Cyclotron induction on above-surface levels in solids

B. P. Smolyakov and N. K. Solovarov

Physicotechnical Institute, Kazan' Branch, USSR Academy of Sciences

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Results are presented of an investigation of cyclotron induction in metal and semiconductor samples located in a high vacuum. A physical model is proposed, based on the assumed existence of long-lived above-surface levels that are unoccupied under normal conditions. The model can explain the principal features of the observed phenomenon.

1. INTRODUCTION

The first observation of resonant microwave induction signals from a number of metals located in a high vacuum ($\sim 10^{-6}$ Torr) at low temperatures (4.2 K) was reported in Ref. 1. It was suggested that the effect is due to cyclotron resonance with surface electrons. We propose here for this phenomenon a physical model based on the premise that there exist surface levels, which are not populated under equilibrium conditions, and on which electrons are localized mainly above the surface of the solid (above-surface levels).[2–6] We propose that microwave-field pulses excite in the skin layer electrons from the Fermi level and from other populated subsurface levels to unoccupied above-surface levels, followed by excitation of resonance cyclotron induction on the latter. This model permits a qualitative explanation of most experimentally observed distinctive properties of induction on metals and semiconductors.

2. APPARATUS AND MEASUREMENT PROCEDURE

A block diagram of the setup employed is shown in Fig. 1. A glass ampule containing the investigated sample was placed at an antinode of the electric component of a microwave field in a cavity resonator. The induction was excited by a pulsed microwave magnetron generator with fixed carrier frequency 9381.4 MHz. The pulse duration was 40 ns with maximum pulse power up to 3 kW. The pulse repetition frequency could be varied from a single shot to 2 kHz. To search for echo signals, provision was made for generating microwave pulse pairs with a minimum interval 0.1 μs. The resonance induction signals were recorded by a traveling-wave-tube receiver with sensitivity $\sim 10^{-12}$ W and adjustable pass band from 10 to 100 MHz. The induction-signal envelope was observed on the screen of a high-speed oscilloscope. Provision was made for plotting the induction intensity vs the constant magnetic field (Fig. 2). To this end, the induction signal was strobed, fed to an integrator, and from it to the Y input of an XY recorder. The X input was a voltage proportional to the linearly varying magnetic field. For the investigation of the possibility of resonance absorption of weak microwave pulses by the excited above-surface levels, the setup included a tunable (9380 ± 90 MHz) magnetron oscillator with pulse power up to 10 W and adjustable duration from 0.1 to 10 μs. The measurements could be made at both room and liquid-helium temperatures. In the latter case, a high vacuum was produced in the ampules with the investigated samples by cryogenic evacuation.

3. EXPERIMENTAL RESULTS

Resonance-induction signals were observed from samples of all the metals investigated by us (in increasing order of atomic weight: Be, Mg, Al, Fe, Co, Ni, Cu, Ag, Cd, In, Sn, La, Ce, Sm, Eu, Tb, Dy, Ho, Er, Au, Hg, Pb and semiconductors InSb, CdHgTe, Ge). Induction was generated only when the sample was placed in the antinode of the electric component of the microwave field of the cavity resonator. Stable signals were observed when the constant magnetic field **H** was perpendicular to the sample surface. An insignificant (a few degrees) deviation from this condition caused the induction signal to vanish. A mandatory condition for signal observation was placement of the sample in a highly

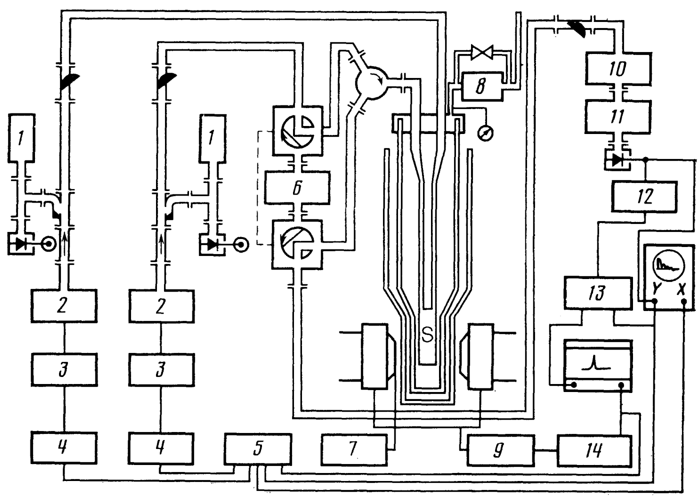

FIG. 1. Block diagram of experimental setup: 1—Wavemeter, 2—pulsed microwave generator, 3—modulator, 4—submodulator, 5—pulse-train generator, 6—rocking-frequency generator, 7—magnetic-induction meter, 8—vacuum pump, 9—magnet power supply, 10—TWT receiver, 11—bandpass filter, 12—broadband amplifier, 13—integrator, 14—scanning block, S—cavity with sample.

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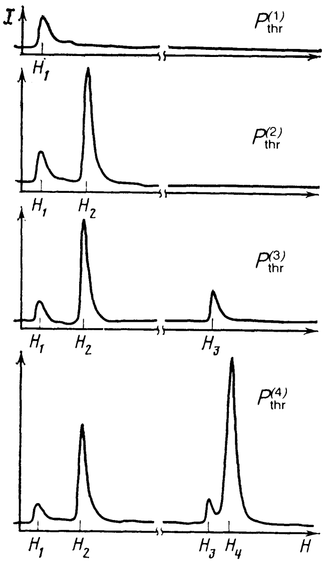

FIG. 2. Cyclotron-induction signal intensity I vs the constant magnetic field H in successive excitation of the signals as $P^{(i)}_{thr}$ is reached for each of the four lines of the Tb sample. $P^{(2)}_{thr}/P^{(1)}_{thr} = 2.7$ dB, $P^{(3)}_{thr}/P^{(2)}_{thr} = 1.6$ dB, $P^{(4)}_{thr}/P^{(3)}_{thr} = 1.8$ dB.

evacuated ampule. In our experiments a vacuum of about 10^{-2} Torr was produced in the ampule with the sample and the two were then placed in a microwave resonator immersed in liquid helium. The main role of the low temperature was apparently to obtain a high vacuum. Favoring this assumption is that induction signals were observed also at room temperature if a vacuum $\sim 10^{-6}$–10^{-7} Torr was attained in the ampule. Higher pressure weakened the induction signals and shortened their duration.

The effect was patently resonant in the constant magnetic field for both metals and semiconductors. The line widths of all the investigated samples did not exceed 10–20 Oe, the spectral width of the exciting microwave pulse. For each substance there was a definite number (from one to four) of values of the constant magnetic field at which resonance excitation of induction signals took place.

Another distinctive feature of the induction was that its excitation started at a certain (threshold) microwave-pulse power. Each line i of a substance had its own threshold power $P^{(i)}_{thr}$. At a specific sample orientation relative to the electric component $\mathscr{E}$ of the alternating field and relative to the direction of the magnetic field **H** the values; of $P^{(i)}_{thr}$ for a given substance are constant. The threshold power corresponds to a constant value of the resonant magnetic field ($P^{(i)}_{thr} \leftrightarrow H_i$). If the sample is characterized by several resonance lines with different values of $P^{(i)}_{thr}$ ($P^{(1)}_{thr} \leqslant P^{(2)}_{thr} \leqslant ... P^{max}_{thr}$), the lines were excited in succession as the microwave-pulse power was increased, as $P^{(i)}_{thr}$ was reached for each line. At $P \geqslant P^{max}_{thr}$, all the lines are excited, i.e., as the magnetic field goes through the resonance values H_i, all the resonance induction signals are observed. For Tb at 4.2 K, for example, induction signals were observed at four values of the magnetic field (in Oe)(: $H_1 = 3082.2$. $H_2 = 3102.2$ $H_3 = 3529.6$. $H_4 = 3545.1$ (Fig. 2). In this case, the value of P^{max}_{thr} on the sample surface was produced by a microwave generator with a pulse power 1.2 kW. Excitation of the maximum induction signal for a given line i required an optimal microwave-pulse power exceeding a threshold value ($P^{(i)}_{opt} > P^{(i)}_{thr}$). With further increase of the intensity of the microwave-field electric component in the cavity, an additional resonance induction signal was observed for most investigated samples at one and the same constant magnetic field $H_0 = 3351.3$ Oe.

Experiments were performed to determine the influence of a constant electric field on the character of the observed induction. An electric field up to 3 kV/cm was produced between two metallic samples in a high vacuum at room temperature. The metallic samples had different electron work functions. A lower induction-excitation threshold $P^{(i)}_{thr}$ was observed for the sample to which the negative potential was applied. When the field direction was reversed, the value of $P^{(i)}_{thr}$ for the same line increased. This points to an electronic origin of the observed phenomenon.

For most investigated samples, the decrease of the resonance-induction signal was modulated in amplitude, and the depth of the modulation reached 100%. (see Fig. 3), indicating the presence of two lines close in frequency, with almost equal amplitude of the induction signals. The modulation frequency did not exceed 100 kHz and behaved differently, with change of the microwave-pulse power and repetition frequency, for the different lines and samples. For example, when the power in Ge was decreased by 8 dB the moduation frequency was lowered from 10 to 7.5 kHz. When H_i was increased within the line-width range, the modulation frequency decreased from 10 to 8 kHz in the case of Ge).

Attempts were made to observe echo signals by applying microwave pulses of equal amplitude to different substances. No echo was observed at any possible interval between the microwave pulses. Production of gradients of the constant magnetic field at the sample did not change the

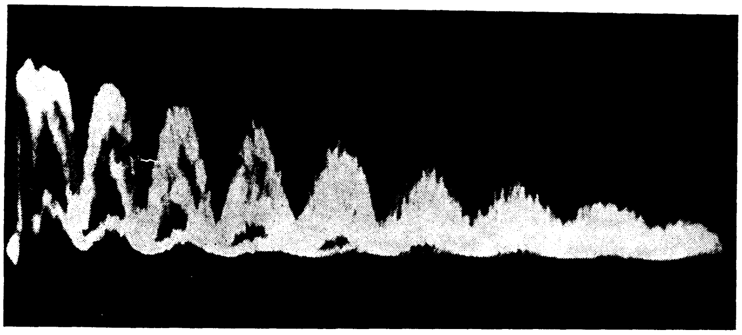

FIG. 3. Oscillogram of cyclotron-induction signal for Cu sample. The induction falloff is amplitude-modulated at a frequency 10^4 Hz; $H = 3382$ Oe, $T = 4.2$ K.

duration of the resonance induction. This indicates that the induction fall-off is little influenced by inhomogeneity of the constant magnetic field. When the power of the second microwave pulse was decreased, the pulse was resonantly absorbed in intervals exceeding the fall-off time of the induction signal. When the interval was increased, the absorption decreased with a time constant up to $\sim 10^{-2}$ s. No absorption was observed in response to single low-power microwave pulses. We emphasize that a second low-power resonance pulse is absorbed only if the first pulse has reached a power $P^{(i)}_{\rm thr}$ and is followed by observation of an induction signal.

The hypothesis advanced in Ref. 1 concerning the role of the surface in the observed effect is confirmed by a number of additional experiments on the change of the induction characteristics with change of the surface properties. Mechanical treatment of the surfaces of metals (Al, Cu, Sn) to produce a rough surface did not change the induction signals. Removal of the oxide surface from a copper sample by annealing in vacuum for a day at 950 °C increased $P^{(i)}_{\rm thr}$ by 2.5 dB and changed the resonant value of the magnetic field. A subsequent annealing of the sample in air (for several times then minutes), which restored to oxide film, restored also the initial values of $P^{(i)}_{\rm thr}$ and H_i. The use of vacuum-tube cathodes, which have high electron work functions, as the investigated samples revealed a correlation between the electron work function and the threshold power for the excitation of the resonance induction.

4. MODEL OF CYCLOTRON-INDUCTION EXCITATION

The proposed physical model is based on the presently accepted theory of the surface energy levels of solids.[4–6] The structure of such levels is sufficiently well described with the aid of the model of two-dimensional almost free electrons.[4] The main feature of electrons on surface levels is the almost free character of their motion in the surface plane. This property of the electrons determines the feasibility, in principle, of observing for them cyclotron resonance and cyclotron induction when samples are placed in a constant magnetic field perpendicular to the sample surface. Observation of cyclotron frequency and of cyclotron induction on a given level is possible if the level population is sufficient to excite a signal having a power exceeding the threshold sensitivity of the recording apparatus, and the lifetime of the electron on the level exceeds the period of the cyclotron motion of the electron. A condition of importance for the observation of cyclotron induction is also that the phase-relaxation time be long enough compared with the "dead" time of the receiving apparatus.

Figure 4 shows schematically the structure of the surface energy levels of electrons on the sample-vacuum interface, with an indication of their localization relative to the sample surface.[4,5] The motion of the electron on an above-surface level constitutes, in the semiclassical model, a series of sequential periodic reflections from the surface and of returns to the surface in the field of the image potential.[7,8] In the simplest case, when the wave-vector component and the electron energy of motion in the plane of the surface are zero ($k_\parallel = 0, E_\parallel = 0$), the electron executes periodic oscillations in a direction perpendicular to the surface. Quantization of the periodic motion determines the allowed values of the electron motion perpendicular to the surface ($E_{i\perp}$), meaning

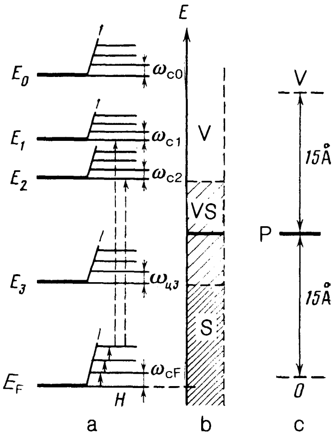

FIG. 4. Above-surface energy-level scheme[4,5]: a) E_F—Fermi level of sample electrons, E_0—vacuum level corresponding to the free electron, $E_{1,2}$—above-surface levels, E_3—subsurface level. Cyclotron splitting of the levels in a magnetic field is shown, ω_{ci} is the cyclotron frequency of the electron on the level i. b) Region of energy values at which the electrons are localized: S—in sample, VS—part in vacuum and part in sample, V—in vacuum. c) Near-surface region in which the electrons are localized: P—last atom plane, V—vacuum, S—sample volume.

the energy of the surface image levels[7,8] (called "reflecting states" in Russian). If $k_\parallel \neq 0$ and $E_\parallel \neq 0$, a translational motion of the electron along the surface is added to the oscillations perpendicular to the surface. Application of a constant magnetic field perpendicular to the sample surface limits the translational motion of the electron to the size of the cyclotron orbit, in accordance with the values of $k_\parallel$ and $E_\parallel$, and splits each level into an infinite almost equidistant sequence of cyclotron Landau levels with a splitting frequency $\omega_{ic} = eH/m_i^* c$ (see Fig. 4).

The course of the excitation of resonance induction can be described as follows. An electron in the skin layer on the Fermi level or on one of the populated subsurface layers in accelerated by the microwave-field electric component located in the surface plane, if the field frequency ω coincides with the electron cyclotron frequency on this level ($\omega = \omega_c$). The electron acquires energy by resonant interaction with the microwave field during the mean free path time or the electron-relaxation time (τ_r). When the energy acquired by the electron in the time τ_r coincides with the energy difference between one of the above-surface levels and the initial level, resonant population of the corresponding above-surface level takes place (the transitions are indicated by arrows in Fig. 4). Note that the term "resonant" population does not have here the usual meaning of frequency resonance. The electron cyclotron-motion frequency changes little in the course of the acceleration, in view of the relativistic increase of the effective mass. What we have here is energy resonance, viz., the electron cyclotron-motion energy E_c on the initial level E_F (for the sake of argument, we refer here to the Fermi level) becomes comparable with the energy difference, $E_c = E_i - E_F$

In this model, the microwave field raises in the course of the cyclotron resonance the energy of the electron motion, parallel to the surface, in the skin layer and over the sample surface. The mechanism of conversion from energy parallel to perpendicular to the surface ($E_\parallel \to E_\perp$) and accordingly the excitation of electrons to above-surface levels can be naturally related to electron scattering from the surface and from the skin layer, accompanied by a change of the wave vector. The electron lifetime on the ith above-surface reflecting layer is determined by the mechanisms that alter $E_{i\perp}$. Principal among them, in the absence of an external alternating field, are collisions with the gas molecules and with the surface. A high vacuum over the surface of the sample weakens the action of the first mechanism. The effectiveness of the second mechanism is determined by the surface-finish quality. For a real rough surface there is little likelihood of existence of long-lived reflecting states with $E_{\parallel} \neq 0$ in the absence of a magnetic field perpendicular to the surface. Application of such a field restricts the regions of electron reflection from the surface to the dimensions of the cyclotron orbit. The existence of long-lived above-surface reflecting electrons can then be connected with the "smooth" surface sections that have linear dimensions exceeding the diameter of the electron cyclotron orbit. The action of a frequency-resonant microwave pulse on electrons of a sufficiently long-lived above-surface level makes the electron cyclotron motion coherent, as manifested by a cyclotron-induction signal.[9]

The threshold character of the excitation of the induction signals is connected in the described model with the fact that the electrons on the populated subsurface level acquire within a time τ_r a cyclotron energy sufficient to populate the above-surface levels. It is natural to assume that the presence of paired splitting of most observed lines, as manifested in the modulation of the induction signal, is connected with the spin splitting of the electron levels in the magnetic field. Further investigations are needed, however, to construct a model that explains the modulation.

5. DISCUSSION OF EXPERIMENTAL RESULTS

The duration $\tau \sim 10^{-7}$ s of the employed microwave pulses exceeded significantly the characteristic times of collisional relaxation of the electrons on the Fermi level ($\tau_r < 10^{-10}$ s at 4.2 K, Ref. 10). We propose therefore that during the time of action of the pulse the electrons are excited to above-surface levels in a time $\Delta t \leqslant \tau_r \ll \tau$, while the remaining part of the pulse phases-in the cyclotron motion of the electrons on these levels. In accordance with the model assumed, the threshold power needed to populate an above-surface level can be estimated from the cyclotron energy imparted to the electrons in the resonant field within a time τ_r:

$$E_c = \mathscr{E}^2 e^2 \tau_p^2 / 2m^*, \tag{1}$$

where $\mathscr{E}$ is the intensity of the circularly polarized electric component of the microwave field in the skin layer of the sample. In our case we have $\mathscr{E} \leqslant 10^4$ V/cm in the empty cavity, and assuming $m^* = m_0$ (m_0 is the mass of the free electron) and $\tau_r < 10^{-10}$ s, we get $E_c < 100$ eV.

The methods most widely used at present to investigate solid-state surface-level spectra are low-energy electron diffraction (LEED)[11] and angular photoelectron (direct and inverse) spectroscopy.[11,12] The deviations of the energies of various surface states from the Fermi energies, obtained by these method, are limited to ~ 10 eV for the investigated solids. According to the estimate above, such values of electron cyclotron energies are attainable under our experimental conditions. An exact quantitative comparison of the published data on the surface-level spectra with our results is difficult in view of the various methods used to prepare the sample surfaces. In our case only the copper sample surfaces were specially treated. In addition, it is difficult to measure exactly the absolute values of the strength of the microwave electric field component on the surface of a sample in a cavity.

When the energy imparted to the electrons reaches the work function, electrons are removed to the vacuum. Indeed, with increase of the microwave pulse power, we observed for most investigated samples a line with H_0 corresponding to ω_{c0} of the free electrons. It should be noted that phenomena of like physical meaning were observed in Refs. 13 and 14 and were called cyclotron resonance with the secondary electrons induced by the microwave field. In these references are reported, for the cw mode and a high vacuum in the empty cavity, narrow (~ 0.2 Oe) absorption lines corresponding to one free-electron cyclotron resonance. The effect had, for the alternating-field power, a threshold that depended on the cavity-wall material (Cu, Ag). The model used for the explanation was based on the assumption that the hypothetical "primary" free electrons present in the vacuum near the cavity walls acquire an additional energy (~ 100 eV) by absorbing a few photons from the alternating field, and when these electrons collide with the cavity wall they knock out more than one electron (secondary electron emission). Cyclotron resonance with the knocked-out secondary electrons, is observed when their number reaches a certain threshold value. The threshold was found to be lowered by photoelectron emission induced by ultraviolet irradiation of the cavity walls, and it was demonstrated that the changes of the electron work function with change of the surface property (adsorption of known atoms, application of an electric field) are observable.

The above-surface levels considered by us can in principle be populated also by secondary electron emission. The cyclotron energy that can be imparted to the primary electron is limited, however, by the width $\Delta\omega$ of the microwave spectrum. Cyclotron acceleration of an electron is possible so long as the microwave-field spectrum contains frequencies corresponding to the electron frequency of the electron in a given constant magnetic field. The decrease of the cyclotron frequency by the relativistic increase (Δm) of the electron effective mass cannot exceed $\Delta\omega$ in cyclotron acceleration. Therefore the maximum cyclotron energy acquired by an electron in a resonance microwave field of frequency ω and with a spectrum width $\Delta\omega (\Delta\omega/\omega \ll 1)$ is given by

$$\Delta E_c = \Delta mc^2 = \Delta\omega m^* c^2 / (\omega - \Delta\omega) \approx \Delta\omega m^* c^2 / \omega. \tag{2}$$

The spectrum width $\Delta\omega$ was determined in our experiments by the duration of the microwave pulses; $\Delta\omega \approx \tau^{-1} \sim 10^7$ s^{-1} and $\Delta E_c \leqslant 5 \cdot 10^2$ eV. In the cw regime, usually, $\Delta\omega < 10^6$ s^{-1} and $\Delta E_c < 50$ eV. Although these estimates do not deny the possibility of a secondary electron emission mechanism in the population of above-surface (including vacuum) levels, we regard it as less probable than direct cyclotron excitations of electrons from populated subsurface levels. In this case the presence of threshold and optimal microwave–field powers upon excitation of the induction can be attributed to the spread of the electron mean free

TABLE I. Effective masses and resonant magnetic fields for electrons on various above-surface levels.

	m_1^*	H_1, Oe	m_2^*	H_2, Oe	m_3^*	H_3, Oe	m_4^*	H_4, Oe
Be	0.997	3342.4	1.008	3380.2	–	–	–	–
Al	0.997	3343.6	1.010	3382.1	1.025	3436.4	–	–
Fe	1.048	3510.2	1.065	3669.7	–	–	–	–
Cu	0.997	3344.1	1.010	3381.9	–	–	–	–
Ag	1.025	3435.2	1.003	3364.5	1.010	3382.1	–	–
In	0.997	3344.1	1.003	3362.1	–	–	–	–
La	1.003	3361.8	–	–	–	–	–	–
Au	1.003	3361.4	1.006	3372.5	–	–	–	–
Hg	1.003	3362.1	1.006	3370.4	–	–	–	–
Pb	0.997	3344.1	1.010	3382.1	–	–	–	–
Ce	1.006	3370.6	1.046	3503.9	–	–	–	–
Pr	1.003	3362.2	1.017	3408.6	–	–	–	–
Eu	1.003	3362.2	1.006	3372.5	–	–	–	–
Tb	0.919	3082.2	0.926	3102.2	1.053	3529.6	1.058	3545.1

Note m^* is in units of the free-electron mass.

time on the initial subsurface level. Assuming also the distribution to be Gaussian with a width $\Delta = 2(2 \ln 2)^{1/2}\sigma$, the probability of the cyclotron energies imparted to the electrons are determined from (1) to be

$$g(E_c) \sim \mathscr{E}^2 \tau_r^2 \exp[-(\tau_r - \tau_r^0)^2/2\sigma^2], \quad \tau_r \leqslant \tau, \tag{3}$$

where τ_r^0 is the electron mean free path time. Correspondence of the maximum of this curve to the energy difference between the initial and populated levels ($E_c^{\max} = W_i - E_F$) will determine the optimal population conditions and the maximum induction signal. For example, in the case of the observable four lines of Tb the relative change of powers of the exciting pulses corresponding to $P_{\text{opt}}^{(i)}$ and $P_{\text{thr}}^{(i)}$ is equal to

i	1	2	3	4
H_i, Oe	3082.2	3102.2	3529.6	3545
$P_{\text{opt}}^{(i)}/P_{\text{thr}}^{(i)}$, dB	1.75	1.5	2	1.75

It is known from the experimental investigations that the effective masses of the electrons on the surface levels differ from the mass of the free electrons and from the effective masses of the electrons inside the sample.[8,16] In our experiment the difference between the effective masses of the electrons on different above-surface levels manifests itself in the presence of several constant-magnetic-field values at which an induction signal is observed for a given substance. The electron effective masses listed in the table were obtained by us at $P = P_{\text{thr}}^{(i)}$ for the investigated line and for a pulse repetition frequency 1000 Hz. Note that when the pulse power is increased above the threshold or when the pulse repetition frequency is increased, an insignificant shift of the lines in terms of the magnetic field was observed.

The cyclotron-resonance linewidth on the Fermi level ($\sim \tau_r^{-1} > 10^{10}$ s^{-1}) greatly exceeds the linewidths of the observed cyclotron induction ($\sim 10^7$ s^{-1}). This explains why this experimental procedure, with a fixed microwave frequency spectrum, makes it possible to observe several induction signals with different resonant values of the constant magnetic field. For induction to be observable in the described model, it is necessary that the cyclotron frequency of the electrons on the initial occupied subsurface level, the cyclotron frequency of the electrons on the above-surface level, and the microwave-generator frequency coincide at the same value of the magnetic field.

It follows from estimates based on Eq. (1) that under cyclotron-resonance conditions, at the pulse powers employed, the electrons acquire on the above-surface level, in a time shorter than 10^{-10} s, an energy $\sim$1–10 eV sufficient for a transition to higher energy levels. Consequently, the electron lifetime on a resonant level during the action of the pulses does not exceed the indicated value. It follows hence that the observed induction signals are excited only by the trailing edges of the pulses. This is apparently one of the reasons why there are no cyclotron-echo signals in the described experiments. In fact, the leading edge of the second pulse shifts the electrons that contribute to the induction signal over to other levels, erasing thereby the phase memory of the first pulse.

6. CONCLUSION

We have demonstrated the possibility of resonant investigation of the energy structure of above-surface levels of metals and semiconductors. In our opinion, the method of observing cyclotron induction offers definite advantages over the method widely used to investigate surface levels,[4–6,11] since it permits direct meaurements of the effective masses, of the phase-relaxation times, and of the lifetimes of the electrons on the above-surface levels. It is promising to study the entire spectrum of the above-surface levels by combining the cyclotron-induction method with optical illumination of the sample surfaces. This approach will apparently make possible a study of the above-surface levels of dielectrics. An obvious promising task is a consistent study, with the aid of cyclotron induction, of the energy structure and the properties of above-surface levels of samples with specially treated surfaces.

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