Anisotropy of the electron-impurity transport relaxation time of indium

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Precision measurements of the anisotropy of relaxation of helicons were made for single-crystal spherical resonators made of indium and of dilute indium-base alloys. It was found that on a background anisotropy associated with the topology of the Fermi surface there were superimposed additional singularities which depended on the type of impurity and were governed by the anisotropic electron-impurity scattering. It was demonstrated that measurements of the helicon relaxation anisotropy could be used to investigate electron-impurity scattering in metals.

The transport properties of metals in the case of anisotropic scattering of conduction electrons by impurity atoms do not obey the majority of the approximate rules formulated in earlier stages of studies of transport phenomena. For example, the scattering anisotropy is used to account for deviations from the Köhler rule,[1] deviations from the Matthiessen rule dependent on the nature of impurities (see, for example, Ref. 2), and absence in real metals of the Peierls exponential fall of the electrical resistance.[3] It therefore follows that detailed information on electron-impurity scattering and its anisotropy is needed to describe transport phenomena in real metals. Experimental information on the scattering is essential also for the development of a theory of the electron-impurity interaction in metals. Unfortunately, these phenomena simply signal a possible anisotropy of the electron-impurity scattering and it is difficult to extract any information on the anisotropy itself from these phenomena.

At present there is essentially only one experimental method for investigating the electron-impurity scattering anisotropy of metals and it is based on the de Haas–van Alphen effect.[4] In spite of major capabilities of this method, it is subject to a number of serious limitations, which make it essential to supplement it by other independent methods.

We shall report precision measurements of the magnetoresistance anisotropy of indium in a strong magnetic field by the method of a helicon resonance[5] under conditions of predominance of the electron-impurity scattering and we shall show that a background anisotropy associated with the topology of the Fermi surface of indium has superimposed additional singularities that depend on the nature of the scattering impurity and are governed by the electron-impurity scattering anisotropy. We shall demonstrate that such measurements can be used as the basis of a method for investigating the anisotropy of the probabilities of electron-impurity scattering in metals with a known shape of the Fermi surface. Therefore, the helicon resonance measurements complement the data deduced from measurements of the scattering anisotropy carried out using the effects governed by extremal-path electrons, particularly the de Haas–van Alphen effect. The low sensitivity of the transport relaxation time to the scattering by dislocations and small-angle boundaries makes it possible to carry out measurements at low concentrations of impurities in the range where the solubility is complete. Measurements were made using impurity concentrations of 10^{-3} at.% or less. It should be pointed out that studies of the electron-impurity scattering in polyvalent metals, particularly in indium, with such a low impurity concentration are practically impossible if the de Haas–van Alphen effect is to be used because of the experimental difficulties resulting from the Fermi surface topology and from the dominant contribution of the small-angle processes of the scattering by crystal lattice defects.

EXPERIMENTAL METHOD AND PREPARATION OF SAMPLES

We used the helicon resonance method in which the samples that served as resonators were single-crystal indium spheres 10 mm in diameter. This eliminated at least two factors that increase the errors in the anisotropy measurements: electrical contacts and influence of the shape of a sample.

These measurements were made at a temperature of 1.3 K in magnetic fields up to 40 kG. We determined the Q factor of the fundamental helicon resonance (1,0) in the transverse geometry,[5] related to the components of the resistance tensor by[6]

$$Q=\alpha\left[\frac{\rho_{xx}+\rho_{yy}+\beta\rho_{zz}}{4\rho_{xy}}\right]^{-1}=\alpha(\rho_H/\rho_{xy})^{-1}, \qquad (1)$$

where the factors α and β are independent, apart from small corrections of the order of $(\omega_c\tau)^{-2}$, of the conductivity or magnetic field, and amount to 0.25 and 2.0, respectively. In Eq. (1) we ignored the contribution of the mixed longitudinal-transverse components of the resistivity, because estimates indicated that the components and the coefficients in front of them were small.

Our experiments satisfied the condition of the local limit ($l\ll\lambda\approx 2d$; l, λ, and d are, respectively, the mean free path of electrons, the helicon wavelength, and the diameter of a sample).

The experimental geometry is shown in Fig. 1. We recorded a signal which was cophasal with the current in an

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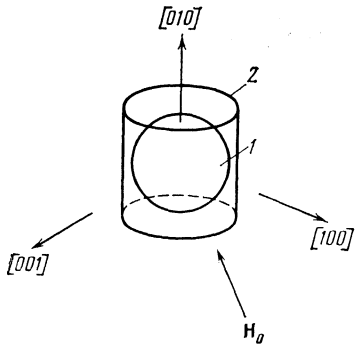

FIG. 1. Experimental geometry: 1) single-crystal spherical resonator; 2) detection (measuring) coil.

excitation coil. The excitation frequencies were 1–10 Hz. Special attention was paid to eliminate acoustohelicon effects.[7,8] Examples of experimental records of helicon resonances in spherical resonators are given in our earlier paper.[6]

A sample was rotated about an axis which coincided with the excitation and detection coils to within less than 0.5°. The rotation axis was perpendicular to a static magnetic field with an accuracy of at least 1°. The selected crystallographic axis of a sample was made parallel to the rotation axis to within better than 0.5° by an optical (laser) method.[9] The precision of the orientation of a sample was checked additionally in the course of an experiment by measuring the frequencies of the de Haas–van Alphen oscillations. The original indium samples of very high purity and with a resistivity ratio $\rho(300\text{ K})/\rho(1.3\text{ K}) \approx 500\,000$ were prepared by us using a method similar to that described in Ref. 10. The dopants were pure Zn, Ga, Pb, and Bi with distribution coefficients close to unity. The dopant concentration was selected to be in the range 10^{-2}–10^{-4} at.%, so that, firstly, a strong magnetic field condition ($\omega_c\tau \gg 1$) was satisfied in all cases and Eq. (1) was obeyed quite well and, secondly, the scattering by the dopant predominated. In reality, all the samples satisfied the condition $\omega_c\tau > 15$ and the contribution made to the helicon relaxation by the scattering on accidental impurities in the original samples did not exceed 3%.

The investigated alloys were prepared by a method of Ref. 11. The impurity concentration was monitored by chemical analysis methods. Special measures were taken to ensure that the impurity distribution in the samples was homogeneous. Single-crystal spheres were grown in a demountable quartz mold in which the surfaces were of the optical quality.

EXPERIMENTAL RESULTS

Figure 2 (curves a–e) showed the normalized, to the [001] direction, angular dependences of the helicon relaxation in the investigated samples obtained by rotation of a static magnetic field in the (100) plane. Clearly, in addition to common features such as deep minima along the [100] and [001] symmetry directions, the anisotropy exhibited also significant differences. There were changes in the depths and widths of the minima, new kinks appeared in the curves, etc. For example, the minimum along the [100] direction in the case of InGa was much deeper and wider than the corresponding minimum in the case of InZn. The angular dependences for the InBi and InPb alloys had inflections near the [101] direction, whereas the dependences for InGa and InZn showed no such inflections.

Variation of the impurity concentration altered the anisotropy in a similar manner, i.e., within the limits of the experimental error the angular dependences normalized to the [001] direction coincided. It should be stressed that these singularities of the anisotropy were not associated with changes in the Fermi surface parameters because of introduction of impurities. We confirmed this by measuring the de Haas–van Alphen oscillation frequencies, which for all the investigated samples agreed with high precision with the frequencies obtained for the pure samples. This was in agreement with the data reported for otheer metals, according to which impurities present in very low concentrations have a negligible effect on the electron energy spectrum.

The dependences of the helicon relaxation on the impurity concentration c were linear for all the investigated impurities, which was an indication that we were operating in the range in which impurities were fully dissolved. The slopes of the impurity concentration dependences were used to calculate the values of ρ_H/c, which were proportional to the transport scattering cross sections of conduction electrons. Their values for the [001] direction are listed in Table I.

This investigation was carried out at 1.3 K and the contribution to the anisotropy made by the scattering of electrons by phonons could be ignored, as deduced from the temperature dependences of ρ_H. When the magnetic field was altered by a factor of 2–3, the width of the helicon resonance line of all the samples and, consequently, the resistivity ρ_H and its anisotropy did not change within the limits of the experimental error and saturation was observed for the diagonal components of the resistivity tensor.

The magnetoresistance, i.e., the increase $\Delta\rho$ in the resistivity on application of a strong magnetic field, depended both on the direction–in accordance with the anisotropy (Fig. 2)–and on the nature of the dissolved impurity. Therefore, the Köhler rule was not obeyed by our experimental results, although the deviations from this rule were considerably smaller (Table I) than in the case of other metallic systems.[1]

DISCUSSION OF RESULTS

In the case of the isotropic electron–impurity scattering in the absence of magnetic breakthrough,[12] Eq. (1) describing the Q factor of a helicon resonance can be reduced to the following form[6]:

$$Q^{-1} = (\Phi/H)\,\tau_{\text{eff}}^{-1}, \tag{2}$$

where τ_{eff} is the effective relaxation time and Φ is a function governed by the parameters of the energy spectrum of conduction electrons in the host metal and dependent only on the orientation of the static magnetic field $\mathbf{H}$ relative to the crystallographic axes. This means that in the isotropic scat-

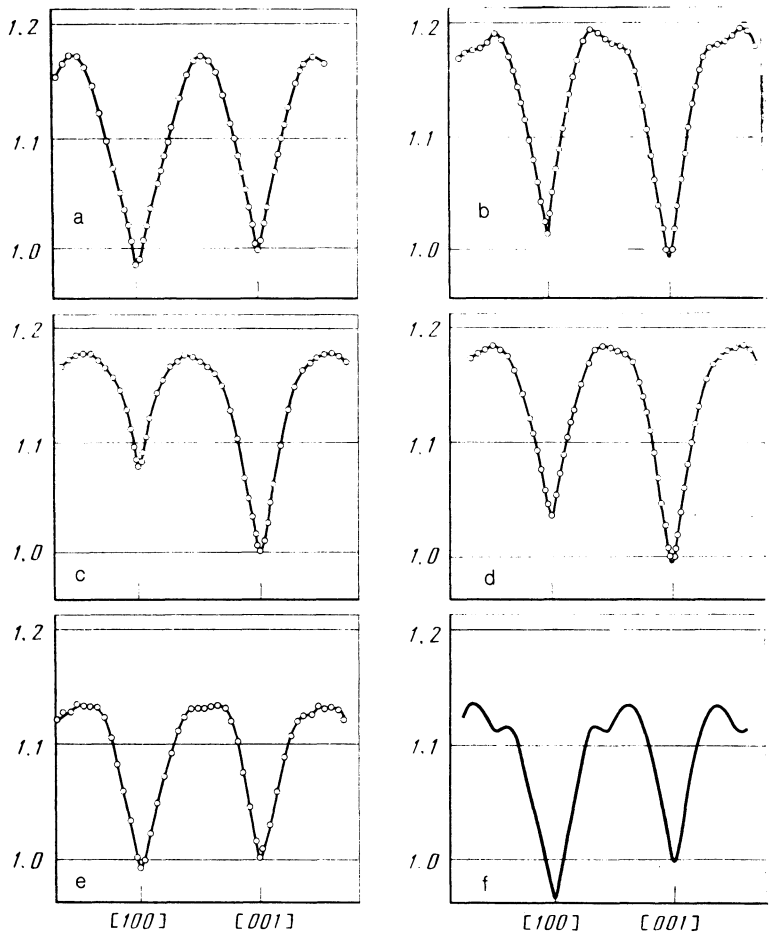

FIG. 2. Anisotropy of the helicon relaxation in single-crystal spheres of indium doped with various impurities (all curves normalized to the [001] direction): a) InGa alloy; b) InBi; c) InZn; d) InPb; e) commercial indium of the In-000 grade; f) theoretical curve representing the relaxation anisotropy in the case of the isotropic electron-impurity scattering.

tering case the angular dependence of Q^{-1} normalized to some direction should be a universal function for all the samples.

Figure 2f shows the angular dependence of Q^{-1} calculated using the known Fermi surface of indium and the isotropic scattering approximation, including the results of calculations of the components ρ_{ik} (Ref. 13). This angular dependence was used to deduce, with the aid of Eq. (2), the angular dependence of τ_{eff}^{-1} (Fig. 3) and to determine more readily the different contributions of the various impurities to the anisotropy.

The fact that the curves in Figs. 3 and 2 are not similar shows that the electron-impurity relaxation time of conduction electrons in a magnetic field defined in the usual way (see, for example, Ref. 14) is anisotropic and provides definite evidence of a strong anisotropy of the probability of the electron-impurity scattering in indium.

The problem of calculation of Q^{-1} in the case of the anisotropic elastic electron-impurity scattering reduces to the solution of the Boltzmann transport equation with a collision integral characterized by a relatively simple form[15]:

$$J = -\int (g-g')\,W(\mathbf{k},\mathbf{k}')\,d\mathbf{k}', \tag{3}$$

where g and g' are the values of the nonequilibrium correction to the distribution function in the initial ($\mathbf{k}$) and final ($\mathbf{k}'$) states of an electron; $W(\mathbf{k},\mathbf{k}')$ is the anisotropic scattering probability. However, even in this case the solution of the transport equation with an arbitrary form of $W(\mathbf{k},\mathbf{k}')$ meets with well-known difficulties.[15]

TABLE I.

	ρ_H/c, $\mu\Omega$·cm.(at.%)$^{-1}$	ρ_0/c, $\mu\Omega$·cm.(at.%)$^{-1}$*	$\Delta\rho/\rho_0$	η_0	η_1	η_2	η_3
Zn	0.084	0.03	1.8±0.2	0.063	0.032	0.006	0.001
Ga	0.365	0.14	1.6±0.1	–	–	–	–
Pb	1.345	0.58	1.3±0.05	0.154	0.144	0.047	0.007
Bi	4.04	1.72	1.3±0.1	0.233	0.254	0 099	0.017

*from Ref. 21

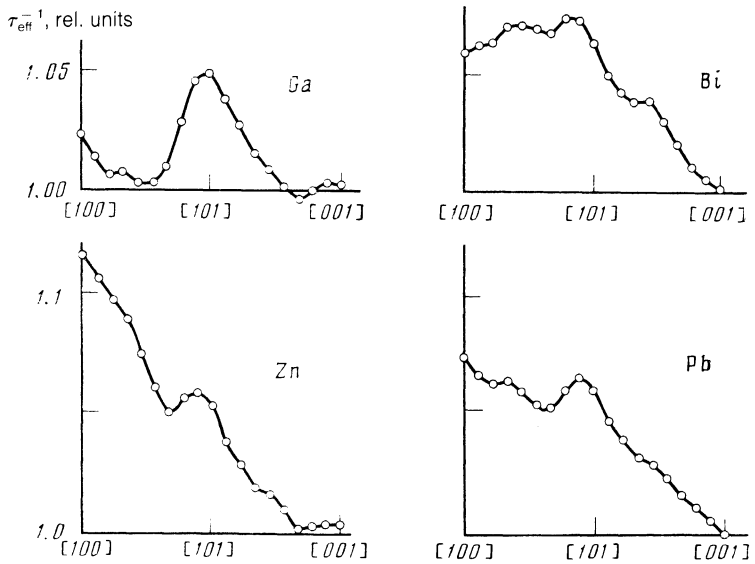

FIG. 3. Normalized angular dependences of the quantity τ_{eff}^{-1} [see Eq. (2)] plotted for various impurities in indium.

In some concrete situations when g and W are of special form the problem can be solved quite simply and the solutions can be expressed in terms of one anisotropic relaxation time. One of such situations is that when the "incoming" term in the collision integral vanishes:

$$J' = \int g' W(\mathbf{k}, \mathbf{k}')\, d\mathbf{k}' = 0. \tag{4}$$

In addition to the well-studied case when the nonequilibrium correction is localized, for example, in the case of the anomalous skin effect,[16] the condition (4) may be satisfied also in a static homogeneous electric field on condition that $W(\mathbf{k},\mathbf{k}') = W(\mathbf{k}, -\mathbf{k}')$ (Ref. 15). This is true, apart from the trivial case of the isotropic scattering ($W = \text{const}$), if for example only the phase shift predominates (in terms of the partial wave method) in the scattering process.[17] The relaxation time then depends on the initial state of an electron:

$$1/\tau(\mathbf{k}) = \int W(\mathbf{k}, \mathbf{k}')\, d\mathbf{k}'. \tag{5}$$

It therefore follows that the relationship between the phase shifts is of considerable importance in the analysis and solution of the transport equation. We calculated the phase shifts for the scattering of electrons in indium by Pb, Bi, and Zn impurities (see Table I) employing the Friedel-Blatt model in which the form of the scattering potential is considered to be a spherically symmetric rectangular well, the parameters of which are governed by the impurity valence and by the lattice deformation in the vicinity of an impurity.[18] According to several investigations, the model is in good agreement with the experimental results such as, for example, measurements of the electrical resistivity of metals with impurities of a valence other than those of the host metal.[19] In several investigations it has also been pointed out that the model is much less satisfactory when describing the scattering by isovalent impurities (see, for example, Ref. 20), so that in calculations of the phase shifts for gallium which is an isovalent impurity in indium, we need more rigorous models. In these calculations we used the values of the Blatt correction given in Ref. 21. It should be pointed out that the relationship between the phase shifts was not very sensitive to the Blatt correction, in contrast to the absolute values of the shifts and the impurity resistivity calculated from them. This increases the reliability of the conclusions which are drawn below.

The probability W of the scattering by impurities in simple metals, such as indium, can be written as follows[22,23]:

$$W(\mathbf{k}, \mathbf{k}') \propto \left| \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \exp(i\eta_l) \sin \eta_l a_{lm}(\mathbf{k})\, a_{lm}^*(\mathbf{k}') \right|^2 \delta(E_{\mathbf{k}} - E_{\mathbf{k}'}), \tag{6}$$

where a^* and a are the coefficients governed by the host metal and independent of the type of impurity; n_l are the phase shifts.

Using Eqs. (3) and (6), we can draw certain conclusions without solving the transport equation. For example, in the case of small phase shifts, if the ratios of the shifts characterized by different values of l are the same for a given group of impurities, we can say that the anisotropy of ρ_{ik}, and, consequently, of the helicon relaxation due to interaction with these impurities should be the same, because in this case the collision integrals for impurities are simply proportional to one another. We can see from Table I that this situation, in agreement with the adopted model, applies to the Bi-Pb impurity pair for which the ratio of the first two dominant phase shifts is unity to within 10%. The helicon relaxation anisotropy is also similar for these two impurities (see Figs. 2 and 3) and this impurity pair satisfies the Köhler rule (Table I).

An analysis of the transport equation was made in Ref. 23 using a collision integral corresponding to the "pure" s scattering in aluminum and corrections to the resistivity were calculated for the case when the p scattering was "acti-

vated." These numerical calculations showed that in the case of the s shift, which was twice as large as the p shift, there were no qualitative changes due to the allowance for the p scattering and the electrical conductivity was described by the relaxation time, the anisotropy of which depended weakly on the nature of the nonequilibrium correction and the value of which was close to that deduced from Eq. (5). We can see from Table I that such a situation occurs when conduction electrons are scattered by the impurity atoms of zinc in indium because in this case the phase shift corresponding to the s scattering is approximately twice as large as the p shift and ten times as large as the d shift. Although calculations similar to those for aluminum have not yet been carried out for indium, we can use the similarity of the electron structures of these two metals to assume that the relaxation time of indium given by Eq. (5) describes at least qualitatively the anisotropy of the resistivity and helicon relaxation in the case of scattering by the zinc impurity. In the τ approximation and in the limit of a strong magnetic field the resistivity tensor components are related to the scattering probability by the following expressions:

$$\rho_{xx}=C_1\int m_c\langle\nu P_y^2\rangle dp_z, \quad \rho_{yy}=C_1\int m_c\langle\nu P_x^2\rangle dp_z,$$
$$\rho_{zz}=C_2\left[\int\frac{m_c\langle v_z^2\rangle}{\langle\nu\rangle}dp_z\right]^{-1}, \quad \rho_{xy}=\frac{H}{(n_e-n_h)ec}, \tag{7}$$

where $P_i=p_i-\langle p_i\nu\rangle/\langle\nu\rangle$ is the component of the electron momentum which oscillates along an orbit; p_z is the projection of the electron momentum along the magnetic field direction; v_z is the electron velocity in the field direction; $\langle\cdots\rangle$ denotes averaging over the angle φ on the Larmor orbit;

$$C_1=4\pi/h^3(n_e-n_h)^2e^2, \quad C_2=h^3/4\pi e^2;$$

$\tau^{-1}(\mathbf{k})=\nu(p_z,\varphi)$ is the frequency of collisions with impurities dependent on the position of an electron on the Fermi surface and defined above [see Eq. (5)].

Equations (1) and (7) with the known parameters of the energy spectrum represent an integral equation relating the results of measurements of the helicon resonance Q factor for different directions of the magnetic field to the anisotropic collision frequency of conduction electrons $\nu(\mathbf{k})$.

We are now faced with the inverse problem of finding the anisotropic frequency of the electron-impurity collisions from the results of measurements of the anisotropy of the helicon resonance Q factor in a spherical single-crystal resonator.

An account of the theory and method for solving inverse problems, including an analysis of the problems of the existence, uniqueness, and stability of the solutions can be found, for example, in Ref. 24. We shall adopt the usual method of solution by inspection.[25] The procedure adopted consisted of the following steps: a) construction of an approximate model of $\nu(\mathbf{k},\nu_i)$ satisfying the Fermi surface symmetry of the metal and dependent on unknown parameters ν_i; b) selection of the parameters ν_i which give the best agreement between the anisotropy of the helicon relaxation $Q^{-1}(\nu_i)$ calculated from Eq. (7) and that found experimentally.

The Fermi surface of indium in the second zone, governing the conductivity of indium,[13] can be divided in the first approximation into regions of three types (Fig. 4a): region I representing hexagonal "cups" where the wave functions of electrons differ little from the wave functions of free electrons; regions II and III representing quadrilateral "cups" perpendicular to the [001] and [100] axes. It is assumed that electrons are perturbed most strongly in the region where the Fermi surface approaches closely the boundaries of the Brillouin zone, i.e., at the edges of the Fermi surface near the quadrilateral "cups."[26]

Bearing in mind these points, we can construct the following approximate model for the anisotropic ν:

$$\nu=\begin{cases}\nu_1=\text{const} & \text{in region I}\\ \nu_2\exp[-{}^1/_2(\Delta_2/\sigma)^2]+\nu_1 & \text{in region II,}\\ \nu_3\exp[-{}^1/_2(\Delta_3/\sigma)^2]+\nu_1 & \text{in region III,}\end{cases} \tag{8}$$

where Δ_2 annd Δ_3 are the distances of a point on the Fermi surface from the edges in regions II and III, respectively; σ are the angular dimensions of an extremum near the edges. An analysis shows that in the case of indium we have $\sigma=10$–$15°$. Therefore, bearing in mind that ν_1 can be calculated from the absolute value of Q^{-1}, our model is described simply by two parameters ν_2 and ν_3.

Figure 4b shows the angular dependence of τ_{eff}^{-1}, which occurs in Eq. (2) and depends on the magnetic field direction, calculated for the InZn alloy using the model described by Eq. (8); Fig. 4c shows the corresponding anisotropy of the frequency of the electron-impurity collisions on central sections of the Fermi surface in the (110) and (010) planes. The collision frequency anisotropy is independent of the magnetic field direction and is a characteristic of the alloy.

We shall now consider how this scattering pattern is in agreement with the existing ideas on the symmetry of the wave functions of electrons on the Fermi surface of indium. According to Ref. 23, the electron states have the p symmetry on all the edges of the Fermi surface of indium, apart from the edges of the quadrilateral "cups" perpendicular to [001], where the s-type states are concentrated. Consequently, in the case of the s scatterers the maximum of the probability $\nu(\mathbf{k})$ should be located near such a "cup" and the minima should be on the remaining edges. This is in good agreement with our results for InZn, which are dominated by the s scattering (Table I) and the probability of the scattering of electrons by "cups" perpendicular to [001] as maxima (Fig. 4c). The scattering probability then changes severalfold, depending on the position of an electron on the Fermi surface.

The results of the present investigation thus demonstrate that it is possible to obtain detailed information on the scattering of conduction electrons on impurities from measurements of helicon relaxation in the range of impurity concentrations where similar investigations by other methods are difficult. The precision of such measurements is sufficient to reveal fine features of the angular dependences of the relaxation, which are due to the nature of the scattering. A fuller interpretation of the experimental results will require further development of the methods for solving the trans-

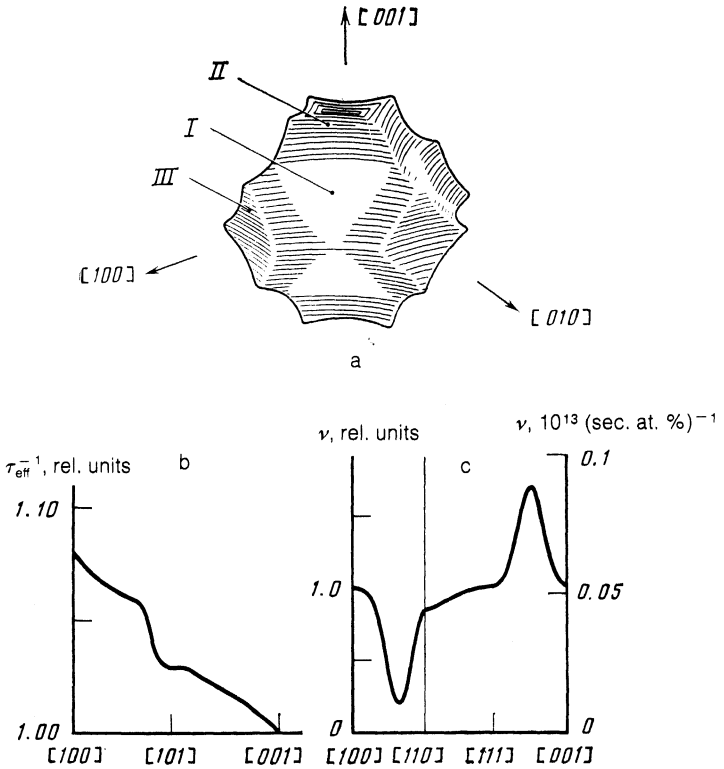

FIG. 4. a) Fermi surface of indium. Regions with very different wave functions of electrons are shown. b) Angular distribution of τ_{eff}^{-1} calculated for the alloy InZn in the case of the anisotropic electron-impurity scattering using the parameters given in Fig. 4c.

port equation in the case when the electron-impurity scattering is anisotropic.

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