cm}^{-2}$, x) $\approx (E_{\perp}/48)^{1/2}$ $\times 5.3 \times 10^8$ cm⁻², and for the remaining points $n = n_{max}$. The dashed lines in the upper part of the figure represents ΔH values computed from the formula (20) for electrons above ³He and above ⁴He for 18.5 GHz and above ⁴He for 37.7 GHz. The dotdash lines are theoretical δH curves taken from Ref. 12. The continuous lines are approximations to the measured values, obtained by the method of least squares.

width at $T \sim 1.3$ K in our experiments for $E_{\perp} \leq 200-300$ V/cm was 600 ± 50 Oe, which virtually coincides with the results of Ref. 4. When E_{\perp} was increased, the line width increased according to an almost linear law, and attained a value of 1300 ± 100 Oe in a 1.3-kV/cm field.

When the temperature was decreased from 1.3 to 0.4 K, the line width for small values of E_{\perp} decreased by a factor of 70-80. This was connected with the rapid decrease of the vapor density above the liquid ⁴He, which governed the scattering processes at $T \ge 1$ K (Ref. 4). At lower temperatures, the electrons are scattered by the thermal vibrations of the liquid surface—by the ripplons.

The measurements for ³He were carried out only at the lowest attainable temperature, which, in our cryostat, was T = 0.36 K. At this temperature $\Delta H(E_{\perp} \rightarrow 0)$ = 20 ± 1 Oe, and, according to estimates, is determined by the scattering by the vapor.⁶ At higher temperatures we could not observe any electrons above the liquid 3 He, which was apparently connected with the possibility of its ebullition.

We present the measurements of the dependence $\Delta H(E_{\perp})$ for $E_{\perp} \leq E_{cr}$, corresponding to the loss of stability by the charged liquid surface. The E_{cr} values for both ⁴He and ³He were close to the values given by theory⁷ and the measured values given in Ref. 8.

As E_{\perp} is increased, the line width at first decreases slightly (Fig. 3), and then, starting from $E_{\perp} \sim 50 \text{ V/cm}$, increases monotonically. In fields $E_{\perp} \geq 500 \text{ V/cm}$, the dependence $\Delta H(E_{\perp})$ is close to being quadratic: ΔH $\propto \Delta H_0 + \alpha E_{\perp}^2$ (Fig. 4).

The value of α decreases as the temperature decreases almost in proportion to *T*. When the frequency of the measuring signal was increased by a factor of two, α decreased by a factor of 2.2. The value of α for ³He is $2.4 \pm 10\%$ times higher than its value for ⁴He at the corresponding temperature, which virtually coincides with the ratio, $\sigma_4/\sigma_3 = 0.375/0.158 = 2.37$, of the surface-tension coefficients. When the surface is incompletely charged (the electron density at the center of the cavity is equal to $(0.3 - 0.2)n_{max}$, $E_1 \ge 1 \text{ kV/cm}$), the values of ΔH turn out to be roughly two times less than the values for the maximum density (Fig. 4).

When E_1 is increased, the resonance curve shifts towards the region of weaker fields. The resonance shift δH does not depend on temperature and the frequency of the measuring signal (Fig. 4). When the electron concentration is decreased at the same values of E_1 , δH decreases slightly. For electrons above ³He, the values of δH are roughly three times greater. The $\delta H(E_1)$ dependences for electrons above ⁴He and $B_1^2 \ge 2 \times 10^5 (V/cm)^2$, the experimental points in the case when $n = n_{max}$ lie, to within the measurement errors, on the straight line described by the equation

$$\delta H[\text{Oe}] = 8.4 - 7.8 \cdot 10^{-3} E_{\perp}^{2} [\text{V}^{2}/\text{cm}^{2}] .$$
(15)

For $E_1^2 < 2 \times 10^5$ (V/cm)², the shift of the resonance occurs significantly more slowly. For ³He, in the entire investigated field range,

$$\delta H[\text{Oe}] = -2.04 \cdot 10^{-4} E_{\perp}^{2} [\text{V}^{2}/\text{cm}^{2}] .$$
(16)

Notice that the ratio of the coefficients attached to E_{\perp}^2 is 2.6±10%, i.e., close to σ_4/σ_3 .

As is well known, electrons localized above liquid helium are easily superheated by the field of the measuring signal.^{1,9} Taking this into account, we performed experiments to determine the effect of the microwave power, P, dissipated by the electrons on the width and location of the cyclotron-resonance line. In Fig. 5 we give the results of the measurement of $\Delta H(P)/\Delta H(P \rightarrow 0)$ and $\delta H(P)/\delta H(P \rightarrow 0)$ as functions of the quantity $P_{\rm res}/n$, i.e., the power absorbable at resonance by one electron. The qualitative behavior of the dependences are identical at all values of E_1 : when the electrons are heated relative to the helium,²⁾ the resonance line narrows slightly, while the resonance shift decreases to zero and then changes sign. It should, however, be noted that the ΔH values given in Fig. 5 are,



FIG. 5. Dependence of the relative line width $\Delta H(P)/\Delta H(P \rightarrow 0)$ and the resonance shift $\delta H(P)/\delta H(P \rightarrow 0)$ on the SHF-signal power, *P*, dissipated at resonance by one electron. The clamping field E_1 is equal to: **O**) 22, \blacklozenge 56; \blacklozenge) 460, \blacktriangle) 590, \diamondsuit) 780, and \bigtriangleup) 1100 V/cm. The error in the absolute calibration for *P* is $\sim \pm 3$ dB.

in a sense, conditional. Indeed, the energy absorbed by, and, hence, the temperature of, the electrons depend on the detuning from the exact resonance. Clearly, this should lead to the distortion of the line shape. In principle, this circumstance can be taken into account, but since the obtained results are only qualitative in nature, we neglected the possible corrections, and assumed the line shape to be Lorentzian in all the cases.

As is evident from Fig. 5, there exists quite a broad range of P values at which the measurement results do not depend on the signal power. All the ΔH and δH measurements that were performed at the frequency of 18.5 GHz, the results of which are shown in Figs. 3 and 4. were performed under these conditions. Since the sensitivity of the receiver at the frequency of 37.7 GHz was significantly lower, the conditions for low power (i.e., for slight superheating) were attained in these experiments only in the strongest fields, in which the possible corrections to ΔH and δH due to superheating did not exceed 5-10%. The noticeable deviation of the δH values at this frequency for $E_1^2 \sim 0.5 - 1 \ (kV/cm)^2$ from the corresponding values on the stright line in Fig. 4 is, apparently, connected with the superheating of the electrons.

4. OBSERVATION OF THE PHOTORESONANCE

To observe the photoresonance through the variation of the conductivity, we fed to the cavity through a waveguide of circular cross section (see Fig. 1) microwave radiation with $\lambda \approx 2 \, \text{mm}$ from a backward-wave tube (BWT). Since the characteristic dimensions of the cavity were much greater than λ , and the radiation coming out of the waveguide was polarized in such a way that the vector \vec{E} lay essentially in the plane of the liquid surface, while for the stimulation of the transitions to be possible it was necessary that $\vec{E} || N$, the photoconductivity was observed at a relatively high level of dissipated microwave power $\sim 1 - 0.1 \ \mu W$ (this quantity was estimated from the heating of the cavity).

The observations were carried out in the following

manner. The magnetic field got frozen in when it attained the value corresponding to the maximum absorption of a measuring signal of frequency 18.5 GHz as a result of the cyclotron resonance. Then, decreasing E_1 , we measured the cyclotron absorption as a function of E_1 . Since F_{11} is strongly E_1 dependent,³ a suitably chosen F will allow the coincidence of the frequencies: $F = F_{1,1}(E_1)$. The monotonic dependence $T(E_1)$, which is due to the decrease of $n \propto E_1$, exhibits a resonance siggularity at the corresponding value of E_1 (see the curves 1 and 2 in Fig. 6). As a rule, the amplitude of the resonance correction to the absorption was <10%. Therefore, it was more convenient to investigate the photoresonance after modulating the power radiated by the BWT by feeding a voltage potential of frequency f_{mod} to the focusing electrode, and separating out the envelope of the signal of frequency 18.5 GHz with the aid of a narrow-band amplifier with a synchronous detector (see the curves 3 and 4 in Fig. 6). This is how we measured the resonance-transition spectra, examples of which are given for electrons localized above ⁴He and ³He in Figs. 7 and 8. The natural-frequency spectrum, $F_{1,i}(E_i)$, for ³He, as measured by us, is shown in Fig. 9. The dependences $F_{11}(E_1)$ for ⁴He had already been measured by Grimes *et al.*,³ and, using their data on the dependence $F_{12}(U_1)$, we were able in each experiment to determine the coefficient of proportionality between U_{\perp} and E_{\perp} , i.e., the depth of the liquid layer.

The halfwidth of the photoresonance at halfheight for ⁴He at T = 0.43 K was 0.5 - 0.8 V/cm in the range $E_{\perp} \approx 12 - 65$ V/cm for the 1 - 2 transition and 0.3 ± 0.1 and 0.1 V/cm (for $E_{\perp} < 17$ V/cm) for the 1 - 3 and 1 - 4 transitions, respectively. Using the known values of $\partial F_{11} / \partial E_{\perp}$, we find that the resonance halfwidth in terms of frequency does not depend on E_{\perp} , and is equal to $\Delta F_{11} = 0.45 \pm 0.12$ GHz.

For ³He the resonance halfwidth at T = 0.36 K with respect to the field was (1.6 ± 0.3) V/cm for the 1-3 transition, 1.1 V/cm for the 1-4 transition. In terms of frequency, $\Delta F_{11} = (1.65 \pm 0.25)$ GHz, i.e., 3-4 times greater than for ⁴He.



FIG. 6. Dependence on U_{\perp} of the amplitude of a signal of frequency 18.5 GHz passing through the cavity when $H = H_e$ and a signal of frequency 140.1 GHz from the BWT has been fed to the cavity: 1), 2) constant component; 3), 4) variable component for a BWT modulation frequency of 60 and 2 kHz respectively. The line 4) was recorded with an amplification factor three times smaller than for the line 3).



FIG. 7. Recording of the photoresonance spectrum of electrons above 4 He.

The nondependence of the resonance width on E_1 allows us to assert that the broadening is not caused by the nonuniformity of the clamping field. The proportionality of the amplitude of the photoresonance to, and the nondependence of its width on, the power of the twomillimeter signal indicate that saturation effects do not play a role. The measured deviation of the frequency F, caused by the pulsations in the power supply of the BWT and the spurious frequency modulation, did not exceed 0.1 GHz, i.e., its contribution to ΔF_{11} is small. Thus, the given values of ΔF_{11} characterize the natural line width under experimental conditions.

The use of modulation of the two-millimeter radiation allows the investigation of the relaxation time, τ_{E} , of the excited electrons, since the signal height at the frequency $f_{\text{mod}} \gg 1/2\pi\tau_E$ is proportional to $(2\pi f_{\text{mod}}\tau_E)^{-1}$. The rapid decrease of the variable component of the photoresonance signal was observed, starting from f_{mod} ≈ 10 kHz (see Fig. 6). From the recordings given in Fig. 6, with account taken of a small correction for the amplitude-frequency characteristics of the receiver, we found with an accuracy of 50% that $\tau_{R} = 13 \ \mu \text{sec.}$ The measurement accuracy in our case was limited largely by the fact that, when f_{mod} was changed, the shape of the envelope of the signal generated by the BWT changed as a result of sharp drop in the frequency characteristic of the modulator, whose basic part was a transformer with a ferrite core.

If E_{\perp} is fixed, such that the amplitude of the variable photoresonance signal has its maximum value, then the dependence of the amplitude on the magnetic field is, as is easy to see, given by the formula





FIG. 8. Recording of the photoresonance spectrum of electrons above 3 He.



FIG. 9. Dependence of the $1 \rightarrow 3$, $1 \rightarrow 4$, and $1 \rightarrow 5$ transition frequencies on the clamping field for electrons localized above ³He (\bigcirc); •) for ⁴He according to Ref. 3.

i.e., it is determined by the difference between two resonance lines with widths $\Delta_1 H$, which is for excited electrons, and $\Delta_1 H$, which is for electrons in the ground state.

By measuring $\Delta_1 H$ in an independent experiment in which the cyclotron-resonance line width is determined from recordings of the type shown in Fig. 10, we can determine $\Delta_1 H$. The obtained $\Delta_2 H$ values are shown in Fig. 3. According to Fig. 10, $\Delta_3 H^{\approx} \Delta_2 H$. For electrons above ³He and $E_1 = 45$ V/cm, we have obtained $\Delta_3 H$ = $(0.9 \pm 0.1) \times \Delta_1 H$.

5. DISCUSSION OF THE RESULTS

1. Variation of the effective mass. Since the publication of Ref. 5, in which the discovery of the cyclotron-resonance shift in strong clamping fields was reported, Cheng and Platzman¹² have published a paper devoted to the theory of this effect. These authors proceeded from Shikin and Monarkha's¹ idea that a liquid surface under an electron localized in a plane by a mag-



FIG. 10. Recording of cyclotron-resonance signal from electrons above ⁴He at T = 0.45 K for f = 18.5 GHz (1); 2) same when a signal of frequency 135 GHz is fed to the cavity; 3), 4) dependence on the magnetic field of the variable component of a signal that has passed through the cavity at BWT modulation frequence $f_{\rm mod} = 1$ kHz when radiation from the BWT with frequency 135 GHz (3, the $1 \rightarrow 2$ transition) and 169, 2 GHz (4, the $1 \rightarrow 3$ transition) is fed to the cavity. The dashed curve depicts the line shape computed with $\Delta_2 H/\Delta_1 H = 0.5$; $\Delta_3 H/\Delta_1 H = 0.5$. The value of $E_1 = 12.8$ V/cm.

netic field in an area³⁾ $S = \pi L_H^2$ is deformed.

Since the deformation of the liquid surface occurs slowly, the deformation is preserved during the transition of the electron from the first Landau level to the second. But since the area of the orbit increases in the process, the electron should change its position with respect to the z coordinate, which requires extra energy. As a result, the resonance occurs at such value of the *H* field for which, according to Ref. 12, the cyclotron frequency is lower than the electromagnetic-field frequency by the amount $\delta\Omega = e^2 E_1^{-2}/8\pi\hbar\sigma$, or

$$\delta H = -mec E_{\perp}^2 / 8\pi \hbar \sigma. \tag{18}$$

It should be noted that, in certain aspects, the formula (18) is in accord with experiment: indeed, in a broad range of E_1 -field values, $\delta H \propto E_1^2$ is frequency and temperature independent. The ratio of the slopes of the dependences $\delta H(E_1^2)$ for electrons above ⁴He and ³He is inversely proportional to the ratio of the surface-tension coefficients. However, the theory does not predict the observed (see Fig. 4) dependence—admittedly a weak one—of δH on the electron concentration. Further, the numerical δH values computed from (18) turn out to be roughly five times smaller than the experimentally obtained values.

In principle, the concentration dependence can be related, as has already been noted by the present author,⁵ with the finite dimensions of the cavity. As a result of this, the resonance should correspond to the frequency of the plasma oscillations with wave vector $\sim 2\pi/R$. According to Fukuyama's¹³ calculation, the plasma shift of the frequency for $n = n_{max}$ is equal to

$$\delta\Omega = -\omega_t^2/2\omega, \ \omega_t^2 = 2\pi n e^2 q/m = E_\perp e q/m;$$
⁽¹⁹⁾

in this case δH is proportional to E_1 , and, for $E_1 = 1$ kV/cm, is numerically equal to ~3 Oe, i.e., is an order-of-magnitude smaller than the difference between the resonance-shift values for the maximum, and a low, electron density.

Another possibility is connected with the fact that the electron-electron interaction can lead to a greater spatial localization of the electrons than a magnetic field can^{14} (see also Ref. 15). Evidently, this should affect the magnitude of the deformation and the cyclotron-resonance frequency shift. The construction of a theory of this effect falls outside the framework of our paper.

Let us point out another hypotehtical possibility of the deformation's being greater than what is given in Ref. 12. A liquid boundary is not sharp. According to some theoretical investigations, it is quite diffuse, so that, for example, for ³He the density varies from zero to the density in the volume over a distance ~15 Å.¹⁶ Clearly, when deformations ~10⁻⁹ cm are in question, allowance for the compression of the transition layer under pressure can be important from the point of view of the electron.

The behavior of $\delta H(E_{\perp}^2)$ for $E_{\perp}^2 < 2 \times 10^5 (V/cm)^2$ is of interest. By itself the kink in the dependence could have been interpreted as a transition from the mechanism described by the formula (18) to a more rapid growth of

 δH as *n* increases. But the qualitative difference in behavior of electrons localized above ³He and electrons localized above ⁴He indicates the necessity to seek another interpretation for the phenomenon. It is possible that the cause of the slowed-down variation of δH for electrons above ⁴He is the weak attenuation of oscillations in superfluid helium, as a result of which the deformation of the surface does not have time to get established during an electron-relaxation time ~10⁻⁸ sec. This question, however, requires a systematic theoretical investigation.

2. The cyclotron-resonance line width. As noted in Refs. 5 and 6, the line width is, up to a factor of 2, quite well described by the formula obtained in Ref. 17 for τ . However, as the subsequent experiments described in this paper have shown, this agreement is accidental, and is observed only in the limited range of clamping fields $20 \leq E_1 \leq 200 - 300 \text{ V/cm}$ (Fig. 3). Although the shape of the dependence $\Delta H \propto E_1^2$ is retained at higher values of E_1 , the corresponding ΔH values depend on temperature, frequency, and the electron concentration (Fig. 4).

In our opinion, the cause of the disagreement with the theory consists in the following. In Ref. 17, in the computation of τ , the electron-momentum-dependent collisions rate $\nu(p)$ was integrated over all the momenta in accordance with the Maxwell distribution. However, a strong magnetic field with $\hbar\Omega > kT$ leads to a situation in which virtually all the electrons turn out to be in one and the same quantum state—at the ground Landau level. All of them will then have the characteristic momentum $p_H^2 \approx \hbar^2/L_H^2 = e\hbar H/2c$. Therefore, the scattering of the electrons under conditions of quantization is rather described directly by the formula (14) from Ref. 17 for $\nu(p)$ with the value $p \approx p_H$ substituted into it. This formula in strong E_1 fields assumes the form

$$v(p) = \frac{1}{\tau(p)} = \frac{mkTe^{*}E_{\perp}^{*}}{2\hbar\sigma p^{2}} = \frac{mckTeE_{\perp}^{*}}{\hbar^{2}\sigma H},$$
(20)

which correctly reflects the dependence of ΔH on T, $\Omega = eH/mc$, and σ . Numerical agreement with experiment for $n = n_{\text{max}}$ is attained if the result given by the formula (20) is divided by two.

However, this approach needs to be developed substantially further. In the first place, the fact that ΔH is not the same for the cases of total and partial charges (Fig. 4) clearly indicates the necessity for us to take the interelectron interaction into consideration. Secondly, the same formula (14) from Ref. 17 with $p = p_H$ gives in weak clamping fields too low values for ΔH (see Fig. 3).⁴⁾ And, finally, it does not reflect the decreasing section on the $\Delta H(E_1)$ curve. In principle, the growth of ΔH with decreasing E_1 could reflect the superheating of the electrons, which leads, according to Ref. 18, to the growth of the collision rate. As will be shown below, however, there was virtually no superheating in our case.

A probable explanation is this: when an electron is scattered by a ripplon with $q \approx p_H/\hbar$, the center of its orbit shifts a distance $y \sim cp_H/eH$. If this displacement

occurs in the field of other electrons, then the energy is changed by an amount $\sim e^2 y^2/d^3 \approx e \hbar c n^{3/2}/H$ (*d* is the interelectron distance). Clearly, scattering accompanied by a change in the position of the orbit center is possible if this energy is less than $\hbar \omega_q$. Remembering that $n = E_1/2\pi e$, we can easily figure out the fact that, for $E_1 \geq 15$ V/cm, the energy-conservation requirement imposes limitations on the possible scattering processes: at high E_1 only those collisions are possible which little change the position of the center of the electron's orbit. It is this limitation that apparently leads to the decrease of ΔH for $E_1 \sim 20 - 30$ V/cm.

Clearly, these considerations should be taken into account in the investigation of the scattering processes in strong E_{\perp} fields. However, judging by the fact that in this case the collision rate increases with increasing electron concentration, a very important role should be played by the process of energy-momentum transfer by the ripplons to the plasma oscillations. It should be noted that the role of purely electron-electron collisions is apparently insignificant, since they should have led to the same result for electrons above ³He and ⁴He. The fact that

 ΔH (³He)/ ΔH (⁴He) = σ_4/σ_3 ,

unambiguously indicates the participation of ripplons in the scattering processes.

3. Energy relaxation. Scattering by the ripplons having $q \approx p_H/\hbar \approx 10^5$ cm⁻¹ and frequency $\omega_q \approx 3 \times 10^7$ sec⁻¹ $\ll \Omega \approx 10^{11}$ sec⁻¹ cannot induce transitions between the Landau levels. Therefore, the energy relaxation is connected with the production of two ripplons with ω_q = $\Omega/2$ and oppositely directed momenta.¹⁹ This mechanism leads to clamping-field-independent characteristic time of relaxation in terms of energy, which is not at variance with experiment (Fig. 5) if we assume that the same degree of heating leads, for different E_{\perp} , to the same relative change in ΔH and δH .

The absence of a theory relating ΔH and δH with the temperature of the electron gas (strictly speaking, as shown above, there is no satisfactory theory even for electrons in equilibrium with helium) does not allow us at present to draw any quantitative conclusions from the experimental results shown in Fig. 5. However, using the results of the photoconductivity studies, we can make some estimates. Indeed, according to our measurements, the line width of the cyclotron resonance associated with the stimulation of the 1 + 2 transition coincides with the line width corresponding to the excitation of the $1 \rightarrow 3$ transition. At the same time, taking account of the fact that an electron in the l=3 state is located considerably farther away from the surface than an electron in the l=2 state (see the Introduction), we can expect that its interaction with the ripplons will be significantly weakened. The fact that a narrowing of the cyclotron-resonance line is not observed apparently indicates that the electron relaxes to the ground state with respect to l during a period of time determined by collisions with loss of momentum, i.e., to the Landau level⁵⁾ $n \approx 2\pi F_{11}/\Omega$. [Let us recall that for electrons above ⁴He, F_{12} is close to F_{13} under experimental conditions (Fig. 10).] In this case we should expect for the cyclotron-resonance line width roughly the same value as for the resonance that occurs upon heating electrons to the temperature $kT_e = n\hbar\Omega$. Let us take into consideration this circumstance, as well as the fact that the cyclotron-resonance line width decreases roughly by a factor of 2 upon the excitation of the photoresonance (Figs. 3 and 10). Then from the dependence $\Delta H(P)$ for $E_1 = 22 \text{ V/cm}$ in Fig. 5 we find that the electron temperature is raised by ~6 K upon the absorption of power ~10⁻¹⁰ erg/sec electron. Assuming that the superheat is proportional to the power (i.e., the relaxation time is temperature independent), we obtain for the superheat under the conditions of the measurements ($P \approx 10^{-12}$ -10^{-13} erg/sec·electron) discussed in Subsecs. 1 and 2 of this section the estimate $\Delta T_e \approx 6 \times 10^{-2} - 6 \times 10^{-3}$ K. Under the same assumptions, we obtain with the aid of the equation of energy balance the estimate $\tau_{E} \approx kT_{e}/P$ = 10^{-5} sec, which, considering the crudeness of the estimate, is in excellent agreement with the measured value, $\tau_{E} = 13 \ \mu sec$, given in Sec. 3. Thus, our argument turns out to be intrinsically consistent.

In Ref. 19 an estimate is obtained for τ_E for the case of two-ripplon production: $\tau_E = 10^{-6}$ sec. Allowing for the dependence of the theoretical value on the specific model and the highly approximate character of the experimental determination of τ_E , this value can be considered to be satisfactorily close to the experimental value.

4. The photoresonance line width. Resonance transitions in the hydrogenlike spectrum have been observed before^{3,20} only for electrons above ⁴He at $T \ge 1.2$ K. The halfwidth of the resonance line under these conditions was determined by the collisions with the atoms of the vapor, and was proportional to the vapor density, being equal²⁰ to 1.3 GHz at T = 1.2 K. Let us note that the halfwidth of the cyclotron-resonance line under these conditions has the same value: $\Delta \Omega / 2\pi \approx 1.1$ GHz. According to the calculation carried out in Ref. 21, as the temperature is lowered to 0.4 K, at which temperature the scattering by the ripplons becomes dominant, the halfwidth of the photoresonance line should decrease drastically and become equal to ~10 MHz. Contrary to this prediction, we have observed photoresonance for electrons above ⁴He with a line halfwidth ~0.45 GHz.⁶) It should be noted that this value is also much greater than the cyclotron-resonance line halfwidth at T = 0.4 K, this width being equal to ~30 MHz in weak clamping fields.

For electrons above ³He at 0.4 K, the scattering by the vapor atoms predominates in weak clamping fields. Decreasing the value experimentally obtained by Grimes *et al.*³ for the photoresonance line halfwidth in proportion to the vapor density and the parameter ratio γ_4/γ_3 (see the Introduction), we find that in this case we can expect the value $\Delta F_{11} \approx 60$ MHz, which is ~30 times smaller than the measured value $\Delta F_{11} = 1.6$ GHz. It is interesting to note that the cyclotron-resonance line halfwidth, which, as before, is much smaller than the photoresonance line halfwidth, is three times greater than for electrons above ⁴He. Thus, there is correlation between the photoresonance and cyclotron-resonance line widths for electrons above 3 He and 4 He.

At present the cause of such a large broadening of the photoresonance line is totally unclear. Judging by the results of Ref. 20, it is hardly connected with the electron-electron interaction. This question needs to be further investigated theoretically and experimentally. In particular, it should be ascertained whether a strong magnetic field that leads to the complete quantization of the electron spectrum has an effect on the line width.⁷ On the face of it, the magnetic field cannot affect the motion along the z-coordinate axis in any way, since for $H \| N$ the vector potential does not contain z, and the Schrödinger equation splits into two equations describing the motion in the direction perpendicular to, and the motion along, the liquid surface, respectively.¹ But the magnetic field has an effect on the spectrum of the plasma oscillations¹³ and, thereby, on the electronelectron interaction. The formulated problem can be solved unambiguously only by direct photoresonance measurements at low temperatures in zero magnetic field.

6. CONCLUSION

The performed investigations of the cyclotron resonance and photoresonance of electrons localized above liquid ³He and ⁴He have allowed us to reveal a number of distinctive features in their behavior that are due to the presence of a strong clamping field, the quantization of their spectrum, and their superheating relative to the liquid helium. The dependence of the effective mass on the clamping field and the electron concentration has been established. The effect of the quantization on the cyclotron-resonance line width, ΔH , has been elucidated, and an anomalous behavior of $\Delta H(E_{\perp})$ at low values of E_1 has been found. The resonance-line shift was observed to change sign during the heating of the electrons: if at electron temperatures <1 K the switching on of the clamping field leads to a shift of the cyclotron resonance toward the region of weaker Hfields, when they are heated to $T \ge 10$ K the shift of the resonance is toward the region of higher fields. It has been found that the photoresonance line width remains quite considerable at low temperatures, and exceeds the theoretically predicted value by almost two orders of magnitude.

The relationships observed in the investigation of the cyclotron resonance can be qualitatively interpreted. It has, however, been found that the change in the effective mass significantly exceeds the change predicted by theory,¹² and that the scattering of the electrons cannot be described by a theory that neglects the quantization of the spectrum and the finite density of the electrons. Thus, the performed measurements primarily raise for theory new problems whose solution can make subsequent experiments more purposeful. At the same time we can even now expect interesting information to be obtained in experiments performed at $T \approx 0.1 - 0.01$ K, when we can, in accordance with our results, expect appreciable narrowing of the cyclotron-resonance line at both low and high values of E_1 . Measurements

in this temperature region would allow us not only to raise the accuracy of the determination of the dependence $\delta H(E_1)$, but also to establish with great reliability the role of the electron-electron interaction, whose direct effect on the behavior of the electrons has so far not been determined.

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- ¹⁾Somewhat more tedious calculations show that the formula (11) is valid in the strong-coupling case as well.
- ²⁾The maximum microwave-signal power dissipated in the cavity was $\sim 10^{-7}$ W; this could lead to the superheating of the helium by $\sim 10^{-5}$ K.
- ³⁾Since the Landau-level splitting $\hbar \Omega > kT$, it is sufficient to consider only electrons in the ground state.
- ⁴⁾This is possibly connected with the fact that the parameter in powers of which the electron-ripplon interaction potential is expanded in Ref. 17 is in fact not too small, or with the fact that the liquid boundary is highly diffuse.¹⁶
- ⁵⁾Such a relaxation scheme also correlates with the result obtained for electrons above ³He: for scattering by the gas, the computed cyclotron-resonance line width in the l=3 state is 5.6 times smaller than the value in the l=1 state. The measured ratio $\Delta_3 H/\Delta_1 H=0.9$ (see Sec. 4).
- ⁶⁾It is interesting to note that in Ref. 3 the authors extrapolate the measured width to zero vapor density and find a residual line width of $0.3\sqrt{3} \approx 0.5$ GHz, which they ascribe to the effect of the nonuniformity of the clamping electric field.
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Nuclear quadrupole spin-lattice relaxation mechanism due to reorientation of the molecular fragments between unequal potential wells in a crystal

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A comparative analysis is made of the effectiveness of the magnetic and quadrupole modulation mechanisms of spin-lattice relaxation for quadrupole nuclei following reorientation of a neighboring molecular fragment that moves between equilibrium positions separated by unequal potential barriers. The results of the analysis are used to interpret the experimental data obtained for $T_1(T)$ in a $CL_3P = NCCl(CF_3)_2$ crystal by the method of nuclear quadrupole resonance on the chlorine nuclei of a $CCl(CF_3)_2$ fragment moving in a potential with unequal wells and of the PCl_3 group that is nonvalently bound to this fragment.

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The reorientation motion of atom groups in a crystal can act on the relaxation of neighboring nuclei by modulating either the magnetic or the electric interactions. For reorientations in an "equal-well" potential, it was shown in a number of $cases^{1-4}$ that modulation of the electric quadrupole interaction with the moving neighbors leads to a more effective relaxation mechanism for nuclei with large quadrupole interactions (halogens in covalent bonds, in contrast, e.g., to nitrogen nuclei⁵). The purpose of the present paper is to elucidate the distinguishing features of various modulation mechanisms and their effectiveness in reorientation motion of atomic groups between unequal potential wells. The modulation due to "unequal-well" motion was observed by us experimentally in the crystal Cl₃P = $NCCl(CF_s)_2$. In this compound one observes the influence of reorientational motion of the asymmetrical fragment $CCl(CF_3)_2$ between three equilibrium positions separated by unequal potential barriers, on the spinlattice relaxation of the chlorine nuclei in the $PC1_3$ group.⁶ Therefore in the exposition that follows all the concrete numerical estimates that follow from the theoretical analysis are made by us for chlorine nuclei (spin 3/2) and are valid for a large class of chlorinecontaining compounds.

1. MODULATION OF MAGNETIC DIPOLE-DIPOLE INTERACTONS

Let a nucleus with spin I and nonzero electric quadrupole moment be coupled to n neighboring magnetic nuclei having a spin S and a magnetic dipole-dipole interaction described by the Hamiltonian

$$\mathscr{H}_{aa} = \gamma_i \gamma_s \hbar^2 \sum_{i}^{n} \frac{(\mathbf{IS}_i) r_i^3 - 3(\mathbf{Ir}_i) (\mathbf{S}_i \mathbf{r}_i)}{r_i^5} = f(r_i, \theta_i, \varphi_i).$$
(1)

Here r_i , θ_i , φ_i are the polar coordinates of the radius vector \mathbf{r}_i joining the quadrupole nucleus with the *i*th magnetic nucleus in the system of the principal axes of the tensor of the electric field gradient (EFG) at the quadrupole nucleus. If the cause of the nuclear spinlattice relaxation are random modulations of this interaction, which result from the reorientational molecular motions, then the probability of the relaxation transition between states *m* and *m'* for a quadrupole nucleus with I = 3/2 is given by⁷

$$W_{mm'} = \hbar^2 \int_{-\infty}^{\infty} g_{mm'}(\tau) \exp\left(-i\omega_{\mathbf{q}}\tau\right) d\tau, \qquad (2)$$

where $g_{mm'} = \operatorname{Sp} \mathscr{H}_{mm'}(0) \mathscr{H}_{m'm}(\tau)$ is the correlation function (the summation under the spur sign is carried out over all the states of the magnetic nuclei), $\omega_q = 2\pi v_Q$, v_Q is the frequency of the nuclear quadrupole resonance (NQR) of the considered nucleus.

In analogy with Ref. 5 we can obtain for the probabilities W and W of one-quantum and two-quantum transitions, respectively, in the general case of arbitrary EFG symmetry, the expressions

$$W_{i} = \frac{9}{32} \left(1 + \frac{2}{1 + \eta^{2}/3} \right) \frac{(\gamma_{i} \gamma_{s} \hbar)^{2}}{\omega_{q}} \sum_{i} \frac{1 - \cos^{i} \theta_{i}}{r_{i}^{*}} F, \qquad (3)$$

$$W_2 = \frac{9}{4} \left(1 - \frac{1}{1 + \eta^2/3} \right) \frac{(\gamma_i \gamma_s \hbar)^2}{\omega_Q} \sum_i \frac{\sin^2 \theta_i \cos^2 \theta_i}{r_i^6} F, \qquad (4)$$

where η is the asymmetry parameter of the EFG tensor, F is a function of the dimensionless parameter $\omega_0 \tau_c$,

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